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(54) Title
HETEROCYCLIC NEOPHANE ANALOGS, PROCESSES FOR THEIR PREPARATION AND THEIR USE AS PEST-COMBATING AGENTS

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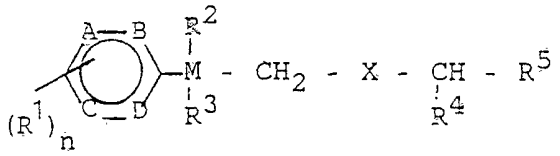
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(56) Prior Art Documents
AU 566515 27061/84 C07D 213/64 A01N 43/40 C07C 43/295
AU 31680/89 C07D 213/64 239/38 253/06 409/12

(57) Claim

1. A compound of the formula 1 and its stereoisomers



(I)

wherein

A, B, C or D independently of one another denote CH or N, in which at least one of the symbols A, B, C or D must correspond to a nitrogen atom,

M denotes a carbon ~~or silicon~~ atom,

X denotes CH_2 or oxygen,

R^1 denotes a radical bonded to a carbon atom from the series consisting of H, (C_1-C_4)alkyl, halogen, (C_2-C_6)-alkenyl, (C_2-C_6)alkynyl, (C_3-C_7)cycloalkyl, phenyl, phenoxy, $((C_1-C_5)$alkoxy, (C_2-C_4)alkenyloxy, (C_2-C_4)-alkynyloxy, (C_1-C_4)alkylthio, (C_3-C_7)cycloalkyloxy,

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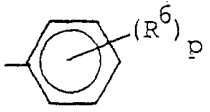
(C_1-C_4)halogenoalkyl, (C_1-C_4)alkoxy-(C_1-C_4)alkyl, (C_1-C_3)halogenoalkoxy, (C_1-C_3)halogenoalkylthio, halogeno-(C_1-C_4)alkoxy-(C_1-C_4)alkyl, (C_1-C_4)alkylthio-(C_1-C_4)alkyl, (C_1-C_4)alkoxy-(C_1-C_4)alkoxy, halogeno-(C_1-C_4)alkoxy-(C_1-C_4)alkoxy, (C_2-C_4)alkenyloxy-(C_1-C_4)-alkoxy, halogeno-(C_2-C_4)alkenyloxy, (C_1-C_4)alkoxy-(C_1-C_4)-alkylthio, (C_1-C_4)alkylthio-(C_1-C_4)alkoxy, (C_1-C_4)alkylthio-(C_1-C_4)alkylthio,

R^2 and R^3 independently of one another denote (C_1-C_3)-alkyl, (C_2-C_8)alkenyl or phenyl, or R^2 and R^3 denote an alkylene chain which - together with the quaternary central atom (M) - forms an unsubstituted or fluorine substituted ring having three ring members (for M = C),

R^4 denotes -H, F, $-CCl_3$, $-C\equiv CH$, (C_1-C_4)alkyl,

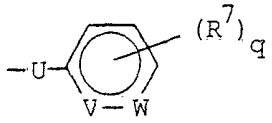
$-\overset{\overset{S}{\|}}{C}-NH_2$,

R^5 denotes a phenyl radical of the formula (A)



(A)

wherein R^6 independently of one another denote H, halogen, (C_1-C_4)alkyl, (C_1-C_4)alkoxy or (C_1-C_4)halogenoalkyl, phenyl or N-pyrrolyl or a radical of the formula (B)



(B)

wherein R^7 independently of one another denote H, halogen, (C_1-C_4)alkyl, (C_1-C_4)alkoxy or (C_1-C_4)halogenoalkyl,

U denotes $\equiv CH_2-$, $\overset{\diagdown}{\underset{\diagup}{C}}=O$, -O- or -S-,

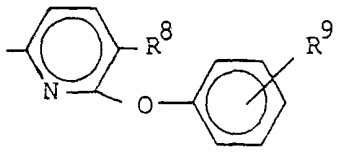
V or W denotes CH or N, in which W must denote CH where V denotes N and vice versa,

p denotes an integer from 1 to 5,

.../3

q denotes 1 or 2, or

a mono- or disubstituted pyridyl group of the formula (C)

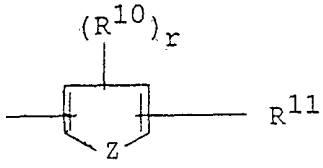


(C)

wherein R^8 denotes halogen or H and

R^9 denotes hydrogen or halogen with the exception of iodine, $(C_1\text{-}C_4)$alkyl, $(C_1\text{-}C_4)$alkoxy or $(C_1\text{-}C_4)$ halogenoalkyl, or

a substituted thienyl of the formula (D)



(D)

wherein Z denotes S,

R^{10} denotes H, halogen, $(C_1\text{-}C_4)$alkyl, $(C_1\text{-}C_4)$alkoxy, $(C_1\text{-}C_4)$halogenoalkyl, CN or NO_2,

R^{11} denotes benzyl, propargyl, allyl or phenoxy,

r denotes 1 or 2, and

n denotes 0, 1 or 2.



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COMPLETE SPECIFICATION

(ORIGINAL)

Class Int. Class

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Complete Specification Lodged:
Accepted:
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Priority :

Related Art :

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Complete Specification for the invention entitled:

HETEROCYCLIC NEOPHANE ANALOGS, PROCESSES FOR THEIR PREPARATION AND THEIR USE AS PEST-COMBATING AGENTS

The following statement is a full description of this invention, including the best method of performing it known to us

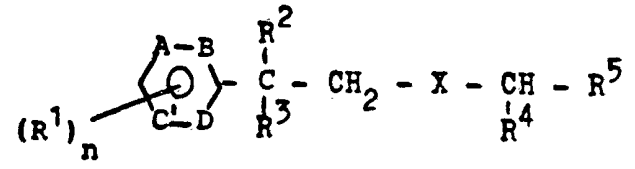
Description

Heterocyclic neophane analogs, processes for their preparation and their use as pest-combating agents

The insecticidal action of certain benzyl neophyl ether compounds and their hydrocarbon analogs has been disclosed in DE-A-3,117,510 and DE-A-3,317,908.

New heterocyclic neophane compounds having outstanding insecticidal, acaricidal and nematocidal properties have been found, which are clearly superior in their use properties to various conventional preparations.

The present invention therefore relates to the compounds of the formula (I) and their stereoisomers



(I)

wherein

A, B, C' or D independently of one another denote CH or N, in which at least one of the symbols A, B, C' or D must correspond to a nitrogen atom,

X denotes CH_2 or oxygen,

R^1 denotes a radical bonded to a carbon atom from the series comprising H, (C_1-C_4)-alkyl, tri(C_1-C_4)-alkylsilyl, halogen, nitro, cyano, (C_2-C_6)alkenyl, (C_2-C_6)-alkynyl, amino, (C_3-C_7)cycloalkyl, phenyl, phenoxy, (C_1-C_5)alkoxy, (C_2-C_4)alkenyloxy, (C_2-C_4)alkynyloxy, hydroxycarbonyl, (C_1-C_4)-alkylthio, (C_3-C_7)cycloalkyloxy, (C_1-C_6)alkylcarbonyl, (C_1-C_4)alkoxycarbonyl, (C_2-C_4)alkenyloxycarbonyl, (C_3-C_5)alkynyloxycarbonyl, (C_1-C_4)-halogenoalkyl, (C_1-C_4)-alkoxy(C_1-C_4)alkyl, (C_1-C_3)-halogenoalkoxy, (C_1-C_3)halogenoalkylthio, halogeno-(C_1-C_4)alkoxy(C_1-C_4)alkyl, (C_1-C_4)-alkylthio-(C_1-C_4)alkyl, (C_1-C_4)alkoxy(C_1-C_4)alkoxy,

alogeno(C_1-C_4)alkoxy(C_1-C_4)alkoxy, (C_2-C_4)alkenyloxy-(C_1-C_4)alkoxy, halogeno(C_2-C_4)alkenyloxy, (C_1-C_4)-alkoxy(C_1-C_4)alkylthio, (C_1-C_4)alkylthio(C_1-C_4)alkoxy, (C_1-C_4)alkylthio(C_1-C_4)alkylthio, halogeno(C_1-C_4)-alkoxycarbonyl, halogeno(C_2-C_4)alkenyloxycarbonyl or di(C_1-C_6-alkyl)-amino or two radicals R^1 when they are positioned ortho to one another together denote a methylenedioxy, ethylenedioxy or (C_3-C_5)alkylene radical,

R^2 and R^3 independently of one another denote (C_1-C_3)-alkyl, (C_2-C_8)alkenyl or phenyl, or R^2 and R^3 denote an alkylene chain which - together with the quaternary carbon atom - forms an unsubstituted or fluorine-substituted ring having three to six ring members,

R^4 denotes -H, F, -CN, $-CCl_3$, $-C\equiv CH$, (C_1-C_4)alkyl, $-\overset{\overset{N}{\|}}{C}-NH_2$, S

R^5 denotes pyridyl, furyl or thienyl which can all be substituted, phthalimidyl, di(C_1-C_4)-alkylmaleimidyl, thiophthalimidyl, dihydrophthalimidyl, tetrahydrophthalimidyl or substituted phenyl,

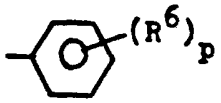
or R^4 and R^5 - together with the carbon atom bridging them - denote an optionally substituted indanyl, cyclopentenoyl or cyclopentenyl radical and n denotes 0, 1 or 2.

R^1 preferably denotes hydrogen, halogen, (C_1-C_4)alkyl, (C_2-C_6)-alkenyl, (C_3-C_7)cycloalkyl, (C_1-C_5)alkoxy, (C_2-C_4)alkenyloxy, (C_1-C_4)alkylthio, (C_1-C_4)halogenoalkyl, (C_1-C_3)halogenoalkoxy or - 2 R^1 radicals together - denote methylenedioxy, in which R^1 is positioned in particular in the 3- or 4-position of the heterocyclic radical.

R^2 and R^3 preferably stand for a (C_1-C_3)alkyl radical such as methyl, ethyl, i-propyl and n-propyl or preferably denote, together with the carbon atom bridging them, an unsubstituted or mono- or difluorinated cyclopropyl ring.

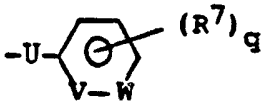
R^4 preferably stands for hydrogen, fluorine, cyano or (C_1-C_4)-alkyl, particularly preferably for hydrogen.

Substituted phenyl R^5 preferably stands for a phenyl radical of the formula (A)



(A)

wherein R^6 independently of one another denote H, halogen, (C_1-C_4)alkyl, (C_1-C_4)alkoxy, (C_1-C_4)halogenoalkyl, phenyl or N-pyrrolyl or a radical of the formula (B)



(B)

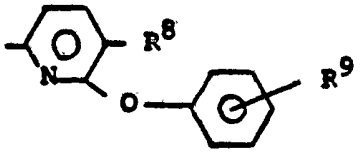
wherein R^7 independently of one another denote H, halogen, (C_1-C_4)alkyl, (C_1-C_4)alkoxy or (C_1-C_4)halogenoalkyl; U denotes $\equiv CH_2-$, $>C=O$, -O- or -S-, preferably -O-; V or W denotes CH or N, in which W must denote CH where V denotes N and vice versa,

p denotes an integer from 1 to 5, in particular 1 or 2 or in the case where R^6 = fluorine denotes to 5 and in the case where R^6 corresponds to the group (B), in particular denotes 1 or 2 and

q denotes 1 or 2.

Of these radicals, radicals of the formula (A) are of particular significance for R^5, wherein R^6 denotes H or 4-fluorine and additionally a radical of the formula (B) which is positioned in the 3-position of the phenyl radical of (A).

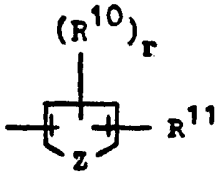
Optionally substituted pyridyl R^5 is, in particular, a mono- or disubstituted pyridyl group of the general formula (C)



(C)

wherein R^8 denotes halogen, in particular fluorine, or H and R^9 denotes hydrogen or halogen with the exception of iodine, (C_1-C_4)alkyl, (C_1-C_4)alkoxy or (C_1-C_4)halogenoalkyl.

Optionally substituted thienyl R^5 or furyl R^5 is, in particular, a heterocyclic ring of the general formula (D)



(D)

wherein Z denotes O or S,
R^{10} denotes H, halogen, (C_1-C_4)alkyl, (C_1-C_4)alkoxy, (C_1-C_4)halogenoalkyl, CN or NO_2,
R^{11} denotes optionally substituted benzyl, propargyl, allyl or phenoxy and
r denotes 1 or 2.

Substituted phenyl radicals for R^5 are of particular importance for the invention.

The following radicals are given as typical examples of the group R^5:
pentafluorophenyl, 5-benzyl-3-furyl, 4-phenoxyphenyl, 3-phenoxyphenyl, 3-(4-fluorophenoxy)phenyl, 3-(4-chlorophenoxy)phenyl, 3-(4-bromophenoxy)phenyl, 3-(3-fluorophenoxy)phenyl, 3-(3-chlorophenoxy)phenyl, 3-(3-bromophenoxy)phenyl, 3-(2-fluorophenoxy)phenyl, 3-(2-chlorophenoxy)phenyl, 3-(2-bromophenoxy)phenyl, 3-(4-methylphenoxy)phenyl, 3-(3-methylphenoxy)phenyl, 3-(2-methylphenoxy)phenyl, 3-(4-methoxyphenoxy)phenyl, 3-(3-methoxyphenoxy)phenyl, 3-(2-methoxyphenoxy)phenyl, 3-(4-ethoxyphenoxy)phenyl, 3-(phenylthio)phenyl, 3-(4-fluorophenylthio)phenyl, 3-(3-fluorophenylthio)phenyl, 3-benzoylphenyl, 3-benzylphenyl, 3-(4-fluorobenzyl)phenyl, 3-(4-chlorobenzyl)phenyl, 3-(3,5-dichlorophenoxy)phenyl, 3-(3,4-dichlorophenoxy)phenyl, 3-(4-chloro-2-methyl-

phenoxy)phenyl, 3-(2-chloro-5-methylphenoxy)phenyl, 3-(4-chloro-5-methylphenoxy)phenyl, 3-(4-ethylphenoxy)-phenyl, 3-(3-chloro-5-methoxyphenoxy)phenyl, 3-(2,5-dichlorophenoxy)phenyl, 3-(3,5-dichlorobenzoyl)phenyl, 3-(3,4-dichlorobenzoyl)phenyl, 3-(4-methylbenzyl)phenyl, 3-(4-isopropoxyphenoxy)phenyl, 4-fluoro-3-phenoxyphenyl, 4-chloro-3-phenoxyphenyl, 4-bromo-3-phenoxyphenyl, 4-fluoro-3-(4-fluorophenoxy)phenyl, 4-fluoro-3-(4-chloro-phenoxy)phenyl, 4-fluoro-3-(4-bromophenoxy)-phenyl, 4-fluoro-3-(4-methylphenoxy)phenyl, 4-fluoro-3-(4-methoxy-phenoxy)phenyl, 4-fluoro-3-(3-fluorophenoxy)phenyl, 4-fluoro-3-(3-chlorophenoxy)phenyl, 4-fluoro-3-(3-bromo-phenoxy)phenyl, 4-fluoro-3-(3-methoxyphenoxy)phenyl, 4-fluoro-3-(4-ethoxyphenoxy)phenyl, 4-fluoro-3-(2-fluoro-phenoxy)phenyl, 3-methoxy-5-phenoxyphenyl, 2-fluoro-3-phenoxyphenyl, 2-fluoro-3-(4-fluorophenoxy)phenyl, 2-fluoro-3-(3-fluorophenoxy)phenyl, 2-fluoro-3-(2-fluoro-phenoxy)phenyl, 3-fluoro-5-(4-fluorophenoxy)phenyl, 3-fluoro-5-(3-fluorophenoxy)phenyl, 3-fluoro-5-(2-fluoro-phenoxy)phenyl, 4-methyl-3-phenoxyphenyl, 3-fluoro-5-(4-methylphenoxy)phenyl, 3-fluoro-5-(3-methoxyphenoxy)-phenyl, 2-fluoro-5-(4-fluorophenoxy)phenyl, 2-fluoro-5-(3-fluorophenoxy)phenyl, 2-fluoro-5-(2-fluorophenoxy)-phenyl, 2-chloro-3-phenoxyphenyl, 3-fluoro-5-phenoxy-phenyl, 2-fluoro-5-phenoxyphenyl, 2-chloro-5-phenoxy-phenyl, 2-bromo-5-phenoxyphenyl, 4-chloro-3-(3-methyl-phenoxy)phenyl, 4-chloro-3-(4-fluorophenoxy)phenyl, 3-chloro-5-phenoxyphenyl, 3-bromo-5-phenoxyphenyl, 4-bromo-3-phenoxyphenyl, 4-trifluoromethyl-3-phenoxyphenyl, 4-fluoro-3-phenylthiophenyl, 4-fluoro-3-benzylphenyl, 3-(2-pyridyloxy)phenyl, 3-(3-pyridyloxy)phenyl, 4-fluoro-3-(2-pyridyloxy)phenyl, 4-chloro-3-(2-pyridyloxy)phenyl, 4-bromo-3-(2-pyridyloxy)phenyl, 4-methyl-3-(2-pyridyloxy)-phenyl, 4-fluoro-3-(3-pyridyloxy)phenyl, 4-chloro-3-(3-pyridyloxy)phenyl, 4-bromo-3-(3-pyridyloxy)phenyl, 4-methyl-3-(3-pyridyloxyphenyl), 2-methyl-3-phenylphenyl, 2-methyl-3-(N-pyrrolyl)phenyl, 6-phenoxy-2-pyridyl, 6-(4-fluorophenoxy)-2-pyridyl, 6-(4-chlorophenoxy)-2-pyridyl, 6-(4-bromophenoxy)-2-pyridyl, 6-(4-methylphenoxy)-

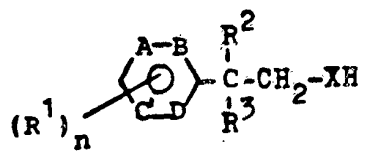
2-pyridyl, 6-(4-methoxyphenoxy)-2-pyridyl, 6-(4-ethoxyphenoxy)-2-pyridyl, 6-(3-fluorophenoxy)-2-pyridyl, 6-(3-chlorophenoxy)-2-pyridyl, 6-(3-bromophenoxy)-2-pyridyl, 6-(3-methoxyphenoxy)-2-pyridyl, 6-(2-fluorophenoxy)-2-pyridyl, 6-(2-chlorophenoxy)-2-pyridyl, 6-(2-bromophenoxy)-2-pyridyl, 5-propargyl-3-furyl, N-phthalimidyl, N-3,4,5,6-phthalimidyl, 2-methyl-5-propargyl-3-furyl, 4-t-butylphenyl, 4-methylphenyl, 4-isopropylphenyl, 4-(2-chloro-4-trifluoromethyl-2-pyridyloxy)phenyl, 4-cyclohexylphenyl, 4-difluoromethoxyphenyl, 4-biphenylyl, 4-trimethylsilylphenyl and 4-phenoxy-2-thienyl.

Further typical examples of the group $-\underset{R^4}{\underset{|}{CH}}-R^5$ are:

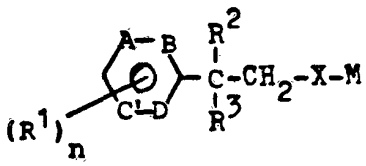
2-allyl-3-methylcyclopent-2-en-1-on-4-yl and 4-phenylindan-2-yl.

The present invention also relates to a process for the preparation of the compounds of the general formula (I), wherein

a) for compounds having X = O a compound of the formula (II) or (III)

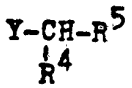


(II)



(III)

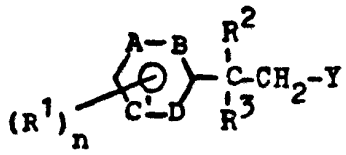
is condensed with an alkylating agent of the formula IV



(IV)

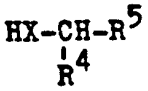
wherein Y denotes a nucleofugic leaving group such as, for example, halogen or sulfonate and M corresponds to an alkali metal equivalent or an alkaline earth metal equivalent, in particular Li, Na, K or Mg,
if appropriate in the presence of a base,
or

b) for compounds having X = O a compound of the formula (V)



(V)

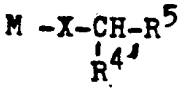
is condensed with an XH-acid compound of the type (VI)



(VI)

in the presence of a base

or with an organometallic compound of the type VII

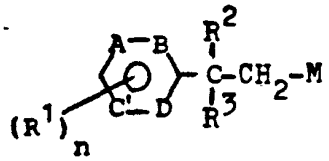


(VII)

having $R^{4'}$ = H or (C_1-C_4)alkyl

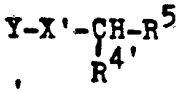
or

c) for compounds having X = CH_2 a compound of the general formula VIII



(VIII)

is condensed with a compound of the type (IX)

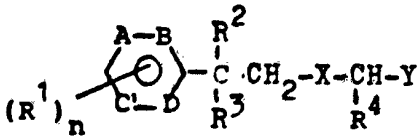


(IX)

having X' = CH_2,

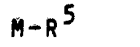
or

d) a compound of the formula (X)



(X)

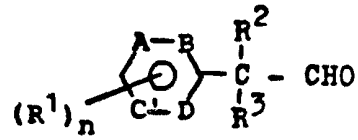
is condensed with an organometallic reagent of the type (XI)



(XI)

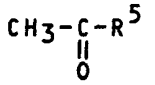
or

e) for compounds having R^4 = H and X = CH_2 an aldehyde of the formula (XII)



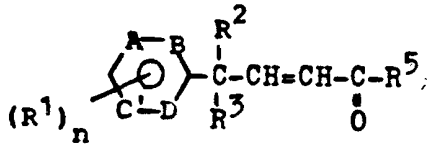
(XII)

is condensed with a ketone of the formula (XIII)



(XIII)

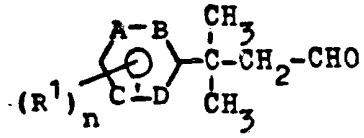
to form an intermediate (XIV)



(XIV)

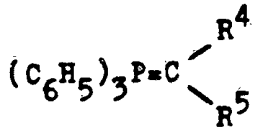
and this is subsequently reduced in a customary manner, or

f) for compounds having X = CH_2 an aldehyde of the formula (XV)

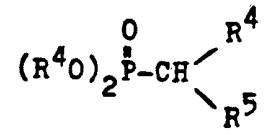


(XV)

is condensed with an ylide of the formula (XVI) or a phosphonate of the formula (XVII)

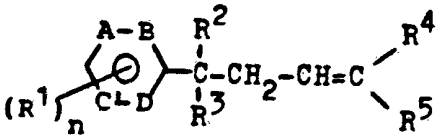


(XVI)



(XVII)

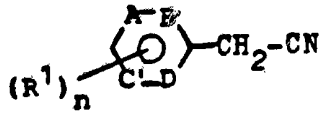
to form an intermediate (XVIII)



(XVIII)

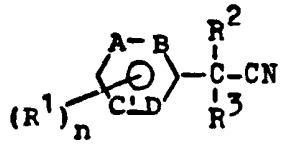
and this is subsequently reacted with a reductant to form the final product (I) having X = CH_2.

The substances (II) or (III) to be used as starting compounds in preparation process a) can be obtained, for example, by a multi-step synthesis from the heterocyclic rings (XIX)



(XIX)

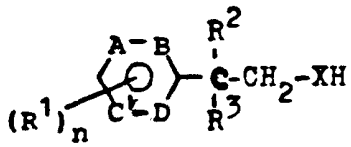
by first introducing the two radicals R^2 and R^3 stepwise with the aid of strong bases such as, for example, sodium hydride and alkylating agents such as alkyl iodide or dialkyl sulfate, by means of which the intermediate (XX) is obtained



(XX)

this is then first hydrolyzed to form the corresponding carboxylic acid and finally reduced using strong reducents such as, for example, $LiAlH_4$ to form the desired alcohol (II).

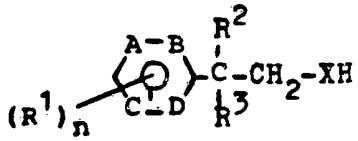
The heterocyclic rings of the type V to be used as starting compounds in preparation process b) can be obtained by halogenation of the alcohols (II) (X = O)



(II)

which is carried out by standard methods.

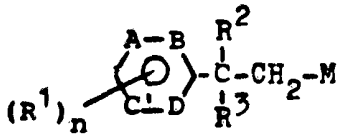
The aldehydes of the general formula (XII) required as starting compounds in preparation process e) can be obtained in a simple manner by oxidation of the corresponding alcohols (II) (X = O), (preparation described further above)



(II)

using the oxidant pyridine x SO_3.

The aldehydes of the general formula (XV) required as starting compounds in preparation process f) are obtained inter alia by carbonylation of the metal compounds (VIII)



(VIII)

which are obtainable from the corresponding halides (V) with, for example, N,N-dimethylformamide.

The precursors and intermediates which are not described in greater detail can be prepared by customary standard methods. Thus all mentioned organometallic intermediates can be obtained by customary metallization reaction such as hydrogen/metal exchange or - preferably - halogen/metal exchange in all its variations.

The mentioned process variations c) and d) are preferably carried out in a diluent whose nature depends upon the type of organometallic compound employed. Those diluents which are suitable in particular are aliphatic and aromatic hydrocarbons such as, for example, pentane, hexane, heptane, cyclohexane, petroleum ether, benzine, ligroin, benzene, toluene and xylene, ethers such as, for example, diethyl ether and dibutyl ether, glycol dimethyl ether, diglycol dimethyl ether, tetrahydrofuran and dioxane, and finally all possible mixtures of the previously mentioned solvents.

The reaction temperature in the abovementioned process variations is between -75°C and +150°C, preferably between -75°C and +105°C. The starting materials are customarily employed in equimolar amounts. An excess of one or the other reaction component is possible, however.

For process variations a), b), e) and f) mentioned further above, essentially the same applies as for variations c) and d). By employing the educts of the type (II) and (VI) and also the carbonyl compounds (XII) and (XIII), it is still possible to employ additional diluents, however. Thus, suitable diluents in these cases are also ketones such as acetone, methyl ethyl ketone, methyl isopropyl ketone and methyl isobutyl ketone, esters such as methyl acetate and ethyl acetate, nitriles such as, for example, acetonitrile and propionitrile, amides such as, for example, dimethylformamide, dimethylacetamide and N-methylpyrrolidone, and also dimethyl sulfoxide, tetramethylenesulfone and hexamethylphosphoric triamide. Bases used are inorganic bases such as, for example, hydroxides,

hydrides, carbonates or hydrogen carbonates of alkali metals or alkaline earth metals, and also organic bases such as, for example, pyridine, triethylamine, N,N-diisopropylethylamine or diazabicyclooctane.

The isolation and, where appropriate, purification of the compounds of the formula (I) takes place according to generally customary methods, for example by evaporating the solvent (if appropriate under reduced pressure) and subsequent distillation or chromatography or by dividing the crude product between two phases and subsequent customary workup from there on.

The compounds of the general formula (I) are readily soluble in most organic solvents.

