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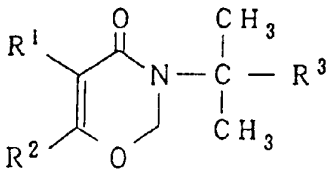
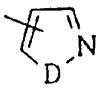
(54) **Title:** CYCLIC AMIDE COMPOUNDS, PROCESS FOR THEIR PRODUCTION AND HERBICIDAL COMPOSITIONS CONTAINING THEM



(I)

(a)

(b)



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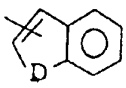

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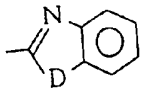

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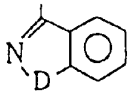

(g)

(57) **Abstract**

The present invention provides a cyclic amide compound of formula (I): wherein R^1 is a phenyl group which may be substituted, R^2 is a hydrogen atom or an alkyl groupe which may be substituted by a halogen atom, and R^3 is (a) which may be substituted, (b) which may be substituted, (c) which may be substituted, (d) which may be substituted, (e) which may be substituted, (f) which may be substituted, or (g) which may be substituted, wherein D is an oxygen atom, a sulfur atom or $-N(R^4)-$, wherein R^4 is an alkyl group, a process for producing such a compound, a herbicidal composition containing such a compound as an active ingredient, a herbicidal method and an intermediate for the production of such a compound. The cyclic amide compound of the present invention is useful as a herbicidally active ingredient to selectively kill weeds without presenting a phytotoxicity against crop plants.

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DESCRIPTION

TITLE OF THE INVENTION

CYCLIC AMIDE COMPOUNDS, PROCESS FOR THEIR PRODUCTION AND HERBICIDAL COMPOSITIONS CONTAINING THEM

TECHNICAL FIELD

The present invention relates to cyclic amide compounds of the formula (I) given hereinafter, a process for their production, their use as herbicides, and intermediates useful for their production, herbicidal compositions comprising the cyclic amide compounds and other herbicidally active components, and a herbicidal method which comprises applying such herbicidal compositions to plants.

BACKGROUND ART

Heretofore, it has been desired to develop herbicides which are capable of selectively killing noxious weeds without presenting a phytotoxicity to crop plants. Various researches have been made to develop such herbicides, and a number of selective herbicides have been published, but they are not necessarily satisfactory. Under the circumstances, it is still desired to develop a superior herbicide.

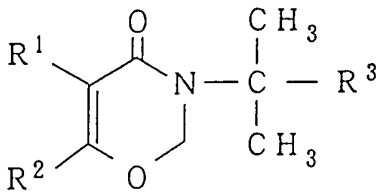
On the other hand, Japanese Unexamined Patent Publication No. 89485/1992 discloses cyclic amide derivatives having herbicidal activities, but such derivatives are different in the chemical structure from

the compounds of the present invention represented by the formula (I) given hereinafter.

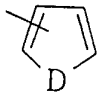
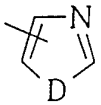
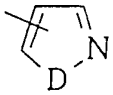
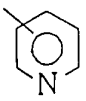
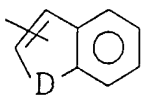
DISCLOSURE OF THE INVENTION

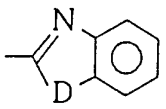
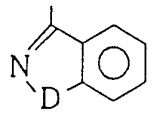
The present inventors have conducted extensive studies on cyclic amide compounds with an aim to develop an excellent herbicide and as a result, have found that compounds of the formula (I) given hereinafter have excellent herbicidal effects and selectivity and thus are capable of effectively controlling noxious weeds without presenting a phytotoxicity to certain crop plants. The present invention has been accomplished on the basis of this discovery.

Thus, the present invention provides a cyclic amide compound of the formula (I):



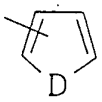
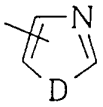
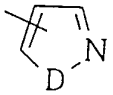
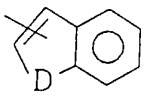
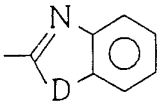
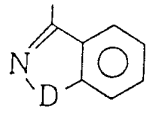
(I)

wherein R^1 is a phenyl group which may be substituted, R^2 is a hydrogen atom or an alkyl group which may be substituted by a halogen atom, and R^3 is  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,

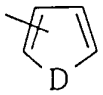
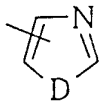
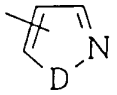
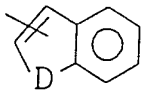
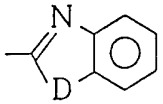
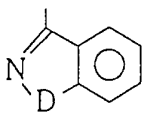
 which may be substituted, or  which may be substituted, wherein D is an oxygen atom, a sulfur atom or $-N(R^4)-$, wherein R^4 is an alkyl group.

The present invention further provides a process for producing such a cyclic amide compound, herbicidal compositions containing it, herbicidal methods of applying such herbicidal compositions and an intermediate useful for its production.

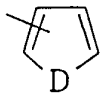
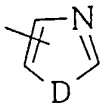
BEST MODE FOR CARRYING OUT THE INVENTION

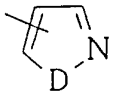
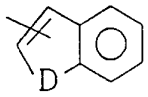
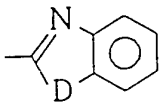
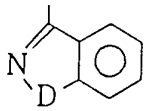
In the above formula (I), the substituent for the phenyl group which may be substituted, for R^1, may, for example, be a halogen atom; an alkyl group which may be substituted by a halogen atom; an alkoxy group which may be substituted by a halogen atom; an alkylthio group which may be substituted by a halogen atom or an alkylsulfonyl group which may be substituted by a halogen atom. Likewise, the substituent for each of  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted, and  which may be substituted, wherein D is as defined above, for R^3, may, for example, be a halogen

atom; an alkyl group which may be substituted by a halogen atom; an alkoxy group which may be substituted by a halogen atom; an alkylthio group which may be substituted by a halogen atom; an alkylsulfonyl group which may be substituted by a halogen atom; an aryl group which may be substituted by a halogen atom or a halogenoalkyl group; a heteroaryl group which may be substituted by a halogen atom or a halogenoalkyl group; an aryloxy group which may be substituted by a halogen atom or a halogenoalkyl group; or a heteroaryloxy group which may be substituted by a halogen atom or a halogenoalkyl group. The above aryl group or aryl moiety may, for example, be a phenyl group or a naphthyl group, and the above heteroaryl group or heteroaryl moiety may, for example, be a pyridyl group, a pyrrole group, a thienyl group or a furyl group. Further, the number of such substituents may be one or more, and in the case of a plurality of substituents, such substituents may be the same or different.

Further, in the definition of  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted, and 

which may be substituted, wherein D is as defined above, for R^3 in the formula (I),

 includes, for example, furan-2-yl, furan-3-yl, thiophen-2-yl, thiophen-3-yl, 1-methylpyrrol-2-yl and 1-methylpyrrol-3-yl;  includes, for example, thiazol-2-yl, oxazol-2-yl, 1-methyl-imidazol-2-yl, thiazol-4-yl, oxazol-4-yl, 1-methylimidazol-4-yl, thiazol-5-yl, oxazol-5-yl and 1-ethylimidazol-5-yl;

 includes, for example, isothiazol-3-yl, isoxazol-3-yl, 1-methylpyrazol-3-yl, isothiazol-4-yl, isoxazol-4-yl, 1-methylpyrazol-4-yl, isothiazol-5-yl, isoxazol-5-yl and 1-methylpyrazol-5-yl;  includes, for example, benzofuran-2-yl, benzofuran-3-yl, benzothiophen-2-yl, benzothiophen-3-yl, 1-methyl-indol-2-yl and 1-methyl-indol-3-yl;  includes, for example, benzoxazol-2-yl, benzothiazol-2-yl and 1-methylbenzimidazol-2-yl;  includes, for example, benzisothiazol-3-yl, benzisoxazol-3-yl and 1-methyl-benzopyrazol-3-yl.

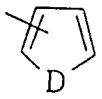
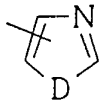
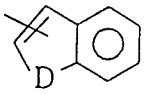
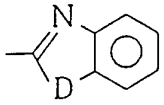
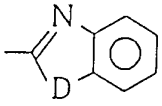
The alkyl group or alkyl moiety in the definition of R^1, R^2, R^3 and R^4 in the formula (I) may, for example, be a C_{1-8}, preferably C_{1-4}, linear or branched alkyl group such as a methyl group, an ethyl group, a propyl group, an isopropyl group, a tert-butyl group, a pentyl group or

an octyl group. The halogen atom for R^1, R^2 or R^3 or the halogen atom as a substituent may be fluorine, chlorine, bromine or iodine. The number of halogen atoms as substituents may be one or more, and in the case of a plurality of such substituted halogen atoms, they may be the same or different.

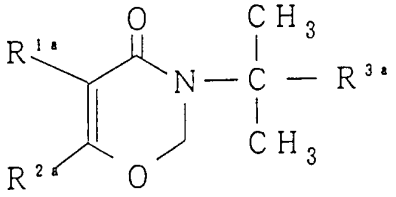
Preferred embodiments of the cyclic amide compound of the formula (I) will be given below.

(1) R^1 is preferably a phenyl group.

(2) R^2 is preferably an alkyl group which may be substituted by a halogen atom, more preferably an unsubstituted alkyl group.

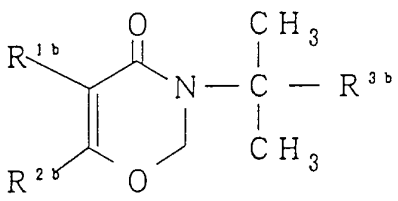
(3) R^3 is preferably  which may be substituted,  which may be substituted,  which may be substituted or  which may be substituted, wherein D is as defined above, more preferably  which may be substituted wherein D is as defined above.

(4) Among compounds of the formula (1), typical compounds may be represented by the following formula:



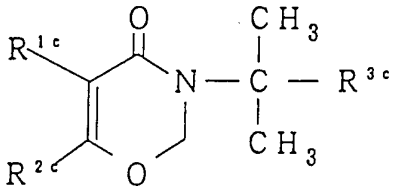
wherein R^{1a} is a phenyl group which may be substituted by a substituent selected from the group consisting of a halogen atom, a C_{1-4} alkyl group, a halogen-substituted C_{1-4} alkyl group and a C_{1-4} alkoxy group, R^{2a} is a C_{1-4} alkyl group which may be substituted by a halogen atom, and R^{3a} is a furan-2-yl group, a furan-3-yl group, a thiophen-2-yl group, a thiophen-3-yl group, a 1-methylpyrrol-2-yl group, a 1-methylpyrrol-3-yl group, a thiazol-2-yl group, a thiazol-4-yl group, a thiazol-5-yl group, an oxazol-2-yl group, an oxazol-4-yl group, an oxazol-5-yl group, a 1-methyl-imidazol-2-yl group, a 1-methyl-imidazol-4-yl group, a 1-ethyl-imidazol-5-yl group, a benzofuran-2-yl group, a benzofuran-3-yl group, a benzothiophen-2-yl group, a benzothiophen-3-yl group, a 1-methylindol-2-yl group, a 1-methylindol-3-yl group, a benzothiazol-2-yl group, a benzoxazol-2-yl group or a 1-methylbenzimidazol-2-yl group, provided that such substituents may be substituted by from 1 to 4 substituents selected from the group consisting of a halogen atom, a C_{1-4} alkyl group, a halogen-substituted C_{1-4} alkyl group and a C_{1-4} alkoxy group.

(5) Among compounds of the formula (1), more typical compounds may be represented by the formula:



wherein R^{1b} is a phenyl group which may be substituted by a substituent selected from the group consisting of a halogen atom, a C_{1-4} alkyl group, a halogen-substituted C_{1-4} alkyl group and a C_{1-4} alkoxy group, R^{2b} is a C_{1-4} alkyl group which may be substituted by a halogen atom, and R^{3b} is a benzothiazol-2-yl group, a benzoxazol-2-yl group or a 1-methylbenzimidazol-2-yl group, provided that these substituents may be substituted by from 1 to 4 substituents selected from the group consisting of a halogen atom, a C_{1-4} alkyl group, a halogen-substituted C_{1-4} alkyl group and a C_{1-4} alkoxy group.

(6) Among compounds of the formula (1), still more typical compounds may be represented by the following formula:



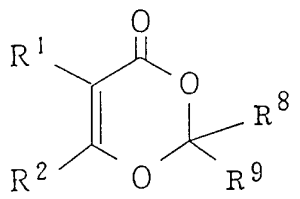
wherein R^{1c} is a phenyl group which may be substituted by a substituent selected from the group consisting of a halogen atom, a C_{1-4} alkyl group, a halogen-substituted C_{1-4} alkyl group and a C_{1-4} alkoxy group, R^{2c} is a C_{1-4} alkyl group which may be substituted by a halogen atom, and R^{3c} is a benzothiazol-2-yl group, which may be substituted by from 1 to 4 substituents selected from the group consisting of a halogen atom, a C_{1-4} alkyl group, a halogen-substituted C_{1-4} alkyl group and a C_{1-4} alkoxy group.

(7) Among cyclic amide compounds of the formula (I), the following compounds are most preferred.

3-[[1-(benzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(5-fluorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(7-chlorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihyro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(4-chlorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(7-bromobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(7-chlorobenzothiazol-2-yl)-1-methyl]ethyl]-6-ethyl-2,3-dihydro-5-phenyl-4H-1,3-oxazin-4-one, 2,3-dihyro-6-methyl-3-[[1-(7-methylbenzothiazol-2-yl)-1-methyl]ethyl]-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(4-fluorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, or 3-[[1-(4,7-dichlorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one.

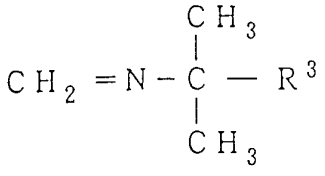
The compound of the formula (I) can be prepared, for example, by a process represented by the following reaction (A).

(A)

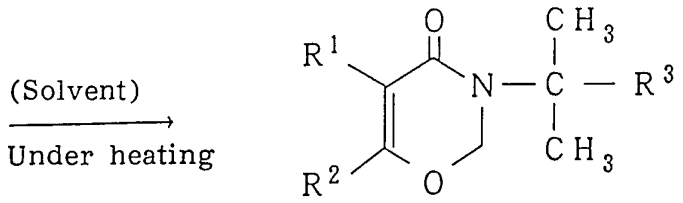


(II)

+



(III)



(I)

In the above formulas, R^1, R^2 and R^3 are as defined above, and each of R^8 and R^9 is an alkyl group.

The reaction (A) is usually carried out in the presence of a solvent. The solvent may, for example, be an aromatic hydrocarbon such as benzene, toluene, xylene or chlorobenzene; a cyclic or non-cyclic aliphatic hydrocarbon such as carbon tetrachloride, chloroform, dichloromethane, dichloroethane, trichloroethane, hexane or cyclohexane; an ether such as dioxane or tetrahydrofuran; an ester such as methyl acetate or ethyl acetate; or an aprotic polar solvent such as dimethylsulfoxide, sulforane, dimethylacetamide, dimethylformamide, N-methylpyrrolidone or pyridine.

The reaction (A) is carried out under heating, and the reaction temperature is usually from 30 to 300°C, preferably from 50 to 200°C. The reaction time is usually from 0.01 to 100 hours, preferably from 0.01 to 20 hours.

The compound of the formula (II) is a known compound or can readily be produced by known methods disclosed in e.g. Japanese Unexamined Patent Publications No. 89485/1992 and No. 172485/1984, Chem. Pharm. Bull., vol.

31, No. 6, 1896-1901 (1983), and ditto, vol. 32, No. 10, 3848-3856 (1984).

The compound of the formula (III) can be produced, for example, from R^3-CN or a nitrile compound of the formula (VII). The compound of the formula (III) can be obtained by introducing a methylene group to the amino group of the compound of the formula (IV). For the introduction of this methylene group, conventional methylene-introducing reactions commonly used in this field can widely be employed. For example, a method of using formalin or paraformaldehyde may be mentioned.

The compound of the formula (IV) may be synthesized from R^3-CN by means of a Grignard reagent or other reagents commonly used in this field, or may be prepared by converting a carbonylamide compound of the formula (V) to an amino compound by a Hofmann rearrangement reaction. Here, for the Hofmann rearrangement reaction, a suitable method commonly used in this field can be employed. Such a Hofmann rearrangement reaction may be conducted, for example, by treatment in the presence of an alkali-hypohalite.

