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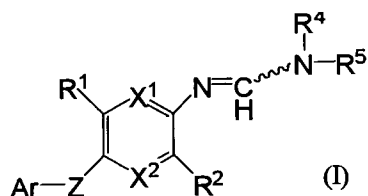
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(54) Title: PYRIDYLAMIDINE COMPOUND OR SALT THEREOF, AND AGRICULTURAL OR HORTICULTURAL FUNGICIDE COMPOSITION CONTAINING THE SAME AS ACTIVE INGREDIENT



(57) Abstract: An agricultural or horticultural fungicide composition having stabilized high plant disease control effect is provided. The agricultural or horticultural fungicide composition contains a pyridylamidine compound represented by the formula (I): or a salt thereof as an active ingredient. Symbols in the formula (I) are defined in the description.

DESCRIPTION

PYRIDYLAMIDINE COMPOUND OR SALT THEREOF, AND AGRICULTURAL OR HORTICULTURAL FUNGICIDE COMPOSITION CONTAINING THE SAME AS ACTIVE INGREDIENT

Technical Field

The present invention relates to a novel pyridylamide compound having fungicidal activity or a salt thereof, and an agricultural or horticultural fungicide composition containing the same as an active ingredient.

Background Art

Patent Document 1 discloses 6-chloro-3-cyano-2-dimethylamino-methyleneamino-5-(pyrid-4-yl)pyridine. Patent Document 2 discloses on page 51 5-dimethylamino-methylene-2-(5-chloro-pyrid-3-yloxy)pyridine as Comp. BH. Patent Document 3 discloses on pages 42-44 N,N-dimethyl-N'-[3-trifluoromethyl-5-(4-trifluoromethyl-phenyl)-pyridin-2-yl]formamidine, N'-[5-(4-chloro-phenyl)-3-methylpyridin-2-yl]-N,N'-dimethyl-formamidine, and N,N-dimethyl-N'-[3-methyl-5-(4-trifluoromethyl-phenyl)-pyridin-2-yl]formamidine. Furthermore, N'-[6-(4-fluorophenoxy)-3-pyridinyl]-N,N-dimethylformamidine is recorded in Chemical Library (Maybridge). However, these compounds are different from the compounds of the present invention.

Patent Document 4 discloses that certain pyridylamide compounds can be used as agricultural or horticultural fungicides. However, Patent Document 4 does not specifically disclose the pyridylamide compounds or salts thereof according to the present invention.

Patent Document 1	DD279016
Patent Document 2	WO2000/46203
Patent Document 3	WO2007/039439
Patent Document 4	WO2008/101682

Disclosure of Invention

Problems to be solved by the invention

Many agricultural or horticultural fungicides conventionally provided each has the characteristic in its plant pathogen control effect. Some agricultural or horticultural fungicide compositions are slightly poor in therapeutic effect as compared

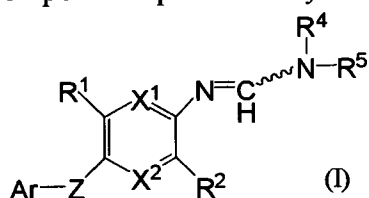
with preventive effect or are relatively short in residual efficacy, and depending on the application embodiments, may show only practically insufficient control effect to plant pathogens. Therefore, creation and production of a novel compound having strong plant pathogen control effect are desired.

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Means for solving the problems

As a result of investigations to solve the above-described problems, the present inventors have found that use of a compound represented by the formula (I) as an active ingredient exhibits excellent control effect to various diseases, particularly powdery mildew, scab and graymold of cereals, disease caused by sclerotinia of pulse, and downy mildew, late blight, and anthracnose of vegetables, and have completed the present invention. Namely, the present invention relates to a pyridylamidine compound represented by the formula (I):

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wherein one of X^1 and X^2 represents N, and the other represents CR^3 ; Ar represents phenyl which may be substituted with R^6 , 3,4-methylenedioxyphenyl, naphthyl, 5,6,7,8-tetrahydro-2-naphthyl, or pyridyl which may be substituted with R^6 ; Z represents a bond, O or OCH_2 ; R^1 , R^2 , R^3 and R^6 each independently represents a hydrogen atom, C_{1-6} alkyl, halo(C_{1-6} alkyl), halogen, C_{1-6} alkoxy, halo(C_{1-6} alkoxy), cyano, nitro, (C_{1-6} alkyl)carbonyl, (C_{1-6} alkoxy)carbonyl, or amino which may be substituted with C_{1-6} alkyl; R^4 and R^5 each independently represents C_{1-6} alkyl, provided that (1) a compound wherein X^1 is CH, X^2 is N, and both R^4 and R^5 are methyl, and (2) 6-chloro-3-cyano-2-dimethylamino-methyleneamino-5-(pyrid-4-yl)pyridine are excluded; or a salt thereof. Furthermore, the present invention relates to an agricultural or horticultural fungicide composition containing the pyridylamidine compound represented by the above formula (I) or a salt thereof, as an active ingredient.

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C_{1-6} alkyl or C_{1-6} alkyl moiety in the formula (I) includes linear or branched alkyl or alkyl moiety, such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl or t-butyl.

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Fluorine, chlorine, bromine or iodine is used as the halogen in the formula (I), and fluorine, chlorine or bromine is preferably used.

C_{1-6} alkoxy or C_{1-6} alkoxy moiety in the formula (I) includes linear or branched alkoxy or alkoxy moiety, such as methoxy, ethoxy, propoxy, isopropoxy, butoxy, isobutoxy or t-butoxy.

The salt of the pyridylamidine compound of the formula (I) generally includes

conventional salts. Examples of the salt include salts with a mineral acid such as hydrochloric acid, hydrobromic acid or sulfuric acid; salts with an organocarboxylic acid such as tartaric acid, formic acid, acetic acid, citric acid, fumaric acid, maleic acid, trichloroacetic acid or trifluoroacetic acid; and salts with a sulfonic acid such as methanesulfonic acid, benzenesulfonic acid, p-toluenesulfonic acid, mesitylenesulfonic acid or naphthalenesulfonic acid.

Geometric isomers (E-form and Z-form) are present in the pyridylamidine compound of the formula (I). Those isomers and mixtures are all encompassed in the present invention.

Effect of the invention

The pyridylamidine compound of the formula (I) or a salt thereof shows excellent effect as an active ingredient of an agricultural or horticultural fungicide composition.

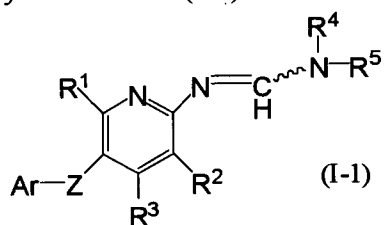
Best Mode for Carrying Out the Invention

Among the pyridylamidine compounds of the formula (I) or salts thereof according to the present invention, the preferable embodiments are described below.

[1] A pyridylamidine compound represented by the formula (I) or a salt thereof.

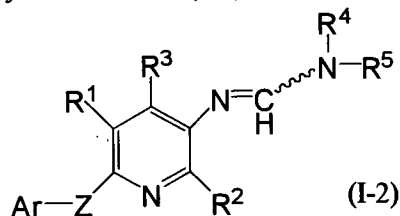
[2] The pyridylamidine compound according to the above [1], wherein in the formula (I), (1) X^1 is N, X^2 is CR^3 , and Z is a single bond, O or OCH_2 , or (2) X^1 is CH, X^2 is N, and Z is a bond, or a salt thereof.

[3] The pyridylamidine compound according to the above [1], which is represented by the formula (I-1):



wherein Ar represents phenyl which may be substituted with R^6 , 3,4-methylenedioxyphenyl, naphthyl, 5,6,7,8-tetrahydro-2-naphthyl, or pyridyl which may be substituted with R^6 ; Z represents a bond, O or OCH_2 ; R^1 , R^2 , R^3 and R^6 each independently represents a hydrogen atom, C_{1-6} alkyl, halo(C_{1-6} alkyl), halogen, C_{1-6} alkoxy, halo(C_{1-6} alkoxy), cyano, nitro, (C_{1-6} alkyl)carbonyl, (C_{1-6} alkoxy)carbonyl, or amino which may be substituted with C_{1-6} alkyl; R^4 and R^5 each independently represents C_{1-6} alkyl, provided that 6-chloro-3-cyano-2-dimethylamino-methyleneamino-5-(pyrid-4-yl)pyridine is excluded; or a salt thereof.

[4] The pyridylamidine compound according to the above [1], which is represented by the formula (I-2):



wherein Ar represents phenyl which may be substituted with R⁶, 3,4-

methylenedioxyphenyl, naphthyl, 5,6,7,8-tetrahydro-2-naphthyl, or pyridyl which may be substituted with R⁶; Z represents a bond, O or OCH₂; R¹, R², R³ and R⁶ each

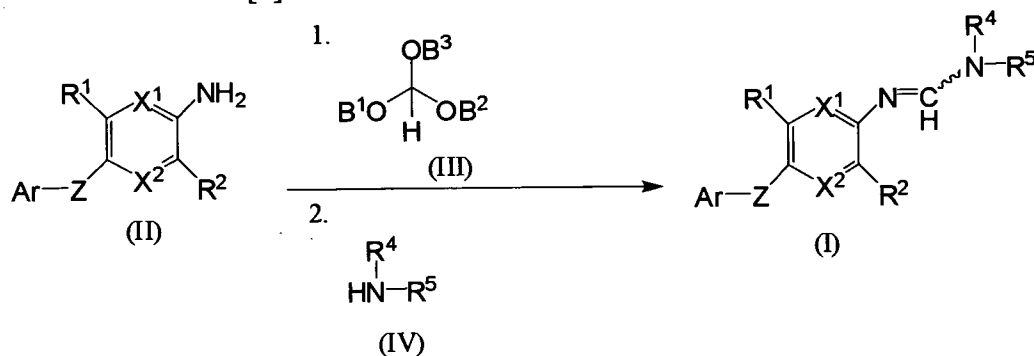
independently represents a hydrogen atom, C₁₋₆ alkyl, halo(C₁₋₆ alkyl), halogen, C₁₋₆ alkoxy, halo(C₁₋₆ alkoxy), cyano, nitro, (C₁₋₆ alkyl)carbonyl, (C₁₋₆ alkoxy)carbonyl, or amino which may be substituted with C₁₋₆ alkyl; R⁴ and R⁵ each independently

represents C₁₋₆ alkyl, provided that the compound wherein both R⁴ and R⁵ are methyl is excluded; or a salt thereof.

[5] The pyridylamidine compound according to the above [4], wherein Z is a bond or a salt thereof.

The compound of the formula (1) can be produced by the methods of production methods [1] to [3] described below and methods based on Synthesis Examples 1 to 4 described hereinafter. The compound of the formula (I) obtained in each production method can be isolated and purified by applying ordinary chemical operation such as extraction, concentration, distillation, crystallization, filtration, recrystallization or various chromatography.

Production Method [1]



In production method [1], Ar, Z, R¹, R², R⁴, R⁵, X¹ and X² have the same meanings as described above; and B¹, B² and B³, each independently represents alkyl or cycloalkyl.

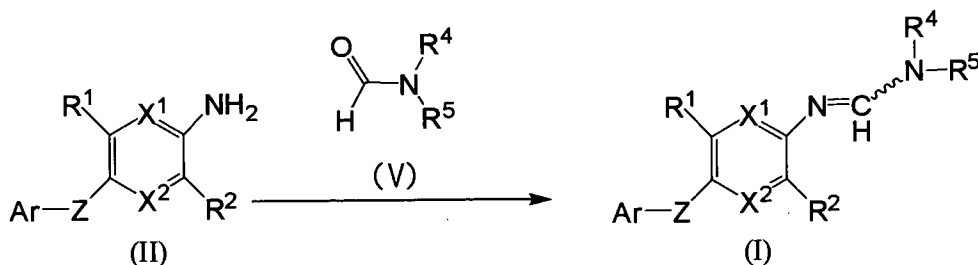
In the production method [1], the compound of the formula (I) can be produced

by reacting a compound of the formula (II) with ortho triester of the formula (III), and reacting the resulting reaction mixture with a compound of the formula (IV). In this reaction, each the ortho triester of the formula (III) and the compound of the formula (IV) is usually employed in an amount of from 0.8 to 80 equivalents, preferably from 5 to 50 equivalents, per mole of the compound of the formula (II).

The reaction between the compound of the formula (II) and the ortho triester of the formula (III) in the first step of the production method [1] is conducted in the presence of an acid or a base. Examples of the acid that can be used include p-toluenesulfonic acid, benzenesulfonic acid, methanesulfonic acid and hydrochloric acid. Examples of the base that can be used include inorganic bases, for example, carbonates of alkali metals, such as sodium carbonate or potassium carbonate; hydrogen carbonates of alkali metals, such as sodium hydrogen carbonate; carbonates of alkaline earth metals, such as calcium carbonate; hydroxides of alkali metals, such as sodium hydroxide or potassium hydroxide; or hydroxides of alkaline earth metals, such as calcium hydroxide; and organic bases, such as trimethylamine, pyridine, diazabicyclooctane or diazabicycloundecene. The reaction may be conducted in the presence of an inert solvent, if necessary. Examples of the inert solvent include hydrocarbon solvents such as hexane, heptane, cyclohexane, methylcyclohexane, benzene, toluene, xylene or decalin; halogenated hydrocarbon solvents such as chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride or trichloroethane; ether solvents such as diethyl ether, diisopropyl ether, methyl-t-butyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole; nitrile solvents such as acetonitrile, propionitrile or n- or iso-butyronitrile; amide solvents such as N,N-dimethylformamide, N,N-dimethylacetamide or N-methylpyrrolidine; mixed solvents of those; and mixed solvents of those and water. The reaction is usually conducted at usually from 0 to 150°C, preferably from 100 to 120°C.

The reaction mixture obtained by the reaction in the first step of the production method [1] is allowed to react with a compound of the formula (IV). The reaction in the second step of the production method [1] is conducted in the presence of an inert solvent. As the inert solvent can use the same inert solvents as used in the reaction in the first step of the production method [1] in this production method can be used. The reaction is usually conducted at from 0 to 150°C, preferably from 10 to 50°C.

Production Method [2]

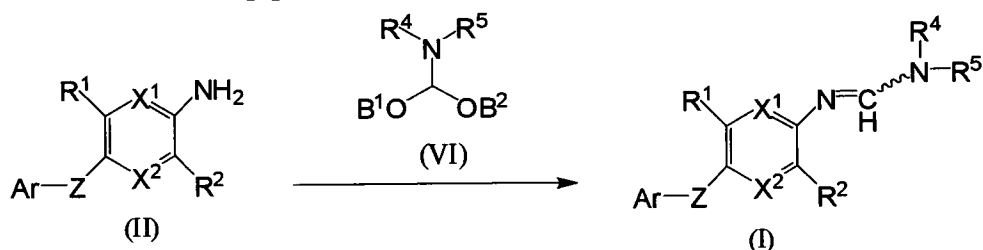


In production method [2], Ar, Z, R¹, R², R⁴, R⁵, X¹ and X² have the same meanings as described above.

In the production method [2], the compound of the formula (I) can be produced
 5 by reacting the compound of the formula (II) with an amide compound of the formula (V). This reaction is conducted in the presence of a halogenating agent. The halogenating agent includes PCl₅, PCl₃, POCl₃ and SOCl₂. Furthermore, the reaction is conducted in the presence of an inert solvent. As the inert solvent, solvents that do not adversely affect the reaction, can be used. For example, the inert solvents
 10 described in the production method [1] can be used. In the reaction, the amide compound of the formula (V) is usually used in an amount of from 0.8 to 50 equivalents, preferably from 1 to 10 equivalents, per mole of the compound of the formula (II). This reaction is usually conducted at from 0 to 150°C, preferably from 20 to 120°C.

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Production Method [3]



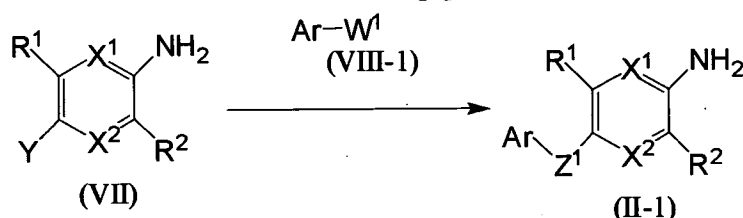
In production method [3], Ar, Z, R¹, R², R⁴, R⁵, X¹, X², B¹ and B² have the same meanings as described above.

In the production method [3], the compound of the formula (I) can be produced
 20 by reacting the compound of the formula (II) with an aminoacetal compound of the formula (VI). This reaction is conducted in the presence of an acid or a base. As the acid or base, the same acids or bases as used in the reaction in the first step of the production method [1], can be used. Furthermore, the reaction is conducted in the
 25 presence of an inert solvent. As the inert solvent, solvents that do not adversely affect the reaction, can be used. For example, the inert solvents described in the production

method [1] can be used. In the reaction, the aminoacetal compound of the formula (VI) is usually used in an amount of from 0.8 to 50 equivalents, preferably from 1 to 10 equivalents, per mole of the compound of the formula (II). The reaction is usually conducted at from 0 to 150°C, preferably from 20 to 120°C.

The compound of the formula (II) as the raw material in the production methods [1] to [3] can be produced according to the following intermediate production methods [1] to [3]. The compound of the formula (II) obtained in each production method can be isolated and purified by applying ordinary chemical operation such as extraction, concentration, distillation, crystallization, filtration, recrystallization or various chromatography.

Intermediate Production Method [1]



In intermediate production method [1], Ar, R¹, R², X¹ and X² have the same meanings as described above; Y is halogen, tosyloxy, SOMe, mesyloxy, or trifluoromethane sulfonyloxy; Z¹ is a bond or O; and W¹ is hydroxyl; boron atom which may be bonded to alkoxy or hydroxyl; silicon atom which may be bonded to alkoxy, hydroxyl or hydrocarbon; metal atom which may be bonded to halogen or hydrocarbon; or complex salt contains plural metal atoms. The metal atom includes Li, Mg, Zn, Sn, Al, Ni, Cu, Zr, and so on.

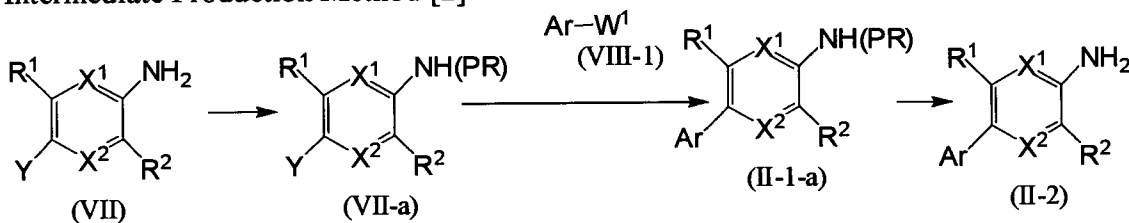
In the intermediate production method [1], a compound of the formula (II-1) can be produced by reacting an aminopyridine compound of the formula (VII) with a compound of the formula (VIII-1) in the presence of a transition metal catalyst. For example, the compound of the formula (II-1) can be produced by a reaction such as Suzuki-Miyaura coupling, Stille coupling, Negishi coupling or Kumada coupling (Angew. Chem. Int. Ed. 2002, 41, 4176-4211). The transition metal catalyst used in the reaction includes the following catalysts. Specific examples of the catalyst include tetrakis(triphenylphosphine)palladium, bis(dibenzylideneacetone)palladium, tetrakis(dibenzylideneacetone)dipalladium, palladium acetate-triphenylphosphine, palladium acetate-tricyclohexylphosphine, dichloropalladium-1,1'-bis(dicyclohexylphosphino)ferrocene, tetrakis(triphenylphosphine)nickel, bis(1,5-cyclooctadiene)nickel, nickel

acetylacetonate, dichlorobis(triphenylphosphine)nickel, tetrakis(triphenylphosphine)platinum, and copper sulfate. When the transition metal catalyst is a complex, the transition metal catalyst previously isolated may be used, or may be used by mixing a ligand in an optional reaction solvent without isolation.