The active compounds are suitable for combating animal pests, in particular insects, arachnida and nematodes, encountered in agriculture, in forestry, in the protection of stored products and of materials, and in the hygiene field, and well tolerated by plants and have favorable toxicity to warm-blooded animals. They are active against normally sensitive and resistant species and against all or some stages of development. The above-mentioned pests include:

From the order of the Isopoda, for example, Oniscus asellus, Armadillidium vulgare and Porcellio scaber. From the order of the Diplopoda, for example, Blaniulus guttulatus. From the order of the Chilopoda, for example, Geophilus carpophagus and Scutigera spec. From the order of the Symphyla, for example, Scutigerella immaculata. From the order of the Thysanura, for example, Lepisma saccharina. From the order of the Collembola, for example, Onychiurus armatus. From the order of the Orthoptera, for example, Blatta orientalis, Periplaneta americana, Leucophaea maderae, Blatella germanica, Acheta domesticus, Gryllotalpa spp., Locusta migratoria migratorioides, Melanoplus differentialis and Schistocerca gregaria. From the order of the Dermaptera, for example, Forficula

auricularia. From the order of the Isoptera, for example, Reticulitermes spp.. From the order of the Anoplura, for example, Phylloera vastatrix, Pemphigus spp., Pediculus humanus corporis, Haematopinus spp. and Linognathus spp.. From the order of the Mallophaga, for example, Trichodectes spp. and Damalinea spp.. From the order of the Thysanoptera, for example, Hercinothrips femoralis and Thrips tabaci. From the order of the Heteroptera, for example, Eurygaster spp., Dysdercus intermedius, Piesma quadrata, Cimex lectularius, Rhodnius prolixus and Triatoma spp.. From the order of the Homoptera, for example, Aleurodes brassicae, Bemisia tabaci, Trialeurodes vaporariorum, Aphis gossypii, Bravicornyne brassicae, Cryptomyzus ribis, Doralis fabae, Doralis pomi, Eriosoma lanigerum, Hyalopterus arundinis, Macrosiphium avenae, Myzus spp., Phorodon humuli, Rhopalosiphum padi, Empoasca spp., Euscelus bilobatus, Nephotettix cincticeps, Lecanium corni, Saissetia oleae, Laodelphax striatellus, Nilaparvata lugens, Aonidiella auroantii, Aspidiotus hederae, Pseudococcus spp. and Psylla spp.. From the order of the Lepidoptera, for example, Pectinophora gossypiella, Bupalus piniarius, Cheimatobia brumata, Lithocolletis blancardella, Hyponomeuta padella, Plutella maculipennis, Malacosoma neustria, Euproctis chrysorrhoea, Lymantria spp., Buceulatrix thurberiella, Phyllocnistis citrella, Agrotis spp., Euxoa spp., Feltia spp., Earias insulana, Heliothis spp., Laphygma exigua, Mamestra brassicae, Panolis flammea, Prodenia litura, Spodoptera spp., Trichoplusia ni, Carpocapsa pomonella, Pieris spp., Chilo spp., Pyrausta nubilalis, Ephestia kuehniella, Galleria mellonella, Cacoecia podana, Capua reticulana, Choristoneura fumiferana, Clysia ambiguella, Homona magnanima and Tortrix viridana. From the order of the Coleoptera, for example, Anobium punctatum, Rhizopertha dominica, Bruchidius obtectus, Acanthoscelides obtectus, Hylotrupes bajulus, Agelastica alni, Leptinotarsa decemlineata, Phaedon cochlearieae, Diabrotica spp., Psylliodes chrysocephala, Epilachna varivestis, Atomaria spp., Oryzaephilus surinamensis, Anthonomus spp., Sitophilus spp., Otiorrhynchus sulcatus,

Cosmopolites sordidus, Ceuthorrhynchus assimilis, Hypera postica, Dermestes spp., Trogoderma spp., Anthrenus spp., Attagenus spp., Lyctus spp., Meligethes aeneus, Ptinus spp., Niptus hololeucus, Gibbium psylloides, Tribolium spp., Tenebrio molitor, Agriotes spp., Conoderus spp., Melolontha melolontha, Amphimallon solstitialis and Costelytra zealandica. From the order of the Hymenoptera, for example, Diprion spp., Hoplocampa spp., Lasius spp., Monomorium pharaonis and Vespa spp.. From the order of the Diptera, for example, Aedes spp., Anopheles spp., Culex spp., Drosophila melanogaster, Musca spp., Fannia spp., Calliphora erythrocephala, Lucilia spp., Chrysomyia spp., Cuterebra spp., Gastrophilus spp., Hyppobosca spp., Stomoxys spp., Oestrus spp., Hypoderma spp., Tabanus spp., Tannia spp., Bibio hortulanus, Oscinella frit, Phorbia spp., Pegomyia hyoscyami, Ceratitis capitata, Dacus oleae and Tipula paludosa. From the order of the Siphonaptera, for example, Xenopsylla cheopis and Ceratophyllus spp. From the order of the Arachnida, for example, Scorpio maurus and Latrodectus mactans.

From the order of the Acarina, for example, Acarus siro, Argas spp., Ornithodoros spp., Dermanyssus gallinae, Eriophyes ribis, Phyllocoptruta oleivora, Boophilus spp., Rhipicephalus spp., Amblyomma spp., Hyalomma spp., Ixodes spp., Psoroptes spp., Chorioptes spp., Sarcoptes spp., Tarsonemus spp., Bryobia praetiosa, Panonychus spp. and Tetranychus spp..

Furthermore, the compounds have an excellent action against nematodes which are harmful to plants, for example those of the Meloidogyne, Heterodera, Ditylenchus, Aphelenchoides, Radopholus, Globodera, Pratylenchus, Longidorus and Xiphinema genera. The invention also relates to agents which contain the compounds of the formula I in addition to suitable formulation auxiliaries.

The agents according to the invention contain the active compounds of the formula I, in general from 1 - 95% by

weight. They can be used in the customary preparations as wettable powders, emulsifiable concentrates, sprayable solutions, dusting agents or granules.

Wettable powders are preparations which are uniformly dispersible in water, which, besides the active compound, in addition to a diluent or inert substance contain wetting agents, for example polyoxyethylated alkyl phenols, polyoxyethylated fatty alcohols, alkyl sulfonates or alkyl phenol sulfonates and dispersing agents, for example sodium ligninsulfonate, sodium 2,2'-dinaphthylmethane-6,6'-disulfonate, sodium dibutylnaphthalenesulfonate and also sodium oleylmethyltaurate.

Emulsifiable concentrates are prepared by dissolving the active compound in an organic solvent, for example butanol, cyclohexanone, dimethylformamide, xylene and also higher-boiling aromatics or hydrocarbons with the addition of one or more emulsifiers. Examples of emulsifiers which can be used are:
calcium alkylarylsulfonates, such as calcium dodecylbenzenesulfonate or non-ionic emulsifiers, such as fatty acid polyglycol esters, alkylaryl polyglycol ethers, fatty alcohol polyglycol ethers, propylene oxide/ethylene oxide condensation products, alkyl polyethers, sorbitan fatty acid esters, polyoxyethylene sorbitan fatty acid esters or polyoxyethylene sorbitol esters.

Dusting agents are obtained by grinding the active compound with finely divided solid substances, for example talc, natural clays such as kaolin, bentonite, poryphillite or diatomaceous earth. Granules can be prepared either by spraying the active compound onto adsorptive, granulated inert material or by applying concentrates of active compound by means of adhesives, for example polyvinyl alcohol, sodium polyacrylates and also mineral oils, to the surface of supports such as sand, kaolinites, or granulated inert material. Suitable active compounds can also be prepared in the customary manner for the

preparation of fertilizer granules - if desired mixed with fertilizers.

The active compounds according to the invention can be present in their commerically available formulations and in the use forms, prepared from these formulations, as a mixture with other active compounds, such as insecticides, baits, sterilizing agents, acaricides, nematicides, fungicides, growth-regulating substances or herbicides.

The insecticides include, for example, phosphates, carbamates, carboxylates, formamidines, tin compounds, substances produced by microorganisms and insect growth regulators, inter alia.

Preferred components for the mixture are

1. from the phosphorus compounds group
azinphos-ethyl, azinphos-methyl, 1-(4-chlorophenyl-4-(O-ethyl,S-propyl)phosphoryloxypyrazole (TIA 230), chlorpyrifos, coumaphos, demeton, demeton-S-methyl, diazinon, etrimfos, fenitrothion, fenthion, parathion-methyl, phosalone, pirimiphos-ethyl, pirimiphos-methyl, profenofos, prothiofos, sulprofos, triazophos and trichlorphon.

2. From the carbamate group
aldicarb, bendiocarb, BPMC (2-(1-methylpropyl)phenyl-methylcarbamate), butocarboxim, butoxicarboxim, carbaryl, carbofuran, carbosulfan, cloethocarb, isoprocarb, methomyl, oxamyl, pirimicarb, promecarb, propoxur and thiodicarb.

3. From the carboxylates group
allethrin, alphametrin, bioallethrin, bioresmethrin, cycloprothrin, cyfluthrin, cyhalothrin, cypermethrin, deltamethrin, α-cyano-3-phenyl-2-methylbenzyl 2,2-dimethyl-3-(2-chloro-2-trifluoromethylvinyl)-cyclopropane-

carboxylate (FMC 54800), fenpropathrin, fenfluthrin, fenvalerate, flucythrinate, flumethrin, fluvalinate, permethrin, resmethrin and tralomethrin.

4. From the amidine group
amitraz and chlordimeform;

5. From the tin compounds group
azocyclotin, cyhexatin and fenbutatin oxide

6. From the growth regulators group
fenoxycarb, flufenoxuron, buprofezin, cyromazine, flubenzimin, diflubenzuron, triflumuron, N-(((3,5-dichloro-4-(1,1,2,2-tetrafluoroethoxy)phenyl)aminocarbonyl)-2,6-difluorobenzamide (XRD 473), tetraflubenzuron, chlorfluazuron, flucylooxuron, hydroprene, methoprene, N-(2-fluorobenzoyl)-N'-(4-(2-chloro-4-trifluoromethyl-1-phenylmercapto)phenyl)urea, N-(2-chlorobenzoyl)-N'-(4-(2-chloro-4-trifluoromethyl-1-phenylmercapto)phenyl)urea, N-(2,6-difluorobenzoyl)-N'-(4-(2-chloro-4-trifluoromethyl-1-phenylmercapto)-phenyl)urea, ethyl N-(N-(2-chloro-4-trifluoromethyl-1-phenylmercapto)phenyl)carbamoyl)-2-fluorobenzocarboximidate, ethyl N-(N-(2-chloro-4-trifluoromethyl-1-phenylmercapto)phenyl)carbamoyl)-2-chlorobenzocarboximidate and ethyl N-(N-(2-chloro-4-trifluoromethyl-1-phenylmercapto)phenyl)carbamoyl)-2,6-difluorobenzocarboximidate.

7. Miscellaneous
α- and β-acermectins, Bacillus thuringiensis, bensultap, binapacryl, bisclofentezin, buprofezin, cartap, cyromacin, dicofol, endosulfan, ethoproxyfen, fenoxycarb, hexythiazox, 3- 2-(4-ethoxyphenyl)-2-methyl-propoxymethyl -1,3-diphenyl ether (MTI-500), 5- 4-(4-ethoxyphenyl-4-methylpentyl -2-fluoro-1, 3-diphenyl ether (MTI-800), 3-(2-chlorophenyl)-3-hydroxy-2-(2-phenyl-4-thiazolyl)-propenenitrile (SN 72129), thiocyclam, core polyhedral viruses and granulosis viruses.

The active compound content of the use forms prepared from the commercially available formulations can vary within wide ranges. The active compound concentration of the use forms can be from 0.0000001 up to 100% by weight of active compound, preferably between 0.00001 and 1% by weight.

Use takes place in a customary manner appropriate for the use forms.

The active compounds according to the invention are also suitable for combating ectoparasites and endoparasites, preferably ectoparasitic insects, from the veterinary medicinal sector or the animal husbandry sector.

The use of the active compounds according to the invention occurs in a known manner here, such as by dermal use in the form of, for example, dipping, spraying, pouring on and spotting on and pow-dering.

A. Formulation examples

a) A dusting agent is obtained by mixing 10 parts by weight of active compound and 90 parts by weight of talc as the inert substance and pulverizing in a hammer mill.

b) An easily water-dispersible, wettable powder is obtained by mixing 25 parts by weight of active compound, 65 parts by weight of kaolin-containing quartz as the inert substance, 10 parts by weight of potassium ligninsulfonate and 1 part by weight of sodium oleoylmethyltaurate as the wetting and dispersing agent and grinding the mixture in a pinned disk mill.

c) A dispersion concentrate which is easily dispersible in water is prepared by mixing 20 parts by weight of active compound with 6 parts by weight of alkylphenol polyglycol ether (Triton X 207), 3 parts by

weight of isotridecanol polyglycol ether (8 EO) and 71 parts by weight of paraffinic mineral oil (boiling range, for example, about 255 to above 377°C) and grinding the mixture in a ball mill to a fineness of under 5 microns.

d) An emulsifiable concentrate can be prepared from 15 parts by weight of active compound, 75 parts by weight of cyclohexanone as the solvent and 10 parts by weight of oxyethylated nonylphenol (10 EO) as the emulsifier.

e) Granules can be prepared from 2 to 15 parts by weight of active compound and an inert granule support material such as attapulgite, pumice granules and/or quartz sand.

B. Chemical examples

Example 1: 2-Methyl-2-(6-chloro-pyrid-3-yl)-propyl 3-phenoxy-benzyl ether (compound No. 18 Table 1)

1.1) 6-Chloropyridine-3-methanol

69.5 g (0.5 mol) of 6-hydroxynicotinic acid are introduced into a mixture of 200 g (1 mol) of phosphorus pentachloride and 233 g (1.5 mol) of phosphorus oxychloride at room temperature, the mixture is stirred at 95°C for 5 hours and excess phosphorus chlorides are then removed in vacuo. The crystalline residue is introduced into a solution of 75 g (1.98 mol) of sodium borohydride in 1,000 ml of water, the temperature being maintained at 30°C at most. The mixture is then stirred at room temperature until the completion of gas evolution and is filtered with suction from solid material, and the filtrate is extracted using ether.

After removing the solvent from the organic phase,

44.5 g (62% of theory) of oily crude product are obtained. The residue is distilled at 0.4 mbar (b.p. 110-115°C) to give 36.6 g (51% of theory).

1.2) 2-Chloro-5-chloromethyl-pyridine

85 ml of thionyl chloride are added cautiously to 14.4 g (0.1 mol) of 6-chloro-pyridine-3-methanol. 8.1 g (0.1 mol) of pyridine are then added dropwise and the mixture is heated under reflux until gas evolution is complete. Excess thionyl chloride is removed in vacuo and the residue is poured onto ice. The precipitate is filtered off with suction and dried. Yield 10.6 g (65.5% of theory), m.p.: 38-40°C. The crude product can be used further without purification.

1.3) 6-Chloro-pyridine-3-acetonitrile

25.6 g of sodium cyanide (0.522 mol) are dissolved in 123 g of dimethyl sulfoxide, the mixture is heated to 90°C and 68.9 g (0.43 mol) of 2-chloro-5-chloromethyl-pyridine are introduced in portions. During this, the temperature rises to 130°C. The mixture is allowed to cool to room temperature, poured into water and extracted using ether. The solvent is removed and the crude product is distilled at 1.0 mbar (130-135°C). M.p.: 80°C. Yield: 41.1 g (63% of theory).

1.4) 2-(6-Chloro-pyrid-3-yl)-2-methyl-propanenitrile

24 g (0.8 mol) of 80% strength sodium hydride (in white oil) are suspended in 640 ml of tetrahydrofuran and 56 g (0.37 mol) of 6-chloro-pyridine-3-acetonitrile, dissolved in 160 ml of tetrahydrofuran, are added dropwise. After completion of gas evolution, 105.3 g (0.74 mol) of methyl iodide, dissolved in 160 ml of tetrahydrofuran, are added dropwise at 0 - 20°C. The mixture is stirred at room temperature for a further 3 hours and then poured into 860 ml of water. The crude product is

extracted using ether and, after removing the solvent, is distilled at 0.3 mbar (b.p. 110-120°C). 49.1 g (73.5% of theory) of a yellowish oil are thus obtained, which crystallizes on standing. M.p.: 63-65°C.

1.5) 2-(6-Chloro-pyrid-3-yl)-2-methyl-propanol

3.6 g (20 mmol) of 2-(6-chloro-pyrid-3-yl)-2-methyl-propionitrile are introduced into 50 ml of toluene, and 25 ml of a diisobutylaluminum hydride solution (1 M in toluene) is added dropwise without cooling, by means of which the internal temperature rises to 45°C. The mixture is stirred for 2 hours at room temperature. 10 ml of methanol, 50 ml of water and 90 ml of 2 N sulfuric acid are then cautiously added successively, and the mixture is stirred for 2 hours at room temperature and finally for 2 hours at 50°C. The toluene phase is separated off and the solvent is removed. The residue is dissolved in 10 ml of methanol and a solution of 0.1 g of sodium and 0.4 g (10.6 mmol) of sodium borohydride in 10 ml of methanol are added. The mixture is stirred for 1 hour at room temperature and acidified with 0.7 g of acetic acid, and the solvent is removed. The residue is boiled with ethyl acetate, the ester phase is boiled off and the solvent is removed in vacuo. The oily crude product is distilled in a bulb tube; 0.15 mbar, apparatus temperature 170°C. Yield 2.5 g (67% of theory) of a colorless oil.

1.6) 2-(6-Chloro-pyrid-3-yl)-2-methyl-propyl 3-phenoxy-benzyl ether

1.9 g (10.2 mmol) of 2-(6-chloro-pyrid-3-yl)-2-methyl-propanol are dissolved in 15 ml of toluene, 2.7 g (10.3 mmol) of 3-phenoxy-benzyl bromide are added and a solution of 0.6 g (1.8 mmol) of tetra-n-butylammonium hydrogen sulfate in 5 g of 50% strength sodium hydroxide solution is added. The mixture is stirred at 70-80°C for 2 hours, mixed with a mixture of 2 ml of methanol and 2 ml of concentrated ammonia water and finally all distributed

between water and toluene. The organic phase is washed with ammonium chloride solution, the solvent is removed and the crude product is purified through a silica gel column. 2.4 g (62.9% of theory) of a weakly yellowish oil are thus obtained. The oil is distilled in the bulb tube at 0.03 mbar and 230-240°C apparatus temperature.

Example 2: 1-(6-Chloro-pyrid-3-yl)-cyclopropyl)-methyl 3-phenoxybenzyl ether (compound No. 513 in Table 2)

2.1) 1-(6-Chloro-pyrid-3-yl)-cyclopropane-1-carbonitrile (corresponds to Process 1.4 in Example 1).

6.5 g (0.217 mol) of 80% strength sodium hydride in white oil are suspended in 170 ml of tetrahydrofuran and a solution of 15.3 g (0.1 mol) of 6-chloro-pyridine-3-acetonitrile in 60 ml of tetrahydrofuran is added dropwise. The mixture is stirred until completion of gas evolution, cooled and a solution of 14.3 g (0.1 mol) of 1-bromo-2-chloro-ethane in 15 ml of tetrahydrofuran are added dropwise at 5-20°C. The mixture is stirred for 2 hours at room temperature, poured into concentrated sodium chloride solution and extracted using ether. The crude product, freed from solvent, is taken up in ethyl acetate and filtered through a bed of 200 g of silica gel. 14.3 g of a brownish, semi-crystalline material are thus obtained, which is purified by bulb tube distillation. 5.8 g (32.5% of theory) of an oil, which solidifies on cooling, are distilled at an apparatus temperature of 130-140°C and 0.08 mbar.

This substance can be converted into compound No. 513 (Table 2) in analogy to Processes 1.5 and 1.6 mentioned in Example 1.

Example 3: (2-(6-Ethoxy-pyridazin-3-yl)-2-methyl-propyl) 3-phenoxy-benzyl ether (compound No. 343 in Table 1)

3.1) 2-(6-Hydroxy-pyridazin-3-yl)-propanenitrile

60.4 g (2.02 mol) of 80% strength sodium hydride in white oil are suspended in 2 liters of tetrahydrofuran and 311.8 g (2.02 mol) of tert.-butyl 2-cyanopropionate are added dropwise at 20-30°C. During the course of 45 minutes this clear solution is added dropwise to a solution of 298 g (2.0 mol) of 3,6-dichloropyridazine and the mixture is heated under reflux for 2 hours. 317 ml of acetic acid are then added and the solvent is removed in vacuo. The residue is taken up in a mixture of 6,400 ml of acetic acid, 250 ml of acetic anhydride and 32 g of p-toluenesulfonic acid monohydrate, heated under reflux for 3 hours and again brought to dryness in vacuo. The residue is suspended in a little water, adjusted to pH 5 with sodium hydroxide solution and the water removed by evaporation. The residue is extracted hot using ethyl acetate. 99.7 g (33% of theory) of crude product are thus obtained, which can be employed for the following reaction without further processing.

3.2) 2-(6-Chloro-pyridazin-3-yl)-propanenitrile

1.5 g (10 mmol) of 2-(6-hydroxy-pyridazin-3-yl)-propanenitrile are mixed with 5 g of phosphorus oxychloride and the mixture is stirred at 80-90°C for 45 minutes. The mixture is poured onto 30 g of ice, adjusted to pH 6 using 2N NaOH and extracted using ether. After removal of the solvent, an oil remains which solidifies on standing. Yield: 1 g (59.7% of theory); m.p.: 65°C from hexane.

3.3) 2-(6-Chloro-pyridazin-3-yl)-2-methyl-propanenitrile

3.9 g (0.131 mol) of 80% strength sodium hydride in white

oil are suspended in 380 ml of tetrahydrofuran and a solution of 18.9 g (0.113 mol) of 2-(6-chloro-pyridazin-3-yl) propanenitrile in 80 ml of tetrahydrofuran is added at 15-20°C. After completion of gas evolution, a solution of 16.3 g (0.115 mol) of methyl iodide in 50 ml of tetrahydrofuran is added dropwise and the mixture is stirred for 1 hour at room temperature. The mixture is poured into 1.4 liters of concentrated sodium chloride solution and extracted with ether. The crude product which remains after removing the solvent is purified by bulb tube distillation.
Apparatus temperature: 180-190°C, 0.1 mbar.
Yield 8.4 g (41% of theory) oil which solidifies on standing.
M.p.: 107-111°C.

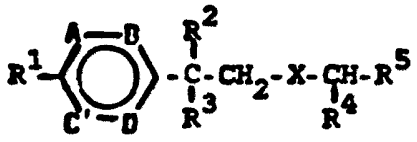
This nitrile can be converted into 2-(6-chloro-pyridazin-3-yl)-2-methyl-propanol in analogy to Process 1.5 mentioned in Example 1.

3.4) 2-(6-Ethoxy-pyridazin-3-yl)-2-methyl-propanol

1.6 g (0.053 mol) of 80% strength sodium hydride in white oil are suspended in 50 ml of dimethyl sulfoxide and 2.5 g (0.055 mol) of ethanol are added dropwise at 15-20°C. After completion of gas evolution, a solution of 1.0 g (0.005 mol) of 2-(6-chloro-pyridazin-3-yl)-2-methylpropanol in 20 ml of dimethyl sulfoxide is allowed to run in an the mixure is warmed to 90-100°C for 1.5 hours. After cooling, the mixture is poured into concentrated sodium chloride solution and extracted with ether. After removing the solvent, a yellow oil remains as the crude product. Yield: 0.6 g (61% of theory).

This crude product can be converted into compound No. 343 of Table 1 directly and in analogy to Process 1.6 mentioned in Example 1.

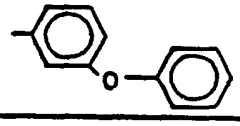
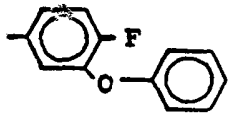
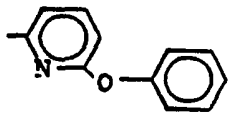
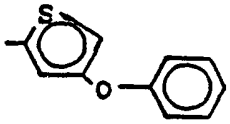
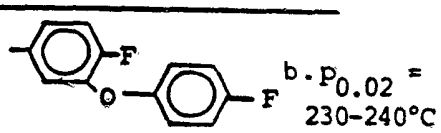
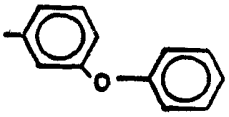
The preparation of the following compounds of the formula I



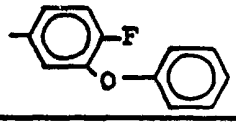
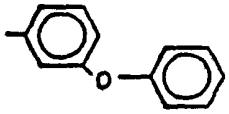
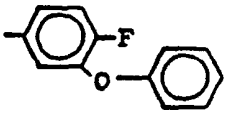
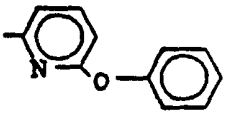
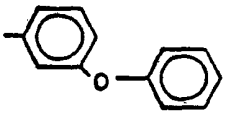
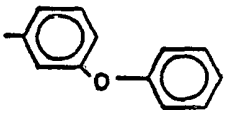
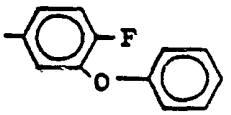
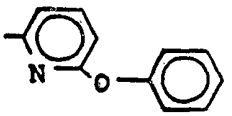
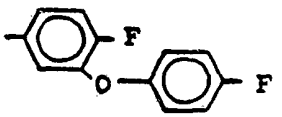
(I)

(Tables 1 and 2) having X = O takes place analogously to the abovementioned examples. Compounds having X = CH_2 are preferably obtained by Processes e) and f).

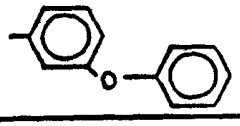
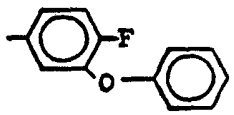
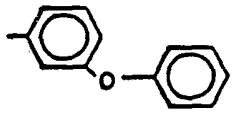
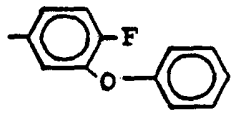
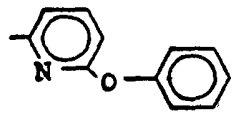
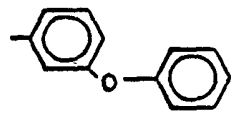
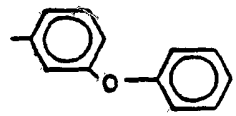
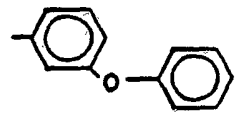
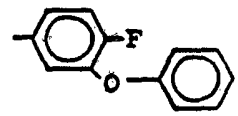
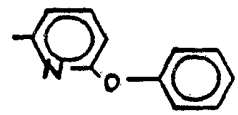
Table 1 R^2 and R^3 = CH_3, A = N, CH or C-R^1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
1	N	EtO	CH	CH	CH	CH_2	H		b.$P_{0.01}$ = 225-230°C
2	N	EtO	CH	CH	CH	CH_2	H		b.$P_{0.05}$ = 240-245°C
3	N	EtO	CH	CH	CH	CH_2	H		
4	N	EtO	CH	CH	CH	CH_2	H		
5	N	EtO	CH	CH	CH	CH_2	H		b.$P_{0.02}$ = 230-240°C
6	N	EtO	CH	CH	CH	CH_2	CN		

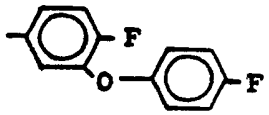
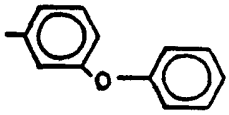
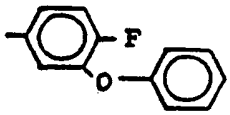
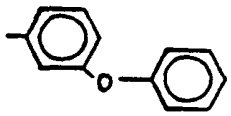
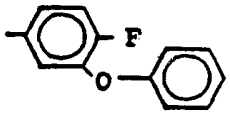
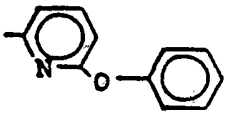
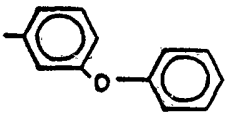
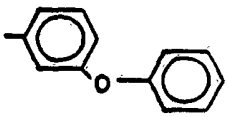
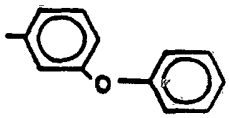
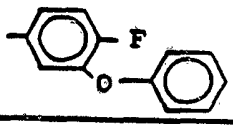
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
7	N	EtO	CH	CH	CH	CH_2	CN		
8	N	EtO	CH	CH	CH	O	H		$b.P_{0.005}$ = 210-220°C
9	N	EtO	CH	CH	CH	O	H		$b.P_{0.1}$ = 230-235°C
10	N	EtO	CH	CH	CH	O	H		
11	N	EtO	CH	CH	CH	O	CN		
12	N	Cl	CH	CH	CH	CH_2	H		
13	N	Cl	CH	CH	CH	CH_2	H		
14	N	Cl	CH	CH	CH	CH_2	H		
15	N	Cl	CH	CH	CH	CH_2	H		

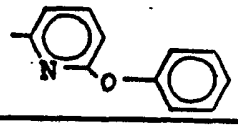
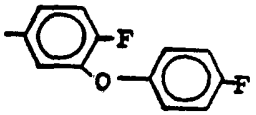
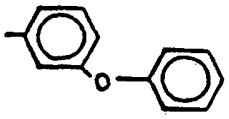
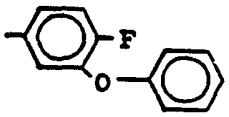
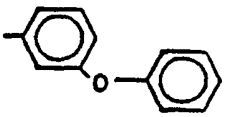
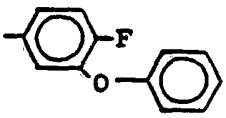
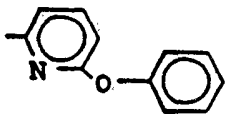
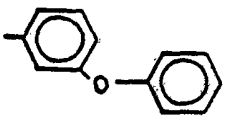
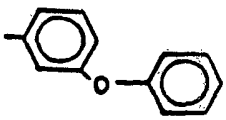
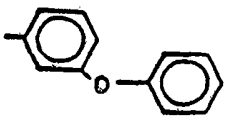
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
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17	N	Cl	CH	CH	CH	CH_2	CN		
18	N	Cl	CH	CH	CH	O	H		b.$P_{0.03}$ = 230–240°C
19	N	Cl	CH	CH	CH	O	H		b.$P_{0.01}$ = 225–230°C
20	N	Cl	CH	CH	CH	O	H		
21	N	Cl	CH	CH	CH	O	CN		
22	N	Cl	CH	CH	CH	O	CH_3		
23	N	H_3CO	CH	CH	CH	CH_2	H		
24	N	H_3CO	CH	CH	CH	CH_2	H		
25	N	H_3CO	CH	CH	CH	CH_2	H		

