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

Canadian Intellectual Property Office
An agency of Industry Canada

CA 2380864 C 2005/07/26

(11)(21) 2 380 864

(12) BREVET CANADIEN
CANADIAN PATENT

(13) C

(86) Date de dépôt PCT/PCT Filing Date: 2000/08/16
(87) Date publication PCT/PCT Publication Date: 2001/02/22
(45) Date de délivrance/Issue Date: 2005/07/26
(85) Entrée phase nationale/National Entry: 2002/01/31
(86) N° demande PCT/PCT Application No.: KR 2000/000906
(87) N° publication PCT/PCT Publication No.: 2001/012585
(30) Priorités/Priorities: 1999/08/16 (1999/33724) KR;
1999/08/16 (1999/33722) KR

(51) Cl.Int.^{7}/Int.Cl.^{7} C07C 229/20, C07C 217/48

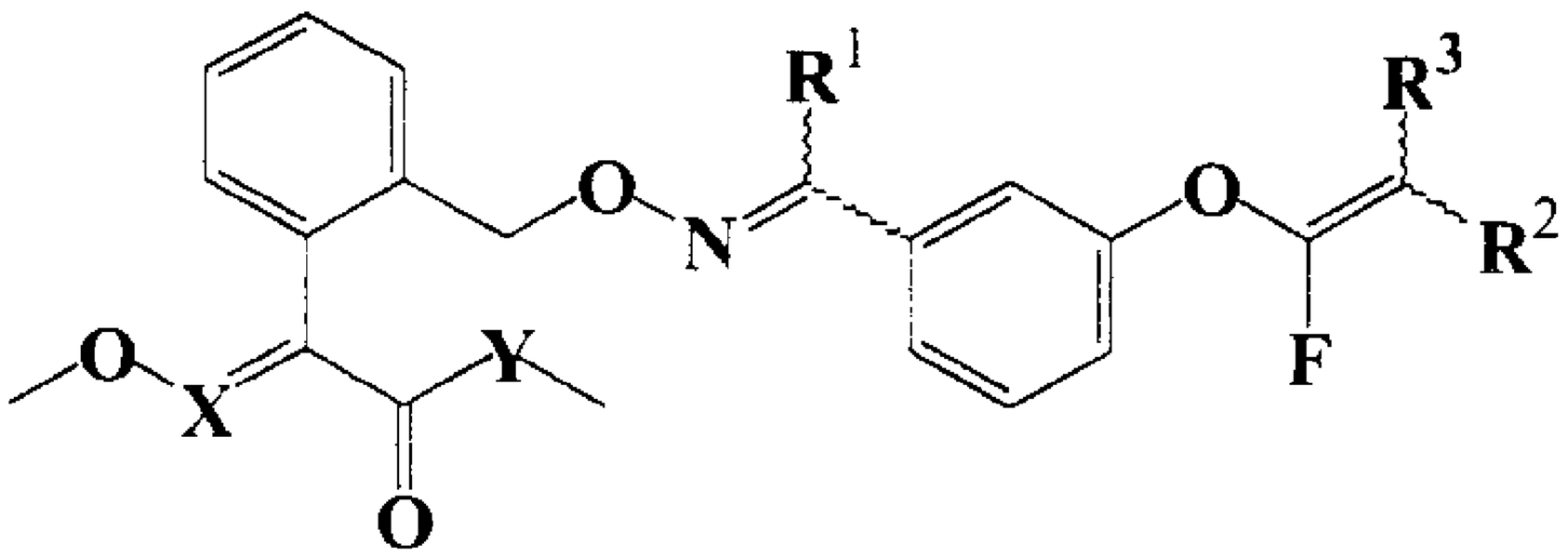
(72) Inventeurs/Inventors:
KIM, BUM TAE, KR;
PARK, NO KYUN, KR;
CHOI, GYUNG JA, KR;
KIM, JIN CHEOL, KR;
PAK, CHWANG SIEK, KR

(73) Propriétaire/Owner:
KOREA RESEARCH INSTITUTE OF CHEMICAL TECHNOLOGY, KR

(74) Agent: CASSAN MACLEAN

(54) Titre : COMPOSES FONGICIDES COMPRENANT UNE FRACTION FLUOROVINYL- OU FLUOROPROPENYL-OXYPHENYLOXIME, ET LEUR PROCEDE DE PREPARATION

(54) Title: FUNGICIDAL COMPOUNDS HAVING A FLUOROVINYL- OR FLUOROPROPENYL-OXYPHENYLOXIME MOIETY AND PROCESS FOR THE PREPARATION THEREOF



(I)

(57) Abrégé/Abstract:
A fungicidal compound of formula (I) having a fluorovinye- or fluoropropenyl-oxyphenyloxime moiety and stereoisomers thereof are useful for protecting crops from fungal diseases: wherein, X is CH or N; Y is O or NH; R^1 is hydrogen, C_{1-4} alkyl, or halogen-substituted C_{1-4} alkyl, R^2 is a phenyl group optionally carrying one or more substituents selected from the group consisting of C_{1-4} alkyl, C_{1-4} alkoxy, methylenedioxy and halogen; or a naphthyl group; and R^3 is hydrogen or CF_3.

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
22 February 2001 (22.02.2001)

PCT

(10) International Publication Number
WO 01/12585 A1

(51) International Patent Classification[7]: C07C 229/20, 217/48

(21) International Application Number: PCT/KR00/00906

(22) International Filing Date: 16 August 2000 (16.08.2000)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:

1999/33722	16 August 1999 (16.08.1999)	KR
1999/33724	16 August 1999 (16.08.1999)	KR

(71) Applicant *(for all designated States except US)*: **KOREA RESEARCH INSTITUTE OF CHEMICAL TECHNOLOGY** [KR/KR]; 100, Jang-dong, Yuseong-gu, Daejeon 305-343 (KR).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: **KIM, Bum, Tae** [KR/KR]; 15-806 Samsung Apt., #175-1 Ohryu-dong, Jung-gu, Daejeon 301-120 (KR). **PARK, No, Kyun** [KR/KR]; 111-302 Kyungnam Apt., #409 Doma-2-dong, Seo-gu, Daejeon 302-162 (KR). **CHOI, Gyung, Ja** [KR/KR]; 104-1005 Narae Apt., #462-4 Jeonmin-dong, Yuseong-gu, Daejeon 305-390 (KR). **KIM, Jin, Cheol** [KR/KR]; 8-202 Kongdongkwanri Apt., #431 Doryong-dong, Yuseong-gu, Daejeon 305-340 (KR). **PAK, Chwang, Siek** [KR/KR]; #383-6 Doryong-dong, Yuseong-gu, Daejeon 305-340 (KR).

(74) Agents: **JANG, Seong, Ku** et al.; 17th Floor, KEC Building, #275-7 Yangjae-dong, Seocho-ku, Seoul 137-130 (KR).

(81) Designated States *(national)*: AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, DE, DK, DM, DZ, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

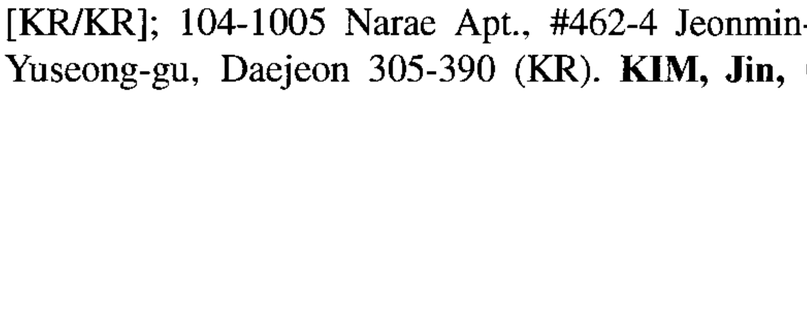
(84) Designated States *(regional)*: ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published:
— *With international search report.*

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

WO 01/12585 A1

(54) Title: FUNGICIDAL COMPOUNDS HAVING A FLUOROVINYL- OR FLUOROPROPENYL-OXYPHENYLOXIME MOIETY AND PROCESS FOR THE PREPARATION THEREOF



(I)

R^3 is hydrogen or CF_3.

(57) Abstract: A fungicidal compound of formula (I) having a fluorovinye- or fluoropropenyl-oxyphenyloxime moiety and stereoisomers thereof are useful for protecting crops from fungal diseases: wherein, X is CH or N; Y is O or NH; R^1 is hydrogen, C_{1-4} alkyl, or halogen-substituted C_{1-4} alkyl, R^2 is a phenyl group optionally carrying one or more substituents selected from the group consisting of C_{1-4} alkyl, C_{1-4} alkoxy, methylenedioxy and halogen; or a naphthyl group; and

FUNGICIDAL COMPOUNDS HAVING A FLUOROVINYL- OR FLUOROPROPENYL-OXYPHENYLOXIME MOIETY AND PROCESS FOR THE PREPARATION THEREOF

Field of the Invention

The present invention relates to novel fungicidal compounds having a fluorovinyl- or fluoropropenyl-oxyphenyloxime moiety, a process for preparing same and a fungicidal composition containing same as an active ingredient.

Description of the Prior Art

A number of fungicidal compounds have been in practical use to protect crops from various pathogenic fungi; and they may be classified into several groups according to their similar structural features. However, the repetitive use of a fungicide over a long period induces the appearance of new fungal strains resistant not only to the particular fungicide but also to related fungicides having common structural features. For this reason, continuous efforts have been undertaken to develop novel fungicides.

Such efforts have led to the development of new fungicides, e.g., propenoic esters derived from strobilurin (U.S. Patent No. 4,994,495; WO 94/19331; U.S. Patent No. 5,003,101) and other propenoic ester fungicides disclosed in EP A 0 278 595 (Zeneca), EP A 0 782 982 (Novatis), WO 96/33164 (Ciba-Geigy), WO 96/33164 (Rhone-Poulenc Agro), WO 98/56774 (BASF), WO 99/06379 (BASF), WO 99/23066 (Agrevo UK), German Patent Nos. 724,200 and 732,846 (both BASF), and British Patent No. 22,893 (Agrevo UK). However, these propenoic ester derivatives still have the problem of limited fungicidal activity.

Summary of the Invention

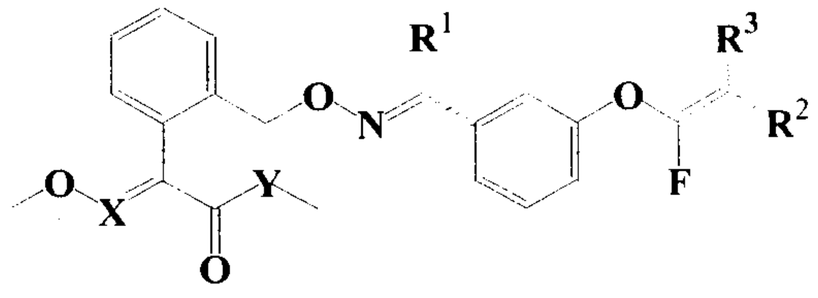
Accordingly, it is a primary object of the present invention to provide a novel compound having a high fungicidal activity against a wide spectrum of plant pathogenic fungi.

