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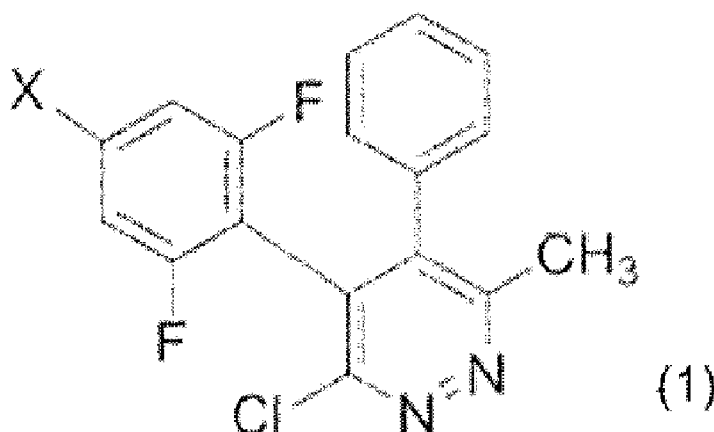
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(54) Title: SYNERGISTIC FUNGICIDAL COMPOSITION FOR CONTROLLING PLANT DISEASES



(57) Abstract: The present invention provides a composition for controlling a plant disease comprising a pyridazine compound represented by formula (1) wherein X represents a fluoro atom or a hydrogen atom, and at least one amidine compound selected from the following group (A), and a method for controlling a plant disease, comprising a step of applying each effective amount of the above-mentioned pyridazine compound represented by the formula (1) and the at least one amidine compound selected from the following group (A) to a plant or soil for cultivating the plant, Group (A): a group consisting of N'-{4-[(4,5-dichlorothiazol-2-yl)oxy]-2,5-dimethylphenyl}-N-ethyl-N-methylmethane imidamide and N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane imidamide, the composition and method having excellent control efficacies against plant diseases.

## Description

### Title of Invention:

### SYNERGISTIC FUNGICIDAL COMPOSITION FOR CONTROLLING PLANT DISEASES

#### Technical Field

[0001] This application claims priority to and the benefit of Japanese Patent Application No. 2016-048178 filed on March 11, 2016, the entire contents of which are incorporated herein by reference.

The present invention relates to a composition for controlling plant diseases and a method for controlling plant diseases.

#### Background Art

[0002] Hitherto, some compounds have been known as an active ingredient for a composition for controlling plant diseases (see Patent Literatures 1, 2 and 3).

#### Citation List

##### Patent Literature

- [0003] [PTL 1] WO 2012/020772 pamphlet  
[PTL 2] WO 2007/031513 pamphlet  
[PTL 3] WO 2008/110313 pamphlet

#### Summary of Invention

##### Technical Problem

[0004] An object of the present invention is to provide a composition for controlling plant diseases and a method for controlling plant diseases, each having an excellent control efficacy on plant diseases.

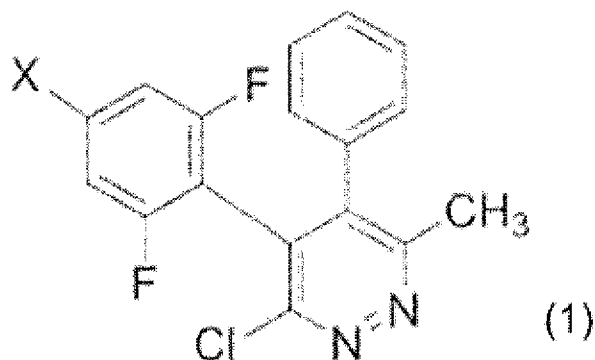
##### Solution to Problem

[0005] The present inventors have intensively studied to find out a composition for controlling plant diseases and a method for controlling plant diseases, each having an excellent control efficacy on plant diseases. As a result, they have found out that a composition comprising a pyridazine compound represented by the below-mentioned formula (1) and at least one amidine compound selected from the below-mentioned group (A) shows an excellent control efficacy on plant diseases.

That is, the present invention provides the followings:

- [1] A composition for controlling a plant disease comprising a pyridazine compound represented by formula (1):

[Chem.1]



wherein X represents a fluoro atom or a hydrogen atom, and  
at least one amidine compound selected from the following group (A),

Group (A): a group consisting of

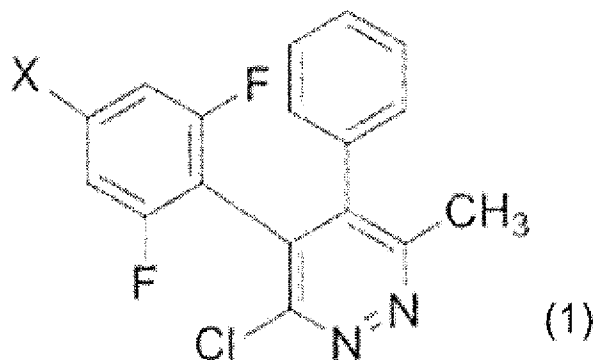
N'-{4-[(4,5-dichlorothiazol-2-yl)oxy]-2,5-dimethylphenyl}-N-ethyl-N-methylmethane  
imidamide and N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane  
imidamide.

[2] The composition for controlling a plant disease described in [1] wherein the  
pyridazine compound is the compound represented by formula (1) wherein X  
represents a hydrogen atom.

[3] The composition for controlling a plant disease described in [1] or [2] wherein a  
weight ratio of the pyridazine compound to the amidine compound is within the range  
of 1:0.0125 to 1:500.