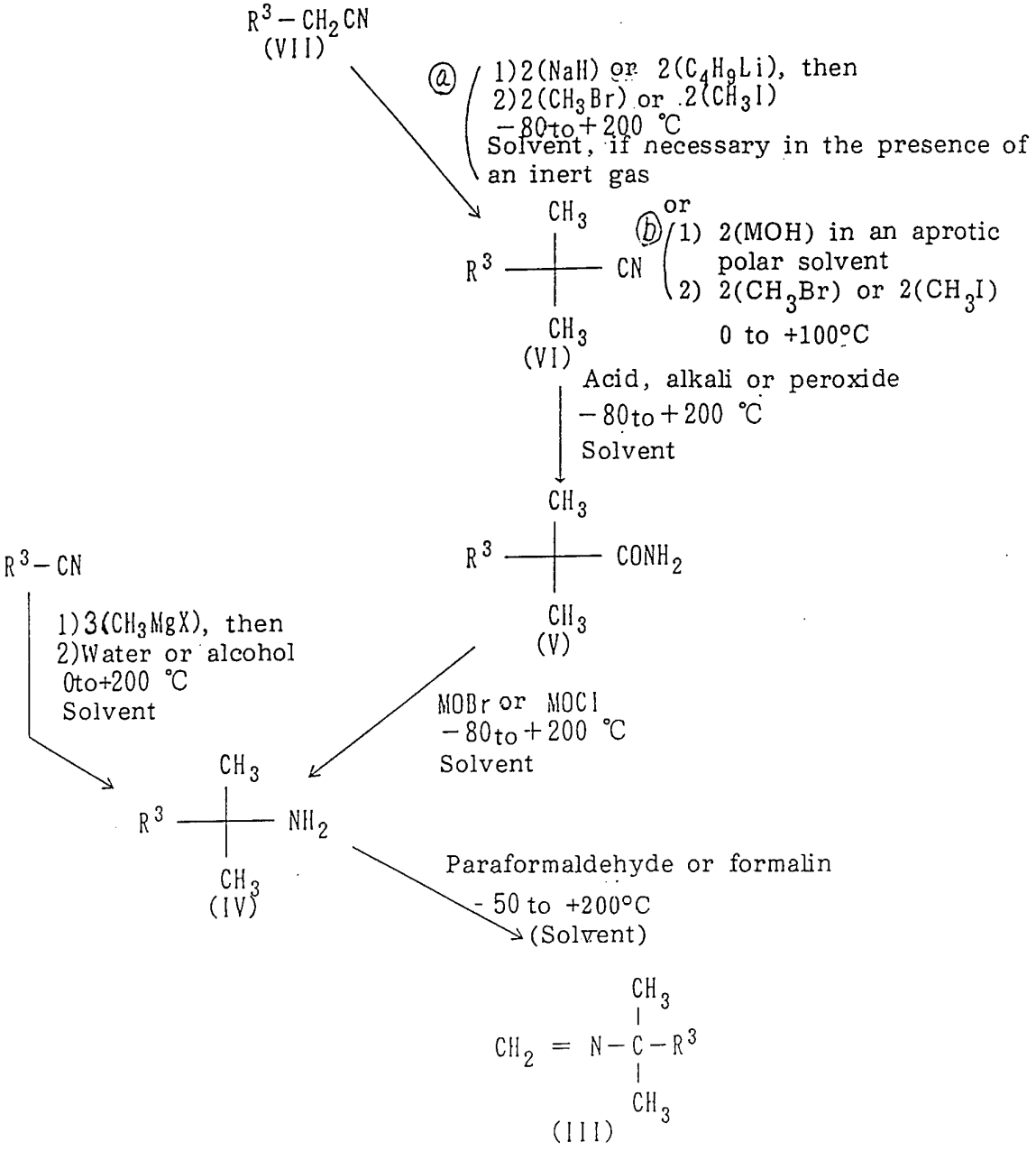
The carbonylamide compound of the formula (V) can be obtained by hydrolyzing the nitrile group of the compound of the formula (VI). This hydrolysis may be carried out by a method commonly used in this field. For example, treatment with an acid, alkali or peroxide in the presence of water, may be mentioned, and reagents as

described hereinafter may be employed for this purpose.

The nitrile compound of the formula (VI) can be prepared by introducing a methyl group to the methylene group adjacent to the nitrile group of the compound of the formula (VII). For the introduction of the methyl group, a suitable method commonly used in this field, may be employed. For example, a methyl halide may be reacted in the presence of a strong alkali. Otherwise, a methyl halide may be reacted in the presence of an alkali metal compound.

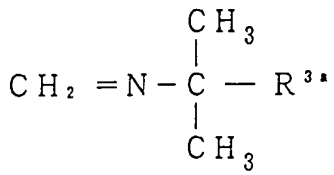
For example, the compound of the formula (III) can be produced by a method as represented by the following reaction (B). In some cases, this compound of the formula (III) may exist in an equilibrium state with its trimer.

(B)


R3 – CH2CN
(VII)
ⓐ 1)2(NaH) or 2(C4H9Li), then
2)2(CH3Br) or 2(CH3I)
–80to+200 ℃
Solvent, if necessary in the presence of an inert gas
or
ⓑ 1) 2(MOH) in an aprotic polar solvent
2) 2(CH3Br) or 2(CH3I)
0 to +100°C
(VI)
Acid, alkali or peroxide
–80to+200 ℃
Solvent
(V)
R3 – CN
1)3(CH3MgX), then
2)Water or alcohol
0to+200 ℃
Solvent
MOBr or MOCl
–80to+200 ℃
Solvent
(IV)
Paraformaldehyde or formalin
- 50 to +200°C
(Solvent)
(III)

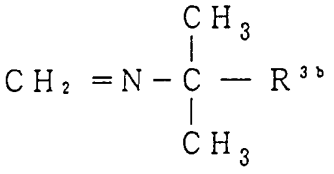
In the above formulas, R^3 is as defined above, M is a sodium atom or a potassium atom, and X is chlorine, bromine or iodine, provided that R^3 in the formula R^3-CN does not include a group substituted by a bromine atom or an iodine atom.

Among compounds of the formula (III), typical compounds may be represented by the following formula:



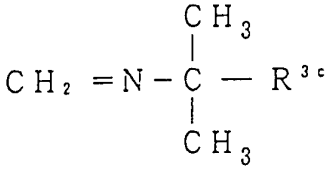
wherein R^{3a} is as defined above.

Among compounds of the formula (III), more typical compounds may be represented by the formula:



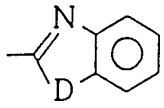
wherein R^{3b} is as defined above.

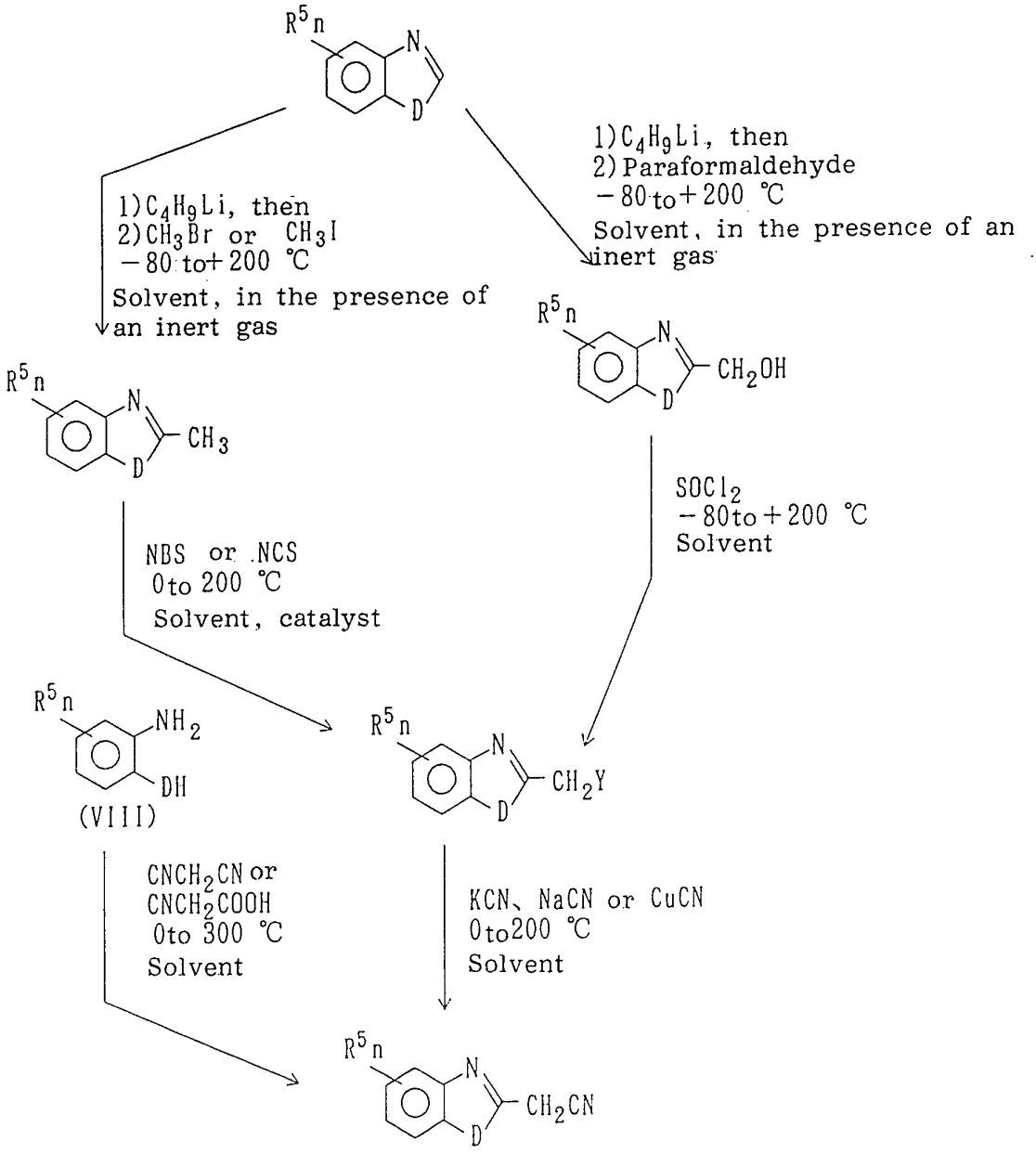
Further, among compounds of the formula (III), still more typical compounds may be represented by the following formula:



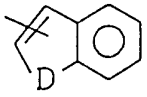
wherein R^{3c} is as defined above.

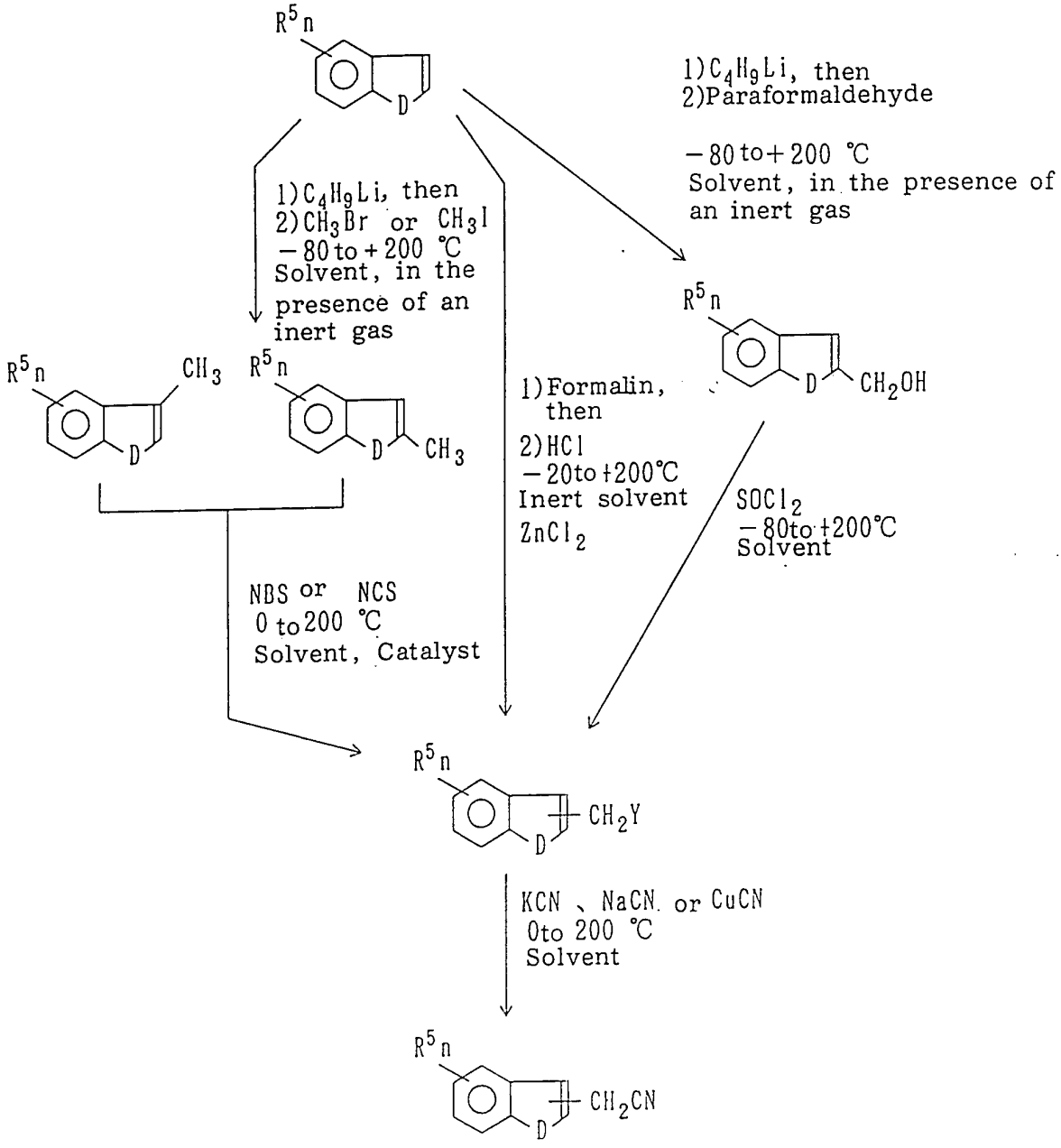
In the above reaction (B), the compound of the formula (VII) can be produced, for example, by methods represented by the following reactions (C) to (F).

(C) When R^3 is  which may be substituted:

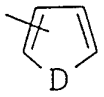


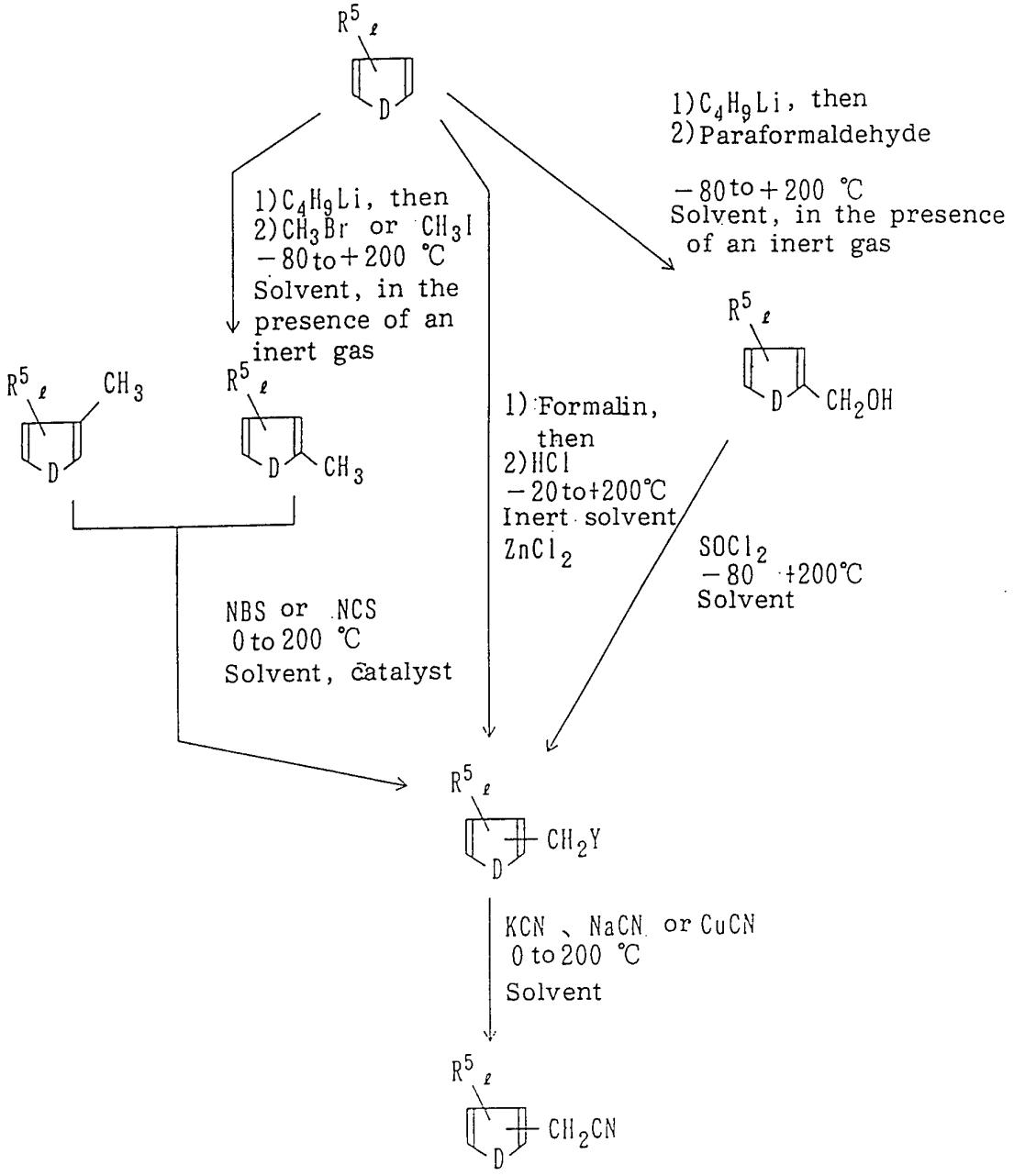
(VII-1)

(D) When R^3 is  which may be substituted:

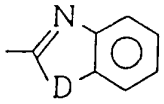


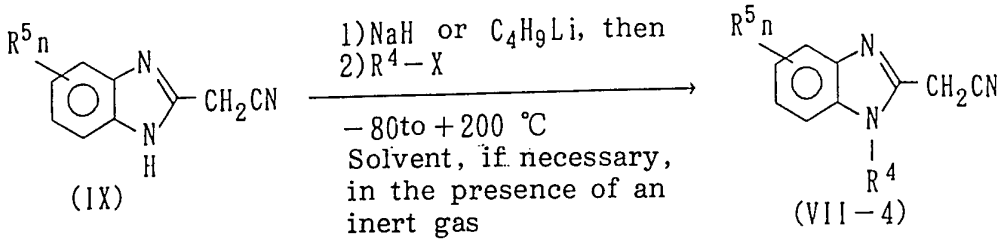
(VII-2)

(E) When R^3 is  which may be substituted:

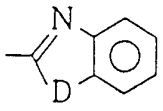


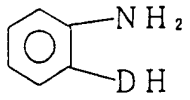
(VII-3)

(F) When R^3 is  which may be substituted, and D is $-N(R^4)-$, wherein R^4 is an alkyl group:

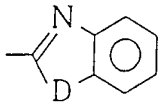


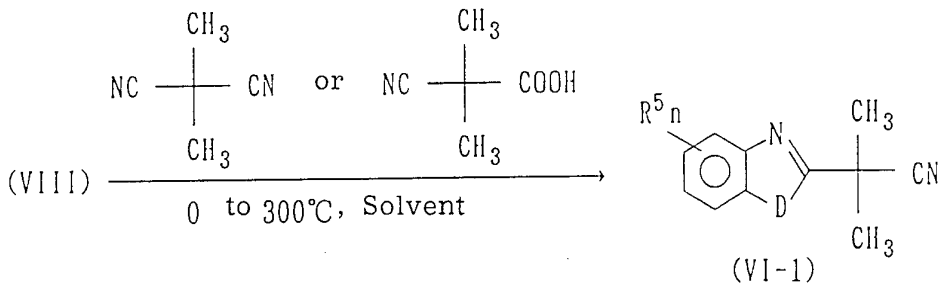
In the formulas for the reactions (C) to (F), D, X and R^4 are as defined above, R^5 is a hydrogen atom; a halogen atom; an alkyl group which may be substituted by a halogen atom; an alkoxy group which may be substituted by a halogen atom; an alkylthio group which may be substituted by a halogen atom; an alkylsulfonyl group which may be substituted by a halogen atom; an aryl group which may be substituted by a halogen atom or a halogenoalkyl group; a heteroaryl group which may be substituted by a halogen atom or a halogenoalkyl group; an aryloxy group which may be substituted by a halogen atom or a halogenoalkyl group; or a heteroaryloxy group which may be substituted by a halogen atom or a halogenoalkyl group, Y is a chlorine atom or a bromine atom, n is an integer of from 1 to 4, and ℓ is an integer of from 1 to 3. When n or ℓ is an integer of 2 or more, the plurality of R^5 may be the same or different. NBS represents N-bromosuccinimide, and NCS represents N-chlorosuccinimide.