It is another object of the present invention to provide a process for the preparation of said compound.

It is a further object of the present invention to provide a fungicidal composition containing said compound.

In accordance with one aspect of the present invention, there are provided a novel

compound of formula (I) and stereoisomers thereof:



(I)

wherein,

X is CH or N;

Y is O or NH;

R^1 is hydrogen, C_{1-4} alkyl, or halogen-substituted C_{1-4} alkyl;

R^2 is a phenyl group optionally carrying one or more substituents selected from the group consisting of C_{1-4} alkyl, C_{1-4} alkoxy, methylenedioxy and halogen; or a naphthyl group; and

R^3 is hydrogen or CF_3.

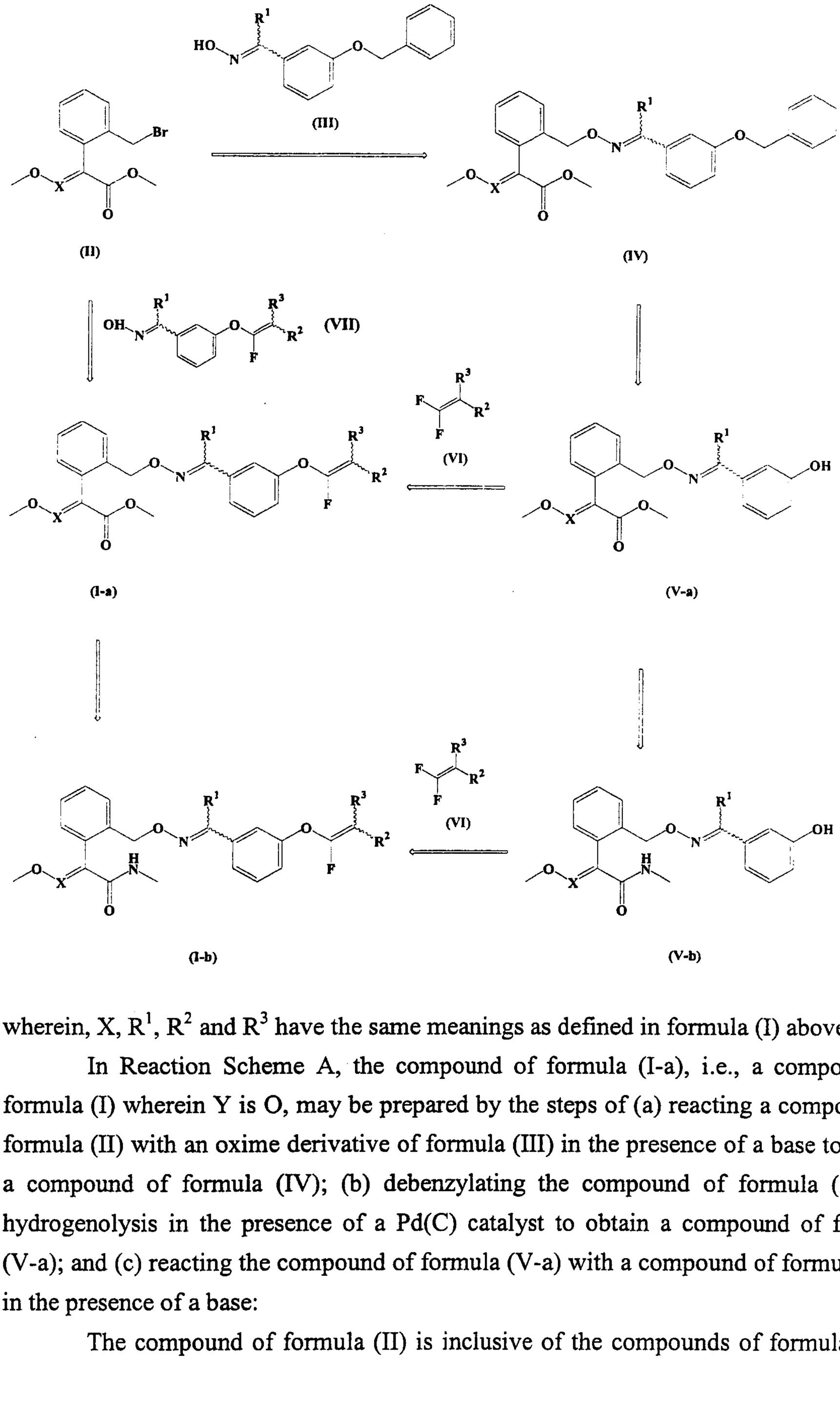
Detailed Description of the Invention

The structure of the compound of formula (I) of the present invention is characterized by the fluorovinyl- or fluoropropenyl-oxyphenyloxime moiety, and depending on whether X is CH or N, it may also be classified as a propenoic acid derivative (X=CH) or as an iminophenylacetic acid derivative (X=N).

Among the compounds of the present invention, preferred are those wherein R^1 is hydrogen or methyl, and R^2 is phenyl group, or Cl- or F-substituted phenyl group.

The compound of formula (I) of the present invention may be prepared, as shown in Reaction Scheme A:

Reaction Scheme A

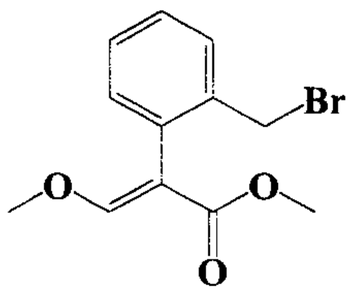


wherein, X, R^1, R^2 and R^3 have the same meanings as defined in formula (I) above.

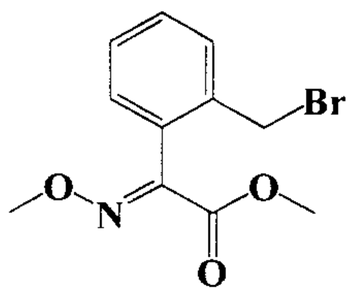
In Reaction Scheme A, the compound of formula (I-a), i.e., a compound of formula (I) wherein Y is O, may be prepared by the steps of (a) reacting a compound of formula (II) with an oxime derivative of formula (III) in the presence of a base to obtain a compound of formula (IV); (b) debenzylating the compound of formula (IV) by hydrogenolysis in the presence of a Pd(C) catalyst to obtain a compound of formula (V-a); and (c) reacting the compound of formula (V-a) with a compound of formula (VI) in the presence of a base:

The compound of formula (II) is inclusive of the compounds of formula (II-a)

(X=CH) and formula (II-b) (X=N):



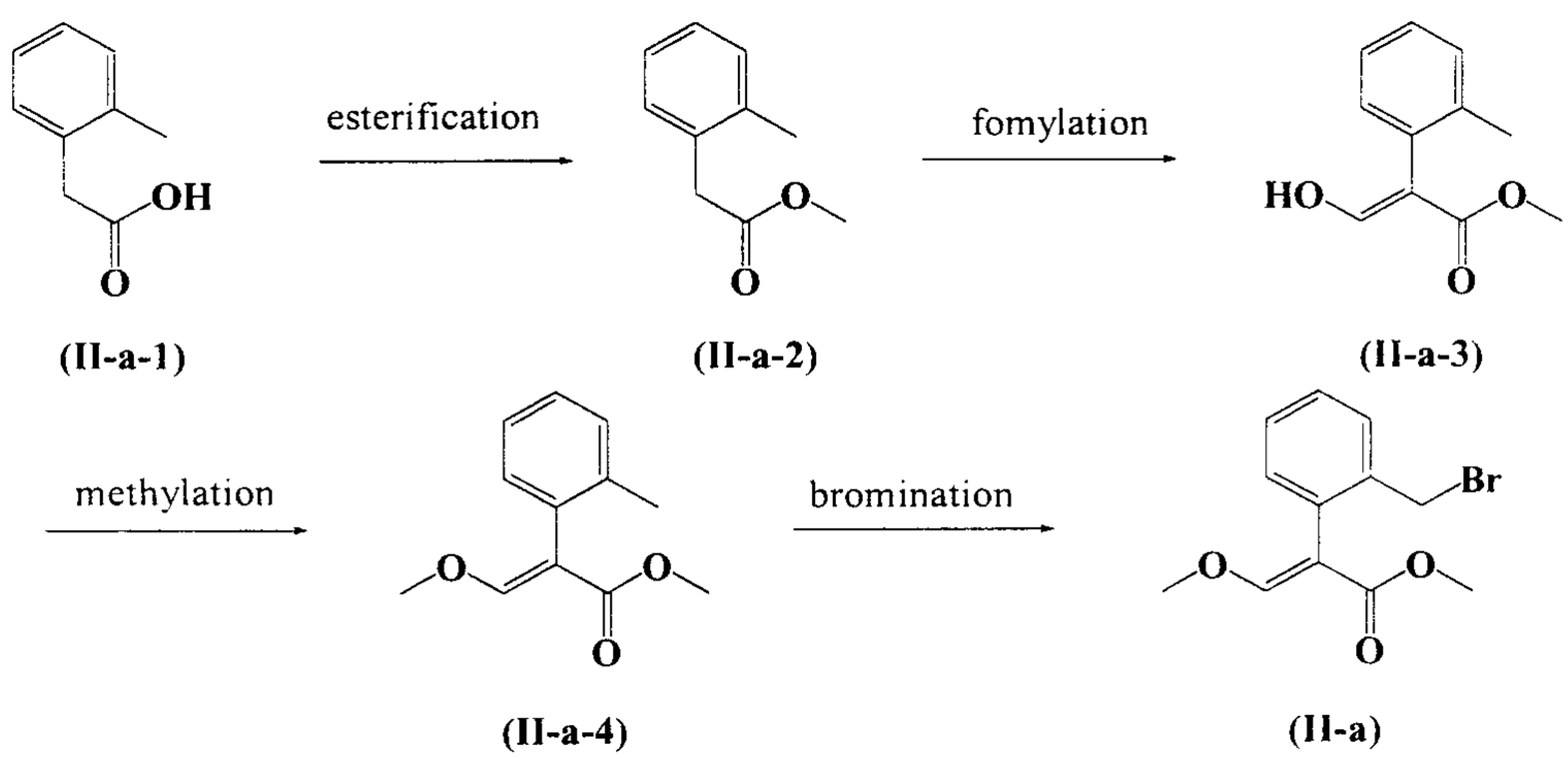
(II-a)



(II-b)