[4] A method for controlling a plant disease, comprising a step of applying each  
effective amount of a pyridazine compound represented by formula (1):

[Chem.2]



wherein X represents a fluoro atom or a hydrogen atom, and  
at least one amidine compound selected from the following group (A),

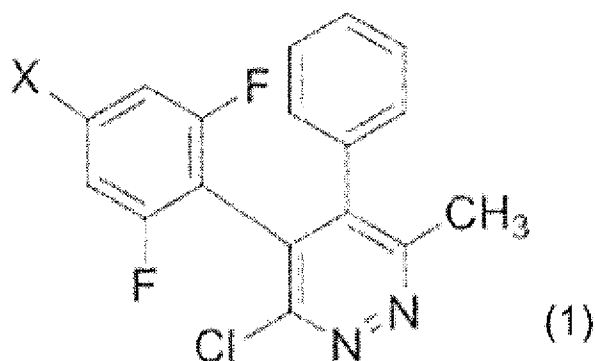
Group (A): a group consisting of

N'-{4-[(4,5-dichlorothiazol-2-yl)oxy]-2,5-dimethylphenyl}-N-ethyl-N-methylmethane imidamide and N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane imidamide,

to a plant or soil for cultivating the plant.

[5] A method for controlling a plant disease, comprising a step of applying each effective amount of a pyridazine compound represented by formula (1):

[Chem.3]



wherein X represents a fluoro atom or a hydrogen atom, and

at least one amidine compound selected from the following group (A),

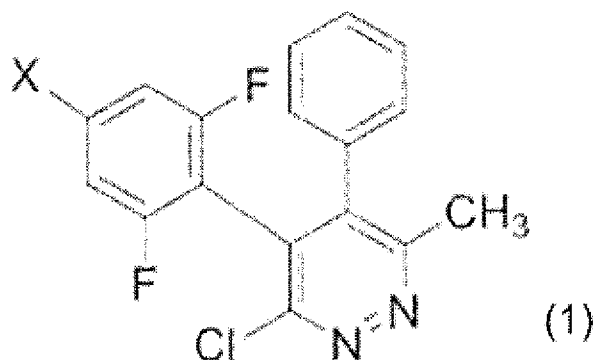
Group (A): a group consisting of

N'-{4-[(4,5-dichlorothiazol-2-yl)oxy]-2,5-dimethylphenyl}-N-ethyl-N-methylmethane imidamide and N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane imidamide,

to a seed.

[6] A combined use of a pyridazine compound represented by formula (1):

[Chem.4]



wherein X represents a fluoro atom or a hydrogen atom, and

at least one amidine compound selected from the following group (A),

Group (A): a group consisting of

N'-{4-[(4,5-dichlorothiazol-2-yl)oxy]-2,5-dimethylphenyl}-N-ethyl-N-methylmethane imidamide and N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane imidamide.

[0006] The present invention can control plant diseases.

### Description of Embodiments

[0007] The composition for controlling plant diseases of the present invention (hereinafter, referred to as "present composition") comprises the above-mentioned pyridazine compound represented by formula (1) (hereinafter, referred to as "present compound 1") and the above-mentioned amidine compound (hereinafter, referred to as "present compound A").

[0008] First, the present compound 1 is described.

[0009] The present compound 1 is a compound described in, for example, WO 2012/020772 pamphlet, and can be prepared according to the process described therein.

[0010] Examples of the present compound 1 include the compounds shown in Table 1 below.

[0011] [Table 1]

| Present compound 1   | Structural Formula |
|----------------------|--------------------|
| Present compound 1-1 |                    |
| Present compound 1-2 |                    |

[0012] Next, the present compound A is described.

[0013] Examples of the present compound A include the following compounds:

N'-{4-[(4,5-dichlorothiazol-2-yl)oxy]-2,5-dimethylphenyl}-N-ethyl-N-methylmethane imidamide (hereinafter, referred to as "present compound A-1"); and

N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane imidamide (hereinafter, referred to as "present compound A-2").

[0014] The present compound A-1 is a compound described in, for example, WO

2007/031513 pamphlet. The present compound A-2 is a compound described in, for example, WO 2008/110313 pamphlet. The present compound A-1 and the present compound A-2 can be prepared according to the process described therein.

[0015] The weight ratio of the present compound 1 to the present compound A is within a range of usually 1:0.0125 to 1:500, preferably 1:0.025 to 1:100, and more preferably 1:0.1 to 1:10.

[0016] Although the present composition may be a mixture as itself of the present compound 1 and the present compound A, the present composition is usually prepared by mixing the present compound 1, the present compound A and an inert carrier, and if necessary, adding a surfactant or other auxiliary agents for formulation, and then formulating into the form of oil solutions, emulsifiable concentrates, flowables, wettable powders, granulated wettable powders, dusts, granules and the others. Such formulations may be used by itself or with an addition of other inert components as an agent for controlling plant diseases.

The present composition may comprises usually 0.1 to 99 % by weight, preferably 0.2 to 90 % by weight, and more preferably 1 to 80 % by weight of the present compound 1 and the present compound A in total.

[0017] Examples of an inert carrier used on formulating include a solid carrier and a liquid carrier, and examples of the solid carrier include finely-divided powders or particles consisting of minerals (for example, kaolin clay, attapulgite clay, bentonite, montmorillonite, acid clay, pyrophyllite, talc, diatomaceous earth, or calcite), natural organic substances (for example, corncob powder, or walnut shell powder), synthetic organic substances (for example, urea), salts (for example, calcium carbonate, or ammonium sulfate), synthetic inorganic substances (for example, synthetic hydrous silicon oxide) and so on. Also, examples of the liquid carrier include aromatic hydrocarbons (for example, xylene, alkyl benzene, or methylnaphthalene), alcohols (for example, 2-propanol, ethylene glycol, propylene glycol, or ethylene glycol monoethyl ether), ketones (for example, acetone, cyclohexanone, or isophorone), vegetable oils (for example, soybean oil, or cotton oils), petroleum-derived aliphatic hydrocarbons, esters, dimethyl sulfoxide, acetonitrile and water.

Examples of the surfactant include anionic surfactant (for example, alkyl sulfate salt, alkylaryl sulfonate salt, dialkyl sulfosuccinate salt, polyoxyethylene alkylaryl ether phosphates, lignin sulfonate, or naphthalene sulfonate formaldehyde polycondensation), nonionic surfactant (for example, polyoxyethylene alkylaryl ether, polyoxyethylene alkyl polyoxypropylene block copolymer, or sorbitan fatty acid ester) and cationic surfactant (for example, alkyltrimethyl ammonium salt).

