Office de la Propriété Intellectuelle du Canada
Un organisme d'Industrie Canada

Canadian Intellectual Property Office
An agency of Industry Canada

CA 2078697 C 2003/12/30

(11)(21) 2 078 697

(12) BREVET CANADIEN
CANADIAN PATENT

(13) C

(22) Date de dépôt/Filing Date: 1992/09/21

(41) Mise à la disp. pub./Open to Public Insp.: 1993/03/31

(45) Date de délivrance/Issue Date: 2003/12/30

(30) Priorités/Priorities: 1991/09/30 (251394/1991) JP;
1991/09/30 (251393/1991) JP;
1992/05/28 (136729/1992) JP

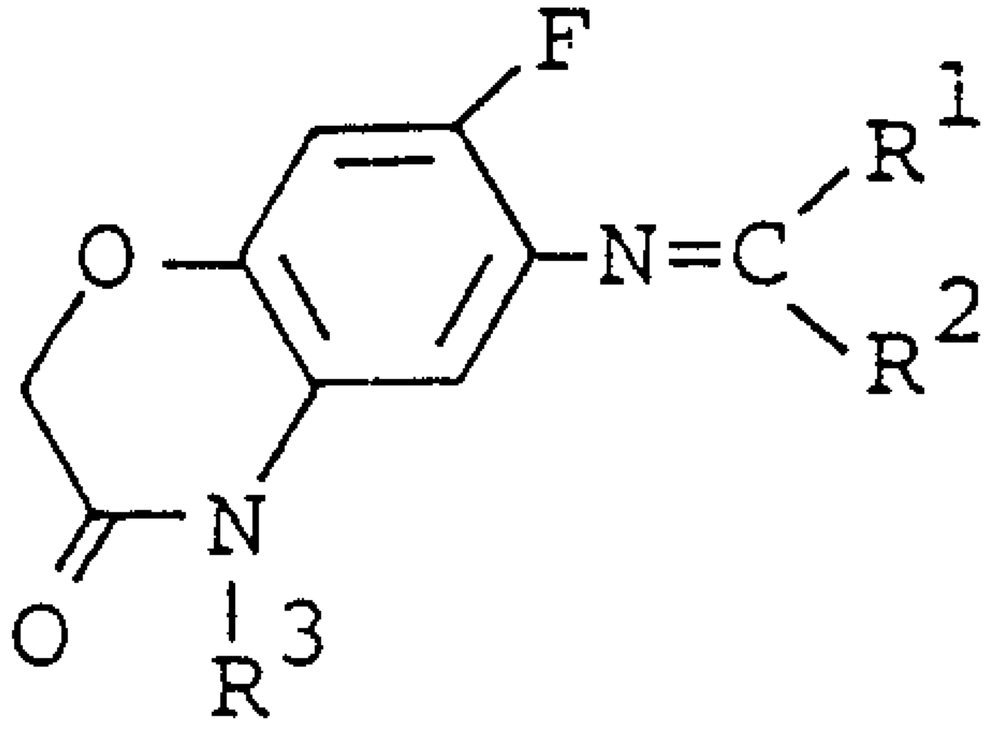
(51) Cl.Int.^{5}/Int.Cl.^{5} C07D 265/36

(72) Inventeurs/Inventors:
MATSUMOTO, MITSUHIRO, JP;
FUNAKI, YUJI, JP

(73) Propriétaire/Owner:
SUMITOMO CHEMICAL COMPANY, LIMITED, JP

(74) Agent: KIRBY EADES GALE BAKER

(54) Titre : COMPOSES A BASE D'IMINE ET LEUR PRODUCTION
(54) Title: IMINE COMPOUNDS AND THEIR PRODUCTION

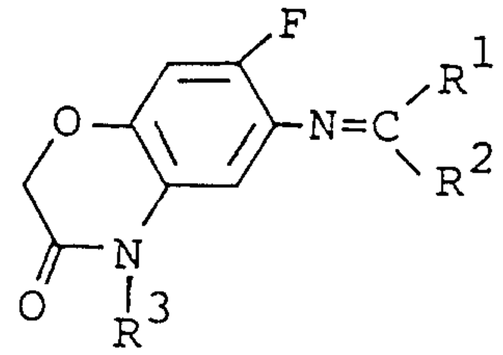


(57) Abrégé/Abstract:
The present invention is directed to an imine compound of the general formula: (see above formula) wherein R^1 and R^2 are the same or different and each represent a C_1-C_5 alkyl group, or R^1 and R^2 are combined together to form a C_4-C_5 alkylene group; R^3 is a C_1-C_5 alkyl group, a C_3-C_4 alkenyl group or a C_3-C_4 alkynyl group. The imine compound of the present invention is useful as an intermediate in the production of agrochemicals and the like. The invention is also directed to an amide compound useful as a raw material for preparation of the imine compound and to production processes for these compounds.

2078697

Abstract

The present invention is directed to an imine compound of the general formula:

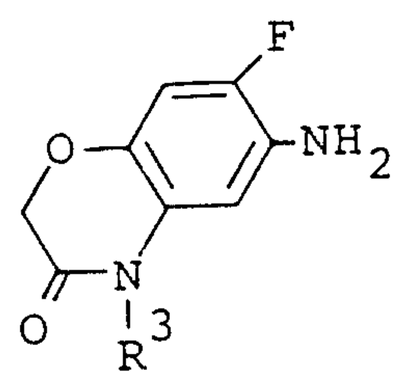


wherein R^1 and R^2 are the same or different and each represent a C_1-C_5 alkyl group, or R^1 and R^2 are combined together to form a C_4-C_5 alkylene group; R^3 is a C_1-C_5 alkyl group, a C_3-C_4 alkenyl group or a C_3-C_4 alkynyl group. The imine compound of the present invention is useful as an intermediate in the production of agrochemicals and the like. The invention is also directed to an amide compound useful as a raw material for preparation of the imine compound and to production processes for these compounds.