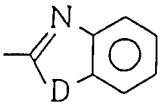
When R^3 is  which may be substituted, the compounds of the formulas (VII) to (III) can be produced by reacting a hetero ring-forming reagent commonly used in this field, such as a nitrile compound, with the compound of the formula:

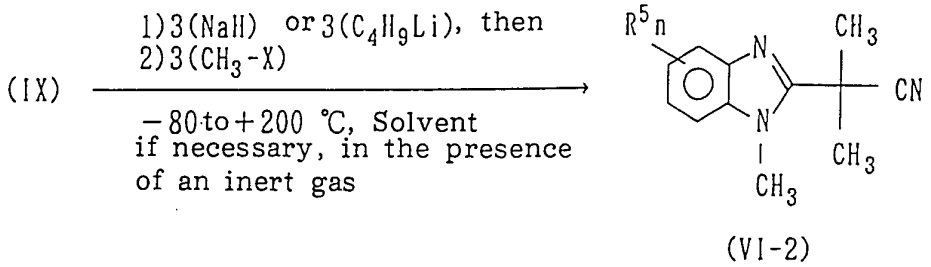


wherein the benzene ring may be substituted. Here, for this hetero ring-forming reaction, a method useful in this field may suitably be employed. For example, the compound of the formula (VII) can be produced by using $CNCH_2CN$ or $CNCH_2COOH$ as the hetero ring-forming reagent under the conditions commonly known in this field. The compound of the formula (VI) can be prepared by a method represented by the reaction (G) or (H), and the compound of the formula (V-1) can be prepared by a method represented by the reaction (I). Further, the compound of the formula (IV) can be prepared by a method represented by the reaction (J).

(G) When R^3 is  which may be substituted:

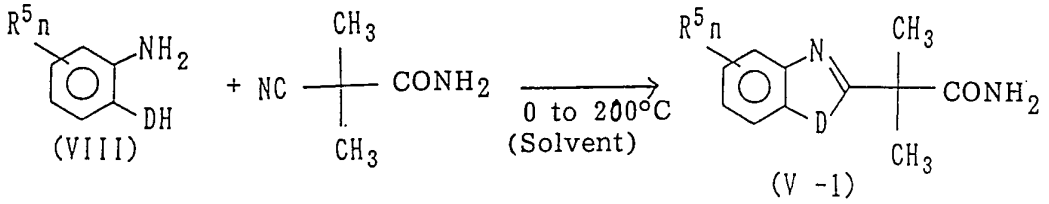


(H) When R^3 is  which may be substituted, and D is $-N(CH_3)-$:



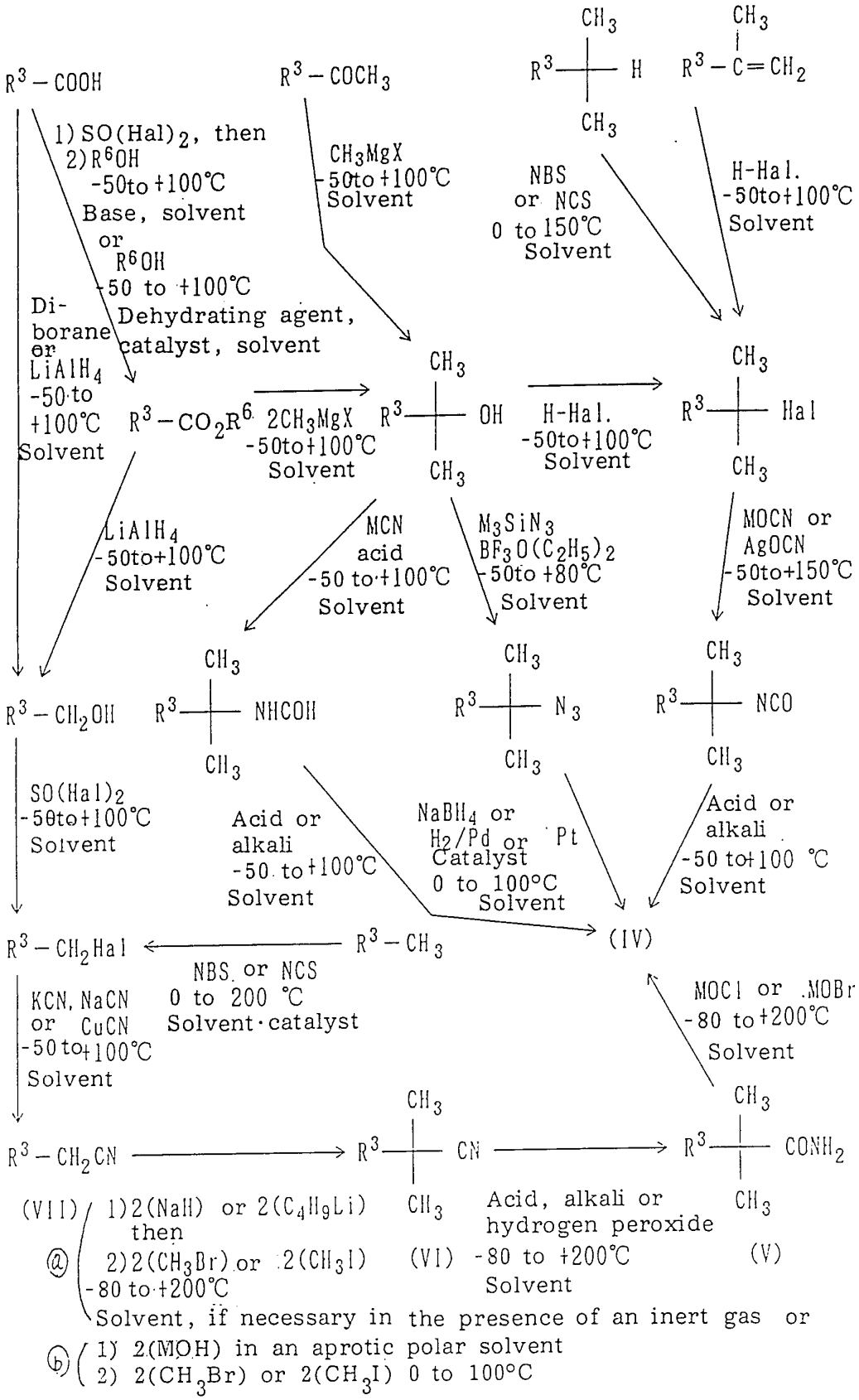
(VI-2)

(I)



(V -1)

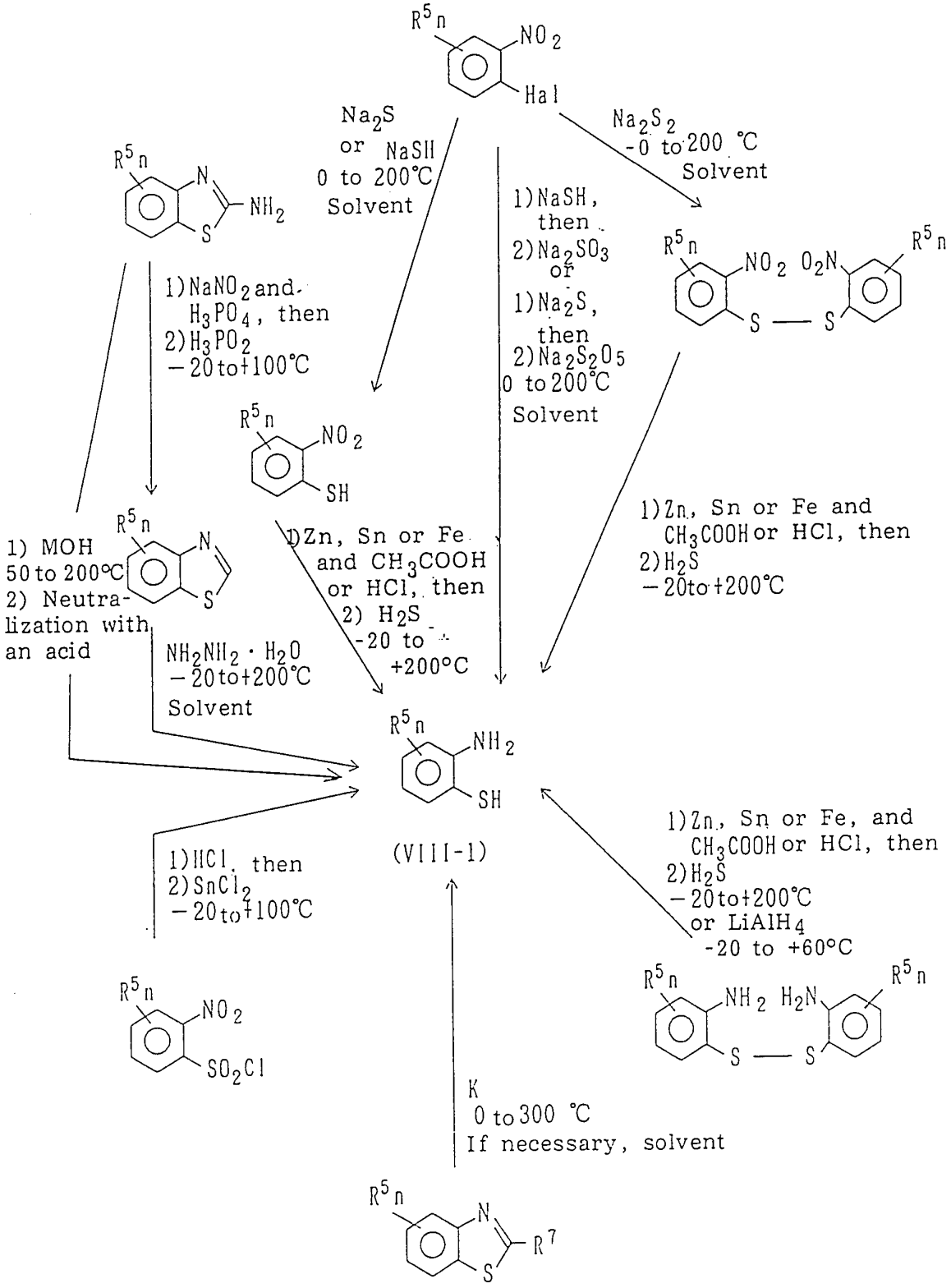
(J)

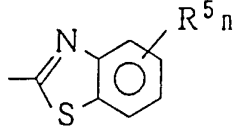

R3 – COOH
R3 – COCH3
R3 – C(CH3)H – H
R3 – C(CH3)=CH2
1) SO(Hal)2, then 2) R6OH -50to+100℃ Base, solvent or R6OH -50 to +100℃
CH3MgX -50to+100℃ Solvent
NBS or NCS 0 to 150℃ Solvent
H-Hal. -50to+100℃ Solvent
Di-borane or LiAlH4 -50 to +100℃ Solvent
Dehydrating agent, catalyst, solvent
R3 – CO2R6
2CH3MgX -50to+100℃ Solvent
R3 – C(CH3)2 – OH
H-Hal. -50to+100℃ Solvent
R3 – C(CH3)2 – Hal
LiAlH4 -50to+100℃ Solvent
MCN acid -50 to+100℃ Solvent
M3SiN3 BF3O(C2H5)2 -50to +80℃ Solvent
MOCN or AgOCN -50to+150℃ Solvent
R3 – CH2OH
R3 – C(CH3)2 – NHCOH
R3 – C(CH3)2 – N3
R3 – C(CH3)2 – NCO
SO(Hal)2 -50to+100℃ Solvent
Acid or alkali -50 to+100℃ Solvent
NaBH4 or H2/Pd or Pt Catalyst 0 to 100℃ Solvent
Acid or alkali -50 to+100 ℃ Solvent
R3 – CH2Hal
R3 – CH3
(IV)
NBS or NCS 0 to 200 ℃ Solvent·catalyst
KCN, NaCN or CuCN -50 to+100℃ Solvent
MOCl or MOBr -80 to+200℃ Solvent
R3 – CH2CN
R3 – C(CH3)2 – CN
R3 – C(CH3)2 – CONH2
(VII)
ⓐ 1) 2(NaH) or 2(C4H9Li) then 2) 2(CH3Br) or 2(CH3I) -80 to+200℃ Solvent, if necessary in the presence of an inert gas or
ⓑ 1) 2(MOH) in an aprotic polar solvent 2) 2(CH3Br) or 2(CH3I) 0 to 100℃
(VI)
Acid, alkali or hydrogen peroxide -80 to +200℃ Solvent
(V)

In the formulas for the reactions (G), (H) and (I), D, R^5, X and n are as defined above. In the formulas for the reaction (J), R^3, M, X, NBS and NCS are as defined above, Hal is a halogen atom, and R^6 is a halogen atom, an alkoxy group, a benzyloxy group or a phenoxy group.

Further, among compounds of the formula (VIII), those wherein D is a sulfur atom, can be prepared, for example, by a method represented by the reaction (K).

(K)


R^5n
NO_2
Hal
Na_2S
or NaSH
0 to 200°C
Solvent
Na_2S_2
-0 to 200 °C
Solvent
1) NaSH, then
2) Na_2SO_3 or
1) Na_2S, then
2) $Na_2S_2O_5$
0 to 200°C
Solvent
NO_2 O_2N
S — S
NH_2
1) $NaNO_2$ and H_3PO_4, then
2) H_3PO_2
−20 to +100°C
SH
1) Zn, Sn or Fe and CH_3COOH or HCl, then
2) H_2S
−20 to +200°C
1) MOH
50 to 200°C
2) Neutralization with an acid
1) Zn, Sn or Fe and CH_3COOH or HCl, then
2) H_2S
-20 to +200°C
$NH_2NH_2 \cdot H_2O$
−20 to +200°C
Solvent
(VIII-1)
1) HCl, then
2) $SnCl_2$
−20 to +100°C
1) Zn, Sn or Fe, and CH_3COOH or HCl, then
2) H_2S
−20 to +200°C
or $LiAlH_4$
-20 to +60°C
SO_2Cl
H_2N
K
0 to 300 °C
If necessary, solvent
R^7

In the formulas for the reaction (K), Hal, R^5 and n are as defined above, and R^7 is a hydrogen atom, a chlorine atom, a phenyl group, -OH, $-NH_2$ or 

The solvent or the inert solvent to be used in the above reactions (B) to (K) may suitably be selected from aromatic hydrocarbons such as benzene, toluene, xylene and chlorobenzene; cyclic and non-cyclic aliphatic hydrocarbons such as chloroform, carbon tetrachloride, methylene chloride, dichloroethane, trichloroethane, hexane and cyclohexane; ethers such as diethyl ether, dioxane and tetrahydrofuran; nitriles such as acetonitrile, propionitrile and acrylonitrile; esters such as methyl acetate and ethyl acetate; aprotic polar solvents such as dimethylsulfoxide, sulforane, dimethylacetamide, dimethylformamide, N-methylpyrrolidone and pyridine; ketones such as acetone and methyl ethyl ketone; alcohols such as methanol, ethanol and tert-butanol; organic and inorganic acids such as acetic acid, formic acid and hydrochloric acid; and water. The base may suitably be selected from carbonates such as potassium carbonate and sodium carbonate; hydrogen carbonates such as potassium hydrogencarbonate and sodium hydrogencarbonate; metal hydroxides such as potassium hydroxide and sodium hydroxide; tertiary amines such as triethylamine; and pyridines such as pyridine and 4-dimethylaminopyridine. The inert gas may suitably be

selected from such gases as argon, helium and nitrogen. As the dehydrating agent, N,N'-dicyclohexylcarbodiimide may, for example, be mentioned. The catalyst may, for example, be 2,2'-azobisisobutyronitrile, metachloroperbenzoic acid or light. The acid may, for example, be formic acid, hydrochloric acid, hydrobromic acid or sulfuric acid. The alkali may, for example, be potassium hydroxide, sodium hydroxide or sodium metal. The peroxide may, for example, be hydrogen peroxide.

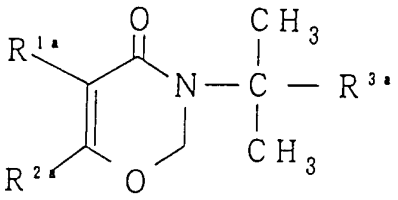
The reducing agent for the nitro group in the reaction (K) may, for example, be tin chloride, sodium sulfide (Na_2S, Na_2S_2, Na_2S_x), sodium hydrosulfide (NaSH), sodium dithionite ($Na_2S_2O_4$), ammonium sulfide ($(NH_4)_2S$) or hydrazine in addition to those described in the above reaction scheme. In a case where a catalytic reduction method is employed, platinum dioxide, Raney nickel, palladium-carbon, rhodium, iron, copper or a hydrogen-transfer catalyst may, for example, be used for the reaction with hydrogen, ammonium formate, alcohol, cyclohexene, formic acid, triethylammonium formate or ammonium chloride.