The reaction is conducted in the presence of an inert solvent and a base. As the inert solvent, the same solvents as used in the production method [1], can be used. As the base, the same bases as used in the reaction in the first step of the production method [1], can be used.

The compound of the formula (VIII-1) is used in an amount of from 0.5 to 15 equivalents, preferably from 0.8 to 8 equivalents, per mole of the aminopyridine compound of the formula (VII). The transition metal catalyst is used in a proportion of from 0 to 50% by weight, preferably from 0 to 10% by weight, based on the weight of the aminopyridine compound of the formula (VII). The base is used in an amount of from 1 to 5 equivalents per mole of the aminopyridine compound of the formula (VII). Reaction temperature of the reaction is from -80 to 200°C, preferably from 20 to 200°C. Furthermore, the reaction is preferably conducted in an inert gas atmosphere such as nitrogen or argon.

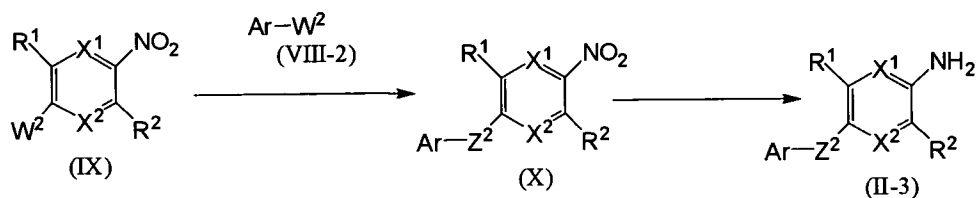
Intermediate Production Method [2]



In intermediate production method [2], Ar , R^1 , R^2 , X^1 , X^2 , Y and W^1 have the same meanings as described above; and PR is a protective group.

In the intermediate production method [2], a protective group is introduced into the aminopyridine compound of the formula (VII) by the ordinary method to prepare an aminopyridine compound of the formula (VII-a), and this compound can be converted to a compound of the formula (II-1-a) in the same manner as in the intermediate production method [1]. Further, when the protective group in the compound of the formula (II-1-a) is eliminated by the ordinary method, the compound of the formula (II-2) can be obtained.

Intermediate Production Method [3]



In intermediate production method [3], Ar, R¹, R², X¹ and X² have the same meanings as described above; Z² is O or OCH₂; and W² is halogen, tosyloxy, SOME, mesyloxy, trifluoromethane sulfonyloxy, hydroxyl or alkoxy.

In the intermediate production method [3], the compound of the formula (II-3) can be produced by reducing a compound of the formula (X). The method of reducing the compound of the formula (X) includes a method of catalytically hydrogenating using a reducing agent containing a transition metal such as palladium, platinum, nickel, rhodium, iridium, ruthenium or chromium, in a catalytic amount, and a method of reducing using tin chloride in the presence of hydrochloric acid. The reduction reaction can be conducted in the presence of a base, if necessary. As the base, the same bases as used in the reaction in the first step of the production method [1] can be used. Reaction temperature in the reduction reaction is from -80 to 100°C, preferably from 20 to 60°C.

The compound of the formula (X) can be produced by reacting a nitropyridine compound of the formula (IX) with a compound of the formula (VIII-2). The compound of the formula (VIII-2) can be used in an amount of from 0.5 to 15 equivalents, preferably from 0.8 to 8 equivalents, per mole of the nitropyridine compound of the formula (IX). Reaction temperature of the reaction is from -80 to 100°C, preferably from 20 to 60°C.

The reaction is conducted in the presence of an inert solvent and a base. As the inert solvent, the same inert solvents as used in the reaction in the second step of the production method [1] can be used. As the base, the same bases as used in the reaction in the first step of the production method [1] can be used.

The pyridylamidine compound represented by the formula (I) or a salt thereof (hereinafter referred to as the compound of the present invention) is useful as an active ingredient in an agricultural or horticultural fungicide composition. It has a great control effect against, for example, diseases of rice (*Oryza sativa*, etc.), such as blast, brown spot, and sheath blight; diseases of cereals (*Hordeum vulgare*, *Triticum aestivum*, etc.), such as powdery mildew, scab, rust, snowmold, loose smut, eye spot, leaf spot, and glume blotch; diseases of citrus (*Citrus spp.*, etc.), such as melanose, and scab; diseases of apple (*Malus pumila*), such as blossom blight, powdery mildew, Alternaria leaf spot, and scab; diseases of peach (*Prunus persica*, etc.), such as scab, and black spot; diseases

of peach (*Prunus persica*, etc.), such as brownrot, scab, and Phomopsis rot; diseases of grape (*Vitis vinifera* spp., etc.), such as anthracnose, ripe rot, powdery mildew, and powdery mildew, and downy mildew; diseases of Japanese persimmon (*Diospyros kaki*, etc.), such as anthracnose, and circular leaf spot; diseases of cucurbit (*Cucumis melo*, etc.), such as anthracnose, powdery mildew, gummy stem blight, and downy mildew; diseases of tomato (*Lycopersicon esculentum*), such as early blight, leaf mold, and late blight; diseases of cruciferous vegetables such as Alternaria diseases, disease of potato (*solanum tuberosum*), such as early blight, and late blight; diseases of strawberry (*Fragaria*, etc.) such as powdery mildew; diseases of various crops, such as graymold, disease caused by sclerotinia, especially powdery mildew of cereals and vegetables, and blast of rice. Furthermore, it has a control effect against soil borne disease caused by phytopathogenic fungus, such as Fusarium, Pythium, Rhizoctonia, Verticillium, and Plasmodiophora.

The compound of the present invention is usually formulated by mixing the compound with various agricultural adjuvants and used in the form of a formulation such as a dust, granules, water-dispersible granules, a wettable powder, a water-based suspension concentrate, an oil-based suspension concentrate, water soluble granules, an emulsifiable concentrate, a soluble concentrate, a paste, an aerosol or an ultra low-volume formulation. However, so long as it is suitable for the purpose of the present invention, it may be formulated into any type of formulation which is commonly used in this field. Such agricultural adjuvants include solid carriers such as diatomaceous earth, slaked lime, calcium carbonate, talc, white carbon, kaoline, bentonite, kaolinite, sericite, clay, sodium carbonate, sodium bicarbonate, mirabilite, zeolite and starch; solvents such as water, toluene, xylene, solvent naphtha, dioxane, acetone, isophorone, methyl isobutyl ketone, chlorobenzene, cyclohexane, dimethyl sulfoxide, N,N-dimethylformamide, dimethylacetamide, N-methyl-2-pyrrolidone, and alcohol; anionic surfactants such as a salt of fatty acid, a benzoate, an alkylsulfosuccinate, a dialkylsulfosuccinate, a polycarboxylate, a salt of alkylsulfuric acid ester, an alkyl sulfate, an alkylaryl sulfate, an alkyl diglycol ether sulfate, a salt of alcohol sulfuric acid ester, an alkyl sulfonate, an alkylaryl sulfonate, an aryl sulfonate, a lignin sulfonate, an alkyldiphenyl ether disulfonate, a polystyrene sulfonate, a salt of alkylphosphoric acid ester, an alkylaryl phosphate, a styrylaryl phosphate, a salt of polyoxyethylene alkyl ether sulfuric acid ester, a polyoxyethylene alkylaryl ether sulfate, a salt of polyoxyethylene alkylaryl ether sulfuric acid ester, a polyoxyethylene alkyl ether phosphate, a salt of polyoxyethylene alkylaryl phosphoric acid ester, and a salt of a condensate of naphthalene sulfonate with formalin; nonionic surfactants such as a sorbitan fatty acid ester, a glycerin fatty acid ester, a fatty acid polyglyceride, a fatty

acid alcohol polyglycol ether, acetylene glycol, acetylene alcohol, an oxyalkylene block polymer, a polyoxyethylene alkyl ether, a polyoxyethylene alkylaryl ether, a polyoxyethylene styrylaryl ether, a polyoxyethylene glycol alkyl ether, a polyethylene glycol, a polyoxyethylene fatty acid ester, a polyoxyethylene sorbitan fatty acid ester, a polyoxyethylene glycerin fatty acid ester, a polyoxyethylene hydrogenated castor oil, and a polyoxypropylene fatty acid ester; 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, cottonseed oil, soybean oil, rapeseed oil, linseed oil, tung oil, and liquid paraffins; and so on. Each of the components as such adjuvants may be one or more suitably selected for use, so long as the purpose of the present invention can thereby be accomplished. Further, various additives which are commonly used, such as a filler, a thickener, an anti-settling agent, an anti-freezing agent, a dispersion stabilizer, a phytotoxicity reducing agent, an anti-mold agent, and so on, may also be employed. The weight ratio of the compound of the present invention to the various agricultural adjuvants is usually from 0.005:99.995 to 95:5, preferably from 0.2:99.8 to 90:10.

Concentration of the compound used of the present invention varies depending on differences in objective crops, use method, preparation form, application amount and the like, and cannot necessarily be defined. However, in the case of foliage treatment, the concentration is usually from 0.1 to 10,000 ppm, preferably from 1 to 2,000 ppm, per the active ingredient. In the case of soil treatment, the concentration is usually from 10 to 100,000 g/ha, preferably from 200 to 20,000 g/ha, per the active ingredient.

Regarding various preparations and application of its dilution, the compound of the invention can be applied by the conventional application methods which are commonly employed, such as spraying (e.g. spraying, jetting, misting, atomizing, powder or grain scattering or dispersing in water), soil application (e.g. mixing or drenching), or surface application (e.g. coating, powdering or covering). Furthermore, the compound can be applied by a so-called ultrahigh concentration and small amount spraying method (ultra low volume). In this method, it is possible to contain the active ingredient in 100%.

If necessary, the compound of the invention can be mixed with other agricultural chemicals, such as other fungicide, an insecticides, a miticides, a nematocides, a soil insect pesticides, an antiviral agent, an attractant, a herbicide, a plant growth regulating agent and in this case, further excellent effect is sometimes exhibited.

The fungicidal active compounds in the above-mentioned other agricultural chemicals include, for example, (by common names, some of them are still in an application stage, or test codes of Japan Plant Protection Association) anilinopyrimidine

compounds such as mepanipyrim, pyrimethanil, cyprodinil and ferimzone; a triazoropyrimidine compound such as 5-chloro-7-(4-methylpiperidin-1-yl)-6-(2,4,6-trifluorophenyl)-[1,2,4]triazolo[1,5-a]pyrimidine; pyridinamine compounds such as fluazinam; azole compounds such as triadimefon, bitertanol, triflumizole, etaconazole, propiconazole, penconazole, flusilazole, myclobutanil, cyproconazole, tebuconazole, hexaconazole, furconazole-cis, prochloraz, metconazole, epoxiconazole, tetraconazole, oxpoconazole fumarate, sipconazole, prothioconazole, triadimenol, flutriafol, difenoconazole, fluquinconazole, fenbuconazole, bromuconazole, diniconazole, tricyclazole, probenazole, simeconazole, pefurazoate, ipconazole and imibenconazole; quinoxaline compounds such as quinomethionate; dithiocarbamate compounds such as maneb, zineb, mancozeb, polycarbamate, metiram, propineb and thiram; organic chlorine compounds such as fthalide, chlorothalonil and quintozone, imidazole compounds such as benomyl, thiophanate-methyl, carbendazim, thiabendazole, fuberiazole and cyazofamid; cyanoacetamide compounds such as cymoxanil; phenylamide compounds such as metalaxyl, metalaxyl-M, mefenoxam, oxadixyl, ofurace, benalaxyl, benalaxyl-M (another name: kiralaxyl, chiralaxyl), furalaxyl and cyprofuram; sulfenic acid compounds such as dichlofluanid; copper compounds such as cupric hydroxide and oxine copper; isoxazole compounds such as hymexazol; organophosphorus compounds such as fosetyl-Al, tolclofos-methyl, S-benzyl, O,O-diisopropylphosphorothioate, O-ethyl, S,S-diphenylphosphorodithioate and aluminum ethylhydrogen phosphonate, edifenphos, iprobenfos; N-halogenothioalkyl compounds such as captan, captafol and folpet; dicarboximide compounds such as procymidone, iprodione and vinclozolin; benzanilide compounds such as flutolanil, mepronil, zoxamid and tiadinil; anilide compounds such as carboxin, oxycarboxin, thifluzamide, penthiopyrad, boscalid, bixafen, fluopyram, isotianil and mixture of 2 *syn*-isomers 3-(difluoromethyl)-1-methyl-N-[(1RS,4SR,9SR)-1,2,3,4-tetrahydro-9-isopropyl-1,4-methanonaphthalen-5-yl]pyrazole-4-carboxamide and 2 *anti*-isomers 3-(difluoromethyl)-1-methyl-N-[(1RS,4SR,9SR)-1,2,3,4-tetrahydro-9-isopropyl-1,4-methanonaphthalen-5-yl]pyrazole-4-carboxamide (isopyrazam); piperazine compounds such as triforine; pyridine compounds such as pyrifenoxy; carbinol compounds such as fenarimol and flutriafol; piperidine compounds such as fenpropidine, morpholine compounds such as fenpropimorph, spiroxamine and tridemorph; organotin compounds such as fentin hydroxide and fentin acetate; urea compounds such as pencycuron; cinnamic acid compounds such as dimethomorph and flumorph; phenylcarbamate compounds such as diethofencarb, cyanopyrrole compounds such as fludioxonil and fenpiclonil; strobilurin compounds such as azoxystrobin, kresoxim-methyl, metominofen, trifloxystrobin, picoxystrobin, oryzastrobin, dimoxystrobin,

pyraclostrobin, and fluoxastrobin; oxazolidinone compounds such as famoxadone, thiazolecarboxamide compounds such as ethaboxam; silylamide compounds such as silthiopham; aminoacid amidecarbamate compounds such as iprovalicarb and methyl[S,(R,S)]-[(3-(N-isopopoxycarbonylvalinyl)-amino]-3-(4-chloro-phenyl)propionate (valiphenal); benthiavalicarb-isopropyl; imidazolidine compounds such as fenamidone; hydroxanilide compounds such as fenhexamid; benzenesulfonamide compounds such as flusulfamide; oxime ether compounds such as cyflufenamid; phenoxyamide compounds such as fenoxanil; antibiotics such as validamycin, kasugamycin and polyoxins; guanidine compounds such as iminoctadine and dodine; 4-quinolionol derivative compounds such as 2,3-dimethyl-6-t-butyl-8-fluoro-4-acetylquinoline; cyanomethylene compounds such as 2-(2-fluoro-5-(trifluoromethyl)phenylthio)-2-(3-(2-methoxyphenyl)thiazolidin-2-yliden)acetonitrile; and other compounds such as pyribencarb, isoprothiolane, Pyroquilon, diclomezine, quinoxifen, propamocarb hydrochloride, chloropicrin, dazomet, metam-sodium, nicobifen, metrafenone, UBF-307, diclocymet, proquinazid, amisulbrom (another name: ambromdole), mandipropamid, fluopicolide, carpropamid, and meptyldinocap.

The active compounds of insect pest control agents such as insecticides, miticides, nematocides or soil pesticides in the above-mentioned other agricultural chemicals, include, for example, (by common names, some of them are still in an application stage, or test codes) organic phosphate compounds such as profenofos, dichlorvos, fenamiphos, fenitrothion, EPN, diazinon, chlorpyrifos, chlorpyrifos-methyl, acephate, prothiofos, fosthiazate, cadusafos, dislufoton, isoxathion, isofenphos, ethion, etrimfos, quinalphos, dimethylvinphos, dimethoate, sulprofos, thiometon, vamidothion, pyraclofos, pyridaphenthion, pirimiphos-methyl, propaphos, phosalone, formothion, malathion, tetrachlovinphos, chlorfenvinphos, cyanophos, trichlorfon, methidathion, phenthoate, ESP, azinphos-methyl, fenthion, heptenophos, methoxychlor, paration, phosphocarb, demeton-S-methyl, monocrotophos, methamidophos, imicyafos, parathion-methyl, terbufos, phospamidon, phosmet and phorate; carbamate compounds such as carbaryl, propoxur, aldicarb, carbofuran, thiodicarb, methomyl, oxamyl, ethiofencarb, pirimicarb, fenobucarb, carbosulfan, benfuracarb, bendiocarb, furathiocarb, isoprocab, metolcarb, xylylcarb, XMC and fenothiocarb; nereistoxin derivatives such as cartap, thiocyclam, bensultap and thiosultap-sodium; organic chlorine compounds such as dicofol, tetradifon, endosulfan, dienochlor and dieldrin; organic metal compounds such as fenbutatin Oxide and cyhexatin, pyrethroid compounds such as fenvalerate, permethrin, cypermethrin, deltamethrin, cyhalothrin, tefluthrin, ethofenprox, flufenprox, cyfluthrin, fenpropathrin, flucythrinate, fluvalinate, cycloprothrin, lambda-cyhalothrin, pyrethrins, esfenvalerate, tetramethrin, resmethrin,

protrifenbute, bifenthrin, zeta-cypermethrin, acrinathrin, alpha-cypermethrin, allethrin, gamma-cyhalothrin, theta-cypermethrin, tau-fluvalinate, tralomethrin, profluthrin, beta-cypermethrin, beta-cyfluthrin, metofluthrin, phenothrin, imidate and flumethrin; benzoylurea compounds such as diflubenzuron, chlorfluazuron, teflubenzuron, flufenoxuron, triflumuron, hexaflumuron, lufenuron, novaluron, noviflumuron, bistrifluron and fluazuron; juvenile hormone-like compounds such as methoprene, pyriproxyfen, fenoxycarb and diofenolan; pyridazinone compounds such as pridaben; pyrazole compounds such as fenpyroximate, fipronil, tebufenpyrad, ethiprole, tolfenpyrad, acetoprole, pyrafluprole and pyriprole; neonicotinoids such as imidacloprid, nitenpyram, acetamiprid, thiacloprid, thiamethoxam, clothianidin, nidinotefuran, dinotefuran and nithiazine; hydrazine compounds such as tebufenozide, methoxyfenozide, chromafenozide and halofenozide; pyridine compounds such as pyridaryl and flonicamid; tetrone acid compounds such as spiroticlofen; strobilurin compounds such as fluacrypyrim; pyridinamine compounds such as flufenimer; dinitro compounds; organic sulfur compounds; urea compounds; triazine compounds; hydrazone compounds; and other compounds such as buprofezin, hexythiazox, amitraz, chlordimeform, silafluofen, triazamate, pymetrozine, pyrimidifen, chlorfenapyr, indoxacarb, acequinocyl, etoxazole, cyromazine, 1,3-dichloropropene, diafenthiuron, benclothiaz, bifenazate, spiromesifen, spirotetramat, propargite, clofentezine, metaflumizone, flubendiamide, cyflumetofen, chlorantraniliprole, cyenopyrafen, pyrifluquinazon, fenazaquin, pyridaben, amidoflumet, chlorobenzoate, sulfluramid, hydramethylnon, metaldehyde, HGW 86, ryanodine, flufenimer, pyridaryl, spiroticlofen, verbutin, thiazolylcinnanonitrile, amidoflumet, AKD-1022, IKA-2000, and the like. Further, microbial agricultural chemicals such as *Bacillus thuringiensis aizawai*, *Bacillus thuringiensis kurstaki*, *Bacillus thuringiensis israelensis*, *Bacillus thuringiensis japonensis*, *Bacillus thuringiensis tenebrionis*, insecticidal crystal protein produced by *Bacillus thuringiensis*, insect viruses, entomopathogenic fungi, and nematophagous fungi; antibiotics or semisynthetic antibiotics such as avermectin, emamectin-benzoate, milbemectin, milbemycin, spinosad, ivermectin, lepidectin, DE-175, abamectin and emamectin and spinetoram; natural products such as azadirachtin and rotenone; and repellents such as DEET may, for example, be mentioned.

