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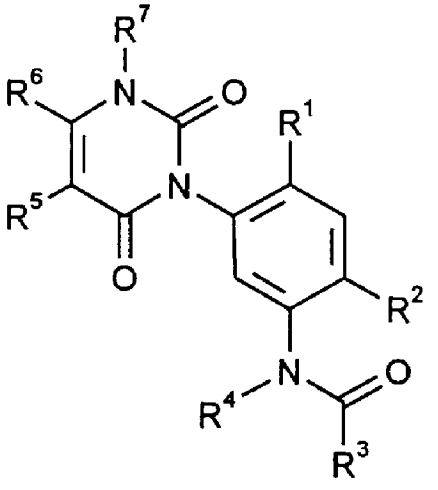
(54) **CARBONYLAMINOPHENYLURACILES SUBSTITUES**
(54) **SUBSTITUTED CARBONYLAMINOPHENYLURACILS**



(I)

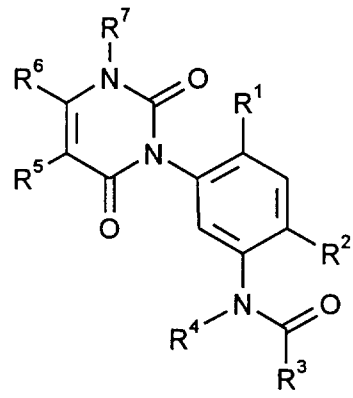
(57) L'invention concerne de nouveaux carbonylaminophényluraciles de la formule générale (I) où R^1, R^2, R^3, R^4, R^5, R^6 et R^7 ont la signification donnée dans la description, leur procédé de production ainsi que leur utilisation comme herbicides.

(57) The invention concerns novel substituted carbonylaminophenyluracils of general formula (I) in which R^1, R^2, R^3, R^4, R^5, R^6 and R^7 have the meanings given in the description. The invention further concerns a method of preparing these compounds and their use as herbicides.

Substituted carbonylaminophenyluracils

Abstract

The invention relates to novel substituted carbonylaminophenyluracils of the general formula (I)



(I)

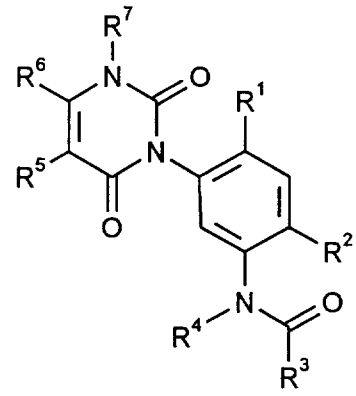
in which R^1, R^2, R^3, R^4, R^5, R^6 and R^7 are each as defined in the description, to the preparation thereof and to their use as herbicides.

Substituted carbonylaminophenyluracils

The invention relates to novel substituted carbonylaminophenyluracils, to the preparation thereof and to their use as herbicides.

Certain substituted aminophenyluracils are known to have herbicidal properties (cf. EP 408 382 / US 5 084 084 / US 5 127 935 / US 5 154 755, EP 563 384 / US 5 356 863, EP 648 749). However, these compounds have hitherto not achieved any significance.

This invention, accordingly, provides the novel substituted carbonylaminophenyluracils of the general formula (I)



(I)

in which

R^1 represents hydrogen, cyano or halogen,

R^2 represents cyano or halogen,

R^3 represents respectively optionally substituted cycloalkyl, cycloalkylalkyl, aryl, arylalkyl, heterocyclyl or heterocyclylalkyl,

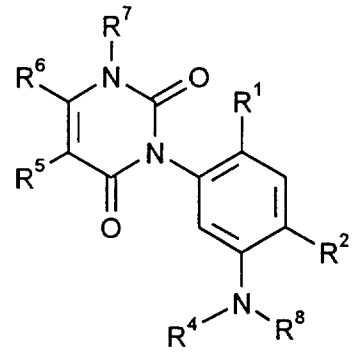
R^4 represents hydrogen or represents respectively optionally substituted alkyl, alkenyl, alkinyl, cycloalkyl, cycloalkyl, aryl, arylalkyl, heterocyclyl or heterocyclylalkyl or represents the grouping -CO-R^3 in which R^3 is as defined above,

R^5 represents hydrogen, halogen or represents respectively optionally substituted alkyl or alkoxy,

R^6 represents optionally substituted alkyl and

R^7 represents hydrogen or represents respectively optionally substituted alkyl, alkoxy, alkenyl or alkinyl.

The novel substituted aminophenyluracils of the general formula (I) are obtained when appropriate aminophenyl-uracils of the general formula (II)



(II)

in which

R^1, R^2, R^4, R^5, R^6 and R^7 are each as defined above and

R^8 represents hydrogen, trifluoroacetyl or alkylsulphonyl,

are reacted with acid derivatives of the general formula (III)

$$R^3\text{-CO-X} \qquad \text{(III)}$$

in which

R^3 is as defined above and

X represents halogen or the grouping -O-CO-R^3,

if appropriate in the presence of a reaction auxiliary and if appropriate in the presence of a diluent.

The novel substituted carbonylaminophenyluracils of the general formula (I) have strong herbicidal activity. The starting materials of the formula (II) are also herbicidal to a certain extent.

In the definitions, the saturated or unsaturated hydrocarbon chains, such as alkyl, alkenyl or alkinyl, are in each case straight-chain or branched.

Halogen generally represents fluorine, chlorine, bromine or iodine, preferably fluorine, chlorine or bromine, in particular fluorine or chlorine.

The invention preferably provides compounds of the formula (I) in which

R^1 represents hydrogen, cyano, fluorine or chlorine,

R^2 represents cyano, fluorine, chlorine or bromine,

R^3 represents respectively optionally cyano-, fluorine-, chlorine-, bromine- or C_1-C_4-alkyl-substituted cycloalkyl or cycloalkylalkyl having 3 to 8 carbon atoms in the cycloalkyl moiety and optionally 1 to 4 carbon atoms in the alkyl moiety,

R^3 furthermore represents phenyl, naphthyl, benzyl, phenylethyl, furyl, tetrahydrofuryl, thienyl, tetrahydrothienyl, oxazolyl, isoxazolyl, pyrazolyl, pyridinyl or quinolinyl, each of which is optionally substituted by fluorine, chlorine, bromine, cyano, nitro, carboxy, carbamoyl, thiocarbamoyl, by C_1-C_4-alkyl, C_1-C_4-alkoxy, C_1-C_4-alkylthio, C_1-C_4-alkylsulphinyl or C_1-C_4-alkylsulphonyl (each of which is optionally substituted by fluorine and/or chlorine), by dimethylaminosulphonyl or diethylaminosulphonyl, by C_1-C_4-alkoxy-carbonyl (which is optionally substituted by fluorine, chlorine, bromine, cyano, methoxy or ethoxy), by phenyl, phenyloxy or phenylthio (each of which is optionally substituted by fluorine, chlorine, bromine, cyano, methyl, methoxy, trifluoromethyl and/or trifluoromethoxy),