The compound of the present invention exhibits excellent herbicidal effects when used as an active ingredient of a herbicidal composition. Gramineous weeds include Echinochloa such as barnyardgrass (Echinochloa oryzicola) or cockspur grass (Panicum crus-galli), Brachiaria such as alexandergrass (Brachiaria

plantaginea) or paragrass (Panicum purpurascen), and Leptochloa such as sprangletop (Leptochloa chinensis) or red sprangletop (Leptochloa panicea). As will be apparent from the Test Examples given hereinafter, it is effective especially for controlling noxious weeds growing in a paddy field, e.g. gramineous weeds such as barnyardgrass (Echinochloa crusgalli) selectively and at a low dose without presenting a phytotoxicity to rice. As compared with conventional herbicides, it is superior in the persistency of the herbicidal effects, whereby constant herbicidal effects can be obtained over a long period of time. Further, when it is used in admixture with or in combination with other herbicidally effective component as will be described hereinafter, it is possible to selectively control not only gramineous weeds but also other weeds including Cyperaceae such as japanese bulrush (Scirpus juncoides), flatsedge (Cyperus serotinus), small-flower umbrellaplant (Cyperus difformis), slender spikerush (Eleocharis acicularis), and water chestnut (Eleocharis kuroguwai), alismataceae such as japanese ribbon wapato (Sagittaria pygmaea), arrow-head (Sagittaria trifolia), and narrowleaf waterplantain (Alisma canaliculatum), pontederiaceae such as monochoria (Monochoria vaginalis) and monochoria species (Monochoria korsakowii), scrophulariaceae such as false pimpernel (Lindernia pyxidaria) and abunome (Dopatrium junceum), and lythraceae such as toothcup

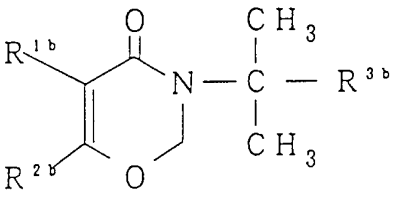
(Rotala indica) and red stem (Ammania multiflora), for a long period of time without presenting a phytotoxicity to rice. Thus, such a composition is useful as a paddy field herbicidal composition.

Among compounds of the formula (I), those represented by the following formula:



wherein R^{1a}, R^{2a} and R^{3a} are as defined above, are capable of controlling noxious weeds especially in a paddy rice field, e.g. gramineous weeds such as barnyardgrass, selectively at a low dose without giving any phytotoxicity to rice plants, and they are excellent in the persistency of the herbicidal effects as compared with conventional herbicides, whereby constant herbicidal effects can be expected over a long period of time.

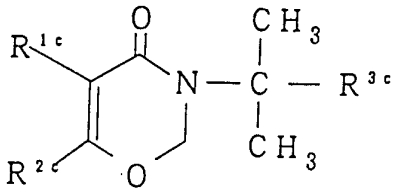
Further, among compounds of the formula (I), those represented by the following formula:



wherein R^{1b}, R^{2b} and R^{3b} are as defined above, are capable of controlling noxious weeds especially in a paddy rice field, e.g. gramineous weeds such as barnyardgrass,

selectively at a low dose without giving any phytotoxicity to rice plants, and they are excellent in the persistency of the herbicidal effects as compared with conventional herbicides, whereby constant herbicidal effects can be expected over a long period of time.

Furthermore, among compounds of the formula (I), still typical compounds of the formula:



wherein R^{1c}, R^{2c} and R^{3c} are as defined above, are capable of controlling noxious weeds especially in a paddy rice field, e.g. gramineous weeds such as barnyardgrass, selectively at a low dose without giving any phytotoxicity to rice plants, and they are excellent in the persistency of the herbicidal effects as compared with conventional herbicides, whereby constant herbicidal effects can be expected over a long period of time.

A herbicidal composition containing the compound of the present invention may be applied to various places including not only paddy fields but also upland fields and non-agricultural fields such as forests, farm roads, open grounds and factory sites. Further, the manner of application may suitably be selected from soil treatments and foliage treatments.

The compound of the present invention is usually

mixed with various agricultural adjuvants and formulated into various formulations such as granules, water dispersible granules, wettable powders, aqueous suspensions, oil suspensions, aqueous solutions, emulsifiable concentrates, tablets or capsules. It can be formulated into any formulations konwn in this field so long as the object of the present invention is satisfied. Such agricultural adjuvants include solid carriers such as diatomaceous earth, hydrated lime, calcium carbonate, talc, white carbon, kaoline, bentonite, jeaklite, clay, and starch; solvents such as water, toluene, xylene, solvent naphtha, dioxane, acetone, isophorone, methyl isobutyl ketone, chlorobenzene, cyclohexane, dimethylsulfoxide, dimethylformamide, N-methyl-2-pyrrolidone, and alcohol; spreaders and surfactants such as sodium alkyl sulfate, sodium alkylbenzene sulfonate, sodium lignin sulfonate, polyoxyethylene alkylaryl ether sulfate, polyoxyethylene glycol alkyl ether, polyoxyethylene lauryl ether, polyoxyethylene alkylaryl ether, an ester of polyoxyethylene aliphatic acid, and an ester of polyoxyethylene sorbitan aliphatic acid; vegetable and mineral oils such as olive oil, kapok oil, castor oil, palm oil, camellia oil, coconut oil, sesame oil, corn oil, rice bran oil, peanut oil, cotton seed oil, soybean oil, rape seed oil, linseed oil, tung oil, and liquid

paraffins. Such adjuvants may be selected from those known in this field so long as the object of the present invention is satisfied. Further, other conventional adjuvants such as bulking agents, thickeners, anti-settling agents, anti-freezing agents, dispersion stabilizers, phytotoxicity-reducing agents, and antifungus agents may be used. Here, the weight ratio of the compound of the present invention to the agricultural adjuvants is usually from 0.1:99.9 to 90:10, preferably from 0.2:99.8 to 80:20.

The dose of the herbicidal composition of the present invention can not generally be defined, since it may vary depending upon the weather condition, the soil condition, the type of the formulation, the types of the weeds to be controlled, the season for the application, etc. However, it is usually applied so that the compound of the present invention would be applied in an amount of from 0.1 to 40 g/a, preferably from 0.5 to 20 g/a.

The herbicidal compositions of the present invention may be used in admixture with or in combination with other agricultural chemicals, fertilizers or phytotoxicity-reducing agents. Said other agricultural chemicals include, for example, herbicides, fungicides, antibiotics, plant hormones and insecticides. In such a case, they may exhibit even better effects or activities. For example, when the compounds of the present invention are used in admixture with or in combination with one or

more other herbicidally active components as will be described hereinafter, synergistic effects may be obtained.

The ratio of the compound of the present invention to such other herbicidally active component can not generally be defined, since it varies depending upon the weather condition, the soil condition, the type of the formulation, the season for the application, the manner of the application, etc. However, at least one such other herbicidally active component may be incorporated usually in an amount of from 0.01 to 100 parts by weight, preferably from 0.02 to 60 parts by weight, per part by weight of the compound of the present invention. The total dose of all the active ingredients is usually from 0.2 to 100 g/a, preferably from 0.5 to 50 g/a.

Now, specific examples of such other herbicidally active components will be given below.

Diphenyl ether compounds such as

2,4,6-trichlorophenyl-4-nitrophenyl ether (common name: chlornitrofen),

5-(2,4-dichlorophenoxy)-2-nitroanisole (common name: chlomethoxyfen), and

methyl 5-(2,4-dichlorophenoxy)-2-nitrobenzoate (common name: bifenox),

heterocyclic compounds such as

5-t-butyl-3-(2,4-dichloro-5-isopropoxyphenyl)-1,3,4-oxadiazol-2(3H)-one (common name: oxadiazon),

3-[(5-cyclopentyloxy-4-chloro-2-fluoro)phenyl]-5-isopropylidene-1,3-oxazolidin-2,4-dione (compound disclosed in papers presented at the 17th meeting of Nippon Noyaku Gakkai, p. 48 (1992)),

S,S-dimethyl 2-difluoromethyl-4-(2-methylpropyl)-6-(trifluoromethyl)-3,5-pyridinethioate (common name: dithiopyr),

exo-1-methyl-4-(1-methylethyl)-2-[(2-methylphenyl)methoxy]-7-oxabicyclo[2,2.1]heptane (common name: cinmethylin),

3,7-dichloroquinolin-8-carboxylic acid (common name: quinclorac),

1-(diethylcarbamoyl)-3-(2,4,6-trimethylphenyl-sulfonyl)-1,2,4-triazole (compound disclosed in WEED RESEARCH (1991) vol. 36, separate number I, papers presented at the 30th meeting, p. 27),

3-isopropyl-2,1,3-benzothiadiazin-4-one-2,2-dioxide (common name: bentazone) and its sodium salt,

2,3-dihydro-3,3-dimethylbenzofuran-5-yl ethanesulfonate (common name: benfuresate), and

methyl 2-[(4,6-dimethoxypyrimidin-2-yl)oxy]-6-[1-(N-methoxyimino)ethyl]benzoate (compound disclosed in Japanese Unexamined Patent Publication No. 134073/1992),

anilide compounds such as

2-chloro-2',6'-diethyl-N-(2-propoxyethyl)acetanilide (common name: pretilachlor),

2-chloro-N-[(3-methoxy-2-thienyl)methyl]-2'6'-dimethylacetanilide (common name: thenylchlor),
2',3'-dichloro-4-ethoxymethoxybenzanilide (common name: etobenzanide),
2-[(benzothiazol-2-yl)oxy]-N-methylacetanilide (common name: mefenacet),
2-(2-naphthoxy)propionanilide (common name: naproanilide), and
2-(2,4-dichloro-3-methylphenoxy)propionanilide (common name: clomeprop),
carbamate compounds such as
S-(4-chlorobenzyl) N,N-diethylthiocarbamate (common name: thiobencarb),
S-ethyl hexahydro-1H-azepin-1-carbothioate (common name: molinate),
S-(1-methyl-1-phenylethyl)piperidine-1-carbothioate (common name: dimepiperate),
S-benzyl N-(1,2-dimethylpropyl)-N-ethylthiocarbamate (common name: esprocarb), and
O-(3-t-butylphenyl) N-(6-methoxy-2-pyridyl)-N-methylthiocarbamate (common name: pyributicarb),
phenoxyalkane compounds such as
(R)-n-butyl 2-[4-(4-cyano-2-fluorophenoxy)phenoxy]-propionate (compound disclosed in Japanese Unexamined Patent Publication No. 65201/1993),
sulfonylurea compounds such as
methyl 2-[[[[[(4,6-dimethoxypyrimidin-2-

yl)amino]carbonyl]amino]sulfonyl]methyl]benzoate (common name: bensulfuron-methyl),

N-[[(4,6-dimethoxypyrimidin-2-yl)amino]carbonyl]-4-ethoxycarbonyl-1-methyl-5-pyrazolesulfonamide (common name: pyrazosulfuron-ethyl),

1-(2-chloroimidazo[1.2-a]pyridin-3-ylsulfonyl)-3-(4,6-dimethoxy-2-pyrimidinyl)urea (common name: imazosulfuron),

3-(4,6-dimethoxy-1,3,5-triazin-2-yl)-1-[2-(2-methoxyethoxy)phenylsulfonyl]urea (common name: cinosulfuron),

5-(2,2-difluoro-2-chloroethoxy)-N-[[(4,6-dimethoxypyrimidin-2-yl)amino]carbonyl]-3-methyl-4-isothiazolesulfonamide (compound disclosed in Japanese Unexamined Patent Publication No. 190887/1988),

N-[[(4,6-dimethoxy-2-pyrimidinyl)amino]carbonyl]-1-methyl-4-(2-methyl-2H-tetrazol-5-yl)-1H-pyrazole-5-sulfonamide (common name: azimsulfuron), and

1-[[2-(cyclopropylcarbonyl)phenyl]sulfamoyl]-3-(4,6-dimethoxy-2-pyrimidinyl)urea (compound disclosed in Japanese Unexamined Patent Publication No. 224567/1992),

pyrazole compounds such as

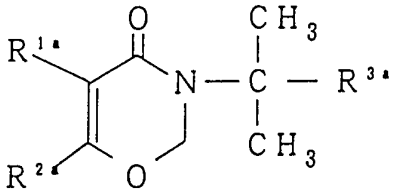
2-[4-(2,4-dichlorobenzoyl)-1,3-dimethylpyrazol-5-yloxy]acetophenone (common name: pyrazoxyfen),

4-(2,4-dichlorobenzoyl)-1,3-dimethyl-5-pyrazolyl-p-

toluenesulfonate (common name: pyrazolate), and
2-[4-(2,4-dichloro-m-toluoyl)-1,3-dimethylpyrazol-5-yloxy]-4'-methylacetophenone (common name: benzofenap),
benzylamide compounds such as
2-bromo-N-(α,α-dimethylbenzyl)-3,3-dimethylbutylamide (common name: bromobutide),
urea compounds such as
1-(α,α-dimethylbenzyl)-3-(p-tolyl)urea (common name: daimuron), and
1-(2-chlorobenzyl)-3-(α,α-dimethylbenzyl)urea (common name: cumyluron),
triazine compounds such as
2-methylthio-4,6-bis(methylamino)-s-triazine (common name: simetryn), and
2-methylthio-4-ethylamino-6-(1',2'-diethylpropylamino)-s-triazine (common name: dimethametryn),
phenyloxy comopunds such as
2,4-dichlorophenoxy acetic acid, and its salt and ester (common name: 2,4-D),
4-(4-chloro-o-tolyloxy)butyric acid, and its salt and ester (common name: MCPB), and
S-ethyl 4-chloro-2-methylphenoxythioacetate (common name: MCPA-thioethyl), and
other compounds such as
2-[2-(3-chlorophenyl)-2,3-epoxypropyl]-2-ethylindan-

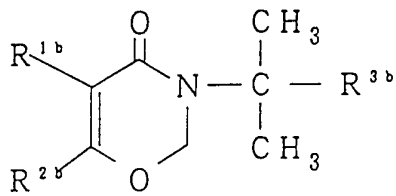
1,3-dione (compound disclosed in Japanese Unexamined Patent Publication No. 304043/1990).

Among compounds of the formula (I), those represented by the following formula:



wherein R^{1a}, R^{2a} and R^{3a} are as defined above, are expected to provide more excellent effects and activities when used in admixture with or in combination with one or more other herbicidal components as mentioned above, whereby they pave a way to control noxious weeds grown in a paddy field, selectively at a low dose without giving any phytotoxicity to rice plants, and constant herbicidal effects are expected to be obtained over a long period of time.

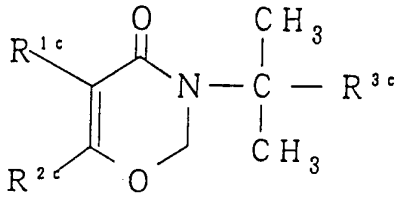
Further, among compounds of the formula (I), those represented by the following formula:



wherein R^{1b}, R^{2b} and R^{3b} are as defined above, are expected to provide more excellent effects and activities when used in admixture with or in combination with one or more other herbicidal components as mentioned above,

whereby they pave a way to control noxious weeds grown in a paddy field, selectively at a low dose without giving any phytotoxicity to rice plants, and constant herbicidal effects are expected to be obtained over a long period of time.

Furthermore, among compounds of the formula (I), still more typical compound of the formula:



wherein R^{1c}, R^{2c} and R^{3c} are as defined above, are expected to provide more excellent effects and activities when used in admixture with or in combination with one or more other herbicidal components as mentioned above, such as a diphenyl ether compound, a heterocyclic compound, an anilide compound, a carbamate compound, a phenoxy alkanoic acid compound, a sulfonylurea compound, a pyrazole compound, a benzylamide compound, a urea compound, a triazine compound, a phenoxy compound and other compounds, whereby they pave a way to control noxious weeds grown in a paddy field, selectively at a low dose without giving any phytotoxicity to rice plants, and constant herbicidal effects are expected to be obtained over a long period of time.

Now, the present invention will be described in further detail with reference to Examples. However, it

should be understood that the present invention is by no means restricted to such specific Examples. Firstly, specific Preparation Examples of the compounds of the present invention will be described.

PREPARATION EXAMPLE 1

Preparation of 3-[[1-(benzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one (Compound No. 1 as identified hereinafter)

(1) 5.1 g of sodium hydride was put into 200 mℓ of dry tetrahydrofuran, and the mixture was cooled to -10 to 0°C to suspend sodium hydride. Then, a solution having 10 g of benzothiazol-2-yl acetonitrile dissolved in 50 mℓ of tetrahydrofuran, was dropwise added thereto at a temperature of from 0 to 10°C, and the mixture was stirred at 70°C for one hour. Then, it was cooled to a temperature of from -10 to 0°C. Then, 17.26 g of iodomethane was dropwise added thereto at a temperature of from -10 to +10°C, and the mixture was reacted at room temperature for 2 nights with stirring.

After completion of the reaction, the reaction product was put into ice water and extracted with ethyl acetate. Then, the extract was washed with a sodium chloride aqueous solution, then dried over anhydrous sodium sulfate and filtered. The solvent of the obtained filtrate was distilled off under reduced pressure, and the residue was purified by column chromatography (developing solvent: toluene) to obtain 10.7 g of 2-

benzothiazol-2-yl)-2-methylpropionitrile having a melting point of from 44 to 46°C.

(2) 10.25 g of 2-(benzothiazol-2-yl)-2-methylpropionitrile was dissolved in 70 mℓ of tert-butanol, and 5.7 g of potassium hydroxide powder was added thereto. The mixture was reacted at 80°C for one hour with stirring.

After completion of the reaction, the the reaction product was cooled to room temperature, put into water and extracted with ethyl acetate. Then, the extract was washed with a sodium chloride aqueous solution and dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure. Precipitated crystals were washed with a mixture of ethyl ether/hexane = 1/1 and dried under reduced pressure to obtain 5.09 g of 2-(benzothiazol-2-yl)-2-methylpropionamide having a melting point of from 133 to 134°C.