Examples of the other auxiliary agents for formulation include water-soluble polymer (for example, polyvinyl alcohol, or polyvinyl pyrrolidone), polysaccharides (for

example, arabic gum, alginic acid and salts thereof, CMC (carboxymethyl-cellulose), or xanthan gum), inorganic substances (for example, aluminum magnesium silicate, or alumina-sol), antiseptic agent, coloring agent, and PAP (isopropyl acid phosphate), and stabilizing agent (for example, BHT (2,6-di-tert-butyl-4-methylphenol)).

[0018] The present composition may also be prepared by separately formulating the present compound 1 and the present compound A into different formulations respectively according to the above-mentioned processes, if necessary, further diluting them with water, thereafter, mixing the separately prepared different formulations or the resultant dilute solutions thereof with each other.

[0019] The present composition may further comprise one or more other fungicide(s) and/or insecticide(s).

[0020] The present composition can be applied to a plant or soil for cultivating the plant to control the plant diseases.

[0021] Examples of the plant diseases which can be controlled by the present invention include the following diseases, but are not limited thereto.

[0022] Rice diseases: blast (*Magnaporthe grisea*), brown spot (*Cochliobolus miyabeanus*), sheath blight (*Rhizoctonia solani*), and bakanae disease (*Gibberella fujikuroi*);

Wheat diseases: powdery mildew (*Erysiphe graminis*), fusarium Head blight (*Fusarium graminearum*, *F. avenaceum*, *F. culmorum*, *Microdochium nivale*), rust (for example, yellow rust (*Puccinia striiformis*), black rust (*P. graminis*), Brown rust (*P. recondita*)), snow mold (*Microdochium nivale*), typhula snow blight (*Typhula* sp.), loose smut (*Ustilago tritici*), stinking smut (*Tilletia caries*), eyespot (*Pseudocercospora herpotrichoides*), Septoria leaf blotch (*Mycosphaerella graminicola*), glume blotch (*Stagonospora nodorum*), and tan spot (*Pyrenophora tritici-repentis*);

Barley diseases: powdery mildew (*Erysiphe graminis*), loose smut (*Fusarium graminearum*, *F. avenaceum*, *F. culmorum*, *Microdochium nivale*), rust (*Puccinia striiformis*, *P. graminis*, *P. hordei*), loose smut (*Ustilago nuda*), scald (*Rhynchosporium secalis*), net blotch (*Pyrenophora teres*), spot blotch (*Cochliobolus sativus*), leaf stripe (*Pyrenophora graminea*), and damping-off caused by rhizoctonia fungus (*Rhizoctonia solani*);

Corn diseases: smut (*Ustilago maydis*), southern leaf blight (*Cochliobolus heterostrophus*), zonate leaf spot (*Gloeocercospora sorghi*), southern rust (*Puccinia polysora*), gray leaf spot (*Cercospora zeae-maydis*), and damping-off caused by rhizoctonia fungus (*Rhizoctonia solani*);

[0023] Citrus diseases: melanose (*Diaporthe citri*), scab (*Elsinoe fawcetti*), fruit rot (*Penicillium digitatum*, *P. italicum*), and Phytophthora disease (*Phytophthora parasitica*, *Phytophthora citrophthora*);

Apple diseases: blossom blight (*Monilinia mali*), canker (*Valsa ceratosperma*), powdery mildew (*Podosphaera leucotricha*), *Alternaria* leaf spot (*Alternaria alternata* apple pathotype), scab (*Venturia inaequalis*), bitter rot (*Colletotrichum acutatum*), and crown rot (*Phytophthora cactorum*);

Pear diseases: scab (*Venturia nashicola*, *V. pirina*), black spot (*Alternaria alternata* Japanese pear pathotype), rust (*Gymnosporangium haraeum*), and *phytophthora* fruit rot, *phytophthora* crown and root rot (*Phytophthora cactorum*) and brown spot (*Stemphilium Vesicarium*);

Peach diseases: brown rot (*Monilinia fructicola*), scab (*Cladosporium carpophilum*), and *Phomopsis* rot (*Phomopsis* sp.);

Grapes diseases: anthracnose (*Elsinoe ampelina*), ripe rot (*Glomerella cingulata*), powdery mildew (*Uncinula necator*), rust (*Phakopsora ampelopsidis*), black rot (*Guignardia bidwellii*), and downy mildew (*Plasmopara viticola*);

Diseases of Japanese persimmon: anthracnose (*Gloeosporium kaki*), and leaf spot (*Cercospora kaki*, *Mycosphaerella nawae*);

Diseases of Cucurbitaceae: anthracnose (*Colletotrichum lagenarium*), powdery mildew (*Sphaerotheca fuliginea*), gummy stem blight (*Mycosphaerella melonis*), *Fusarium* wilt (*Fusarium oxysporum*), downy mildew (*Pseudoperonospora cubensis*), *Phytophthora* rot (*Phytophthora* sp.), and damping-off (*Pythium* sp.);

Tomato diseases: early blight (*Alternaria solani*), leaf mold (*Cladosporium fulvum*), and late blight (*Phytophthora infestans*);

Eggplant disease: brown spot (*Phomopsis vexans*), and powdery mildew (*Erysiphe cichoracearum*);

Diseases of brassica family: *Alternaria* leaf spot (*Alternaria japonica*), white spot (*Cercospora brassicae*), clubroot (*Plasmodiophora brassicae*), and downy mildew (*Peronospora parasitica*);

Welsh onion diseases: rust (*Puccinia allii*), and downy mildew (*Peronospora destructor*);

[0024] Soybean diseases: purple stain (*Cercospora kikuchii*), *Sphaceloma* scab (*Elsinoe glycines*), pod and stem blight (*Diaporthe phaseolorum* var. *sojae*), septoria brown spot (*Septoria glycines*), *Cercospora* leaf spot (*Cercospora sojae*), rust (*Phakopsora pachyrhizi*), *phytophthora* root and stem rot (*Phytophthora sojae*), damping-off caused by *rhizoctonia* fungus (*Rhizoctonia solani*), target spot (*Corynespora asiicola*), and sclerotinia rot (*Sclerotinia sclerotiorum*);