Imine Compounds and Their Production

The present invention relates to novel imine compounds useful as intermediates in the production of agrochemicals and medicaments. It also relates to a process for producing these imine compounds.

Certain kinds of tetrahydrophthalimide derivatives, e.g. 2-[7-fluoro-4-(2-propynyl)-2H-1,4-benzoxazin-3(4H)-on-6-yl]-4,5,6,7-tetrahydroisoindole-1,3-dione are known as agrochemicals having herbicidal activity. It is also known that these derivatives can be produced from benzoxazine compounds of the general formula:

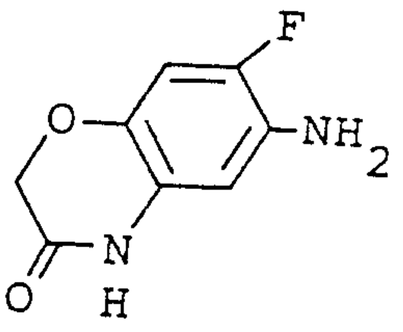


(I)

wherein R^3 is a C_1-C_5 alkyl group, a C_3-C_4 alkenyl group or a C_3-C_4 alkynyl group (JP-A 61-76486).

However, there has been a great demand for further improvements in the conditions and procedures of conventional production processes to produce the benzoxazine compounds (I) with high quality on an industrial scale.

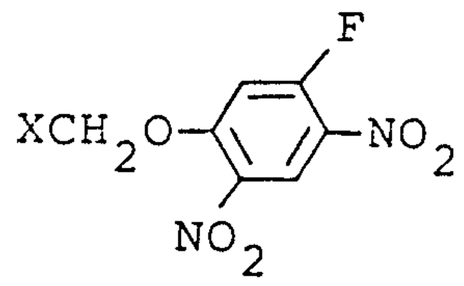
For example, although the benzoxazine compounds (I) can be produced from an aniline compound of the formula:



(II)

(JP-A 62-221677), various side reactions have a tendency to accompany this process because the aniline compound (II) has a reactive amino group. It is not always easy to produce the benzoxazine compounds (I) with high quality on an industrial scale.

Moreover, although the aniline compound (II) as the raw material can be obtained, for example, by catalytic reduction of the fluorodinitrobenzene derivative of the general formula:



(III)

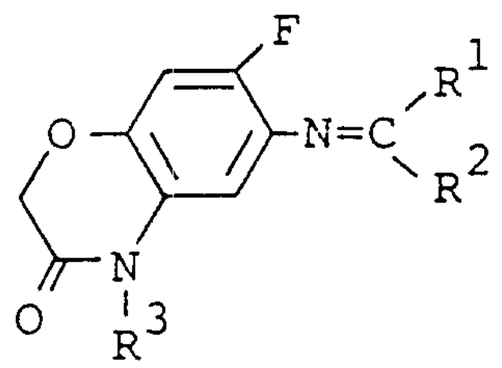
wherein X is a C_1-C_5 alkoxycarbonyl group, a cyano or a carboxyl group (JP-A 62-212375), it is sometimes difficult to separate the aniline compound (II) from the catalyst used, because this compound is slightly soluble in organic solvents which have been widely used for industrial applications.

Under these circumstances, the present inventors have devoted themselves to studying various methods of solving the above problems. These studies have lead to the present invention.

That is, one object of the present invention is to provide novel intermediates for effective production of tetrahydrophthalimide derivatives on an industrial scale.

Another object of the present invention is to provide a process for producing these intermediates.

According to the present invention, there are provided imine compounds of the general formula:

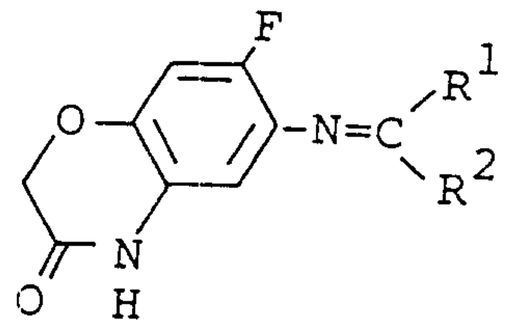


(IV)

wherein R^1 and R^2 are the same or different and each represent a C_1-C_5 alkyl group, or R^1 and R^2 are combined together to form a C_4-C_5 alkylene group; R^3 is a C_1-C_5 alkyl group, a C_3-C_4 alkenyl group or a C_3-C_4 alkynyl group.

Also provided are their raw materials, i.e., amide

compounds of the general formula:



(V)

wherein R^1 and R^2 are as defined above.

Further, production processes for these imine and amide compounds are provided.

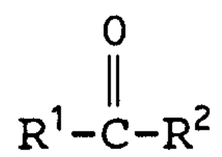
The imine compound (IV) of the present invention can be produced by reacting the amide compound (V) with an electrophilic reagent of the general formula:

$$R^3\text{-}Y \qquad \text{(VI)}$$

wherein R^3 is as defined above and Y is halogen or $Z\text{-}SO_3$ wherein Z is a C_1-C_5 alkoxy group, a C_1-C_5 alkyl group, a C_1-C_5 perfluoroalkyl group or a phenyl group which may be substituted with a halogen atom or a C_1-C_5 alkyl group.