R^4 represents hydrogen, represents respectively optionally cyano-, carboxy-, carbamoyl-, thiocarbamoyl-, halogen-, C_1-C_4-alkyl- or C_1-C_4-alkoxy-carbonyl-substituted alkyl, alkenyl or alkinyl having in each case up to 10 carbon atoms,

R^4 furthermore represents respectively optionally cyano-, fluorine-, chlorine-, bromine- or C_1-C_4-alkyl-substituted cycloalkyl or cycloalkylalkyl having 3 to 8 carbon atoms in the cycloalkyl moiety and optionally 1 to 4 carbon atoms in the alkyl moiety,

R^4 furthermore represents phenyl, naphthyl, benzyl, phenylethyl, furyl, tetrahydrofuryl, thienyl, tetrahydrothienyl, oxazolyl, isoxazolyl, pyrazolyl, pyridinyl or quinolinyl, each of which is optionally substituted by fluorine, chlorine, bromine, cyano, nitro, carboxy, carbamoyl, thiocarbamoyl, by C_1-C_4-alkyl, C_1-C_4-alkoxy, C_1-C_4-alkylthio, C_1-C_4-alkylsulphinyl or C_1-C_4-alkylsulphonyl (each of which is optionally substituted by fluorine and/or chlorine), by dimethylaminosulphonyl or diethylaminosulphonyl, by C_1-C_4-alkoxy-carbonyl (which is optionally substituted by fluorine, chlorine, bromine, cyano, methoxy or ethoxy), by phenyl, phenyloxy or phenylthio (each of which is optionally substituted by fluorine, chlorine, bromine, cyano, methyl, methoxy, trifluoromethyl and/or trifluoromethoxy), or represents the grouping -CO-R^3 in which R^3 has the preferred meaning indicated above,

R^5 represents hydrogen, fluorine, chlorine, bromine or represents respectively optionally fluorine- and/or chlorine-substituted alkyl or alkoxy having in each case 1 to 4 carbon atoms,

R^6 represents optionally fluorine- and/or chlorine-substituted alkyl having 1 to 4 carbon atoms and

R^7 represents hydrogen or represents respectively optionally cyano-, fluorine-, chlorine- or C_1-C_4-alkoxy-substituted alkyl, alkoxy, alkenyl or alkinyl having in each case up to 6 carbon atoms.

The invention in particular provides compounds of the formula (I) in which

R^1 represents hydrogen, fluorine or chlorine,

R^2 represents cyano, fluorine, chlorine or bromine,

R^3 represents respectively optionally cyano-, fluorine-, chlorine-, bromine-, methyl-, ethyl-, n- or i-propyl-substituted cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cyclopropylmethyl, cyclobutylmethyl, cyclopentylmethyl or cyclohexylmethyl,

R^3 furthermore represents phenyl, naphthyl, benzyl, phenylethyl, furyl, tetrahydrofuryl, thienyl, tetrahydrothienyl, oxazolyl, isoxazolyl, pyrazolyl, pyridinyl or quinolinyl, each of which is optionally substituted by fluorine, chlorine, bromine, cyano, nitro, carboxy, carbamoyl, thiocarbamoyl, methyl, ethyl, n- or i-propyl, trifluoromethyl, methoxy, ethoxy, n- or i-propoxy, difluoromethoxy, trifluoromethoxy, methylthio, ethylthio, methylsulphinyl, ethylsulphinyl, methylsulphonyl or ethylsulphonyl, by dimethylaminosulphonyl or diethylaminosulphonyl, by methoxycarbonyl, ethoxycarbonyl, n- or i-propoxycarbonyl, by phenyl, phenyloxy or phenylthio,

R^4 represents hydrogen, represents respectively optionally cyano-, carboxy-, carbamoyl-, thiocarbamoyl-, halogen-, C_1-C_4-alkyl- or C_1-C_4-alkoxycarbonyl-substituted methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, n-, i-, s- or t-pentyl, propenyl, butenyl, pentenyl, propinyl, butinyl or pentinyl,

R^4 furthermore represents respectively optionally cyano-, fluorine-, chlorine-, bromine-, methyl-, ethyl-, n- or i-propyl-substituted cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cyclopropylmethyl, cyclobutylmethyl, cyclopentylmethyl or cyclohexylmethyl,

R^4 furthermore represents respectively optionally fluorine-, chlorine-, bromine-, cyano-, nitro-, carboxy-, carbamoyl-, thiocarbamoyl-, methyl-, ethyl-, n- or i-propyl-, trifluoromethyl-, methoxy-, ethoxy-, n- or i-propoxy-, methylthio-, ethylthio-, methylsulphinyl-, ethylsulphinyl-, methylsulphonyl- or ethylsulphonyl-, dimethylaminosulphonyl- or diethylaminosulphonyl-, methoxycarbonyl-, ethoxycarbonyl-, n- or i-propoxycarbonyl-, phenyl-, phenyloxy- or phenylthio-substituted phenyl, naphthyl, benzyl, phenylethyl, furyl, tetrahydrofuryl, thienyl, tetrahydrothienyl, oxazolyl, isoxazolyl,

pyrazolyl, pyridinyl or quinolinyl, or represents the grouping -CO-R^3 in which R^3 has the meaning indicated above as particularly preferred,

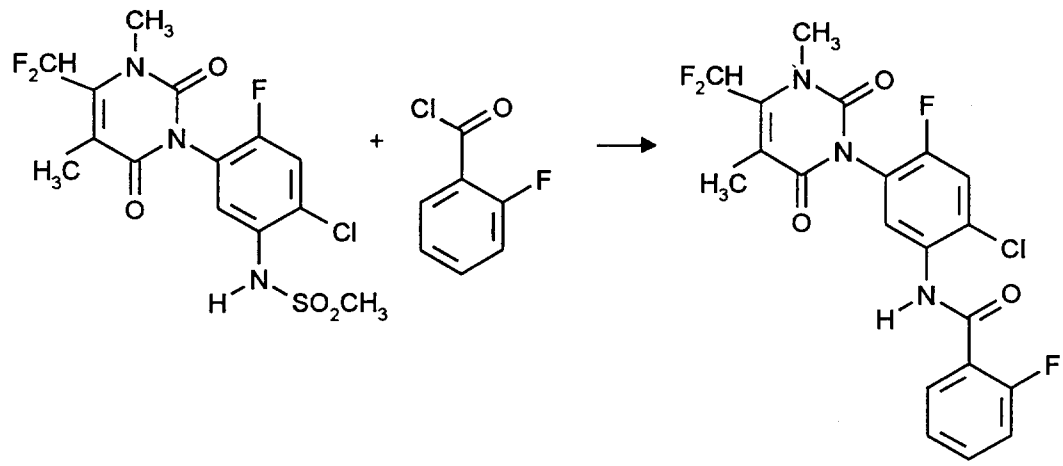
R^5 represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl, methoxy or ethoxy,

R^6 represents methyl, ethyl, difluoromethyl, dichloromethyl, trifluoromethyl, trichloromethyl, chlorodifluoromethyl, fluorodichloromethyl, fluoroethyl, chloroethyl, difluoroethyl, dichloroethyl, chlorofluoroethyl, trifluoroethyl, trichloroethyl, chlorodifluoroethyl, fluorodichloroethyl, tetrafluoroethyl, chlorotrifluoroethyl or pentafluoroethyl and

R^7 represents hydrogen or represents respectively optionally cyano-, fluorine-, chlorine-, methoxy- or ethoxy-substituted methyl, ethyl, n- or i-propyl, n-, i- or s-butyl, methoxy, ethoxy, n- or i-propoxy, n-, i- or s-butoxy, propenyl, butenyl, propinyl or butinyl.