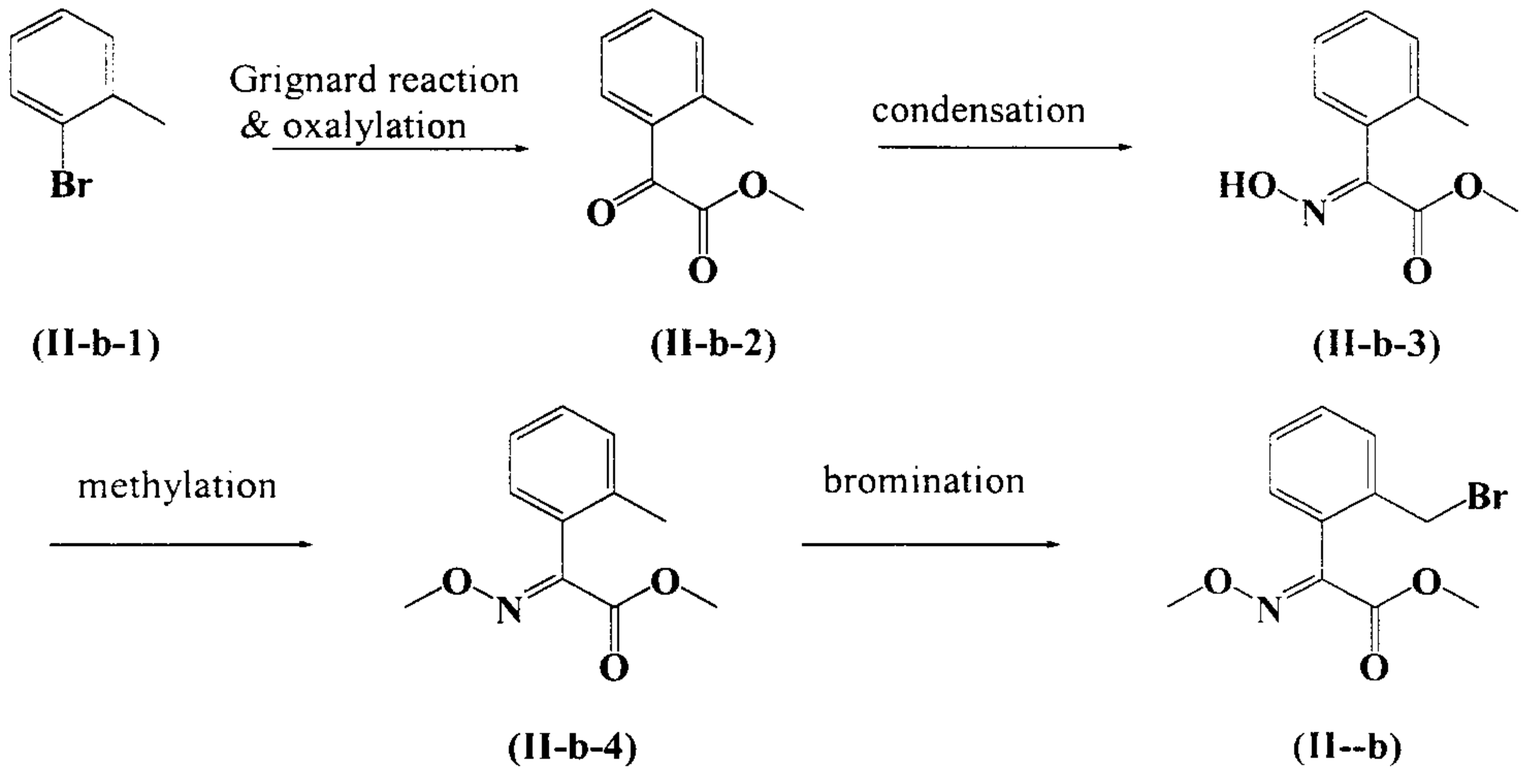
The compound of formula (II-a) may be prepared by esterification, formylation, methylation and bromination of *o*-tolylacetic acid according to a conventional method (Yamada, K. et al., *Tetrahedron Lett.*, 2745(1973); Vyas, G.N. et al., *Org. Syn. Coll., 4*, 836(1963); Kalir, A., *Org. Syn. Coll., 5*, 825(1973); Korean Unexamined Patent Publication Nos. 98-83587 and 99-15785; and World Patent Publication No. WO 99/07665), as shown in Reaction Scheme B:

Reaction Scheme B



The compound of formula (II-b) may be prepared by Grignard reaction, oxalylation, condensation, methylation and bromination of *o*-bromotoluene according to a conventional method (Rambaud, M. et al., *Synthesis*, 564(1988); Korean Unexamined Patent Publication Nos. 98-83587 and 99-15785; and World Patent Publication No. WO 99/07665), as shown in Reaction Scheme C:

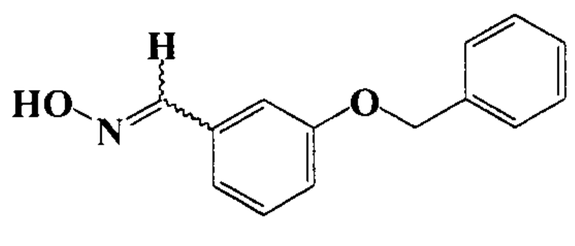
Reaction Scheme C



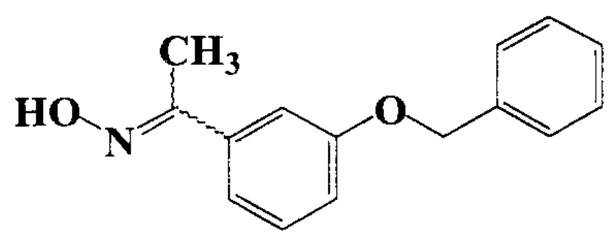
(II-b-1) (II-b-2) (II-b-3)

(II-b-4) (II--b)

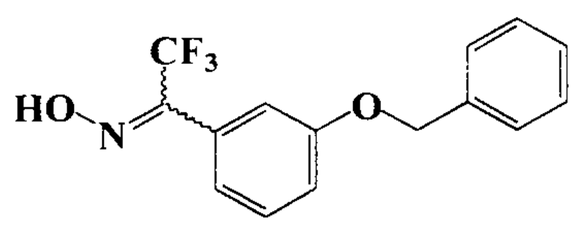
The compound of formula (III), on the other hand, represents, among others, the compounds of formula (III-a) (R^1=H), formula (III-b) (R^1=CH_3) and formula (III-c) (R^1= CF_3):



(III-a)



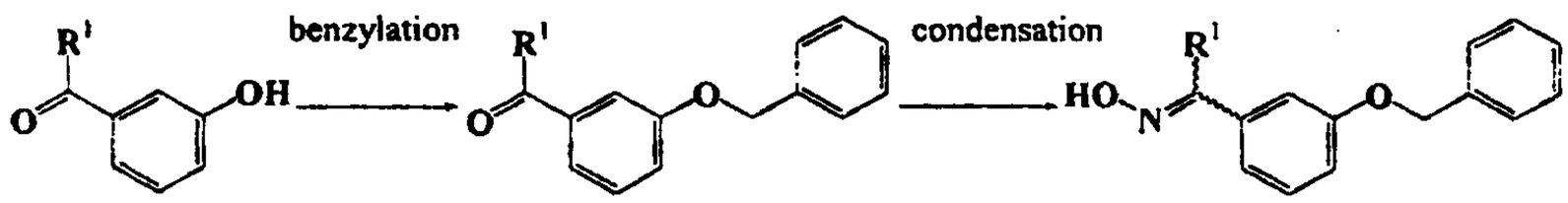
(III-b)



(III-c)

The compounds of formulas (III-a), (III-b) and (III-c) may be prepared by the steps of benzylation and condensation of 3-hydroxybenzaldehyde, 3-hydroxyacetophenone and 3-hydroxy-2'2'2'-trifluoroacetophenone, respectively, in accordance with a conventional method (Kuhn, R. et al., *Chem. Ber. 90*, 203(1957); Fletcher, H. G. et al., *Methods Carbohydr. Chem., II*, 166(1963); Freedman, H. H., et al., *Tetrahedron Lett.*, 3251(1975); Lichtenhaler, F. W., et al., *Tetrahedron Lett.*, 1425(1980); and Sugg, E. E., et al., *J. Org. Chem., 50*, 5032(1985)), as shown in Reaction Scheme D:

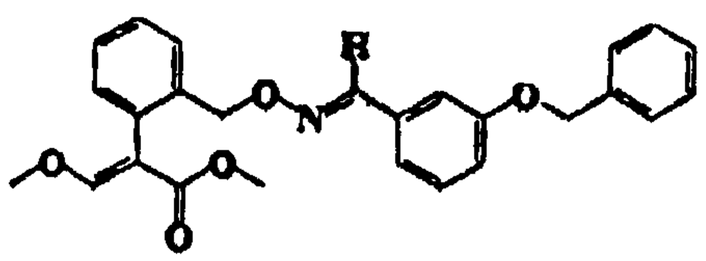
Reaction Scheme D



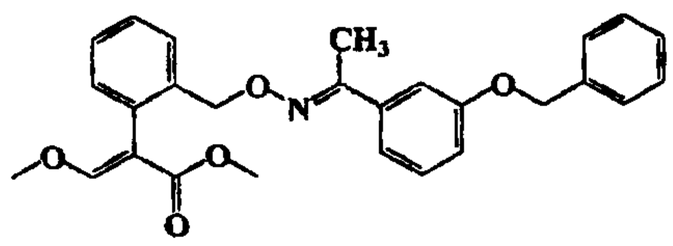
wherein, R^1 has the same meanings as defined in formula (I) above.

In the reaction to prepare the compound of formula (IV), the compound of formula (II) and the compound of formula (III) may be used in equimolar amounts and the base may be used in one or two equivalent amounts. The base may be an inorganic base, e.g., sodium hydride, potassium *t*-butoxide, sodium carbonate or potassium carbonate; or an organic base, e.g., triethyl amine or pyridine. The solvent which may be used in the reaction includes acetone, methyl ethyl ketone, benzene, toluene, tetrahydrofuran, acetonitrile, dichloromethane or dimethyl formamide, and the reaction may be conducted at a temperature ranging from room temperature to 100 ℃. The progress of the reaction is conveniently followed by measuring the disappearance of the compound of formula (II) with thin layer chromatography (TLC).