Examples of the electrophilic reagent (VI) include alkyl halides, e.g. methyl chloride, methyl bromide, methyl iodide, ethyl bromide, ethyl iodide, n-propyl bromide, isopropyl chloride, butyl chloride and isobutyl chloride; alkenyl halides, e.g. allyl chloride and allyl bromide; alkynyl halides, e.g. 2-propynyl chloride, 2-propynyl bromide and 1-methyl-2-propynyl chloride; sulfuric esters, e.g. dimethyl sulfate and diethyl sulfate; methanesulfonates, e.g. 2-propynyl methanesulfonate and 1-methyl-2-propynyl methanesulfonate; benzenesulfonates, e.g. 2-propynyl benzenesulfonate and 1-methyl-2-propynyl benzenesulfonate; toluenesulfonates, e.g. 2-propynyl toluenesulfonate and 1-methyl-2-propynyl toluenesulfonate; chlorobenzenesulfonates, e.g. 2-propynyl chlorobenzenesulfonate and 1-methyl-2-propynyl chlorobenzenesulfonate and 1-methyl-2-propynyl chlorobenzenesulfonate; bromobenzenesulfonates, e.g. 2-propynyl bromobenzenesulfonate and 1-methyl-2-propynyl bromobenzenesulfonate; and triflates, e.g. 2-propynyl triflate and 1-methyl-2-propynyl triflate.

The reaction is usually conducted in a solvent. Typical examples of the solvent are ketones of the general formula:



(VII)

wherein R^1 and R^2 are as defined above, corresponding to a combination of R^1 and R^2 of the amide compounds (V). There can also be used aromatic compounds, e.g. toluene, xylene, monochlorobenzene and dichlorobenzene; aliphatic compounds, e.g. hexane and heptane; dimethylformamide, dimethylsulfoxide, tetrahydrofuran, and mixtures thereof. If necessary, water may be added to the solvent.

The reaction is usually conducted with a base. Examples of the base include hydroxides of alkali metals, e.g. sodium hydroxide and potassium hydroxide; carbonates of alkali metals, e.g. sodium carbonate and potassium carbonate; and organic amines, e.g. triethylamine, pyridine and N,N-diethylaniline.

A catalyst may also be used to increase the effectiveness of the reaction. Examples of the catalyst are quaternary ammonium salts, e.g. tetrabutylammonium bromide, triethylbenzylammonium chloride and tetrabutylammonium hydrogensulfate; quaternary phosphonium salts, e.g. cetyltributylphosphonium bromide and butyltrioctylphosphonium bromide; and crown ethers, e.g. 18-crown-6; and TDA-1.

The electrophilic reagent (VI) is used in an amount of not less than one equivalent, usually from 1 to 2 equivalents, per equivalent of the amide compound (V).

The base is used in an amount of not less than one equivalent, usually from 1 to 5 equivalents, per equivalent of the amide compound (V). The catalyst is usually used in a catalytic amount per equivalent of the amide compound (V).

The reaction temperature is usually in the range of from room temperature to the boiling point of the solvent used.

After completion of the reaction, the desired imine compound (IV) can be obtained by a conventional post-treatment. Moreover, when an excess amount of an

electrophilic reagent is used, for example, the addition of aqueous Na_2SO_3, ammonium hydroxide, methanol or the like to thereby decompose the remaining electrophilic reagent may be effective in some cases for obtaining the compound of the present invention with high quality. The resulting imine compound (IV) is usually used in the subsequent step as it is in solution form, although the imine compound can also be isolated, if necessary, by concentration.

Next, the following will describe a process for producing amide compound (V) which is an intermediate used in the production of the imine compound (IV) of the present invention.

The amide compound (V) can be produced by reacting the aniline compound (II) with the ketone (VII). The aniline compound (II) can be obtained by catalytic reduction of a fluorodinitrobenzene derivative (III) in the presence of a catalyst.

Examples of the catalyst used for the catalytic reduction are platinum dioxide, palladium carbon and Raney nickel. The catalyst is usually used in a range of from a catalytic amount to 10% by weight based on the weight of the fluorodinitrobenzene derivative (III).

The reaction is usually conducted in a solvent. Examples of the solvent include water, organic solvents, e.g. toluene, xylene, methanol, ethanol and isopropanol, and mixtures thereof. The aniline compound (II), which is only slightly soluble in organic solvents, is precipitated as crystals when the catalytic reduction is completed. It is, therefore, not always easy to separate the resulting aniline compound (II) from the catalyst using a conventional filtration technique. Thus, in the process of the present invention, after completion of the catalytic reduction, a mixture of the aniline compound (II) in crystal form and the catalyst is obtained by a suitable technique, e.g. filtration, and this mixture may be used as such for the subsequent imination. Therefore, the solvent may be used in an amount which does not adversely affect the proceeding catalytic reduction. In the

present invention, the solvent is usually used at a weight of 1 to 10 times that of the weight of the fluorodinitrobenzene derivative (III).

The reaction temperature is usually in the range of from room temperature to 150°C.

The reaction pressure is usually in the range of from normal pressure to 30 kg/cm^2.

Next, the following will describe the step of reacting the aniline compound (II) together with the catalyst used for the catalytic reduction with the ketone (VII) to obtain the amide compound (V).

Examples of the ketone (VII) to be used include aliphatic ketones, e.g. acetone, methyl ethyl ketone, methyl isobutyl ketone, diethyl ketone and dibutyl ketone; and alicyclic ketones, e.g. cyclopentanone and cyclohexanone.

The ketone (VII) is used in a range of from a molar concentration equivalent to that of the aniline compound (II) to large excess amounts in which case the ketone (VII) serves as a solvent.

The reaction is usually conducted in a solvent. Examples of the solvent are, in addition to the ketones (VII) as described above, aromatic compounds, e.g. toluene, xylene, monochlorobenzene and dichlorobenzene; halogen compounds, e.g. methylene chloride and dichloroethane; aliphatic compounds, e.g. hexane and heptane; and mixtures thereof. The amount of solvent to be used is not particularly limited.

The reaction temperature is in the range of from room temperature to the boiling point of the solvent used. Depending upon the type of solvent used, the rate at which the reaction proceeds may be increased by removing water from the reaction system under reflux conditions.