EXAMPLES

Now, the present invention will be described in further detail with reference to Examples, but it should be understood that the present invention is by no means restricted thereto. Firstly, Preparation Examples of the compound of the present invention will be described.

Synthesis Example 1

Synthesis of N-ethyl-N-methyl-N'[5-(4-t-butyl-phenyl)-4-methylpyridin-2-yl]formamidine (Compound No.1)

(1) Triethylamine (101.7 mmol, 14 ml) and acetic anhydride (101.7 mmol, 9.6 ml) were added to a methylene chloride solution (250 ml) of 2-amino-4-picoline (92.5 mmol, 10 g) at 0°C, and the temperature was elevated to 40°C, followed by stirring for 2.5 hours. Water (200 ml) was added to the resulting reaction mixture to stop the reaction, and an aqueous layer was extracted with chloroform. An organic layer was washed with saturated brine, dried over anhydrous sodium sulfate, and filtered,

followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to obtain 8.6 g of 2-(acetylamino)-4-picoline (yield 62%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 8.21 (brs, 1H), 8.10 (d, 1H, J= 5.1Hz), 8.03 (s, 1H), 6.86 (dd, 1H, J= 0.9, 5.1Hz), 2.36 (s, 3H), 2.19 (s, 3H).

(2) N-Bromosuccinimide (6.8 mmol, 1.2 g) was added to a N,N'-dimethylformamide solution (20 ml) of 2-(acetylamino)-4-picoline (6.7 mmol, 1 g) at 0°C, followed by stirring at the same temperature for 30 minutes. Water (20 ml) was added to the resulting reaction mixture on ice to stop the reaction, and an aqueous layer was extracted with diethyl ether five times. An organic layer was dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to obtain 1.25 g of 2-(acetylamino)-5-bromo-4-picoline (yield 82%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 8.27 (s, 1H), 8.13 (s, 1H), 7.85 (brs, 1H), 2.41 (s, 3H), 2.20 (s, 3H).

(3) 4-t-Butylphenyl boronic acid (7.21 mmol, 1.28 g), Tris(dibenzylideneacetone)dipalladium(0) (0.20 mmol, 180 mg), tricyclohexylphosphine (1.31 mmol, 368 mg) and cesium carbonate (19.66 mmol, 6.41 g) were successively added to a dioxane solution (30 ml) of 2-(acetylamino)-amino-5-bromo-4-picoline (6.55 mmol, 1.5 g) at room temperature, and the temperature was elevated to 90°C, followed by stirring for 9.5 hours. Water (30 ml) was added to the resulting reaction mixture on ice to stop the reaction, and an aqueous layer was extracted with ethyl acetate. An organic layer was washed with saturated saline, dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to obtain 1.58 g of 2-(acetylamino)-amino-5-(4-t-butylphenyl)-4-picoline (yield 85%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 8.10 (s, 1H), 8.09 (s, 1H), 7.98 (brs, 1H), 7.45 (d, 2H, J=8.4Hz), 7.23 (d, 2H, J=8.7 Hz),

2.33 (s, 3H), 2.22 (s, 3H), 1.36 (s, 9H).

(4) 2N Hydrochloric acid (4 ml) was added to a dioxane solution (20 ml) of 2-(acetylamino)-amino-5-(4-t-butylphenyl)-4-picoline (4.11 mmol, 1.16 g) at room temperature, and the temperature was elevated to 90°C, followed by stirring for 2 hours.

5 An aqueous sodium hydrogen carbonate solution was added to the resulting reaction mixture on ice to stop the reaction, and an aqueous layer was extracted with ethyl acetate. An organic layer was washed with saturated saline, dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to
10 obtain 1.05 g of 2-amino-5-(4-t-butylphenyl)-4-picoline (yield 91%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 7.86 (s, 1H), 7.35 (d, 2H, J=8.7Hz), 7.15 (d, 2H, J= 8.4Hz), 6.35 (s, 1H), 4.28 (brs, 2H), 2.14 (s, 3H), 1.29 (s, 3H).

(5) p-Toluenesulfonic acid monohydrate (0.27 mmol, 46 mg) was added to a
15 trimethyl orthoformate solution (15 ml) of 2-amino-5-(4-t-butylphenyl)-4-picoline (2.71 mmol, 650 mg) at room temperature, followed by stirring under heat refluxing for 2 hours. Trimethyl orthoformate was distilled away under reduced pressure, and methylene chloride (15 ml) and N-ethylmethylaniline (4.06 mmol, 0.18 ml) were added to the resulting reaction mixture at room temperature, followed by stirring for 20 hours. A mixture was distilled away under reduced pressure, and the resulting crude product
20 was purified with column chromatography to obtain 306 mg of the objective product (yield 37%).

Synthesis Example 2

Synthesis of N-ethyl-N-methyl-N' [2,5-dimethyl-6-(4-t-butylphenyl)pyridin-3-yl]formamidine (Compound No. 11)

25 (1) Bromine (68.81 mmol, 3.5 ml) was gradually added dropwise to an aqueous solution of 48% hydrobromic acid (120 ml) of 2-amino-5-bromo-3-nitropyridine (22.93 mmol, 5.0 g) at 0°C, followed by stirring at the same temperature for 1 hour. An aqueous solution prepared by dissolving sodium nitrite (57.34 mmol, 4.0 g) in 60 ml of
30 water was gradually added dropwise, followed by stirring at the same temperature for 1.5 hours. To the resulting reaction mixture, 10 mol/liter sodium hydroxide aqueous solution was added on ice to neutralize, and an aqueous layer was extracted with ethyl acetate. An organic layer was washed with saturated saline, dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced
35 pressure. The resulting crude product was purified with column chromatography to obtain 2.24 g of 2,5-dibromo-3-nitropyridine (yield 35%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 8.66 (d, 2H, J=2.4Hz), 8.27 (d, 2H, J=2.1Hz).

(2) Tetrakis(triphenylphosphine)Palladium(0) (0.57 mmol, 656 mg), potassium carbonate (34.06 mmol, 4.71 g) and trimethylboroxine (17.03 mmol, 4.275 g) were successively added to a dioxane solution (15 ml) of 2,5-dibromo-3-nitropyridine (5.68 mmol, 1.6 g) in argon atmosphere, followed by stirring under heat refluxing for 7.5 hours. Water was added to the resulting reaction mixture on ice, followed by extracting with ethyl acetate. An organic layer was washed with saturated saline, dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to obtain 671 mg of 2,5-dimethyl-3-nitropyridine (yield 78%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 8.53 (s, 1H), 8.07 (s, 1H), 2.80 (s, 3H), 2.41 (s, 3H).

(3) Palladium carbon (65 mg) was added to a methanol solution (10ml) of 2,5-dimethyl-3-nitropyridine (5.52 mmol, 840 mg) at room temperature, followed by stirring in a hydrogen atmosphere for 15 hours. The resulting reaction mixture was filtered, followed by distilling away a solvent under reduced pressure, thereby obtaining 680 mg of 3-amino-2,5-dimethylpyridine. ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 7.76 (s, 1H), 6.75 (s, 1H), 3.54 (brs, 2H), 2.36 (s, 3H), 2.20 (s, 3H).

(4) N-Bromosuccinimide was added to a N,N-dimethylformamide solution (12 ml) of 3-amino-2,5-dimethylpyridine (4.09 mmol, 500 mg) at 0°C, followed by stirring at the same temperature for 30 minutes. Water was added to the resulting reaction mixture on ice to stop the reaction, and an aqueous layer was extracted with ethyl acetate. An organic layer was dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to obtain 710 mg of 3-amino-6-bromo-2,5-dimethylpyridine (yield 86%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 6.80 (s, 1H), 3.18 (brs, 2H), 2.34 (s, 3H), 2.25 (s, 3H).

(5) 4-t-Butylphenylboronic acid (1.79 mmol, 319 mg), tetrakis(triphenylphosphine)Palladium(0) (0.045 mmol, 45 mg) and cesium carbonate (4.48mmol, 1.46 g) were successively added to a dioxane solution (7.5 ml) of 3-amino-5-bromo-2,5-dimethylpyridine (1.49 mmol, 300 mg) in argon atmosphere, and the temperature was elevated to 90°C, followed by stirring for 15 hours. Water was added to the resulting reaction mixture on ice, and an aqueous layer was extracted with ethyl acetate. An organic layer was washed with saturated brine, dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to obtain 350 mg of 3-amino-2,5-dimethyl-6-(4-t-butylphenyl)pyridine (yield 92%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 7.40 (s, 4H), 6.83 (s, 1H),

3.55 (brs, 2H), 2.43 (s, 3H), 2.26 (s, 3H), 1.33 (s, 9H).

(6) 3-Amino-2,5-dimethyl-6-(4-t-butylphenyl)pyridine (1.38 mmol, 350 mg) was used as a raw material, and 220 mg of the objective product was obtained in the same manner as in Synthesis Example 1(5) (yield 49%).

5

Synthesis Example 3

Synthesis of N-methyl-N-ethyl-N'[2-(3-t-butyl-phenoxy)-3-methylpyridin-5-yl]formamidine (Compound No.210)

(1) Sodium hydride (2.61 mmol, 104 mg) was added to a dioxane solution (8 ml) of 3-t-butylphenol (2.26 mmol, 339 mg) at room temperature, followed by stirring in argon atmosphere for 30 minutes. 2-Chloro-3-methyl-5-nitropyridine (1.74 mmol, 300 mg) was added at the same temperature, followed by stirring under heat refluxing for 3 hours. Saturated aqueous ammonium chloride was added to the resulting reaction mixture on ice, and an aqueous layer was extracted with ethyl acetate. An organic layer was washed with saturated brine, dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to obtain 64 mg of 2-(3-t-butylphenoxy)-3-methyl-5-nitropyridine. ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 8.85 (d, 1H, J=2.7Hz), 8.32 (d, 1H, J=2.7 Hz), 7.38(dd, 1H, J=7.8, 7.8Hz), 7.31 (ddd, 1H, J=1.5, 1.8, 8.1Hz), 7.14 (dd, 1H, J=1.8, 2.1Hz), 6.96 (ddd, 1H, J=1.2, 2.1, 7.8Hz), 2.47 (s, 3H), 1.34 (s, 9H).

(2) Palladium carbon (44 mg) was added to a methanol solution (7.5 ml) of 2-(3-t-butylphenoxy)-3-methyl-5-nitropyridine (1.54 mmol, 440 mg) at room temperature, followed by stirring in hydrogen atmosphere for 15 hours. The resulting reaction mixture was filtered, and a solvent was distilled away under reduced pressure, thereby obtaining 350 mg of 5-amino-2-(3-t-butylphenoxy)-3-methylpyridine (yield 89%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 7.57 (d, 1H, J=2.4Hz), 7.22 (d, 1H, J=7.8Hz), 7.06-7.13 (m, 2H), 6.97 (dd, 1H, J=0.6, 2.7Hz), 6.74 (ddd, 1H, J=0.6, 2.4, 8.1Hz), 2.99 (brs, 2H), 2.25(s, 3H), 1.30 (s, 9H).

(3) 5-Amino-2-(3-t-butylphenoxy)-3-methylpyridine (0.59 mmol, 150 mg) was used as a raw material, and 98 mg of the objective product was obtained in the same manner as in Synthesis Example 1(5) (yield 52%).

Synthesis Example 4

Synthesis of N-ethyl-N-methyl-N'[5-(4-chloro-3-trifluoromethylphenoxy)-3,6-dimethylpyridin-2-yl]formamidine (Compound No.247)

(1) 2-Chloro-5-hydroxytrifluoromethylbenzene (11.76 mmol, 2.31 g), copper

sulfate (1.07 mmol, 171 mg) and potassium hydroxide (21.69 mmol, 1.20 mg) were successively added to a dimethoxyethane solution (30 ml) of 2-amino-5-bromo-6-methylpyridine (10.69 mmol, 2.0g) at room temperature, and the temperature was elevated to 160°C, followed by stirring in an autoclave for 16 hours. The temperature of the resulting reaction mixture was returned to room temperature, 28% aqueous ammonia was added, and an aqueous layer was extracted with ethyl acetate. An organic layer was washed with saturated brine, dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to obtain 400 mg of 2-amino-5-(4-chloro-3-trifluoro-methylphenoxy)-6-methylpyridine (yield 12%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 7.38(d, 1H, J=8.7Hz), 7.18 (d, 1H, J=3.0Hz), 7.12 (d, 1H, J=8.7Hz), 6.90 (dd, 1H, J=3.0, 8.7Hz), 6.44 (d, 1H, J=8.7Hz), 4.71 (brs, 2H), 2.25 (s, 3H).

(2) N-Bromosuccinimide (1.01 mmol, 180 mg) was added to an N,N-dimethylformamide solution (3 ml) of 2-amino-5-(4-chloro-3-trifluoromethylphenoxy)-6-methylpyridine (0.99 mmol, 300 mg) at 0°C, followed by stirring for 30 minutes. Water was added to the resulting reaction mixture on ice, and an aqueous layer was extracted with ethyl acetate. An organic layer was washed with saline, dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to obtain 219 mg of 2-amino-3-bromo-5-(4-chloro-3-trifluoromethylphenoxy)-6-methylpyridine (yield 58%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 7.40 (d, 1H, J=8.7Hz), 7.36 (s 1H), 7.20 (d, 1H, J=3.0Hz), 6.90 (dd, 1H, J=3.0, 8.7Hz), 4.97 (s, 2H), 2.22 (s, 3H).

(3) Tetrakis(triphenylphosphine)palladium(0) (0.03 mmol, 33 mg), potassium carbonate (1.72 mmol, 238 mg) and trimethylboroxine (0.86 mmol, 215 mg) were added to a dioxane solution (3 ml) of 2-amino-3-bromo-5-(4-chloro-3-trifluoromethylphenoxy)-6-methylpyridine (0.57 mmol, 219 mg) in an argon atmosphere, followed by stirring under heat refluxing for 7.5 hours. Water was added to the resulting reaction mixture on ice, followed by extracting with ethyl acetate. An organic layer was washed with saturated brine, dried over anhydrous sodium sulfate, and filtered, followed by distilling away a solvent under reduced pressure. The resulting crude product was purified with column chromatography to obtain 145 mg of 2-amino-5-(4-chloro-3-trifluoro-methylphenoxy)-3,6-dimethylpyridine (yield 80%). ¹H NMR δ ppm (300 MHz, CDCl₃) data of this compound are 7.37 (d, 1H, J=9.0Hz), 7.17 (d, 1H, J=2.7Hz), 6.97 (s, 1H), 6.88 (dd, 1H, J=3.0, 9.0Hz), 4.49 (brs, 2H), 2.22 (s, 3H), 2.11 (s, 3H).

(4) 2-Amino-5-(4-chloro-3-trifluoromethylphenoxy)-3,6-dimethylpyridine (0.46mmol, 145 mg) was used as a raw material, and 118 mg of the objective product was obtained in the same manner as in Synthesis Example 1 (5) (yield 67%).

Typical examples of the compound of the above formula (I) will be given in Table 1. These compounds can be prepared by the above-described Preparation Examples 1 to 4 or by the above-described various production methods. In Table 1, No. represents the compound No., Me represents methyl, Et represents ethyl, and Pr(n) represents normal propyl, Pr(i) represents iso-propyl, Bu(n) represents normal butyl, Bu(t) represents tertiary butyl, and the temperature shown as the physical properties is the melting point. Further, the left side of Z combines with Ar. On the other hand, ¹H-NMR data of some of the compounds of the above formula (I) are shown in Table 2.

Table 1

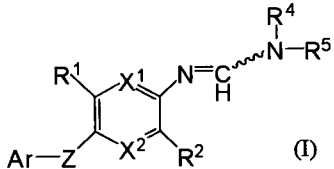
 (I)									
No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
1	4-Bu(t)-phenyl	—	N	CMe	H	H	Me	Et	Oil
2	4-Bu(t)-phenyl	—	N	CH	Me	H	Me	Et	82.0°C
3	4-Bu(t)-phenyl	—	N	CH	H	Me	Me	Et	Oil
4	4-Bu(t)-phenyl	—	N	CCF ₃	H	Me	Me	Et	Oil
5	4-Bu(t)-phenyl	—	N	CH	CF ₃	H	Me	Et	121.1°C
6	4-Bu(t)-phenyl	—	N	CH	CF ₃	Br	Me	Et	135.2°C
7	4-Bu(t)-phenyl	—	N	CH	Me	Me	Me	Et	101.6°C
8	4-Bu(t)-phenyl	—	N	CH	Me	Cl	Me	Et	Oil
9	4-Bu(t)-phenyl	—	N	CH	Me	Br	Me	Et	Oil
10	4-Bu(t)-phenyl	—	CMe	N	H	H	Me	Et	Oil
11	4-Bu(t)-phenyl	—	CH	N	Me	Me	Me	Et	102.0°C
12	4-Bu(t)-phenyl	—	CH	N	Me	H	Me	Et	Oil
13	4-Bu(t)-phenyl	—	CH	N	Me	Br	Me	Et	128.7°C
14	4-CF ₃ -phenyl	—	N	CMe	H	H	Me	Et	93.4°C
15	4-CF ₃ -phenyl	—	N	CH	Me	Me	Me	Et	72.1°C
16	4-CF ₃ -phenyl	—	N	CH	H	Me	Me	Et	Oil
17	4-CF ₃ -phenyl	—	N	CH	Me	H	Me	Et	119.1°C
18	4-CF ₃ -phenyl	—	N	CH	Me	Br	Me	Et	Oil
19	4-CF ₃ -phenyl	—	CMe	N	H	H	Me	Et	Oil
20	4-CF ₃ -phenyl	—	CH	N	Me	Me	Me	Et	Oil
21	4-CF ₃ -phenyl	—	CH	N	Me	H	Me	Et	69.2°C
22	4-CF ₃ -phenyl	—	CH	N	Me	Br	Me	Et	
23	4-CF ₃ -phenyl	—	CH	N	H	NO ₂	Me	Et	
24	3-Cl-4-F-phenyl	—	N	CMe	H	H	Me	Et	Oil
25	3-Cl-4-F-phenyl	—	N	CH	Me	Me	Me	Et	Oil
26	3-Cl-4-F-phenyl	—	N	CH	H	Me	Me	Et	65.8°C
27	3-Cl-4-F-phenyl	—	N	CH	Me	H	Me	Et	Oil
28	3-Cl-4-F-phenyl	—	N	CH	Me	Br	Me	Et	Oil