The radical definitions listed above, whether general or listed in ranges of preference, apply not only to the end products of the formula (I) but also, correspondingly, to the starting materials and/or intermediates required in each case for the preparation. These radical definitions can be combined as desired with one another, thus including combinations between the preferred ranges indicated.

Using, for example, 1-(4-chloro-2-fluoro-5-methylsulphonylamino-phenyl)-3,6-dihydro-2,6-dioxo-3,5-dimethyl-4-difluoromethyl-1(2H)-pyrimidine and 2-fluoro-benzoyl chloride as starting materials, the course of the reaction in the process according to the invention can be illustrated by the following equation:



Formula (II) provides a general definition of the aminophenyluracils to be used as starting materials in the process according to the invention for preparing the compounds of the formula (I). In this formula (II), R^1, R^2, R^4, R^5, R^6 and R^7 each preferably or in particular have that meaning which has already been indicated above, in the description of the compounds of the formula (I) to be prepared according to the invention, as preferred or particularly preferred for R^1, R^2, R^4, R^5, R^6 and R^7; R^8 preferably represents hydrogen, trifluoroacetyl or C_1-C_4-alkyl-sulphonyl.

The starting materials of the formula (II) are known and/or can be prepared by known processes (cf. EP 408 382, EP 648 749, Preparation Examples).

Formula (III) provides a general definition of the acid derivatives further to be used as starting materials in the process according to the invention for preparing the compounds of the formula (I). In this formula (III), R^3 preferably or in particular has that meaning which has already been indicated above, in the description of the compounds of the formula (I) to be prepared according to the invention, as preferred or as particularly preferred for R^3; X preferably represents fluorine, chlorine or bromine, in particular fluorine.

The starting materials of the formula (III) are known chemicals for synthesis.

The process according to the invention for preparing compounds of the formula (I) is preferably carried out in the presence of a suitable reaction auxiliary. Suitable reaction auxiliaries are generally the customary inorganic or organic bases or acid acceptors. These preferably include alkali metal or alkaline earth metal acetates,

amides, carbonates, bicarbonates, hydrides, hydroxides or alkoxides, such as, for example, sodium acetate, potassium acetate or calcium acetate, lithium amide, sodium amide, potassium amide or calcium amide, sodium carbonate, potassium carbonate or calcium carbonate, sodium bicarbonate, potassium bicarbonate, or calcium bicarbonate, lithium hydride, sodium hydride, potassium hydride or calcium hydride, lithium hydroxide, sodium hydroxide, potassium hydroxide or calcium hydroxide, sodium methoxide or potassium methoxide, sodium ethoxide or potassium ethoxide, sodium n- or i-propoxide or potassium n- or i-propoxide, sodium n-, i-, s- or t-butoxide or potassium n-, i-, s- or t-butoxide; furthermore also basic organic nitrogen compounds, such as, for example trimethylamine, triethylamine, tripropylamine, tributylamine, ethyl-diisopropylamine, N,N-dimethyl-cyclohexylamine, dicyclohexylamine, ethyl-dicyclohexylamine, N,N-dimethyl-aniline, N,N-dimethyl-benzylamine, pyridine, 2-methyl-, 3-methyl-, 4-methyl-, 2,4-dimethyl-, 2,6-dimethyl-, 3,4-dimethyl- and 3,5-dimethyl-pyridine, 5-ethyl-2-methyl-pyridine, 4-dimethylamino-pyridine, N-methyl-piperidine, 1,4-diazabicyclo[2,2,2]-octane (DABCO), 1,5-diazabicyclo[4,3,0]-non-5-ene (DBN), and 1,8-diazabicyclo[5,4,0]-undec-7-ene (DBU).

The process according to the invention for preparing compounds of the formula (I) is preferably carried out in the presence of a diluent. Suitable diluents are in general the customary organic solvents. These preferably include aliphatic, alicyclic and aromatic, optionally halogenated hydrocarbons, such as, for example, pentane, hexane, heptane, petroleum ether, ligroin, benzine, benzene, toluene, xylene, chlorobenzene, dichlorobenzene, cyclohexane, methylcyclohexane, dichloromethane (methylene chloride), trichloromethane (chloroform) or carbon tetrachloride, dialkyl ethers, such as, for example, diethyl ether, diisopropyl ether, methyl t-butyl ether (MTBE), ethyl t-butyl ether, methyl t-pentyl ether (TAME), ethyl t-pentyl ether, tetrahydrofuran (THF), 1,4-dioxane, ethylene glycol dimethyl ether or ethylene glycol diethyl ether, diethylene glycol dimethyl ether or diethylene glycol diethyl ether; dialkyl ketones, such as, for example, acetone, butanone (methyl ethyl ketone), methyl i-propyl ketone or methyl i-butyl ketone, nitriles, such as, for example, acetonitrile, propionitrile, butyronitrile or benzonitrile; amides, such as, for example, N,N-dimethyl-formamide (DMF), N,N-dimethyl-acetamide, N-methyl-formanilide, N-methyl-pyrrolidone or hexamethyl-phosphoric triamide; esters, such as, for example, methyl acetate, ethyl acetate, n- or i-propyl acetate, n-, i- or s-butyl acetate; sulphoxides, such as, for example, dimethylsulphoxide; alkanols, such as, for example, methanol, ethanol, n-

or i-propanol, n-, i-, s- or t-butanol, ethylene glycol monomethyl ether or ethylene glycol monoethyl ether, diethylene glycol monomethyl ether or diethylene glycol monoethyl ether; their mixtures with water or pure water.

In the practice of the process according to the invention, the reaction temperatures can be varied over a relatively wide range. Generally, the reaction is carried out at temperatures between 0°C and 150°C, preferably between 10°C and 120°C.

The process according to the invention is generally carried out at atmospheric pressure. However, it is also possible to carry out the process according to the invention under elevated or reduced pressure - generally between 0.1 bar and 10 bar.

In the practice of the process according to the invention, the starting materials are generally employed in approximately equimolar amounts. However, it is also possible to use a relatively large excess of one of the components. The reaction is generally carried out in a suitable diluent in the presence of a reaction auxiliary, and the reaction mixture is generally stirred for several hours at the temperature required. Work-up is carried out by conventional methods (cf. the Preparation Examples).