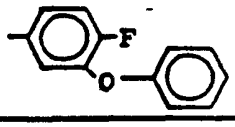
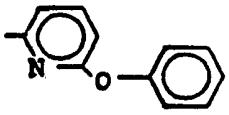
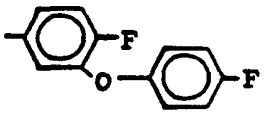
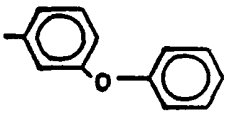
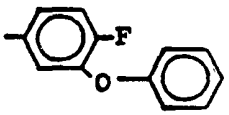
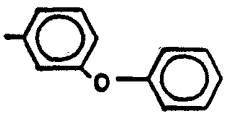
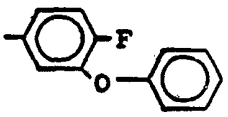
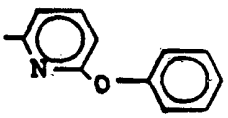
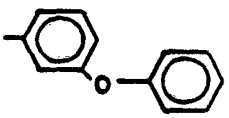
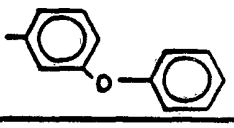
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
26	N	H_3CO	CH	CH	CH	CH_2	H		
27	N	H_3CO	CH	CH	CH	CH_2	CN		
28	N	H_3CO	CH	CH	CH	CH_2	CN		
29	N	H_3CO	CH	CH	CH	O	H		
30	N	H_3CO	CH	CH	CH	O	H		b.$p_{0.07}$ = 220-240°C
31	N	H_3CO	CH	CH	CH	O	H		
32	N	H_3CO	CH	CH	CH	O	CN		
33	N	H_3CO	CH	CH	CH	O	CH_3		
34	N	EtS	CH	CH	CH	CH_2	H		
35	N	EtS	CH	CH	CH	CH_2	H		

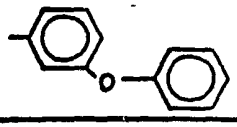
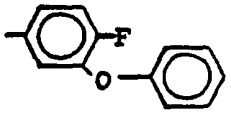
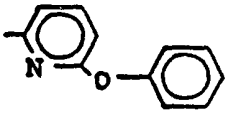
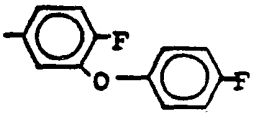
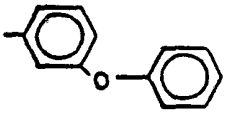
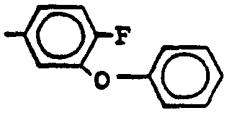
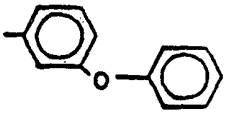
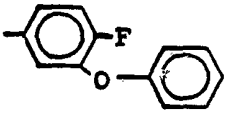
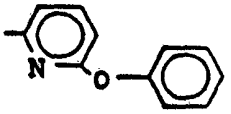
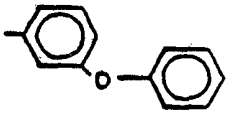
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
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37	N	EtS	CH	CH	CH	CH_2	H		
38	N	EtS	CH	CH	CH	CH_2	CN		
39	N	EtS	CH	CH	CH	CH_2	CN		
40	N	EtS	CH	CH	CH	O	H		
41	N	EtS	CH	CH	CH	O	H		b.$P_{0.01}$ = 220-230°C
42	N	EtS	CH	CH	CH	O	H		
43	N	EtS	CH	CH	CH	O	CN		
44	N	EtS	CH	CH	CH	O	CH_3		
45	N	H_3CS	CH	CH	CH	CH_2	H		

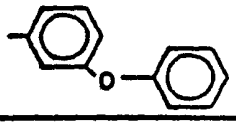
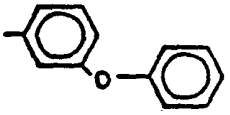
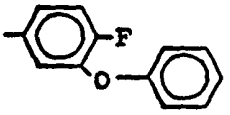
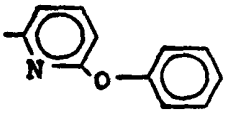
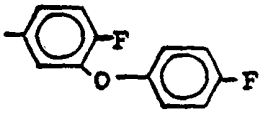
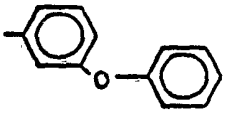
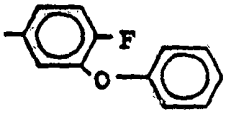
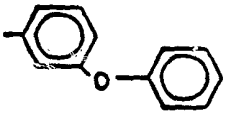
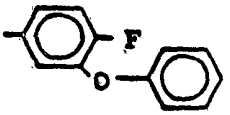
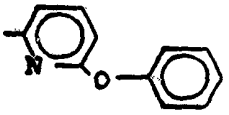
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
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47	N	H_3CS	CH	CH	CH	CH_2	H		
48	N	H_3CS	CH	CH	CH	CH_2	H		
49	N	H_3CS	CH	CH	CH	CH_2	CN		
50	N	H_3CS	CH	CH	CH	CH_2	CN		
51	N	H_3CS	CH	CH	CH	O	H		
52	N	H_3CS	CH	CH	CH	O	H		
53	N	H_3CS	CH	CH	CH	O	H		
54	N	H_3CS	CH	CH	CH	O	CN		
55	N	H_3CS	CH	CH	CH	O	CH_3		

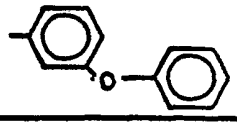
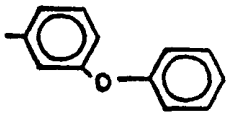
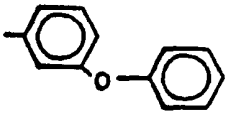
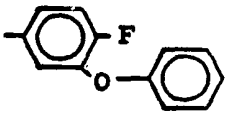
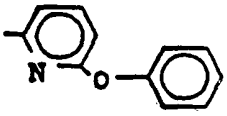
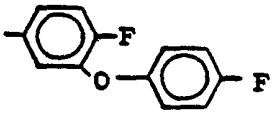
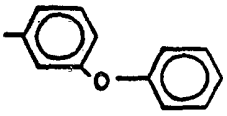
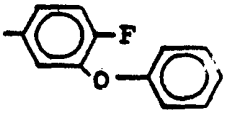
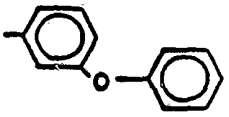
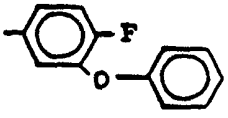
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
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57	CCl	Cl	CH	N	CH	CH_2	H		
58	CCl	Cl	CH	N	CH	CH_2	H		
59	CCl	Cl	CH	N	CH	CH_2	H		
60	CCl	Cl	CH	N	CH	CH_2	CN		
61	CCl	Cl	CH	N	CH	CH_2	CN		
62	CCl	Cl	CH	N	CH	O	H		
63	CCl	Cl	CH	N	CH	O	H		
64	CCl	Cl	CH	N	CH	O	H		
65	CCl	Cl	CH	N	CH	O	CN		

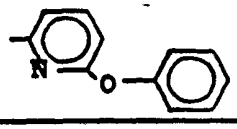
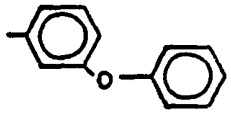
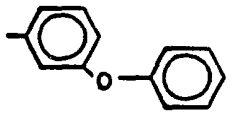
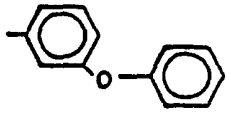
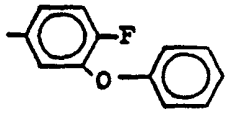
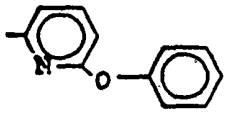
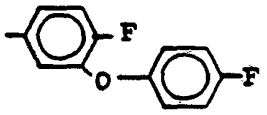
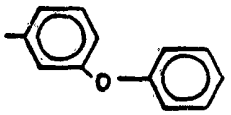
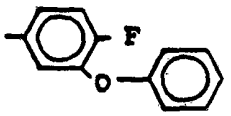
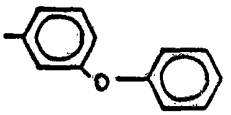
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
66	CCl	Cl	CH	N	CH	O	CH_3		
67	CCl	EtO	CH	N	CH	CH_2	H		
68	CCl	EtO	CH	N	CH	CH_2	H		
69	CCl	EtO	CH	N	CH	CH_2	H		
70	CCl	EtO	CH	N	CH	CH_2	H		
71	CCl	EtO	CH	N	CH	CH_2	CN		
72	CCl	EtO	CH	N	CH	CH_2	CN		
73	CCl	EtO	CH	N	CH	O	H		
74	CCl	EtO	CH	N	CH	O	H		
75	CCl	EtO	CH	N	CH	O	H		

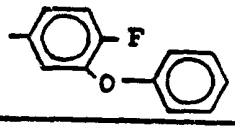
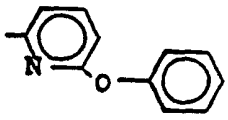
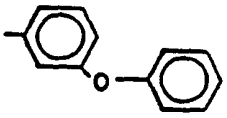
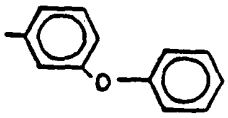
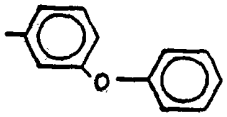
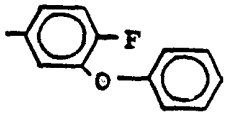
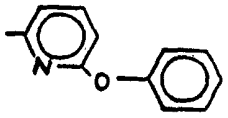
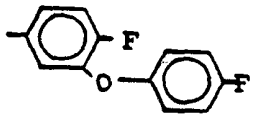
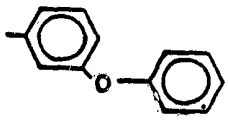
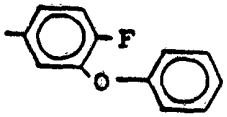
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
76	CCl	EtO	CH	N	CH	O	CN		
77	CCl	EtO	CH	N	CH	O	CH_3		
78	CF	EtO	CH	N	CH	CH_2	H		
79	CF	EtO	CH	N	CH	CH_2	H		
80	CF	EtO	CH	N	CH	CH_2	H		
81	CF	EtO	CH	N	CH	CH_2	H		
82	CF	EtO	CH	N	CH	CH_2	CN		
83	CF	EtO	CH	N	CH	CH_2	CN		
84	CF	EtO	CH	N	CH	O	H		
85	CF	EtO	CH	N	CH	O	H		

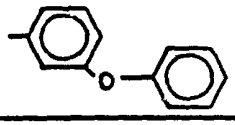
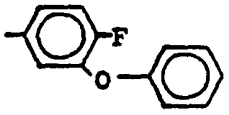
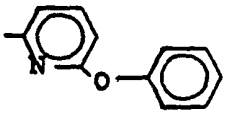
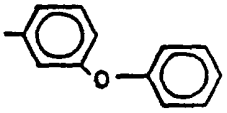
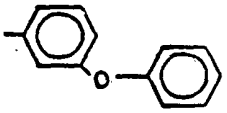
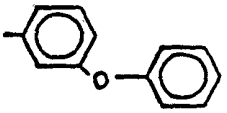
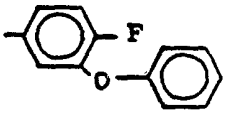
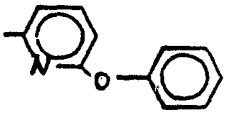
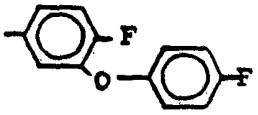
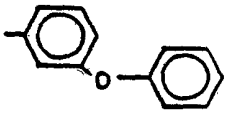
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
86	CF	EtO	CH	N	CH	O	H		
87	CF	EtO	CH	N	CH	O	CN		
88	CF	EtO	CH	N	CH	O	CH_3		
89	C-O-CH_2-O		CH	N	CH	CH_2	H		
90	C-O-CH_2-O		CH	N	CH	CH_2	H		
91	C-O-CH_2-O		CH	N	CH	CH_2	H		
92	C-O-CH_2-O		CH	N	CH	CH_2	H		
93	C-O-CH_2-O		CH	N	CH	CH_2	CN		
94	C-O-CH_2-O		CH	N	CH	CH_2	CN		
95	C-O-CH_2-O		CH	N	CH	O	H		

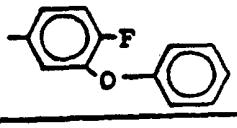
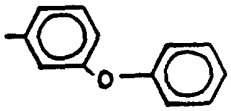
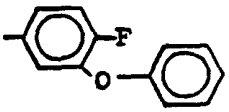
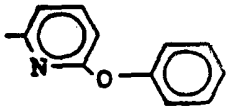
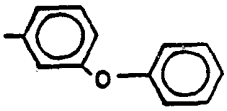
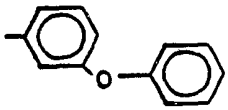
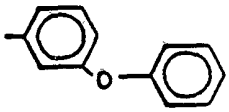
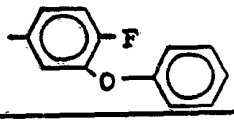
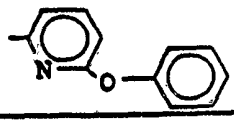
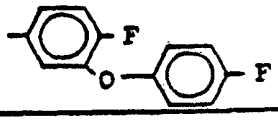
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
96	C-O-CH_2-O		CH	N	CH	O	H		
97	C-O-CH_2-O		CH	N	CH	O	H		
98	C-O-CH_2-O		CH	N	CH	O	CN		
99	C-O-CH_2-O		CH	N	CH	O	CH_3		
100	CH	EtO	N	CH	CH	CH_2	H		b.$P_{0.04}$ = 230-245°C
101	CH	EtO	N	CH	CH	CH_2	H		
102	CH	EtO	N	CH	CH	CH_2	H		
103	CH	EtO	N	CH	CH	CH_2	H		
104	CH	EtO	N	CH	CH	CH_2	CN		
105	CH	EtO	N	CH	CH	CH_2	CN		

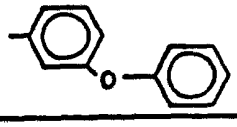
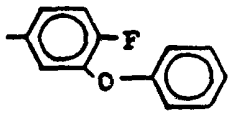
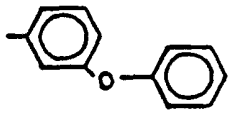
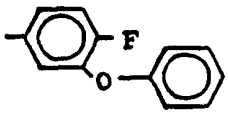
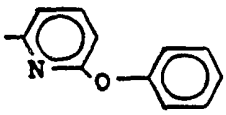
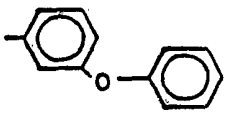
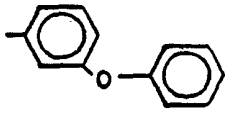
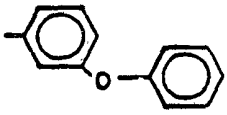
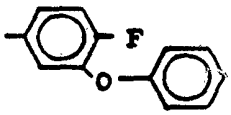
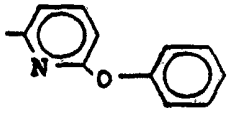
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
106	CH	EtO	N	CH	CH	O	H		
107	CH	EtO	N	CH	CH	O	H		
108	CH	EtO	N	CH	CH	O	H		
109	CH	EtO	N	CH	CH	O	CN		
110	CH	EtO	N	CH	CH	O	CH_3		
111	CH	Cl	N	CH	CH	CH_2	H		
112	CH	Cl	N	CH	CH	CH_2	H		
113	CH	Cl	N	CH	CH	CH_2	H		
114	CH	Cl	N	CH	CH	CH_2	H		
115	CH	Cl	N	CH	CH	CH_2	CN		

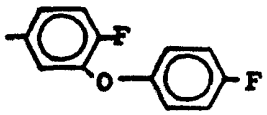
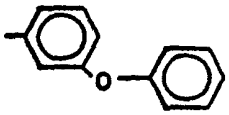
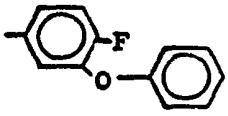
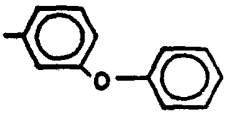
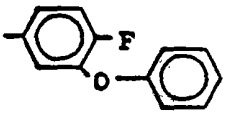
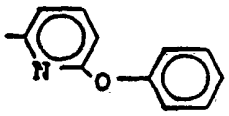
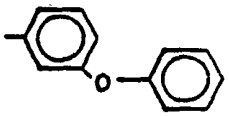
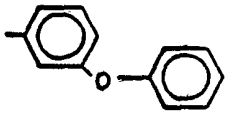
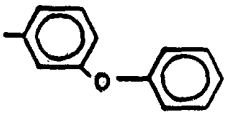
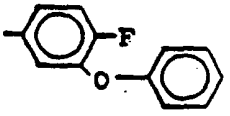
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
116	CH	Cl	N	CH	CH	CH_2	CN		
117	CH	Cl	N	CH	CH	O	H		
118	CH	Cl	N	CH	CH	O	H		
119	CH	Cl	N	CH	CH	O	H		
120	CH	Cl	N	CH	CH	O	CN		
121	CH	Cl	N	CH	CH	O	CH_3		
122	CH	H_3CO	N	CH	CH	CH_2	H		
123	CH	H_3CO	N	CH	CH	CH2	H		
124	CH	H_3CO	N	CH	CH	CH_2	H		
125	CH	H_3CO	N	CH	CH	CH_2	H		

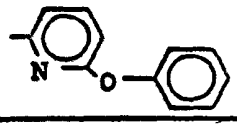
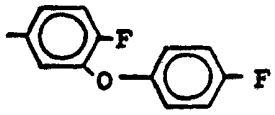
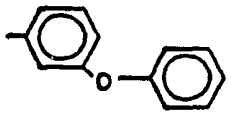
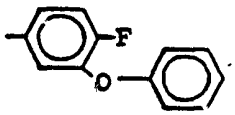
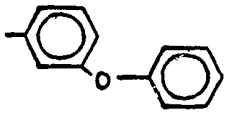
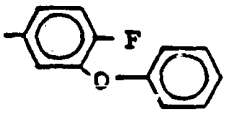
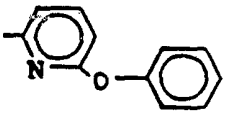
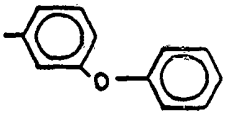
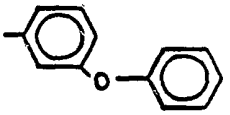
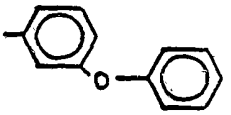
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
126	CH	H_3CO	N	CH	CH	CH_2	CN		
127	CH	H_3CO	N	CH	CH	CH_2	CN		
128	CH	H_3CO	N	CH	CH	O	H		
129	CH	H_3CO	N	CH	CH	O	H		
130	CH	H_3CO	N	CH	CH	O	H		
131	CH	H_3CO	N	CH	CH	O	CN		
132	CH	H_3CO	N	CH	CH	O	CH_3		
133	CH	EtS	N	CH	CH	CH_2	H		
134	CH	EtS	N	CH	CH	CH_2	H		
135	CH	EtS	N	CH	CH	CH_2	H		

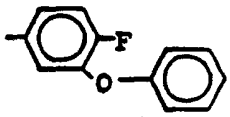
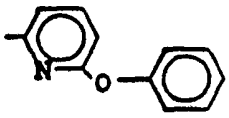
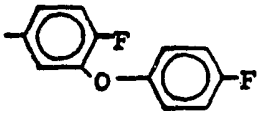
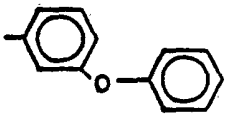
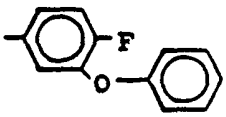
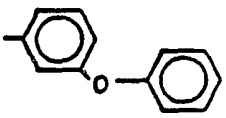
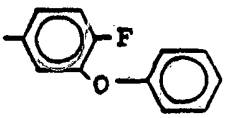
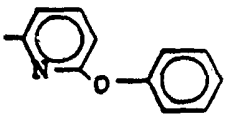
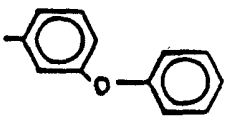
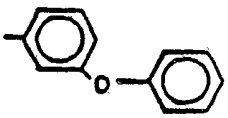
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
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137	CH	EtS	N	CH	CH	CH_2	CN		
138	CH	EtS	N	CH	CH	CH_2	CN		
139	CH	EtS	N	CH	CH	O	H		
140	CH	EtS	N	CH	CH	O	H		
141	CH	EtS	N	CH	CH	O	H		
142	CH	EtS	N	CH	CH	O	CN		
143	CH	EtS	N	CH	CH	O	CH_3		
144	CH	F_2CHO	N	CH	CH	CH_2	H		
145	CH	F_2CHO	N	CH	CH	CH_2	H		

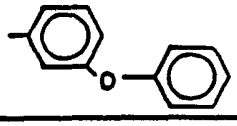
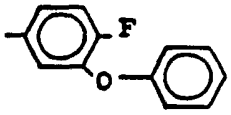
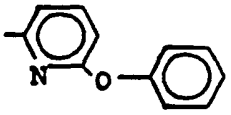
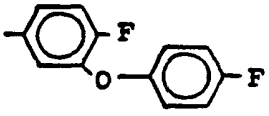
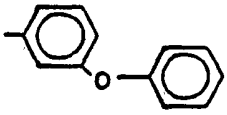
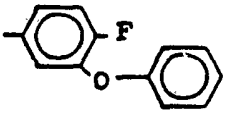
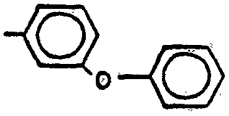
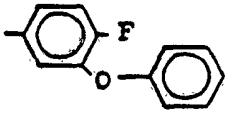
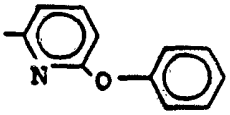
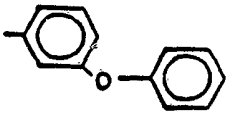
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
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147	CH	F_2CHO	N	CH	CH	CH_2	H		
148	CH	F_2CHO	N	CH	CH	CH_2	CN		
149	CH	F_2CHO	N	CH	CH	CH_2	CN		
150	CH	F_2CHO	N	CH	CH	O	H		
151	CH	F_2CHO	N	CH	CH	O	H		
152	CH	F_2CHO	N	CH	CH	O	H		
153	CH	F_2CHO	N	CH	CH	O	CN		
154	CH	F_2CHO	N	CH	CH	O	CH_3		
155	CCl	CL	N	CH	CH	CH_2	H		

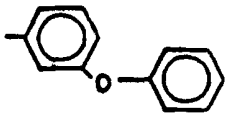
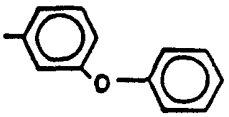
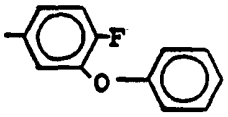
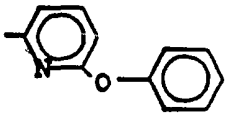
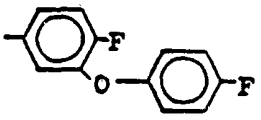
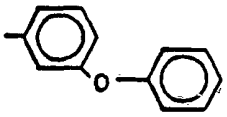
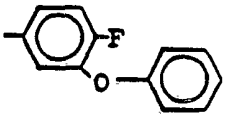
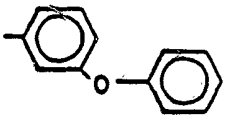
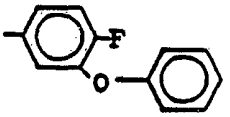
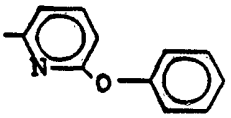
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
156	CCl	Cl	N	CH	CH	CH_2	H		
157	CCl	Cl	N	CH	CH	CH_2	H		
158	CCl	Cl	N	CH	CH	CH_2	H		
159	CCl	Cl	N	CH	CH	CH_2	CN		
160	CCl	Cl	N	CH	CH	CH_2	CN		
161	CCl	Cl	N	CH	CH	O	H		
162	CCl	Cl	N	CH	CH	O	H		
163	CCl	Cl	N	CH	CH	O	H		
164	CCl	Cl	N	CH	CH	O	CN		
165	CCl	Cl	N	CH	CH	O	CH_3		

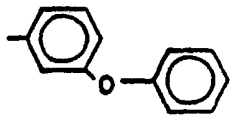
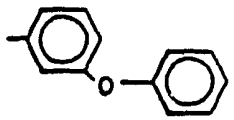
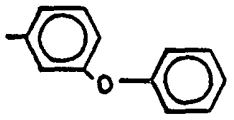
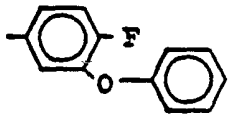
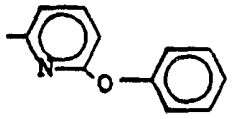
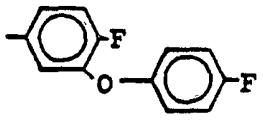
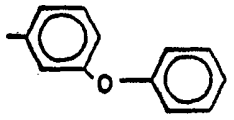
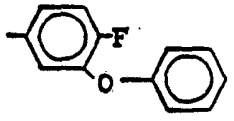
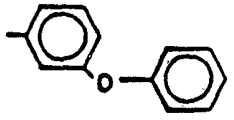
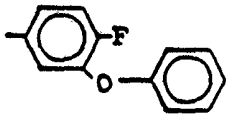
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
166	CCl	EtO	N	CH	CH	CH_2	H		
167	CCl	EtO	N	CH	CH	CH_2	H		
168	CCl	EtO	N	CH	CH	CH_2	H		
169	CCl	EtO	N	CH	CH	CH_2	H		
170	CCl	EtO	N	CH	CH	CH_2	CN		
171	CCl	EtO	N	CH	CH	CH_2	CN		
172	CCl	EtO	N	CH	CH	O	H		
173	CCl	EtO	N	CH	CH	O	H		
174	CCl	EtO	N	CH	CH	O	H		
175	CCl	EtO	N	CH	CH	O	CN		

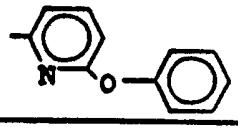
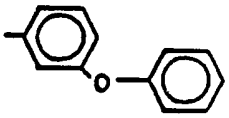
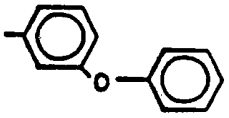
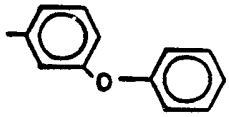
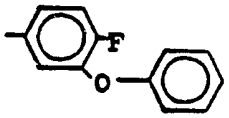
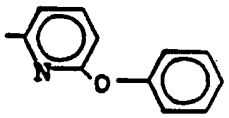
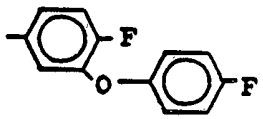
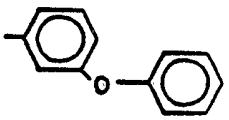
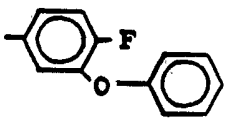
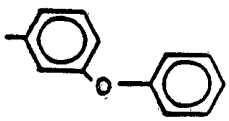
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
176	CCl	EtO	N	CH	CH	O	CH_3		
177	C-O-CH_2-O		N	CH	CH	CH_2	H		
178	C-O-CH_2-O		N	CH	CH	CH_2	H		
179	C-O-CH_2-O		N	CH	CH	CH_2	H		
180	C-O-CH_2-O		N	CH	CH	CH_2	H		
181	C-O-CH_2-O		N	CH	CH	CH_2	CN		
182	C-O-CH_2-O		N	CH	CH	CH_2	CN		
183	C-O-CH_2-O		N	CH	CH	O	H		
184	C-O-CH_2-O		N	CH	CH	O	H		
185	C-O-CH_2-O		N	CH	CH	O	H		

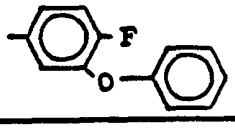
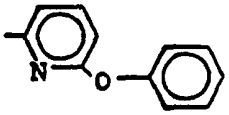
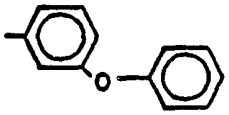
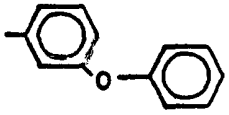
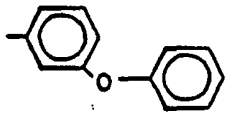
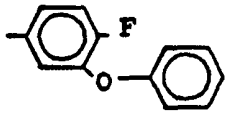
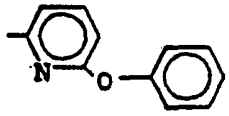
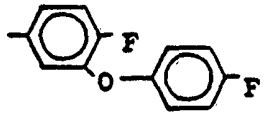
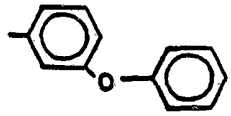
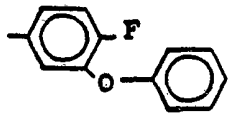
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
186	C-O-CH_2-O	N	CH	CH	O		CN		
187	C-O-CH_2-O	N	CH	CH	O		CH_3		
188	C-O-CH_2-O	CH	CH	N	CH_2		H		
189	C-O-CH_2-O	CH	CH	N	CH_2		H		
190	C-O-CH_2-O	CH	CH	N	CH_2		H		
191	C-O-CH_2-O	CH	CH	N	CH_2		H		
192	C-O-CH_2-O	CH	CH	N	CH_2		CN		
193	C-O-CH_2-O	CH	CH	N	CH_2		CN		
194	C-O-CH_2-O	CH	CH	N	O		H		
195	C-O-CH_2-O	CH	CH	N	O		H		