Kidney bean diseases: anthracnose (*Colletotrichum lindemthianum*);

Peanut diseases: leaf spot (*Cercospora personata*), brown leaf spot (*Cercospora arachidicola*), and southern blight (*Sclerotium rolfsii*);

Garden pea diseases: powdery mildew (*Erysiphe pisi*);

Potato diseases: early blight (*Alternaria solani*), late blight (*Phytophthora infestans*), pink rot (*Phytophthora erythroseptica*), and powdery scab (*Spongospora subterranea* f. sp. *subterranea*);

Strawberry diseases: powdery mildew (*Sphaerotheca humuli*), and anthracnose (*Glomerella cingulata*);

Tea diseases: net blister blight (*Exobasidium reticulatum*), white scab (*Elsinoe leucospila*), gray blight (*Pestalotiopsis* sp.), and anthracnose (*Colletotrichum theae-sinensis*);

Tabacco diseases: brown spot (*Alternaria longipes*), powdery mildew (*Erysiphe cichoracearum*), anthracnose (*Colletotrichum tabacum*), downy mildew (*Peronospora tabacina*), and black shank (*Phytophthora nicotianae*);

Rape seed diseases: sclerotinia rot (*Sclerotinia sclerotiorum*), and rape seed damping-off caused by *Rhizoctonia solani* (*Rhizoctonia solani*);

Cotton diseases: cotton damping-off caused by *Rhizoctonia solani* (*Rhizoctonia solani*);

Sugar beet diseases: cercospora leaf spot (*Cercospora beticola*), leaf blight (*Thanatephorus cucumeris*), root rot (*Thanatephorus cucumeris*), and aphanomyces root rot (*Aphanomyces cochlioides*);

Rose diseases: blackspot (*Diplocarpon rosae*), powdery mildew (*Sphaerotheca pannosa*), and downy mildew (*Peronospora sparsa*);

Chrysanthemum and Asteraceae vegetable diseases: downy mildew (*Bremia lactucae*), leaf blight (*Septoria chrysanthemi-indici*), and white rust (*Puccinia horiana*);

Various plants diseases: diseases caused by *Pythium* spp. (*Pythium aphanidermatum*, *Pythium debaryanum*, *Pythium graminicola*, *Pythium irregulare*, *Pythium ultimum*), Gray mold (*Botrytis cinerea*), and Sclerotinia rot (*Sclerotinia sclerotiorum*);

Japanese radish diseases: *Alternaria* leaf spot (*Alternaria brassicicola*);

Turfgrass diseases: dollar spot (*Sclerotinia homeocarpa*), brown patch, and large patch (*Rhizoctonia solani*);

Banana diseases: Sigatoka disease (*Mycosphaerella fijiensis*, *Mycosphaerella musicola*);

Sunflower diseases: downy mildew (*Plasmopara halstedii*);

Seed diseases or diseases in the early stages of the growth of various plants caused by bacteria of *Aspergillus* spp., *Penicillium* spp., *Fusarium* spp., *Gibberella* spp., *Trichoderma* spp., *Thielaviopsis* spp., *Rhizopus* spp., *Mucor* spp., *Corticium* spp., *Phoma* spp., *Rhizoctonia* spp., *Diplodia* spp.; and

Viral diseases of various plants mediated by *Polymixa* spp. or *Olpidium* spp.

[0025] Examples of the plants to which the present composition can be applied include the followings, but are not limited thereto.

[0026] Crops: corn, rice, wheat, barley, rye, oat, sorghum, cotton, soybean, adzuki bean, kidney bean, peanut, buckwheat, beet, rapeseed, sunflower, sugar cane, tobacco, and the others;

Vegetables: solanaceous vegetables (for example, eggplant, tomato, pimento, pepper, or potato), cucurbitaceous vegetables (for example, cucumber, pumpkin, zucchini, water melon, melon, or squash), cruciferous vegetables (for example, Japanese radish, white turnip, horseradish, kohlrabi, Chinese cabbage, cabbage, leaf mustard, broccoli, or cauliflower), asteraceous vegetables (for example, burdock, crown daisy, artichoke, or lettuce), liliaceous vegetables (for example, welsh onion, onion, garlic, or asparagus), ammiaceous vegetables (for example, carrot, parsley, celery, or parsnip), chenopodiaceous vegetables (for example, spinach, or Swiss chard), lamiaceous vegetables (for example, perilla, mint, or basil), strawberry, sweet potato, glutinous yam, eddoe, and the others;

Flowers;

Foliage plants;

Turfgrass;

Fruits: pomaceous fruits (for example, apple, pear, Japanese pear, Chinese quince, or quince), stone fleshy fruits (for example, peach, plum, nectarine, Japanese apricot (*Prunus mume*), cherry fruit, apricot, or prune), citrus fruits (for example, Citrus unshiu, orange, lemon, lime, or grapefruit), nuts (for example, chestnuts, walnuts, hazelnuts, almond, pistachio, cashew nuts, or macadamia nuts), berry fruits (for example, blueberry, cranberry, blackberry, or raspberry), grapes, Japanese persimmon, olive, Japanese plum, banana, coffee, date palm, coconuts, and the others; and

Trees other than fruit trees: tea, mulberry, flowering plants, roadside trees (for example, ash, birch, dogwood, eucalyptus, ginkgo (*Ginkgo biloba*), lilac, maple, oak (*Quercus*), poplar, Judas tree, Formosan gum (*Liquidambar formosana*), plane tree, zelkova, Japanese arborvitae (*Thuja standishii*), fir wood, hemlock, juniper, pinus, picea, or yew (*Taxus cuspidate*)), and the others.

[0027] The above-mentioned "plant(s)" may include plant(s) whose resistance has been imparted by genetic recombination.

[0028] Exemplary embodiments of the present composition include the followings, but are not limited thereto.