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
29	3-Cl-4-F-phenyl	—	CMe	N	H	H	Me	Et	
30	3-Cl-4-F-phenyl	—	CH	N	Me	Me	Me	Et	
31	3-Cl-4-F-phenyl	—	CH	N	Me	H	Me	Et	
32	3-Cl-4-F-phenyl	—	CH	N	Me	Br	Me	Et	
33	3-Cl-4-F-phenyl	—	CH	N	H	NO ₂	Me	Et	
34	4-OCF ₃ -phenyl	—	N	CMe	H	H	Me	Et	Oil
35	4-OCF ₃ -phenyl	—	N	CH	Me	Me	Me	Et	
36	4-OCF ₃ -phenyl	—	N	CH	H	Me	Me	Et	
37	4-OCF ₃ -phenyl	—	N	CH	Me	H	Me	Et	
38	4-OCF ₃ -phenyl	—	N	CH	Me	Br	Me	Et	
39	4-OCF ₃ -phenyl	—	CMe	N	H	H	Me	Et	
40	4-OCF ₃ -phenyl	—	CH	N	Me	Me	Me	Et	
41	4-OCF ₃ -phenyl	—	CH	N	Me	H	Me	Et	
42	4-OCF ₃ -phenyl	—	CH	N	Me	Br	Me	Et	
43	4-OCF ₃ -phenyl	—	CH	N	H	NO ₂	Me	Et	
44	4-Bu(t)-phenyl	—	N	CH	Et	H	Me	Et	96.5°C
45	4-Bu(t)-phenyl	—	N	CMe	H	H	Me	Pr(l)	Oil
46	4-Bu(t)-phenyl	—	N	CMe	H	H	Bu(n)	Bu(n)	Oil
47	4-Bu(t)-phenyl	—	N	CMe	H	H	Et	Et	Oil
48	4-Bu(t)-phenyl	—	CH	N	H	NO ₂	Me	Et	
49	phenyl	—	N	CMe	H	H	Me	Et	Oil
50	4-methyl-3-nitrophenyl	—	N	CMe	H	H	Me	Et	
51	4-methyl-3-nitrophenyl	—	N	CH	Me	Me	Me	Et	
52	4-methyl-3-nitrophenyl	—	N	CH	H	Me	Me	Et	
53	4-methyl-3-nitrophenyl	—	N	CH	Me	H	Me	Et	
54	4-methyl-3-nitrophenyl	—	N	CH	Me	Br	Me	Et	
55	4-methyl-3-nitrophenyl	—	CMe	N	H	H	Me	Et	
56	4-methyl-3-nitrophenyl	—	CH	N	Me	Me	Me	Et	
57	4-methyl-3-nitrophenyl	—	CH	N	Me	H	Me	Et	
58	4-methyl-3-nitrophenyl	—	CH	N	Me	Br	Me	Et	
59	4-methyl-3-nitrophenyl	—	N	CCF ₃	H	H	Me	Et	
60	4-Bu(t)-phenyl	—	N	CH	OMe	H	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
61	4-Bu(t)-phenyl	—	N	CH	OMe	Me	Me	Et	
62	4-Bu(t)-phenyl	—	N	CH	OEt	H	Me	Et	
63	4-Bu(t)-phenyl	—	N	CH	OEt	Me	Me	Et	
64	phenyl	—	CMe	N	H	H	Me	Et	
65	phenyl	—	N	CH	Me	Me	Me	Et	
66	phenyl	—	N	CH	H	Me	Me	Et	
67	phenyl	—	N	CH	Me	H	Me	Et	
68	phenyl	—	N	CH	Me	Br	Me	Et	
69	phenyl	—	CH	N	Me	Me	Me	Et	
70	phenyl	—	CH	N	Me	H	Me	Et	
71	4-methylphenyl	—	N	CMe	H	H	Me	Et	Oil
72	4-methylphenyl	—	N	CH	Me	Me	Me	Et	Oil
73	4-methylphenyl	—	N	CH	H	Me	Me	Et	
74	4-methylphenyl	—	N	CH	Me	H	Me	Et	Oil
75	4-methylphenyl	—	N	CH	Me	Br	Me	Et	
76	4-methylphenyl	—	CMe	N	H	H	Me	Et	
77	4-methylphenyl	—	CH	N	Me	Me	Me	Et	
78	4-methylphenyl	—	CH	N	Me	H	Me	Et	
79	4-methylphenyl	—	CH	N	Me	Br	Me	Et	
80	4-methylphenyl	—	N	CCF ₃	H	H	Me	Et	
81	4-Pr(i)-phenyl	—	N	CMe	H	H	Me	Et	Oil
82	4-Pr(i)-phenyl	—	N	CH	Me	Me	Me	Et	Oil
83	4-Pr(i)-phenyl	—	N	CH	H	Me	Me	Et	
84	4-Pr(i)-phenyl	—	N	CH	Me	H	Me	Et	57.7°C
85	4-Pr(i)-phenyl	—	N	CH	Me	Br	Me	Et	Oil
86	4-Pr(i)-phenyl	—	CMe	N	H	H	Me	Et	
87	4-Pr(i)-phenyl	—	CH	N	Me	Me	Me	Et	Oil
88	4-Pr(i)-phenyl	—	CH	N	Me	H	Me	Et	Oil
89	4-Pr(i)-phenyl	—	CH	N	Me	Br	Me	Et	
90	4-Pr(i)-phenyl	—	N	CCF ₃	H	H	Me	Et	
91	4-methoxyphenyl	—	N	CMe	H	H	Me	Et	Oil
92	4-methoxyphenyl	—	N	CH	Me	Me	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
93	4-methoxyphenyl	—	N	CH	H	Me	Me	Et	
94	4-methoxyphenyl	—	N	CH	Me	H	Me	Et	
95	4-methoxyphenyl	—	N	CH	Me	Br	Me	Et	
96	4-methoxyphenyl	—	CMe	N	H	H	Me	Et	
97	4-methoxyphenyl	—	CH	N	Me	Me	Me	Et	
98	4-methoxyphenyl	—	CH	N	Me	H	Me	Et	
99	4-methoxyphenyl	—	CH	N	Me	Br	Me	Et	
100	4-methoxyphenyl	—	CH	N	H	NO ₂	Me	Et	
101	4-methoxycarbonyl-phenyl	—	N	CMe	H	H	Me	Et	
102	4-methoxycarbonyl-phenyl	—	N	CH	Me	Me	Me	Et	
103	4-methoxycarbonyl-phenyl	—	N	CH	H	Me	Me	Et	
104	4-methoxycarbonyl-phenyl	—	N	CH	Me	H	Me	Et	
105	4-methoxycarbonyl-phenyl	—	N	CH	Me	Br	Me	Et	
106	4-methoxycarbonyl-phenyl	—	CMe	N	H	H	Me	Et	
107	4-methoxycarbonyl-phenyl	—	CH	N	Me	Me	Me	Et	
108	4-methoxycarbonyl-phenyl	—	CH	N	Me	H	Me	Et	
109	4-methoxycarbonyl-phenyl	—	CH	N	Me	Br	Me	Et	
110	2-Cl-4-CF ₃ -6-pyridyl	O	N	CH	Et	H	Me	Et	
111	2-Cl-4-CF ₃ -6-pyridyl	O	N	CH	Et	Me	Me	Et	
112	2-Cl-4-CF ₃ -6-pyridyl	O	N	CH	CF ₂ H	Me	Me	Et	
113	2-Cl-4-CF ₃ -6-pyridyl	O	N	CH	CF ₂ H	H	Me	Et	
114	2-Cl-4-CF ₃ -6-pyridyl	O	N	CH	H	CF ₂ H	Me	Et	
115	2-Cl-4-CF ₃ -6-pyridyl	O	N	CH	Me	CF ₂ H	Me	Et	
116	3,5-bis(trifluoromethyl)phenyl	OCH ₂	N	CH	Me	H	Me	Et	
117	3,5-bis(trifluoromethyl)phenyl	OCH ₂	N	CH	Me	Me	Me	Et	
118	3,5-bis(trifluoromethyl)phenyl	OCH ₂	N	CH	CF ₂ H	Me	Me	Et	
119	4-CF ₃ -phenyl	—	N	CCF ₃	H	H	Me	Et	
120	2,4,6-trimethylphenyl	—	N	CMe	H	H	Me	Et	
121	2,4,6-trimethylphenyl	—	N	CH	Me	Me	Me	Et	
122	2,4,6-trimethylphenyl	—	N	CH	H	Me	Me	Et	
123	2,4,6-trimethylphenyl	—	N	CH	Me	H	Me	Et	
124	2,4,6-trimethylphenyl	—	N	CH	Me	Br	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
125	2,4,6-trimethylphenyl	—	CMe	N	H	H	Me	Et	
126	2,4,6-trimethylphenyl	—	CH	N	Me	Me	Me	Et	
127	2,4,6-trimethylphenyl	—	CH	N	Me	H	Me	Et	
128	2,4,6-trimethylphenyl	—	CH	N	Me	Br	Me	Et	
129	2,4,6-trimethylphenyl	—	N	CCF ₃	H	H	Me	Et	
130	2,4,6-trimethylphenyl	—	CH	N	H	NO ₂	Me	Et	
131	3,5-dichlorophenyl	—	N	CMe	H	H	Me	Et	
132	3,5-dichlorophenyl	—	N	CH	Me	Me	Me	Et	
133	3,5-dichlorophenyl	—	N	CH	H	Me	Me	Et	
134	3,5-dichlorophenyl	—	N	CH	Me	H	Me	Et	
135	3,5-dichlorophenyl	—	N	CH	Me	Br	Me	Et	
136	3,5-dichlorophenyl	—	CMe	N	H	H	Me	Et	
137	3,5-dichlorophenyl	—	CH	N	Me	Me	Me	Et	
138	3,5-dichlorophenyl	—	CH	N	Me	H	Me	Et	
139	3,5-dichlorophenyl	—	CH	N	Me	Br	Me	Et	
140	4-Bu(n)-phenyl	—	N	CMe	H	H	Me	Et	
141	4-Bu(n)-phenyl	—	N	CH	Me	Me	Me	Et	
142	4-Bu(n)-phenyl	—	N	CH	H	Me	Me	Et	
143	4-Bu(n)-phenyl	—	N	CH	Me	H	Me	Et	
144	4-Bu(n)-phenyl	—	N	CH	Me	Br	Me	Et	
145	4-Bu(n)-phenyl	—	CMe	N	H	H	Me	Et	
146	4-Bu(n)-phenyl	—	CH	N	Me	Me	Me	Et	
147	4-Bu(n)-phenyl	—	CH	N	Me	H	Me	Et	
148	4-Bu(n)-phenyl	—	CH	N	Me	Br	Me	Et	
149	4-acetylphenyl	—	N	CMe	H	H	Me	Et	
150	4-acetylphenyl	—	N	CH	Me	Me	Me	Et	
151	4-acetylphenyl	—	N	CH	H	Me	Me	Et	
152	4-acetylphenyl	—	N	CH	Me	H	Me	Et	
153	4-acetylphenyl	—	N	CH	Me	Br	Me	Et	
154	4-acetylphenyl	—	CMe	N	H	H	Me	Et	
155	4-acetylphenyl	—	CH	N	Me	Me	Me	Et	
156	4-acetylphenyl	—	CH	N	Me	H	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
157	4-acetylphenyl	—	CH	N	Me	Br	Me	Et	
158	3-pyridyl	—	N	CMe	H	H	Me	Et	
159	3-pyridyl	—	N	CH	Me	Me	Me	Et	
160	3-pyridyl	—	N	CH	H	Me	Me	Et	
161	3-pyridyl	—	N	CH	Me	H	Me	Et	
162	3-pyridyl	—	N	CH	Me	Br	Me	Et	
163	3-pyridyl	—	CMe	N	H	H	Me	Et	
164	3-pyridyl	—	CH	N	Me	Me	Me	Et	
165	3-pyridyl	—	CH	N	Me	H	Me	Et	
166	3-pyridyl	—	CH	N	Me	Br	Me	Et	
167	3-pyridyl	—	CH	N	H	NO ₂	Me	Et	
168	4-pyridyl	—	N	CMe	H	H	Me	Et	
169	4-pyridyl	—	N	CH	Me	Me	Me	Et	
170	4-pyridyl	—	N	CH	H	Me	Me	Et	
171	4-pyridyl	—	N	CH	Me	H	Me	Et	
172	4-pyridyl	—	N	CH	Me	Br	Me	Et	
173	4-pyridyl	—	CMe	N	H	H	Me	Et	
174	4-pyridyl	—	CH	N	Me	Me	Me	Et	
175	4-pyridyl	—	CH	N	Me	H	Me	Et	
176	4-pyridyl	—	CH	N	Me	Br	Me	Et	
177	3-pyridyl	O	CH	N	Me	H	Me	Et	Oil
178	3-pyridyl	O	CH	N	Me	Br	Me	Et	
179	3-pyridyl	O	CH	N	Me	Me	Me	Et	
180	2-methyl-5-pyridyl	O	CH	N	Me	H	Me	Et	
181	2-methyl-5-pyridyl	O	CH	N	Me	Br	Me	Et	
182	2-methyl-5-pyridyl	O	CH	N	Me	Me	Me	Et	
183	2-cyano-3-pyridyl	O	CH	N	Me	H	Me	Et	
184	2-cyano-3-pyridyl	O	CH	N	Me	Br	Me	Et	
185	2-cyano-3-pyridyl	O	CH	N	Me	Me	Me	Et	
186	2-methyl-3-pyridyl	O	CH	N	Me	H	Me	Et	
187	2-methyl-3-pyridyl	O	CH	N	Me	Br	Me	Et	
188	2-methyl-3-pyridyl	O	CH	N	Me	Me	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
189	2-methoxy-3-pyridyl	O	CH	N	Me	H	Me	Et	
190	2-methoxy-3-pyridyl	O	CH	N	Me	Br	Me	Et	
191	2-methoxy-3-pyridyl	O	CH	N	Me	Me	Me	Et	
192	2-ethoxy-3-pyridyl	O	CH	N	Me	H	Me	Et	
193	2-ethoxy-3-pyridyl	O	CH	N	Me	Br	Me	Et	
194	2-ethoxy-3-pyridyl	O	CH	N	Me	Me	Me	Et	
195	2-Cl-4-CF ₃ -6-pyridyl	O	N	CMe	H	H	Me	Et	
196	2-Cl-4-CF ₃ -6-pyridyl	O	N	CH	Me	Me	Me	Et	
197	2-Cl-4-CF ₃ -6-pyridyl	O	N	CH	H	Me	Me	Et	
198	2-Cl-4-CF ₃ -6-pyridyl	O	N	CH	Me	H	Me	Et	
199	2-Cl-4-CF ₃ -6-pyridyl	O	N	CH	Me	Br	Me	Et	
200	6-CF ₃ -2-pyridyl	O	N	CMe	H	H	Me	Et	
201	6-CF ₃ -2-pyridyl	O	N	CH	Me	Me	Me	Et	
202	6-CF ₃ -2-pyridyl	O	N	CH	H	Me	Me	Et	
203	6-CF ₃ -2-pyridyl	O	N	CH	Me	H	Me	Et	
204	6-CF ₃ -2-pyridyl	O	N	CH	Me	Br	Me	Et	
205	5-CF ₃ -2-pyridyl	O	N	CMe	H	H	Me	Et	
206	5-CF ₃ -2-pyridyl	O	N	CH	Me	Me	Me	Et	
207	5-CF ₃ -2-pyridyl	O	N	CH	H	Me	Me	Et	
208	5-CF ₃ -2-pyridyl	O	N	CH	Me	H	Me	Et	
209	5-CF ₃ -2-pyridyl	O	N	CH	Me	Br	Me	Et	
210	3-Bu(t)-phenyl	O	CH	N	Me	H	Me	Et	Oil
211	3-Bu(t)-phenyl	O	CH	N	Me	Br	Me	Et	Oil
212	3-Bu(t)-phenyl	O	CH	N	Me	Me	Me	Et	Oil
213	3,5-bis(trifluoromethyl)phenyl	O	CH	N	Me	H	Me	Et	
214	3,5-bis(trifluoromethyl)phenyl	O	CH	N	Me	Br	Me	Et	
215	3,5-bis(trifluoromethyl)phenyl	O	CH	N	Me	Me	Me	Et	
216	4-chloro-3-methylphenyl	O	CH	N	Me	H	Me	Et	
217	4-chloro-3-methylphenyl	O	CH	N	Me	Br	Me	Et	
218	4-chloro-3-methylphenyl	O	CH	N	Me	Me	Me	Et	
219	3-(dimethylamino)phenyl	O	CH	N	Me	H	Me	Et	
220	3-(dimethylamino)phenyl	O	CH	N	Me	Br	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
221	3-(dimethylamino)phenyl	O	CH	N	Me	Me	Me	Et	
222	3-ethoxyphenyl	O	CH	N	Me	H	Me	Et	
223	3-ethoxyphenyl	O	CH	N	Me	Br	Me	Et	
224	3-ethoxyphenyl	O	CH	N	Me	Me	Me	Et	
225	4-F-3-methylphenyl	O	CH	N	Me	H	Me	Et	
226	4-F-3-methylphenyl	O	CH	N	Me	Br	Me	Et	
227	4-F-3-methylphenyl	O	CH	N	Me	Me	Me	Et	
228	3-CF ₃ -phenyl	O	CH	N	Me	H	Me	Et	Oil
229	3-CF ₃ -phenyl	O	CH	N	Me	Br	Me	Et	
230	3-CF ₃ -phenyl	O	CH	N	Me	Me	Me	Et	Oil
231	3,4-dicyanophenyl	O	CH	N	Me	H	Me	Et	
232	3,4-dicyanophenyl	O	CH	N	Me	Br	Me	Et	
233	3,4-dicyanophenyl	O	CH	N	Me	Me	Me	Et	
234	3-Me-4-Pr(i)-phenyl	O	CH	N	Me	H	Me	Et	Oil
235	3-Me-4-Pr(i)-phenyl	O	CH	N	Me	Br	Me	Et	
236	3-Me-4-Pr(i)-phenyl	O	CH	N	Me	Me	Me	Et	Oil
237	3-OMe-5-Me-phenyl	O	CH	N	Me	H	Me	Et	
238	3-OMe-5-Me-phenyl	O	CH	N	Me	Br	Me	Et	
239	3-OMe-5-Me-phenyl	O	CH	N	Me	Me	Me	Et	
240	4-NO ₂ -3-CF ₃ -phenyl	O	CH	N	Me	H	Me	Et	
241	4-NO ₂ -3-CF ₃ -phenyl	O	CH	N	Me	Br	Me	Et	
242	4-NO ₂ -3-CF ₃ -phenyl	O	CH	N	Me	Me	Me	Et	
243	3,4,5-trifluorophenyl	O	CH	N	Me	H	Me	Et	Oil
244	3,4,5-trifluorophenyl	O	CH	N	Me	Br	Me	Et	
245	3,4,5-trifluorophenyl	O	CH	N	Me	Me	Me	Et	Oil
246	4-Cl-3-CF ₃ -phenyl	O	N	CMe	H	H	Me	Et	
247	4-Cl-3-CF ₃ -phenyl	O	N	CH	Me	Me	Me	Et	Oil
248	4-Cl-3-CF ₃ -phenyl	O	N	CH	H	Me	Me	Et	
249	4-Cl-3-CF ₃ -phenyl	O	N	CH	Me	H	Me	Et	Oil
250	4-Cl-3-CF ₃ -phenyl	O	N	CH	Me	Br	Me	Et	
251	3-CF ₃ -phenyl	OCH ₂	CH	N	Me	H	Me	Et	
252	3-CF ₃ -phenyl	OCH ₂	CH	N	Me	Br	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
253	3-CF ₃ -phenyl	OCH ₂	CH	N	Me	Me	Me	Et	Oil
254	3-CF ₃ -phenyl	OCH ₂	N	CMe	H	H	Me	Et	
255	3-CF ₃ -phenyl	OCH ₂	N	CH	Me	Me	Me	Et	
256	3-CF ₃ -phenyl	OCH ₂	N	CH	H	Me	Me	Et	
257	3-CF ₃ -phenyl	OCH ₂	N	CH	Me	H	Me	Et	
258	3-CF ₃ -phenyl	OCH ₂	N	CH	Me	Br	Me	Et	
259	2-Cl-phenyl	OCH ₂	CH	N	Me	H	Me	Et	
260	2-Cl-phenyl	OCH ₂	CH	N	Me	Br	Me	Et	
261	2-Cl-phenyl	OCH ₂	CH	N	Me	Me	Me	Et	
262	2-Cl-phenyl	OCH ₂	N	CMe	H	H	Me	Et	
263	2-Cl-phenyl	OCH ₂	N	CH	Me	Me	Me	Et	
264	2-Cl-phenyl	OCH ₂	N	CH	H	Me	Me	Et	
265	2-Cl-phenyl	OCH ₂	N	CH	Me	H	Me	Et	
266	2-Cl-phenyl	OCH ₂	N	CH	Me	Br	Me	Et	
267	3-Cl-phenyl	OCH ₂	CH	N	Me	H	Me	Et	
268	3-Cl-phenyl	OCH ₂	CH	N	Me	Br	Me	Et	
269	3-Cl-phenyl	OCH ₂	CH	N	Me	Me	Me	Et	
270	3-Cl-phenyl	OCH ₂	N	CMe	H	H	Me	Et	
271	3-Cl-phenyl	OCH ₂	N	CH	Me	Me	Me	Et	
272	3-Cl-phenyl	OCH ₂	N	CH	H	Me	Me	Et	
273	3-Cl-phenyl	OCH ₂	N	CH	Me	H	Me	Et	
274	3-Cl-phenyl	OCH ₂	N	CH	Me	Br	Me	Et	
275	4-Cl-phenyl	OCH ₂	CH	N	Me	H	Me	Et	
276	4-Cl-phenyl	OCH ₂	CH	N	Me	Br	Me	Et	
277	4-Cl-phenyl	OCH ₂	CH	N	Me	Me	Me	Et	
278	4-Cl-phenyl	OCH ₂	N	CMe	H	H	Me	Et	
279	4-Cl-phenyl	OCH ₂	N	CH	Me	Me	Me	Et	
280	4-Cl-phenyl	OCH ₂	N	CH	H	Me	Me	Et	
281	4-Cl-phenyl	OCH ₂	N	CH	Me	H	Me	Et	
282	4-Cl-phenyl	OCH ₂	N	CH	Me	Br	Me	Et	
283	2,6-dichlorophenyl	OCH ₂	CH	N	Me	H	Me	Et	
284	2,6-dichlorophenyl	OCH ₂	CH	N	Me	Br	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
285	2,6-dichlorophenyl	OCH ₂	CH	N	Me	Me	Me	Et	
286	2,6-dichlorophenyl	OCH ₂	N	CMe	H	H	Me	Et	
287	2,6-dichlorophenyl	OCH ₂	N	CH	Me	Me	Me	Et	
288	2,6-dichlorophenyl	OCH ₂	N	CH	H	Me	Me	Et	
289	2,6-dichlorophenyl	OCH ₂	N	CH	Me	H	Me	Et	
290	2,6-dichlorophenyl	OCH ₂	N	CH	Me	Br	Me	Et	
291	2-Me-phenyl	OCH ₂	CH	N	Me	H	Me	Et	
292	2-Me-phenyl	OCH ₂	CH	N	Me	Br	Me	Et	
293	2-Me-phenyl	OCH ₂	CH	N	Me	Me	Me	Et	
294	2-Me-phenyl	OCH ₂	N	CMe	H	H	Me	Et	
295	2-Me-phenyl	OCH ₂	N	CH	Me	Me	Me	Et	
296	2-Me-phenyl	OCH ₂	N	CH	H	Me	Me	Et	
297	2-Me-phenyl	OCH ₂	N	CH	Me	H	Me	Et	
298	2-Me-phenyl	OCH ₂	N	CH	Me	Br	Me	Et	
299	3-NO ₂ -phenyl	OCH ₂	CH	N	Me	H	Me	Et	
300	3-NO ₂ -phenyl	OCH ₂	CH	N	Me	Br	Me	Et	
301	3-NO ₂ -phenyl	OCH ₂	CH	N	Me	Me	Me	Et	
302	3-NO ₂ -phenyl	OCH ₂	N	CMe	H	H	Me	Et	
303	3-NO ₂ -phenyl	OCH ₂	N	CH	Me	Me	Me	Et	
304	3-NO ₂ -phenyl	OCH ₂	N	CH	H	Me	Me	Et	
305	3-NO ₂ -phenyl	OCH ₂	N	CH	Me	H	Me	Et	
306	3-NO ₂ -phenyl	OCH ₂	N	CH	Me	Br	Me	Et	
307	2-Cl-4-F-phenyl	OCH ₂	N	CMe	H	H	Me	Et	
308	2-Cl-4-F-phenyl	OCH ₂	N	CH	Me	Me	Me	Et	
309	2-Cl-4-F-phenyl	OCH ₂	N	CH	H	Me	Me	Et	
310	2-Cl-4-F-phenyl	OCH ₂	N	CH	Me	H	Me	Et	
311	2-Cl-4-F-phenyl	OCH ₂	N	CH	Me	Br	Me	Et	
312	4-Bu(t)-phenyl	—	N	CMe	H	H	Me	Me	99.1°C
313	4-Bu(t)-phenyl	—	N	CH	Et	Br	Me	Et	110.5°C
314	4-Bu(t)-phenyl	—	N	CH	Et	Me	Me	Et	84.2°C
315	4-Bu(t)-phenyl	—	N	CH	Et	Et	Me	Et	
316	4-Bu(t)-phenyl	—	N	CH	Me	Et	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
317	4-Bu(t)-phenyl	—	N	CEt	H	H	Me	Et	
318	4-Bu(t)-phenyl	—	N	CMe	Me	Me	Me	Et	112.7°C
319	4-Bu(t)-phenyl	—	N	CMe	Me	H	Me	Et	102.0°C
320	4-Bu(t)-phenyl	—	N	CMe	H	Me	Me	Et	Oil
321	4-Bu(t)-phenyl	—	N	CH	CF ₃	Me	Me	Et	119.2°C
322	4-Bu(t)-phenyl	—	N	CH	CF ₃	CF ₃	Me	Et	90.7°C
323	4-Bu(t)-phenyl	—	N	CH	Me	CF ₃	Me	Et	Oil
324	4-Bu(t)-phenyl	—	N	CH	Me	Me	Me	Pr(n)	Oil
325	4-Bu(t)-phenyl	—	N	CH	Me	Me	Me	Pr(i)	81.9°C
326	4-Bu(t)-phenyl	—	N	CH	Me	Me	Me	Bu(t)	129.6°C
327	4-Bu(t)-phenyl	—	N	CH	Me	Me	Et	Et	
328	4-Bu(t)-phenyl	—	N	CH	Me	Me	Et	Pr(n)	
329	4-Bu(t)-phenyl	—	N	CH	Me	Me	Et	Pr(i)	Oil
330	4-Bu(t)-phenyl	—	N	CH	Me	Me	Me	Et	>300°C (hydrochloride)
331	4-Bu(t)-phenyl	—	N	CH	H	CF ₂ H	Me	Et	
332	4-Bu(t)-phenyl	—	N	CH	CF ₂ H	H	Me	Et	
333	4-Bu(t)-phenyl	—	N	CH	CF ₂ H	Me	Me	Et	
334	4-Bu(t)-phenyl	—	N	CH	Me	CF ₂ H	Me	Et	
335	2-naphthyl	—	N	CH	Me	H	Me	Et	Oil
336	2-naphthyl	—	N	CH	Me	Me	Me	Et	Oil
337	3-CF ₃ -phenyl	—	N	CH	Me	H	Me	Et	Oil
338	3-CF ₃ -phenyl	—	N	CH	Me	Me	Me	Et	Oil
339	4-(dimethylamino)phenyl	—	N	CH	Me	H	Me	Et	
340	4-(dimethylamino)phenyl	—	N	CH	Me	Me	Me	Et	Oil
341	4-Cl-3-CF ₃ -phenyl	O	CH	N	Me	H	Me	Et	Oil
342	4-Cl-3-CF ₃ -phenyl	O	CH	N	Me	Me	Me	Et	Oil
343	2-naphthyl	O	CH	N	Me	H	Me	Et	Oil
344	2-naphthyl	O	CH	N	Me	Me	Me	Et	Oil
345	3,4-methylenedioxyphenyl	O	CH	N	Me	H	Me	Et	Oil
346	3,4-methylenedioxyphenyl	O	CH	N	Me	Me	Me	Et	Oil
347	5,6,7,8-tetrahydro-2-naphthyl	O	CH	N	Me	H	Me	Et	Oil