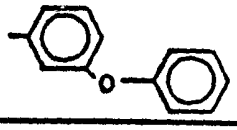
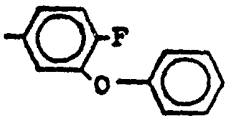
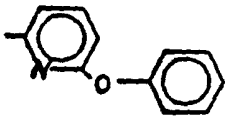
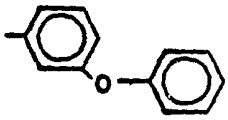
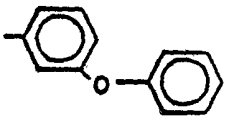
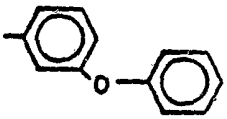
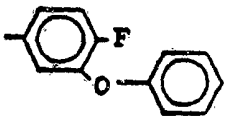
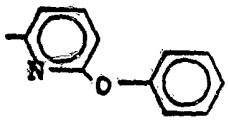
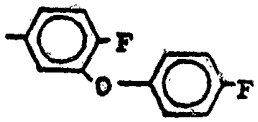
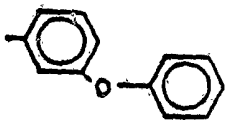
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
196	C-O-CH_2-O		CH	CH	N	O	H		
197	C-O-CH_2-O		CH	CH	N	O	CN		
198	C-O-CH_2-O		CH	CH	N	O	CH_3		
199	N	EtO	CH	N	CH	CH_2	H		b.$P_{0.005}$ = 230-245°C
200	N	EtO	CH	N	CH	CH_2	H		b.$P_{0.01}$ = 230-240°C
201	N	EtO	CH	N	CH	CH_2	H		
202	N	EtO	CH	N	CH	CH_2	H		
203	N	EtO	CH	N	CH	CH_2	CN		
204	N	EtO	CH	N	CH	CH_2	CN		
205	N	EtO	CH	N	CH	O	H		b.$P_{0.05}$ = 230-240°C

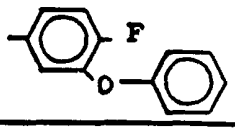
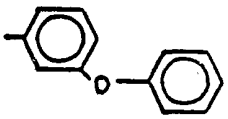
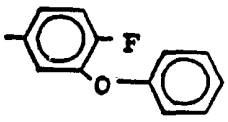
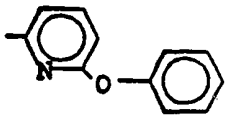
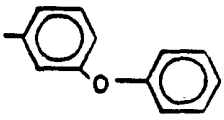
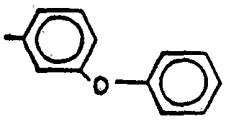
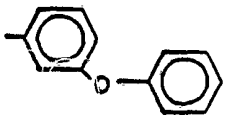
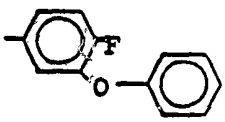
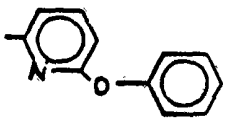
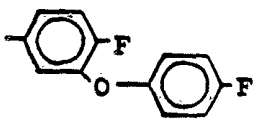
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
206	N	EtO	CH	N	CH	O	H		$b.p_{0.05}$ = 230-235°C
207	N	EtO	CH	N	CH	O	H		
208	N	EtO	CH	N	CH	O	CN		
209	N	EtO	CH	N	CH	O	CH_3		
210	N	Cl	CH	N	CH	CH_2	H		
211	N	Cl	CH	N	CH	CH_2	H		
212	N	Cl	CH	N	CH	CH_2	H		
213	N	Cl	CH	N	CH	CH_2	H		
214	N	Cl	CH	N	CH	CH_2	CN		
215	N	Cl	CH	N	CH	CH_2	CN		

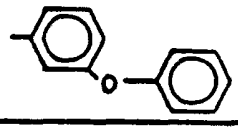
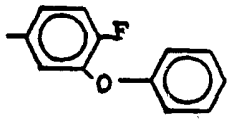
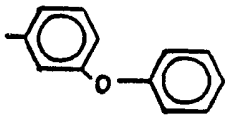
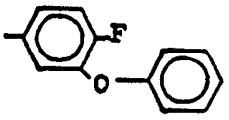
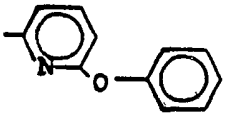
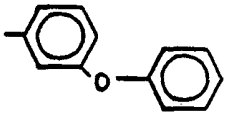
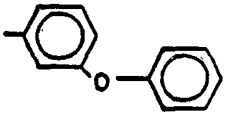
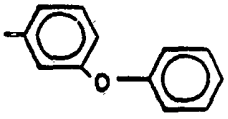
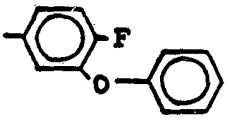
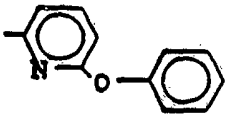
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
216	N	Cl	CH	N	CH	O	H		
217	N	Cl	CH	N	CH	O	H		
218	N	Cl	CH	N	CH	O	H		
219	N	Cl	CH	N	CH	O	CN		
220	N	Cl	CH	N	CH	O	CH_3		
221	N	H_3CO	CH	N	CH	CH_2	H		
222	N	H_3CO	CH	N	CH	CH_3	H		
223	N	H_3CO	CH	N	CH	CH_2	H		
224	N	H_3CO	CH	N	CH	CH_2	H		
225	N	H_3CO	CH	N	CH	CH_2	CN		

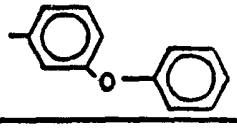
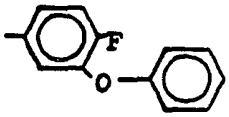
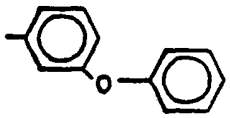
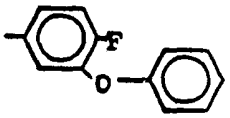
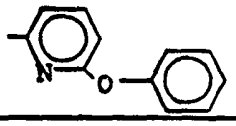
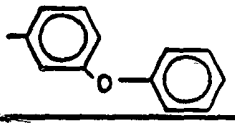
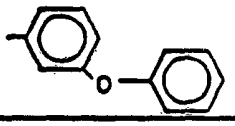
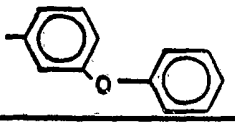
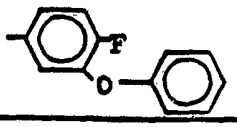
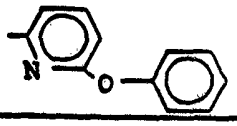
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
226	N	H_3CO	CH	N	CH	CH_2	CN		
227	N	H_3CO	CH	N	CH	O	H		
228	N	H_3CO	CH	N	CH	O	H		
229	N	H_3CO	CH	N	CH	O	H		
230	N	H_3CO	CH	N	CH	O	CN		
231	N	H_3CO	CH	N	CH	O	CH_3		
232	N	EtS	CH	N	CH	CH_2	H		pale yellow oil
233	N	EtS	CH	N	CH	CH_2	H		pale yellow oil
234	N	EtS	CH	N	CH	CH_2	H		
235	N	EtS	CH	N	CH	CH_2	H		

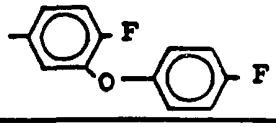
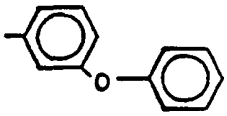
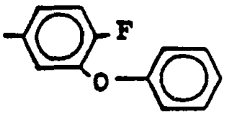
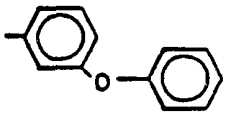
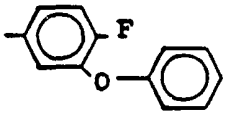
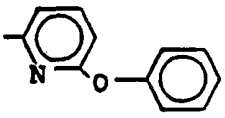
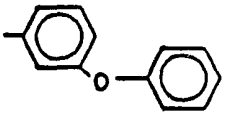
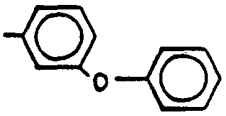
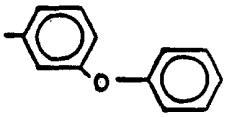
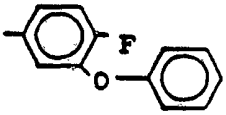
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
236	N	EtS	CH	N	CH	CH_2	CN		
237	N	EtS	CH	N	CH	CH_2	CN		
238	N	EtS	CH	N	CH	O	H		pale yellow oil
239	N	EtS	CH	N	CH	O	H		pale yellow oil
240	N	EtS	CH	N	CH	O	H		
241	N	EtS	CH	N	CH	O	CN		
242	N	EtS	CH	N	CH	O	CH_3		
243	N	n-C_3H_7	CH	N	CH	CH_2	H		
244	N	n-C_3H_7	CH	N	CH	CH_2	H		
245	N	n-C_3H_7	CH	N	CH	CH_2	H		

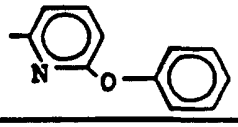
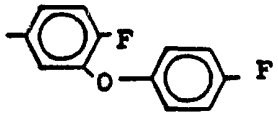
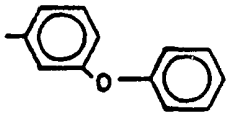
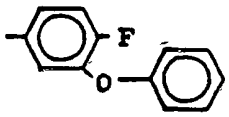
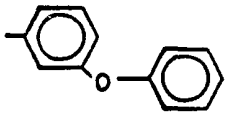
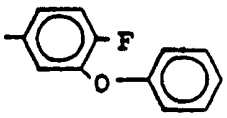
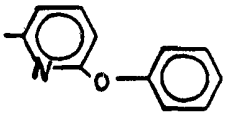
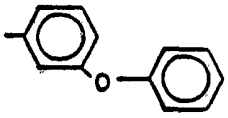
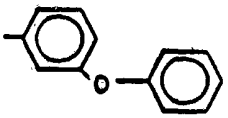
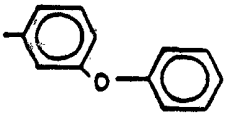
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
247	N	n-C_3H_7	CH	N	CH	CH_2	CN		
248	N	n-C_3H_7	CH	N	CH	CH_2	CN		
249	N	n-C_3H_7	CH	N	CH	O	H		
250	N	n-C_3H_7	CH	N	CH	O	H		
251	N	n-C_3H_7	CH	N	CH	O	H		
252	N	n-C_3H_7	CH	N	CH	O	CN		
253	N	n-C_3H_7	CH	N	CH	O	CH_3		
254	CH	EtO	N	CH	N	CH_2	H		b.$P_{0.05}$ = 225-245°C
255	CH	EtO	N	CH	N	CH_2	H		
256	CH	EtO	N	CH	N	CH_2	H		

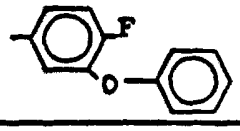
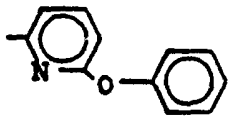
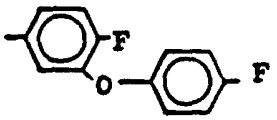
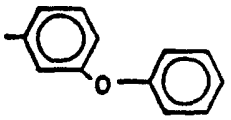
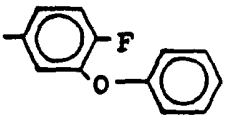
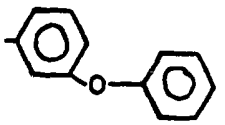
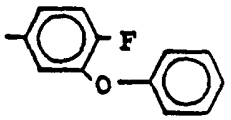
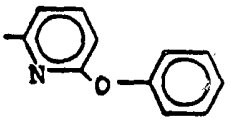
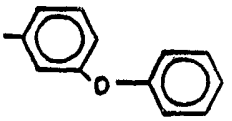
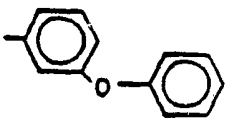
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
257	CH	EtO	N	CH	N	CH_2	H		
258	CH	EtO	N	CH	N	CH_2	CN		
259	CH	EtO	N	CH	N	CH_2	CN		
260	CH	EtO	N	CH	N	O	H		b.$P_{0.03}$ = 230°C
261	CH	EtO	N	CH	N	O	H		
262	CH	EtO	N	CH	N	O	H		
263	CH	EtO	N	CH	N	O	CN		
264	CH	EtO	N	CH	N	O	CH_3		
265	CH	Cl	N	CH	N	CH_2	H		
266	CH	Cl	N	CH	N	CH_2	H		

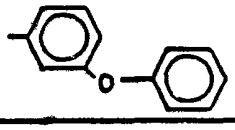
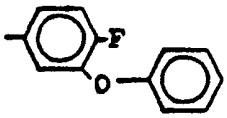
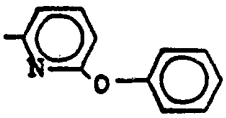
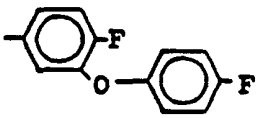
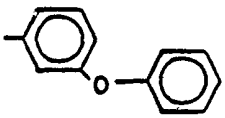
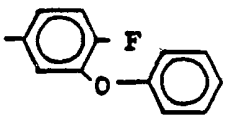
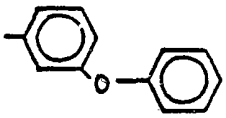
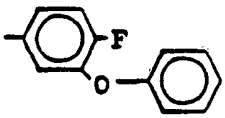
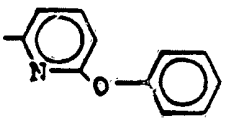
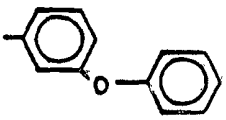
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
267	CH	Cl	N	CH	N	CH_2	H		
268	CH	Cl	N	CH	N	CH_2	H		
269	CH	Cl	N	CH	N	CH_2	CN		
270	CH	Cl	N	CH	N	CH_2	CN		
271	CH	Cl	N	CH	N	O	H		
272	CH	Cl	N	CH	N	O	H		
273	CH	Cl	N	CH	N	O	H		
274	CH	Cl	N	CH	N	O	CN		
275	CH	Cl	N	CH	N	O	CH_3		
276	CH	H_3CO	N	CH	N	CH_2	H		

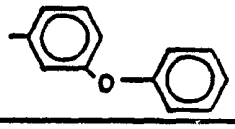
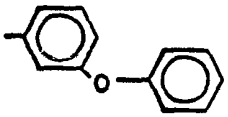
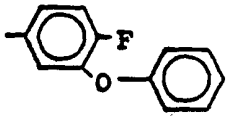
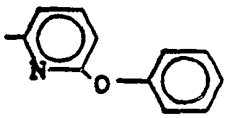
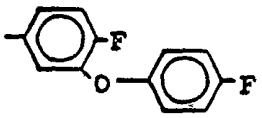
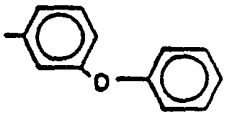
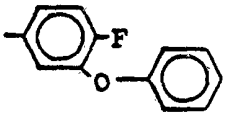
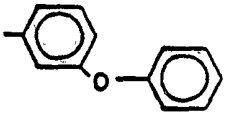
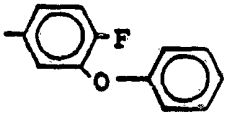
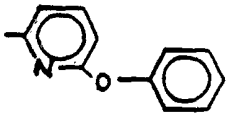
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
277	CH	H_3CO	N	CH	N	CH_2	H		
278	CH	H_3CO	N	CH	N	CH_2	H		
279	CH	H_3CO	N	CH	N	CH_2	H		
280	CH	H_3CO	N	CH	N	CH_2	CN		
281	CH	H_3CO	N	CH	N	CH_2	CN		
282	CH	H_3CO	N	CH	N	O	H		
283	CH	H_3CO	N	CH	N	O	H		
284	CH	H_3CO	N	CH	N	O	H		
285	CH	H_3CO	N	CH	N	O	CN		
286	CH	H_3CO	N	CH	N	O	CH_3		

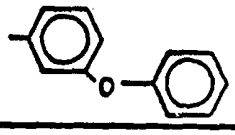
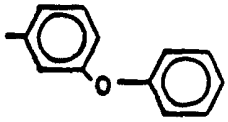
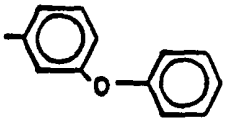
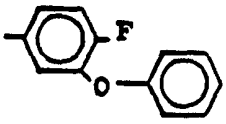
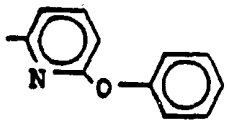
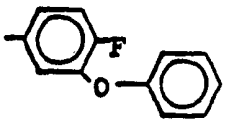
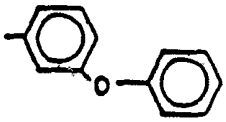
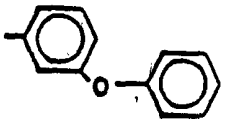
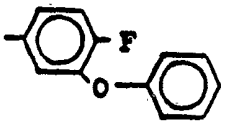
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
287	CH	F_2CHO	N	CH	N	CH_2	H		
288	CH	F_2CHO	N	CH	N	CH_2	H		
289	CH	F_2CHO	N	CH	N	CH_2	H		
290	CH	F_2CHO	N	CH	N	CH_2	H		
291	CH	F_2CHO	N	CH	N	CH_2	CN		
292	CH	F_2CHO	N	CH	N	CH_2	CN		
293	CH	F_2CHO	N	CH	N	O	H		
294	CH	F_2CHO	N	CH	N	O	H		
295	CH	F_2CHO	N	CH	N	O	H		
296	CH	F_2CHO	N	CH	N	O	CN		

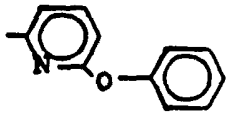
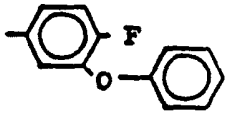
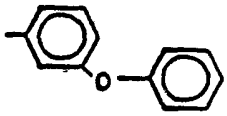
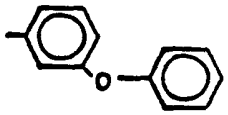
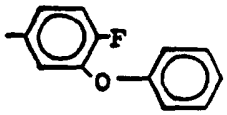
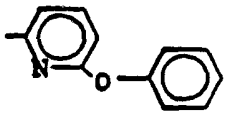
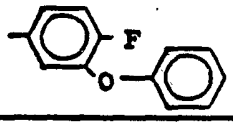
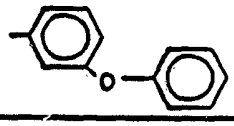
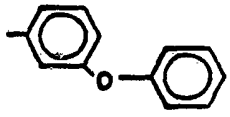
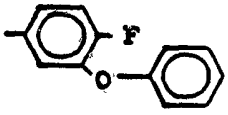
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
297	CH	F_2CHO	N	CH	N	O	CH_3		
298	CH	EtS	N	CH	N	CH_2	H		
299	CH	EtS	N	CH	N	CH_2	H		
300	CH	EtS	N	CH	N	CH_2	H		
301	CH	EtS	N	CH	N	CH_2	H		
302	CH	EtS	N	CH	N	CH_2	CN		
303	CH	EtS	N	CH	N	CH_2	CN		
304	CH	EtS	N	CH	N	O	H		
305	CH	EtS	N	CH	N	O	H		
306	CH	EtS	N	CH	N	O	H		

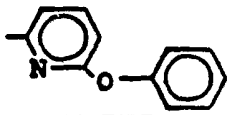
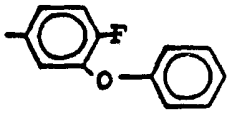
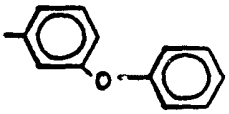
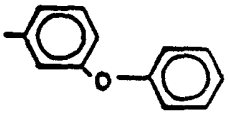
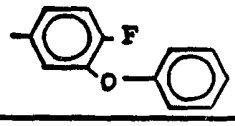
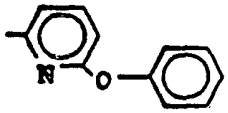
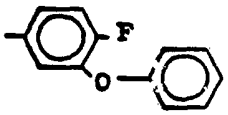
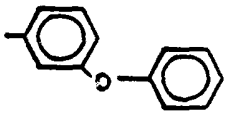
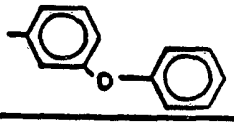
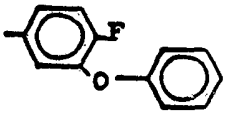
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
307	CH	EtS	N	CH	N	O	CN		
308	CH	EtS	N	CH	N	O	CH_3		
309	N	EtO	CH	CH	N	CH_2	H		
310	N	EtO	CH	CH	N	CH_2	H		
311	N	EtO	CH	CH	N	CH_2	H		
312	N	EtO	CH	CH	N	O	H		pale yellow oil
313	N	EtO	CH	CH	N	O	H		pale yellow oil
314	N	Cl	CH	CH	N	CH_2	H		
315	N	Cl	CH	CH	N	CH_2	H		

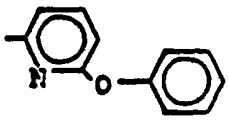
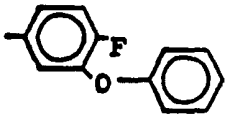
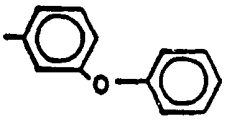
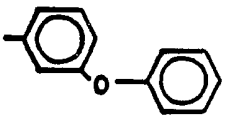
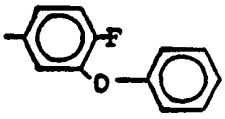
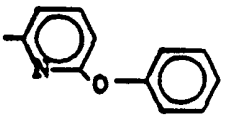
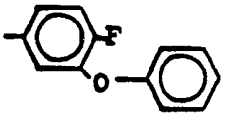
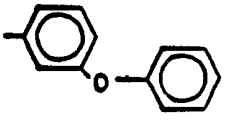
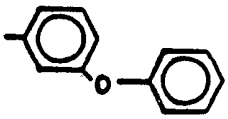
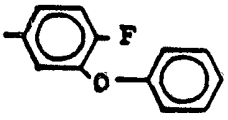
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
316	N	Cl	CH	CH	N	CH_2	H		
317	N	Cl	CH	CH	N	O	H		
318	N	Cl	CH	CH	N	O	H		
319	N	CH_3O	CH	CH	N	CH_2	H		
320	N	CH_3O	CH	CH	N	CH_2	H		
321	N	CH_3O	CH	CH	N	CH_2	H		
322	N	CH_3O	CH	CH	N	O	H		
323	N	CH_3O	CH	CH	N	O	H		
324	N	EtS	CH	CH	N	CH_2	H		
325	N	EtS	CH	CH	N	CH_2	H		

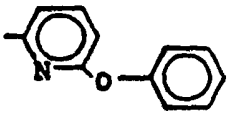
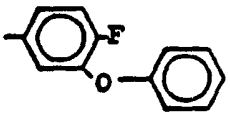
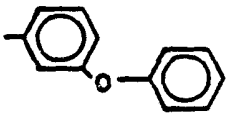
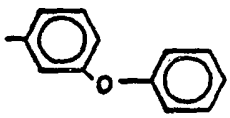
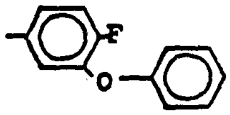
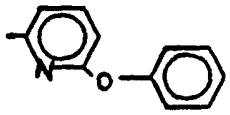
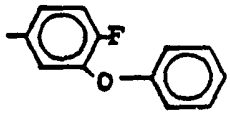
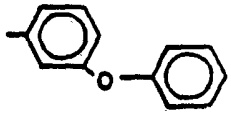
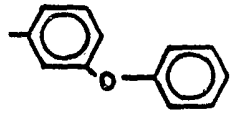
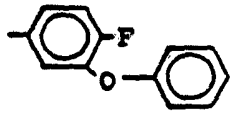
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
326	N	EtS	CH	CH	N	CH_2	H		
327	N	EtS	CH	CH	N	O	H		
328	N	EtS	CH	CH	N	O	H		
329	CCl	EtO	N	N	CH	CH_2	H		
330	CCl	EtO	N	N	CH	CH_2	H		
331	CCl	EtO	N	N	CH	CH_2	H		
332	CCl	EtO	N	N	CH	O	H		
333	CCl	EtO	N	N	CH	O	H		
334	CCl	Cl	N	N	CH	CH_2	H		
335	CCl	Cl	N	N	CH	CH_2	H		

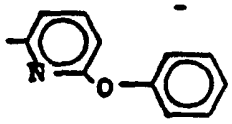
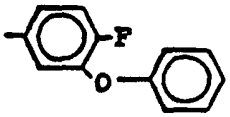
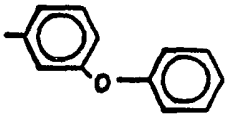
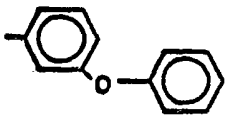
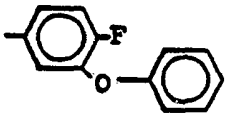
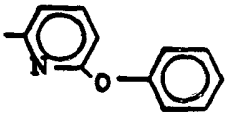
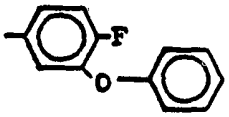
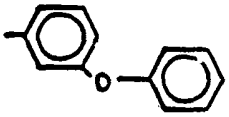
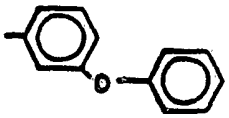
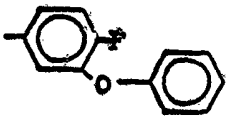
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
336	CCl	Cl	N	N	CH	CH_2	H		
337	CCl	Cl	N	N	CH	O	H		
338	CCl	Cl	N	N	CH	O	H		
339	CH	EtO	CH	N	N	CH_2	H		
340	CH	EtO	CH	N	N	CH_2	H		
341	CH	EtO	CH	N	N	CH_2	H		
342	CH	EtO	CH	N	N	O	H		pale yellow oil
343	CH	EtO	CH	N	N	O	H		pale yellow oil
344	CH	CH_3O	CH	N	N	CH_2	H		
345	CH	CH_3O	CH	N	N	CH_2	H		

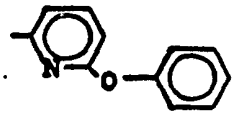
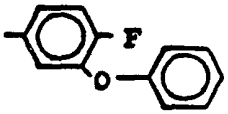
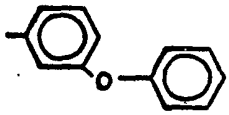
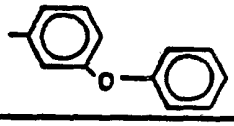
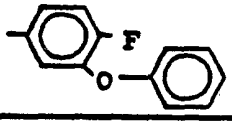
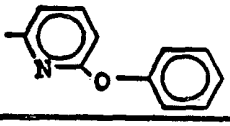
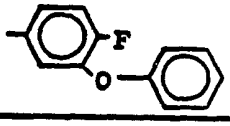
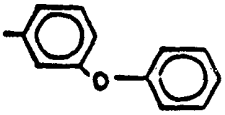
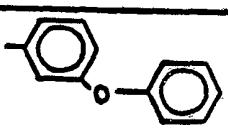
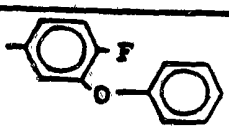
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
346	CH	CH_3O	CH	N	N	CH_2	H		
347	CH	CH_3O	CH	N	N	O	H		
348	CH	CH_3O	CH	N	N	O	H		
349	CH	Cl	CH	N	N	CH_2	H		
350	CH	Cl	CH	N	N	CH_2	H		
351	CH	Cl	CH	N	N	CH_2	H		
352	CH	Cl	CH	N	N	O	H		
353	CH	Cl	CH	N	N	O	H		
354	CH	EtS	CH	N	N	CH_2	H		
355	CH	EtS	CH	N	N	CH_2	H		

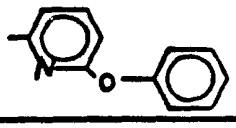
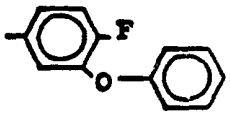
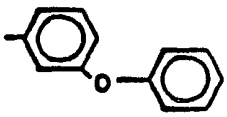
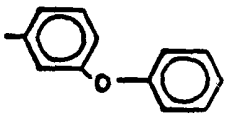
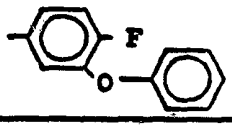
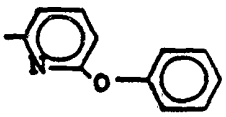
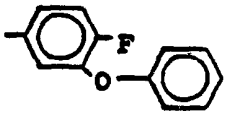
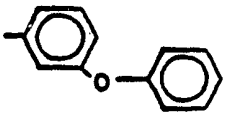
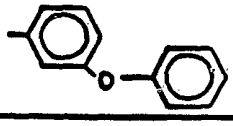
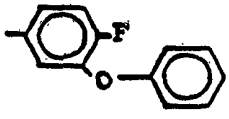
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
356	CH	EtS	CH	N	N	CH_2	H		
357	CH	EtS	CH	N	N	O	H		pale yellow oil
358	CH	EtS	CH	N	N	O	H		pale yellow oil
359	CCl	Cl	CH	N	N	CH_2	H		
360	CCl	Cl	CH	N	N	CH_2	H		
361	CCl	Cl	CH	N	N	CH_2	H		
362	CCl	Cl	CH	N	N	O	H		
363	CCl	Cl	CH	N	N	O	H		
364	CH	EtO	N	N	N	CH_2	H		
365	CH	EtO	N	N	N	CH_2	H		