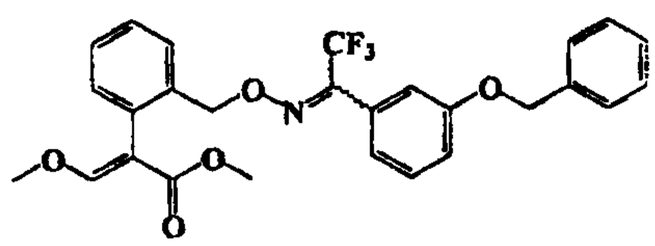
Examples of the compound of formula (IV) include the compounds of formulas (IV-a) (X=CH, R^1=H), (IV-b) (X=CH, R^1=CH_3), (IV-c) (X=CH, R^1=CF_3), (IV-d) (X=N, R^1=H), (IV-e) (X=N, R^1=CH_3) and (IV-f) (X=N, R^1=CF_3), depending on the starting materials used, i.e., depending on any one of the reactions of the compound of formula (II-a) or (II-b) with the compound of formula (III-a), (III-b) or (III-c):



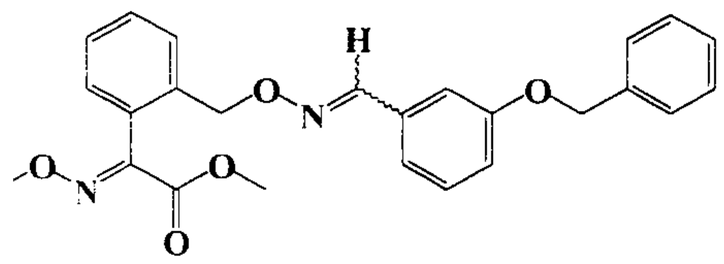
(IV-a)



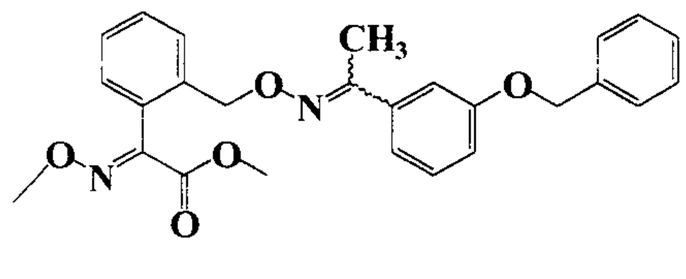
(IV-b)



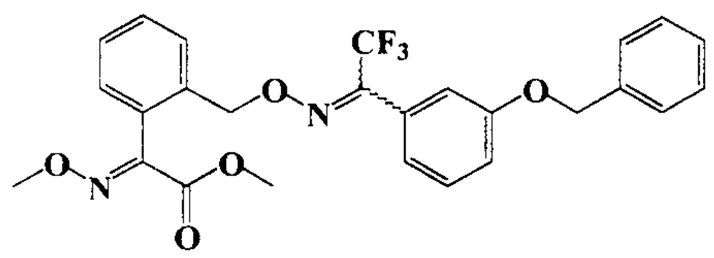
(IV-c)



(IV-d)

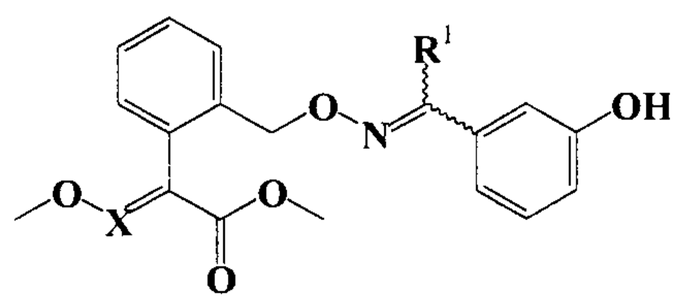


(IV-e)



(IV-f)

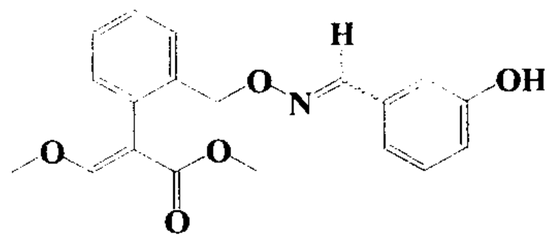
Subsequently, the compound of formula (IV) is debenzylated by hydrogenolysis to obtain a phenolic ester compound of formula (V-a):



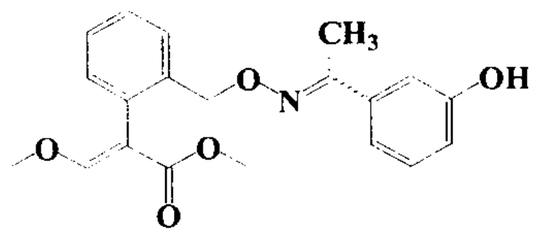
(V-a)

wherein, X and R^1 has the same meanings as defined previously.

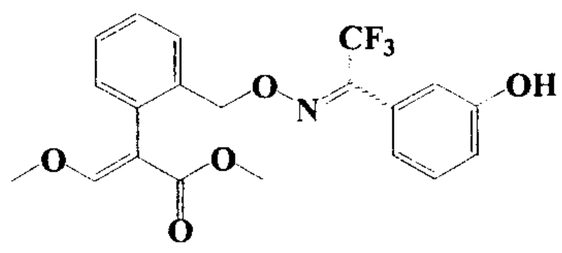
Examples of the compound of formula (V-a) include the compounds of formulas (V-a-1) (X=CH, R^1=H), (V-a-2) (X=CH, R^1=CH_3), (V-a-3) (X=CH, R^1=CF_3), (V-a-4) (X=N, R^1=H), (V-a-5) (X=N, R^1=CH_3) and (V-a-6) (X=N, R^1=CF_3), which correspond to the compounds of formulas (IV-a) to (IV-f), respectively:



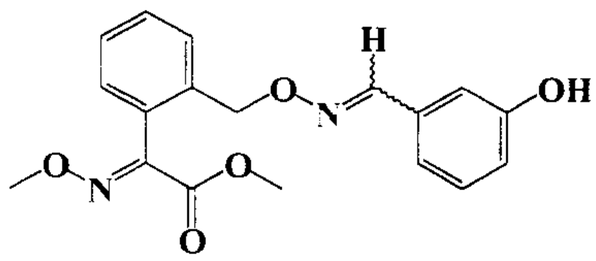
(V-a-1)



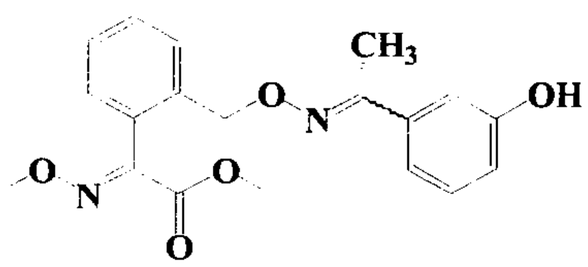
(V-a-2)



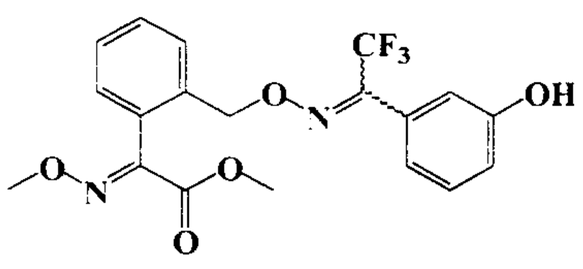
(V-a-3)



(V-a-4)

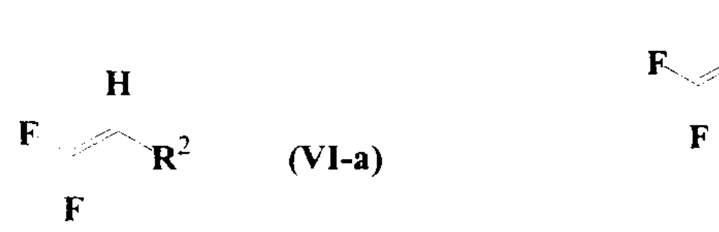


(V-a-5)

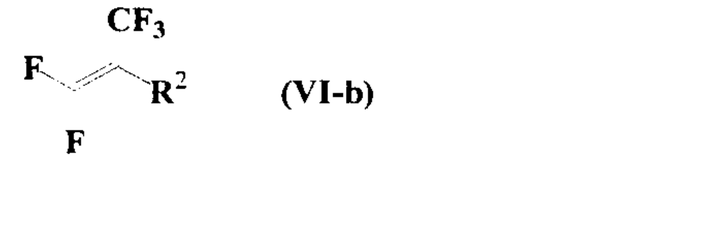


(V-a-6)

On the other hand, the compound of formula (VI) is inclusive of a compound of formula (VI-a), i.e., the compound of formula (VI) wherein R^3 is H, and a compound of formula (VI-b), i.e., the compound of formula (I) wherein R^3 is CF_3.



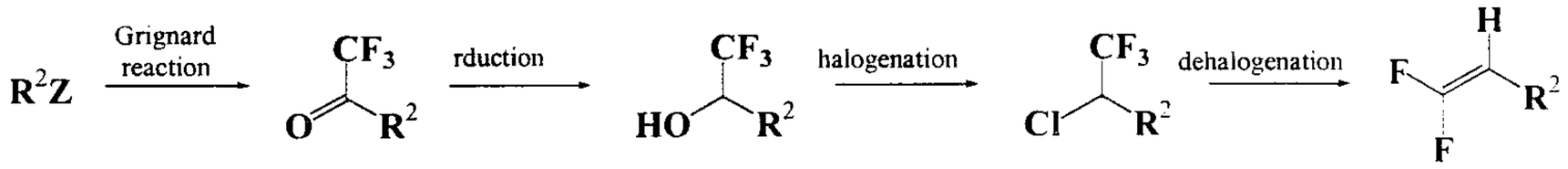
(VI-a)



(VI-b)

The compound of formula (VI-a) may be prepared by a Grignard reaction, reduction, halogenation and dehalogenation of a halide of R^2 according to a conventional method (Herkes, F.E. et al., J. Org. Chem., 32, 1311(1967); and Nemeth, G. et al., J. fluorine Chem., 76, 91(1996)), as shown in Reaction Scheme E:

Reaction Scheme E

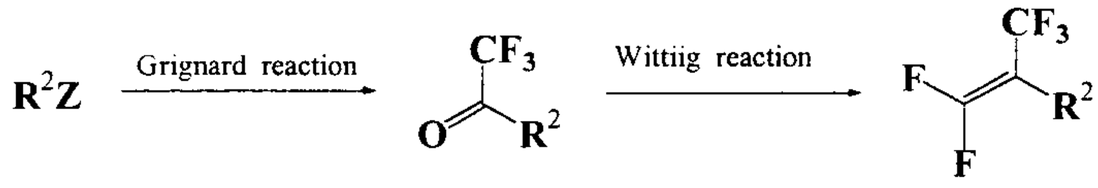


(VI-a)

wherein, R^2 has the same meaning as defined in formula (I) above; and Z represents Cl or F.

Further, a compound of formula (VI-b) may be prepared by a Grignard reaction and Wittig reaction of a halide of R^2 according to a conventional method (Herkes, F.E. et al., *J. Org. Chem., 32,* 1311(1967); and Wheatman. G.A. et al., *J. Org. Chem., 48,* 917(1983)), as shown in Reaction Scheme F:

Reaction Scheme F



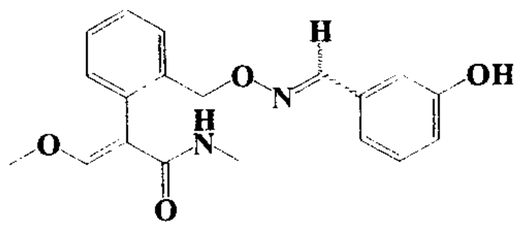
(VI-b)

wherein, R^2 and Z have the same meanings as above.