If necessary, it is also possible to use dehydrating agents, e.g. molecular sieves and calcium chloride, or an acid or a base catalyst, e.g. sulfuric acid, hydrochloric acid, acetic acid, p-toluene-sulfonic acid, piperidine or pyridine. The catalyst is used in a catalytic amount for the aniline compound (II).

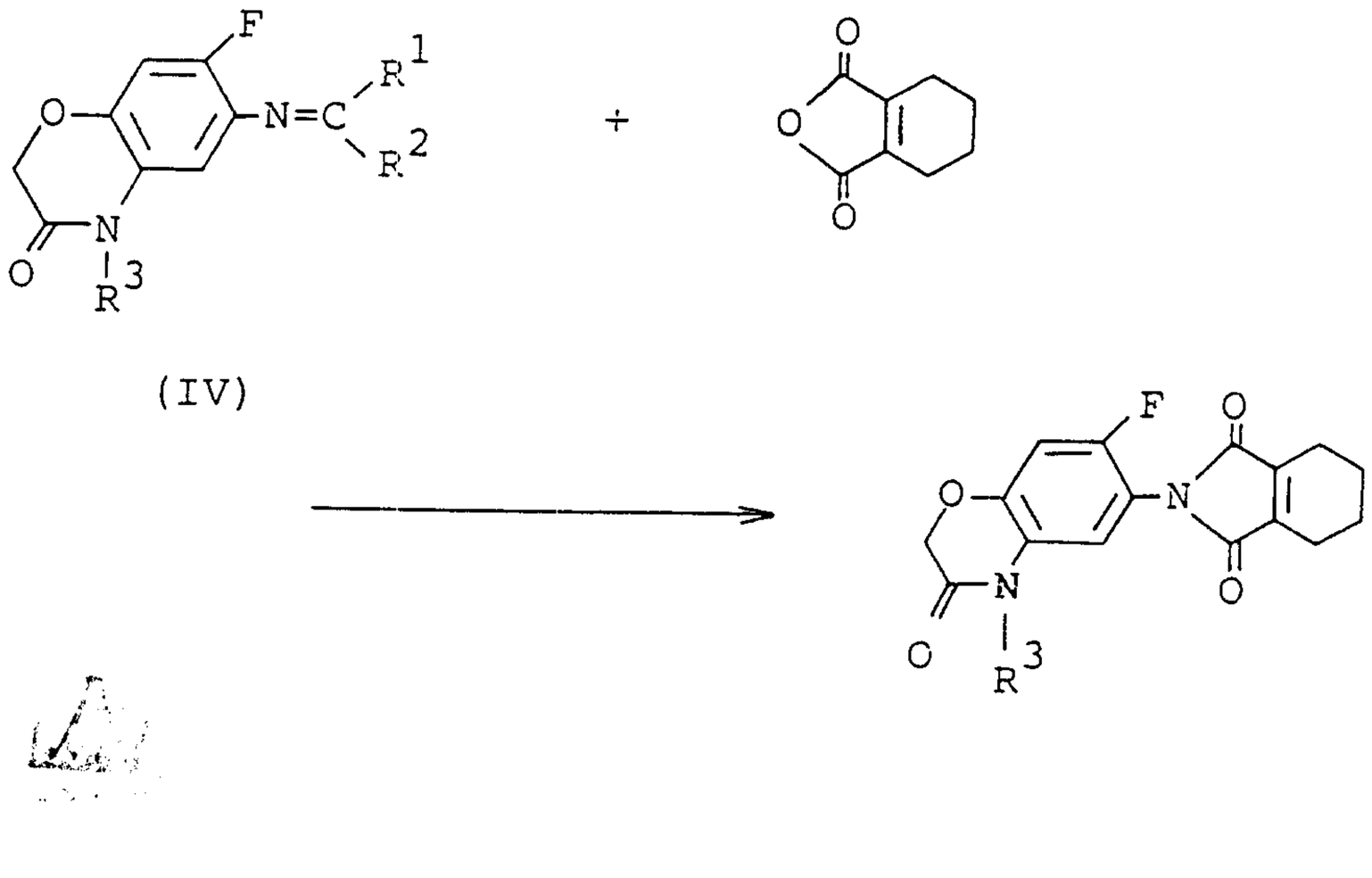
The resulting amide compound (V) is dissolved in the solvent, while the reduction catalyst, which is insoluble in organic solvents, is separated and recovered by a conventional solid-liquid separation technique, e.g. filtration or centrifugation. Thereby the desired amide compound (V) is obtained in solution form.

Although the amide compound (V) obtained in this way is used in the subsequent step of producing the imine compound (IV) as it is in solution form, if necessary, the amide compound (V) can be isolated from the solution by a conventional technique, e.g. concentration.

Examples of the amide compound (V) obtained by the above reaction are 6-(1,3-dimethylbutylidenamino)-7-fluoro-2H-1,4-benzoxazin-3(4H)-one, 6-cyclohexylidenamino-7-fluoro-2H-1,4-benzoxazin-3(4H)-one, 6-(1-ethylpropylidenamino)-7-fluoro-2H-1,4-benzoxazin-3(4H)-one, 6-cyclopentylidenamino-7-fluoro-2H-1,4-benzoxazin-3(4H)-one, 6-(1-methylpropylidenamino)-7-fluoro-2H-1,4-benzoxazin-3(4H)-one and 6-(1-methylbutylidenamino)-7-fluoro-2H-1,4-benzoxazin-3(4H)-one.

The imine compound (IV) and amide compound (V) of the present invention may be present, when R^1 and R^2 are different from each other, in the form of syn- and anti-isomers, based on the double bonding of the imino group, although the abundance ratio thereof is not particularly limited.