(3) An aqueous solution having 2.21 g of sodium hydroxide dissolved in 13 mℓ of water, was cooled to a temperature of from -10 to 0°C, and 469 μℓ of bromine was dropwise added thereto. The mixture was stirred for 30 minutes. Then, 2 g of 2-(benzothiazol-2-yl)-2-methylpropionamide was added thereto, and the mixture was reacted at a temperature of from -10 to 0°C for 2.5 hours with stirring and further at room temperature overnight with stirring.

After completion of the reaction, hydrochloric acid

was dropwise added to the reaction product at a temperature of from -10 to +20°C to adjust the pH to 1. Then, toluene was added thereto, and the mixture was back-extracted with dilute hydrochloric acid (10%). Ammonia was dropwise added to the obtained aqueous layer at a temperature of from -10 to +20°C to adjust the pH to 14. The mixture was extracted with methylene chloride and washed with a sodium chloride aqueous solution. Then, it was dried over anhydrous sodium sulfate, and the solvent was distilled off under reduced pressure to obtain 870 mg of oily 1-(benzothiazol-2-yl)-1-methylethylamine.

(4) 1.02 g of 1-(benzothiazol-2-yl)-1-methylethylamine obtained in the same manner as in the above Step (3) and 195 mg of paraformaldehyde were dissolved in 10 mℓ of dry toluene, and the mixture was reacted at a temperature of from 90 to 100°C for 40 minutes and then further reacted under reflux under an azeotropic dehydrating condition for 7 hours to obtain a solution containing [1-(benzothiazol-2-yl)-1-methyl-N-methylene]ethylamine.

(5) The solution obtained in the above Step (4) was cooled, and 1.4 g of 2,2,6-trimethyl-5-phenyl-4H-1,3-dioxin-4-one was added thereto. The mixture was reacted under reflux for 16 hours and then cooled to room temperature. The solvent was distilled off, and the residue was purified by column chromatography (developing solvent: methylene chloride) to obtain 210 mg of the

desired product as white crystals having a melting point of from 166 to 168°C.

PREPARATION EXAMPLE 2

Preparation of 3-[[1-(5-chlorothienyl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one (Compound No. 6 as identified hereinafter)

(1) 64 g of 2-chlorothiophene, 115 mℓ of a 35% formalin aqueous solution and 11.48 g of zinc chloride were dissolved in 150 mℓ of ethyl ether, and the solution was cooled to -5°C. Then, hydrogen chloride gas was introduced and reacted thereto at a temperature of not higher than 10°C over a period of 2 hours.

After completion of the reaction, the reaction product was put into ice water and extracted with ethyl ether. The extract was washed with a sodium chloride aqueous solution. Then, it was dried over anhydrous sodium sulfate, and then ethyl ether was distilled off at a temperature of not higher than 40°C to obtain 73.8 g of oily brown 2-chloro-5-chloromethylthiophene.

(2) 73.8 g of 2-chloro-5-chloromethylthiophene obtained in the above Step (1), 39.5 g of sodium cyanide, 100 mℓ of acetone and 100 mℓ of water were mixed, and the mixture was reacted at about 60°C for 5.5 hours with stirring.

After completion of the reaction, acetone was distilled off, and the reaction product was extracted with methylene chloride. The extract was washed with a

sodium chloride aqueous solution. Then, it was dried over anhydrous sodium sulfate, and then the solvent was distilled off. The residue was purified by column chromatography (developing solvent: toluene/methylene chloride = 8/2) to obtain 11.37 g of (5-chlorothiophen-2-yl)acetonitrile.

(3) 11.37 g of (5-chlorothiophen-2-yl)acetonitrile obtained in the above Step (2) was dissolved in 300 mℓ of dry tetrahydrofuran, and the solution was cooled to -70°C in an inert gas atmosphere. Then, 104 mℓ of a hexane solution of n-butyl lithium (1.66 M) was dropwise added thereto at a temperature of not higher than -60°C, and the mixture was stirred at a temperature of about -70°C for 1.5 hours. Then, 12.3 mℓ of iodomethane was dropwise added thereto at a temperature of not higher than -55°C, and the mixture was left to stand overnight.

After completion of the reaction, the reaction product was poured into ice water and extracted with ethyl acetate. The extract was washed with a sodium chloride aqueous solution. Then, it was dried over anhydrous sodium sulfate, and then the solvent was distilled off under reduced pressure. The residue was purified by column chromatography (developing solvent: toluene) to obtain 11.2 g oily brown 2-(5-chlorothiophen-2-yl)-2-methylpropionitrile.

(4) 12.3 g of 2-(5-chlorothiophen-2-yl)-2-methylpropionitrile obtained in the above Step (3) and

7.43 g of potassium hydroxide powder were dissolved in 50 mℓ of t-butanol and reacted at 80°C for 2 hours.

After completion of the reaction, the reaction product was cooled to room temperature and extracted with ethyl acetate. The extract was washed a few times with a sodium chloride aqueous solution. Then, it was dried over anhydrous sodium sulfate, and then the solvent was distilled off. The residue was purified by column chromatography (developing solvent: methylene chloride/ethyl acetate = 8/2) to obtain 8.95 g of 2-(5-chlorothiophen-2-yl)-2-methylpropionamide as slightly brown crystals having a melting point of from 90 to 93°C.

(5) 3 g of 2-(5-chlorothiophen-2-yl)-2-methylpropionamide obtained in the above Step (4) was added to an aqueous hypobromous acid solution preliminarily prepared by reacting 30 mℓ of 3N sodium hydroxide and 2.46 g of bromine at 0°C, and the mixture was reacted at a temperature of from 0 to -5°C for 6 hours.

After completion of the reaction, the reaction product was adjusted to pH 1 with hydrochloric acid and washed with toluene. Aqueous ammonia was added to the aqueous layer to make it alkaline and then extracted with methylene chloride. The extract was washed with a sodium chloride aqueous solution. Then, it was dried over anhydrous sodium sulfate. Then, the solvent was distilled off under reduced pressure to obtain 1.6 g of oily 1-(5-chlorothiophen-2-yl)-1-methylethylamine.

(6) 630 mg of 1-(5-chlorothiophen-2-yl)-1-methylethylamine obtained in the above Step (5) and 132 mg of paraformaldehyde were dissolved in 20 mℓ of dry toluene and reacted at a temperature of from 90 to 100°C for 40 minutes and then further reacted under reflux under an azeotropic dehydrating condition for 7 hours to obtain a solution containing [1-(5-chlorothiophen-2-yl)-1-methyl-N-methylene]ethylamine.

(7) The solution obtained in the above Step (6) was cooled. Then, 957 mg of 2,2,6-trimethyl-5-phenyl-4H-1,3-dioxin-4-one was added thereto, and the mixture was reacted under reflux for 18 hours.

After completion of the reaction, the reaction product was cooled to room temperature. Then, the solvent was distilled off, and the residue was purified by column chromatography (developing solvent: toluene/methylene chloride = 1/1) to obtain 499 mg of the desired product as a transparent oily substance.

NMR: (60 MHz, δ: in $CDC\ell_3$) 1.92(9H,s), 5.1(2H,s), 6.75(2H,s), 7.13-7.40(5H,m)

PREPARATION EXAMPLE 3

Preparation of 3-[[1-(4-chlorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one (Compound No. 48 as identified hereinafter)

(1) 9.0 g of 4-chlorobenzothiazole was dissolved in 100 mℓ of ethanol, and 100 mℓ of hydrazine hydrate was added thereto. The mixture was reacted at a refluxing

temperature for 16 hours.

After completion of the reaction, the solvent was distilled off under reduced pressure to obtain 8.0 g of oily 2-amino-3-chlorothiophenol.

(2) 8.0 g of 2-amino-3-chlorothiophenol was dissolved in 15 mℓ of ethanol. Then, 20 mℓ of acetic acid was added to this solution, and then 4.0 g of malononitrile was added thereto. The mixture was reacted at room temperature for 3 hours.

After completion of the reaction, the reaction product was put into water, and precipitated crystals were collected by filtration, then thoroughly washed with water and dried to obtain 8.0 g of (4-chlorobenzothiazol-2-yl)acetonitrile having a melting point of from 119 to 121°C.

(3) 8.0 g of (4-chlorobenzothiazol-2-yl)acetonitrile was dissolved in 100 mℓ of tetrahydrofuran. To this solution, 3.4 g of 60% sodium hydride was gradually added, and then 12 g of methyl iodide was dropwise added thereto. The mixture was reacted at room temperature for 16 hours.

After completion of the reaction, the reaction product was put into water and extracted with ethyl acetate. The extract was dried over anhydrous sodium sulfate. Then, the solvent was distilled off under reduced pressure. The obtained residue was purified by column chromatography (developing solvent: ethyl

acetate/hexane = 1/3) to obtain 6.5 g of 2-(4-chlorobenzothiazol-2-yl)-2-methylpropionitrile having a melting point of from 95 to 97°C.

(4) 6.5 g of 2-(4-chlorobenzothiazol-2-yl)-2-methylpropionitrile was dissolved in 80 mℓ of formic acid. To this solution, hydrogen chloride gas was introduced at a temperature of from 40 to 50°C and reacted for 5 hours.

After completion of the reaction, the reaction mixture was put into water and extracted with ethyl acetate. The extract was dried over anhydrous sodium sulfate. Then, the solvent was distilled off under reduced pressure. The obtained residue was purified by column chromatography (developing solvent: ethyl acetate/hexane = 1/1) to obtain 6.5 g of 2-(4-chlorobenzothiazol-2-yl)-2-methylpropionamide having a melting point of from 92 to 94°C.

(5) 1.2 mℓ of bromine was gradually added at a temperature of from -10 to 0°C to an aqueous solution having 4.7 g of sodium hydroxide dissolved in 60 mℓ of water, and the mixture was reacted for 30 minutes. To the obtained reaction solution, 6.0 g of 2-(4-chlorobenzothiazol-2-yl)-2-methylpropionamide was added, and the mixture was reacted at room temperature for one hour and further reacted at 80°C for one hour.

After completion of the reaction, the reaction product was put into water and extracted with methylene

chloride. The extract was dried over anhydrous sodium sulfate. Then, the solvent was distilled off under reduced pressure to obtain 3.6 g of oily 1-(4-chlorobenzothiazol-2-yl)-1-methylethylamine.

(6) In the same manner as in Steps (4) and (5) in Preparation Example 1, the desired product can be obtained.

PREPARATION EXAMPLE 4

Preparation of 2,3-dihydro-3-[[1-(6-methoxybenzothiazol-2-yl)-1-methyl]ethyl]-6-methyl-5-phenyl-4H-1,3-oxazin-4-one (Compound No. 9 as identified hereinafter)

(1) 3 g of 2-cyano-6-methoxybenzothiazole was dissolved in 100 mℓ of toluene, and 16 mℓ of an ethyl ether solution containing 3 mol/ℓ of methyl magnesium bromide was added thereto. The mixture was reacted at a refluxing temperature for one day. Then, the reaction mixture was returned to room temperature, and 10 mℓ of dry ethanol was added thereto, and the mixture was further reacted. Precipitated crystals were filtered off.

Then, the solvent of the filtration was distilled off, and water was added to the residue, and the mixture was extracted with ethyl acetate. The extract was dried over anhydrous sodium sulfate, and then the solvent was distilled off under reduced pressure. The obtained residue was purified by column chromatography (developing solvent: ethyl acetate) to obtain 0.5 g of oily 1-(6-

methoxybenzothiazol-2-yl)-1-methylethylamine.

(2) 1 g of 1-(6-methoxybenzothiazol-2-yl)-1-methylethylamine obtained in the above Step (1) and 135 mg of paraformaldehyde were dissolved in 10 mℓ of toluene, and the solution was refluxed for one hour. Then, an azeotropic dehydration reaction was carried out for 7 hours to obtain a solution containing [1-(6-methoxybenzothiazol-2-yl)-1-methyl-N-methylene]ethylamine.

(3) The solution obtained in the above Step (2) was cooled. Then, 980 mg of 2,2,6-trimethyl-5-phenyl-4H-1,3-dioxin-4-one was added thereto. The mixture was reacted under reflux overnight.

After completion of the reaction, the reaction mixture was cooled to room temperature. Then, the solvent was distilled off, and the residue was purified by column chromatography (developing solvent: methylene chloride) to obtain 500 mg of the desired product as white crystals having a melting point of from 160 to 162°C.

PREPARATION EXAMPLE 5

Preparation of 2,3-dihydro-6-methyl-3-[[1-methyl-1-(2-pyridyl)]ethyl]-5-phenyl-4H-1,3-oxazin-4-one (Compound No. 8 as identified hereinafter)

(1) 25 g of 2-pyridylacetonitrile was dissolved in 400 mℓ of dry tetrahydrofuran, and the solution was cooled to -10°C in an inert gas atmosphere. Then, 284 mg of a

hexane solution of n-butyl lithium (1.66 M) was dropwise added thereto at a temperature of not higher than 7°C, and the mixture was reacted at room temperature for 1.5 hours. The reaction product was cooled to -5°C. Then, 29 mℓ of iodomethane was dropwise added thereto at a temperature of not higher than 5°C, and then the mixture was reacted at room temperature overnight.

After completion of the reaction, the reaction product was gradually added to ice water and extracted with ethyl acetate and washed with a sodium chloride aqueous solution. Then, it was dried over anhydrous sodium sulfate. Then, the solvent was distilled off under reduced pressure, and the residue was purified by column chromatography (developing solvent: methylene chloride/ethyl acetate = 8/2) to obtain 18.77 g of oily yellow 2-methyl-2-(2-pyridyl)propionitrile.

(2) 19.17 g of 2-methyl-2-(2-pyridyl)propionitrile obtained in the above Step (1) was dropwise added to 30 mℓ of concentrated sulfuric acid at a temperature of not higher than 8°C. The mixture was reacted at room temperature overnight and then gradually poured into ice water. The reaction product was alkalized with aqueous ammonia and then extracted with ethyl acetate. The extract was washed with a sodium chloride aqueous solution. Then, it was dried over anhydrous sodium sulfate. Then, the solvent was distilled off under reduced pressure, and the residue was recrystallized from

toluene to obtain 5.57 g of 2-methyl-2-(2-pyridyl)propionamide (melting point: 99 - 100°C) as white crystals.

(3) 37 mℓ of 3N sodium hydroxide was cooled to -10°C, and 2.93 g of bromine was dropwise added thereto at a temperature of not higher than 0°C. The mixture was stirred at a temperature of not higher than 0°C for 30 minutes. Then, 3 g of 2-methyl-2-(2-pyridyl)propionamide obtained in the above Step (2) was added thereto. The mixture was reacted at a temperature of not higher than 0°C for 8 hours and then stirred overnight. Then, refluxing was conducted for 3 hours, and then the reaction product was cooled. It was then extracted with methylene chloride, and the extract was washed with a sodium chloride aqueous solution. Then, it was dried over anhydrous sodium sulfate, and the solvent was distilled off under reduced pressure to obtain 1.15 g of oily brown 1-methyl-1-(2-pyridyl)ethylamine.

(4) 842 mg of 1-methyl-1-(2-pyridyl)ethylamine obtained the above Step (3) and 186 mg of paraformaldehyde were dissolved in 20 mℓ of dry toluene, and the solution was heated at a temperature of from 80 to 90°C for 30 minutes, and an azeotropic dehydration reaction was conducted under reflux for further 7 hours to obtain a solution containing 1-methyl-N-methylene-1-(2-pyridyl)ethylamine.

(5) The solution obtained in the above Step (4) was

cooled. Then, 1.35 g of 2,2,6-trimethyl-5-phenyl-4H-1,3-dioxin-4-one was added thereto, and the mixture was reacted under reflux for 18 hours.

After completion of the reaction, the reaction product was cooled, and the solvent was distilled off under reduced pressure. The residue was purified by column chromatography (developing solvent: methylene chloride/ethyl acetate = 1/1) to obtain 676 mg of the desired product as a yellow oily substance.

NMR: (60 MHz, δ: in $CDC\ell_3$) 1.77(6H,s), 1.87(3H,s), 5.33(2H,s), 6.83-7.73(9H,m). 8.43-8.60(1H,m)

PREPARATION EXAMPLE 6

Preparation of 3-[[1-(7-chlorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one (Compound No. 3 as identified hereinafter)

(1) 25.3 g of sodium was gradually added to 500 mℓ of dry ethanol. Then, the mixture was cooled, and hydrogen sulfide gas was introduced to obtain an aqueous sodium sulfate solution. Then, 192 g of 2,3-dichloronitrobenzene was gradually added thereto, and the mixture was reacted while raising the temperature to 80°C. The reaction product was cooled again and put into water. The obtained product was subjected to cerite filtration. Then, the aqueous layer was washed with toluene and cooled. It was acidified with hydrochloric acid at a temperature of not higher than 5°C, and obtained crystals were washed with water. These crystals

were dissolved in methylene chloride and washed with a sodium chloride aqueous solution. Then, it was dried over anhydrous sodium sulfate, and then the solvent was distilled off under reduced pressure to obtain 63 g of 2-chloro-6-nitrothiophenol as yellow crystals having a melting point of from 55 to 58°C.

(2) 30 g of 2-chloro-6-nitrothiophenol obtained in the above Step (1) was dissolved in 700 mℓ of acetic acid, and the solution was heated to 70°C in an inert gas atmosphere. Then, 72.3 g of zinc was gradually added and reacted thereto at a temperature of from 80 to 100°C for one hour. Then, the reaction product was cooled, and hydrogen sulfide gas was introduced for one hour and a half to convert excess zinc to zinc sulfide at a temperature of not higher than 35°C. To this solution, 31.3 g of malononitrile was added, and the mixture was reacted at room temperature overnight.

After completion of the reaction, the reaction product was subjected to cerite filtration, and then water was added thereto. The mixture was extracted with methyl chloride, and the extract was washed with water and then dried over anhydrous sodium sulfate. The solvent was distilled off, and the residue was purified by column chromatography (developing solvent: methylene chloride) to obtain 16.3 g of 7-chlorobenzothiazol-2-ylacetonitrile as yellow crystals having a melting point of from 112 to 113°C.