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
348	5,6,7,8-tetrahydro-2-naphthyl	O	CH	N	Me	Me	Me	Et	Oil
349	4-Cl-3-Bu(t)-phenyl	O	CH	N	Me	H	Me	Et	Oil
350	4-Cl-3-Bu(t)-phenyl	O	CH	N	Me	Me	Me	Et	Oil
351	4-Cl-3-Bu(t)-phenyl	O	N	CMe	H	H	Me	Et	
352	4-Cl-3-Bu(t)-phenyl	O	N	CH	Me	H	Me	Et	Oil
353	4-Cl-3-Bu(t)-phenyl	O	N	CH	Me	Me	Me	Et	Oil
354	4-Cl-3-Bu(t)-phenyl	O	N	CH	Et	H	Me	Et	
355	4-Cl-3-Bu(t)-phenyl	O	N	CH	Et	Me	Me	Et	
356	4-Cl-3-Bu(t)-phenyl	O	N	CH	CF ₂ H	H	Me	Et	
357	4-Cl-3-Bu(t)-phenyl	O	N	CH	CF ₂ H	Me	Me	Et	
358	3-Bu(t)-phenyl	O	N	CH	H	H	Me	Et	Oil
359	3-Bu(t)-phenyl	O	N	CH	H	Me	Me	Et	Oil
360	3-Bu(t)-phenyl	O	N	CH	Me	H	Me	Et	
361	3-Bu(t)-phenyl	O	N	CH	Me	Me	Me	Et	Oil
362	3-Bu(t)-phenyl	O	N	CH	CF ₂ H	H	Me	Et	
363	3-Bu(t)-phenyl	O	N	CH	CF ₂ H	Me	Me	Et	
364	2-Bu(t)-phenyl	O	CH	N	Me	H	Me	Et	Oil
365	2-Bu(t)-phenyl	O	CH	N	Me	Me	Me	Et	Oil
366	4-Bu(t)-phenyl	O	CH	N	Me	H	Me	Et	Oil
367	4-Bu(t)-phenyl	O	CH	N	Me	Me	Me	Et	Oil
368	3,4-dichlorophenyl	O	N	CH	Me	H	Me	Et	
369	3,4-dichlorophenyl	O	N	CH	Me	Me	Me	Et	
370	3,4-dichlorophenyl	O	N	CMe	H	H	Me	Et	
371	3,4-dichlorophenyl	O	N	CH	CF ₂ H	H	Me	Et	
372	3,4-dichlorophenyl	O	N	CH	CF ₂ H	Me	Me	Et	
373	3-methoxyphenyl	O	CH	N	Me	H	Me	Et	Oil
374	3-methoxyphenyl	O	CH	N	Me	Me	Me	Et	Oil
375	3-CF ₃ -phenyl	O	N	CMe	H	H	Me	Et	
376	3-CF ₃ -phenyl	O	N	CH	Me	H	Me	Et	
377	3-CF ₃ -phenyl	O	N	CH	Me	Me	Me	Et	
378	3-CF ₃ -phenyl	O	N	CH	CF ₂ H	H	Me	Et	
379	3-CF ₃ -phenyl	O	N	CH	CF ₂ H	Me	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
380	3-CF ₃ -phenyl	O	N	CH	Et	H	Me	Et	
381	3-CF ₃ -phenyl	O	N	CH	Et	Me	Me	Et	
382	3-CF ₃ -phenyl	OCH ₂	CH	N	Me	Cl	Me	Et	semisolid
383	3-CF ₃ -phenyl	OCH ₂	N	CH	CF ₂ H	H	Me	Et	
384	3-CF ₃ -phenyl	OCH ₂	N	CH	CF ₂ H	Me	Me	Et	
385	3-OCF ₃ -phenyl	OCH ₂	CH	N	Me	Me	Me	Et	Oil
386	3-OCF ₃ -phenyl	OCH ₂	N	CH	Me	H	Me	Et	
387	3-OCF ₃ -phenyl	OCH ₂	N	CH	Me	Me	Me	Et	
388	3-OCF ₃ -phenyl	OCH ₂	N	CMe	H	H	Me	Et	
389	3-OCF ₃ -phenyl	OCH ₂	N	CH	CF ₂ H	H	Me	Et	
390	3-OCF ₃ -phenyl	OCH ₂	N	CH	CF ₂ H	Me	Me	Et	
391	3-OCF ₃ -phenyl	OCH ₂	N	CH	Et	H	Me	Et	
392	3-OCF ₃ -phenyl	OCH ₂	N	CH	Et	Me	Me	Et	
393	4-F-3-CF ₃ -phenyl	OCH ₂	CH	N	Me	Me	Me	Et	Oil
394	4-F-3-CF ₃ -phenyl	OCH ₂	N	CH	CF ₂ H	H	Me	Et	
395	4-F-3-CF ₃ -phenyl	OCH ₂	N	CH	CF ₂ H	Me	Me	Et	
386	3-OCF ₃ -phenyl	OCH ₂	N	CH	Me	H	Me	Et	
396	4-F-3-CF ₃ -phenyl	OCH ₂	N	CH	Me	H	Me	Et	
397	4-F-3-CF ₃ -phenyl	OCH ₂	N	CH	Me	Me	Me	Et	
398	4-F-3-CF ₃ -phenyl	OCH ₂	N	CH	Et	H	Me	Et	
399	4-F-3-CF ₃ -phenyl	OCH ₂	N	CH	Et	Me	Me	Et	
400	4-CF ₃ -3-pyridyl	OCH ₂	CH	N	Me	Cl	Me	Et	Oil
401	4-CF ₃ -3-pyridyl	OCH ₂	CH	N	Me	Me	Me	Et	Oil
402	4-CF ₃ -3-pyridyl	OCH ₂	N	CH	CF ₂ H	H	Me	Et	
403	4-CF ₃ -3-pyridyl	OCH ₂	N	CH	CF ₂ H	Me	Me	Et	
404	4-CF ₃ -3-pyridyl	OCH ₂	N	CH	Me	H	Me	Et	
405	4-CF ₃ -3-pyridyl	OCH ₂	N	CH	Me	Me	Me	Et	
406	4-CF ₃ -3-pyridyl	OCH ₂	N	CH	Et	H	Me	Et	
407	4-CF ₃ -3-pyridyl	OCH ₂	N	CH	Et	Me	Me	Et	
408	6-CF ₃ -2-pyridyl	OCH ₂	CH	N	Me	Cl	Me	Et	89.7°C
409	6-CF ₃ -2-pyridyl	OCH ₂	CH	N	Me	Me	Me	Et	67.8°C
410	6-CF ₃ -2-pyridyl	OCH ₂	N	CH	Me	H	Me	Et	

Table 1 continued

No.	Ar	Z	X ¹	X ²	R ¹	R ²	R ⁴	R ⁵	Physical Properties
411	6-CF ₃ -2-pyridyl	OCH ₂	N	CH	Me	Me	Me	Et	
412	6-CF ₃ -2-pyridyl	OCH ₂	N	CMe	H	H	Me	Et	
413	6-CF ₃ -2-pyridyl	OCH ₂	N	CH	CF ₂ H	H	Me	Et	
414	6-CF ₃ -2-pyridyl	OCH ₂	N	CH	CF ₂ H	Me	Me	Et	
415	4-Cl-3-CF ₃ -phenyl	O	N	CH	Et	H	Me	Et	
416	4-Cl-3-CF ₃ -phenyl	O	N	CH	Et	Me	Me	Et	Oil
417	4-Cl-3-CF ₃ -phenyl	O	N	CH	CF ₂ H	H	Me	Et	
418	4-Cl-3-CF ₃ -phenyl	O	N	CH	CF ₂ H	Me	Me	Et	
419	4-Cl-3-CF ₃ -phenyl	O	N	CH	Me	CF ₂ H	Me	Et	
420	3-CF ₃ -phenyl	OCH ₂	N	CH	H	CF ₂ H	Me	Et	
421	3-CF ₃ -phenyl	OCH ₂	N	CH	Me	CF ₂ H	Me	Et	
422	3-OCF ₃ -phenyl	OCH ₂	N	CH	H	CF ₂ H	Me	Et	
423	3-OCF ₃ -phenyl	OCH ₂	N	CH	Me	CF ₂ H	Me	Et	
424	4-F-3-CF ₃ -phenyl	OCH ₂	N	CH	H	CF ₂ H	Me	Et	
425	4-F-3-CF ₃ -phenyl	OCH ₂	N	CH	Me	CF ₂ H	Me	Et	
426	4-CF ₃ -3-pyridyl	OCH ₂	N	CH	H	CF ₂ H	Me	Et	
427	4-CF ₃ -3-pyridyl	OCH ₂	N	CH	Me	CF ₂ H	Me	Et	
428	6-CF ₃ -2-pyridyl	OCH ₂	N	CH	H	CF ₂ H	Me	Et	
429	6-CF ₃ -2-pyridyl	OCH ₂	N	CH	Me	CF ₂ H	Me	Et	
430	3-CF ₃ -phenyl	O	N	CH	H	CF ₂ H	Me	Et	
431	3-CF ₃ -phenyl	O	N	CH	Me	CF ₂ H	Me	Et	
432	3-Bu(t)-phenyl	O	N	CH	Me	CF ₂ H	Me	Et	
433	3-Bu(t)-phenyl	O	N	CH	H	CF ₂ H	Me	Et	
434	4-Cl-3-Bu(t)-phenyl	O	N	CH	Me	CF ₂ H	Me	Et	
435	4-Cl-3-Bu(t)-phenyl	O	N	CH	H	CF ₂ H	Me	Et	