[0029] A present composition wherein the combination of the present compound 1 and the present compound A represents as follows:

A combination of the present compound 1-1 and the present compound A-1;

A combination of the present compound 1-1 and the present compound A-2;

A combination of the present compound 1-2 and the present compound A-1; and

A combination of the present compound 1-2 and the present compound A-2.

[0030] A present composition comprising the present compound 1-1 and either of the present compound A-1 or the present compound A-2 wherein the weight ratio of the present compound 1-1 to either of the present compound A-1 or the present compound A-2 is 1:0.0125 to 1:500;

A present composition comprising the present compound 1-1 and either of the present compound A-1 or the present compound A-2 wherein the weight ratio of the present compound 1-1 to either of the present compound A-1 or the present compound A-2 is 1:0.025 to 1:100;

A present composition comprising the present compound 1-1 and either of the present compound A-1 or the present compound A-2 wherein the weight ratio of the present compound 1-1 to either of the present compound A-1 or the present compound A-2 is 1:0.1 to 1:10;

A present composition comprising the present compound 1-2 and either of the present compound A-1 or the present compound A-2 wherein the weight ratio of the present compound 1-2 to either of the present compound A-1 or the present compound A-2 is 1:0.0125 to 1:500;

A present composition comprising the present compound 1-2 and either of the present compound A-1 or the present compound A-2 wherein the weight ratio of the present compound 1-2 to either of the present compound A-1 or the present compound A-2 is 1:0.025 to 1:100; and

A present composition comprising the present compound 1-2 and either of the present compound A-1 or the present compound A-2 wherein the weight ratio of the present compound 1-2 to either of the present compound A-1 or the present compound A-2 is 1:0.1 to 1:10.

[0031] The method for controlling plant diseases of the present invention (hereinafter, referred to as "control method of the present invention") is carried out by applying each of an effective amount of the present compound 1 and the present compound A to a plant or soil for cultivating the plant. Examples of the plant include foliage of a plant, seeds of a plant and bulbs of a plant. Moreover, the bulbs described herein mean discoid stems, corms, rhizomes, tubers, tuberous, and tuberous roots.

[0032] In the control method of the present invention, the present compound 1 and the present compound A may be applied separately to a plant or soil for cultivating the plant in the same period, but are usually applied as the present composition in terms of a convenience on applying.

[0033] In the control method of the present invention, examples of the method of applying the present compound 1 and the present compound A include foliage treatment, soil treatment, root treatment, and seed treatment.

[0034] Such the foliage treatment includes, for example, a method of applying the present

compound 1 and the present compound A onto surface of a plant to be cultivated by a foliar application or a stem application.

Such the soil treatment includes, for example, soil broadcast, soil incorporation, and irrigation of the agent solution comprising the present compound 1 and the present compound A to soil.

Such the root treatment includes, for example, a method of soaking a whole or a root of the plant into a medicinal solution comprising the present compound 1 and the present compound A, and a method of attaching a solid formulation comprising the present compound 1, the present compound A and the solid carrier to a root of the plant.

Such the seed treatment includes, for example, an applying of the present composition to a seed or a bulb of the plant to be prevented from the plant disease, specifically, for example, spray treatment by spraying a suspension of the present composition in a mist form onto the surface of a seed or the surface of a bulb, smear treatment by applying the wettable powders, the emulsifiable concentrates or the flowables of the present composition with added by small amounts of water or as itself to a seed or a bulb, immersion treatment by immersing a seed into a solution of the present composition for a certain period of time, film-coating treatment and pellet-coating treatment.

[0035] Each dose of the present compound 1 and the present compound A in the control method of the present invention may be varied depending on a kind of plant to be applied, a kind or a frequency of an occurrence of a plant disease as a control subject, a dosage form, an application period, an application method, an application site, a climate condition, and the like. In case of an application to a foliage of the plant or soil for cultivating the plant, a total amount of the present compound 1 and the present compound A is within the range of usually 1 to 500 g, preferably 2 to 200 g, and more preferably 10 to 100 g, per 1000 m<sup>2</sup>. Also a total amount of the present compound 1 and the present compound A in the treatment for seed is within the range of usually 0.001 to 10 g, and preferably 0.01 to 1 g, per 1 kg of seeds.

The emulsifiable concentrates, the wettable powders or the flowables, etc., are usually applied by diluting them with water, and then spreading them. In this case, each concentration of the present compound 1 and the present compound A contains usually 0.0005 to 2% by weight, and preferably 0.005 to 1% by weight of the present compound 1 and the present compound A in total. The dusts or the granules, etc., are usually applied as itself without diluting them.

### Examples

[0036] The present invention is described in more detail below by Formulation Examples and Test Examples, but the present invention should not be limited thereto.

[0037] First, Formulation Examples are described. Herein, "parts" means "parts by weight".

[0038] Formulation Example 1

Five(5) parts of the present compound 1-1, and 5 parts of either of the present compound A-1 or the present compound A-2, 35 parts of a mixture of white carbon and polyoxyethylene alkyl ether sulfate ammonium salt (weight ratio 1:1), and 55 parts of water are mixed, and the resultant solution is then subjected to fine grinding according to a wet grinding method to obtain each flowable.

[0039] Formulation Example 2

Five(5) parts of the present compound 1-2, 5 parts of either of the present compound A-1 or the present compound A-2, 35 parts of a mixture of white carbon and polyoxyethylene alkyl ether sulfate ammonium salt (weight ratio 1:1), and 55 parts of water are mixed, and the resultant solution is then subjected to fine grinding according to a wet grinding method to obtain each flowable.

[0040] Formulation Example 3

Ten(10) parts of the present compound 1-1, 5 parts of either of the present compound A-1 or the present compound A-2, 1.5 parts of sorbitan trioleate, and 28 parts of aqueous solution that contained 2 parts of polyvinyl alcohol are mixed, and the resultant solution is then subjected to fine grinding according to a wet grinding method, and thereto are added 45.50 parts of an aqueous solution that contained 0.05 parts of xanthan gum and 0.1 part of aluminum magnesium silicate, followed by adding 10 parts of propylene glycol, and the mixture is blended by stirring to obtain each flowable.