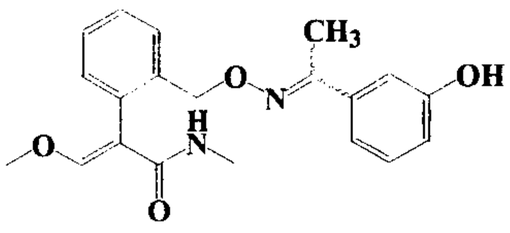
In the step to prepare the compound of formula (I-a) of the present invention by reacting the compound of formula (V-a) with a compound of formula (VI) in the presence of a base, the compounds of formulas (V-a) and (VI) may be used in equimolar amounts and the base may be used in one to two equivalent amounts. The base may be an inorganic base, e.g., sodium hydride, potassium *t*-butoxide, sodium carbonate or potassium carbonate; or an organic base, e.g., triethyl amine or pyridine. The solvent, which may be used in the reaction, is benzene, toluene, tetrahydrofuran, acetonitrile, dichloromethane or dimethyl formamide, and the reaction temperature in the range of room temperature to 100 ℃.

In the preparation of the compound of formula (I-b) by reacting the phenolic ester compound of formula (V-a) with methylamine to obtain a phenolic amide compound of formula (V-b), in a conventional manner, and then reacting the compound of formula (V-b) with a compound of formula (VI) in the presence of a base, methylamine may be preferably employed in an excess amount than the phenolic ester compound used. The above reaction may be conducted in the presence of an alcohol (e.g., methanol), acetonitrile, dichloromethane and dimethyl formamide, at a temperature ranging from room temperature to the boiling point of the solvent used.

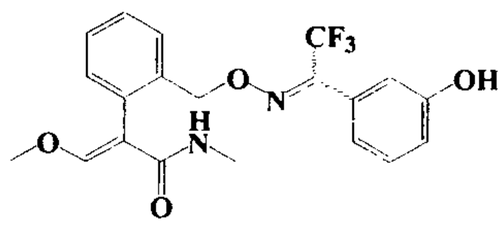
Examples of the compound of formula (V-b) are the compounds of formulas (V-b-1) (X=CH, R^1=H), (V-b-2) (X=CH, R^1=CH_3), (V-b-3) (X=CH, R^1=CF_3), (V-b-4) (X=N, R^1=H), (V-b-5) (X=N, R^1=CH_3) and (V-b-6) (X=N, R^1=CF_3), which correspond to the compounds of formulas (IV-a) to (IV-f), respectively:



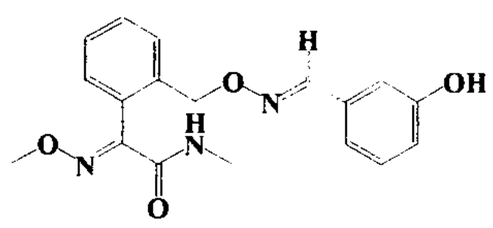
(V-b-1)



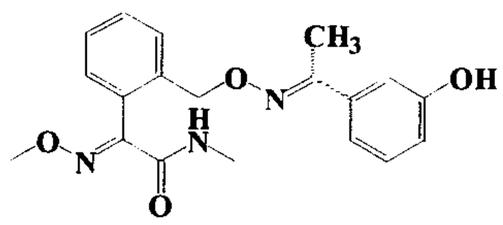
(V-b-2)



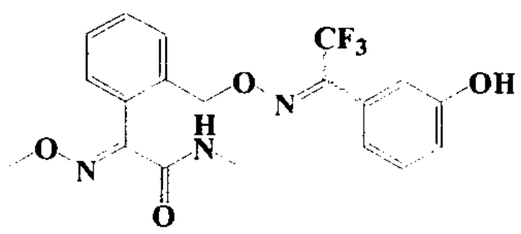
(V-b-3)



(V-b-4)



(V-b-5)



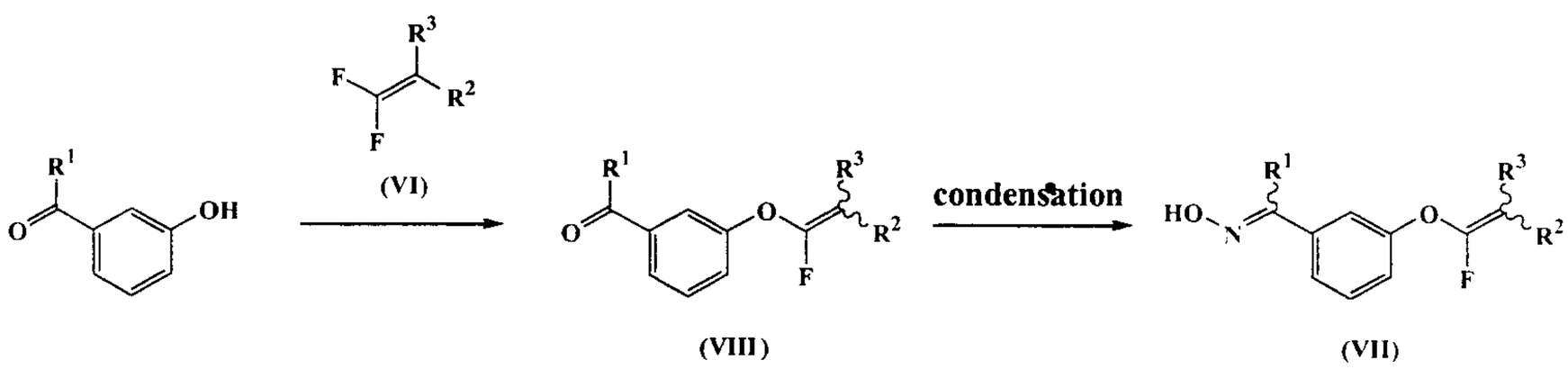
(V-b-6)

On the other hand, the compound of formula (I-a) may be prepared, as shown in the above Reaction Scheme A, by reacting a compound of formula (II) with a compound of formula (VII) in the presence of a base. At this time, the compound of formula (II) and the compound of formula (VII) may be used in equimolar amounts and the base may be used in one or two equivalent amounts. The base may be an inorganic base, e.g., sodium hydride, potassium *t*-butoxide, sodium carbonate or potassium carbonate; or an organic base, e.g., triethylamine or pyridine. The solvent which may be used in the reaction includes acetone, methyl ethyl ketone, benzene, toluene, tetrahydrofuran, acetonitrile, dichloromethane or dimethyl formamide, and the reaction may be conducted at a temperature ranging from room temperature to 100 ℃. The progress of the reaction is conveniently followed by measuring the disappearance of the compound of formula (II) with thin layer chromatography (TLC).

Further, the compound of formula (I-b) may be obtained by reacting the compound of formula (I-a) with methylamine in a conventional manner.

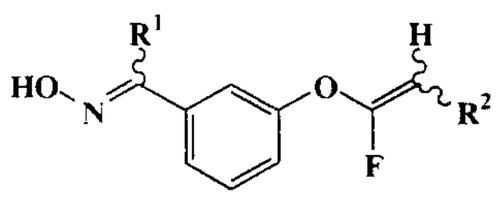
The compound of formula (VII) may be prepared by reaction of 3-hydroxybenzaldehyde, 3-hydroxyacetophenone or 3-hydroxy-2'2'2'-trifluoroacetophenone with a compound of formula (VI) to obtain a compound of formula (VIII) and condensation of the compound of formula (VIII) with hydroxylamine according to a conventional method (Lichtenhaler, F. W., et al., *Tetrahedron Lett.*, 1425(1980); and Sugg, E. E., et al., *J. Org. Chem., 50*, 5032(1985)), as shown in Reaction Scheme G:

Reaction Scheme G

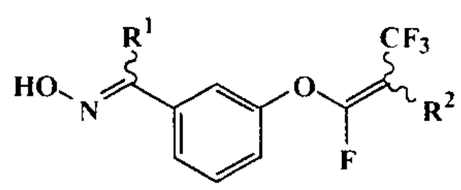


wherein, R^1, R^2 and R^3 have the same meanings as defined in formula (I).

The compound of formula (VII) is inclusive of a compound of formula (VII-a), i.e., the compound of formula (VII) wherein R^3 is H, and a compound of formula (VII-b), i.e., the compound of formula (VII) wherein R^3 is CF_3, which correspond to the compounds of formulas (VI-a) and (VI-b) used as a starting material, respectively:

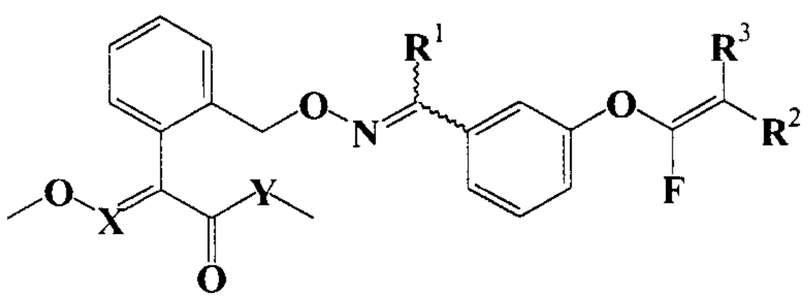


(VII-a)

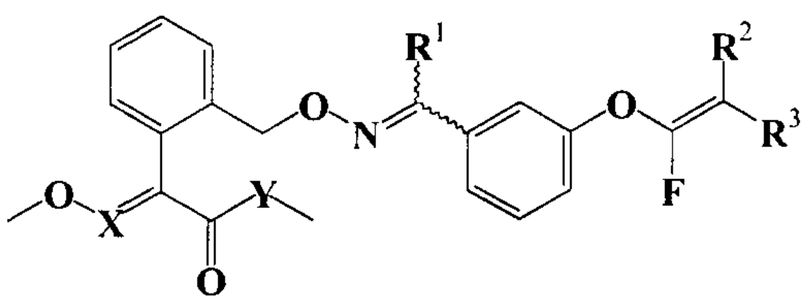


(VII-b)

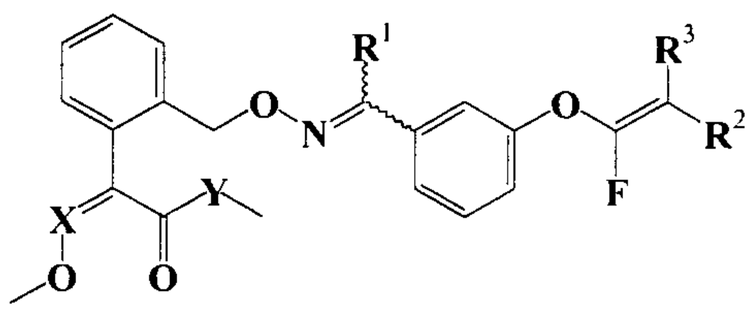
The compound of formula (I) of the present invention has three double bonds, and when one ignores the double bond of the bridging oxime group, there exist four stereoisomers thereof, which, according to the terminology defined in the Cahn-Ingold-Prelog system (J. March, Advanced Organic Chemistry, 3rd Ed., Wiley-Interscience), may be expressed as (*E,E*), (*E,Z*), (*Z,E*) and (*Z,Z*) isomers, which are included within the scope of the present invention.