(3) 16.3 g of 7-chlorobenzothiazol-2-ylacetonitrile obtained in the above Step (2) was suspended in 200 mℓ of dry tetrahydrofuran. This suspension was dropwise added to 6.87 g of sodium hydride (60%) in an inert gas atmosphere. The mixture was reacted at 60°C for one hour and then cooled to -10°C. Then, 11.7 mℓ of iodomethane was added thereto at a temperature of not higher than 10°C, and the mixture was reacted at room temperature for 2 hours.

After completion of the reaction, the reaction product was put into ice water and extracted with ethyl acetate. The extract was washed with a sodium chloride aqueous solution and dried over anhydrous sodium sulfate. Then, the solvent was distilled off, and the residue was purified by column chromatography (developing solvent: methylene chloride) to obtain 17.67 g of 2-(7-chlorobenzothiazol-2-yl)-2-methylpropionitrile as brown crystals (melting point: 69 - 71°C).

(4) 17.67 g of 2-(7-chlorobenzothiazol-2-yl)-2-methylpropionitrile obtained in the above Step (3) was dissolved in 50 mℓ of formic acid, and the solution was heated to a temperature of from 60 to 70°C. Then, hydrogen chloride gas was introduced and reacted thereto for 4 hours.

After completion of the reaction, the reaction product was cooled and then put into water. The mixture was extracted with ethyl acetate, and the extract was

washed with water. The solvent was distilled off, and the obtained crystals were washed with water and then with toluene and dried under reduced pressure to obtain 15.79 g of 2-(7-chlorobenzothiazol-2-yl)-2-methyl-propionamide (melting point: 139 - 141°C).

(5) 2.86 g of sodium hydroxide was dissolved in 20 mℓ of water, and the solution was cooled. Then, 6.69 mℓ of bromine was added thereto at a temperature of not higher than 0°C, and the mixture was stirred for 30 minutes. To this solution, 3 g of 2-(7-chlorobenzothiazol-2-yl)-2-methylpropionamide obtained in the above Step (3) was added, and the mixture was reacted at 70°C for 2 hours.

After completion of the reaction, the reaction mixture was cooled to room temperature and extracted with methylene chloride. The extract was washed with water and then dried over anhydrous sodium sulfate. The solvent was distilled off, and the residue was purified by column chromatography (developing solvent: methylene chloride/ethyl acetate = 6/4) to obtain 2.3 g of semisolid [1-(7-chlorobenzothiazol-2-yl)-1-methyl]ethylamine.

(6) 930 mg of 1-(7-chlorobenzothiazol-2-yl)-1-methylethylamine obtained in the above Step (5) and 151 mg of paraformaldehyde were dissolved in 20 mℓ of dry toluene, and the solution was reacted at a temperature of from 90 to 100°C for 30 minutes. Further, an azeotropic dehydration reaction was carried out under reflux for 7

hours to obtain a solution containing [1-(7-chlorobenzothiazol-2-yl)-1-methyl-N-methylene]ethylamine.

(7) To the solution obtained in the above Step (6), 896 mg of 2,2,6-trimethyl-5-phenyl-4H-1,3-dioxin-4-one was added, and the mixture was reacted at 120°C for 18 hours.

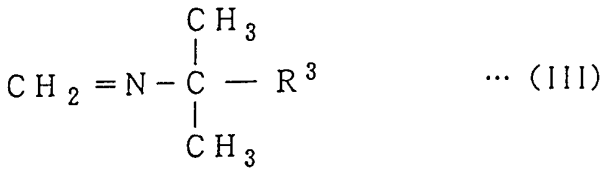
After completion of the reaction, the solvent was distilled off under reduced pressure. Then, the residue was purified twice by column chromatography (developing solvent: methylene chloride) to obtain 270 mg of the desired product as a brown oily substance.

NMR: (60 MHz, δ: in $CDC\ell_3$) 1.90(3H,s), 1.93(6H,s), 5.40(2H,s), 7.17-7.99(8H,m).

Further, the above oily substance was purified three times by the same column chromatography to obtain 110 mg of the desired product as white crystals having a melting point of from 115.5 to 118°C.

The intermediates of the above formulas (III) to (VII) are believed to be novel compounds, and representative examples of these intermediates will be given sequentially in Tables 1-1 to 1-5.

Table 1-1



… (III)

Intermediate No.	R^3
1	Thiophen-2-yl
2	5-Chlorothiophen-2-yl
3	Thiophen-3-yl
4	Benzothiophen-2-yl
5	Benzothiophen-3-yl
6	Benzofuran-2-yl
7	Benzothiazol-2-yl
8	4-Chloro-benzothiazol-2-yl
9	4-Methylbenzothiazol-2-yl
10	4-Methoxybenzothiazol-2-yl
11	6-Methoxybenzothiazol-2-yl
12	5-Chlorobenzothiazol-2-yl
13	6-Chlorobenzothiazol-2-yl
14	5-Fluorobenzothiazol-2-yl
15	5-(Trifluoromethyl)benzothiazol-2-yl
16	Benzoxazol-2-yl
17	5-Chlorobenzoxazol-2-yl
18	6-Methylbenzoxazol-2-yl
19	1-Methylbenzimidazol-2-yl
20	1-n-Butylbenzimidazol-2-yl

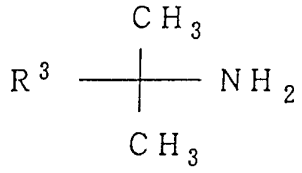
Table 1-1 (continued)

Intermediate No.	R^3
21	5-Chloro-1-methylbenzimidazol-2-yl
22	6-Fluorobenzothiazol-2-yl
23	5-Methylbenzothiazol-2-yl
24	Pyridin-2-yl
25	7-Chlorobenzothiazol-2-yl
26	5-Methylfuran-2-yl
27	1-Methyl-pyrrol-2-yl
28	4-Methylthiazol-2-yl
29	Oxazol-2-yl
30	1-Methylimidazol-2-yl
31	Isothiazol-3-yl
32	Isothiazol-4-yl
33	Isoxazol-5-yl
34	1-Ethylpyrazol-4-yl
35	5-Chlorobenzofuran-2-yl
36	6-Methylbenzothiophen-2-yl
37	1-Methylindol-2-yl
38	1-Ethylindol-3-yl
39	5-Ethoxythiophen-2-yl
40	5-(2,2,2-Trifluoroethoxy)thiophen-2-yl
41	5-Methylthiofuran-2-yl
42	6-Methylsulfonylpyridin-3-yl

Table 1-1 (continued)

Intermediate No.	R^3
43	5-Trifluoromethylpyridin-2-yl
44	3-Chloro-5-trifluoromethylpyridin-2-yl
45	5-Trifluoromethylbenzothiazol-2-yl
46	7-Phenoxybenzothiazol-2-yl
47	5-Phenylthiophen-2-yl
48	Benzisothiazol-3-yl
49	Benzisoxazol-3-yl
50	1-Methylindol-3-yl
51	6-Trifluoromethoxybenzothiazol-2-yl
52	7-Fluorobenzothiazol-2-yl
53	7-Bromobenzothiazol-2-yl
54	5-tert-Butylthiophen-2-yl
55	7-(5-Trifluoromethyl-2-pyridyloxy)-benzothiazol-2-yl
56	5-(5-Trifluoromethyl-2-pyridyloxy)-thiophen-2-yl
57	2-(4-Pyridyloxy)pyridin-5-yl
58	2-Chloropyridin-5-yl
59	7-Methylbenzothiazol-2-yl
60	6-Methylbenzothiazol-2-yl
61	5-Methoxybenzoxazol-2-yl
62	4-Fluorobenzothiazol-2-yl
63	4,7-Dichlorobenzothiazol-2-yl

Table 1-2



… (IV)

Intermediate No.	R^3	Physical properties
101	Thiophen-2-yl	Oily substance
102	5-Chlorothiophen-2-yl	Oily substance
103	Thiophen-3-yl	Oily substance
104	Benzothiophen-2-yl	Oily substance
105	Benzothiophen-3-yl	Oily substance
106	Benzofuran-2-yl	Oily substance
107	Benzothiazol-2-yl	Oily substance
108	4-Chlorobenzothiazol-2-yl	Oily substance
109	4-Methylbenzothiazol-2-yl	Oily substance
110	4-Methoxybenzothiazol-2-yl	Oily substance
111	6-Methoxybenzothiazol-2-yl	Oily substance
112	5-Chlorobenzothiazol-2-yl	m.p.76-77°C
113	6-Chlorobenzothiazol-2-yl	Oily substance
114	5-Fluorobenzothiazol-2-yl	Oily substance
115	5-(Trifluoromethyl)benzothiazol-2-yl	Oily substance
116	Benzoxazol-2-yl	Oily substance
117	5-Chlorobenzoxazol-2-yl	m.p.84-91°C

Table 1-2 (continued)

Intermediate No.	R^3	Physical properties
118	6-Methylbenzoxazol-2-yl	Oily substance
119	1-Methylbenzimidazol-2-yl	m.p.61-65°C
120	1-n-Butylbenzimidazol-2-yl	Oily substance
121	5-Chloro-1-methylbenzimidazol-2-yl	Oily substance
122	6-Fluorobenzothiazol-2-yl	Oily substance
123	5-Methylbenzothiazol-2-yl	Brown solid
124	Pyridin-2-yl	Oily substance
125	7-Chlorobenzothiazol-2-yl	Oily substance
126	5-Methylfuran-2-yl	
127	1-Methyl-pyrrol-2-yl	
128	4-Methylthiazol-2-yl	
129	Oxazol-2-yl	
130	1-Methylimidazol-2-yl	
131	Isothiazol-3-yl	
132	Isothiazol-4-yl	
133	Isoxazol-5-yl	
134	1-Ethylpyrazol-4-yl	
135	5-Chlorobenzofuran-2-yl	
136	6-Methylbenzothiophen-2-yl	
137	1-Methylindol-2-yl	
138	1-Ethylindol-3-yl	

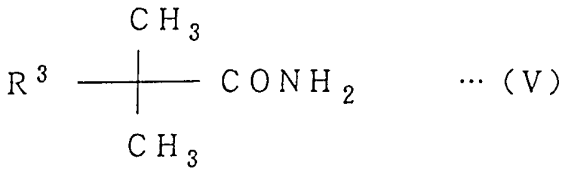
Table 1-2 (continued)

Intermediate No.	R^3	Physical properties
139	5-Ethoxythiophen-2-yl	
140	5-(2,2,2-trifluoroethoxy)-thiophen-2-yl	
141	5-Methylthiofuran-2-yl	
142	6-Methylsulfonylpyridin-3-yl	
143	5-Trifluoromethylpyridin-2-yl	
144	3-Chloro-5-trifluoromethyl-pyridin-2-yl	Oily substance
145	7-Trifluoromethylbenzo-thiazol-2-yl	
146	7-Phenoxybenzothiazol-2-yl	
147	5-Phenylthiophen-2-yl	
148	Benzisothiazol-3-yl	
149	Benzisoxazol-3-yl	
150	1-Methylindol-3-yl	
151	6-Trifluoromethoxybenzo-thiazol-2-yl	
152	7-Fluorobenzothiazol-2-yl	
153	7-Bromobenzothiazol-2-yl	Oily substance
154	5-tert-Butylthiophen-2-yl	Oily substance
155	7-(5-Trifluoromethyl-2-pyridyloxy)benzothiazol-2-yl	
156	5-(5-Trifluoromethyl-2-pyridyloxy)thiophen-2-yl	

Table 1-2 (continued)

Intermediate No.	R^3	Physical properties
157	2-(4-Pyridyloxy)pyridin-5-yl	
158	2-Chloropyridin-5-yl	Oily substance
159	7-Methylbenzothiazol-2-yl	Oily substance
160	6-Methylbenzothiazol-2-yl	m.p.60-61°C
161	5-Methoxybenzoxazol-2-yl	m.p.105-106°C
162	4-Fluorobenzothiazol-2-yl	Oily substance
163	4,7-Dichlorobenzothiazol-2-yl	Oily substance

Table 1-3



… (V)

Intermediate No.	R^3	Physical properties
201	Thiophen-2-yl	m.p.143-144°C
202	5-Chlorothiophen-2-yl	m.p.90-93°C
203	Thiophen-3-yl	m.p.147-152°C
204	Benzothiophen-2-yl	m.p.131-133°C
205	Benzothiophen-3-yl	Oily substance
206	Benzofuran-2-yl	m.p.150-156°C
207	Benzothiazol-2-yl	m.p.133-134°C
208	4-Chlorobenzothiazol-2-yl	m.p.92-94°C
209	4-Methylbenzothiazol-2-yl	Oily substance
210	4-Methoxybenzothiazol-2-yl	m.p.121-123°C
211	6-Methoxybenzothiazol-2-yl	m.p.155-157°C
212	5-Chlorobenzothiazol-2-yl	m.p. 141-142.5°C
213	6-Chlorobenzothiazol-2-yl	m.p.148-150°C
214	5-Fluorobenzothiazol-2-yl	m.p.125-126°C
215	5-(Trifluoromethyl)benzothiazol-2-yl	m.p.140-142°C
216	Benzoxazol-2-yl	m.p.161-163°C
217	5-Chlorobenzoxazol-2-yl	m.p.144-148°C

Table 1-3 (continued)

Intermediate No.	R^3	Physical properties
218	6-Methylbenzoxazol-2-yl	m.p.116-118°C
219	1-Methylbenzimidazol-2-yl	m.p.257-258°C
220	1-n-Butylbenzimidazol-2-yl	m.p.180-181°C
221	5-Chloro-1-methylbenzimidazol-2-yl	m.p.228-230°C
222	6-Fluorobenzothiazol-2-yl	Oily substance
223	5-Methylbenzothiazol-2-yl	m.p.150-152°C
224	Pyridin-2-yl	m.p.99-100°C
225	7-Chlorobenzothiazol-2-yl	m.p.139-141°C
226	5-Methylfuran-2-yl	
227	1-Methyl-pyrrol-2-yl	
228	4-Methylthiazol-2-yl	
229	Oxazol-2-yl	
230	1-Methylimidazol-2-yl	
231	Isothiazol-3-yl	
232	Isothiazol-4-yl	
233	Isoxazol-5-yl	
234	1-Ethylpyrazol-4-yl	
235	5-Chlorobenzofuran-2-yl	
236	6-Methylbenzothiophen-2-yl	
237	1-Methylindol-2-yl	
238	1-Ethylindol-3-yl	

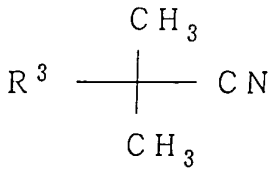
Table 1-3 (continued)

Intermediate No.	R^3	Physical properties
239	5-Ethoxythiophen-2-yl	
240	5-(2,2,2-trifluoroethoxy)-thiophen-2-yl	
241	5-Methylthiofuran-2-yl	
242	6-Methylsulfonylpyridin-3-yl	
243	5-Trifluoromethylpyridin-2-yl	
244	3-Chloro-5-trifluoromethyl-pyridin-2-yl	m.p.118-135°C
245	7-Trifluoromethylbenzo-thiazol-2-yl	
246	7-Phenoxybenzothiazol-2-yl	
247	5-Phenylthiophen-2-yl	
248	Benzisothiazol-3-yl	
249	Benzisoxazol-3-yl	
250	1-Methylindol-3-yl	
251	6-Trifluoromethoxybenzo-thiazol-2-yl	
252	7-Fluorobenzothiazol-2-yl	
253	7-Bromobenzothiazol-2-yl	m.p.133-136°C
254	5-tert-Butylthiophen-2-yl	m.p.95-98°C
255	7-(5-Trifluoromethyl-2-pyridyloxy)benzothiazol-2-yl	
256	5-(5-Trifluoromethyl-2-pyridyloxy)thiophen-2-yl	

Table 1-3 (continued)

Interme-diate No.	R^3	Physical properties
257	2-(4-Pyridyloxy)pyridin-5-yl	
258	2-Chloropyridin-5-yl	m.p.111-113°C
259	7-Methylbenzothiazol-2-yl	m.p.142-143°C
260	6-Methylbenzothiazol-2-yl	m.p.149-151°C
261	5-Methoxybenzoxazol-2-yl	m.p.161-162°C
262	4-Fluorobenzothiazol-2-yl	
263	4,7-Dichlorobenzothiazol-2-yl	

Table 1-4



… (VI)

Intermediate No.	R^3	Physical properties
301	Thiophen-2-yl	b.p.73-77°C/ 7 mmHg
302	5-Chlorothiophen-2-yl	Oily substance
303	Thiophen-3-yl	Oily substance
304	Benzothiophen-2-yl	Oily substance
305	Benzothiophen-3-yl	Oily substance
306	Benzofuran-2-yl	Oily substance
307	Benzothiazol-2-yl	m.p.44-46°C
308	4-Chlorobenzothiazol-2-yl	m.p.95-97°C
309	4-Methylbenzothiazol-2-yl	Oily substance
310	4-Methoxybenzothiazol-2-yl	m.p.89-91°C
311	6-Methoxybenzothiazol-2-yl	Oily substance
312	5-Chlorobenzothiazol-2-yl	m.p.99-101°C
313	6-Chlorobenzothiazol-2-yl	m.p.74-76°C
314	5-Fluorobenzothiazol-2-yl	m.p.86-87°C
315	5-(Trifluoromethyl)benzothiazol-2-yl	Oily substance
316	Benzoxazol-2-yl	Oily substance
317	5-Chlorobenzoxazol-2-yl	m.p.80-83°C

Table 1-4 (continued)

Intermediate No.	R^3	Physical properties
318	6-Methylbenzoxazol-2-yl	m.p.59-61°C
319	1-Methylbenzimidazol-2-yl	m.p.88-91°C
320	1-n-Butylbenzimidazol-2-yl	Oily substance
321	5-Chloro-1-methylbenzimidazol-2-yl	m.p.72-74°C
322	6-Fluorobenzothiazol-2-yl	Oily substance
323	5-Methylbenzothiazol-2-yl	m.p.68-74°C
324	Pyridin-2-yl	Oily substance
325	7-Chlorobenzothiazol-2-yl	m.p.69-71°C
326	5-Methylfuran-2-yl	
327	1-Methyl-pyrrol-2-yl	
328	4-Methylthiazol-2-yl	
329	Oxazol-2-yl	
330	1-Methylimidazol-2-yl	
331	Isothiazol-3-yl	
332	Isothiazol-4-yl	
333	Isoxazol-5-yl	
334	1-Ethylpyrazol-4-yl	
335	5-Chlorobenzofuran-2-yl	
336	6-Methylbenzothiophen-2-yl	
337	1-Methylindol-2-yl	
338	1-Ethylindol-3-yl	