The imine compound (IV) obtained by the process of the present invention can be converted to the above-described tetrahydrophthalimide derivative useful in the production of agrochemicals by the following reaction scheme.



(IV)

wherein R^1, R^2, and R^3 are as defined above.

According to the present invention, the imine compound (IV) can be obtained by simple procedures with high efficiency and high quality, so that the production processes of the present invention are superior to conventional processes from an industrial point of view. The imine compound (IV) and amide compound (V) are useful as intermediates in the production of agrochemicals and the like.

The following Examples will further illustrate the present invention in detail, but are not to be construed to limit the scope thereof.

Example 1

In a 200-ml autoclave, toluene (23 g), methanol (2 g), acetic acid (0.2 g) and 5% palladium carbon (0.3 g) were charged. Then, the atmosphere in the autoclave was replaced with hydrogen gas, after which a solution (100 g) of butyl 5-fluoro-2,4-dinitrophenoxyacetate (20 g) in toluene was added thereto over 3 hours, while feeding hydrogen gas at a pressure of 10 kg/cm^2 and maintaining the inner temperature at 60°C to 70°C.

After confirming that hydrogen gas absorption had terminated, the reaction was stopped and the reaction mixture was filtered at 30°C to 40°C, resulting in a mixture (12 g) of 6-amino-7-fluoro-2H-1,4-benzoxazin-3(4H)-one and palladium carbon.

This mixture was charged in a reaction vessel equipped with a Dean-Stark extractor, to which methyl isobutyl ketone (100 g) and p-toluenesulfonic acid (0.1 g) were added, followed by dehydration under reflux at 80° to 90°C under reduced pressure for 5 hours. After completion of the reaction, the reaction mixture was cooled to 40°C and then filtered, whereby palladium carbon was recovered as a residue on the filter and a solution of 6-(1,3-dimethyl-butylidenamino)-7-fluoro-2H-1,4-benzoxazin-3(4H)-one in methyl isobutyl ketone was obtained as a filtrate.

Gas chromatography analysis showed that the yield from butyl 5-fluoro-2,4-dinitrophenoxyacetate was 92%. This

solution was concentrated under reduced pressure to give the desired compound as crystals having a melting point of 113° to 115°C.

Example 2

A solution of 6-cyclohexylidenamino-7-fluoro-2H-1,4-benzoxazine-3(4H)-one in cyclohexanone was obtained in the same manner as described in Example 1, except that cyclohexanone was used instead of methyl isobutyl ketone.

Gas chromatography analysis showed that the yield from butyl 5-fluoro-2,4-dinitrophenoxyacetate was 93%. This solution was concentrated under reduced pressure to give the desired compound as crystals having a melting point of 196° to 200°C.

^{1}H-NMR ($CDCl_3$) δ ppm: 9.15 (1H, s), 6.75 (1H, d, J=10 Hz), 6.34 (1H, d, J=8 Hz), 4.59 (2H, s), 2.49 (2H, t, J=6 Hz), 2.16 (2H, m), 1.85 (2H, m), 1.67 (4H, m).

Example 3

A solution of 6-(1-ethylpropylidenamino)-7-fluoro-2H-1,4-benzoxaxin-3(4H)-one in diethyl ketone was obtained in the same manner as described in Example 1, except that diethyl ketone was used instead of methyl isobutyl ketone.

Gas chromatography analysis showed that the yield from butyl 5-fluoro-2,4-dinitrophenoxyacetate was 90%. This solution was concentrated under reduced pressure to give the desired compound as crystals having a melting point of 240°C (decomposition).

^{1}H-NMR ($CDCl_3$) δ ppm: 9.16 (1H, s), 6.73 (1H, d, J=10 Hz), 6.29 (1H, d, J=8 Hz), 4.59 (2H, s), 2.48 (2H, q, J=7 Hz), 2.15 (2H, q, J=8 Hz), 1.20 (3H, t, J=Hz), 1.03 (3H, t, J=8Hz).

Examples 4, 5 and 6

The compounds of Examples 4, 5 and 6 having the variables as shown in Table 1 were obtained in the same manner as described in Example 1, except that cyclopentanone, methyl ethyl ketone and methyl propyl ketone were used, respectively, instead of methyl isobutyl ketone as the solvent for imination.

The amide compounds (V) of Examples 1 to 6 had the variables as shown in Table 1.

Table 1

Example	R^1	R^2
1	CH_3	$CH_2CH(CH_3)_2$
2	$-(CH_2)_5-$	
3	C_2H_5	C_2H_5
4	$-(CH_2)_4-$	
5	CH_3	C_2H_5
6	CH_3	$CH_2CH_2CH_3$

Example 7

Anhydrous potassium carbonate (52 g), tetrabutylammonium bromide (5 g) and water (5 g) were added to a solution (600 g) of 6-(1,3-dimethylbutylidenamino)-7-fluoro-2H-1,4-benzoxazin-3(4H)-one (83 g) in methyl isobutyl ketone. To this reaction mixture, a solution (230 g) of 2-propynyl methanesulfonate (48 g) in methyl isobutyl ketone was added at 40°C over 1 hour.

The reaction mixture was stirred at the same temperature for 6 hours, after which 5% aqueous Na_2SO_3 was added thereto and the mixture was further stirred at 60°C for 2 hours to thereby decompose excess 2-propynyl methanesulfonate.

The reaction mixture was allowed to stand and separated to give a solution of 6-(1,3-dimethylbutylidenamino)-7-fluoro-4-(2-propynyl)-2H-1,4-benzoxazin-3(4H)-one (90 g) in methyl isobutyl ketone (yield: 95%).

The resulting solution was dried over magnesium sulfate and filtered. While paying attention to moisture absorption, methyl isobutyl ketone was removed from the filtrate at a temperature of lower than 30°C under reduced pressure with a vacuum pump to give the desired compound as an oil.