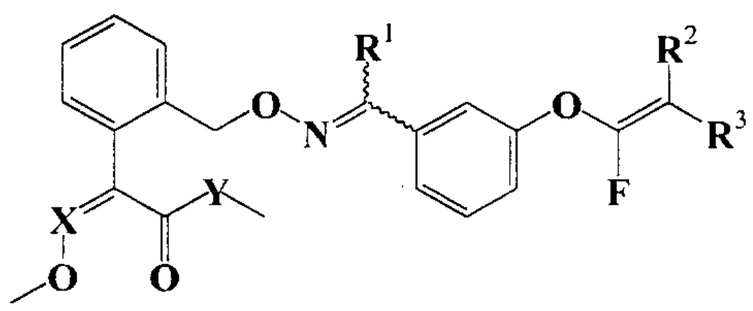
(when R^3 is H, (*E,Z*) isomer; and when R^3 is CF_3, (*E,E*) isomer)



(when R^3 is H, (*E,E*) isomer; and when R^3 is CF_3, (*E,Z*) isomer)



(when R^3 is H, (*Z,Z*) isomer; and when R^3 is CF_3, (*Z,E*) isomer)



(when R^3 is H, (*Z,E*) isomer; and when R^3 is CF_3, (*Z,Z*) isomer)

wherein, X, Y, R^1, R^2 and R^3 have the same meanings as defined above.

In case a mixture of the *E* and *Z* isomers of the compound of formula (II) is used in the reaction shown in Reaction Scheme A, the compound of the present invention is obtained as a mixture of the above four isomers wherein the (*E,E*) and (*E,Z*) isomers predominate with minor amounts of the (*Z,E*) and (*Z,Z*) isomers.

However, in case only the *E* isomer of the compound of formula (II-a-4) or (II-b-4) is used, the compound of formula (I) of the present invention is obtained as a mixture of the (*E,E*) and (*E,Z*) isomers, as is confirmed by ^{1}H-NMR or ^{19}F-NMR analysis.

According to the ^{1}H-NMR analysis (reference compound, TMS) of the compound of formula (I) of the present invention wherein R^3 is hydrogen, a hydrogen of vinyl group of (*E,E*) isomer is shown as a doublet having a coupling constant of 5 to 6 Hz at 5.5 to 5.8 ppm, while that of (*E,Z*) isomer is represented as a doublet having a coupling constant of 30 Hz at 5.0 to 5.4 ppm. The ratio of the (*E,E*) isomer to (*E,Z*) isomer is about 2:1 which may be calculated from integration on the ^{1}H-NMR spectroscopy. This result can be confirmed by the ^{19}F-NMR analysis. The ^{19}F-NMR analysis of the compound (I) of the present invention wherein R^3 is hydrogen, the fluorine substituent on the vinyl group of the (*E,E*) isomer is shown as a doublet having a coupling constant of 5.5 Hz at –83.3 ppm, while that of the (*E,Z*) isomer is represented by a doublet having a coupling constant of 28.6 Hz at –83.1 ppm; and, the (*E,E*) to (*E,Z*) isomer ratio is also confirmed to be about 2:1 from integration on the ^{19}F-NMR spectroscopy.

According to the ^{19}F-NMR analysis data of the compound of formula (I) of the present invention wherein R^3 is CF_3, the vinyl fluorine and the fluorine of CF_3 of the

(*E,E*) isomer are, respectively, a quartet having a coupling constant of 12.2 Hz at -75.9 ppm and a doublet having a coupling constant of 12.3 Hz at -58.7 ppm, while those of the (*E,Z*) isomer are, respectively, a quartet having a coupling constant of 23.9 Hz at –76.3 ppm and a doublet having a coupling constant of 24.7 Hz at -58.5 ppm. The (*E,E*) to (*E,Z*) isomer ratio is about 1:2 based on the integration of fluorine peaks.

The compound of the present invention has a broad spectrum of fungicidal activity against various plant pathogenic fungus, e.g. *Pyricularia oryzae* Carvara KA301 which causes Rice Blast, *Rhizoctonia solani* AG-1 which causes Rice Sheath Blight, *Botrytis cinerae* which causes Cucumber Gray Mold Rot, *Phytophthora infestans* which causes Tomato Late Blight, *Puccinia recondita* which causes Wheat Leaf Rust and *Erysiphe graminis* which causes Barley Powdery Mildew.

Accordingly, the present invention also includes within its scope fungicidal compositions comprising one or more of the compounds of formula (I) or stereoisomer thereof as an active ingredient, in association with fungicidally acceptable carriers.

The fungicidal compositions of the invention may be formulated in various forms such as an emulsion, aqueous dispersion, powder and granules which may contain conventional additives. The compound of the formula (I) may be used in an amount of 10 to 90 % on the basis of the weight of an emulsion or aqueous dispersion, and 0.1 to 10% on the basis of the weight of granules.

Fungicidally acceptable carrier that may be used in the present invention is a liquid carrier, e.g., water, an alcohol(ethanol, ethylene glycol, glycerine), ketone(acetone, methylethylketone), ether(dioxane, tetrahydrofuran, cellosolve), aliphatic hydrocarbon(gasoline, Kerosene), halogenated hydrocarbon(chloroform, carbon tetrachloride), amide(dimethylformamide), ester(ethyl acetate, butyl acetate, fatty glycerine ester) and acetonitrile; and a solid carrier, e.g., mineral particle(Kaoline, clay, bentonite, dolomite, talc, silica, sand) and vegetable powder(shrubs).

The additive that may be used in the fungicidal composition of the present invention includes an emulsifier, adhesive, dispersion agent or permeating agent, e.g., nonionic, anionic or cationic interface active agent(fatty acid sodium salt, polyoxy alkyl ester, alkyl sulfonate ester). Further, an agrochemically active ingredient, e.g., an insecticide, herbicide, plant growth regulator, germicide, and fertilizer, may be added in the composition of the present invention.

The following Preparation and Examples are given for the purpose of illustration only and are not intended to limit the scope of the invention.

Preparation 1: Preparation of methyl (2*E*)-3-methoxy-2-(2'-bromomethyl) phenyl-2-propenoate (compound of formula (II-a))

Step 1: Preparation of methyl *o*-tolylacetate

30.0 g (0.2 mol) of *o*-tolyl acetic acid was dissolved in 100 ml of methanol, 5 ml of concentrated sulfuric acid was added thereto and the resulting solution was stirred with heating for 6 to 12 hours. The resulting solution was cooled and the solvent was removed under a reduced pressure to obtain a residue. The residue was washed twice with water and extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure. The residue thus obtained was subjected to column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 32.15 g (yield 98 %) of the title compound as a colorless liquid.s

^{1}H-NMR ($CDCl_3$, TMS) δ (ppm) 7.21-7.01(m, 4H), 3.61(s, 3H), 3.60(s, 2H), 2.35(s, 3H)

MS (m/e): 164(M+, 42), 133(100), 31(82)

Step 2: Preparation of methyl 3-hydroxy-2-(2'-methyl)phenyl-2- propenoate

24.6 g (0.15 mol) of the compound obtained in Step 1 and 24.3 g (0.45 mol) of sodium methoxide were added to 300 ml of toluene, and 27 g (0.45 mol) of methylformate was added dropwise thereto over a period of 1 hour while cooling and stirring. The resulting solution was stirred at room temperature for 12 hours and extracted twice or three times with water. The combined aqueous layer was acidified with concentrated hydrochloric acid and then extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and then the solvent was removed under a reduced pressure to obtain a residue. The residue was subjected to column chromatography using a mixture of *n*-hexane and ethyl acetate (9:1) as an eluent to obtain 27.36 g (yield 95 %) of the title compound as a colorless liquid.

^{1}H-NMR ($CDCl_3$, TMS) δ (ppm) 11.92(d, 1H), 7.32-7.01(m, 4H), 3.71(s, 3H), 2.21(s, 3H)

MS (m/e): 192(M+, 26), 160(52), 132(48), 84(100)

Step 3: Preparation of methyl 3-methoxy-2-(2'-methyl)phenyl-2- propenoate

19.2 g (0.1 mol) of the compound obtained in Step 2, 15.12 g (0.12 mol) of dimethyl sulfate and 13.82 g (0.1 mol) of potassium carbonate were added to 200 ml of acetone, and the resulting solution was stirred for 12 hours with heating. The solvent was removed under a reduced pressure and the residue was extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure to obtain a residue. The residue was subjected to column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 17.1 g (yield 83 %) of the title compound having two isomers as a colorless

liquid.

The title compound thus obtained was composed of 82% *E* isomer and 18% *Z* isomer.

< *E* isomer (upper spot)>

^{1}H-NMR ($CDCl_3$, TMS) δ (ppm) 7.51(s, 1H), 7.35-6.98(m, 4H), 3.79(s, 3H), 3.68(s, 3H), 2.21(s, 3H)

MS (m/e): 206(M^+, 10), 176(73), 117(100), 77(57)

< *Z* isomer (down spot)>

^{1}H-NMR ($CDCl_3$, TMS) δ (ppm) 7.34-6.98(m, 4H), 6.50(s, 1H), 3.85(s, 3H), 3.68(s, 3H), 2.21(s, 3H)

MS (m/e): 206(M^+, 8), 176(100), 117(92), 77(30)

These isomers were separated and the *E* isomer was used in the following step.

Step 4: Preparation of methyl (2*E*)-3-methoxy-2-(2'-bromomethyl)phenyl -2-propenoate

18.54 g (0.09 mol) of methyl (2 *E*)-3-methoxy-2-(2'-methyl)phenyl- 2-propenoate obtained in Step 3 and 16.0 g (0.09 mol) of *N*-bromosuccinimide were added to 100 ml of carbon tetrachloride. Then, 0.16 g (1 mmol) of 2,2'-azo-bis-isobutyronitrile was added thereto, and the resulting solution was stirred for 12 hours with heating. The resultant solution was cooled and filtered to remove succinimide. The solvent was removed under a reduced pressure and an oily residue thus obtained was subjected to column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 21.73 g (yield 85 %) of the title compound as a colorless solid.

Melting Point: 64-65 ℃

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 7.63(s, 1H), 7.51-7.09(m, 4H), 4.40(s, 2H), 3.82(s, 3H), 3.69(s, 3H)

MS (m/e): 284(M^+, 10), 253(12), 205(21), 173(38), 145(100)

Preparation 2: Preparation of methyl (2 *E*)-2-methoxyimino-2-(2'-bromomethyl)phenylacetate (compound of formula (II-b))

Step 1: Preparation of methyl 2-methylbenzoylformate

5.1 g (0.21 mol) of magnesium was placed in 300 ml of dry ether and 34.18 g (0.2 mol) of 2-bromotoluene was added dropwise thereto under a nitrogen atmosphere to prepare a Grignard reagent. The Grignard reagent solution was cooled to -78 ℃ and 23.6 g (0.2 mol) of dimethyl oxalate was added dropwise thereto. The resulting solution was stirred for 30 minutes, mixed with crushed ice, acidified with 20% hydrochloric acid and then extracted with ether. The organic layer was washed three

times with water, dried over magnesium sulfate, and the solvent was removed under a reduced pressure to obtain a residue. The residue was subjected to column chromatography using a mixture of *n*-hexane and ethyl acetate (9:1) as an eluent to obtain 24.2 g (yield 68 %) of the title compound as a colorless liquid.