Table 1-4 (continued)

Intermediate No.	R^3	Physical properties
339	5-Ethoxythiophen-2-yl	
340	5-(2,2,2-trifluoroethoxy)-thiophen-2-yl	
341	5-Methylthiofuran-2-yl	
342	6-Methylsulfonylpyridin-3-yl	
343	5-Trifluoromethylpyridin-2-yl	
344	3-Chloro-5-trifluoromethyl-pyridin-2-yl	Oily substance
345	7-Trifluoromethylbenzo-thiazol-2-yl	
346	7-Phenoxybenzothiazol-2-yl	
347	5-Phenylthiophen-2-yl	
348	benzisothiazol-3-yl	
349	Benzisoxazol-3-yl	
350	1-Methylindol-3-yl	
351	6-Trifluoromethoxybenzo-thiazol-2-yl	
352	7-Fluorobenzothiazol-2-yl	
353	7-Bromobenzothiazol-2-yl	m.p.74-75°C
354	5-tert-Butylthiophen-2-yl	Oily substance
355	7-(5-Trifluoromethyl-2-pyridyloxy)benzothiazol-2-yl	

Table 1-4 (continued)

Intermediate No.	R^3	Physical properties
356	5-(5-Trifluoromethyl-2-pyridyloxy)thiophen-2-yl	
357	2-(4-pyridyloxy)pyridin-5-yl	
358	2-Chloropyridin-5-yl	
359	7-Methylbenzothiazol-2-yl	Oily substance
360	6-Methylbenzothiazol-2-yl	m.p.69-70°C
361	5-Methoxybenzoxazol-2-yl	Oily substance
362	4-Fluorobenzothiazol-2-yl	
363	4,7-Dichlorobenzothiazol-2-yl	

Table 1-5

$R^3 - CH_2CN$ (VII)

Intermediate No.	R^3	Physical properties
401	5-Chlorothiophen-2-yl	Oily substance
402	Benzothiophen-2-yl	Brown crystals
403	Benzofuran-2-yl	m.p.50-52°C
404	4-Chlorobenzothiazol-2-yl	m.p.119-121°C
405	4-Methylbenzothiazol-2-yl	m.p.168-170°C
406	4-Methoxybenzothiazol-2-yl	m.p.138-140°C
407	6-Methoxybenzothiazol-2-yl	m.p.70-72°C
408	5-Chlorobenzothiazol-2-yl	m.p.150-152°c
409	6-Chlorobenzothiazol-2-yl	m.p.72-74°C
410	5-Fluorobenzothiazol-2-yl	m.p.124-128°C
411	5-(Trifluoromethyl)benzothiazol-2-yl	m.p.90-92°C
412	Benzoxazol-2-yl	m.p.65-67°C
413	5-Chlorobenzoxazol-2-yl	m.p.107-110°C
414	6-Methylbenzoxazol-2-yl	m.p. 115-116.5°C
415	1-Methylbenzimidazol-2-yl	
416	1-n-Butylbenzimidazol-2-yl	Oily substance
417	5-Chloro-1-Methylbenzimidazol-2-yl	
418	6-Fluorobenzothiazol-2-yl	Oily substance
419	5-Methylbenzothiazol-2-yl	Oily substance

Table 1-5 (continued)

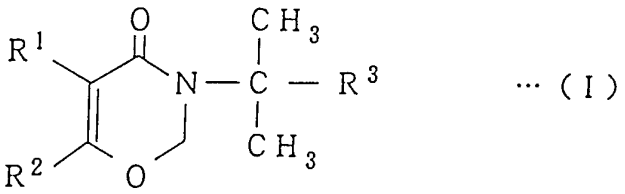
Intermediate No.	R^3	Physical properties
420	7-Chlorobenzothiazol-2-yl	m.p.112-113°C
421	5-Methylfuran-2-yl	
422	1-Methyl-pyrrol-2-yl	
423	4-Methylthiazol-2-yl	
424	Oxazol-2-yl	
425	1-Methylimidazol-2-yl	
426	Isothiazol-3-yl	
427	Isothiazol-4-yl	
428	Isoxazol-5-yl	
429	1-Ethylpyrazol-4-yl	
430	5-Chlorobenzofuran-2-yl	
431	6-Methylbenzothiophen-2-yl	
432	1-Methylindol-2-yl	
433	1-Ethylindol-3-yl	
434	5-Ethoxythiophen-2-yl	
435	5-(2,2,2-Trifluoroethoxy)-thiophen-2-yl	
436	5-Methylthiofuran-2-yl	
437	6-Methylsulfonylpyridin-3-yl	
438	5-Trifluoromethylpyridin-2-yl	
439	3-Chloro-5-trifluoromethyl-pyridin-2-yl	Oily substance
440	7-Trifluoromethylbenzo-thiazol-2-yl	

Table 1-5 (continued)

Intermediate No.	R^3	Physical properties
441	7-Phenoxybenzothiazol-2-yl	
442	5-Phenylthiophen-2-yl	
443	Benzisothiazol-3-yl	
444	Benzisoxazol-3-yl	
445	1-Methylindol-3-yl	
446	6-Trifluoromethoxybenzothiazol-2-yl	
447	7-Fluorobenzothiazol-2-yl	
448	7-Bromobenzothiazol-2-yl	m.p.116-118°C
449	5-tert-Butylthiophen-2-yl	Oily substance
450	7-(5-Trifluoromethyl-2-pyridyloxy)benzothiazol-2-yl	
451	5-(5-Trifluoromethyl-2-pyridyloxy)thiophen-2-yl	
452	2-(4-Pyridyloxy)pyridin-5-yl	
453	2-Chloropyridin-5-yl	
454	7-Methylbenzothiazol-2-yl	m.p.66-70°C
455	6-Methylbenzothiazol-2-yl	m.p.83-85°C
456	5-Methoxybenzoxazol-2-yl	m.p.82-84°C
457	4-Fluorobenzothiazol-2-yl	
458	4,7-Dichlorobenzothiazol-2-yl	

Now, typical examples of the compound of the formula (I) of the present invention will be given in Table 2.

Table 2



… (I)

Compound No.	R^1	R^2	R^3	Physical properties
1	Phenyl	Methyl	Benzothiazol-2-yl	m.p.166-168°C
2	Phenyl	Methyl	5-Fluorobenzothiazol-2-yl	m.p. 154.5-156°C
3	Phenyl	Methyl	7-Chlorobenzothiazol-2-yl	m.p. 115.5-118°C
4	Phenyl	Methyl	Benzoxazol-2-yl	Yellow oily substance
5	Phenyl	Methyl	Benzothiophen-2-yl	Transparent oily substance
6	Phenyl	Methyl	5-Chlorothiophen-2-yl	Transparent oily substance
7	Phenyl	Methyl	Thiophen-3-yl	m.p.85-88°C
8	Phenyl	Methyl	Pyridin-2-yl	Yellow oily substance
9	Phenyl	Methyl	6-Methoxybenzothiazol-2-yl	m.p.160-162°C
10	Phenyl	Methyl	5-Methylfuran-2-yl	
11	Phenyl	Methyl	1-Methylpyrrol-2-yl	
12	Phenyl	Methyl	4-Methylthiazol-2-yl	
13	Phenyl	Methyl	Oxazol-2-yl	

Table 2 (continued)

Compound No.	R^1	R^2	R^3	Physical properties
14	Phenyl	Methyl	1-Methylimidazol-2-yl	
15	Phenyl	Methyl	Isothiazol-3-yl	
16	Phenyl	Methyl	Isothiazol-4-yl	
17	Phenyl	Methyl	Isoxazol-5-yl	
18	Phenyl	Methyl	1-Ethylpyrazol-4-yl	
19	Phenyl	Methyl	5-Chlorobenzofuran-2-yl	
20	Phenyl	Methyl	6-Methylbenzothiophen-2-yl	
21	Phenyl	Methyl	1-Methylindol-2-yl	
22	Phenyl	Methyl	1-Ethylindol-3-yl	
23	Phenyl	Methyl	5-Ethoxythiophen-2-yl	
24	Phenyl	Methyl	5-(2,2,2-trifluoroethoxy)-thiophen-2-yl	
25	Phenyl	Methyl	5-Methylthiofuran-2-yl	
26	Phenyl	Methyl	6-Methylsulfonylpyridin-3-yl	
27	Phenyl	Methyl	5-Trifluoromethylpyridin-2-yl	Oily substance
28	Phenyl	Methyl	3-Chloro-5-trifluoro-methylpyridin-2-yl	m.p.90-95°C
29	Phenyl	Methyl	6-Chlorobenzothiazol-2-yl	Yellow oily substance
30	Phenyl	Methyl	6-Fluorobenzothiazol-2-yl	Yellow oily substance

Table 2 (continued)

Compound No.	R^1	R^2	R^3	Physical properties
31	Phenyl	Methyl	5-Methylbenzothiazol-2-yl	m.p.121-123°C
32	Phenyl	Methyl	6-Methoxybenzoxazol-2-yl	
33	Phenyl	Methyl	7-Trifluoromethylbenzothiazol-2-yl	
34	Phenyl	Methyl	7-Phenoxybenzothiazol-2-yl	
35	Phenyl	Methyl	5-Phenylthiophen-2-yl	
36	Phenyl	Methyl	Benzisothiazol-3-yl	
37	Phenyl	Methyl	Benzisoxazol-3-yl	
38	Phenyl	Methyl	1-Methylindol-3-yl	
39	2-Fluorophenyl	Methyl	7-Chlorobenzothiazol-2-yl	
40	Phenyl	Trifluoromethyl	Benzothiazol-2-yl	
41	2-Trifluoromethylphenyl	Methyl	7-chlorobenzothiazol-2-yl	
42	2-Methoxyphenyl	Ethyl	Thiophen-2-yl	
43	4-Methylthiophenyl	Methyl	Furan-2-yl	
44	3-Methylsulfonylphenyl	Methyl	Thiophen-3-yl	

Table 2 (continued)

Compound No.	R^1	R^2	R^3	Physical properties
45	Phenyl	Propyl	Benzothiazol-2-yl	Oily substance
46	Phenyl	t-Butyl	Benzothiazol-2-yl	
47	Phenyl	Methyl	6-Trifluoromethoxybenzothiazol-2-yl	
48	Phenyl	Methyl	4-Chlorobenzothiazol-2-yl	m.p.165-167°C
49	Phenyl	Methyl	5-Chlorobenzothiazol-2-yl	m.p.145-148°C
50	Phenyl	Methyl	7-Fluorobenzothiazol-2-yl	
51	Phenyl	Methyl	7-Bromobenzothiazol-2-yl	m.p.190-191°C
52	Phenyl	Methyl	5-t-Butylthiophen-2-yl	
53	2-fluorophenyl	Ethyl	7-Chlorobenzothiazol-2-yl	
54	2-fluorophenyl	Methyl	7-(5-Trifluoromethyl-2-pyridyloxy)benzothiazol-2-yl	
55	Phenyl	Methyl	5-(5-Trifluoromethyl-2-pyridyloxy)thiophen-2-yl	
56	Phenyl	Methyl	2-(4-pyridyloxy)pyridin-5-yl	
57	Phenyl	Methyl	2-Chloropyridin-5-yl	Oily substance
58	Phenyl	Ethyl	Benzothiazol-2-yl	Oily substance
59	Phenyl	Ethyl	7-Chlorobenzothiazol-2-yl	Oily substance
60	Phenyl	Propyl	7-Chlorobenzothiazol-2-yl	Oily substance

Table 2 (continued)

Compound No.	R^1	R^2	R^3	Physical properties
61	Phenyl	Methyl	7-Methylbenzothiazol-2-yl	Oily substance
62	Phenyl	Methyl	6-Methylbenzothiazol-2-yl	m.p.154-155°C
63	Phenyl	Methyl	5-Methoxybenzoxazol-2-yl	m.p.165-167°C
64	Phenyl	Methyl	4-Methylbenzothiazol-2-yl	m.p.154-159°C
65	Phenyl	Methyl	4-Methoxybenzothiazol-2-yl	m.p.55-60°C
66	Phenyl	Methyl	4-Fluorobenzothiazol-2-yl	m.p.137-142°C
67	Phenyl	Methyl	4,7-Dichlorobenzothiazol-2-yl	m.p.154-157°C
68	Phenyl	Methyl	6-Bromo-4-fluorobenzothiazol-2-yl	m.p.57-63°C

Now, Test Examples of the present invention will be given.

TEST EXAMPLE 1

Paddy field soil was put into a 1/10,000are pot, and seeds of barnyardgrass (Echinochloa crusgalli) and japanese bulrush (Scirpus juncoides) were sown and slightly covered with soil. Then, the pot was left to stand still in a greenhouse in a state where the irrigated water depth was from 0.5 to 1 cm, and two days later, tubers of japanese ribbon wapato (Sagittaria pygmaea) were planted. Into a separate 1/10,000are pot, soil was put in the same manner, then irrigated, paddled and levelled, and next day, rice seedlings grown to a 2 leaf state were transplanted at a rate of 2 seedlings per pot. Thereafter, the irrigated water depth was maintained at a level of from 3 to 4 cm, and when barnyardgrass and japanese bulrush reached a 0.5 leaf stage, japanese ribbon wapato reached to a primary leaf stage and rice reached four days old after transplantation, an aqueous diluted solution of a wettable powder having the compound of the present invention formulated in accordance with a usual formulation method, was uniformly dropwise applied by a pipette so that the dose of the active ingredient would be at a predetermined level.

On the 17 to 20th day after the application of the herbicide, the growth of the plants was visually observed

and the herbicidal effects and phytotoxicity were evaluated on the basis of the following standards, and the results are shown in Table 3.

Herbicidal effects (growth-controlling degree:%)		Phytotoxicity
5	(100)	Nil
t	(96-99)	Slight
$5-_4$	(91-95)	Small
5-4	(85-90)	Moderate
4-5	(81-84)	Substantial
4	(71-80)	
3	(51-70)	
2	(31-50)	
1	(0-30)	

Table 3

Compound No.	Dose of active ingredient (g/a)	Herbicidal effects			Phototoxicity to rice
		EC	SJ	SP	
1	10 5	5 5	5 5	4 2	Nil Nil
2	10 5	5 5	5 5-4	4 3	Slight Nil
3	10 5	5 5	2 1	1 1	Nil Nil
4	10 5	5 5	5 5	3 3	Slight Nil
5	10 5	5 5	1 1	1 1	Nil Nil
6	10 5	5 5	5 5-4	3 3	Nil Nil
7	10 5	5 5	5 4	2 1	Slight Nil
8	10 5	5 5-4	5-4 3	2 1	Nil Nil
31	10 5	5 5	3 3	2 1	Nil Nil
48	10 5	5 5	3 2	2 2	Nil Nil
51	10 5	5 5	1 1	1 1	Slight Nil
59	10 5	5 5	2 2	3 3	Nil Nil

Table 3 (continued)

Compound No.	Dose of active ingredient (g/a)	Herbicidal effects			Phototoxicity to rice
		EC	SJ	SP	
61	10	5	4	5-4	Slight
	5	5	4	4	Nil
66	10	5	5-4	5-4	Slight
	5	5	5-4	4	Nil
67	10	5	1	1	Nil
	5	5	1	1	Nil

Note:
EC: barnyardgrass (Echinochloa crusgalli)
SH: japanese bulrush (Scirpus juncoides)
SP: japanese ribbon wapato (Sagittaria pygmaea)

TEST EXAMPLE 2

Paddy field soil was put into a 1/10,000are pot, and seeds of barnyardgrass (Echinochloa crusgalli) were sown and slightly covered with the soil. Then, the pot was left to stand in a greenhouse in such a state that the irrigated water depth was from 0.5 to 1 cm. When its leaf stage reached a 2 leaf stage, the irrigated water depth was changed to from 3 to 4 cm, and an aqueous diluted solution of a wettable powder having the compound of the present invention formulated in accordance with a conventional formulation method, was uniformly dropwise applied by a pipette, so that the dose of the active ingredient would be at a predetermined level.

On the 18 to 22nd day after the application of the herbicide, the growth was visually observed, and the herbicidal effects were evaluated based on the same standards as in Test Example 1, and the results are shown in Table 4.

Table 4

Compound No.	Dose of active ingredient (g/a)	Herbicidal effects
		EC
1	10 5 2.5	5 5 5
2	10 5 2.5	5 5 5
3	10 5 2.5	5 5 5
4	10 5	5 5-4
5	10 5 2.5	5 5 5
48	10 5 2.5	5 5 5
51	10 5 2.5	5 5 5
59	10 5	5 5
61	10 5 2.5	5 5 5
66	10 5 2.5	5 5 5

Note:
EC: barnyardgrass (Echinochloa crusgalli)

Now, Formulation Examples of the present invention will be given.

FORMULATION EXAMPLE 1

(1) Compound No. 1	4.01 parts by weight
(2) bentonite	30.00 parts by weight
(3) calcium carbonate	61.49 parts by weight
(4) TOXANON GR-31A (manufactured by Sanyo Chemical Industries Ltd.)	3.00 parts by weight
(5) Calcium lignin sulfonate	1.50 parts by weight

Preliminarily pulverized components (1), (2) and (3) were mixed, and components (4) and (5) and water were mixed thereto, and the mixture was extruded and granulated, followed by drying and size-adjusting to obtain granules.

FORMULATION EXAMPLE 2

(1) Jeeklite	78 parts by weight
(2) Lavelin S (manufactured by Dai-ichi Kogyo Seiyaku Co., Ltd.)	2 parts by weight
(3) Sorpol 5039 (manufactured by Toho Chemical Industry Co., Ltd.)	5 parts by weight
(4) Amorphous silicon dioxide	15 parts by weight

A mixture of the above components and Compound No. 2 were mixed at a weight ratio of 9:1 to obtain a wettable powder.