^{1}H-NMR ($CDCl_3$) δ ppm: 6.78 (1H, d), 6.66 (1H, d), 4.65 (2H, d), 4.62 (2H, d), 2.35 (2H, d), 2.25 (1H, t), 2.15 (1H, m), 1.83 (3H, s), 1.02 (6H, t).

MS: 302 (M^+); (MW 302.35).

Example 8

Anhydrous potassium carbonate (52 g), tetrabutylammonium hydrogensulfate (2 g) and water (5 g) were added to a solution (600 g) of 6-(1,3-dimethylbutylidenamino)-7-fluoro-2H-1,4-benzoxazin-3(4H)-one (83 g) in methyl isobutyl ketone. To this reaction mixture, a solution (230 g) of 2-propynyl methanesulfonate (48 g) in methyl isobutyl ketone was added at 40°C over 1 hour.

This reaction mixture was stirred at the same temperature for 6 hours, after which 5% aqueous Na_2SO_3 was added thereto and the mixture was further stirred at 60°C for 2 hours to thereby decompose excess 2-propynyl methanesulfonate.

The reaction mixture was allowed to stand and separated to give a solution of 6-(1,3-dimethylbutylidenamino)7-fluoro-(2-propynyl)-2H-1,4-benzoxazin-3(4H)-one (90 g) in methyl isobutyl ketone (yield: 95%).

Example 9

Anhydrous potassium carbonate (52 g), tetrabutylammonium bromide (5 g) and water (5 g) were added to a solution (600 g) of 6-cyclohexylidenamino-7-fluoro-2H-1,4-benzoxazin-3(4H)-one (83 g) in cyclohexanone. To this reaction mixture, a solution (230 g) of 2-propynyl methanesulfonate (48 g) in cyclohexanone was added at 40°C over 1 hour.

The reaction mixture was stirred at the same temperature for 6 hours, after which 5% aqueous Na_2SO_3 was added thereto and the mixture was further stirred at 60°C for 2 hours to thereby decompose excess 2-propynyl methanesulfonate.

The reaction mixture was allowed to stand and separated to give a solution of 6-cyclohexylidenamino-7-fluoro-4-(2-propynyl)-2H-1,4-benzoxazin-3(4H)-one (91 g) in cyclohexanone (yield : 96%).

The resulting solution was dried over magnesium sulfate and filtered. Then, cyclohexanone was removed from the filtrate under reduced pressure with a vacuum pump to give the desired compound as an oil.

^{1}H-NMR ($CDCl_3$) δ ppm: 6.77 (1H, d, J=10 Hz), 6.67 (1H, d, J=7 Hz), 4.65 (2H, d, J=3 Hz), 4.61 (2H, d), 2.52 (2H, t, J=6 Hz), 2.26 (1H, t, J=3 Hz), 2.20 (2H, m), 1.86 (2H, m), 1.69 (4H, m).

Example 10

Anhydrous potassium carbonate (52 g), tetrabutylammonium bromide (5 g) and water (5 g) were added to a solution (600 g) of 6-(1-ethylpropylidenamino)-7-fluoro-2H-1,4-benzoxazin-3(4H)-one (79 g) in diethyl ketone. To this reaction mixture, a solution of (230 g) of 2-propynyl methanesulfonate (48 g) in diethyl ketone was added at 40°C over 1 hour.

The reaction mixture was stirred at the same temperature for 6 hours, after which 5% aqueous Na_2SO_3 was added thereto and the mixture was further stirred at 60°C for 2 hours to thereby decompose excess 2-propynyl methanesulfonate.

The reaction mixture was allowed to stand and separated to give a solution of 6-(1-ethylpropylidenamino)-7-fluoro-4-(2-propynyl)-2H-1,4-benzoxazin-3(4H)-one (83 g) in methyl isobutyl ketone (yield: 92%).

The resulting solution was dried over magnesium sulfate and filtered. Then, diethyl ketone was removed from the filtrate under reduced pressure with a vacuum pump to give the desired compound as an oil.

^{1}H-NMR ($CDCl_3$) δ ppm: 6.77 (1H, d, J=10 Hz), 6.65 (1H, d, J=7 Hz), 4.60 (2H, d, J=3 Hz), 4.62 (2H, s), 2.52 (2H, q, J=7 Hz), 2.24 (1H, q, J=3Hz), 2.18 (2H, m), 1.23 (3H, t, J=7 Hz), 1.06 (3H, t, J=7 Hz).

Examples 11 to 15

The imine compounds of Examples 12, 14 and 15 were obtained in the same manner as described in Example 7, except that 1-methyl-2-propynyl methanesulfonate, allyl methanesulfonate and n-propyl methanesulfonate were used, respectively, instead of 2-propynyl methanesulfonate. The

compounds of Examples 4 and 6 were reacted with 2-propynyl methanesulfonate to give the imine compounds of Examples 11 and 13, respectively.

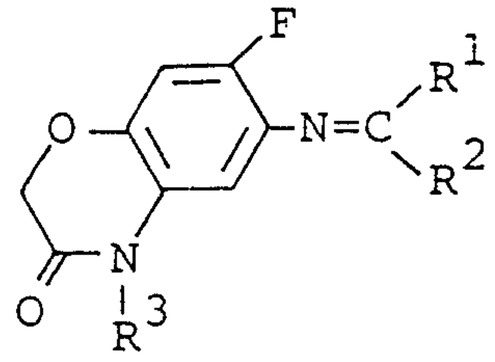
The imine compounds (IV) of Examples 7 to 15 had the variables as shown in Table 2.