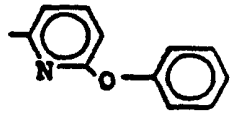
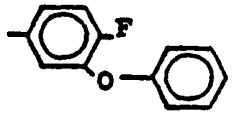
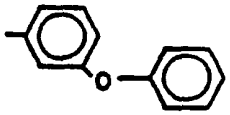
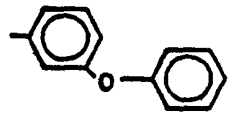
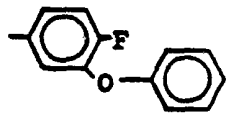
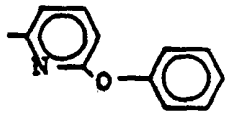
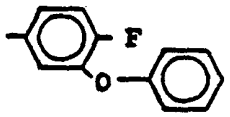
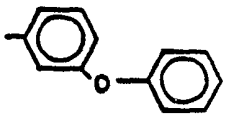
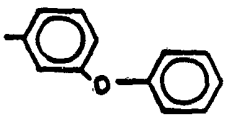
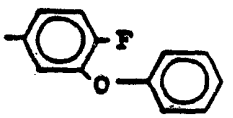
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
366	CH	EtO	N	N	N	CH_2	H		
367	CH	EtO	N	N	N	O	H		
368	CH	EtO	N	N	N	O	H		
369	CH	CH_3O	N	N	N	CH_2	H		
370	CH	CH_3O	N	N	N	CH_2	H		
371	CH	CH_3O	N	N	N	CH_2	H		
372	CH	CH_3O	N	N	N	O	H		
373	CH	CH_3O	N	N	N	O	H		
374	CH	Cl	N	N	N	CH_2	H		
375	CH	Cl	N	N	N	CH_2	H		

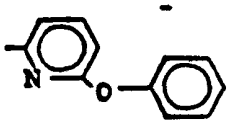
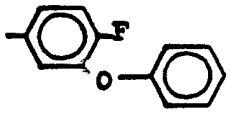
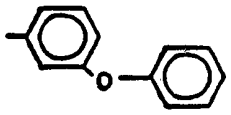
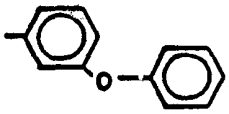
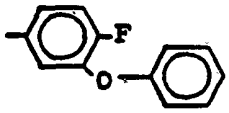
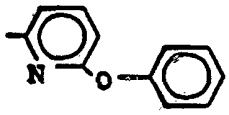
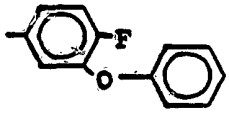
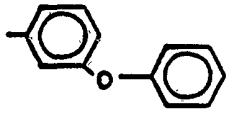
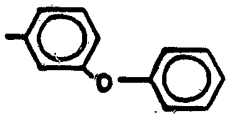
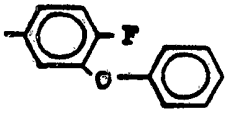
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
376	CH	Cl	N	N	N	CH_2	H		
377	CH	Cl	N	N	N	O	H		
378	CH	Cl	N	N	N	O	H		
379	CH	EtS	N	N	N	CH_2	H		
380	CH	EtS	N	N	N	CH_2	H		
381	CH	EtS	N	N	N	CH_2	H		
382	CH	EtS	N	N	N	O	H		
383	CH	EtS	N	N	N	O	H		
384	N	EtS	N	N	N	CH_2	H		
385	N	EtO	N	N	CH	CH_2	H		

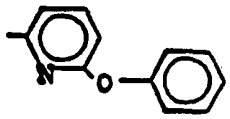
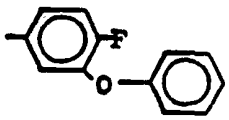
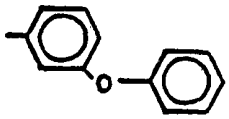
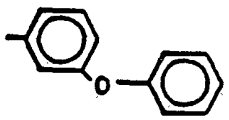
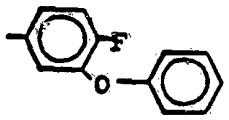
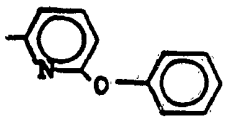
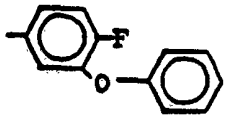
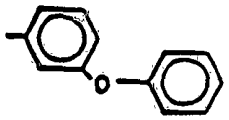
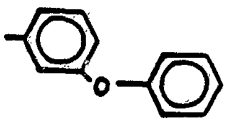
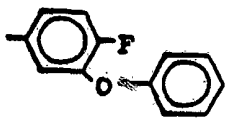
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
386	N	EtO	N	N	CH	CH_2	H		
387	N	EtO	N	N	N	O	H		
388	N	EtO	N	N	CH	O	H		
389	N	CH_3O	N	N	CH	CH_2	H		
390	N	CH_3O	N	N	CH	CH_2	H		
391	N	CH_3O	N	N	CH	CH_2	H		
392	N	CH_3O	N	N	CH	O	H		
393	N	CH_3O	N	N	CH	O	H		
394	N	Cl	N	N	CH	CH_2	H		
395	N	Cl	N	N	CH	CH_2	H		

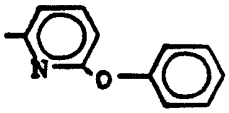
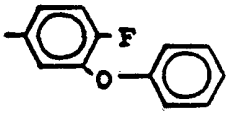
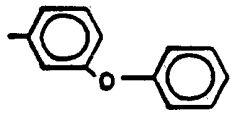
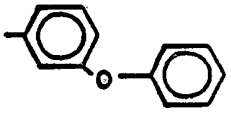
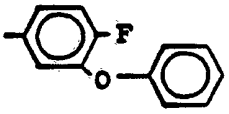
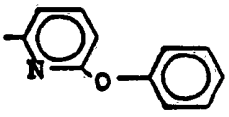
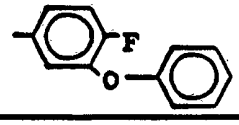
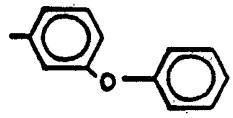
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
396	N	Cl	N	N	CH	CH_2	H		
397	N	Cl	N	N	CH	O	H		
398	N	Cl	N	N	CH	O	H		
399	N	EtS	N	N	CH	CH_2	H		
400	N	EtS	N	N	CH	CH_2	H		
401	N	EtS	N	N	CH	CH_2	H		
402	N	EtS	N	N	CH	O	H		
403	N	EtS	N	N	CH	O	H		
404	N	EtO	N	N	N	CH_2	H		
405	N	EtO	N	N	N	CH_2	H		

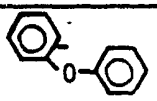
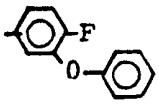
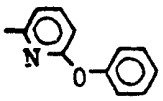
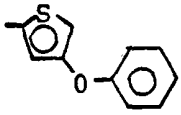
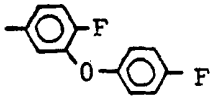
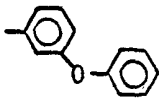
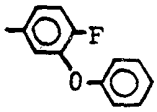
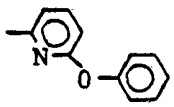
Continuation of Table 1

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
406	N	EtO	N	N	N	CH_2	H		
407	N	EtO	N	N	N	O	H		
408	N	EtO	N	N	N	O	H		
409	N	CH_3O	N	N	N	CH_2	H		
410	N	CH_3O	N	N	N	CH_2	H		
411	N	CH_3O	N	N	N	CH_2	H		
412	N	CH_3O	N	N	N	O	H		
413	N	CH_3O	N	N	N	O	H		
414	N	Cl	N	N	N	CH_2	H		
415	N	Cl	N	N	N	CH_2	H		

Continuation of Table 1

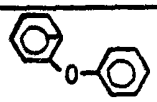
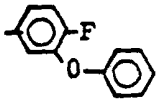
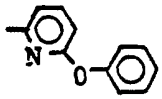
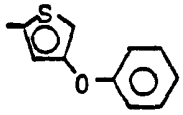
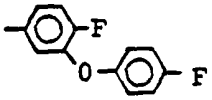
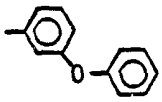
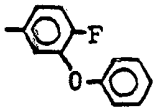
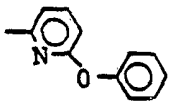
Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
416	N	Cl	N	N	N	CH_2	H		
417	N	Cl	N	N	N	O	H		
418	N	Cl	N	N	N	O	H		
419	N	EtS	N	N	N	CH_2	H		
420	N	EtS	N	N	N	CH_2	H		
421	N	EtS	N	N	N	CH_2	H		
422	N	EtS	N	N	N	O	H		
423	N	EtS	N	N	N	O	H		

Continuation of Table 1

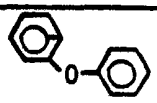
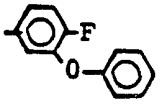
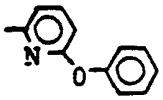
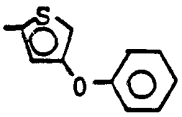
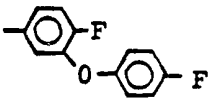
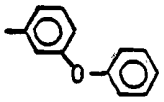
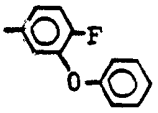
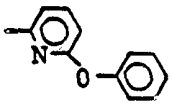
Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
424	N	F_3C-CH_2-O-	CH	CH	CH	CH_2	H		b.p 0.01 =225-235°C
425	N	F_3C-CH_2-O-	CH	CH	CH	CH_2	H		b.p 0.01 = 230-235°C
426	N	F_3C-CH_2-O-	CH	CH	CH	CH_2	H		b.p. 0.001 = 225-230°C
427	N	F_3C-CH_2-O-	CH	CH	CH	CH_2	H		b.p. 0.001 = 220-225°C
428	N	F_3C-CH_2-O-	CH	CH	CH	CH_2	H		
429	N	F_3C-CH_2-O-	CH	CH	CH	O	H		b.p 0.02 =230-240°C
430	N	F_3C-CH_2-O-	CH	CH	CH	O	H		b.p 0.04 =230-240°C
431	N	F_3C-CH_2-O-	CH	CH	CH	O	H		

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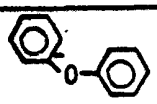
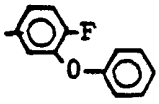
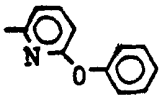
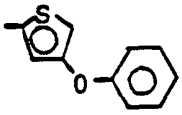
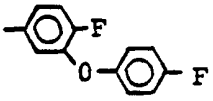
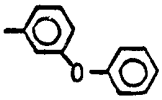
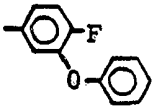
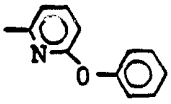
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
432	CH	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
433	CH	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
434	CH	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
435	CH	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
436	CH	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
437	CH	F_3C-CH_2-O-	N	CH	CH	O	H		
438	CH	F_3C-CH_2-O-	N	CH	CH	O	H		
439	CH	F_3C-CH_2-O-	N	CH	CH	O	H		

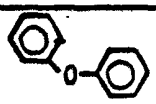
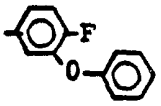
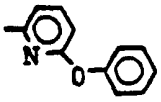
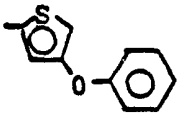
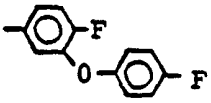
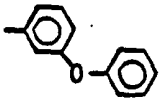
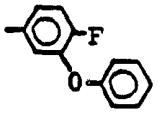
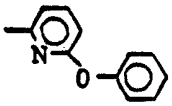
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
440	N	F_3C-CH_2-O-	CH	N	CH	CH_2	H		b.p 0.01 =240-245°C
441	N	F_3C-CH_2-O-	CH	N	CH	CH_2	H		b.p 0.01 =240-245°C
442	N	F_3C-CH_2-O-	CH	N	CH	CH_2	H		
443	N	F_3C-CH_2-O-	CH	N	CH	CH_2	H		
444	N	F_3C-CH_2-O-	CH	N	CH	CH_2	H		
445	N	F_3C-CH_2-O-	CH	N	CH	O	H		pale yellow oil
446	N	F_3C-CH_2-O-	CH	N	CH	O	H		pale yellow oil
447	N	F_3C-CH_2-O-	CH	N	CH	O	H		

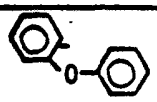
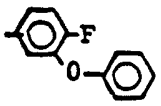
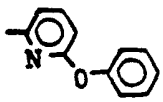
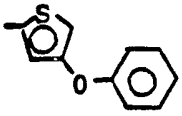
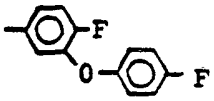
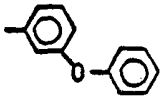
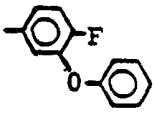
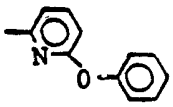
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
448	CH	F_3C-CH_2-O-	N	CH	N	CH_2	H		
449	CH	F_3C-CH_2-O-	N	CH	N	CH_2	H		
450	CH	F_3C-CH_2-O-	N	CH	N	CH_2	H		
451	CH	F_3C-CH_2-O-	N	CH	N	CH_2	H		
452	CH	F_3C-CH_2-O-	N	CH	N	CH_2	H		
453	CH	F_3C-CH_2-O-	N	CH	N	O	H		
454	CH	F_3C-CH_2-O-	N	CH	N	O	H		
455	CH	F_3C-CH_2-O-	N	CH	N	O	H		

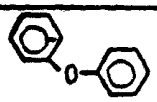
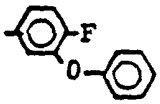
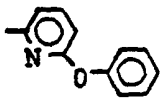
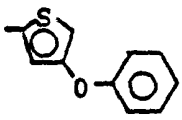
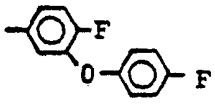
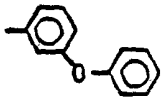
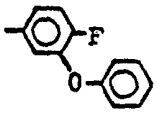
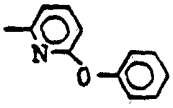
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
456	N	F_3C-CH_2-O-	N	CH	CH	CH_2	H		pale yellow oil
457	N	F_3C-CH_2-O-	N	CH	CH	CH_2	H		pale yellow oil
458	N	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
459	N	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
460	N	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
461	N	F_3C-CH_2-O-	N	CH	CH	O	H		pale yellow oil
462	N	F_3C-CH_2-O-	N	CH	CH	O	H		pale yellow oil
463	N	F_3C-CH_2-O-	N	CH	CH	O	H		

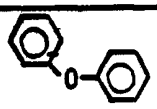
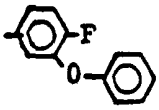
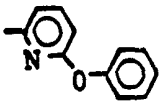
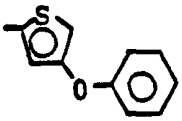
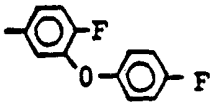
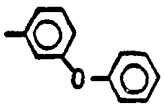
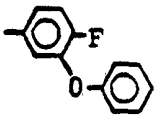
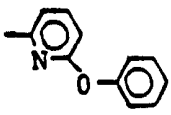
Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
464	N	F_3C-CH_2-O-	CH	CH	N	CH_2	H		pale yellow oil
465	N	F_3C-CH_2-O-	CH	CH	N	CH_2	H		pale yellow oil
466	N	F_3C-CH_2-O-	CH	CH	N	CH_2	H		
467	N	F_3C-CH_2-O-	CH	CH	N	CH_2	H		
468	N	F_3C-CH_2-O-	CH	CH	N	CH_2	H		
469	N	F_3C-CH_2-O-	CH	CH	N	O	H		pale yellow oil
470	N	F_3C-CH_2-O-	CH	CH	N	O	H		pale yellow oil
471	N	F_3C-CH_2-O-	CH	CH	N	O	H		

Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
472	N	F_3C-CH_2-O-	N	N	CH	CH_2	H		
473	N	F_3C-CH_2-O-	N	N	CH	CH_2	H		
474	N	F_3C-CH_2-O-	N	N	CH	CH_2	H		
475	N	F_3C-CH_2-O-	N	N	CH	CH_2	H		
476	N	F_3C-CH_2-O-	N	N	CH	CH_2	H		
477	N	F_3C-CH_2-O-	N	N	CH	0	H		
478	N	F_3C-CH_2-O-	N	N	CH	0	H		
479	N	F_3C-CH_2-O-	N	N	CH	0	H		

Continuation of Table 1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
480	N	F_3C-CH_2-O-	N	CH	N	CH_2	H		
481	N	F_3C-CH_2-O-	N	CH	N	CH_2	H		
482	N	F_3C-CH_2-O-	N	CH	N	CH_2	H		
483	N	F_3C-CH_2-O-	N	CH	N	CH_2	H		
484	N	F_3C-CH_2-O-	N	CH	N	CH_2	H		
485	N	F_3C-CH_2-O-	N	CH	N	O	H		
486	N	F_3C-CH_2-O-	N	CH	N	O	H		
487	N	F_3C-CH_2-O-	N	CH	N	O	H		

Continuation of Table 1

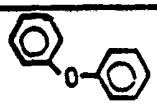
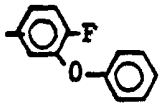
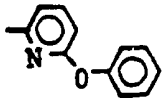
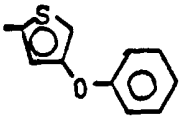
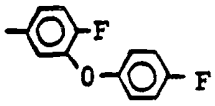
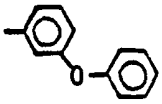
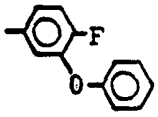
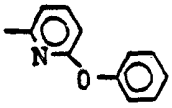
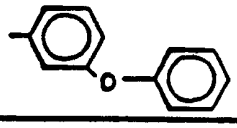
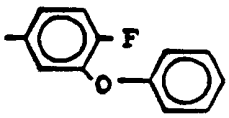
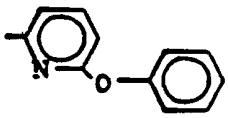
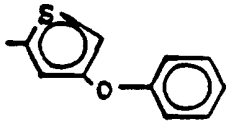
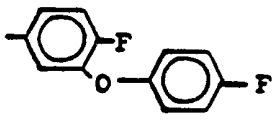
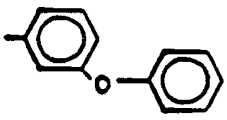
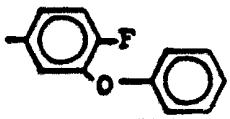
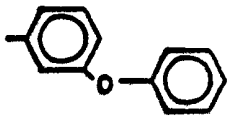
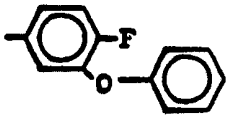
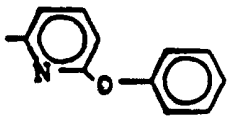
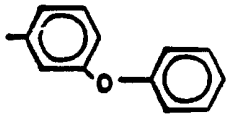
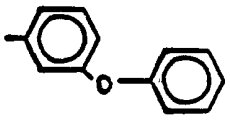
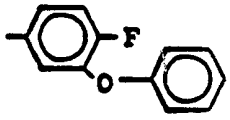
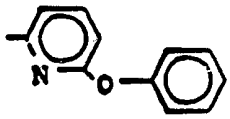
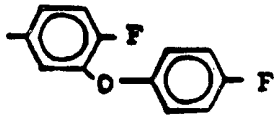
Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
488	N	F_3C-CH_2-O-	N	N	N	CH_2	H		
489	N	F_3C-CH_2-O-	N	N	N	CH_2	H		
490	N	F_3C-CH_2-O-	N	N	N	CH_2	H		
491	N	F_3C-CH_2-O-	N	N	N	CH_2	H		
492	N	F_3C-CH_2-O-	N	N	N	CH_2	H		
493	N	F_3C-CH_2-O-	N	N	N	O	H		
494	N	F_3C-CH_2-O-	N	N	N	O	H		
495	N	F_3C-CH_2-O-	N	N	N	O	H		

Table 2

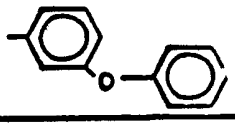
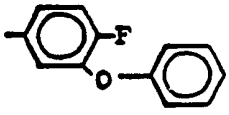
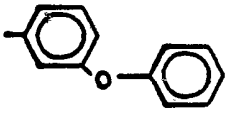
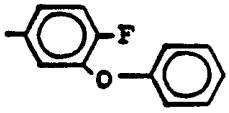
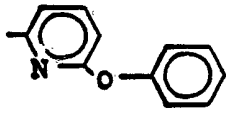
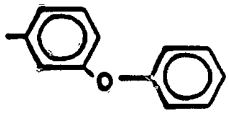
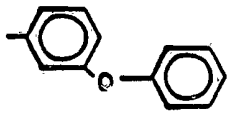
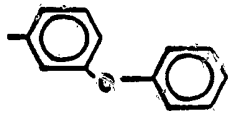
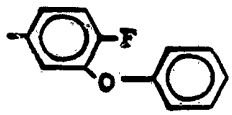
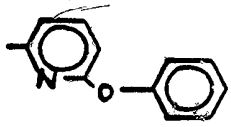
Compounds of the formula I having R^2 and R^3 = CH_2-CH_2; A = N, CH or CR^1

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
496	N	EtO	CH	CH	CH	CH_2	H		
497	N	EtO	CH	CH	CH	CH_2	H		
498	N	EtO	CH	CH	CH	CH_2	H		
499	N	EtO	CH	CH	CH	CH_2	H		
500	N	EtO	CH	CH	CH	CH_2	H		
501	N	EtO	CH	CH	CH	CH_2	CN		

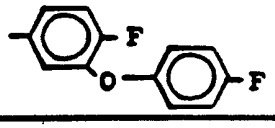
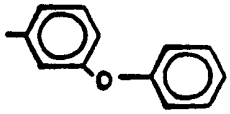
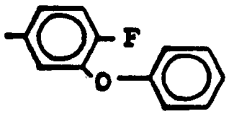
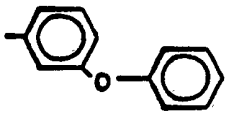
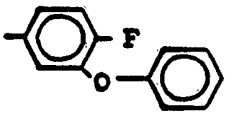
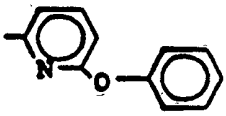
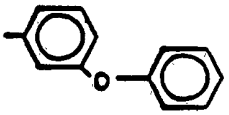
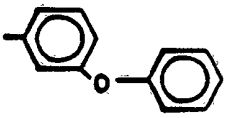
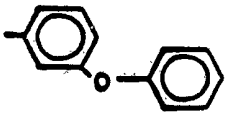
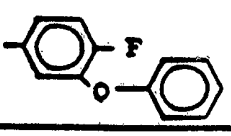
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
502	N	EtO	CH	CH	CH	CH_2	CN		
503	N	EtO	CH	CH	CH	O	H		pale yellow oil
504	N	EtO	CH	CH	CH	O	H		
505	N	EtO	CH	CH	CH	O	H		
506	N	EtO	CH	CH	CH	O	CN		
507	N	Cl	CH	CH	CH	CH_2	H		
508	N	Cl	CH	CH	CH	CH_2	H		
509	N	Cl	CH	CH	CH	CH_2	H		
510	N	Cl	CH	CH	CH	CH_2	H		

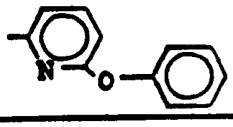
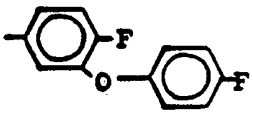
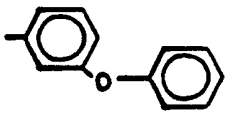
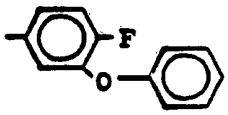
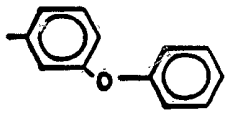
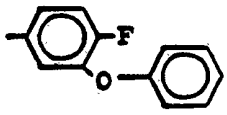
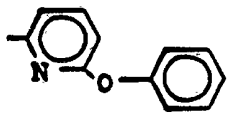
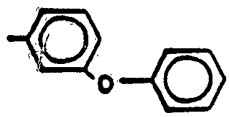
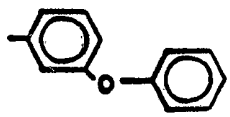
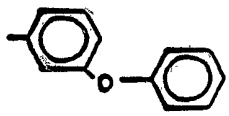
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
511	N	Cl	CH	CH	CH	CH_2	CN		
512	N	Cl	CH	CH	CH	CH_2	CN		
513	N	Cl	CH	CH	CH	O	H		pale yellow oil
514	N	Cl	CH	CH	CH	O	H		
515	N	Cl	CH	CH	CH	O	H		
516	N	Cl	CH	CH	CH	O	CN		
517	N	Cl	CH	CH	CH	O	CH_3		
518	N	H_3CO	CH	CH	CH	CH_2	H		
519	N	H_3CO	CH	CH	CH	CH_2	H		
520	N	H_3CO	CH	CH	CH	CH_2	H		

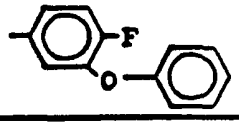
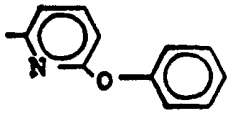
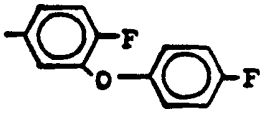
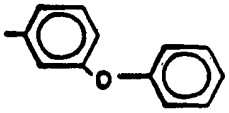
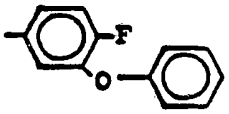
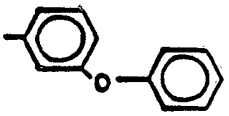
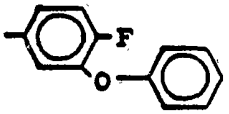
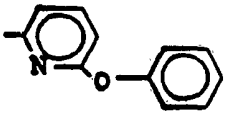
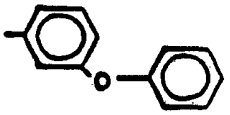
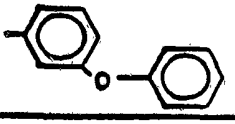
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
521	N	H_3CO	CH	CH	CH	CH_2	H		
522	N	H_3CO	CH	CH	CH	CH_2	CN		
523	N	H_3CO	CH	CH	CH	CH_2	CN		
524	N	H_3CO	CH	CH	CH	O	H		
525	N	H_3CO	CH	CH	CH	O	H		
526	N	H_3CO	CH	CH	CH	O	H		
527	N	H_3CO	CH	CH	CH	O	CN		
528	N	H_3CO	CH	CH	CH	O	CH_3		
529	N	EtS	CH	CH	CH	CH_2	H		
530	N	EtS	CH	CH	CH	CH_2	H		

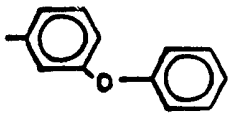
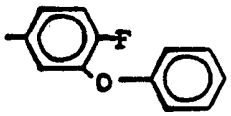
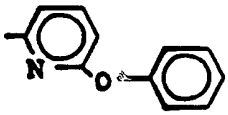
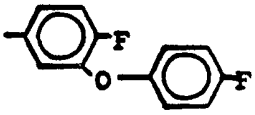
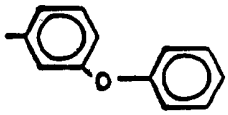
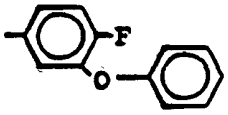
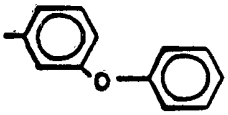
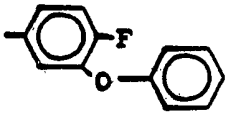
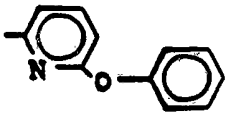
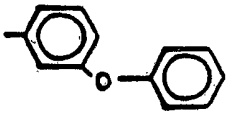
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
531	N	EtS	CH	CH	CH	CH_2	H		
532	N	EtS	CH	CH	CH	CH_2	H		
533	N	EtS	CH	CH	CH	CH_2	CN		
534	N	EtS	CH	CH	CH	CH_2	CN		
535	N	EtS	CH	CH	CH	O	H		
536	N	EtS	CH	CH	CH	O	H		
537	N	EtS	CH	CH	CH	O	H		
538	N	EtS	CH	CH	CH	O	CN		
539	N	EtS	CH	CH	CH	O	CH_3		
540	N	H_3CS	CH	CH	CH	CH_2	H		