[0041] Formulation Example 4

Ten(10) parts of the present compound 1-2, 5 parts of either of the present compound A-1 or the present compound A-2, 1.5 parts of sorbitan trioleate, and 28 parts of aqueous solution that contained 2 parts of polyvinyl alcohol are mixed, and the resultant solution is then subjected to fine grinding according to a wet grinding method, and thereto are added 45.50 parts of an aqueous solution that contained 0.05 parts of xanthan gum and 0.1 part of aluminum magnesium silicate, followed by adding 10 parts of propylene glycol, and the mixture is blended by stirring to obtain each flowable.

[0042] Formulation Example 5

Ten(10) parts of the present compound 1-1, 40 parts of either of the present compound A-1 or the present compound A-2, 3 parts of calcium lignosulfonate, 2 parts of sodium lauryl sulfate and 45 parts of synthetic hydrous silicon oxide are fully ground and mixed to obtain each wettable powder.

[0043] Formulation Example 6

Ten(10) parts of the present compound 1-2, 40 parts of either of the present compound A-1 or the present compound A-2, 3 parts of calcium lignosulfonate, 2 parts

of sodium lauryl sulfate and 45 parts of synthetic hydrous silicon oxide are fully ground and mixed to obtain each wettable powder.

[0044] Formulation Example 7

Five(5) parts of the present compound 1-1, 5 parts of either of the present compound A-1 or the present compound A-2, 14 parts of polyoxyethylene styryl phenyl ether, 6 parts of dodecylbenzene sulfonic acid calcium salt, and 70 parts of xylene are mixed fully to obtain each formulation.

[0045] Formulation Example 8

Five(5) parts of the present compound 1-2, 5 parts of either of the present compound A-1 or the present compound A-2, 14 parts of polyoxyethylene styryl phenyl ether, 6 parts of dodecylbenzene sulfonic acid calcium salt, and 70 parts of xylene are mixed fully to obtain each formulation.

[0046] Next, Test Examples are described.

[0047] Test Example 1

Either of the present compound 1-1 or the present compound 1-2, and either of the present compound A-1 or the present compound A-2 are mixed, and each of the resultant mixtures is diluted with dimethyl sulfoxide such that each concentration of the present compound 1, etc., and the present compound A, etc., is 10 ppm. The resultant dilute solution is dispensed into a microtiter plate (with 96 wells) in 1  $\mu$ l portion thereof per well. Thereto is then dispensed 150  $\mu$ l of a potato dextrose broth medium (PDB medium) to which conidia of Septoria leaf blotch (*Mycosphaerella graminicola*) is inoculated in advance. This plate is cultured at 18°C for four days, thereby allowing Septoria leaf blotch to undergo proliferation, and the absorbance at 550 nm of each well of the microtiter plate is then measured to examine a degree of growth of the Septoria leaf blotch.

The efficacy is calculated on the basis of the obtained degree of growth of the treated group and the untreated group, respectively, by the following "Equation 1". From the test results, a high efficacy is acknowledged.

Equation 1

$$\text{Efficacy (\%)} = 100 \times (X - Y) / X$$

X: Degree of growth of fungus in the untreated group

Y: Degree of growth of fungus in the treated group

[0048] Test Example 2

A plastic pot is filled with soil and thereto wheat (cultivar. Shirogane) is seeded and the plants are grown in a greenhouse for ten days. Each of the present compound 1-1 or the present compound 1-2, and either of the present compound A-1 or the present compound A-2 are made to a formulation according to a method described in any one of the above-mentioned Formulation Examples 1 to 8, and each of the resultant for-

mulation is diluted with water such that each concentration of the present compound 1, etc., and the present compound A, etc., respectively is 100 ppm. The resultant dilute solution is sprayed to foliar parts so as to adhere adequately onto the surfaces of leaves of the above wheats. After spraying the dilute solution, the plants are air-dried, and one day after the application, an aqueous suspension of uredospores of Brown rust (*Puccinia recondita*) is inoculated by spraying thereto. After the inoculation, the plants are placed at 27°C under humid condition for one day, and are then cultivated under lighting for ten to fifteen days, and a lesion area is examined (hereinafter referred to as "treated group").

Whereas, wheats are cultivated similarly to the treated group except that no foliage application of the above-mentioned agent solutions are done (hereinafter referred to as "untreated group"). A lesion area of Brown rust is examined similarly to the above-mentioned treated group.

From each of the lesion area of the treated group and the untreated group respectively, the efficacy of the treated group is calculated by the following Equation 2. From the test results, a high efficacy is acknowledged.

Equation 2

Efficacy (%) = [1 - (lesion area of the treated group) / (lesion area of the untreated group)] × 100

[0049] Test Example 3

Each of the present compound 1-1 and the present compound A-2 was dissolved into dimethyl sulfoxide such that each concentration of these compounds was adjusted to one hundred fifty times as much as the concentration indicated in the below-mentioned Table 2. The resultant agent solution was dispensed into each microtiter plate (with 96 wells) in 1 µl portion thereof per well. 149 µl of YBG medium to which conidia of *Septoria leaf blotch* (*Mycosphaerella graminicola*) was inoculated in advance (which was prepared by dissolving 10 g of yeast extract, 10 g of Bacto Peptone, and 20 mL of glycerol into 1 L of water, followed by sterilizing the medium) was dispensed into each of the well to which the agent solution was dispensed. The plate was cultivated at 18°C for four days, thereby allowing *Septoria leaf blotch* (*Mycosphaerella graminicola*) to undergo proliferation, and the absorbance at 550 nm of each well of the microtiter plate was then measured to examine a degree of growth of *Septoria leaf blotch* (hereinafter referred to as "treated group").

Whereas, *Septoria leaf blotch* was proliferated similarly to the case of the treated group except that dimethyl sulfoxide was used instead of the agent solution, and the degree of the growth was examined (hereinafter referred to as "untreated group"). The efficacy was calculated from each of the obtained degree of growth of the treated group and the untreated group respectively by the following "Equation 1".