Table 2

Example	R^1		R^2	R^3
7	CH_3		$CH_2CH(CH_3)_2$	$CH_2C\equiv CH$
8	CH_3		$CH_2CH(CH_3)_2$	$CH_2C\equiv CH$
9		$-(CH_2)_5-$		$CH_2C\equiv CH$
10	C_2H_5		C_2H_5	$CH_2C\equiv CH$
11		$-(CH_2)_4-$		$CH_2C\equiv CH$
12	CH_3		$CH_2CH(CH_3)_2$	$CH(CH_3)C\equiv CH$
13	CH_3		$CH_2CH_2CH_3$	$CH_2C\equiv CH$
14	CH_3		$CH_2CH(CH_3)_2$	$CH_2CH=CH_2$
15	CH_3		$CH_2CH(CH_3)_2$	$CH_2CH_2CH_3$

Claims:

1. An imine compound of the general formula:



(IV)

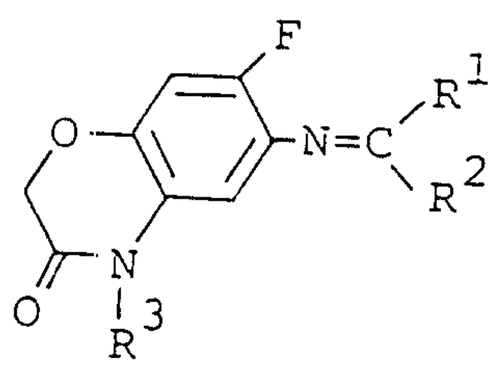
wherein R^1 and R^2 are the same or different and each represent a C_1-C_5 alkyl group, or R^1 and R^2 are combined together to form a C_4-C_5 alkylene group; R^3 is a C_1-C_5 alkyl group, a C_3-C_4 alkenyl group or a C_3-C_4 alkynyl group.

2. An amine compound according to claim 1, wherein R^1 is a methyl group and R^2 is an isobutyl group.

3. An imine compound according to claim 1, wherein R^3 is a 2-propynyl group.

4. An imine compound according to claim 1, wherein R^1 is a methyl group, R^2 is an isobutyl group and R^3 is a 2-propynyl group.

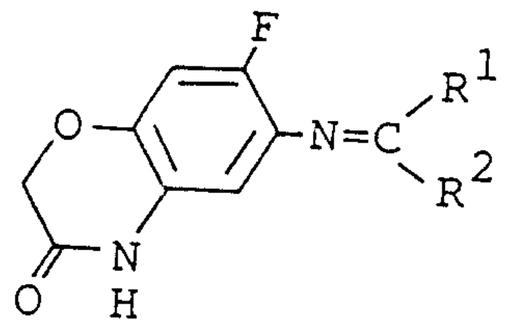
5. A process for producing an imine compound of the general formula:



(IV)

wherein R^1 and R^2 are the same or different and each represent a C_1-C_5 alkyl group, or R^1 and R^2 are combined together to form a C_4-C_5 alkylene group; R^3 is a C_1-C_5 alkyl group, a C_3-C_4 alkenyl group or a C_3-C_4 alkynyl group, which comprises the

steps of reacting an amide compound of the general formula:



(V)

wherein R^1 and R^2 are as defined above with an electrophilic reagent of the general formula:

$$R^3\text{-}Y \qquad \text{(VI)}$$

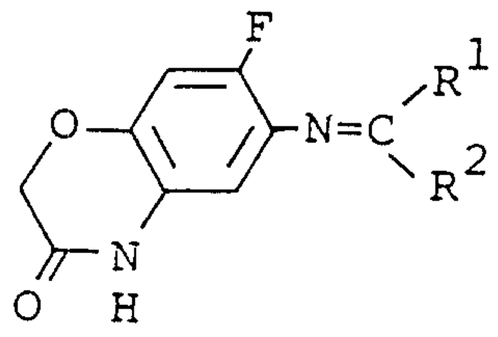
wherein R^3 is as defined above and Y is halogen or $Z\text{-}SO_3$ wherein Z is a C_1-C_5 alkoxy group, a C_1-C_5 alkyl group, a C_1-C_5 perfluoroalkyl group or a phenyl group which may be substituted with a halogen atom or a C_1-C_5 alkyl group.

6. A process according to claim 5, wherein R^1 is a methyl group and R^2 is an isobutyl group.

7. A process according to claim 5, wherein R^3 is a 2-propynyl group.

8. A process according to claim 5, wherein R^1 is a methyl group, R^2 is an isobutyl group and R^3 is a 2-propynyl group.

9. An amide compound of the general formula:

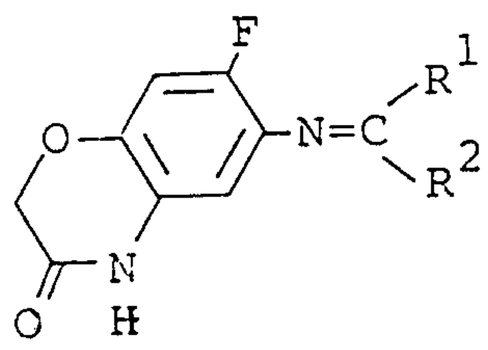


(V)

wherein R^1 and R^2 are the same or different and each represent a C_1-C_5 alkyl group, or R^1 and R^2 are combined together to form a C_4-C_5 alkylene group.

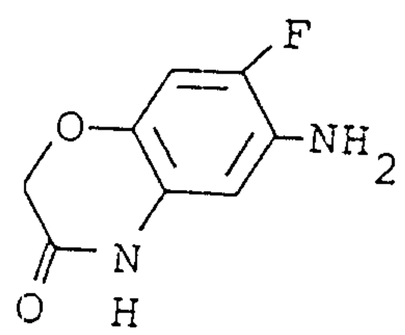
10. An amide compound according to claim 9, wherein R^1 is a methyl group and R^2 is an isobutyl group.