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 7.88-7.01(m, 4H), 3.98(s, 3H), 2.65(s, 3H)

MS (m/e): 178(M^+, 21), 119(100), 91(71), 65(37)

Step 2: Preparation of methyl 2-methoxyimino-2-(2'-methyl)phenylacetate

8.35 g (0.1 mol) of O-methylhydroxylamine hydrochloride and 8.1 ml (0.1 mol) of pyridine were added to 100 ml of methanol, and then, 17.8 g (0.1 mol) of the compound obtained in Step 1 was added thereto. The resulting solution was stirred for 12 hours with heating and concentrated under a reduced pressure. The resultant solution was mixed with water and extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure to obtain a residue. The residue was subjected to column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 19.04 g (yield 92 %) of the title compound as a colorless liquid.

The title compound thus obtained was composed of 50% *Z* isomer and 50% *E* isomer. The Z isomer was a liquid and the *E* isomer was a solid obtained by recrystallization in n-hexane. The structure of *E* isomer was identified by X-ray crystallography.

< *Z* isomer(upper spot) >

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 7.41-7.15(m, 4H), 4.01(s, 3H), 3.85(s, 3H), 2.45(s, 3H)

MS (m/e): 207(M^+, 8), 176(41), 116(100), 89(62)

< *E* isomer(down spot)>

m.p.: 63-64 ℃

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 7.38-7.05(m, 4H), 4.04(s, 3H), 3.85(s, 3H), 2.19(s, 3H)

MS (m/e): 207(M^+, 11), 176(82), 116(100), 89(70)

The *E* isomer was employed in following step.

Step 3: Preparation of methyl (2 *E*)-2-methoxyimino-2-(2'-bromomethyl) phenylacetate

9.0 g (0.0435 mol) of methyl (2 *E*)-2-methoxyimino-2-(2'-methyl)-phenyl acetate obtained in Step 2 and 7.74 g (0.0435 mol) of *N*-bromosuccinimide were added to 50 ml of carbon tetrachloride, and then, 0.16 g (1 mmol) of 2,2'-azo-bis- isobutyronitrile was added thereto. The resulting solution was stirred for 12 hours with heating, solvent was

removed under a reduced pressure and obtained an oily residue which was subjected to silica gel column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 11.08 g (yield 90 %) of the title compound as a colorless liquid.

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 7.62-7.01(m, 4H), 4.39(s, 2H), 4.04(s, 3H), 3.85(s, 3H)

MS (m/e): 285(M^+, 46), 252(35), 175(100), 146(94), 116(78)

Preparation 3: Preparation of 3-benzyloxybenzaldoxime (compound of formula (III-a))

Step 1: Preparation of 3-benzyloxybenzaldehyde

24.2 g (0.2 mol) of 3-hydroxybenzaldehyde and 25.32 g (0.2 mol) of benzyl chloride were placed in 500 ml of acetone and 21.2 g (0.2 mol) of sodium carbonate was added thereto. The resulting solution was stirred for 12 to 24 hours with heating, cooled to room temperature. The solvent was removed under a reduced pressure and the residue thus obtained was washed with water and the extracted with ethyl acetate twice. The organic layer was dried and the solvent was removed under a reduced pressure to obtain a residue. The residue was subjected to column chromatography using a mixture of *n*-hexane and ethyl acetate (9:1) as an eluent to obtain 35.6 g (yield 84 %) of the title compound as a colorless liquid.

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 10.01(s, 1H), 7.67-7.18(m, 9H), 5.14(s, 2H)

MS (m/e): 212(M^+, 32), 121(73), 91(100)

Step 2: Preparation of 3-benzyloxybenzaldoxime

31.8 g (0.15 mol) of the compound obtained in Step 1 and 11.47 g (0.165 mol) of hydroxylamine hydrochloride were placed in 200 ml of methyl alcohol, and 13.35 ml (0.165 mol) of pyridine was added thereto. The resulting solution was refluxed for 1 hour, and then was mixed with water and extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure to obtain a white-colored residue. The residue was washed with 100 ml of *n*-hexane to obtain 30.3 g (yield 89 %) of the title compound.

m.p.: 58-59 ℃

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 8.62(b, 1H), 8.18(s, 1H), 7.54-7.02(m.9H), 5.13(s, 2H)

MS (m/e): 227(M^+, 32), 91(100), 65(45)

Preparations 4 and 5: Preparation of 3-benzyloxyphenylmethyloxime (compound of formula (III-b)) and 3-benzyloxyphenyltrifluoromethyloxime (compound of formula (III-c))

The procedure of Preparation 3 was repeated except that 3-hydroxyphenyl methyl ketone and 3-hydroxyphenyl trifluoromethyl ketone were used in place of 3-hydroxybenzaldehyde, to obtain the title compounds.

The analysis data of the compounds prepared in Preparations 3 to 5 are listed in Table 1.

Table 1

Prep. No.	R^1	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	Mass (m/e) (M, int.)	M.P. (℃)	Yield (%)	Product
3	H	10,01(s, 1H), 7.67-7.18(m, 9H), 5.14(s, 2H)	212(32), 121(73), 91(100)	-	84	Intermediate
4	CH_3	7.82-7.02(m, 9H), 5.14(s, 2H), 2.28(s, 3H)	226(37), 135(63), 91(100)	-	86	
5	CF_3	8.10-7.01(m, 9H), 5.15(s, 2H)	280(42), 211(23), 91(100), 65(47)	-	74	
3	H	8.62(b, 1H), 8.18(s, 1H), 7.54-7.02(m, 9H), 5.13(s, 2H)	227(32), 91(100), 65(45)	58-59	89	Compound of formula (III)
4	CH_3	8.62(b, 1H), 7.51- 6.96(m, 9H), 5.10(s, 2H), 2.29(s, 3H)	241(35), 91(100), 65(45)	61-62	84	
5	CF_3	9.02(b, 1H), 7.61- 6.94(m, 9H), 5.12(s, 2H)	295(48), 91(100), 65(18)	72-73	82	

Preparation 6: Preparation of methyl (2*E*)-3-methoxy-2-[2-(((3-benzyloxyphenyl)imino)oxy)methylphenyl]-propenoate (compound of formula (IV-a))

5.7 g (0.02 mol) of methyl (2*E*)-3-methoxy-2-(2'-bromomethyl)phenyl-2-propenoate and 4.54 g (0.02 mol) of 3-benyloxybenzaldoxime prepared in preparation 3 were placed in 50 ml of acetone and 2.76 g (0.02 mol) of potassium carbonate was added thereto. The resulting solution was refluxed for 24 hours and cooled to room temperature, and the solvent was removed under a reduced pressure. The residue thus obtained was mixed with water and extracted with ethyl acetate with three times. The organic layer was dried over magnesium sulfate and filtered. The filtrate was subjected to column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 5.86 g (yield 68 %) of the title compound as a brown liquid.

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 8.01(s, 1H), 7.52(s, 1H), 7.51-6.82(m, 13H), 5.18(s, 2H), 5.04(s, 2H), 3.73(s, 3H), 3.61(s, 3H)

MS (m/e): 431(M^+, 21), 205(39), 189(50), 145(100), 91(67)

Preparations 7 to 11: Preparation of the compounds of formulas (IV-b) to (IV-f) as intermediates

The procedure of Preparation 6 was repeated except that methyl (2*E*)-3-methoxy-2-(2'-bromomethyl)phenyl-2-propenoate obtained in Preparation 1 and methyl (2*E*)-2-methoxyimino-2-(2'-bromomethyl)phenylacetate obtained in Preparation 2, and the oxime compounds obtained in Preparations 3 to 5 to obtain the compounds of formulas (IV-b) to (IV-f) as intermediates.

The analysis data of the compounds prepared in Preparations 6 to 11 are listed in Table 2.

Table 2

Prep. No.	X	R^1	^{1}H-NMR($CDCl_3$, TMS)δ (ppm)	Mass(m/e)(M, int.)	Yield (%)
6	CH	H	8.01(s, 1H), 7.52(s, 1H), 7.51-6.82(m, 13H), 5.18(s, 2H), 5.04(s, 2H), 3.73(s, 3H), 3.61(s, 3H)	431(21), 205(39), 189(50), 145(100), 91(67)	68
7	CH	CH_3	7.59(s, 1H), 7.57-7.12(m, 13H), 5.17(s, 2H), 5.03(s, 2H), 3.79(s, 3H), 3.67(s, 3H), 2.20(s, 3H)	445(37), 205(47), 189(37), 145(100), 91(87)	64
8	CH	CF_3	7.57(s, 1H), 7.48-7.01(m, 13H), 5.19(s, 2H), 5.01(s, 2H), 3.74(s, 3H), 3.62(s, 3H)	499(33), 278(57), 205(76), 189(66), 145(100), 91(89)	77
9	N	H	8.06(s, 1H), 7.55-6.99(m, 13H), 5.17(s, 2H), 5.09(s, 2H), 3.91(s, 3H), 3.71(s, 3H)	432(27), 401(38), 227(51), 206(47), 84(100), 47(79)	67
10	N	CH_3	7.64-6.87(m, 13H), 5.16(s, 2H), 5.05(s, 2H), 3.92(s, 3H), 3.73(s, 3H), 2.14(s, 3H)	446(31), 227(57), 206(53), 84(100)	61
11	N	CF_3	7.71-6.90(m, 13H), 5.18(s, 2H), 5.04(s, 2H), 3.91(s, 3H), 3.72(s, 3H)	500(43), 278(51), 222(64), 107(100)	72

Preparation 12: Preparation of methyl (2*E*)-3-methoxy-2-[2-(((3-hydroxyphenyl)imino)oxy)methylphenyl]-propenoate (compound of formula (V-a-1))

5.17 g (0.012 mol) of methyl (2*E*)-3-methoxy-2-[2-(((3-benzyloxyphenyl)imino)oxy)methylphenyl]-propenoate was dissolved in 50 ml of methyl alcohol, and a catalytic amount (25 mg, 0.01 mmol) of 5% palladium on

activated carbon was added thereto. The resulting mixture was reacted with stirring for 6 hours under a hydrogen pressure in a hydrogenation reactor. The reaction mixture solution was filtered to remove the activated carbon components, and the solvent was removed under a reduced pressure. The residue thus obtained was subjected to column chromatography using a mixture of *n*-hexane and ethyl acetate (2:1) as an eluent to obtain 3.64 g (yield 89 %) of the title compound as a brown liquid.

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 8.02(s, 1H), 7.54(s, 1H), 7.53-6.84(m, 8H), 5.18(s, 2H), 6.48(b, 1H), 5.14(s, 2H), 3.78(s, 3H), 3.67(s, 3H)

MS (m/e): 341(M^+, 41), 250(37), 189(57), 145(100), 103(20)

Preparations 13 to 17: Preparation of the compounds of formulas (V-a-2) to (V-a-6) as intermediates

The procedure of Preparation 12 was repeated using the intermediates obtained in Preparations 7 to 11 to obtain the phenolic ester compounds of formulas (V-a-2 to (V-a-6) as intermediates.