The active compounds according to the invention can be used as defoliants, desiccants, haulm killers and, especially, as weed-killers. By weeds in the broadest sense, there are to be understood all plants which grow in locations where they are undesirable. Whether the substances according to the invention act as total or selective herbicides depends essentially on the amount used. The active compounds according to the invention can be used, for example, in connection with the following plants:

Dicotyledonous weeds of the genera: Sinapis, Lepidium, Galium, Stellaria, Matricaria, Anthemis, Galinsoga, Chenopodium, Urtica, Senecio, Amaranthus, Portulaca, Xanthium, Convolvulus, Ipomoea, Polygonum, Sesbania, Ambrosia, Cirsium, Carduus, Sonchus, Solanum, Rorippa, Rotala, Lindernia, Lamium, Veronica, Abutilon, Emex, Datura, Viola, Galeopsis, Papaver, Centaurea, Trifolium, Ranunculus and Taraxacum.

Dicotyledonous crops of the genera: Gossypium, Glycine, Beta, Daucus, Phaseolus, Pisum, Solanum, Linum, Ipomoea, Vicia, Nicotiana, Lycopersicon, Arachis, Brassica, Lactuca, Cucumis and Cucurbita.

Monocotyledonous weeds of the genera: Echinochloa, Setaria, Panicum, Digitaria, Phleum, Poa, Festuca, Eleusine, Brachiaria, Lolium, Bromus, Avena, Cyperus, Sorghum, Agropyron, Cynodon, Monochoria, Fimbristylis, Sagittaria, Eleocharis, Scirpus, Paspalum, Ischaemum, Sphenoclea, Dactyloctenium, Agrostis, Alopecurus and Apera.

Monocotyledonous crops of the genera: Oryza, Zea, Triticum, Hordeum, Avena, Secale, Sorghum, Panicum, Saccharum, Ananas, Asparagus and Allium.

However, the use of the active compounds according to the invention is in no way restricted to these genera, but also extends in the same manner to other plants.

The active compounds according to the invention are suitable, depending on the concentration, for the total control of weeds, for example on industrial terrain and railway tracks, and on paths and squares with or without tree plantings. Equally, the compounds can be employed for controlling weeds in perennial cultures, for example forests, decorative tree plantings, orchards, vineyards, citrus groves, nut orchards, banana plantations, coffee plantations, tea plantations, rubber plantations, oil palm plantations, cocoa plantations, soft fruit plantings and hopfields, on lawns, turf and pasture-land, and for the selective control of weeds in annual cultures.

The active compounds of the formula (I) according to the invention are suitable in particular for selectively controlling monocotyledonous and dicotyledonous weeds in monocotyledonous and dicotyledonous crops, both pre-emergence and post-emergence.

The active compounds according to the invention can be converted into the customary formulations, such as solutions, emulsions, wettable powders, suspensions, powders, dusting agents, pastes, soluble powders, granules, suspo-emulsion concentrates, natural and synthetic materials impregnated with active compound, and very fine capsules in polymeric substances.

These formulations are produced in a known manner, for example by mixing the active compounds with extenders, that is liquid solvents and/or solid carriers, optionally with the use of surfactants, that is emulsifiers and/or dispersing agents and/or foam-forming agents.

In the case of the use of water as an extender, organic solvents can, for example, also be used as auxiliary solvents. Essentially, suitable liquid solvents are: aromatics, such as xylene, toluene or alkylnaphthalenes, chlorinated aromatics and chlorinated aliphatic hydrocarbons, such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic hydrocarbons, such as cyclohexane or paraffins, for example petroleum fractions, mineral and vegetable oils, alcohols, such as butanol or glycol as well as their ethers and esters, ketones, such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents, such as dimethylformamide and dimethyl sulphoxide, as well as water.

Suitable solid carriers are:
for example ammonium salts and ground natural minerals, such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals, such as finely divided silica, alumina and silicates, suitable solid carriers for granules are: for example crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, as well as synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, maize cobs and tobacco stalks; suitable emulsifiers and/or foam-forming agents are: for example nonionic and anionic emulsifiers, such as polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulphonates, alkyl sulphates, aryl-sulphonates as well as protein hydrolysates; suitable dispersing agents are: for example lignin-sulphite waste liquors and methylcellulose.

Tackifiers such as carboxymethylcellulose and natural and synthetic polymers in the form of powders, granules or latexes, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, as well as natural phospholipids, such as cephalins and lecithins, and synthetic phospholipids, can be used in the formulations. Further additives can be mineral and vegetable oils.

It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyes, such as alizarin dyes, azo

dyes and metal phthalocyanine dyes, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

The formulations in general contain between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.

For controlling weeds, the active compounds according to the invention, as such or in the form of their formulations, can also be used as mixtures with known herbicides, finished formulations or tank mixes being possible.

Possible components for the mixtures are known herbicides, for example anilides, such as diflufenican and propanil; arylcarboxylic acids, such as dichloropicolinic acid, dicamba and picloram; aryloxyalkanoic acids, such as 2,4-D, 2,4-DB, 2,4-DP, fluroxypyr, MCPA, MCPP and triclopyr; aryloxy-phenoxy-alkanoic esters, such as diclofop-methyl, fenoxaprop-ethyl, fluazifop-butyl, haloxyfop-methyl and quizalofop-ethyl; azinones, such as chloridazon and norflurazon; carbamates, such as chlorpropham, desmedipham, phenmedipham and propham; chloroacetanilides, such as alachlor, acetochlor, butachlor, metazachlor, metolachlor, pretilachlor and propachlor; dinitroanilines, such as oryzalin, pendimethalin and trifluralin; diphenyl ethers, such as acifluorfen, bifenox, fluoroglycofen, fomesafen, halosafen, lactofen and oxyfluorfen; ureas, such as chlorotoluron, diuron, fluometuron, isoproturon, linuron and methabenzthiazuron; hydroxylamines, such as alloxydim, clethodim, cycloxydim, sethoxydim and tralkoxydim; imidazolinones, such as imazethapyr, imazamethabenz, imazapyr and imazaquin; nitriles, such as bromoxynil, dichlobenil and ioxynil; oxyacetamides, such as mefenacet; sulphonylureas, such as amidosulfuron, bensulfuron-methyl, chlorimuron-ethyl, chlorsulfuron, cinosulfuron, metsulfuron-methyl, nicosulfuron, primisulfuron, pyrazosulfuron-ethyl, thifensulfuron-methyl, triasulfuron and tribenuron-methyl; thiocarbamates, such as butylate, cycloate, diallate, EPTC, esprocarb, molinate, prosulfocarb, thiobencarb and triallate; triazines, such as atrazine, cyanazine, simazine, simetryne, terbutryne and terbutylazine; triazinones, such as hexazinone, metamitron and metribuzin; and others, such as aminotriazole, benfuresate, bentazone, cinmethylin, clomazone, clopyralid, difenzoquat, dithiopyr, ethofumesate, fluorochloridone, glufosinate, glyphosate, isoxaben, pyridate, quinchlorac, quinmerac, sulphosate and tridiphane.

Mixtures with other known active compounds, such as fungicides, insecticides, acaricides, nematicides, bird repellents, plant nutrients and agents which improve soil structure, are also possible.