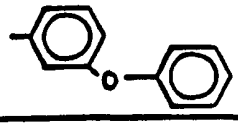
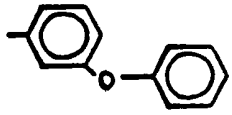
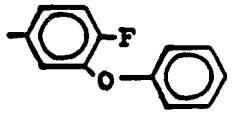
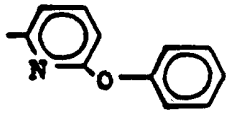
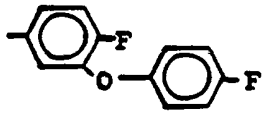
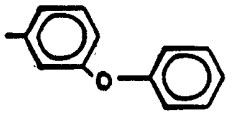
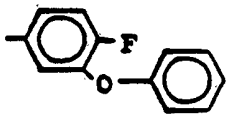
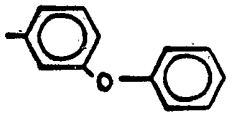
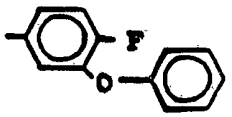
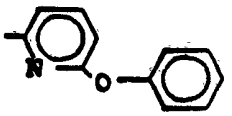
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
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542	N	H_3CS	CH	CH	CH	CH_2	H		
543	N	H_3CS	CH	CH	CH	CH_2	H		
544	N	H_3CS	CH	CH	CH	CH_2	CN		
545	N	H_3CS	CH	CH	CH	CH_2	CN		
546	N	H_3CS	CH	CH	CH	O	H		
547	N	H_3CS	CH	CH	CH	O	H		
548	N	H_3CS	CH	CH	CH	O	H		
549	N	H_3CS	CH	CH	CH	O	CN		
550	N	H_3CS	CH	CH	CH	O	CH_3		

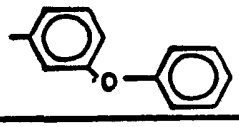
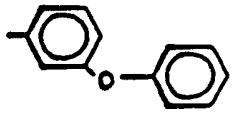
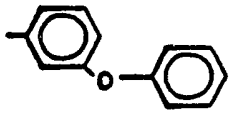
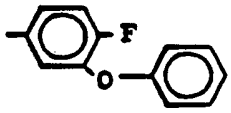
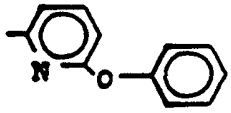
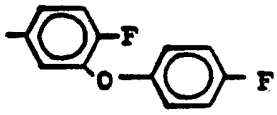
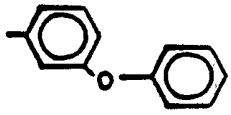
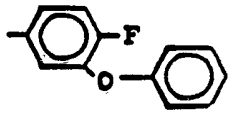
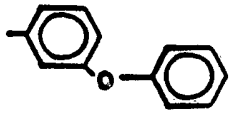
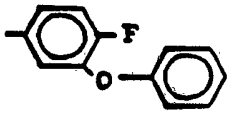
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
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552	CCl	Cl	CH	N	CH	CH_2	H		
553	CCl	Cl	CH	N	CH	CH_2	H		
554	CCl	Cl	CH	N	CH	CH_2	H		
555	CCl	Cl	CH	N	CH	CH_2	CN		
556	CCl	Cl	CH	N	CH	CH_2	CN		
557	CCl	Cl	CH	N	CH	O	H		
558	CCl	Cl	CH	N	CH	O	H		
559	CCl	Cl	CH	N	CH	O	H		
560	CCl	Cl	CH	N	CH	O	CN		

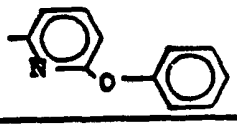
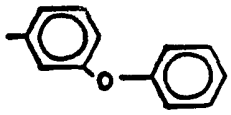
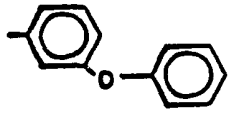
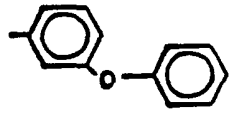
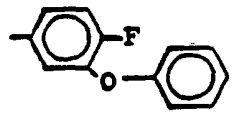
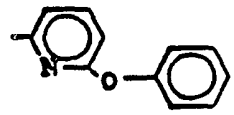
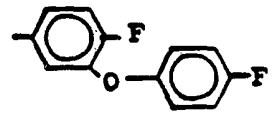
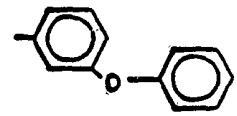
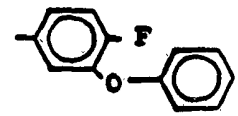
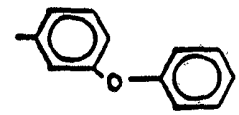
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
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562	CCl	EtO	CH	N	CH	CH_2	H		
563	CCl	EtO	CH	N	CH	CH_2	H		
564	CCl	EtO	CH	N	CH	CH_2	H		
565	CCl	EtO	CH	N	CH	CH_2	H		
566	CCl	EtO	CH	N	CH	CH_2	CN		
567	CCl	EtO	CH	N	CH	CH_2	CN		
568	CCl	EtO	CH	N	CH	O	H		
569	CCl	EtO	CH	N	CH	O	H		
570	CCl	EtO	CH	N	CH	O	H		

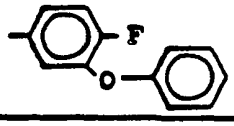
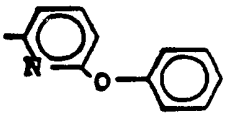
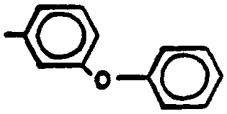
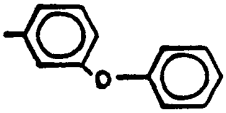
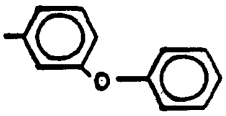
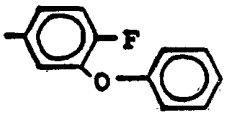
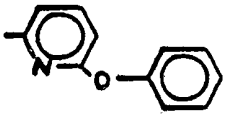
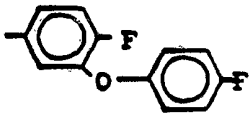
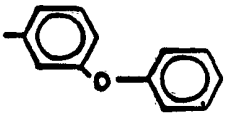
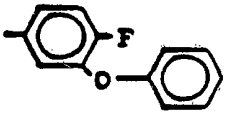
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
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572	CCl	EtO	CH	N	CH	O	CH_3		
573	CF	EtO	CH	N	CH	CH_2	H		
574	CF	EtO	CH	N	CH	CH_2	H		
575	CF	EtO	CH	N	CH	CH_2	H		
576	CF	EtO	CH	N	CH	CH_2	H		
577	CF	EtO	CH	N	CH	CH_2	CN		
578	CF	EtO	CH	N	CH	CH_2	CN		
579	CF	EtO	CH	N	CH	O	H		
580	CF	EtO	CH	N	CH	O	H		

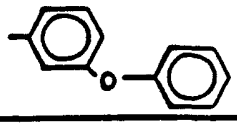
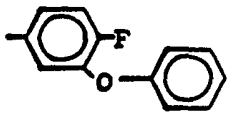
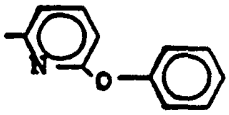
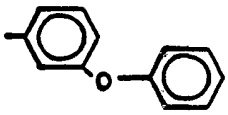
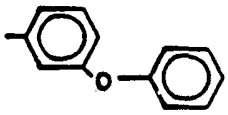
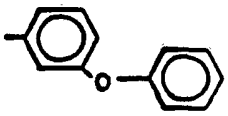
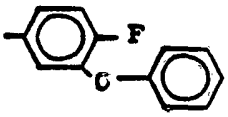
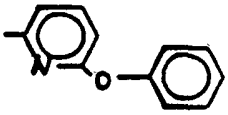
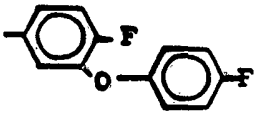
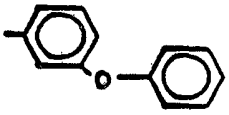
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
581	CF	EtO	CH	N	CH	O	H		
582	CF	EtO	CH	N	CH	O	CN		
583	CF	EtO	CH	N	CH	O	CH_3		
584	C-O-CH_2-O		CH	N	CH	CH_2	H		
585	C-O-CH_2-O		CH	N	CH	CH_2	H		
586	C-O-CH_2-O		CH	N	CH	CH_2	H		
587	C-O-CH_2-O		CH	N	CH	CH_2	H		
588	C-O-CH_2-O		CH	N	CH	CH_2	CN		
589	C-O-CH_2-O		CH	N	CH	CH_2	CN		
590	C-O-CH_2-O		CH	N	CH	O	H		

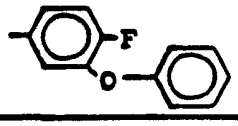
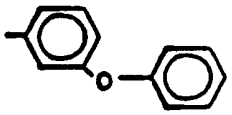
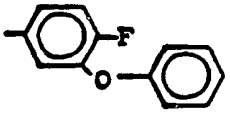
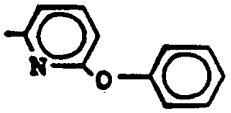
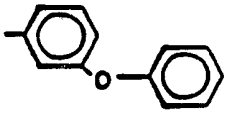
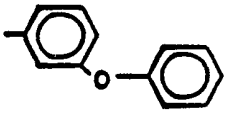
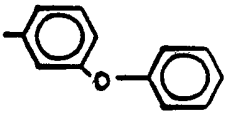
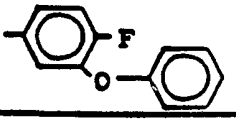
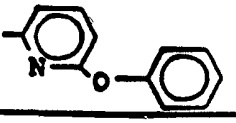
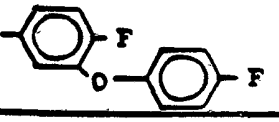
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
591	C-O-CH_2-O	CH	N	CH	O	H			
592	C-O-CH_2-O	CH	N	CH	O	H			
593	C-O-CH_2-O	CH	N	CH	O	CN			
594	C-O-CH_2-O	CH	N	CH	O	CH_3			
595		CH	EtO	N	CH	CH	CH_2	H	
596		CH	EtO	N	CH	CH	CH_2	H	
597		CH	EtO	N	CH	CH	CH_2	H	
598		CH	EtO	N	CH	CH	CH_2	H	
599		CH	EtO	N	CH	CH	CH_2	CN	
600		CH	EtO	N	CH	CH	CH_2	CN	

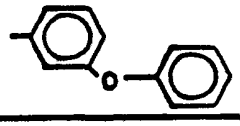
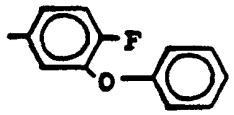
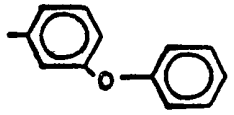
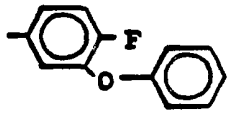
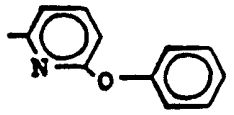
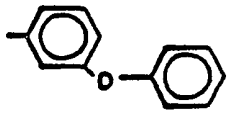
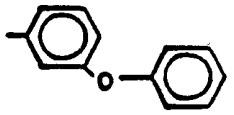
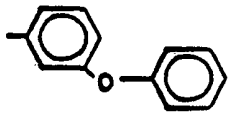
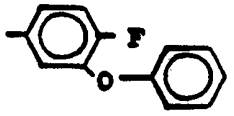
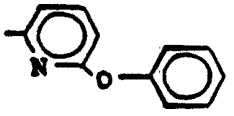
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
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602	CH	EtO	N	CH	CH	O	H		
603	CH	EtO	N	CH	CH	O	H		
604	CH	EtO	N	CH	CH	O	CN		
605	CH	EtO	N	CH	CH	O	CH_3		
606	CH	Cl	N	CH	CH	CH_2	H		
607	CH	Cl	N	CH	CH	CH_2	H		
608	CH	Cl	N	CH	CH	CH_2	H		
609	CH	Cl	N	CH	CH	CH_2	H		
610	CH	Cl	N	CH	CH	CH_2	CN		

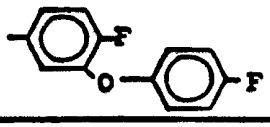
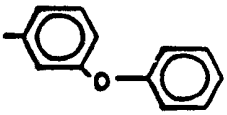
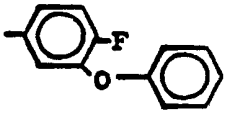
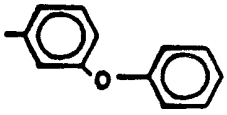
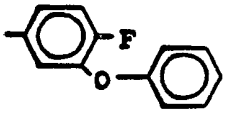
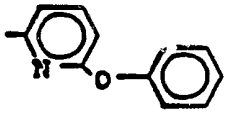
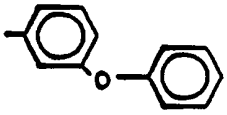
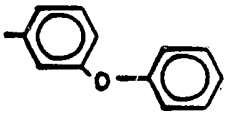
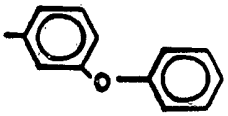
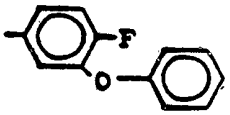
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
611	CH	Cl	N	CH	CH	CH_2	CN		
612	CH	Cl	N	CH	CH	O	H		
613	CH	Cl	N	CH	CH	O	H		
614	CH	Cl	N	CH	CH	O	H		
615	CH	Cl	N	CH	CH	O	CN		
616	CH	Cl	N	CH	CH	O	CH_3		
617	CH	H_3CO	N	CH	CH	CH_2	H		
618	CH	H_3CO	N	CH	CH	CH2	H		
619	CH	H_3CO	N	CH	CH	CH_2	H		
620	CH	H_3CO	N	CH	CH	CH_2	H		

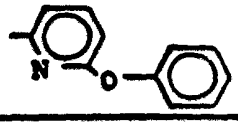
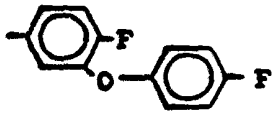
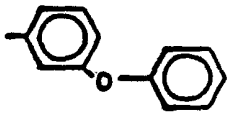
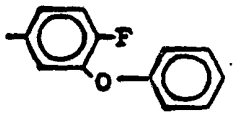
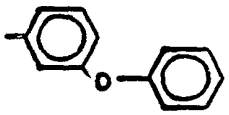
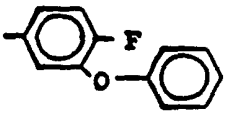
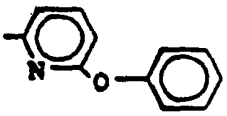
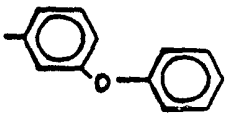
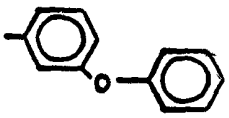
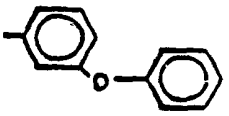
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
621	CH	H_3CO	N	CH	CH	CH_2	CN		
622	CH	H_3CO	N	CH	CH	CH_2	CN		
623	CH	H_3CO	N	CH	CH	O	H		
624	CH	H_3CO	N	CH	CH	O	H		
625	CH	H_3CO	N	CH	CH	O	H		
626	CH	H_3CO	N	CH	CH	O	CN		
627	CH	H_3CO	N	CH	CH	O	CH_3		
628	CH	EtS	N	CH	CH	CH_2	H		
629	CH	EtS	N	CH	CH	CH_2	H		
630	CH	EtS	N	CH	CH	CH_2	H		

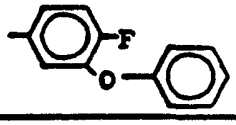
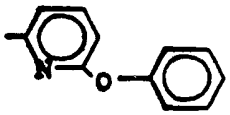
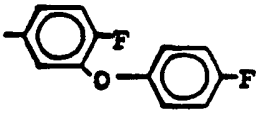
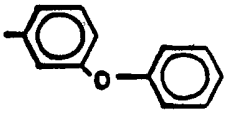
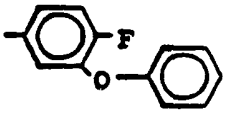
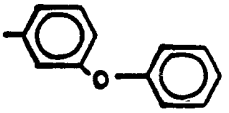
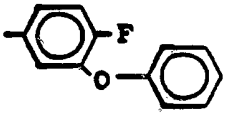
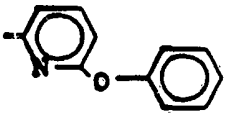
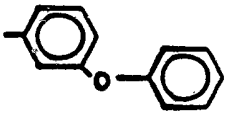
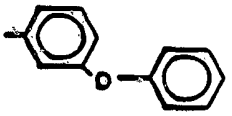
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
631	CH	EtS	N	CH	CH	CH_2	H		
632	CH	EtS	N	CH	CH	CH_2	CN		
633	CH	EtS	N	CH	CH	CH_2	CN		
634	CH	EtS	N	CH	CH	O	H		
635	CH	EtS	N	CH	CH	O	H		
636	CH	EtS	N	CH	CH	O	H		
637	CH	EtS	N	CH	CH	O	CN		
638	CH	EtS	N	CH	CH	O	CH_3		
639	CH	F_2CHO	N	CH	CH	CH_2	H		
640	CH	F_2CHO	N	CH	CH	CH_2	H		

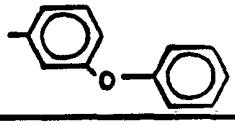
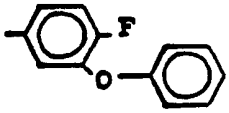
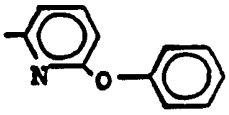
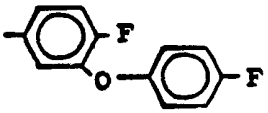
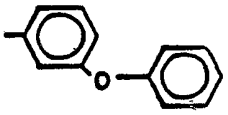
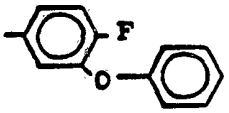
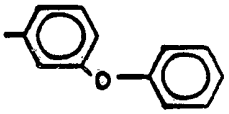
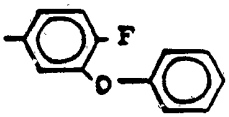
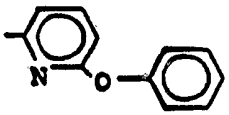
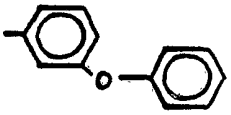
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
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642	CH	F_2CHO	N	CH	CH	CH_2	H		
643	CH	F_2CHO	N	CH	CH	CH_2	CN		
644	CH	F_2CHO	N	CH	CH	CH_2	CN		
645	CH	F_2CHO	N	CH	CH	O	H		
646	CH	F_2CHO	N	CH	CH	O	H		
647	CH	F_2CHO	N	CH	CH	O	H		
648	CH	F_2CHO	N	CH	CH	O	CN		
649	CH	F_2CHO	N	CH	CH	O	CH_3		
650	CCl	CL	N	CH	CH	CH_2	H		

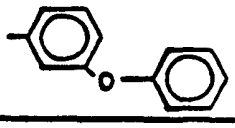
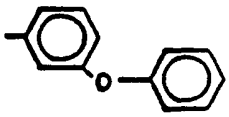
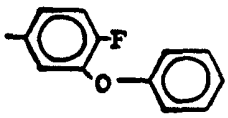
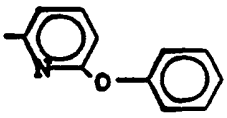
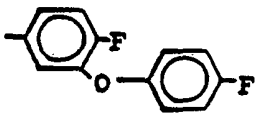
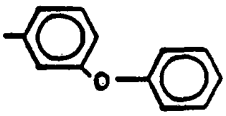
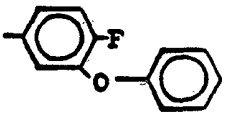
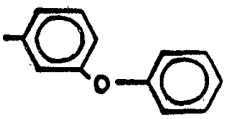
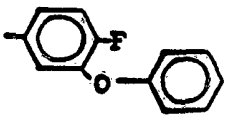
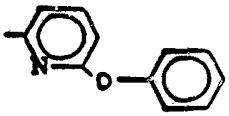
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
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652	CCl	Cl	N	CH	CH	CH_2	H		
653	CCl	Cl	N	CH	CH	CH_2	H		
654	CCl	Cl	N	CH	CH	CH_2	CN		
655	CCl	Cl	N	CH	CH	CH_2	CN		
656	CCl	Cl	N	CH	CH	O	H		
657	CCl	Cl	N	CH	CH	O	H		
658	CCl	Cl	N	CH	CH	O	H		
659	CCl	Cl	N	CH	CH	O	CN		
660	CCl	Cl	N	CH	CH	O	CH_3		

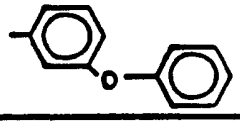
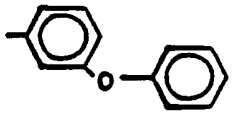
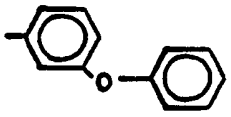
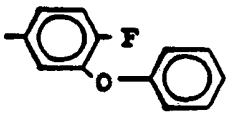
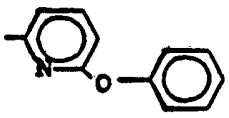
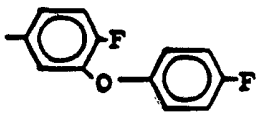
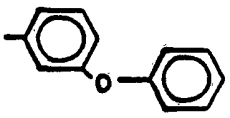
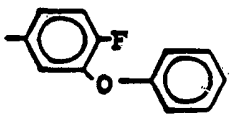
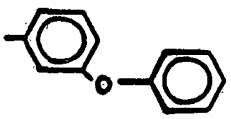
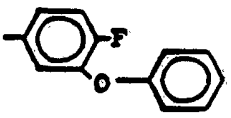
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
661	CCl	EtO	N	CH	CH	CH_2	H		
662	CCl	EtO	N	CH	CH	CH_2	H		
663	CCl	EtO	N	CH	CH	CH_2	H		
664	CCl	EtO	N	CH	CH	CH_2	H		
665	CCl	EtO	N	CH	CH	CH_2	CN		
666	CCl	EtO	N	CH	CH	CH_2	CN		
667	CCl	EtO	N	CH	CH	O	H		
668	CCl	EtO	N	CH	CH	O	H		
669	CCl	EtO	N	CH	CH	O	H		
670	CCl	EtO	N	CH	CH	O	CN		

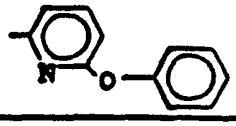
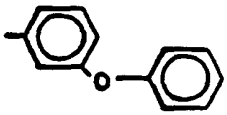
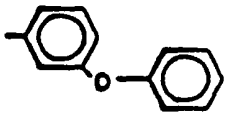
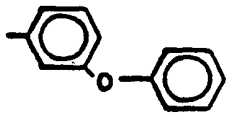
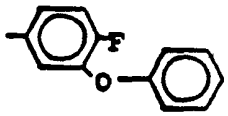
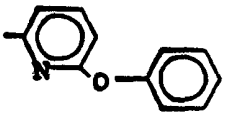
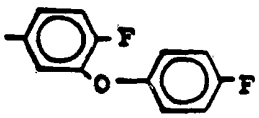
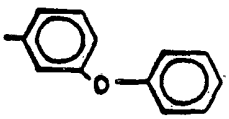
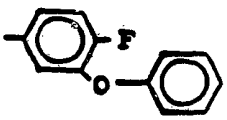
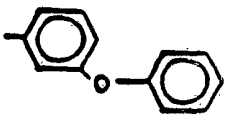
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
671	CCl	EtO	N	CH	CH	O	CH_3		
672	C-O-CH_2-O		N	CH	CH	CH_2	H		
673	C-O-CH_2-O		N	CH	CH	CH_2	H		
674	C-O-CH_2-O		N	CH	CH	CH_2	H		
675	C-O-CH_2-O		N	CH	CH	CH_2	H		
676	C-O-CH_2-O		N	CH	CH	CH_2	CN		
677	C-O-CH_2-O		N	CH	CH	CH_2	CN		
678	C-O-CH_2-O		N	CH	CH	O	H		
679	C-O-CH_2-O		N	CH	CH	O	H		
680	C-O-CH_2-O		N	CH	CH	O	H		

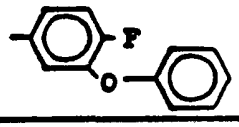
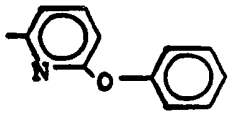
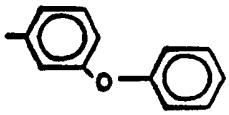
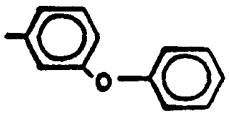
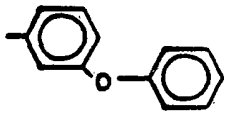
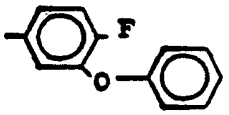
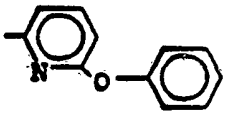
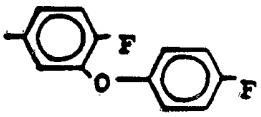
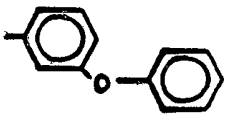
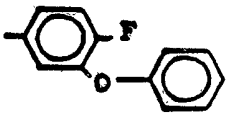
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
681	C-O-CH_2-O	N	CH	CH	O		CN		
682	C-O-CH_2-O	N	CH	CH	O		CH_3		
683	C-O-CH_2-O	CH	CH	N	CH_2		H		
684	C-O-CH_2-O	CH	CH	N	CH_2		H		
685	C-O-CH_2-O	CH	CH	N	CH_2		H		
686	C-O-CH_2-O	CH	CH	N	CH_2		H		
687	C-O-CH_2-O	CH	CH	N	CH_2		CN		
688	C-O-CH_2-O	CH	CH	N	CH_2		CN		
689	C-O-CH_2-O	CH	CH	N	O		H		
690	C-O-CH_2-O	CH	CH	N	O		H		

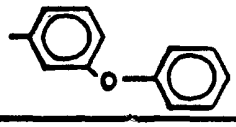
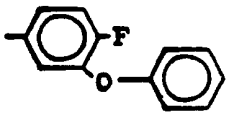
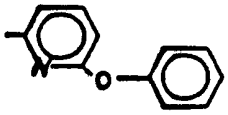
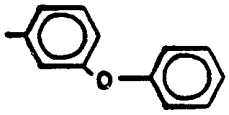
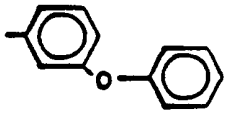
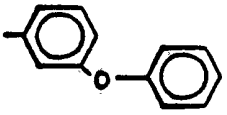
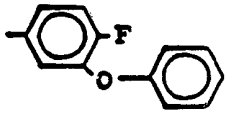
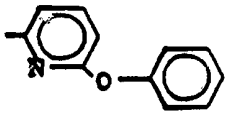
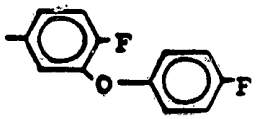
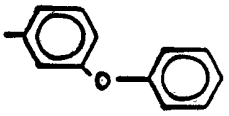
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
691	C-O-CH_2-O		CH	CH	N	O	H		
692	C-O-CH_2-O		CH	CH	N	O	CN		
693	C-O-CH_2-O		CH	CH	N	O	CH_3		
694	N	EtO	CH	N	CH	CH_2	H		
695	N	EtO	CH	N	CH	CH_2	H		
696	N	EtO	CH	N	CH	CH_2	H		
697	N	EtO	CH	N	CH	CH_2	H		
698	N	EtO	CH	N	CH	CH_2	CN		
699	N	EtO	CH	N	CH	CH_2	CN		
700	N	EtO	CH	N	CH	O	H		

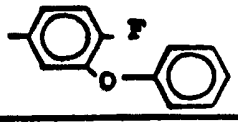
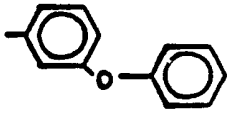
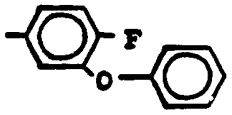
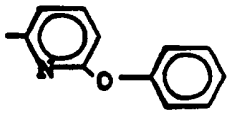
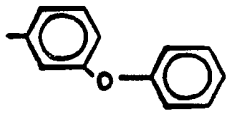
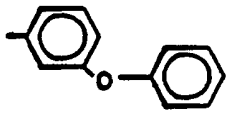
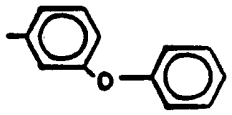
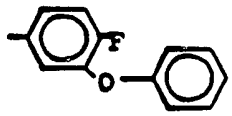
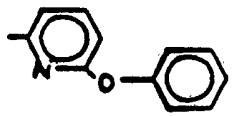
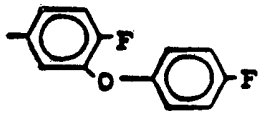
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
701	N	EtO	CH	N	CH	O	H		
702	N	EtO	CH	N	CH	O	H		
703	N	EtO	CH	N	CH	O	CN		
704	N	EtO	CH	N	CH	O	CH_3		
705	N	Cl	CH	N	CH	CH_2	H		
706	N	Cl	CH	N	CH	CH_2	H		
707	N	Cl	CH	N	CH	CH_2	H		
708	N	Cl	CH	N	CH	CH_2	H		
709	N	Cl	CH	N	CH	CH_2	CN		
710	N	Cl	CH	N	CH	CH_2	CN		