From the test results, it was acknowledged that a synergistic effect was shown in the mixed-use group of the present compound 1-1 and the present compound A-2 in comparison with the case of each of the single-use group of the above-mentioned compounds respectively.

Equation 1

$$\text{Efficacy (\%)} = 100 \times (X - Y) / X$$

X: Degree of growth of fungus in the untreated group

Y: Degree of growth of fungus in the treated group

[0050] [Table 2]

| Testing compound                            | Concentration in medium (ppm) | Mixing ratio (Present compound 1-1 : Present compound A-2) | Efficacy (%) |
|---------------------------------------------|-------------------------------|------------------------------------------------------------|--------------|
| Present compound 1-1                        | 0.005                         | —                                                          | 4            |
| Present compound A-2                        | 0.05                          | —                                                          | 65           |
| Present compound 1-1 + Present compound A-2 | 0.1 + 0.01                    | 1:0.1                                                      | 100          |
| Present compound 1-1 + Present compound A-2 | 0.005 + 0.05                  | 1:10                                                       | 100          |

[0051] Test Example 4

Five(5) parts of the present compound 1-1, 35 parts of a mixture of white carbon and ammonium polyoxyethylene alkyl ether sulfate (the weight ratio of 1:1) and 55 parts of water were mixed, and the mixture was then finely-ground by a wet grinding method to obtain a flowable comprising the present compound 1-1. Separately, the flowable comprising the present compound A-2 was prepared according to the above-mentioned similar method except that the present compound A-2 was used instead of the present compound 1-1.

Each of the flowable comprising the present compound 1-1 and the flowable comprising the present compound A-2 was diluted with water and if necessary, the resultant dilute solutions were mixed each other such that each concentration of the respective compounds in the dilution solution was adjusted to the concentration indicated in Table 3, to prepare the dilution solutions, respectively.

A plastic pot was filled with soil, and thereto wheat (cultivar. Shirogane) was seeded,

and the plants were grown in a greenhouse for ten days. The above-mentioned dilution solutions were sprayed to foliar parts so as to adhere adequately onto the surfaces of leaves of the above wheats. After spraying the dilute solution, the plants were air-dried, and one day after the application, an aqueous suspension of uredospores of Brown rust (*Puccinia recondita*) was inoculated by spraying thereto. After the inoculation, the plants were placed at 27°C under humid condition for one day, and were then cultivated under lighting for ten days, and a lesion area was examined (hereinafter referred to as "lesion area of treated group").

Whereas, wheats were cultivated similarly to the treated group except that no foliage application of the above-mentioned agent solution was done, and Brown rust were inoculated, and the lesion area thereof was examined (hereinafter referred to as "lesion area of the untreated group").

From each of the lesion area of the treated group and the untreated group, respectively, the efficacy of the treated group was calculated by the following Equation 2.

From the test results, it was acknowledged that a synergistic effect was shown in the mixed-use group of the present compound 1-1 and the present compound A-2 in comparison with the case of each of the single-use group of the above-mentioned compounds respectively.

Equation 2

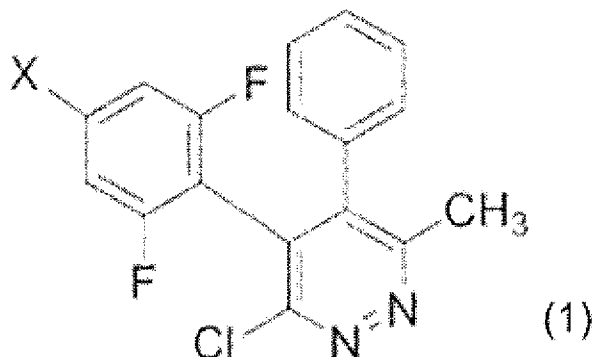
Efficacy (%) = [1- (lesion area of the treated group) / (lesion area of the untreated group)] × 100

[0052] [Table 3]

| Testing compound                            | Concentration in agent solution (ppm) | Mixing ratio (Present compound 1-1 : Present compound A-2) | Efficacy (%) |
|---------------------------------------------|---------------------------------------|------------------------------------------------------------|--------------|
| Present compound 1-1                        | 0.01                                  | -                                                          | 31           |
| Present compound 1-1                        | 0.0001                                | -                                                          | 20           |
| Present compound A-2                        | 0.001                                 | -                                                          | 20           |
| Present compound 1-1 + Present compound A-2 | 0.01 + 0.001                          | 1:0.1                                                      | 70           |
| Present compound 1-1 + Present compound A-2 | 0.0001 + 0.001                        | 1:10                                                       | 62           |

## Claims

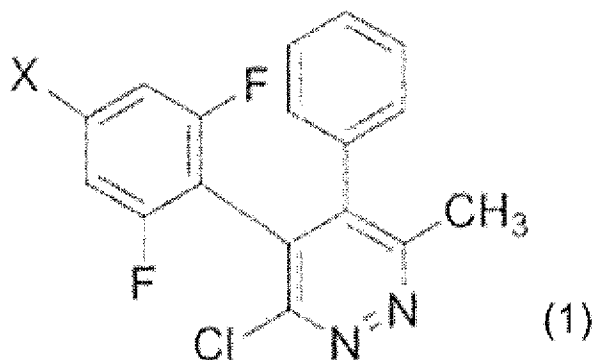
- [Claim 1] A composition for controlling a plant disease comprising a pyridazine compound represented by formula (1):  
[Chem.1]



wherein X represents a fluoro atom or a hydrogen atom, and at least one amidine compound selected from the following group (A),  
Group (A): a group consisting of  
N'-{4-[(4,5-dichlorothiazol-2-yl)oxy]-2,5-dimethylphenyl}-N-ethyl-N-methylmethane imidamide and  
N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane imidamide.