The active compounds can be used as such, in the form of their formulations or in the use forms prepared therefrom by further dilution, such as ready-to-use solutions, suspensions, emulsions, powders, pastes and granules. They are used in the customary manner, for example by watering, spraying, atomizing or scattering.

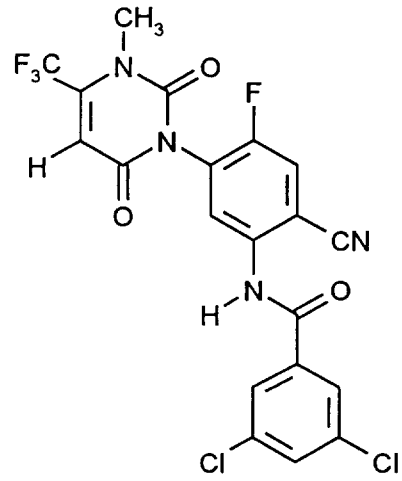
The active compounds according to the invention can be applied either before or after emergence of the plants. They can also be incorporated into the soil before sowing.

The amount of active compound used can vary within a substantial range. It depends essentially on the nature of the desired effect. In general, the amounts used are between 10 g and 10 kg of active compound per hectare of soil surface, preferably between 50 g and 5 kg per ha.

The preparation and use of the active compounds according to the invention can be seen from the examples below.

Preparation Examples:

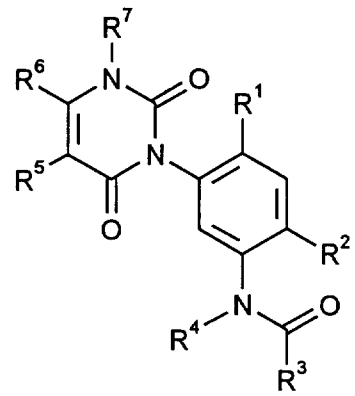
Example 1



With stirring, 1.3 g (6 mmol) of 3,5-dichloro-benzoyl chloride are added to a mixture of 2.1 g (5 mmol) of 1-(4-cyano-2-fluoro-5-ethylsulphonylamino-phenyl)-3,6-dihydro-2,6-dioxo-3-methyl-4-trifluoromethyl-1(2H)-pyrimidine, 1 ml of triethylamine and 50 ml of acetonitrile, and the reaction mixture is stirred at 20°C for 24 hours. The mixture is then concentrated using waterpump vacuum, the residue is shaken with 1N hydrochloric acid/ethyl acetate and the organic phase is separated off, dried using sodium sulphate and filtered. The filtrate is concentrated using waterpump vacuum, the residue is digested with diethyl ether/petroleum ether and the crystalline product is isolated by filtration with suction.

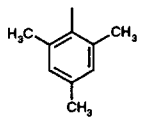
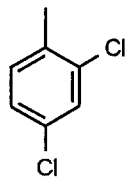
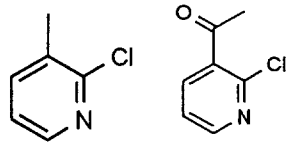
0.90 g (30% of theory) of 1-[4-cyano-2-fluoro-5-(3,5-dichloro-benzoylamino)-phenyl]-3,6-dihydro-2,6-dioxo-3-methyl-4-trifluoromethyl-1(2H)-pyrimidine of melting point 212°C is obtained.

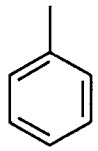
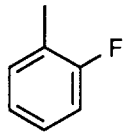
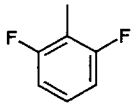
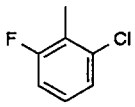
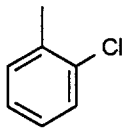
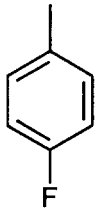
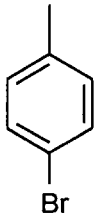
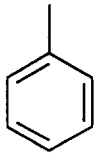
By the method of Example 1, and according to the general description of the process according to the invention, it is also possible to prepare, for example, the compounds of the formula (I) listed in Table 1 below.

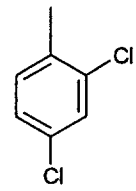
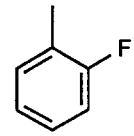
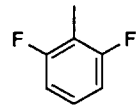
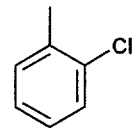
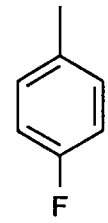
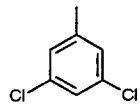
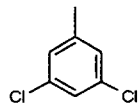
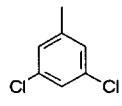
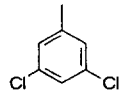


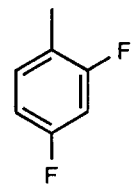
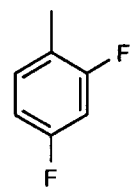
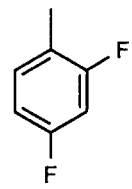
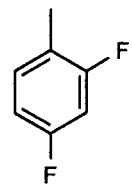
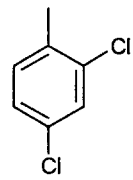
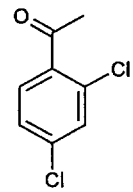
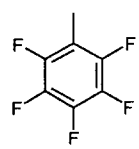
(I)

Table 1: Examples of compounds of the formula (I)

Ex. No.	R^1	R^2	R^3	R^4	R^5	R^6	R^7	Melting point (°C)
2	F	CN		H	H	CF_3	CH_3	155
3	F	CN		H	H	CF_3	CH_3	218
4	F	CN		H	H	CF_3	CH_3	136

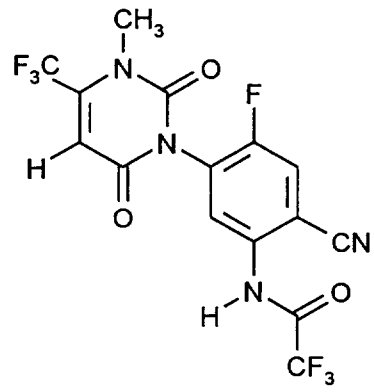
Ex. No.	R^1	R^2	R^3	R^4	R^5	R^6	R^7	Melting point (°C)
5	F	CN		H	H	CF_3	CH_3	
6	F	CN		H	H	CF_3	CH_3	
7	F	CN		H	H	CF_3	CH_3	
8	F	CN		H	H	CF_3	CH_3	
9	F	CN		H	H	CF_3	CH_3	
10	F	CN		H	H	CF_3	CH_3	
11	F	CN		H	H	CF_3	CH_3	
12	F	$CSNH_2$		H	H	CF_3	CH_3	

Ex. No.	R^1	R^2	R^3	R^4	R^5	R^6	R^7	Melting point (°C)
13	F	$CSNH_2$		H	H	CF_3	CH_3	
14	F	$CSNH_2$		H	H	CF_3	CH_3	
15	F	$CSNH_2$		H	H	CF_3	CH_3	
16	F	$CSNH_2$		H	H	CF_3	CH_3	
17	F	$CSNH_2$		H	H	CF_3	CH_3	
18	F	$CSNH_2$		H	H	CF_3	CH_3	
19	F	CN		H	H	CF_2Cl	CH_3	
20	F	CN		H	CH_3	CF_3	CH_3	
21	F	$CSNH_2$		H	CH_3	CF_3	CH_3	