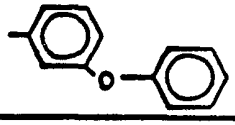
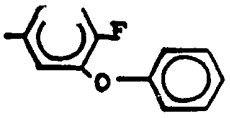
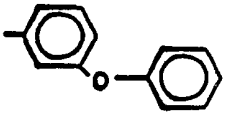
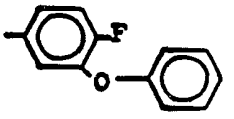
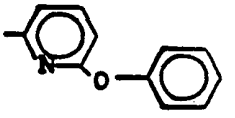
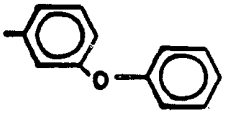
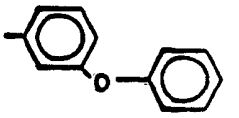
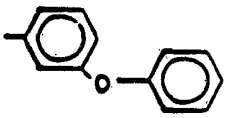
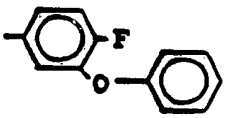
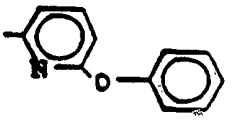
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
711	N	Cl	CH	N	CH	O	H		
712	N	Cl	CH	N	CH	O	H		
713	N	Cl	CH	N	CH	O	H		
714	N	Cl	CH	N	CH	O	CN		
715	N	Cl	CH	N	CH	O	CH_3		
716	N	H_3CO	CH	N	CH	CH_2	H		
717	N	H_3CO	CH	N	CH	CH_3	H		
718	N	H_3CO	CH	N	CH	CH_2	H		
719	N	H_3CO	CH	N	CH	CH_2	H		
720	N	H_3CO	CH	N	CH	CH_2	CN		

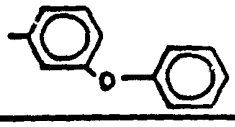
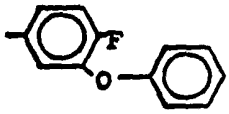
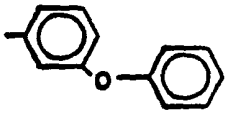
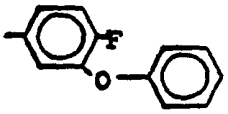
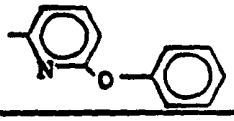
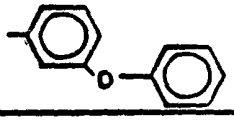
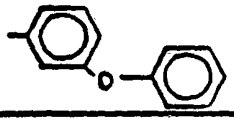
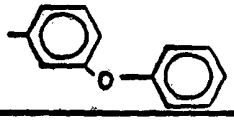
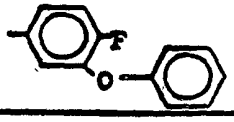
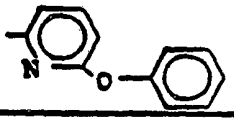
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
721	N	H_3CO	CH	N	CH	CH_2	CN		
722	N	H_3CO	CH	N	CH	O	H		
723	N	H_3CO	CH	N	CH	O	H		
724	N	H_3CO	CH	N	CH	O	H		
725	N	H_3CO	CH	N	CH	O	CN		
726	N	H_3CO	CH	N	CH	O	CH_3		
727	N	EtS	CH	N	CH	CH_2	H		
728	N	EtS	CH	N	CH	CH_2	H		
729	N	EtS	CH	N	CH	CH_2	H		
730	N	EtS	CH	N	CH	CH_2	H		

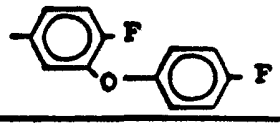
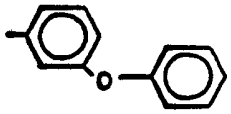
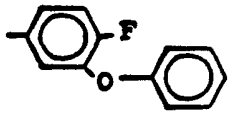
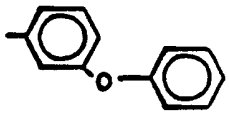
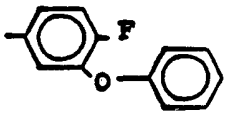
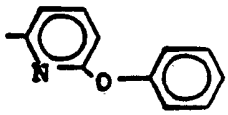
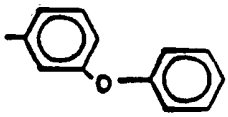
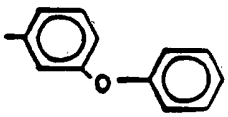
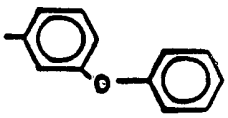
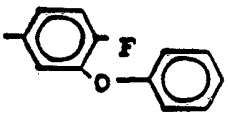
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
731	N	EtS	CH	N	CH	CH_2	CN		
732	N	EtS	CH	N	CH	CH_2	CN		
733	N	EtS	CH	N	CH	O	H		
734	N	EtS	CH	N	CH	O	H		
735	N	EtS	CH	N	CH	O	H		
736	N	EtS	CH	N	CH	O	CN		
737	N	EtS	CH	N	CH	O	CH_3		
738	N	n-C_3H_7	CH	N	CH	CH_2	H		
739	N	n-C_3H_7	CH	N	CH	CH_2	H		
740	N	n-C_3H_7	CH	N	CH	CH_2	H		

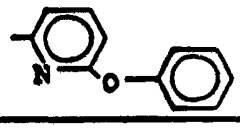
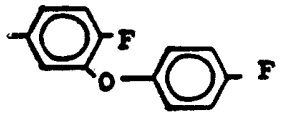
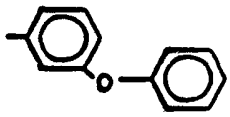
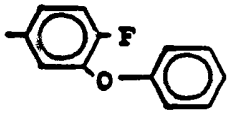
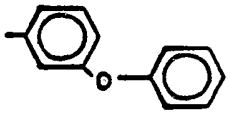
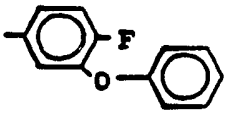
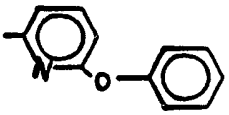
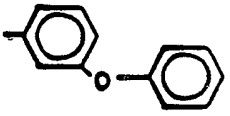
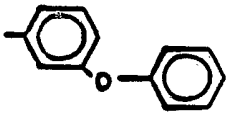
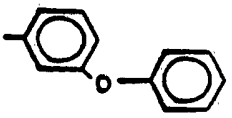
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
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742	N	n-C_3H_7	CH	N	CH	CH_2	CN		
743	N	n-C_3H_7	CH	N	CH	O	H		
744	N	n-C_3H_7	CH	N	CH	O	H		
745	N	n-C_3H7	CH	N	CH	O	H		
746	N	n-C_3H_7	CH	N	CH	O	CN		
747	N	n-C_3H_7	CH	N	CH	O	CH_3		
748	CH	EtO	N	CH	N	CH_2	H		
749	CH	EtO	N	CH	N	CH_2	H		
750	CH	EtO	N	CH	N	CH_2	H		

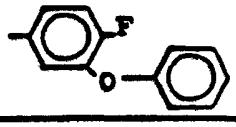
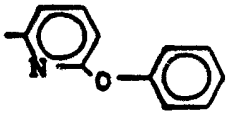
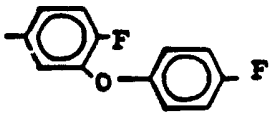
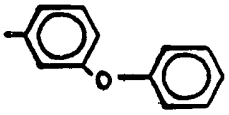
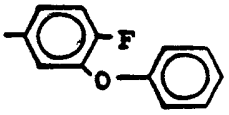
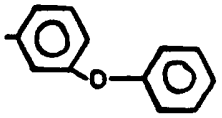
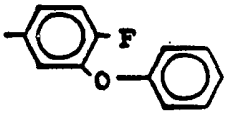
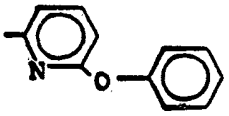
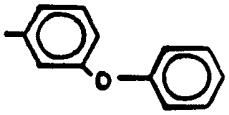
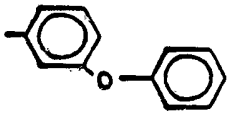
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
751	CH	EtO	N	CH	N	CH_2	H		
752	CH	EtO	N	CH	N	CH_2	CN		
753	CH	EtO	N	CH	N	CH_2	CN		
754	CH	EtO	N	CH	N	O	H		
755	CH	EtO	N	CH	N	O	H		
756	CH	EtO	N	CH	N	O	H		
757	CH	EtO	N	CH	N	O	CN		
758	CH	EtO	N	CH	N	O	CH_3		
759	CH	Cl	N	CH	N	CH_2	H		
760	CH	Cl	N	CH	N	CH_2	H		

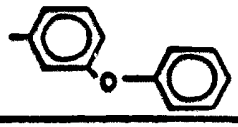
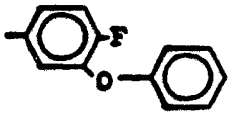
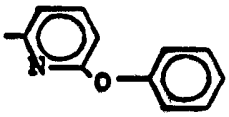
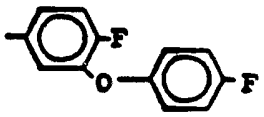
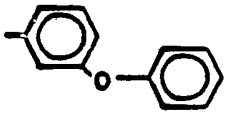
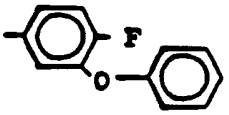
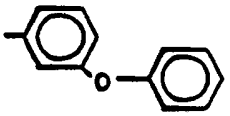
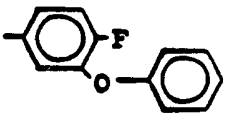
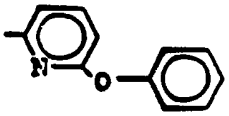
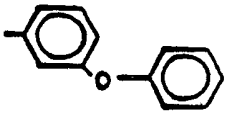
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
761	CH	Cl	N	CH	N	CH_2	H		
762	CH	Cl	N	CH	N	CH_2	H		
763	CH	Cl	N	CH	N	CH_2	CN		
764	CH	Cl	N	CH	N	CH_2	CN		
765	CH	Cl	N	CH	N	O	H		
776	CH	Cl	N	CH	N	O	H		
777	CH	Cl	N	CH	N	O	H		
778	CH	Cl	N	CH	N	O	CN		
779	CH	Cl	N	CH	N	O	CH_3		
780	CH	H_3CO	N	CH	N	CH_2	H		

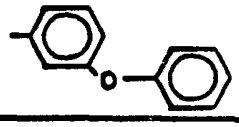
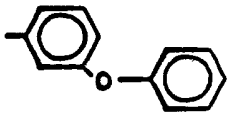
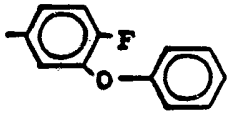
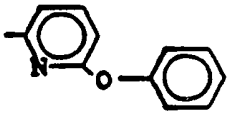
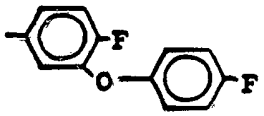
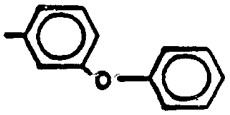
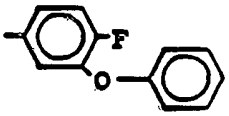
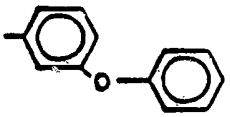
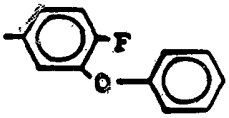
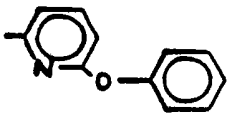
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
781	CH	H_3CO	N	CH	N	CH_2	H		
782	CH	H_3CO	N	CH	N	CH_2	H		
783	CH	H_3CO	N	CH	N	CH_2	H		
784	CH	H_3CO	N	CH	N	CH_2	CN		
785	CH	H_3CO	N	CH	N	CH_2	CN		
786	CH	H_3CO	N	CH	N	O	H		
787	CH	H_3CO	N	CH	N	O	H		
788	CH	H_3CO	N	CH	N	O	H		
789	CH	H_3CO	N	CH	N	O	CN		
790	CH	H_3CO	N	CH	N	O	CH_3		

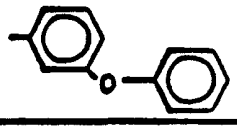
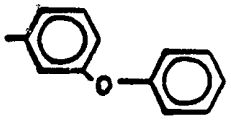
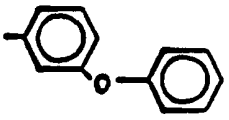
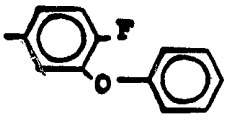
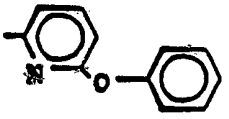
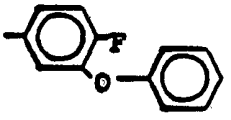
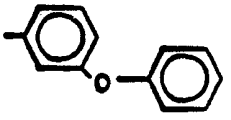
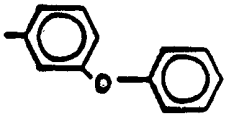
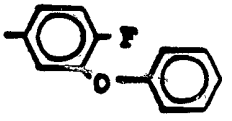
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
791	CH	F_2CHO	N	CH	N	CH_2	H		
792	CH	F_2CHO	N	CH	N	CH_2	H		
793	CH	F_2CHO	N	CH	N	CH_2	H		
794	CH	F_2CHO	N	CH	N	CH_2	H		
795	CH	F_2CHO	N	CH	N	CH_2	CN		
796	CH	F_2CHO	N	CH	N	CH_2	CN		
797	CH	F_2CHO	N	CH	N	O	H		
798	CH	F_2CHO	N	CH	N	O	H		
799	CH	F_2CHO	N	CH	N	O	H		
800	CH	F_2CHO	N	CH	N	O	CN		

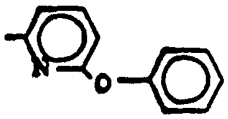
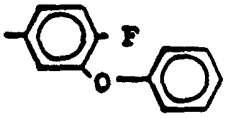
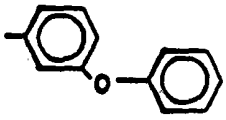
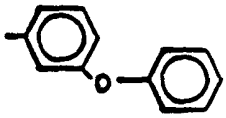
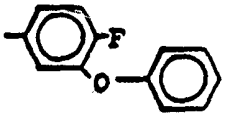
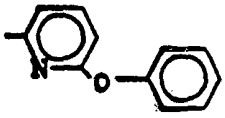
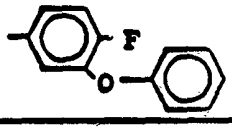
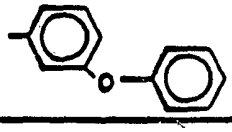
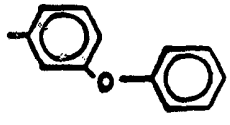
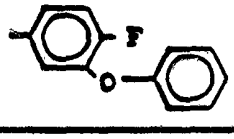
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
801	CH	F_2CHO	N	CH	N	O	CH_3		
802	CH	EtS	N	CH	N	CH_2	H		
803	CH	EtS	N	CH	N	CH_2	H		
804	CH	EtS	N	CH	N	CH_2	H		
805	CH	EtS	N	CH	N	CH_2	H		
806	CH	EtS	N	CH	N	CH_2	CN		
807	CH	EtS	N	CH	N	CH_2	CN		
808	CH	EtS	N	CH	N	O	H		
809	CH	EtS	N	CH	N	O	H		
810	CH	EtS	N	CH	N	O	H		

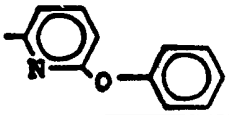
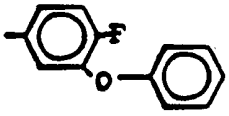
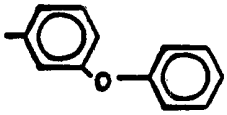
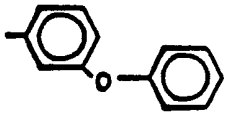
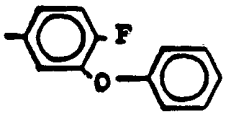
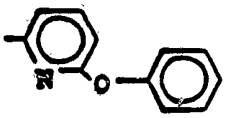
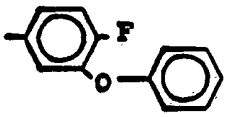
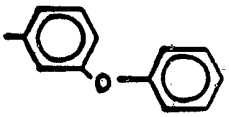
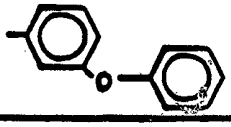
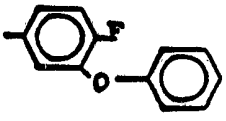
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
811	CH	EtS	N	CH	N	O	CN		
812	CH	EtS	N	CH	N	O	CH_3		
813	N	EtO	CH	CH	N	CH_2	H		
814	N	EtO	CH	CH	N	CH_2	H		
815	N	EtO	CH	CH	N	CH_2	H		
816	N	EtO	CH	CH	N	O	H		
817	N	EtO	CH	CH	N	O	H		
818	N	Cl	CH	CH	N	CH_2	H		
819	N	Cl	CH	CH	N	CH_2	H		

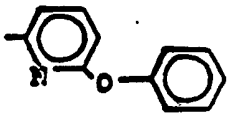
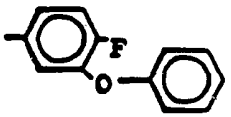
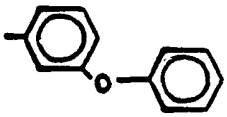
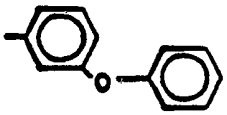
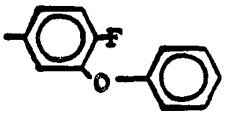
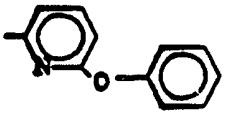
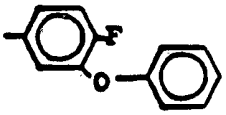
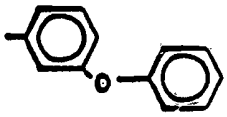
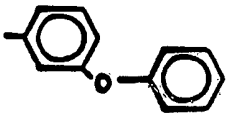
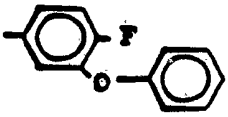
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
820	N	Cl	CH	CH	N	CH_2	H		
821	N	Cl	CH	CH	N	O	H		
822	N	Cl	CH	CH	N	O	H		
823	N	CH_3O	CH	CH	N	CH_2	H		
824	N	CH_3O	CH	CH	N	CH_2	H		
825	N	CH_3O	CH	CH	N	CH_2	H		
826	N	CH_3O	CH	CH	N	O	H		
827	N	CH_3O	CH	CH	N	O	H		
828	N	EtS	CH	CH	N	CH_2	H		
829	N	EtS	CH	CH	N	CH_2	H		

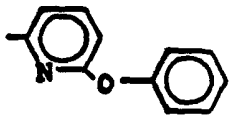
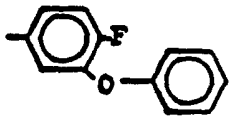
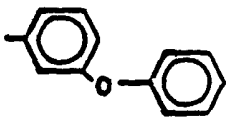
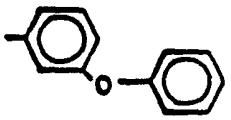
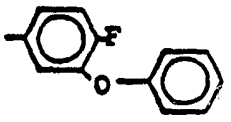
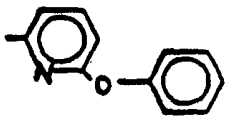
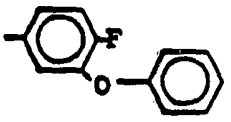
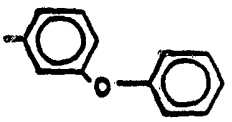
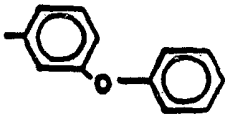
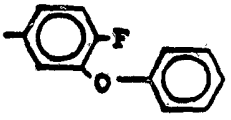
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
830	N	EtS	CH	CH	N	CH_2	H		
831	N	EtS	CH	CH	N	O	H		
832	N	EtS	CH	CH	N	O	H		
833	CCl	EtO	N	N	CH	CH_2	H		
834	CCl	EtO	N	N	CH	CH_2	H		
835	CCl	EtO	N	N	CH	CH_2	H		
836	CCl	EtO	N	N	CH	O	H		
837	CCl	EtO	N	N	CH	O	H		
838	CCl	Cl	N	N	CH	CH_2	H		
839	CCl	Cl	N	N	CH	CH_2	H		

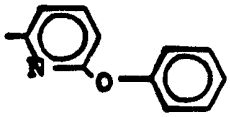
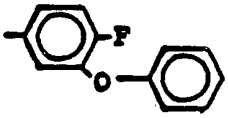
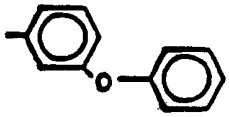
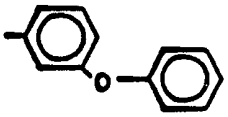
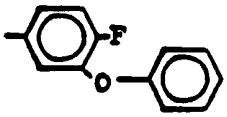
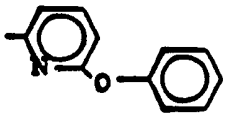
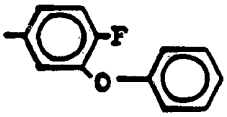
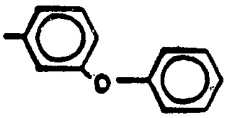
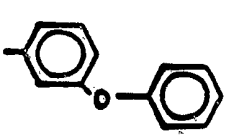
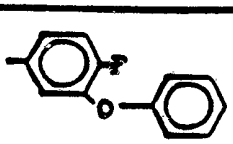
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
840	CCl	Cl	N	N	CH	CH_2	H		
841	CCl	Cl	N	N	CH	O	H		
842	CCl	Cl	N	N	CH	O	H		
843	CH	EtO	CH	N	N	CH_2	H		
844	CH	EtO	CH	N	N	CH_2	H		
845	CH	EtO	CH	N	N	CH_2	H		
846	CH	EtO	CH	N	N	O	H		
847	CH	EtO	CH	N	N	O	H		
848	CH	CH_3O	CH	N	N	CH_2	H		
849	CH	CH_3O	CH	N	N	CH_2	H		

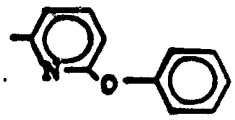
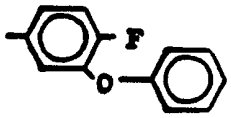
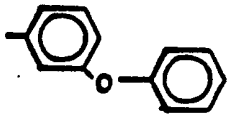
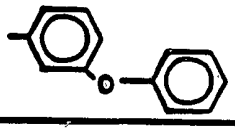
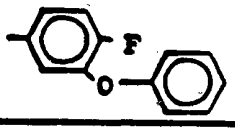
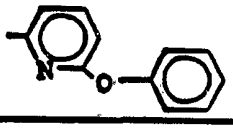
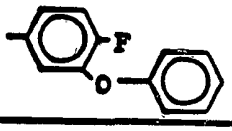
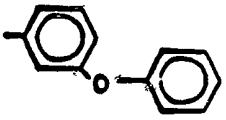
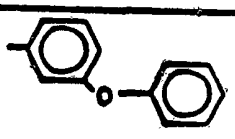
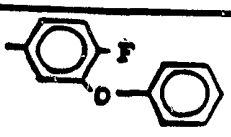
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
850	CH	CH_3O	CH	N	N	CH_2	H		
851	CH	CH_3O	CH	N	N	O	H		
852	CH	CH_3O	CH	N	N	O	H		
853	CH	Cl	CH	N	N	CH_2	H		
854	CH	Cl	CH	N	N	CH_2	H		
855	CH	Cl	CH	N	N	CH_2	H		
856	CH	Cl	CH	N	N	O	H		
857	CH	Cl	CH	N	N	O	H		
858	CH	EtS	CH	N	N	CH_2	H		
859	CH	EtS	CH	N	N	CH_2	H		

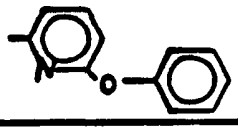
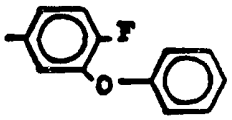
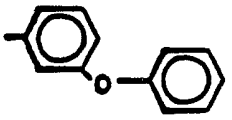
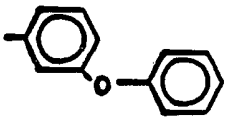
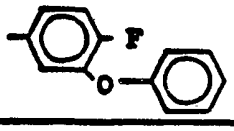
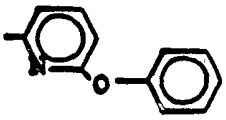
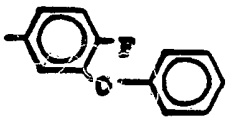
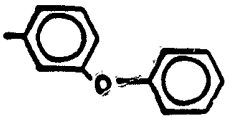
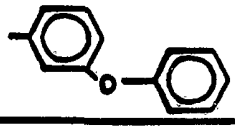
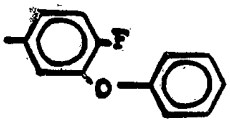
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
860	CH	EtS	CH	N	N	CH_2	H		
861	CH	EtS	CH	N	N	O	H		
862	CH	EtS	CH	N	N	O	H		
863	CCl	Cl	CH	N	N	CH_2	H		
864	CCl	Cl	CH	N	N	CH_2	H		
865	CCl	Cl	CH	N	N	CH_2	H		
866	CCl	Cl	CH	N	N	O	H		
857	CCl	Cl	CH	N	N	O	H		
868	CH	EtO	N	N	N	CH_2	H		
869	CH	EtO	N	N	N	CH_2	H		

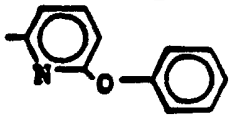
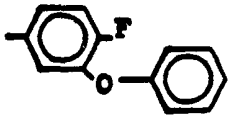
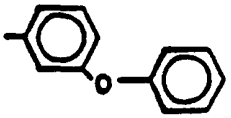
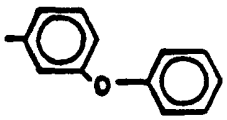
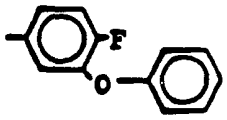
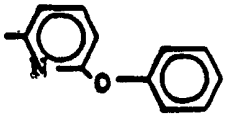
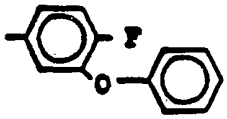
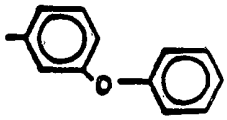
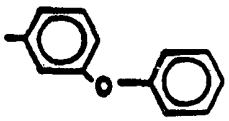
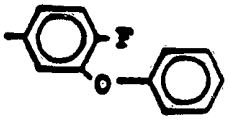
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
870	CH	EtO	N	N	N	CH_2	H		
871	CH	EtO	N	N	N	O	H		
872	CH	EtO	N	N	N	O	H		
873	CH	CH_3O	N	N	N	CH_2	H		
874	CH	CH_3O	N	N	N	CH_2	H		
875	CH	CH_3O	N	N	N	CH_2	H		
876	CH	CH_3O	N	N	N	O	H		
877	CH	CH_3O	N	N	N	O	H		
878	CH	Cl	N	N	N	CH_2	H		
879	CH	Cl	N	N	N	CH_2	H		

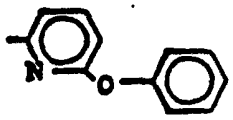
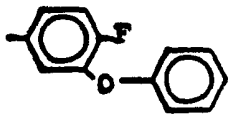
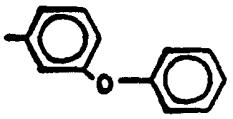
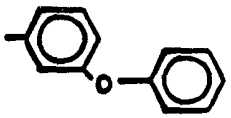
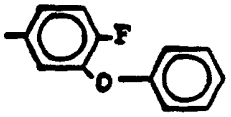
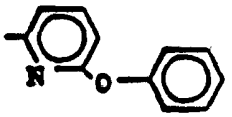
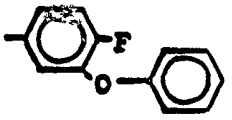
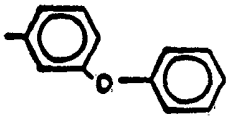
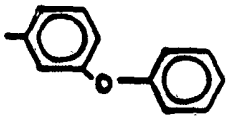
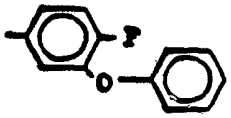
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
880	CH	Cl	N	N	N	CH_2	H		
881	CH	Cl	N	N	N	O	H		
882	CH	Cl	N	N	N	O	H		
883	CH	EtS	N	N	N	CH_2	H		
884	CH	EtS	N	N	N	CH_2	H		
885	CH	EtS	N	N	N	CH_2	H		
886	CH	EtS	N	N	N	O	H		
887	CH	EtS	N	N	N	O	H		
888	N	EtS	N	N	N	CH_2	H		
889	N	EtO	N	N	CH	CH_2	H		