FORMULATION EXAMPLE 3

(1) Compound No. 3	0.81 part by weight
(2) Bentonite	30.00 parts by weight
(3) Calcium carbonate	64.69 parts by weight

(4) TOXANON GR-31A (as mentioned above) 3.00 parts by weight

(5) Calcium lignin sulfonate 1.50 parts by weight

Preliminarily pulverized components (1), (2) and (3) were mixed, and components (4) and (5) and water were added and mixed, and the mixture was extruded and granulated, followed by drying and size-adjusting to obtain granules.

FORMULATION EXAMPLE 4

(1) Compound No. 3 30.0 parts by weight

(2) Jeeklite 60.0 parts by weight

(3) NK WG-1 (manufactured by Takemoto Oil and Fat Co., Ltd.) 5.0 parts by weight

(4) NK FS-7 (manufactured by Takemoto Oil and Fat Co., Ltd.) 5.0 parts by weight

Components (1), (2) and (3) were mixed and subjected to a pulverizer, and then component (4) was added, and the mixture was kneaded and then extruded and granulated, followed by drying and size-adjusting to obtain a granular wettable powder.

FORMULATION EXAMPLE 5

(1) Compound No. 3 1.30 parts by weight

(2) Pyrazoxyphene 16.25 parts by weight

(3) Bromobutyde 10.73 parts by weight

(4) Soprophor FL (manufactured by RHONE POULENC) 2.00 parts by weight

(5) Sorpol 355 (manufactured by Toho Chemical Co., Ltd.) 1.50 parts by weight

(6) IP solvent 1620 (manufactured by Idemitsu Petrochemical Co.,Ltd.) 32.00 parts by weight

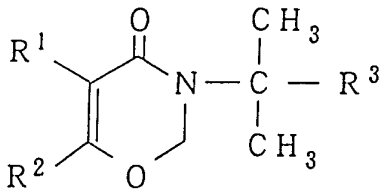
(7) Ethylene glycol 6.00 parts by weight

(8) Water 30.22 parts by weight

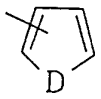
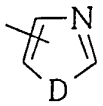
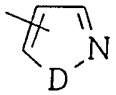
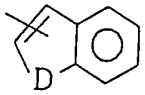
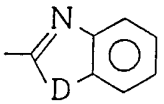
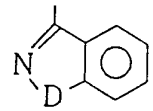
The above components were mixed and pulverized by a wet pulverizer (Dynomil) to obtain an aqueous suspension.

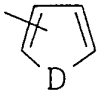
CLAIMS:

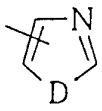
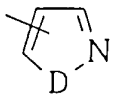
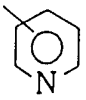
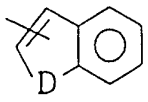
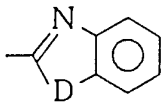
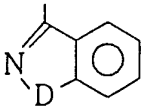
1. A cyclic amide compound of the formula (I):

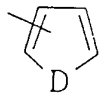
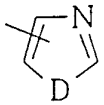
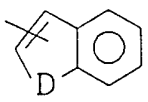
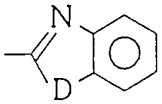


(I)

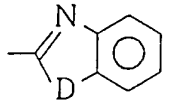
wherein R^1 is a phenyl group which may be substituted, R^2 is a hydrogen atom or an alkyl group which may be substituted by a halogen atom, and R^3 is  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted, or  which may be substituted, wherein D is an oxygen atom, a sulfur atom or $-N(R^4)-$, wherein R^4 is an alkyl group.

2. The cyclic amide compound according to Claim 1, wherein the substituent for the phenyl group which may be substituted, for R^1 in the formula (I), is a halogen atom; an alkyl group which may be substituted by a halogen atom; an alkoxy group which may be substituted by a halogen atom; an alkylthio group which may be substituted by a halogen atom; or an alkylsulfonyl group which may be substituted by a halogen atom, and the substituent for  which may be substituted,

 which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted, or  which may be substituted, wherein D is as defined above, for R^3 in the formula (1), is a halogen atom; an alkyl group which may be substituted by a halogen atom; an alkoxy group which may be substituted by a halogen atom; an alkylthio group which may be substituted by a halogen atom; an alkylsulfonyl group which may be substituted by a halogen atom; an aryl group which may be substituted by a halogen atom or a halogenoalkyl group; a heteroaryl group which may be substituted by a halogen atom or a halogenoalkyl group; an aryloxy group which may be substituted by a halogen atom or a halogenoalkyl group; or a heteroaryloxy group which may be substituted by a halogen atom or a halogenoalkyl group.

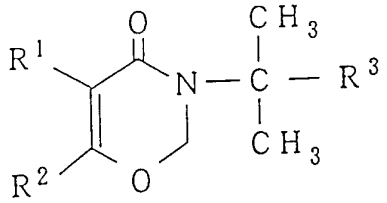
3. The cyclic amide compound according to Claim 1, wherein in the formula (I), R^2 is an alkyl group which may be substituted by a halogen atom, and R^3 is  which may be substituted,  which may be substituted,  which may be substituted, or  which may be substituted, wherein D is as

defined above.

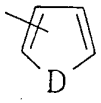
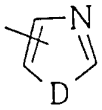
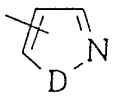
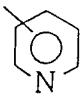
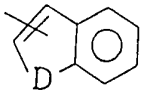
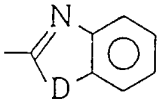
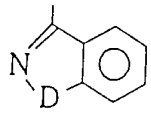
4. The cyclic amide compound according to Claim 1, wherein in the formula (I), R^1 is a phenyl group, R^2 is an alkyl group, and R^3 is  which may be substituted, wherein D is as defined above.

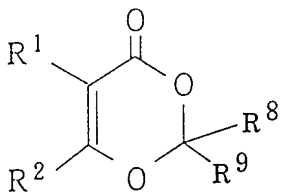
5. The cyclic amide compound according to Claim 1, which is 3-[[1-(benzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(5-fluorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(7-chlorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihyro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(4-chlorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(7-bromobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(7-chlorobenzothiazol-2-yl)-1-methyl]ethyl]-6-ethyl-2,3-dihydro-5-phenyl-4H-1,3-oxazin-4-one, 2,3-dihyro-6-methyl-3-[[1-(7-methylbenzothiazol-2-yl)-1-methyl]ethyl]-5-phenyl-4H-1,3-oxazin-4-one, 3-[[1-(4-fluorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one, or 3-[[1-(4,7-dichlorobenzothiazol-2-yl)-1-methyl]ethyl]-2,3-dihydro-6-methyl-5-phenyl-4H-1,3-oxazin-4-one.

6. A process for producing a cyclic amide compound of the formula (I):



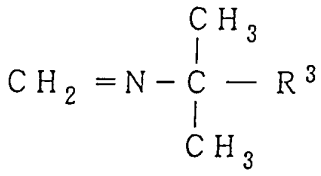
(I)

wherein R^1 is a phenyl group which may be substituted, R^2 is a hydrogen atom or an alkyl group which may be substituted by a halogen atom, and R^3 is  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted, or  which may be substituted, wherein D is an oxygen atom, a sulfur atom or $-N(R^4)-$, wherein R^4 is an alkyl group, which comprises reacting a dioxine compound of the formula (II):



(II)

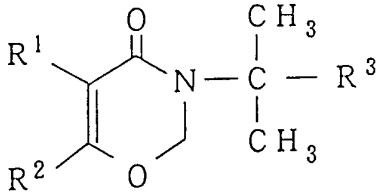
wherein R^1 and R^2 are as defined above, and each of R^8 and R^9 is an alkyl group, with an N-methyleneamine compound of the formula (III):



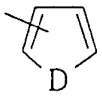
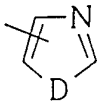
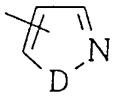
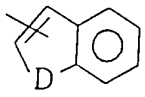
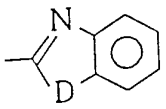
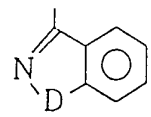
(III)

wherein R^3 is as defined above.

7. A herbicidal composition comprising a herbicidally effective amount of at least one member selected from the group consisting of cyclic amide compounds of the formula (I):

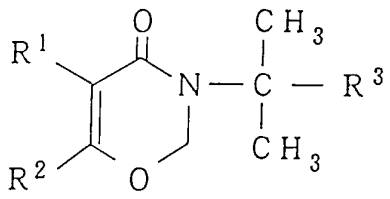


(I)

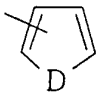
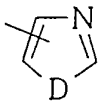
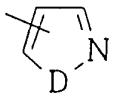
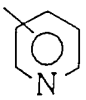
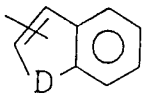
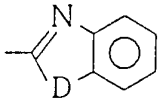
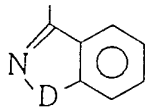
wherein R^1 is a phenyl group which may be substituted, R^2 is a hydrogen atom or an alkyl group which may be substituted by a halogen atom, and R^3 is  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted, or  which may be substituted, wherein D is an oxygen atom, a sulfur atom or $-N(R^4)-$, wherein R^4 is an alkyl group, and an agricultural adjuvant.

8. A herbicidal composition comprising a herbicidally effective amount of at least one member selected from the

group consisting of cyclic amide compounds of the formula (I):



(I)

wherein R^1 is a phenyl group which may be substituted, R^2 is a hydrogen atom or an alkyl group which may be substituted by a halogen atom, and R^3 is  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted, or  which may be substituted, wherein D is an oxygen atom, a sulfur atom or $-N(R^4)-$, wherein R^4 is an alkyl group, a herbicidally effective amount of at least one other herbicidally active ingredient selected from the group consisting of 2,4,6-trichlorophenyl-4-nitrophenyl ether, 5-(2,4-dichlorophenoxy)-2-nitroanisole, methyl 5-(2,4-dichlorophenoxy)-2-nitrobenzoate, 5-t-butyl-3-(2,4-dichloro-5-isopropoxyphenyl)-1,3,4-oxadiazol-2(3H)-one, 3-[5-cyclopentyloxy-4-chloro-2-fluoro)phenyl]-5-isopropylidene-1,3-oxazolidine-2,4-dione, S,S-dimethyl 2-difluoromethyl-4-(2-methylpropyl)-6-(trifluoromethyl)-

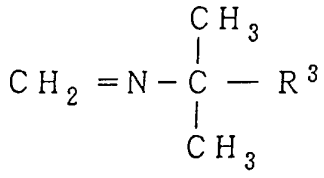
3,5-pyridinedicarbothioate, exo-1-methyl-4-(1-methylethyl)-2-[(2-methylphenyl)methoxy]-7-oxabicyclo[2,2,1]heptane, 3,7-dichloroquinoline-8-carboxylic acid, 1-(diethylcarbamoyl)-3-(2,4,6-trimethylphenylsulfonyl)-1,2,4-triazole, 3-isopropyl-2,1,3-benzothiadiazin-4-one-2,2-dioxide and its sodium salt, 2,3-dihydro-3,3,-dimethylbenzofuran-5-ylethanesulfonate, methyl 2-[(4,6-dimethoxypyrimidin-2-yl)oxy]-6-[1-(N-methoxyimino)ethyl]benzoate, 2-chloro-2',6'-diethyl-N-(2-propoxyethyl)acetanilide, 2-chloro-N-[(3-methoxy-2-thienyl)methyl]-2',6'-dimethylacetanilide, 2',3'-dichloro-4-ethoxymethoxybenzanilide, 2-[(benzothiazol-2-yl)oxy]-N-methylacetanilide, 2-(2-naphthoxy)propionanilide, 2-(2,4-dichloro-3-methylphenoxy)propionanilide, S-(4-chlorobenzyl)-N,N-diethylthiocarbamate, S-ethyl hexahydro-1H-azepin-1-carbothioate, S-(1-methyl-1-phenylethyl)piperidine-1-carbothioate, S-benzyl N-(1,2-dimethylpropyl)-N-ethylthiocarbamate, O-3-t-butylphenyl N-(6-methoxy-2-pyridyl)-N-methylthiocarbamate, (R) n-butyl 2-[4-(4-cyano-2-fluorophenoxy)phenoxy]propionate, methyl 2-[[[[[(4,6-dimethoxypyrimidin-2-yl)amino]carbonyl]amino]sulfonyl]methyl]benzoate, N-[[(4,6-dimethoxypyrimidin-2-yl)amino]carbonyl]-4-ethoxycarbonyl-1-methyl-5-pyrazolesulfonamide, 1-(2-chloroimidazo[1,2-a]pyridin-3-ylsulfonyl)-3-(4,6-dimethoxy-2-pyrimidinyl)urea, 3-(4,6-dimethoxy-1,3,5-

triazin-2-yl)-1-[2-(2-methoxyethoxy)phenylsulfonyl]urea, 5-(2,2-difluoro-2-chloroethoxy)-N-[[(4,6-dimethoxypyrimidin-2-yl)amino]carbonyl]-3-methyl-4-isothiazolesulfonamide, N-[[(4,6-dimethoxy-2-pyrimidinyl)amino]carbonyl]-1-methyl-4-(2-methyl-2H-tetrazol-5-yl)-1H-pyrazole-5-sulfonamide, 1-[[2-(cyclopropylcarbonyl)phenyl]-sulfamoyl]-3-(4,6-dimethoxy-2-pyrimidinyl)urea, 2-[4-(2,4-dichlorobenzoyl)-1,3-dimethylpyrazol-5-yloxy]acetophenone, 4-(2,4-dichlorobenzoyl)-1,3-dimethyl-5-pyrazolyl-p-toluenesulfonate, 2-[4-(2,4-dichloro-m-toluoyl)-1,3-dimethylpyrazol-5-yloxy]-4'-methylacetophenone, 2-bromo-N-(α,α-dimethylbenzyl)-3,3-dimethylbutylamide, 1-(α,α-dimethylbenzyl)-3-(p-tolyl)urea, 1-(2-chlorobenzyl)-3-(α,α-dimethylbenzyl)urea, 2-methylthio-4,6-bis-(methylamino)-s-triazine, 2-methylthio-4-ethylamino-6-(1',2'-diethylpropylamino)-s-triazine, 2,4-dichlorophenoxy acetic acid and its salt and ester, 4-(4-chloro-o-tolyloxy)butyric acid and its salt and ester, S-ethyl 4-chloro-2-methylphenoxythioacetate, and 2-[2-(3-chlorophenyl)-2,3-epoxypropyl]-2-ethylindan-1,3-dione, and an agricultural adjuvant.

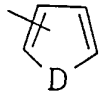
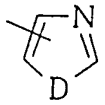
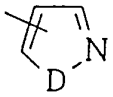
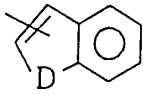
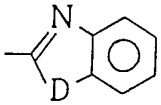
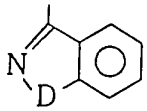
9. A herbicidal method which comprises applying the herbicidal composition of Claim 7 to plants.

10. A herbicidal method which comprises applying the herbicidal composition of Claim 8 to plants.

11. An N-methyleneamine compound of the formula (III):



(III)

wherein R^3 is  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted,  which may be substituted, or  which may be substituted,

wherein D is an oxygen atom, a sulfur atom or $-N(R^4)-$,

wherein R^4 is an alkyl group.

INTERNATIONAL SEARCH REPORT

Int onal Application No
PCT/JP 93/01815

A. CLASSIFICATION OF SUBJECT MATTER
IPC 5 C07D413/06 C07D417/06 C07D277/64 C07D333/28 C07D213/38
A01N43/86

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 5 C07D A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP,A,4 089 485 (DAICEL CHEMICAL IND. LTD.) 23 March 1992 cited in the application see tables I-3 (compounds 27-34), I-4 (compounds 35-37, 39-41), I-8 (compounds 76-83), I-9 (compound 84)	1-10
A	see compounds of formula II	11

Y	EP,A,0 372 586 (KUMIAI CHEMICAL INDUSTRY CO. & IHARA CHEMICAL INDUSTRY CO.) 13 June 1990 see the whole document	1-10

Y	US,A,4 138 243 (BÖHNER B. ET AL.) 6 February 1979 see the whole document	1-10

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

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"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
21 March 1994	14. 04. 94

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax (+31-70) 340-3016	Hartrampf, G

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INTERNATIONAL SEARCH REPORT

Intr onal Application No
PCT/JP 93/01815

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US,A,4 220 655 (BÖHNER B. ET AL.) 2 September 1980 see the whole document	1-10
P,Y	JP,A,5 201 811 (DAICEL CHEMICAL IND. LTD. & MITSUBISHI PETROCHEMICAL CO. LTD.) 10 August 1993 see compounds of formula I	1-10
P,Y	WO,A,93 15064 (MITSUBISHI PETROCHEMICAL CO. LTD. & DAICEL CHEMICAL INDUSTRIES LTD.) 5 August 1993 see compounds of formula I, tables 1, 3, 4	1-10
A	see compounds of formula III, table 7	11
A	JP,A,59 172 485 (KATO, TETSUZO) 29 September 1984 cited in the application see compounds of formula II, table on page 819	1-11

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

Information on patent family members

Int onal Application No
PCT/JP 93/01815

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
JP-A-4089485	23-03-92	NONE		
EP-A-0372586	13-06-90	US-A-	5006157	09-04-91
		US-A-	5076834	31-12-91
		JP-A-	3204855	06-09-91
US-A-4138243	06-02-79	CH-A-	619931	31-10-80
		BE-A-	857684	13-02-78
		CA-A-	1084494	26-08-80
		DE-A-	2735841	16-02-78
		GB-A-	1582964	21-01-81
		JP-A-	53023965	06-03-78
		NL-A-	7708937	14-02-78
		US-A-	4220655	02-09-80
US-A-4220655	02-09-80	CH-A-	619931	31-10-80
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		CA-A-	1084494	26-08-80
		DE-A-	2735841	16-02-78
		GB-A-	1582964	21-01-81
		JP-A-	53023965	06-03-78
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JP-A-5201811	10-08-93	NONE		
WO-A-9315064	05-08-93	AU-B-	3367893	01-09-93
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