Ex. No.	R^1	R^2	R^3	R^4	R^5	R^6	R^7	Melting point (°C)
22	F	CN		H	H	CF_3	CH_3	
23	F	CN		H	CH_3	CF_3	CH_3	
24	F	CN		H	H	CF_3	C_2H_5	
25	F	$CSNH_2$		H	H	CF_3	CH_3	
26	F	CN			H	CF_3	CH_3	127
27	F	CN		H	H	CF_3	CH_3	116

Starting materials of the formula (II):

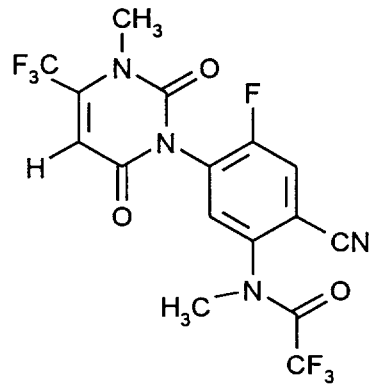
Example (II-1)



A mixture of 1.67 g (4 mmol) of 1-(4-cyano-2-fluoro-5-methylsulphonylamino-phenyl)-3,6-dihydro-2,6-dioxo-3-methyl-4-trifluoromethyl-1(2H)-pyrimidine, 1.05 g (5 mmol) of trifluoroacetic anhydride, 1.5 g of triethylamine and 20 ml of acetonitrile is stirred at 20°C for 30 minutes and then concentrated using water-pump vacuum. The residue is shaken with 1N hydrochloric acid/ethyl acetate and the organic phase is separated off, dried using sodium sulphate and filtered. The filtrate is concentrated using waterpump vacuum and the residue is worked up by column chromatography (silica gel, hexane/ethyl acetate, vol.: 4:1).

1.0 g (59% of theory) of 1-(4-cyano-2-fluoro-5-trifluoroacetylaminophenyl)-3,6-dihydro-2,6-dioxo-3-methyl-4-trifluoromethyl-1(2H)-pyrimidine of melting point 105°C is obtained.

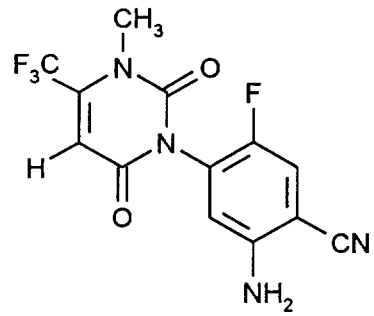
Example (II-2)



A mixture of 2.19 g (5 mmol) of 1-(4-cyano-2-fluoro-5-trifluoroacetylamino-phenyl)-3,6-dihydro-2,6-dioxo-3-methyl-4-trifluoromethyl-1(2H)-pyrimidine, 0.76 g (6 mmol) of dimethyl sulphate, 0.83 g of potassium carbonate and 50 ml of acetone is heated under reflux for 3 hours and then concentrated using waterpump vacuum. The residue is stirred with 1N hydrochloric acid/diethyl ether and the crystalline product is then isolated by filtration with suction.

1.4 g (64% of theory) of 1-[4-cyano-2-fluoro-5-(N-methyl-N-trifluoroacetylamino)-phenyl]-3,6-dihydro-2,6-dioxo-3-methyl-4-trifluoromethyl-1(2H)-pyrimidine of melting point 202°C are obtained.

Example (II-3)



With stirring, 0.17 g (1.2 mmol) of pivaloyl chloride is added to a mixture of 0.50 g (1.2 mmol) of 1-(4-cyano-2-fluoro-5-trifluoroacetylamino-phenyl)-3,6-dihydro-2,6-dioxo-3-methyl-4-trifluoromethyl-1(2H)-pyrimidine, 1 ml of triethyl-amine and 50 ml of acetonitrile, and the reaction mixture is stirred for 18 hours at 20°C and for a further 15 hours at 60°C. The mixture is then concentrated using waterpump vacuum, the residue is shaken with 1N hydrochloric acid/ethyl acetate and the organic phase is separated off, dried using sodium sulphate and filtered. The filtrate is concentrated using waterpump vacuum and the residue is worked up by column chromatography (silica gel, chloroform/ethyl acetate, vol.: 1:1).

In addition to unreacted 1-(4-cyano-2-fluoro-5-trifluoroacetylamino-phenyl)-3,6-dihydro-2,6-dioxo-3-methyl-4-trifluoromethyl-1(2H)-pyrimidine (first fraction: 0.30 g), 0.2 g (50% of theory) of 1-(4-cyano-2-fluoro-5-amino-phenyl)-3,6-dihydro-2,6-dioxo-3-methyl-4-trifluoromethyl-1(2H)-pyrimidine is obtained as second fraction. Melting point: 195°C.

The compound to be prepared according to Example (II-3) has not been disclosed in the literature; as a novel compound, it forms part of the subject matter of the present application.

Use Examples:

Example A

Pre-emergence test

Solvent:	5 parts by weight of acetone
Emulsifier:	1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, the stated amount of emulsifier is added and the concentrate is diluted with water to the desired concentration.

Seeds of the test plants are sown in normal soil and, after about 24 hours, watered with the preparation of the active compound. Advantageously, the amount of water per unit area is kept constant. The active compound concentration in the preparation is not important, only the active compound application rate per unit area being critical.

After three weeks, the degree of damage to the plants is rated in % damage in comparison with the development of the untreated control.

The figures denote:

0%	=	no effect (like untreated control)
100%	=	total destruction

In this test, for example, the compounds of Preparation Example 1, 2, 3, 4, (II-1) and (II-2) exhibit, at application rates between 125 g and 4,000 g, strong activity against weeds such as Alopecurus (70-95%), Avena (80-100%), Setaria (100%), Abutilon (100%), Amaranthus (95-100%) and Sinapis (100%).

Example B

Post-emergence test

Solvent: 5 parts by weight of acetone
Emulsifier: 1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, the stated amount of emulsifier is added and the concentrate is diluted with water to the desired concentration.

Test plants which have a height of 5-15 cm are sprayed with the preparation of the active compound in such a way as to apply the particular amounts of active compound desired per unit area.

After three weeks, the degree of damage to the plants is rated in % damage in comparison with the development of the untreated control.

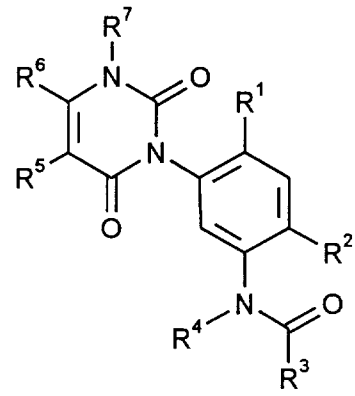
The figures denote:

0% = no effect (like untreated control)
100% = total destruction

In this test, for example, the compounds of Preparation Example 1, 2, 3, 4, (II-1) and (II-2) exhibit, at application rates of 125 g to 2,000 g, strong activity against weeds such as Abutilon (100%), Amaranthus (90-100%) and Sinapis (80-100%).