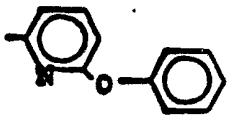
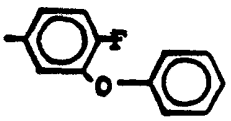
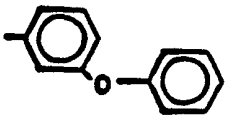
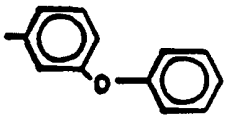
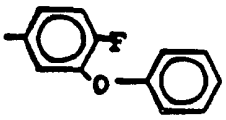
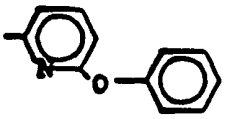
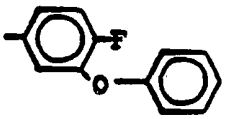
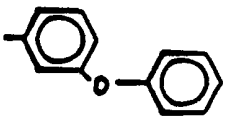
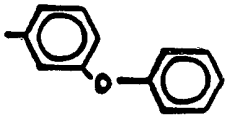
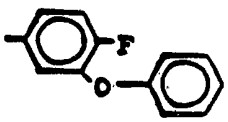
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
890	N	EtO	N	N	CH	CH_2	H		
891	N	EtO	N	N	N	O	H		
892	N	EtO	N	N	CH	O	H		
893	N	CH_3O	N	N	CH	CH_2	H		
894	N	CH_3O	N	N	CH	CH_2	H		
895	N	CH_3O	N	N	CH	CH_2	H		
896	N	CH_3O	N	N	CH	O	H		
897	N	CH_3O	N	N	CH	O	H		
898	N	Cl	N	N	CH	CH_2	H		
899	N	Cl	N	N	CH	CH_2	H		

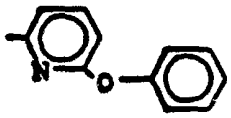
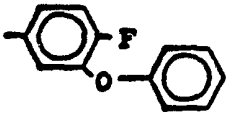
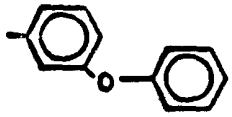
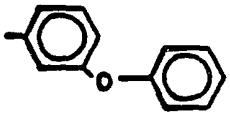
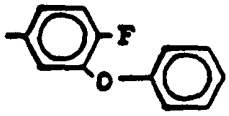
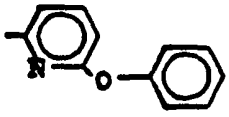
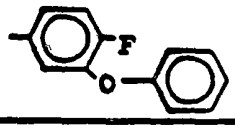
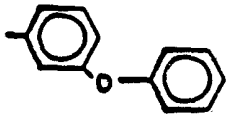
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
900	N	Cl	N	N	CH	CH_2	H		
901	N	Cl	N	N	CH	O	H		
902	N	Cl	N	N	CH	O	H		
903	N	EtS	N	N	CH	CH_2	H		
904	N	EtS	N	N	CH	CH_2	H		
905	N	EtS	N	N	CH	CH_2	H		
906	N	EtS	N	N	CH	O	H		
907	N	EtS	N	N	CH	O	H		
908	N	EtO	N	N	N	CH_2	H		
909	N	EtO	N	N	N	CH_2	H		

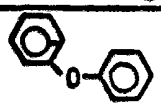
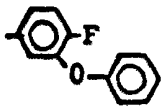
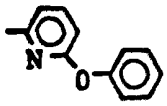
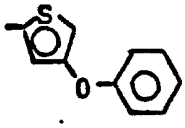
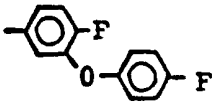
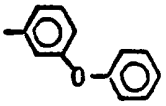
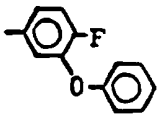
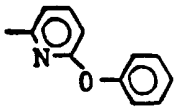
Continuation of Table 2

Compound No.	A	R^1	B	C	D	X	R^4	R^5	Physical data
910	N	EtO	N	N	N	CH_2	H		
911	N	EtO	N	N	N	O	H		
912	N	EtO	N	N	N	O	H		
913	N	CH_3O	N	N	N	CH_2	H		
914	N	CH_3O	N	N	N	CH_2	H		
915	N	CH_3O	N	N	N	CH_2	H		
916	N	CH_3O	N	N	N	O	H		
917	N	CH_3O	N	N	N	O	H		
918	N	Cl	N	N	N	CH_2	H		
919	N	Cl	N	N	N	CH_2	H		

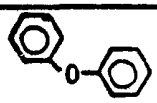
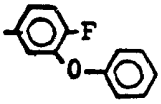
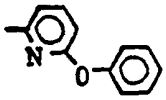
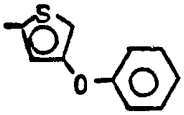
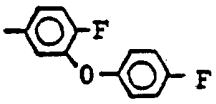
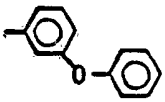
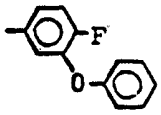
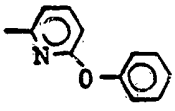
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
920	N	Cl	N	N	N	CH_2	H		
921	N	Cl	N	N	N	O	H		
923	N	Cl	N	N	N	O	H		
924	N	EtS	N	N	N	CH_2	H		
925	N	EtS	N	N	N	CH_2	H		
926	N	EtS	N	N	N	CH_2	H		
927	N	EtS	N	N	N	O	H		
928	N	EtS	N	N	N	O	H		

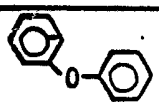
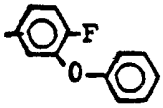
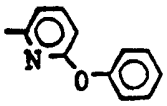
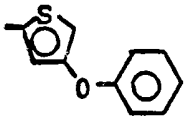
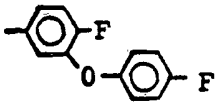
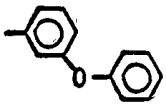
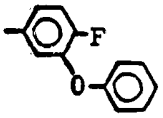
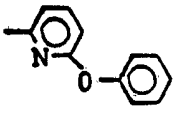
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
929	N	F_3C-CH_2-O-	CH	CH	CH	CH_2	H		
930	N	F_3C-CH_2-O-	CH	CH	CH	CH_2	H		
931	N	F_3C-CH_2-O-	CH	CH	CH	CH_2	H		
932	N	F_3C-CH_2-O-	CH	CH	CH	CH_2	H		
933	N	F_3C-CH_2-O-	CH	CH	CH	CH_2	H		
934	N	F_3C-CH_2-O-	CH	CH	CH	O	H		
935	N	F_3C-CH_2-O-	CH	CH	CH	O	H		
936	N	F_3C-CH_2-O-	CH	CH	CH	O	H		

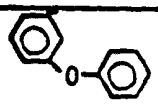
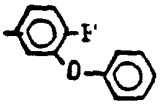
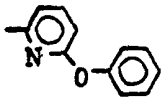
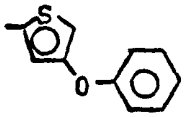
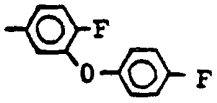
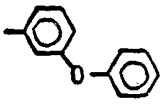
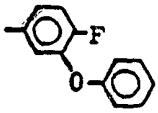
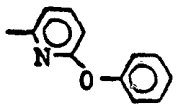
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
937	CH	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
938	CH	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
939	CH	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
940	CH	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
941	CH	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
942	CH	F_3C-CH_2-O-	N	CH	CH	O	H		
943	CH	F_3C-CH_2-O-	N	CH	CH	O	H		
944	CH	F_3C-CH_2-O-	N	CH	CH	O	H		

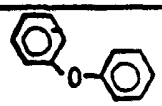
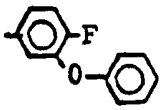
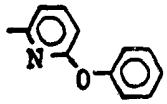
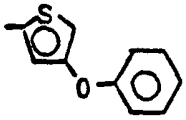
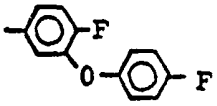
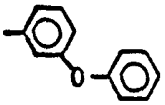
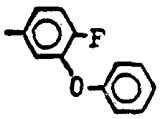
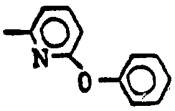
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
945	N	F_3C-CH_2-O-	CH	N	CH	CH_2	H		
946	N	F_3C-CH_2-O-	CH	N	CH	CH_2	H		
947	N	F_3C-CH_2-O-	CH	N	CH	CH_2	H		
948	N	F_3C-CH_2-O-	CH	N	CH	CH_2	H		
949	N	F_3C-CH_2-O-	CH	N	CH	CH_2	H		
950	N	F_3C-CH_2-O-	CH	N	CH	O	H		
951	N	F_3C-CH_2-O-	CH	N	CH	O	H		
952	N	F_3C-CH_2-O-	CH	N	CH	O	H		

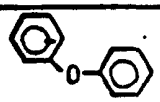
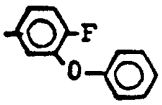
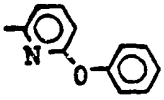
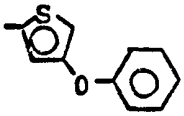
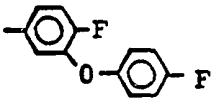
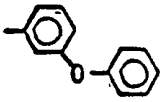
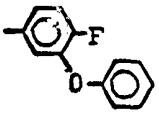
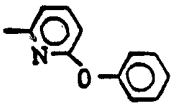
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
953	CH	F_3C-CH_2-O-	N	CH	N	CH_2	H		
954	CH	F_3C-CH_2-O-	N	CH	N	CH_2	H		
955	CH	F_3C-CH_2-O-	N	CH	N	CH_2	H		
956	CH	F_3C-CH_2-O-	N	CH	N	CH_2	H		
957	CH	F_3C-CH_2-O-	N	CH	N	CH_2	H		
958	CH	F_3C-CH_2-O-	N	CH	N	O	H		
959	CH	F_3C-CH_2-O-	N	CH	N	O	H		
960	CH	F_3C-CH_2-O-	N	CH	N	O	H		

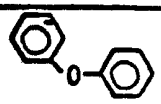
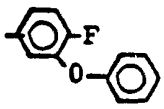
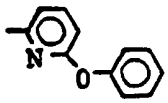
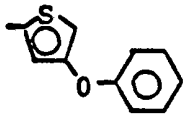
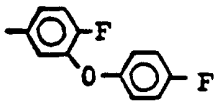
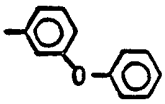
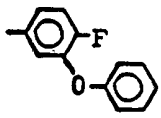
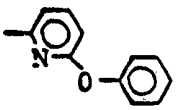
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
961	N	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
962	N	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
963	N	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
964	N	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
965	N	F_3C-CH_2-O-	N	CH	CH	CH_2	H		
966	N	F_3C-CH_2-O-	N	CH	CH	O	H		
967	N	F_3C-CH_2-O-	N	CH	CH	O	H		
968	N	F_3C-CH_2-O-	N	CH	CH	O	H		

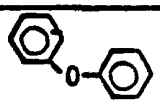
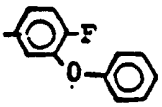
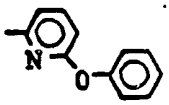
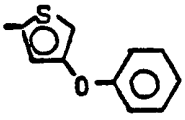
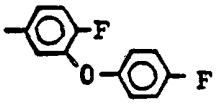
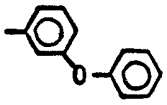
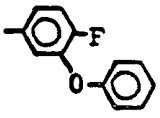
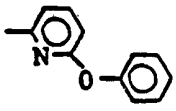
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
969	N	F_3C-CH_2-O-	CH	CH	N	CH_2	H		
970	N	F_3C-CH_2-O-	CH	CH	N	CH_2	H		
971	N	F_3C-CH_2-O-	CH	CH	N	CH_2	H		
972	N	F_3C-CH_2-O-	CH	CH	N	CH_2	H		
973	N	F_3C-CH_2-O-	CH	CH	N	CH_2	H		
974	N	F_3C-CH_2-O-	CH	CH	N	O	H		
975	N	F_3C-CH_2-O-	CH	CH	N	O	H		
976	N	F_3C-CH_2-O-	CH	CH	N	O	H		

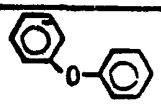
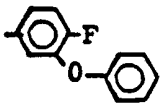
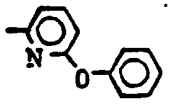
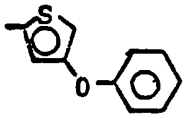
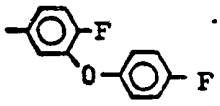
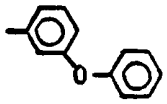
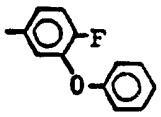
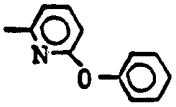
Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
977	N	F_3C-CH_2-O-	N	N	CH	CH_2	H		
978	N	F_3C-CH_2-O-	N	N	CH	CH_2	H		
979	N	F_3C-CH_2-O-	N	N	CH	CH_2	H		
980	N	F_3C-CH_2-O-	N	N	CH	CH_2	H		
981	N	F_3C-CH_2-O-	N	N	CH	CH_2	H		
982	N	F_3C-CH_2-O-	N	N	CH	O	H		
983	N	F_3C-CH_2-O-	N	N	CH	O	H		
984	N	F_3C-CH_2-O-	N	N	CH	O	H		

Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
985	N	F_3C-CH_2-O-	N	CH	N	CH_2	H		
986	N	F_3C-CH_2-O-	N	CH	N	CH_2	H		
987	N	F_3C-CH_2-O-	N	CH	N	CH_2	H		
988	N	F_3C-CH_2-O-	N	CH	N	CH_2	H		
989	N	F_3C-CH_2-O-	N	CH	N	CH_2	H		
990	N	F_3C-CH_2-O-	N	CH	N	O	H		
991	N	F_3C-CH_2-O-	N	CH	N	O	H		
992	N	F_3C-CH_2-O-	N	CH	N	O	H		

Continuation of Table 2

Compound No.	A	R^1	B	C'	D	X	R^4	R^5	Physical data
993	N	F_3C-CH_2-O-	N	N	N	CH_2	H		
994	N	F_3C-CH_2-O-	N	N	N	CH_2	H		
995	N	F_3C-CH_2-O-	N	N	N	CH_2	H		
996	N	F_3C-CH_2-O-	N	N	N	CH_2	H		
997	N	F_3C-CH_2-O-	N	N	N	CH_2	H		
998	N	F_3C-CH_2-O-	N	N	N	O	H		
999	N	F_3C-CH_2-O-	N	N	N	O	H		
1000	N	F_3C-CH_2-O-	N	N	N	O	H		

C. Biological examples

Example 1

Broad beans (Vicia faba) strongly infested with cowpea aphid (Aphis craccivora) were sprayed with aqueous dilutions of emulsion concentrates having active compound contents of 1,000 ppm until the stage of the onset of dripping. After 3 days, the mortality was in each case 100% in the preparations containing the active compounds of Examples 1, 2, 5, 8, 9, 18, 19, 41, 199, 200, 205, 206, 232, 238, 239, 312, 342, 343, 357, 424, 425, 429, 430, 440, 441, 445, 446, 462, 497, 503, 504 and 513.

Example 2

Bean plants (Phaseolus vulgaris) heavily infested with the glasshouse whitefly (Trialeurodes Vaporarium) were sprayed with aqueous dilutions of emulsion concentrates (1,000 ppm active compound content) until the onset of dripping. After placing the plants in a greenhouse, microscopic examination took place after 14 days, with the result in each case of 100% mortality in the preparations containing the active compounds of Examples 2, 8, 9, 18, 206, 239, 424, 425, 429, 430, 440, 441, 445, 446 and 503.

Example 3

Test procedure:	analogous to Example 2
Test animal:	Tetranychus urticae (glasshouse red spider mite)
Test plants:	Phaseolus vulgaris (dwarf bean)
Application rate:	1,000 ppm active compound in the spraying liquor

The activity determined after 8 days resulted in 100% mortality for the compounds 425, 429, 430, 445 and 446.

Example 4

Bean plants (Phaseolus vulgaris) heavily infested by the citrus mealybug (Pseudococcus citri) were sprayed with aqeuous dilutions of emulsion concentrates (in each case 1,000 ppm active compound in the spraying liquor) up to the stage of the onset of dripping.

After a standing time of 7 days in a greenhouse, examination took place at 20-25°C.

100% mortality was established for the compounds according to Examples 2, 5, 8, 9, 18, 19, 41, 200, 205, 206, 238, 342, 424, 429, 430, 446, 503 and 504.

Example 5

Large milkweed bugs (Oncopeltus fasciatus) were treated with aqueous dilutions of emulsion concentrates (in each case 1,000 ppm of active compound in the spraying liquor) of active compounds from Examples 2, 9, 19, 206, 425, 430, 441, 446 and 504. The bugs were subsequently placed in containers provided with air-permeable lids at room temperature.

The mortality was determined 5 days after the treatment and amounted to 100% in each individual case.

Example 6

The bottom insides of Petri dishes coated with synthetic nutrient were sprayed with 3 ml in each case of an aqueous emulsion containing 2,000 ppm of active compound after solidification of the feed paste. After drying of the spray coating and introduction of 10 larvae of the common cotton worm (Prodenia litura), the dishes were stored for 7 days at 21°C and the degree of action of the respective compound was then determined (expressed in % mortality). The compounds 1, 2, 5, 8, 9, 18,

19, 41, 200, 205, 206, 239, 424, 425, 430, 445, 446, 503 and 504 produced an activity of 100% in each case in this test.

Example 7

Bean leaves (Phaseolus vulgaris) were treated with an aqueous emulsion of the compound from Example 1 in a concentration of 1,000 ppm (with respect to active compound) and placed in observation cages with identically treated larvae of the Mexican bean beetle (Epilachna varivestis). An analysis after 48 hours showed 100% destruction of the test animals. The compounds according to Examples 2, 8, 9, 18, 19, 41, 200, 206, 239, 342, 424, 425, 429, 430, 441, 445, 446, 497, 503 and 504 proved to be equally active.

Example 8

1 ml of the active compound from Example 1 in acetone having a concentration of 1,000 ppm was uniformly applied to the inner side of the lid and bottom of a Petri dish by means of a pipette and the dishes were left open until the solvent had completely evaporated. 10 house flies (Musca domestica) each were then placed in the Petri dishes, the dishes were closed with the lids and after 3 hours a 100% destruction of the test animals was determined. The compounds according to Examples 2, 8, 9, 18, 19, 41, 200, 206, 238, 312, 424, 425, 429, 430, 440, 441, 445, 446, 503, 504 and 513 proved to be equally active.

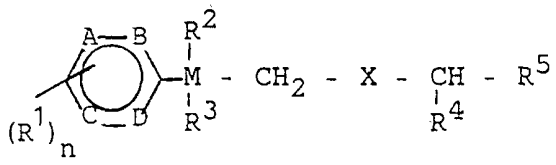
Example 9

1 ml in each case of an active compound solution in acetone having a concentration of 2,000 ppm was uniformly applied to the inner side of the lid and the bottom of a Petri dish with the aid of a pipette. After complete evaporation of the solvent, 10 larvae (L4) of the

German cockroach (Blatella germanica) were placed in each Petri dish and the dishes were closed with lids. After 72 hours, the action (expressed in % mortality) was determined. The compounds 1, 2, 8, 9, 18, 19, 41, 200, 206, 239, 312, 424, 425, 429, 430, 441, 446, 503, 504 and 513 showed activity of 100% in each case in this test.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A compound of the formula 1 and its stereoisomers



(I)

wherein

A, B, C or D independently of one another denote CH or N, in which at least one of the symbols A, B, C or D must correspond to a nitrogen atom,

M denotes a carbon ~~or silicon~~ atom,

X denotes CH_2 or oxygen,

R^1 denotes a radical bonded to a carbon atom from the series consisting of H, (C_1-C_4)alkyl, halogen, (C_2-C_6)-alkenyl, (C_2-C_6)alkynyl, (C_3-C_7)cycloalkyl, phenyl, phenoxy, $((C_1-C_5)$alkoxy, (C_2-C_4)alkenyloxy, (C_2-C_4)-alkynyloxy, (C_1-C_4)alkylthio, (C_3-C_7)cycloalkyloxy, (C_1-C_4)halogenoalkyl, (C_1-C_4)alkoxy-(C_1-C_4)alkyl, (C_1-C_3)halogenoalkoxy, (C_1-C_3)halogenoalkylthio, halogeno-(C_1-C_4)alkoxy-(C_1-C_4)alkyl, (C_1-C_4)alkylthio-(C_1-C_4)alkyl, (C_1-C_4)alkoxy-(C_1-C_4)alkoxy, halogeno-(C_1-C_4)alkoxy-(C_1-C_4)alkoxy, (C_2-C_4)alkenyloxy-(C_1-C_4)-alkoxy, halogeno-(C_2-C_4)alkenyloxy, (C_1-C_4)alkoxy-(C_1-C_4)-alkylthio, (C_1-C_4)alkylthio-(C_1-C_4)alkoxy, (C_1-C_4)alkylthio-(C_1-C_4)alkylthio,

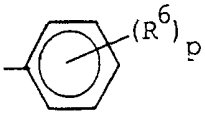
R^2 and R^3 independently of one another denote (C_1-C_3)-alkyl, (C_2-C_8)alkenyl or phenyl, or R^2 and R^3 denote an alkylene chain which - together with the quaternary

central atom (M) - forms an unsubstituted or fluorine substituted ring having three ring members (for M = C),

R^4 denotes -H, F, $-CCl_3$, $-C\equiv CH$, (C_1-C_4)alkyl,

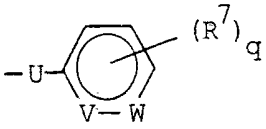
$-\overset{\overset{S}{\|}}{C}-NH_2$,

R^5 denotes a phenyl radical of the formula (A)



(A)

wherein R^6 independently of one another denote H, halogen, (C_1-C_4)alkyl, (C_1-C_4)alkoxy or (C_1-C_4)halogenoalkyl, phenyl or N-pyrrolyl or a radical of the formula (B)



(B)

wherein R^7 independently of one another denote H, halogen, (C_1-C_4)alkyl, (C_1-C_4)alkoxy or (C_1-C_4)halogenoalkyl,

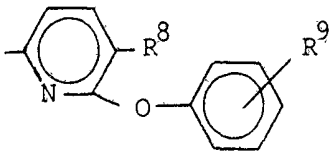
U denotes $\equiv CH_2-$, $>C=O$, -O- or -S-,

V or W denotes CH or N, in which W must denote CH where V denotes N and vice versa,

p denotes an integer from 1 to 5,

q denotes 1 or 2, or

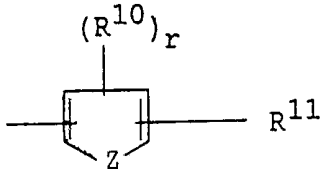
a mono- or disubstituted pyridyl group of the formula (C)



(C)

wherein R^8 denotes halogen or H and

R^9 denotes hydrogen or halogen with the exception of iodine, (C_1-C_4)alkyl, (C_1-C_4)alkoxy or (C_1-C_4) halogenoalkyl, or a substituted thienyl of the formula (D)



(D)

wherein Z denotes S,

R^{10} denotes H, halogen, (C_1-C_4)alkyl, (C_1-C_4)alkoxy, (C_1-C_4)halogenoalkyl, CN or NO_2,

R^{11} denotes benzyl, propargyl, allyl or phenoxy,

r denotes 1 or 2, and

n denotes 0, 1 or 2.

~~$=C-NH_2$,~~

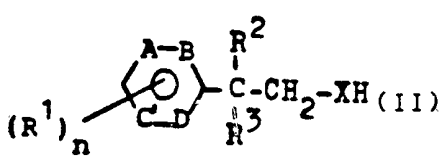
~~-S-~~

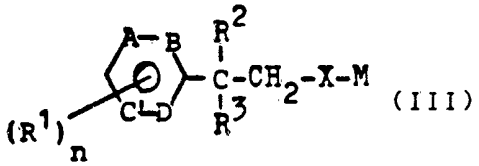
~~R^5 denotes pyridyl, furyl or thienyl which can all be~~ substituted, phthalimidyl, di(C_1-C_4)-alkylmaleimidyl, thiophthalimidyl, dihydrophthalimidyl, tetrahydrophthalimidyl or substituted phenyl,

or R^4 and R^5 - together with the carbon atom bridging them - denote an optionally substituted indanyl, cyclopentenoyl or cyclopentenyl radical and n denotes 0, 1 ~~or 2~~.

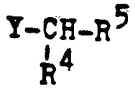
2. A process for the preparation of the compounds of the formula I wherein

a) for compounds having X = O a compound of the formula (II) or (III)


(II)


(III)

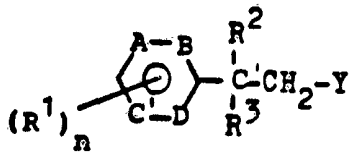
- is condensed with an alkylating agent of the formula IV


(IV)

wherein Y denotes a nucleofugic leaving group such as, for example, halogen or sulfonate and M corresponds to an alkali metal equivalent or an alkaline earth metal equivalent, in particular Li, Na, K or Mg, if appropriate in the presence of a base,

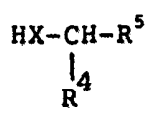
or

b) for compounds having X = O a compound of the formula (V)


(V)

is condensed with an XH-acid compound of the type (VI)

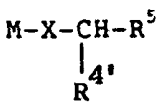
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(VI)

in the presence of a base

or with an organometallic compound of the type VII

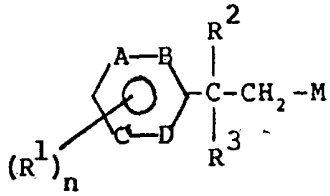


(VII)

having $R^{4'}$ = H or (C_1-C_4) alkyl

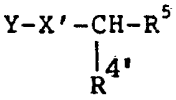
wherein Y and M have the meanings defined in claim 2a, or

c) for compounds having X = CH_2 a compound of the formula VIII



(VIII)

is condensed with a compound of the type (IX)

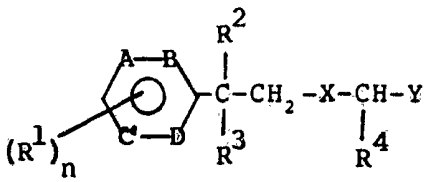


(IX)

having X' = CH_2,

wherein Y and M have the meanings defined in claim 2a, or

d) a compound of the formula (X)



(X)

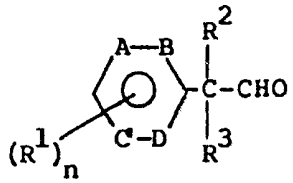
is condensed with an organometallic reagent of the type (XI)

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$M-R^5$

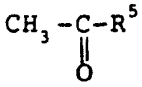
wherein Y and M have the meanings defined in claim 2a, or

e) for compounds having R^4 = H and X - CH_2 an aldehyde of the formula (XII)



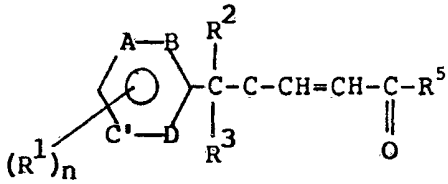
(XII)

is condensed with a ketone of the formula (XIII)



(XIII)

to form an intermediate (XIV)

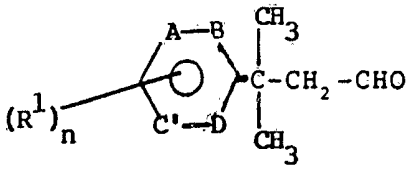


(XIV)

and this is subsequently reduced in a customary manner,

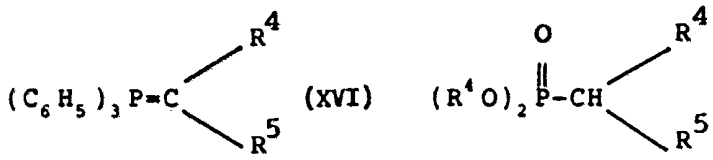
or

f) for compounds having X = CH_2 an aldehyde of the formula (XV)



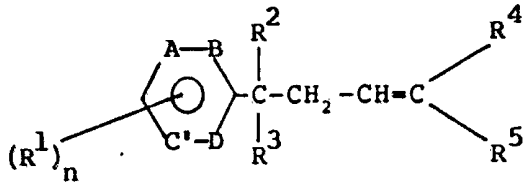
(XV)

is condensed with an ylide of the formula (XVI) or a phosphonate of the formula (XVII)



(XVI) (XVII)

to form an intermediate (XVIII)



(XVIII)

and this is subsequently reacted with a reductant to form the final product (I) having X = CH_2, the reaction temperature in the abovementioned process variations being between -75°C and +105°C.

3. An insecticidal or acaricidal agent which contains a compound of the formula I in an active amount and customary formulation auxiliaries.

4. A process for combating insects or acarids which comprises treating them or their environment with an active amount of a compound of the formula I.

DATED this 7th day of November, 1990.

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