- [Claim 2] The composition for controlling a plant disease according to claim 1 wherein the pyridazine compound is the compound represented by formula (1) wherein X represents a hydrogen atom.
- [Claim 3] The composition for controlling a plant disease according to claim 1 or 2 wherein a weight ratio of the pyridazine compound to the amidine compound is within the range of 1:0.0125 to 1:500.
- [Claim 4] A method for controlling a plant disease, comprising a step of applying each effective amount of a pyridazine compound represented by formula (1):

[Chem.2]

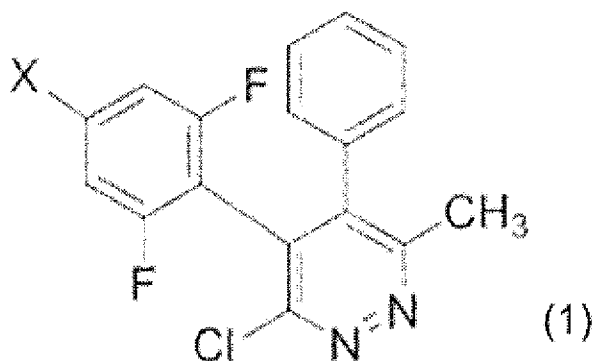


wherein X represents a fluoro atom or a hydrogen atom, and  
 at least one amidine compound selected from the following group (A),  
 Group (A): a group consisting of  
 N'-{4-[(4,5-dichlorothiazol-2-yl)oxy]-2,5-dimethylphenyl}-N-ethyl-N-methylmethane imidamide and  
 N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane imidamide,  
 to a plant or soil for cultivating the plant.

[Claim 5]

A method for controlling a plant disease, comprising a step of applying  
 each effective amount of a pyridazine compound represented by  
 formula (1):

[Chem.3]

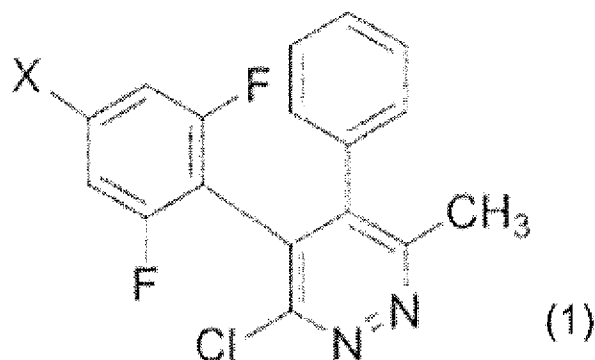


wherein X represents a fluoro atom or a hydrogen atom, and  
 at least one amidine compound selected from the following group (A),  
 Group (A): a group consisting of  
 N'-{4-[(4,5-dichlorothiazol-2-yl)oxy]-2,5-dimethylphenyl}-N-ethyl-N-methylmethane imidamide and  
 N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane imidamide

N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane  
imidamide,  
to a seed.

[Claim 6]

A combined use of a pyridazine compound represented by formula (1):  
[Chem.4]



wherein X represents a fluoro atom or a hydrogen atom, and  
at least one amidine compound selected from the following group (A),  
Group (A): a group consisting of  
N'-{4-[(4,5-dichlorothiazol-2-yl)oxy]-2,5-dimethylphenyl}-N-ethyl-N-  
methylmethane imidamide and  
N'-(2,5-dimethyl-4-phenoxyphenyl)-N-ethyl-N-methylmethane  
imidamide.

# INTERNATIONAL SEARCH REPORT

International application No  
PCT/JP2017/009717

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|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------|
| <b>A. CLASSIFICATION OF SUBJECT MATTER</b><br>INV.    A01N37/52            A01N43/58            A01N43/78            A01N25/04            A01N25/14<br>A01P3/00            A01N25/00<br>ADD.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       |                                                                                                                       |                                                                                                                               |
| According to International Patent Classification (IPC) or to both national classification and IPC                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  |                                                                                                                       |                                                                                                                               |
| <b>B. FIELDS SEARCHED</b><br>Minimum documentation searched (classification system followed by classification symbols)<br>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 practicable, search terms used)<br>EPO-Internal, WPI Data, CHEM ABS Data                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |                                                                                                                       |                                                                                                                               |
| <b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      |                                                                                                                       |                                                                                                                               |
| Category*                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                          | Citation of document, with indication, where appropriate, of the relevant passages                                    | Relevant to claim No.                                                                                                         |
| X                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  | EP 2 014 167 A1 (BAYER CROPSCIENCE AG [DE]) 14 January 2009 (2009-01-14) columns 3-5<br>-----                         | 1-6                                                                                                                           |
| X                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  | US 2015/181878 A1 (WACHENDORFF-NEUMANN ULRIKE [DE] ET AL) 2 July 2015 (2015-07-02) paragraphs [0023], [0034]<br>----- | 1-6                                                                                                                           |
| X,P                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                | EP 3 011 832 A1 (BAYER CROPSCIENCE AG [DE]) 27 April 2016 (2016-04-27) paragraph [0008]<br>-----                      | 1-6                                                                                                                           |
| <div style="display: flex; justify-content: space-between;"> <div> <input type="checkbox"/> Further documents are listed in the continuation of Box C.         </div> <div> <input checked="" type="checkbox"/> See patent family annex.         </div> </div>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |                                                                                                                       |                                                                                                                               |
| <div style="display: flex;"> <div style="flex: 1;"> <p>* Special categories of cited documents :</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier application or patent but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="flex: 1;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"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</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</p> <p>"&amp;" document member of the same patent family</p> </div> </div> |                                                                                                                       |                                                                                                                               |
| Date of the actual completion of the international search<br><br><div style="text-align: center; font-size: 1.2em;">5 May 2017</div>                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                                                                                                                       | Date of mailing of the international search report<br><br><div style="text-align: center; font-size: 1.2em;">15/05/2017</div> |
| Name and mailing address of the ISA/<br>European Patent Office, P.B. 5818 Patentlaan 2<br>NL - 2280 HV Rijswijk<br>Tel. (+31-70) 340-2040,<br>Fax: (+31-70) 340-3016                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |                                                                                                                       | Authorized officer<br><br><div style="text-align: center; font-size: 1.2em;">Sawicki, Marcin</div>                            |

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/JP2017/009717

| Patent document<br>cited in search report | Publication<br>date | Patent family<br>member(s) | Publication<br>date     |
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