Patent Claims

1. Carbonylaminophenyluracils of the general formula (I)



(I)

in which

R^1 represents hydrogen, cyano or halogen,

R^2 represents cyano or halogen,

R^3 represents respectively optionally substituted cycloalkyl, cycloalkyl-alkyl, aryl, arylalkyl, heterocyclyl or heterocyclylalkyl,

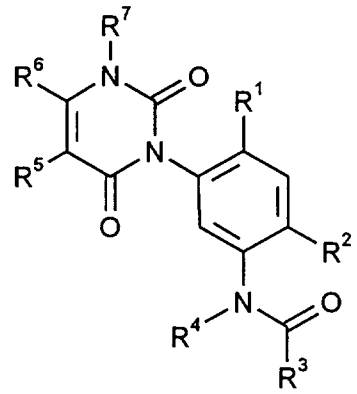
R^4 represents hydrogen or represents respectively optionally substituted alkyl, alkenyl, alkinyl, cycloalkyl, cycloalkyl, aryl, arylalkyl, heterocyclyl or heterocyclylalkyl or represents the grouping -CO-R^3 in which R^3 is as defined above,

R^5 represents hydrogen, halogen or represents respectively optionally substituted alkyl or alkoxy,

R^6 represents optionally substituted alkyl and

R^7 represents hydrogen or represents respectively optionally substituted alkyl, alkoxy, alkenyl or alkinyl.

2. Process for preparing aminophenyluracils of the general formula (I)

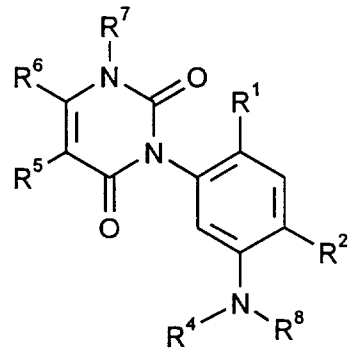


(I)

in which

R^1, R^2, R^3, R^4, R^5, R^6 and R^7 are each as defined in Claim 1,

characterized in that appropriate aminophenyl-uracils of the general formula (II)



(II)

in which

R^1, R^2, R^4, R^5, R^6 and R^7 are each as defined above and

R^8 represents hydrogen, trifluoroacetyl or alkylsulphonyl,

are reacted with acid derivatives of the general formula (III)

$$R^3\text{-CO-X} \qquad \text{(III)}$$

in which

R^3 is as defined above and

X represents halogen or the grouping -O-CO-R^3,

if appropriate in the presence of a reaction auxiliary and if appropriate in the presence of a diluent.

3. Carbonylaminophenyluracils of the general formula (I) according to Claim 1, characterized in that

R^1 represents hydrogen, cyano, fluorine or chlorine,

R^2 represents cyano, fluorine, chlorine or bromine,

R^3 represents respectively optionally cyano-, fluorine-, chlorine-, bromine- or C_1-C_4-alkyl-substituted cycloalkyl or cycloalkylalkyl having 3 to 8 carbon atoms in the cycloalkyl moiety and optionally 1 to 4 carbon atoms in the alkyl moiety,

R^3 furthermore represents phenyl, naphthyl, benzyl, phenylethyl, furyl, tetrahydrofuryl, thienyl, tetrahydrothienyl, oxazolyl, isoxazolyl, pyrazolyl, pyridinyl or quinolinyl, each of which is optionally substituted by fluorine, chlorine, bromine, cyano, nitro, carboxy, carbamoyl, thiocarbamoyl, by C_1-C_4-alkyl, C_1-C_4-alkoxy, C_1-C_4-alkylthio, C_1-C_4-alkylsulphinyl or C_1-C_4-alkylsulphonyl (each of which is optionally substituted by fluorine and/or chlorine), by dimethylaminosulphonyl or diethylaminosulphonyl, by C_1-C_4-alkoxy-carbonyl (which is optionally substituted by fluorine, chlorine, bromine, cyano, methoxy or ethoxy), by phenyl, phenyloxy or phenylthio (each of which is optionally substituted by fluorine, chlorine, bromine, cyano, methyl, methoxy, trifluoromethyl and/or trifluoromethoxy),

R^4 represents hydrogen, represents respectively optionally cyano-, carboxy-, carbamoyl-, thiocarbamoyl-, halogen-, C_1-C_4-alkyl- or C_1-C_4-alkoxy-carbonyl-substituted alkyl, alkenyl or alkinyl having in each case up to 10 carbon atoms,

R^4 furthermore represents respectively optionally cyano-, fluorine-, chlorine-, bromine- or C_1-C_4-alkyl-substituted cycloalkyl or cycloalkylalkyl having 3 to 8 carbon atoms in the cycloalkyl moiety and optionally 1 to 4 carbon atoms in the alkyl moiety,

R^4 furthermore represents phenyl, naphthyl, benzyl, phenylethyl, furyl, tetrahydrofuryl, thienyl, tetrahydrothienyl, oxazolyl, isoxazolyl, pyrazolyl, pyridinyl or quinolinyl, each of which is optionally substituted by fluorine, chlorine, bromine, cyano, nitro, carboxy, carbamoyl, thiocarbamoyl, by C_1-C_4-alkyl, C_1-C_4-alkoxy, C_1-C_4-alkylthio, C_1-C_4-alkylsulphinyl or C_1-C_4-alkylsulphonyl (each of which is optionally substituted by fluorine and/or chlorine), by dimethylaminosulphonyl or diethylaminosulphonyl, by C_1-C_4-alkoxy-carbonyl (which is optionally substituted by fluorine, chlorine, bromine, cyano, methoxy or ethoxy), by phenyl, phenyloxy or phenylthio (each of which is optionally substituted by fluorine, chlorine, bromine, cyano, methyl, methoxy, trifluoromethyl and/or trifluoromethoxy), or represents the grouping -CO-R^3 in which R^3 has the preferred meaning indicated above,

R^5 represents hydrogen, fluorine, chlorine, bromine or represents respectively optionally fluorine- and/or chlorine-substituted alkyl or alkoxy having in each case 1 to 4 carbon atoms,

R^6 represents optionally fluorine- and/or chlorine-substituted alkyl having 1 to 4 carbon atoms and

R^7 represents hydrogen or represents respectively optionally cyano-, fluorine-, chlorine- or C_1-C_4-alkoxy-substituted alkyl, alkoxy, alkenyl or alkinyl having in each case up to 6 carbon atoms.