Table 2

No.	¹ H-NMR δppm (Solvent : CDCl ₃ / 300MHz)
1	8.51, 8.39(s, 1H), 8.09(s, 1H), 7.43(d, 2H, J=6.6Hz), 7.25(d, 2H, J=7.2Hz), 6.87(s, 1H), 3.39, 3.60(q, 2H, J=7.2, 6.9Hz), 3.08, 3.05(s, 3H), 2.25(s, 3H), 1.36(s, 9H), 1.24(t, 3H, J=7.2Hz)
2	8.46, 8.36(s, 1H), 7.40-7.44(m, 3H), 7.23-7.26(m, 2H), 6.89(d, 1H, J=7.5Hz), 3.41, 3.64(q, 2H, J=7.2Hz), 3.12, 3.09(s, 3H), 2.46(s, 3H), 1.36(s, 9H), 1.24(t, 3H, J=7.2Hz)
3	8.51, 8.36(s, 1H), 8.32(d, 1H, J=1.8Hz), 7.62(d, 1H, J=2.4Hz), 7.50(d, 2H, J=8.7Hz), 7.45(d, 2H, J=8.4z), 3.41, 3.62(q, 2H, J=7.2, 6.9Hz), 3.10, 3.08(s, 3H), 2.36(s, 3H), 1.35(s, 9H), 1.26(t, 3H, J=7.2Hz)
4	8.59, 8.48(s, 1H), 8.22(s, 1H), 7.42(d, 2H, J=8.7Hz), 7.30(s, 1H), 7.26(d, 2H, J=8.1z), 3.43, 3.64(q, 2H, J=7.2Hz), 3.13, 3.11(s, 3H), 1.36(s, 9H), 1.27(t, 3H, J=7.2Hz)
5	8.62, 8.53(s, 1H), 7.52(d, 1H, 8.4Hz), 7.41(d, 2H, J=8.7Hz), 7.24(d, 2H, 8.1Hz), 7.12(d, 1H, J=8.1z), 3.44, 3.63(q, 2H, J=7.2Hz), 3.12(s, 3H), 1.36(s, 9H), 1.28(t, 3H, J=7.2Hz)
6	8.62, 8.47(s, 1H), 7.82, 7.81(s, 1H), 7.41(d, 2H, J=8.4Hz), 7.24(d, 2H, 8.1Hz), 3.48, 3.71(q, 2H, J=7.2, 6.9Hz), 3.21, 3.14(s, 3H), 1.35(s, 9H), 1.30(t, 3H, J=7.2Hz)
7	8.42, 8.28(s, 1H), 7.42(d, 2H, J=8.4Hz), 7.27(s, 1H), 7.27(d, 2H, 8.1Hz), 3.38, 3.60(q, 2H, J=7.2, 6.6Hz), 3.08, 3.06(s, 3H), 2.44(s, 3H), 2.30(s, 3H), 1.37(s, 9H), 1.24(t, 3H, J=6.9Hz)
8	8.51, 8.34(s, 1H), 7.50(s, 1H), 7.42(d, 2H, J=8.4Hz), 7.24(d, 2H, J=8.4Hz), 3.42, 3.66(q, 2H, J=7.2Hz), 3.14, 3.08(s, 3H), 2.41(s, 3H), 1.35(s, 9H), 1.26(t, 3H, J=7.2Hz)
9	8.50, 8.32(s, 1H), 7.67(s, 1H), 7.42(d, 2H, J=8.4Hz), 7.24(d, 2H, J=8.4Hz), 3.41, 3.65(q, 2H, J=7.2Hz), 3.14, 3.08(s, 3H), 2.40(s, 3H), 1.35(s, 9H), 1.26(t, 3H, J=7.2Hz)
10	8.04(s, 1H), 7.89(s, 1H), 7.86(s, 1H), 7.48(d, 2H, J=8.7Hz), 7.45(d, 2H, J=8.7Hz), 3.34(m, 2H), 3.03(s, 3H), 2.33(s, 3H), 1.35(s, 9H), 1.23(t, 3H, J=7.2Hz)
11	7.40-7.49(m, 5H), 6.93(s, 1H), 3.35(m, 2H), 3.04(s, 3H), 2.53(s, 3H), 2.30(s, 3H), 1.34(s, 9H), 1.23(t, 3H, J=7.2Hz)

Table 2 continued

No.	¹ H-NMR δppm (Solvent : CDCl ₃ / 300MHz)
12	8.16(s, 1H), 7.61(brs, 1H), 7.43(m, 4H), 7.15(s, 1H), 3.32(m, 2H), 3.01(s, 3H), 2.33(s, 3H), 1.33(s, 9H), 1.20(t, 3H, J=7.2Hz)
13	7.57, 7.40-7.48(brs, 1H), 7.40-7.48(m, 4H), 7.07, 7.02(brs, 1H), 3.36, 3.59(q, 2H, J=6.6Hz), 3.11, 3.05(s, 3H), 2.32(s, 3H), 1.34(s, 9H), 1.27(t, 3H, J=7.2Hz)
14	8.54, 8.42(s, 1H), 8.06(s, 1H), 7.67(d, 2H, J=7.8Hz), 7.43(d, 2H, J=8.1Hz), 6.89, 6.88(s, 1H), 3.40, 3.61(q, 2H, J=7.2Hz), 3.09, 3.07(s, 3H), 2.23(s, 3H), 1.24(t, 3H, J=7.2Hz)
15	8.43, 8.29(s, 1H), 7.65(d, 2H, J=8.4Hz), 7.43(d, 2H, J=8.1Hz), 7.24(s, 1H), 3.40, 3.61(q, 2H, J=7.2, 6.9Hz), 3.09, 3.04(s, H), 2.39(s, 3H), 2.30(s, 3H), 1.26(t, 3H, J=7.2Hz)
16	8.51, 8.36(s, 1H), 8.34(d, 1H, J=2.1Hz), 7.66(m, 4H), 7.62(d, 1H, J=1.8Hz), 3.41, 3.63(q, 2H, J=7.2Hz), 3.11, 3.08(s, 3H), 2.37, 2.36(s, 3H), 1.26(t, 3H, J=7.2Hz)
17	8.45, 8.35(s, 1H), 7.66(d, 2H, J=8.1Hz), 7.44(d, 2H, J=8.1Hz), 7.35(d, 1H, J=8.1Hz), 6.84(d, 1H, J=8.1Hz), 3.41, 3.61(q, 2H, J=7.2, 7.5Hz), 3.09(s, 3H), 2.43(s, 3H), 1.26(t, 3H, J=7.2Hz)
18	8.53, 8.35(s, 1H), 7.67(d, 2H, J=7.2Hz), 7.65(s, 1H), 7.43(d, 2H, J=8.4Hz), 3.43, 3.66(q, 2H, J=7.2Hz), 3.16, 3.10(s, 3H), 2.36(s, 3H), 1.28(t, 3H, J=7.2Hz)
19	8.06(m, 3H), 7.67(d, 2H, J=7.8Hz), 7.55(s, 1H), 7.55, 7.47(s, 1H), 3.34, 3.35(m, 2H), 3.05(s, 3H), 2.35(s, 3H), 1.24(t, 3H, J=7.2Hz).
20	7.66(d, 2H, J=8.7Hz), 7.62(d, 2H, J=8.4Hz), 7.51(brs, 1H), 6.97(s, 1H), 3.36, 3.55(m, 2H), 3.06(s, 3H), 2.54(s, 3H), 2.27(s, 3H), 1.24(t, 3H, J=7.2Hz)
21	8.19(s, 1H), 7.68(d, 2H, J=9.0Hz), 7.64(d, 2H, J=8.4Hz), 7.21(s, 1H), 3.36, 3.56(m, 2H), 3.05(s, 3H), 2.33(s, 3H), 1.24(t, 3H, J=7.2Hz)
24	8.51, 8.39(s, 1H), 8.01(s, 1H), 7.33(d, 2H, J=7.5Hz), 7.16(m, 2H, J=8.1Hz), 6.86, 6.83(s, 1H), 3.32, 3.59(q, 2H, J=7.2Hz), 3.08, 3.06(s, 3H), 2.20(s, 3H), 1.23(t, 3H, J=7.2Hz)
25	8.44, 8.30(s, 1H), 7.34(d, 1H, J=7.5Hz), 7.21(s, 1H), 7.16(m, 2H), 3.40, 3.60(q, 2H, J=7.2Hz), 3.08(s, 3H), 2.38(s, 3H), 2.28, 2.27(s, 3H), 1.25, 1.22(t, 3H, J=7.2Hz)

Table 2 continued

No.	¹ H-NMR δppm (Solvent : CDCl ₃ / 300MHz)
26	8.51, 8.36(s, 1H), 8.22(d, 1H, J=2.4Hz), 7.54(m, 2H), 7.37(m, 1H), 7.17(dd, 1H, J=8.4, 9.0Hz), 3.41, 3.61(q, 2H, J=7.2Hz), 3.10, 3.08(s, 3H), 2.35, 2.33(s, 3H), 1.25, 1.22(t, 3H, J=7.2Hz)
27	8.54, 8.45(s, 1H), 7.36(m, 2H), 7.16(m, 2H), 7.06(m, 1H), 3.41, 3.72(q, 2H, J=7.2Hz), 3.21, 3.14(s, 3H), 2.41(s, 3H), 1.29(t, 3H, J=7.2Hz)
28	8.55, 8.37(s, 1H), 7.63(s, 1H), 7.34(m, 1H), 7.17(m, 2H), 3.44, 3.66(q, 2H, J=7.2Hz), 3.15, 3.11(s, 3H), 2.36(s, 3H), 1.28(t, 3H, J=7.2Hz)
34	8.52, 8.40(s, 1H), 8.05(s, 1H), 7.33(d, 2H, J=6.3Hz), 7.25(d, 2H, J=7.5Hz), 6.86(s, 1H), 3.38, 3.59(q, 2H, J=7.2, 6.6Hz), 3.08(s, 3H), 2.21(s, 3H), 1.23(t, 3H, J=7.2Hz)
44	8.48, 8.39(s, 1H), 7.41(d, 2H, J=8.1Hz), 7.38(d, 1H, J=7.2Hz), 7.24(d, 2H, J=8.4Hz), 6.79(d, 1H, J=8.1Hz), 3.39, 3.61(q, 2H, J=7.2, 6.9Hz), 3.09, 3.07(s, 3H), 2.73(q, 2H, J=7.5Hz), 1.36(s, 9H), 1.25(t, 3H, J=7.5Hz), 1.21(t, 3H, J=7.5Hz)
45	8.59, 8.39(s, 1H), 8.10(s, 1H), 7.43(d, 2H, J=8.4Hz), 7.25(d, 2H, J=8.4Hz), 6.86, 6.84(s, 1H), 3.79(qq, 1H, J=6.6Hz), 2.99, 2.94(s, 3H), 2.25(s, 3H), 1.36(s, 9H), 1.27(d, 6H, J=6.6Hz)
46	8.46(s, 1H), 8.10(s, 1H), 7.43(d, 2H, J=8.1Hz), 7.25(d, 2H, J=8.1Hz), 6.84(s, 1H), 3.52(t, 2H, J=7.2Hz), 3.28(t, 2H, J=7.2Hz), 2.25(s, 3H), 1.61(m, 4H), 1.36(s, 9H), 1.27-1.41(m, 4H), 0.95(t, 3H, J=6.6Hz), 0.93(t, 3H, J=6.9Hz)
47	8.45(s, 1H), 8.10(s, 1H), 7.43(d, 2H, J=8.4Hz), 7.26(d, 2H, J=8.1Hz), 6.85(s, 1H), 3.59(q, 2H, J=6.9Hz), 3.37(q, 2H, J=7.2Hz), 2.25(s, 3H), 1.36(s, 9H), 1.23(t, 6H, J=7.2Hz)
49	8.56, 8.44(s, 1H), 8.08(s, 1H), 7.30-7.55(m, 5H), 6.96, 6.93(s, 1H), 3.41, 3.64(q, 2H, J=7.2Hz), 3.13, 3.09(s, 3H), 2.24(s, 3H), 1.25(t, 3H, J=7.2Hz)
71	8.53, 8.41(s, 1H), 8.07(s, 1H), 7.23(d, 2H, J=9.0Hz), 7.20(d, 2H, J=8.7Hz), 6.90, 6.88(s, 1H), 3.39, 3.61(q, 2H, J=7.2Hz), 3.10, 3.07(s, 3H), 2.40(s, 3H), 2.23(s, 3H), 1.24(t, 3H, J=6.9Hz)
72	8.45, 8.32(s, 1H), 7.26(s, 1H), 7.21(s, 4H), 3.41, 3.60(q, 2H, J=7.2, 6.9Hz), 3.08(s, 3H), 2.43(s, 3H), 2.39(s, 3H), 2.29(s, 3H), 1.26(t, 3H, J=7.2Hz)
74	8.42, 8.32(s, 1H), 7.39(d, 1H, J=8.1Hz), 7.21(s, 4H), 6.82(d, 1H, J=8.1Hz), 3.39, 3.61(q, 2H, J=7.2Hz), 3.08, 3.07(s, 3H), 2.47(s, 3H), 2.39(s, 3H), 1.25(t, 3H, J=7.2Hz)

Table 2 continued

No.	¹ H-NMR δppm (Solvent : CDCl ₃ / 300MHz)
81	8.53, 8.41(s, 1H), 8.09(s, 1H), 7.28(d, 2H, J=8.7Hz), 7.13(d, 2H, J=8.4Hz), 6.91, 6.89(s, 1H), 3.39, 3.61(q, 2H, J=7.2Hz), 3.10, 3.07(s, 3H), 2.95(qq, 1H, J=6.9, 6.9Hz), 2.25(s, 3H), 1.29(d, 6H, J=6.9Hz), 1.24(t, 3H, J=7.2Hz)
82	8.44, 8.30(s, 1H), 7.25(s, 5H), 3.40, 3.60(q, 2H, J=6.9Hz), 3.08(s, 3H), 2.95(qq, 1H, J=6.9, 6.9Hz), 2.43(s, 3H), 2.29(s, 3H), 1.29(d, 6H, J=6.9Hz), 1.25(t, 3H, J=7.2Hz)
84	8.44, 8.34(s, 1H), 7.42(d, 1H, J=8.1Hz), 7.25(s, 4H), 6.86(d, 1H, J=8.1Hz), 3.40, 3.62(q, 2H, J=7.2Hz), 3.10, 3.07(s, 3H), 2.95(qq, 1H, J=6.9, 6.9Hz), 2.45(s, 3H), 1.29(d, 6H, J=6.9Hz), 1.25(t, 3H, J=7.2Hz)
85	8.51, 8.33(s, 1H), 7.67(s, 1H), 7.27(d, 2H, J=8.1Hz), 7.22(d, 2H, J=8.4Hz), 3.42, 3.65(q, 2H, J=7.2Hz), 3.14, 3.09(s, 3H), 2.95(qq, 1H, J=6.9, 6.9Hz), 2.40(s, 3H), 1.29(d, 6H, J=6.9Hz), 1.27(m, 3H)
87	7.47(brs, 1H), 7.43(dd, 1H, J=1.8, 8.4Hz), 7.42(dd, 1H, J=1.8, 8.4Hz), 7.25(d, 2H, J=8.4Hz), 6.91(s, 1H), 3.34(m, 2H), 3.03(s, 3H), 2.93(qq, 1H, J=6.9, 6.9Hz), 2.52(s, 3H), 2.28(s, 3H), 1.26(d, 6H, J=7.2Hz), 1.23(t, 3H, J=7.2Hz)
88	8.17(d, 1H, J=2.1Hz), 7.64, 7.54(brs, 1H), 7.443(dd, 1H, J=1.8, 8.4Hz), 7.437(dd, J=1.8, 8.4Hz), 7.27(d, 2H, J=8.4Hz), 7.19(d, 1H, J=2.1Hz), 3.35, 3.55(m, 2H), 3.04(s, 3H), 2.94(qq, 1H, J=6.9, 6.9Hz), 2.34(s, 3H), 1.27(d, 6H, J=7.2Hz), 1.23(t, 3H, J=7.2Hz)
91	8.51, 8.39(s, 1H), 8.06(s, 1H), 7.24(d, 2H, J=9.0Hz), 6.97(d, 2H, J=9.0Hz), 6.86, 6.84(s, 1H), 3.84(s, 3H), 3.38, 3.59(q, 2H, J=7.2, 6.9Hz), 3.08, 3.06(s, 3H), 2.23(s, 3H), 1.23(t, 3H, J=7.2Hz)
177	8.42(d, 1H, J=2.4 Hz), 8.35(d, 1H, J=4.5Hz), 7.61(d, 1H, J=2.4 Hz), 7.52(brs, 1H), 7.40(ddd, 1H, J=1.2, 2.7, 8.4Hz), 7.27(dd, 1H, J=4.5, 8.4 Hz), 7.21(d, 1H, J=1.8 Hz), 3.31(m, 2H), 2.98(s, 3H), 2.28(s, 3H), 1.20(t, 3H, J=7.2Hz)
210	7.67(d, 1H, J=2.4 Hz), 7.53(brs, 1H), 7.20-7.28(m, 2H), 7.12(ddd, 1H, J=1.8, 1.8, 7.8 Hz), 7.09(dd, 1H, J=1.5, 2.1Hz), 6.80(ddd, 1H, J=2.1, 2.4, 8.1Hz), 3.32(m, 2H), 3.00(s, 3H), 2.28(s, 3H), 1.30(s, 9H), 1.21(t, 3H, J=7.2Hz)

Table 2 continued

No.	¹ H-NMR δppm (Solvent : CDCl ₃ / 300MHz)
211	7.51, 7.38(s, 1H), 7.21(d, 1H, J=8.4Hz), 7.09-7.11(m, 3H), 6.80(dd, 1H, J=1.2, 7.8Hz), 3.26, 3.54(q, 2H, J=6.9Hz, m, 2H), 3.05(s, 3H), 2.22(s, 3H), 1.30(s, 9H), 1.23(t, 3H, J=7.2Hz)
212	7.44(brs, 1H), 7.18(dd, 2H, J=7.5, 7.8Hz), 7.04(m, 2H), 6.97(s, 1H), 6.66(dd, 1H, J=2.1, 8.1Hz), 3.35(m, 2H), 3.01(s, 3H), 2.37(s, 3H), 2.18(s, 3H), 1.28(s, 9H), 1.22(t, 3H, J=7.2Hz)
228	7.67(d, 1H, J=2.7Hz), 7.56(s, 1H), 7.44(dd, 1H, J=7.8, 8.1Hz), 7.35(d, 1H, J=7.8Hz), 7.28(d, 1H, J=5.1Hz), 7.22(d, 1H, J=8.1Hz), 3.33(m, 2H), 3.03(s, 3H), 2.28(s, 3H), 1.22(t, 3H, J=7.2Hz)
230	7.45(brs, 1H), 7.39(dd, 1H, J=7.8, 8.1Hz), 7.28(d, 1H, J=8.4Hz), 7.21(s, 1H), 7.14(dd, 1H, J=1.8, 8.1Hz), 7.01(s, 1H), 3.35(m, 2H), 3.04(s, 3H), 2.37(s, 3H), 2.18(s, 3H), 1.23(t, 3H, J=7.2Hz)
234	7.66(d, 1H, J=2.7Hz), 7.52(brs, 1H), 7.19(m, 2H), 6.83(m, 2H), 3.31(m, 2H), 3.08(qq, 1H, J=6.9, 6.9Hz), 3.00(s, 3H), 2.29(s, 3H), 2.27(s, 3H), 1.20(t, 3H, J=7.2Hz), 1.20(d, 6H, J=6.9Hz)
236	7.44(brs, 1H), 7.12(d, 1H, J=8.4Hz), 6.96(s, 1H), 6.70-6.76(m, 2H), 3.35(m, 2H), 3.07(qq, 1H, J=6.9, 6.9Hz), 3.01(s, 3H), 2.37(s, 3H), 2.26(s, 3H), 2.17(s, 3H), 1.22(t, 3H, J=7.2Hz), 1.19(d, 6H, J=6.9Hz)
243	7.65(d, 1H, J=2.4Hz), 7.53(brs, 1H), 7.22(s, 1H, J=2.1Hz), 6.69(m, 2H), 3.32, 3.51(m, 2H), 3.01(s, 3H), 2.24(s, 3H), 1.22(t, 3H, J=7.2Hz)
245	7.44(brs, 1H), 6.98(s, 1H), 6.60(m, 2H), 3.34, 3.49(m, 2H), 3.01(s, 9H), 2.37(s, 3H), 2.15(s, 3H), 1.22(t, 3H, J=7.2Hz)
247	8.40, 8.26(s, 1H), 7.37(d, 1H, J=8.7Hz), 7.18(d, 1H, J=3.0Hz), 7.03(s, 1H), 6.90(dd, 1H, J=3.0, 8.7Hz), 3.40, 3.60(q, 2H, J=7.2Hz), 3.08(s, 3H), 2.27(s, 6H), 1.26(t, 3H, J=7.2Hz)
249	8.48, 8.37(s, 1H), 7.39(d, 1H, J=8.7Hz), 7.18(m, 2H), 7.02(m, 1H), 6.91(dd, 1H, J=2.7, 8.7Hz), 3.44, 3.68(q, 2H, J=7.2, 6.6Hz), 3.17, 3.12(s, 3H), 2.32(s, 3H), 1.28(t, 3H, J=7.2Hz)