11. A process for producing an amide compound of the general formula:



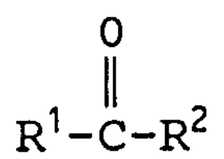
(V)

wherein R^1 and R^2 are the same or different and each represent a C_1-C_5 alkyl group, or R^1 and R^2 are combined together to form a C_4-C_5 alkylene group, which comprises the steps of reacting an aniline compound (II) of the general formula:



(II)

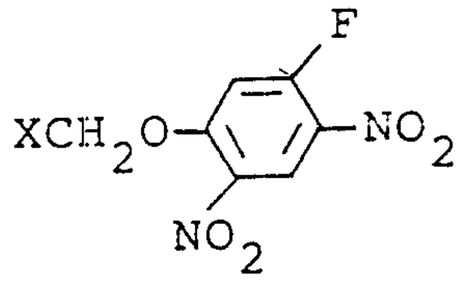
with a ketone compound of the general formula:



(VII)

wherein R^1 and R^2 are as defined above.

12. A process according to claim 11, wherein the aniline compound (II) is obtained by catalytic reduction of a fluorodinitrobenzene derivative of the general formula:



(III)

wherein X is a C_1-C_5 alkoxycarbonyl group, a cyano or a carboxyl group in the presence of a catalyst, and then reacting with the ketone compound (VII) without being

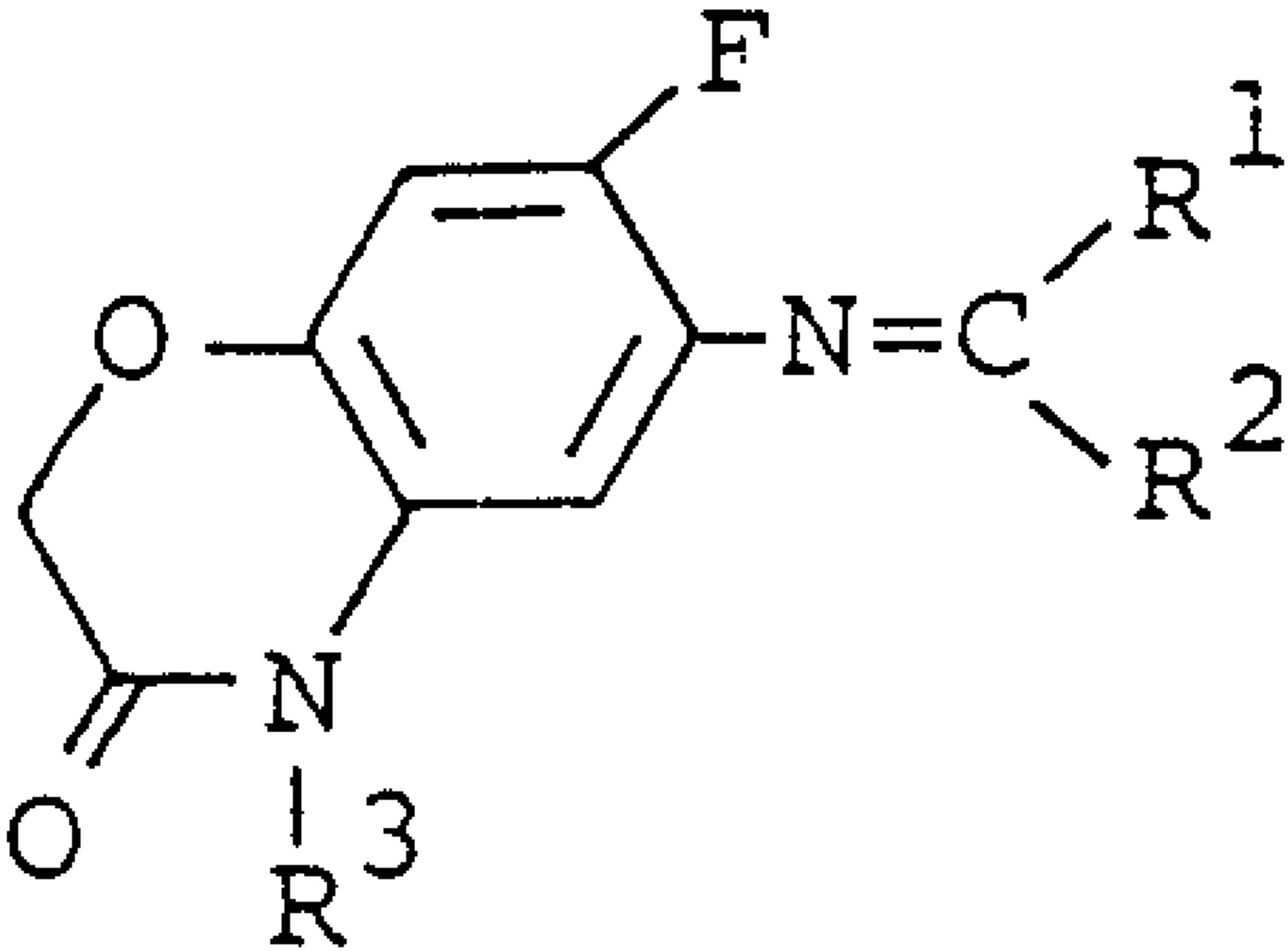
separated from the catalyst to form the amide compound (V), followed by removal of the catalyst.

13. A process according to claim 11, wherein R^1 is a methyl group and R^2 is an isobutyl group.

14. A process according to claim 12, wherein R^1 is a methyl group and R_2 is an isobutyl group.

15. A process according to claim 12, wherein X is a butoxycarbonyl group.

16. A process according to claim 12, wherein the reduction catalyst is palladium carbon, platinum dioxide or Raney-nickel™.


F
O
N
C
R1
R2
O
N
R3