4. Carbonylaminophenyluracils of the general formula (I) according to Claim 1, characterized in that

R^1 represents hydrogen, fluorine or chlorine,

R^2 represents cyano, fluorine, chlorine or bromine,

R^3 represents respectively optionally cyano-, fluorine-, chlorine-, bromine-, methyl-, ethyl-, n- or i-propyl-substituted cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cyclopropylmethyl, cyclobutylmethyl, cyclopentylmethyl or cyclohexylmethyl,

R^3 furthermore represents phenyl, naphthyl, benzyl, phenylethyl, furyl, tetrahydrofuryl, thienyl, tetrahydrothienyl, oxazolyl, isoxazolyl, pyrazolyl, pyridinyl or quinolinyl, each of which is optionally substituted by fluorine, chlorine, bromine, cyano, nitro, carboxy, carbamoyl, thiocarbamoyl, methyl, ethyl, n- or i-propyl, trifluoromethyl, methoxy, ethoxy, n- or i-propoxy, difluoromethoxy, trifluoromethoxy, methylthio, ethylthio, methylsulphinyl, ethylsulphinyl, methylsulphonyl or ethylsulphonyl, by dimethylaminosulphonyl or diethylaminosulphonyl, by methoxycarbonyl, ethoxycarbonyl, n- or i-propoxycarbonyl, by phenyl, phenyloxy or phenylthio,

R^4 represents hydrogen, represents respectively optionally cyano-, carboxy-, carbamoyl-, thiocarbamoyl-, halogen-, C_1-C_4-alkyl- or C_1-C_4-alkoxy-carbonyl-substituted methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, n-, i-, s- or t-pentyl, propenyl, butenyl, pentenyl, propinyl, butinyl or pentinyl,

R^4 furthermore represents respectively optionally cyano-, fluorine-, chlorine-, bromine-, methyl-, ethyl-, n- or i-propyl-substituted cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cyclopropylmethyl, cyclobutylmethyl, cyclopentylmethyl or cyclohexylmethyl,

R^4 furthermore represents respectively optionally fluorine-, chlorine-, bromine-, cyano-, nitro-, carboxy-, carbamoyl-, thiocarbamoyl-, methyl-, ethyl-, n- or i-propyl-, trifluoromethyl-, methoxy-, ethoxy-, n- or i-propoxy-, methylthio-, ethylthio-, methylsulphinyl-, ethylsulphinyl-, methylsulphonyl- or ethylsulphonyl-, dimethylaminosulphonyl- or diethylaminosulphonyl-, methoxycarbonyl-, ethoxycarbonyl-, n- or i-propoxycarbonyl-, phenyl-, phenyloxy- or phenylthio-substituted phenyl, naphthyl, benzyl, phenylethyl, furyl, tetrahydrofuryl, thienyl, tetrahydrothienyl, oxazolyl, isoxazolyl,

pyrazolyl, pyridinyl or quinolinyl, or represents the grouping $-CO-R^3$ in which R^3 has the meaning indicated above as particularly preferred,

R^5 represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl, methoxy or ethoxy,

R^6 represents methyl, ethyl, difluoromethyl, dichloromethyl, trifluoromethyl, trichloromethyl, chlorodifluoromethyl, fluorodichloromethyl, fluoroethyl, chloroethyl, difluoroethyl, dichloroethyl, chlorofluoroethyl, trifluoroethyl, trichloroethyl, chlorodifluoroethyl, fluorodichloroethyl, tetrafluoroethyl, chlorotrifluoroethyl or pentafluoroethyl and

R^7 represents hydrogen or represents respectively optionally cyano-, fluorine-, chlorine-, methoxy- or ethoxy-substituted methyl, ethyl, n- or i-propyl, n-, i- or s-butyl, methoxy, ethoxy, n- or i-propoxy, n-, i- or s-butoxy, propenyl, butenyl, propinyl or butinyl.

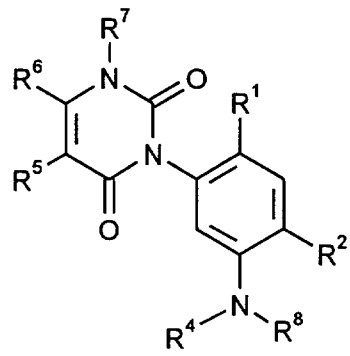
5. Herbicides, characterized by a content of at least one carbonylaminophenyluracil of the general formula (I) according to Claims 1 to 4.

6. Method for controlling undesirable plants, characterized in that carbonylaminophenyluracils of the general formula (I) according to Claims 1 to 4 are allowed to act on undesirable plants and/or their habitat.

7. Use of carbonylaminophenyluracils of the general formula (I) according to Claims 1 to 4 for controlling undesirable plants.

8. Process for preparing herbicides, characterized in that carbonylaminophenyluracils of the general formula (I) according to Claims 1 to 4 are mixed with extenders and/or surfactants.

9. Aminophenyluracil of the general formula (II)



(II)

in which

R^1 represents fluorine,

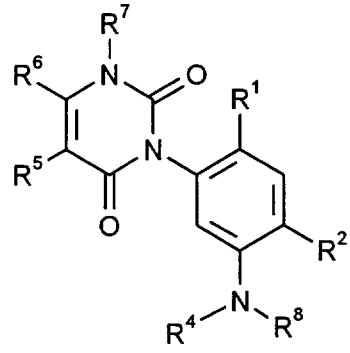
R^2 represents cyano,

R^4, R^5 and R^8 each represent hydrogen,

R^6 represents trifluoromethyl and

R^7 represents methyl.

10. Use of aminophenyluracils of the general formula (II)



(II)

in which

R^1 represents hydrogen, cyano or halogen,

R^2 represents cyano or halogen,

R^3 represents respectively optionally substituted cycloalkyl, cycloalkylalkyl, aryl, arylalkyl, heterocyclyl or heterocyclylalkyl,

R^4 represents hydrogen or represents respectively optionally substituted alkyl, alkenyl, alkinyl, cycloalkyl, cycloalkyl, aryl, arylalkyl, heterocyclyl or heterocyclylalkyl or represents the grouping -CO-R^3 in which R^3 is as defined above,

R^5 represents hydrogen, halogen or represents respectively optionally substituted alkyl or alkoxy,

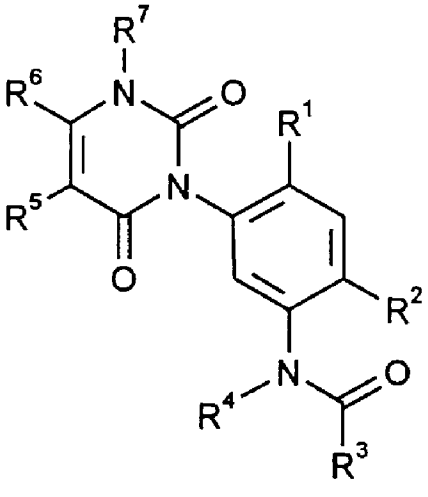
R^6 represents optionally substituted alkyl and

R^7 represents hydrogen or represents respectively optionally substituted alkyl, alkoxy, alkenyl or alkinyl, and

R^8 represents hydrogen, trifluoroacetyl or alkylsulphonyl

for controlling undesirable plants.

Fetherstonhaugh & Co.,
Ottawa, Canada
Patent Agents



(I)