Table 2 continued

No.	¹ H-NMR δppm (Solvent : CDCl ₃ / 300MHz)
253	7.76(s, 1H), 7.65(d, 1H, J=7.2Hz), 7.53(d, 1H, J=7.8Hz), 7.47(d, 1H, J=7.8 Hz), 7.38(s, 1H), 6.88(s, 1H), 5.43(s, 2H), 3.34(m, 2H), 2.99(s, 3H), 2.38(s, 3H), 2.18(s, 3H), 1.20(t, 3H, J=7.2Hz)
313	8.56, 8.38(s, 1H), 7.65(s, 1H), 7.41(d, 2H, J=8.1Hz), 7.22(d, 2H, J=8.1Hz), 3.42, 3.66(q, 2H, J=7.2Hz), 3.15, 3.09(s, 3H), 2.67(q, 2H, J=7.2Hz), 1.36(s, 9H), 1.27(t, 3H, J=7.2Hz), 1.19(t, 3H, J=7.5Hz)
314	8.46, 8.33(s, 1H), 7.40(d, 2H, J=8.4Hz), 7.24(d, 2H, J=8.4Hz), 7.23(s, 1H), 3.39, 3.60(q, 2H, J=6.9Hz), 3.08(s, 3H), 2.70(q, 2H, J=7.5Hz), 2.28(s, 3H), 1.36(s, 9H), 1.22(t, 3H, J=8.1Hz), 1.20(t, 3H, J=7.5Hz)
318	8.37(s, 1H), 7.40(dd, 1H, J=2.1, 8.4Hz), 7.39(dd, 1H, J=2.1, 8.4Hz), 7.05(d, 1H, J=2.1, 8.4Hz), 7.04(dd, 1H, J=2.1, 8.4Hz), 3.37, 3.59(q, 2H, J=6.9Hz), 3.07(s, 3H), 2.29(s, 3H), 2.16(s, 3H), 1.93(s, 3H), 1.36(s, 9H), 1.23(t, 3H, J=7.2Hz)
319	8.40, 8.31(s, 1H), 7.40(d, 2H, J=8.4Hz), 7.07(d, 2H, J=8.4Hz), 6.70(s, 1H), 3.38, 3.59(q, 2H, J=7.2, 6.9Hz), 3.06(s, 3H), 2.20(s, 3H), 1.99(s, 3H), 1.36(s, 9H), 1.23(t, 3H, J=7.2Hz)
320	8.42, 8.27(s, 1H), 7.98(s, 1H), 7.432(dd, 1H, J=2.1, 8.7H), 7.425(dd, 1H, J=2.1, 8.7Hz), 7.233(dd, 1H, J=2.1, 8.4Hz), 7.226(dd, 1H, J=2.1, 8.4Hz), 3.38, 3.61(q, 2H, J=7.2Hz), 3.09, 3.05(s, 3H), 2.36(s, 3H), 2.19(s, 3H), 1.37(s, 9H), 1.26(t, 3H, J=7.2Hz)
321	8.57, 8.45(s, 1H), 7.403(dd, 1H, J=2.4, 8.7Hz), 7.395(dd, 1H, J=2.1, 8.7Hz), 7.36(s, 1H), 7.24(d, 2H, J=9.0Hz), 3.43, 3.63(q, 2H, J=7.2Hz), 3.11(s, 3H), 2.35, 2.33(s, 3H), 1.36(s, 9H), 1.27(t, 3H, J=7.2Hz)
322	8.72, 8.60(s, 1H), 7.80(s, 1H), 7.43(d, 2H, J=8.4Hz), 7.21(d, 2H, J=8.4Hz), 3.47, 3.64(q, 2H, J=7.2Hz), 3.15(s, 3H), 1.36(s, 9H), 1.26(t, 3H, J=7.2Hz)
323	8.59, 8.44(s, 1H), 7.67(s, 1H), 7.44(d, 2H, J=8.1Hz), 7.25(d, 2H, J=8.4Hz), 3.42, 3.61(q, 2H, J=7.2Hz), 3.11, 3.10(s, 3H), 2.46(s, 3H), 1.36(s, 9H), 1.27(t, 3H, J=7.2Hz)

Table 2 continued

No.	¹ H-NMR δppm (Solvent : CDCl ₃ / 300MHz)
324	8.38, 8.32(s, 1H), 7.41(d, 2H, J=8.4Hz), 7.25(m, 3H), 3.28, 3.51(t, 2H, J=6.9Hz), 3.07(s, 3H), 2.42(s, 3H), 2.28(s, 3H), 1.67(m, 2H), 1.36(s, 9H), 0.92(t, 3H, J=7.5Hz)
325	8.51, 8.26(s, 1H), 7.41(dd, 1H, J=2.1, 8.4Hz), 7.40(dd, 1H, J=2.1, 8.4 Hz), 7.26(m, 3H), 3.81(qq, 1H, J=6.9, 6.9Hz), 2.99, 2.94(s, 3H), 2.42(s, 3H), 2.28(s, 3H), 1.36(s, 9H), 1.28(d, 3H, J=6.9Hz), 1.27(d, 3H, J=6.9Hz)
326	8.74(s, 1H), 7.41(d, 2H, J=8.4Hz), 7.24-7.27(m, 3H), 3.10(s, 3H), 2.42(s, 3H), 2.29(s, 3H), 1.41(s, 9H), 1.36(s, 9H), 1.26(t, 3H, J=7.2Hz)
329	7.44(d, 2H, J=8.1Hz), 7.40, 7.32(s, 1H), 7.23(s, 1H), 7.22(d, 1H, J=8.1Hz), 3.81(qq, 1H, J=6.9, 6.9Hz), 3.51(q, 2H, J=7.5Hz), 2.42(s, 3H), 2.28(s, 3H), 1.36(s, 9H), 1.28(d, 3H, J=6.9Hz), 1.27(d, 3H, J=6.9Hz), 1.26(t, 3H, J=7.2Hz)
330	9.01, 8.90(s, 1H), 7.82, 7.79(s, 1H), 7.50(d, 2H, J=8.1Hz), 7.21(d, 2H, J=8.4Hz), 3.64, 3.94(q, 2H, J=7.2, 7.5Hz), 3.47, 3.43(s, 3H), 1.46(t, 3H, J=7.2Hz), 1.37(s, 9H)
336	8.50, 8.37(s, 1H), 7.84-7.88(m, 3H), 7.76(s, 1H), 7.44-7.54(m, 3H), 7.38(s, 1H), 3.43, 3.63(q, 2H, J=7.2Hz), 3.10(s, 3H), 2.48(s, 3H), 2.33(s, 3H), 1.27(t, 3H, J=7.2Hz)
337	8.52, 8.42(s, 1H), 7.41-7.60(m, 5H), 7.00(m, 1H), 3.45, 3.69(q, 2H, J=7.2Hz), 3.17, 3.12(s, 3H), 2.42(s, 3H), 1.28(t, 3H, J=7.2Hz).
338	8.46, 8.33(s, 1H), 7.49-7.57(m, 5H), 3.41, 3.61(q, 2H, J=7.2, 6.9Hz), 3.09(s, 3H), 2.40(s, 3H), 2.30(s, 3H), 1.26(t, 3H, J=7.2Hz)
341	7.63(d, 1H, J=2.4Hz), 7.54, 7.49(brs, 1H), 7.44(d, 1H, J=8.7Hz), 7.39(d, 1H, J=3.0Hz), 7.23(d, 1H, J=1.8Hz), 7.17(dd, 1H, J=2.7, 9.0Hz), 3.32, 3.50(m, 2H), 3.01(s, 3H), 2.27(s, 3H), 1.21(t, 3H, J=7.2Hz)
342	7.44(brs, 1H), 7.39(d, 1H, J=8.7Hz), 7.33(d, 1H, J=3.0Hz), 7.09(dd, 1H, J=3.0, 8.7Hz), 6.99(s, 1H), 3.34, 3.50(m, 2H), 3.02(s, 3H), 2.36(s, 3H), 2.18(s, 3H), 1.22(t, 3H, J=7.2Hz)
343	7.81(m, 2H), 7.71(m, 2H), 7.55(brs, 1H), 7.35-7.45(m, 3H), 7.27(m, 2H), 3.33, 3.51(m, 2H), 3.02(s, 3H), 2.31(s, 3H), 1.22(t, 3H, J=7.2Hz)
344	7.78(d, 2H, J=8.7Hz), 7.64(d, 1H, J=8.1Hz), 7.47(brs, 1H), 7.31-7.43(m, 2H), 7.26(dd, 1H, J=2.1, 8.7Hz), 7.15(d, 1H, J=2.1Hz), 7.02(s, 1H), 3.35(m, 2H), 3.02(s, 3H), 2.40(s, 3H), 2.19(s, 3H), 1.23(t, 3H, J=6.9Hz)

Table 2 continued

No.	¹ H-NMR δppm (Solvent : CDCl ₃ / 300MHz)
345	7.62(d, 1H, J=2.4Hz), 7.50(brs, 1H), 7.18(d, 1H, J=2.1Hz), 6.76(d, 1H, J=8.1Hz), 6.60(d, 1H, 2.4Hz), 6.51(dd, 1H, J=2.4, 8.4Hz), 5.95(s, 2H), 3.31(m, 2H), 2.99(s, 3H), 2.27(s, 3H), 1.20(t, 3H, J=7.2Hz)
346	7.41(s, 1H), 6.95(s, 1H), 6.70(d, 1H, J=8.7Hz), 6.55(d, 1H, J=2.4Hz), 6.40(dd, 1H, J=2.4, 8.4Hz), 5.93(s, 2H), 3.34(brs, 2H), 3.00(s, 3H), 2.35(s, 3H), 2.17(s, 3H), 1.21(t, 3H, J=7.2Hz)
347	7.64(d, 1H, J=2.4Hz), 7.51(s, 1H), 7.19(d, 1H, J=2.4Hz), 7.02(s, 1H, J=8.4Hz), 6.77(dd, 1H, J=2.1, 8.4Hz), 6.74(d, 1H, J=2.1Hz), 3.31(m, 2H), 2.99(s, 3H), 2.73(m, 4H), 2.27(s, 3H), 1.77(tt, 4H, J=3.0, 3.0Hz), 1.20(t, 3H, J=7.2Hz)
348	7.43(s, 1H), 6.95(m, 2H), 6.68(dd, 1H, J=2.4, 8.4Hz), 6.61(d, 1H, J=2.1Hz), 3.35(m, 2H), 3.01(s, 9H), 2.69(m, 4H), 2.37(s, 3H), 2.17(s, 3H), 1.75(tt, 4H, J=3.0, 3.3Hz), 1.22(t, 3H, J=7.2Hz)
349	7.65(d, 1H, J=2.7Hz), 7.53(brs, 1H), 7.27(d, 1H, J=8.7Hz), 7.22(d, 1H, J=2.1Hz), 7.15(d, 1H, J=2.7Hz), 6.76(dd, 1H, J=2.7, 8.7Hz), 3.32(m, 2H), 3.01(s, 3H), 2.27(s, 3H), 1.45(s, 9H), 1.21(t, 3H, J=7.2Hz)
350	7.43(s, 1H), 7.20(d, 1H, J=8.7Hz), 7.15(d, 1H, J=3.0Hz), 6.97(s, 1H), 6.63(dd, 1H, J=3.0, 8.7Hz), 3.35(m, 1H), 3.01(s, 3H), 2.36(s, 3H), 2.18(s, 3H), 1.44(s, 9H), 1.22(t, 3H, J=7.2Hz)
352	8.39, 8.29(s, 1H), 7.20(d, 1H, J=8.4Hz), 7.15(d, 1H, J=8.7Hz), 7.03(d, 1H, J=3.0Hz), 6.81(d, J=8.1Hz), 6.52(dd, 1H, J=3.0, 8.7Hz), 3.39, 3.60(q, 2H, J=7.2Hz), 3.08(s, 3H), 2.35(s, 3H), 1.43(s, 9H), 1.25(t, 3H, J=7.2Hz)
353	8.48, 8.34(s, 1H), 7.19(d, 1H, J=8.7Hz), 7.07(s, 1H), 7.04(d, 1H, J=2.7Hz), 6.49(dd, 1H, J=3.0, 8.7Hz), 3.44, 3.61(q, 2H, J=6.9, 6.0Hz), 3.10(s, 3H), 2.36(s, 3H), 2.26(s, 3H), 1.44(s, 9H), 1.25(t, 3H, J=6.3Hz)
358	8.44, 8.31(s, 1H), 8.06(d, 1H, J=2.7Hz), 7.26(dd, 1H, J=2.7, 8.7Hz), 7.21(d, 1H, J=8.4Hz), 7.09(dd, 1H, J=0.9, 7.8Hz), 7.03(dd, 1H, J=1.8, 2.4Hz), 6.94(d, 1H, J=8.7Hz), 6.73(dd, 1H, J=2.4, 8.1Hz), 3.38, 3.58(q, 2H, J=7.2Hz), 3.07(s, 3H), 1.28(s, 9H), 1.24(t, 3H, J=7.2Hz)

Table 2 continued

No.	¹ H-NMR δppm (Solvent : CDCl ₃ / 300MHz)
359	8.40(s, 1H), 7.90(d, 1H, J=3.0Hz), 7.21(dd, 1H, J=7.8, 8.4Hz), 7.15(d, 1H, J=3.0Hz), 7.05(dd, 1H, J=0.9, 7.8Hz), 7.04(dd, 1H, J=1.5, 2.4Hz), 6.71(ddd, 1H, J=2.4, 2.4, 8.1Hz), 3.38, 3.60(q, 2H, J=7.2Hz), 3.08(s, 3H), 2.29(s, 3H), 1.29(s, 9H), 1.23(t, 3H, J=7.2Hz)
361	8.37, 8.23(s, 1H), 7.18(dd, 1H, J=7.8, 8.1Hz), 7.05(s, 1H), 7.04(m, 1H), 6.97(dd, 1H, J=1.8, 2.4Hz), 6.58(ddd, 1H, J=2.4, 2.4, 8.1Hz), 3.38, 3.58(q, 2H, J=7.2Hz), 3.06(s, 3H), 2.32(s, 3H), 2.24(s, 3H), 1.29(s, 9H), 1.26(m, 3H)
364	7.64(d, 1H, J=2.4Hz), 7.52(brs, 1H), 7.40(dd, 1H, J=1.8, 8.4Hz), 7.22(d, 1H, J=2.7Hz), 7.12(ddd, 1H, J=1.8, 7.5, 7.8Hz), 7.05(ddd, 1H, J=1.5, 7.2, 7.8Hz), 6.81(dd, 1H, J=1.5, 8.1Hz), 3.33(m, 2H), 2.99(s, 3H), 2.32(s, 3H), 1.41(s, 9H), 1.20(t, 3H, J=7.2Hz)
365	7.43(s, 1H), 7.37(dd, 1H, J=1.8, 7.5Hz), 7.08(ddd, 1H, J=1.5, 7.2, 7.8Hz), 6.98(ddd, 1H, J=1.5, 7.2, 7.8Hz), 6.97(s, 1H), 6.69(dd, 1H, J=1.5, 7.8Hz), 3.36(m, 2H), 3.00(s, 3H), 2.34(s, 3H), 2.21(s, 3H), 1.46(s, 9H), 1.22(t, 3H, J=7.2Hz)
366	7.65(d, 1H, J=2.7Hz), 7.51(brs, 1H), 7.35(dd, 1H, J=2.1, 8.7Hz), 7.34(dd, 1H, J=2.1, 8.7Hz), 7.21(d, 1H, J=2.4Hz), 6.97(dd, 1H, J=2.1, 8.7Hz), 6.96(dd, 1H, J=2.1, 8.7Hz), 3.32(m, 2H), 3.00(s, 3H), 2.27(s, 3H), 1.31(s, 9H), 1.21(t, 3H, J=7.2Hz)
367	7.43(s, 1H), 7.28(d, 2H, J=9.0Hz), 6.96(s, 1H), 6.87(d, 2H, J=8.7Hz), 3.36(m, 2H), 3.01(s, 3H), 2.37(s, 3H), 2.18(s, 3H), 1.29(s, 9H), 1.22(t, 3H, J=7.2Hz)
373	7.68(d, 1H, J=2.4Hz), 7.53(brs, 1H), 7.19-7.25(m, 2H), 6.64(ddd, 1H, J=2.1, 2.4, 8.1Hz), 6.56-6.60(m, 2H), 3.77(s, 3H), 3.32(m, 2H), 3.00(s, 3H), 2.56(s, 3H), 1.21(t, 3H, J=7.2Hz)
374	7.43(brs, 1H), 7.15(dd, 1H, J=8.1, 8.1Hz), 6.97(s, 1H), 6.57(ddd, 1H, J=2.4, 2.4, 8.4Hz), 6.47-6.52(m, 2H), 3.75(s, 3H), 3.34(s, 3H), 3.01(s, 3H), 2.38(s, 3H), 2.16(s, 3H), 1.22(t, 3H, J=7.2Hz)
382	7.74(s, 1H), 7.64(d, 1H, J=7.5Hz), 7.56(d, 1H, J=7.8Hz), 7.49(d, 1H, J=7.5Hz), 7.45, 7.38(s, 1H), 7.06(s, 1H), 5.40(s, 2H), 3.31, 3.53(m, 2H), 3.04(s, 3H), 2.18(s, 3H), 1.23(t, 3H, J=7.2Hz)

Table 2 continued

No.	¹ H-NMR δppm (Solvent : CDCl ₃ / 300MHz)
385	7.36(m, 4H), 7.12(d, 1H, J=6.9Hz), 6.88(s, 1H), 5.39(s, 2H), 3.35(m, 2H), 2.99(s, 3H), 2.38(s, 3H), 2.18(s, 3H), 1.26(t, 3H, J=7.2Hz)
393	7.74(d, 1H, J=6.9Hz), 7.63(m, 1H), 7.61(s, 1H), 7.16(m, 1H), 6.94(s, 1H), 5.37(s, 2H), 3.39(brs, 2H), 3.06(s, 3H), 2.39(s, 3H), 2.15(s, 3H), 1.23(t, 3H, J=7.2Hz)
400	8.97(s, 1H), 8.74(d, 1H, J=5.1Hz), 7.55(d, 1H, J=5.1Hz), 7.53, 7.40(s, 1H), 7.20(s, 1H), 5.55(s, 2H), 3.36, 3.60(m, 2H), 3.14, 3.04(s, 3H), 2.16(s, 3H), 1.25(t, 3H, J=7.2Hz)
401	8.99(s, 1H), 8.70(d, 1H, J=5.1Hz), 7.53(d, 1H, J=5.1Hz), 7.38(s, 1H), 6.96(s, 1H), 5.60(s, 2H), 3.41(m, 2H), 3.08(s, 3H), 2.37(s, 3H), 2.16(s, 3H), 1.23(t, 3H, J=7.2Hz)
408	7.86(dd, 1H, J=7.8, 8.1Hz), 7.66(d, 1H, J=7.8Hz), 7.58(d, 1H, J=7.8Hz), 7.52, 7.40(s, 1H), 7.16(s, 1H), 5.55(s, 2H), 3.34, 3.57(m, 2H), 3.09, 3.01(s, 3H), 2.22(s, 3H), 1.24(t, 3H, J=7.2Hz)
409	7.83(dd, 1H, J=7.8, 8.1Hz), 7.66(d, 1H, J=7.8Hz), 7.55(d, 1H, J=7.8Hz), 7.37(s, 1H), 6.90(s, 1H), 5.58(s, 2H), 3.35(m, 2H), 2.99(s, 3H), 2.32(s, 3H), 2.22(s, 3H), 1.20(t, 3H, J=7.2Hz)
416	8.50, 8.37(s, 1H), 7.38(d, 1H, J=9.0Hz), 7.21(d, 1H, J=2.7Hz), 7.04(s, 1H), 6.92(dd, 1H, J=2.7, 9.0Hz), 3.44, 3.63(q, m, 2H, J=7.2Hz), 3.13, 3.11(s, 3H), 2.65(q, 2H, J=7.2Hz), 2.29, 2.27(s, 3H), 1.28(t, 3H, J=7.2Hz), 1.19(t, 3H, J=7.5Hz)

Test examples of the composition of the invention are described below. In each test, a control index was according to the following standard.

[Control index]

[Degree of disease: visual observation]

- | | | |
|----|----|---|
| 5 | 5: | Lesion or spore formation is not observed at all. |
| | 4: | Lesion area, the number of lesion or spore formation area is less than 10% of non-treatment area. |
| 10 | 3: | Lesion area, the number of lesion or spore formation area is less than 40% of non-treatment area. |
| | 2: | Lesion area, the number of lesion or spore formation area is less than 70% of non-treatment area. |
| | 1: | Lesion area, the number of lesion or spore formation area is greater or equal to 70% of non-treatment area. |

15

Test Example 1: Test on preventative effects against wheat powdery mildew

Wheat (breed: Norin 61) was cultivated in a plastic pot having a diameter of 7.5 cm, and when reached 1.5 leaf stage, 10 ml of a chemical in which the pyridylamidine compound of the formula (I) or a salt thereof had been adjusted to a predetermined concentration was sprayed with a spray gun. After drying the chemical (the day of treatment), conidiospores of mildew (*Erysiphe graminis*) were spread inoculated, and held in a temperature-controlled room at 20°C. Six to seven days after the inoculation, spore formation area was investigated, and a control index was obtained according to the evaluation standard described above. Test was carried out for the above-mentioned compound Nos. 1, 2, 3, 4, 5, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 19, 20, 21, 25, 26, 27, 34, 44, 45, 47, 49, 71, 72, 81, 82, 84, 85, 87, 88, 210, 211, 212, 228, 230, 234, 236, 245, 247, 253, 312, 314, 318, 319, 320, 321, 323, 324, 325, 326, 329, 330, 338, 340, 341, 342, 344, 348, 349, 350, 352, 353, 358, 359, 361, 364, 365, 366, 367, 374, 382, 385, 393, 400, 401, 409 and 416, whereby all compounds show a control index of at least 4 at 200 ppm.

Test Example 2: Test on preventative effects against wheat glume blotch

Wheat (breed: Norin 61) was cultivated in a plastic pot having a diameter of 7.5 cm, and when reached 1.5 leaf stage, 10 ml of a chemical in which the pyridylamidine compound of the formula (I) or a salt thereof had been adjusted to a predetermined concentration was sprayed with a spray gun. After drying the chemical (the day of treatment), conidiospores suspension of *Septoria nodorum* were spread by jet inoculation, held in the inoculation chamber at 20°C for 72 hours and then held in a temperature-controlled room at 20°C. Six to seven days after the inoculation, the number of lesion was investigated, and a control index was obtained according to the evaluation standard described above. Test was carried out for the above-mentioned compound Nos. 1, 5, 7, 8, 9, 12, 15, 16, 17, 18, 20, 21, 24, 25, 26, 27, 44, 45, 46, 47, 49, 71, 74, 82, 84, 87, 88, 212, 228, 234, 236, 247, 253, 312, 314, 318, 319, 320, 324, 325, 326, 329, 330, 335, 336, 340, 343, 344, 348, 350, 352, 353, 358, 359, 374, 382, 385, 393, 400, 409, and 416, whereby all compounds show a control index of at least 4 at 200 ppm.

Test Example 3: Test on preventative effects against brown rust

Wheat (breed: Norin 61) was cultivated in a plastic pot having a diameter of 7.5 cm, and when reached 1.5 leaf stage, 10 ml of a chemical in which the pyridylamidine compound of the formula (I) or a salt thereof had been adjusted to a predetermined concentration was sprayed with a spray gun. After drying the chemical

(the day of treatment), uredospores of *Puccinia recondita* were spread inoculated and held in a temperature-controlled room at 20°C. Six to seven days after the inoculation, spore formation area was investigated, and a control index was obtained according to the evaluation standard described above. Test was carried out for the above-mentioned
5 compound Nos. 1, 3, 7, 10, 11, 13, 15, 19, 20, 25, 82, 87, 211, 212, 230, 236, 245, 247, 253, 314, 318, 320, 324, 325, 326, 329, 330, 336, 338, 341, 342, 344, 346, 348, 350, 352, 353, 358, 359, 361, 364, 365, 367, 374, 382, 385, 393, 400, 401, 408, 409, 416, whereby all compounds show a control index of at least 4 at 200 ppm.

10 Test Example 4: Test on preventative effects against tomato late blight

Tomato (breed: Yellow Pear) was cultivated in a plastic pot having a diameter of 7.5 cm, and when reached 3 leaf stage, 10 ml of a chemical in which the pyridylamidine compound of the formula (I) or a salt thereof had been adjusted to a predetermined concentration was sprayed with a spray gun. After drying the chemical
15 (the following day of treatment), zoospore suspension of *Pythophthora infestans* were spread by jet inoculation, held in the inoculation chamber at 20°C for 18 hours and then held in a temperature-controlled room at 20°C. Three days after the inoculation, lesion area was investigated, and a control index was obtained according to the evaluation standard described above. Test was carried out for the above-mentioned
20 compound Nos. 1, 2, 12, 17, 34, 47, 81, 84, 234, 319, 343 and 347, whereby all compounds show a control index of at least 4 at 400 ppm.

Test Example 5: Test on preventative effects against rice blast

Rice (breed: Nihonbare) was cultivated in a plastic pot having a diameter of 7.5
25 cm, and when reached 2 leaf stage, 10 ml of a chemical in which the pyridylamidine compound of the formula (I) or a salt thereof had been adjusted to a predetermined concentration was sprayed with a spray gun. After drying the chemical (the day of treatment), conidiospores suspension of *Pyricularia oryzae* were spread by jet inoculation, held in the inoculation chamber at 20°C for 72 hours and then held in a
30 temperature-controlled room at 20°C. Six to seven days after the inoculation, the number of lesion was investigated, and a control index was obtained according to the evaluation standard described above. Test was carried out for the above-mentioned compound Nos. 1, 2, 3, 7, 8, 9, 11, 12, 15, 19, 20, 44, 47, 71, 72, 82, 84, 85, 87, 88, 91, 212, 228, 230, 245, 247, 253, 312, 314, 318, 319, 320, 324, 325, 326, 330, 335, 341,
35 344, 347, 349, 352, 353, 358, 361, 364, 365, 366, 367, 393 and 400, whereby all compounds show a control index of at least 4 at 200 ppm.

Test Example 6: Test on preventative effects against cucumber powdery mildew

Cucumber (breed: Sagamihanziro) was cultivated in a plastic pot having a diameter of 7.5 cm, and when reached 1.5 leaf stage, 10 ml of a chemical in which the pyridylamidine compound of the formula (I) or a salt thereof had been adjusted to a predetermined concentration was sprayed with a spray gun. After drying the chemical (the day of treatment), conidiospores suspension of *Sphaerotheca fuliginia* were spread by jet inoculation and held in a temperature-controlled room at 20°C. Seven to ten days after the inoculation, spore formation area was investigated, and a control index was obtained according to the evaluation standard described above. Test was carried out for the above-mentioned compound Nos. 1, 2, 7, 10, 11, 12, 13, 15, 17, 18, 19, 20, 21, 44, 47, 81, 82, 84, 87, 211, 212, 230, 236, 243, 245, 247, 253, 313, 314, 318, 319, 320, 324, 325, 326, 329, 330, 336, 338, 341, 342, 344, 346, 347, 348, 349, 350, 352, 353, 361, 365, 367, 374, 382, 385, 393, 400, 401, 408, 409 and 416, whereby all compounds show a control index of at least 4 at 200 ppm.

Formulation Example 1

(1) Compound of the present invention	20 parts by weight
(2) Clay	72 parts by weight
(3) Sodium lignin sulfonate	8 parts by weight

The above components are uniformly mixed to obtain a wettable powder.

Formulation Example 2

(1) Compound of the present invention	5 parts by weight
(2) Talc	95 parts by weight

The above components are uniformly mixed to obtain a dust.

Formulation Example 3

(1) Compound of the present invention	20 parts by weight
(2) N,N'-dimethylacetamide	20 parts by weight
(3) Polyoxyethylene alkyl phenyl ether	10 parts by weight
(4) Xylene	50 parts by weight

The above components are uniformly mixed and dissolved to obtain an emulsifiable concentrate.

Formulation Example 4

(1) Clay	68 parts by weight
(2) Sodium lignin sulfonate	2 parts by weight

- | | |
|--|--------------------|
| (3) Polyoxyethylene alkyl aryl ether sulfate | 5 parts by weight |
| (4) Fine silica | 25 parts by weight |

A mixture of the above components and the compound of the present invention are mixed in a weight ratio of 4:1 to obtain a wettable powder.

5

Formulation Example 5

- | | |
|--|----------------------|
| (1) Compound of the present invention | 50 parts by weight |
| (2) Oxylated polyalkylphenyl phosphate-triethanolamine | 2 parts by weight |
| (3) Silicone | 0.2 part by weight |
| 10 (4) Water | 47.8 parts by weight |

The above components are uniformly mixed and pulverized to obtain a stock, solution, and

- | | |
|------------------------------|----------------------|
| (5) Sodium polycarboxylate | 5 parts by weight |
| (6) Anhydrous sodium sulfate | 42.8 parts by weight |
- 15 are further added thereto, followed by uniform mixing, granulation and drying to obtain a water-dispersible granules.

Formulation Example 6

- | | |
|--|----------------------|
| (1) Compound of the present invention | 5 parts by weight |
| 20 (2) Polyoxyethylene octylphenyl ether | 1 part by weight |
| (3) Phosphate of polyoxyethylene | 0.1 part by weight |
| (4) Particulate calcium carbonate | 93.9 parts by weight |

The above components (1) to (3) are preliminarily mixed uniformly and diluted with a proper amount of acetone, the diluted mixture is sprayed on the component (4),
25 and acetone is removed to obtain granules.

Formulation Example 7

- | | |
|---------------------------------------|----------------------|
| (1) Compound of the present invention | 2.5 parts by weight |
| (2) N-methyl-2-pyrrolidone | 2.5 parts by weight |
| 30 (3) Soybean oil | 95.0 parts by weight |

The above components are uniformly mixed and dissolved to obtain an ultra low volume formulation.

Formulation Example 8

- | | |
|--|--------------------|
| 35 (1) Compound of the present invention | 20 parts by weight |
| (2) Oxylated polyalkylphenyl phosphate triethanolamine | 2 parts by weight |
| (3) Silicone | 0.2 part by weight |

- | | |
|---------------------|----------------------|
| (4) Xanthan gum | 0.1 part by weight |
| (5) Ethylene glycol | 5 parts by weight |
| (6) Water | 72.7 parts by weight |

5 The above components are uniformly mixed and pulverized to obtain a water-based suspension concentrate.

 While the invention has been described in detail and with reference to specific embodiments thereof, it will be apparent to one skill in the art that various changes and modifications can be made therein without departing from the spirit and scope thereof.

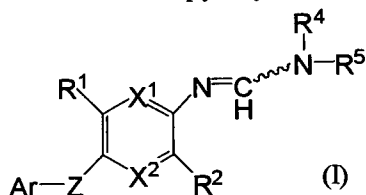
10 This application is based on Japanese patent application No. 2008-004575 filed on January 11, 2008, the entire contents of which are incorporated herein by reference. All references cited herein are incorporated in their entirety.

Industrial Applicability

15 The pyridylamidine compound of the formula (I) or a salt thereof shows excellent effect as an active ingredient of an agricultural or horticultural fungicide composition.

CLAIMS

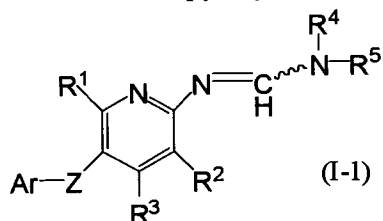
1. A pyridylamidine compound represented by the formula (I):



5 wherein one of X^1 and X^2 represents N, and the other represents CR^3 ; Ar represents phenyl which may be substituted with R^6 , 3,4-methylenedioxyphenyl, naphthyl, 5,6,7,8-tetrahydro-2-naphthyl, or pyridyl which may be substituted with R^6 ; Z represents a bond, O or OCH_2 ; R^1 , R^2 , R^3 and R^6 each independently represents a hydrogen atom, C_{1-6} alkyl, halo(C_{1-6} alkyl), halogen, C_{1-6} alkoxy, halo(C_{1-6} alkoxy), cyano, nitro, (C_{1-6} alkyl)carbonyl, (C_{1-6} alkoxy)carbonyl, or amino which may be substituted with C_{1-6} alkyl; R^4 and R^5 each independently represents C_{1-6} alkyl, provided that (1) a compound wherein X^1 is CH, X^2 is N, and both R^4 and R^5 are methyl, and (2) 6-chloro-3-cyano-2-dimethylamino-methyleneamino-5-(pyrid-4-yl)pyridine are excluded; or a salt thereof.

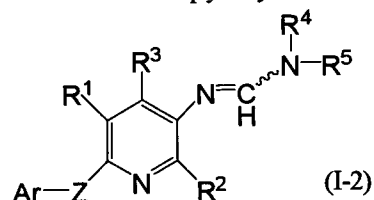
15 2. The pyridylamidine compound according to claim 1, wherein in the formula (I), (1) X^1 is N, X^2 is CR^3 , and Z is a bond, O or OCH_2 , or (2) X^1 is CH, X^2 is N, and Z is a single bond, or a salt thereof.

3. A pyridylamidine compound represented by the formula (I-1):



20 wherein Ar represents phenyl which may be substituted with R^6 , 3,4-methylenedioxyphenyl, naphthyl, 5,6,7,8-tetrahydro-2-naphthyl, or pyridyl which may be substituted with R^6 ; Z represents a bond, O or OCH_2 ; R^1 , R^2 , R^3 and R^6 each independently represents a hydrogen atom, C_{1-6} alkyl, halo(C_{1-6} alkyl), halogen, C_{1-6} alkoxy, halo(C_{1-6} alkoxy), cyano, nitro, (C_{1-6} alkyl)carbonyl, (C_{1-6} alkoxy)carbonyl, or amino which may be substituted with C_{1-6} alkyl; R^4 and R^5 each independently represents C_{1-6} alkyl, provided that 6-chloro-3-cyano-2-dimethylamino-methyleneamino-5-(pyrid-4-yl)pyridine is excluded; or a salt thereof.

4. A pyridylamidine compound represented by the formula (I-2):



wherein Ar represents phenyl which may be substituted with R⁶, 3,4-methylenedioxyphenyl, naphthyl, 5,6,7,8-tetrahydro-2-naphthyl, or pyridyl which may be substituted with R⁶; Z represents a bond, O or OCH₂; R¹, R², R³ and R⁶ each independently represents a hydrogen atom, C₁₋₆ alkyl, halo(C₁₋₆ alkyl), halogen, C₁₋₆ alkoxy, halo(C₁₋₆ alkoxy), cyano, nitro, (C₁₋₆ alkyl)carbonyl, (C₁₋₆ alkoxy)carbonyl, or amino which may be substituted with C₁₋₆ alkyl; R⁴ and R⁵ each independently represents C₁₋₆ alkyl, provided that the compound wherein both R⁴ and R⁵ are methyl is excluded; or a salt thereof.

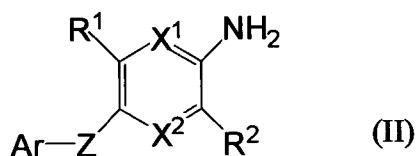
5. The pyridylamidine compound according to claim 4, wherein Z is a bond, or a salt thereof.

6. An agricultural or horticultural fungicide composition comprising the pyridylamidine compound represented by the formula (I) according to claim 1 or a salt thereof as an active ingredient.

7. A control method of plant diseases, comprising applying the pyridylamidine compound represented by the formula (I) according to claim 1 or a salt thereof to an agricultural or horticultural plant.

8. A method for producing the pyridylamidine compound represented by the formula (I) according to claim 1 or a salt thereof, comprising:

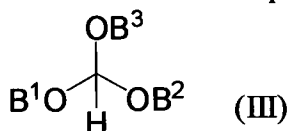
reacting a compound represented by the formula (II):



wherein one of X¹ and X² represents N, and the other represents CR³; Ar represents phenyl which may be substituted with R⁶, 3,4-methylenedioxyphenyl, naphthyl, 5,6,7,8-tetrahydro-2-naphthyl, or pyridyl which may be substituted with R⁶; Z represents a bond, O or OCH₂; R¹, R², R³ and R⁶ each independently represents a hydrogen atom,

C₁₋₆ alkyl, halo(C₁₋₆ alkyl), halogen, C₁₋₆ alkoxy, halo(C₁₋₆ alkoxy), cyano, nitro, (C₁₋₆ alkyl)carbonyl, (C₁₋₆ alkoxy)carbonyl, or amino which may be substituted with C₁₋₆ alkyl;

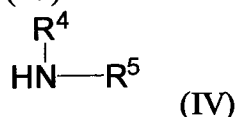
with ortho triester represented by the formula (III):



wherein B¹, B² and B³ each independently represents alkyl or cycloalkyl, to obtain a reaction mixture; and

reacting the reaction mixture with a compound represented by the formula

(IV):



wherein R⁴ and R⁵ each independently represents a hydrogen atom, C₁₋₆ alkyl, halo(C₁₋₆ alkyl), halogen, C₁₋₆ alkoxy, halo(C₁₋₆ alkoxy), cyano, nitro, (C₁₋₆ alkyl)carbonyl, (C₁₋₆ alkoxy)carbonyl, or amino which may be substituted with C₁₋₆ alkyl.

9. The production method according to claim 8, wherein the compound represented by the formula (II) and the ortho triester represented by the formula (III) are allowed to react in the presence of an acid or a base.

10. The production method according to claim 8, wherein the reaction mixture obtained by reacting the compound represented by the formula (II) with the ortho triester represented by the formula (III), and the compound represented by the formula (IV) are allowed to react in the presence of an inert solvent.

INTERNATIONAL SEARCH REPORT

International application No

PCT/JP2009/050602

A. CLASSIFICATION OF SUBJECT MATTER

INV. A01N43/40 C07D405/02 C07D213/72

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)

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)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 0 314 429 A (ICI AMERICA INC [US]) 3 May 1989 (1989-05-03) the whole document	1-10
X	WO 2007/039439 A (HOFFMANN LA ROCHE [CH]; GATTI MCARTHUR SILVIA [CH]; GOETSCHI ERWIN [CH] 12 April 2007 (2007-04-12) cited in the application examples C32-C36	1-3,6
Y	examples C32-C36	8-10
Y	WO 2007/031523 A (BAYER CROPS SCIENCE AG [DE]; LUEMMEN PETER [DE]; KUNZ KLAUS [DE]; GREUL) 22 March 2007 (2007-03-22) scheme1 page 4	8-10
A	the whole document	1-7

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☒ Further documents are listed in the continuation of Box C.☒ See patent family annex.

* Special categories of cited documents :

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- "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

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"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

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"&" document member of the same patent family

Date of the actual completion of the international search

2 April 2009

Date of mailing of the international search report

08/04/2009

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INTERNATIONAL SEARCH REPORT

International application No
PCT/JP2009/050602

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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A	DD 279 016 A1 (AKAD WISSENSCHAFTEN DDR [DD]) 23 May 1990 (1990-05-23) cited in the application the whole document -----	1-3,8-10
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Information on patent family members

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