The analysis data of the compounds prepared in Preparations 12 to 17 are listed in Table 3.

Table 3

Prep. No.	X	R^1	^{1}H-NMR($CDCl_3$, TMS)δ (ppm)	Mass(m/e)(M, int.)	Yield (%)
12	CH	H	8.02(s, 1H), 7.54(s, 1H), 7.53-6.84(m, 8H), 6.48(b, 1H), 5.14(s, 2H), 3.78(s, 3H), 3.67(s, 3H)	341(41), 250(37), 189(57), 145(100), 103(20)	89
13	CH	CH_3	7.59(s, 1H), 7.53-6.98(m, 8H), 5.71(b, 1H), 5.13(s, 2H), 3.79(s, 3H), 3.67(s, 3H), 2.19(s, 3H)	355(33), 205(35), 189(37), 145(100), 134(69)	83
14	CH	CF_3	7.58(s, 1H), 7.54-6.81(m, 8H), 6.55(b, 1H), 5.16(s, 2H), 3.78(s, 3H), 3.66(s, 3H)	409(31), 221(38), 205(62), 189(99), 145(100), 131(38)	85
15	N	H	7.98(s, 1H), 7.56-6.77(m, 8H), 6.54(b, 1H), 5.11(s, 2H), 3.91(s, 3H), 3.72(s, 3H)	342(32), 206(76), 138(100), 59(46)	87
16	N	CH_3	7.50-6.78(m, 8H), 6.55(b, 1H), 5.10(s, 2H), 3.99(s, 3H), 3.76(s, 3H), 2.14(s, 3H)	356(43), 222(30), 131(75), 116(100), 59(42)	87
17	N	CF_3	7.61-6.81(m, 8H), 6.38(b, 1H), 5.12(s, 2H), 3.98(s, 3H), 3.77(s, 3H)	410(37), 222(38), 206(33), 131(65), 116(100), 59(62)	80

Preparation 18: Preparation of *N*-methyl (2E)-3-methoxy-2-[2-(((3-hydroxyphenyl)imino)oxy)methylphenyl]-propenamide (compound of formula (V-b-1))

3.41 g (0.01 mol) of methyl (2E)-3-methoxy-2-[2-(((3-hydroxyphenyl)imino)oxy)methylphenyl]-propenoate was dissolved in 50 ml of methyl alcohol, and 40 ml of 40% methylamine aqueous solution was added thereto. The resulting solution was stirred for 12 hours, and the solvent was removed under a reduced pressure. The residue thus obtained was extracted with ethyl acetate. The organic layer was dried and the solvent was removed under a reduced pressure. The residue thus obtained was subjected to column chromatography using a mixture of *n*-hexane and ethyl acetate (2:1) as an eluent to obtain 2.90 g (yield 85 %) of the title compound as a brown liquid.

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 8.01(s, 1H), 7.55(s, 1H), 7.54-6.88m, 8H), 6.53(b, 1H), 6.34(b, 1H), 5.142s, 2H), 3.81(s, 3H), 2.79(d, 3H)

MS (m/e): 340(M^+, 38), 188(100), 144(72)

Preparations 19 to 23: Preparation of the compounds of formulas (V-b-2) to (V-b-6)

The procedure of Preparation 18 was repeated using the intermediates obtained in Preparations 13 to 17 to obtain the phenolic amide compounds of formulas (V-b-2 to (V-b-6) as intermediates.

The analysis data of the compounds prepared in Preparations 18 to 23 are listed in Table 4.

Table 4

Prep. No.	X	R^1	^{1}H-NMR($CDCl_3$, TMS)δ (ppm)	Mass(m/e)(M, int.)	Yield (%)
18	CH	H	8.01(s, 1H), 7.55(s, 1H), 7.54-6.88(m, 8H), 6.53(b, 1H), 6.34(b, 1H), 5.12(s, 2H), 3.81(s, 3H), 2.79(d, 3H)	340(38), 188(100), 144(72)	85
19	CH	CH_3	7.57(s, 1H), 7.56-6.38(m, 8H), 6.23(b, 1H), 5.12(s, 2H), 5.87(b, 1H), 5.17(s, 2H), 3.79(s, 3H), 2.83(d, 3H), 1.18(s, 3H)	354(26), 188(100), 144(58), 65(49)	87
20	CH	CF_3	7.55(s, 1H), 7.54-6.88(m, 8H), 6.27(b, 1H), 5.12(s, 2H), 5.37 (b, 1H), 5.13(s, 2H), 3.80(s, 3H), 2.86(d, 3H)	408(37), 188(100), 144(64)	83
21	N	H	8.00(s, 1H), 7.57-7.08(m, 8H), 6.74(b, 1H), 6.48(b, 1H), 5.12(s, 2H), 5.09(s, 2H), 3.93(s, 3H),2.86(d, 3H)	341(44), 241(37), 222(870), 132(63), 58(100)	84
22	N	CH_3	7.58-7.01(m, 8H), 6.82(b, 1H), 6.54(b, 1H), 5.12(s, 2H), 5.08 (s, 2H), 3.95(s, 3H), 2.81(d, 3H), 2.13(s, 3H)	355(51), 221(50), 132(82), 116(99), 58(100)	87
23	N	CF_3	7.64-6.92(m, 8H), 6.80(b, 1H), 6.42(b, 1H), 5.12(s, 2H), 5.10 (s, 2H), 3.83(s, 3H), 2.84(d, 3H)	409(42), 379(64), 321(100), 132(78), 88(62)	86

Preparation 24: Preparation of 2,2-difluorostyrene (compound of formula (VI-a))

Step 1: Preparation of 2,2,2-trifluoromethyl phenyl ketone

5.1 g of magnesium(0.21 mol) was placed in 300 ml of dry diethyl ether and 31.4 g of bromobenzene (0.2 mol) was added dropwise thereto under a nitrogen atmosphere to prepare a Grignard reagent. The Grignard reagent solution was cooled to -78 ℃ and 28.4 g of ethyl trifluoroacetate (0.2 mol) was added dropwise thereto. The resulting solution was stirred for 1 hour, mixed with an crushed ice, acidified with a concentrated hydrochloric acid and then extracted three times with diethyl ether. The organic layer was dried over magnesium sulfate and the solvent was removed under a

reduced pressure to obtain a residue. The residue was distilled at 64 to 65 ℃/33 mmHg to obtain 24.74 g (yield 71%) of the title compound as a colorless oil.

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 7.52-7.12(m, 5H)

MS (m/e): 174(M^+, 21), 105(100), 77(82), 69(54)

Step 2: Preparation of 1-hydroxy-2,2,2-trifluoroethylbenzene

12.2 g (0.07 mol) of compound obtained in Step 1 was dissolved in 150 ml of methanol and 1.32 g (0.035 mol) of sodium borohydride was added dropwise thereto for 30 minutes. The resulting solution was stirred at room temperature for 2 hours and the solvent was removed. Ethyl acetate was added thereto and the resultant solution was washed three times with water. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure to obtain a residue. The residue was was distilled at 50 to 51 ℃/1 mmHg to obtain 12.07 g (yield 98%) of the title compound as a colorless liquid.

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 7.54-7.13(m, 5H), 4.87(q, 1H), 4.29(brs, 1H)

MS (m/e): 176(M^+, 39), 107(26), 79(91)

Step 3: Preparation of 1-chloro-2,2,2-trifluoroethylbenzene

11.97 g (0.068 mol) of the compound obtained in Step 2 and 83 g (0.7 mol) of thionyl chloride were added to 100 ml of toluene and the mixture was stirred with heating for 12 hours. The resulting solution was cooled and washed with water. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure to obtain a residue. The residue was subjected to silica gel column chromatography using *n*-hexane as an eluent to obtain 9.9 g (yield 72%) of the title compound as a colorless liquid.

^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 7.62-7.15(m, 5H), 5.10(q, 1H)

MS (m/e): 194(M^+, 94), 125(100), 83(30), 44(81)

Step 4: Preparation of 2,2-difluorostyrene

9.7 g (0.05 mol) of the compound obtained in Step 3 was dissolved in 50 ml of dry tetrahydrofuran, and then, 3.27 g (0.05 mol) of activated zinc was added thereto. The resulting solution was refluxed for 12 hours while stirring and heating. The resulting solution was cooled and filtered to remove precipitated salts. The solvent was removed under a reduced pressure and the residue was distilled at 58 to 59 ℃/49 mmHg to obtain 6.09 g (yield 87%) of the title compound as a colorless oil.

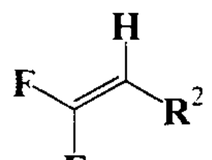
^{1}H -NMR ($CDCl_3$, TMS) δ (ppm) 7.45-7.10(m, 5H), 5.20(dd, 1H, *J*=26Hz, *J*=4Hz)

MS (m/e): 140(M^+, 100), 120(26), 84(16), 44(32)

Preparations 25 to 40

The procedure of Preparation 24 was repeated to obtain various fluorinated vinyl compounds of formula (VI-a). The ^{1}H-NMR and MS analysis data of the compounds obtained in Preparations 24 to 40 are shown in the following Table 5.

Table 5: Substituted 2,2-difluorostyrene



(VI-a)

Prep. No.	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e)(m/e) (M, int)	Yield (%)	Boiling Point (mmHg)
24	C_6H_5-	7.45-7.10(m, 5H), 5.20(dd, 1H, *J* = 26Hz, 4Hz)	140(100), 120(26), 84(16), 44(32)	87	58-59 (49)
25	3-CH_3-C_6H_4-	7.48-6.92(m, 4H), 5.21(dd, 1H, *J* = 26Hz, 4Hz), 2.28(s, 3H)	154(39), 135(29), 64(41), 45(100)	87	Column
26	4-CH_3-C_6H_4-	7.45-6.89(m, 4H), 5.26(dd, 1H, *J* = 26Hz, 4Hz), 2.27(s, 3H)	154(100), 135(62), 45(54)	86	Column
27	4-C_2H_5-C_6H_4-	7.39-7.06(m, 4H), 5.25(dd, 1H, *J* = 26Hz, 4Hz), 2.62(q, 2H), 1.21(t, 3H)	168(33), 153(100), 133(25), 84(34)	75	Column
28	4-*n*-C_4H_9-C_6H_4-	7.24-7.10(m, 4H), 5,20(dd, 1H, *J* = 26Hz, 4Hz), 2.54(t, 2H), 1.62-1.21(m, 4H), 0.92(t, 3H)	196(100), 177(64), 158(32)	77	Column
29	3,4-$(CH_3)_2$-C_6H_3-	7.32-7.02(m, 3H), 5.27(dd, 1H, *J* = 26Hz, 4Hz), 2.22(s, 6H)	168(32), 133(49), 44(100)	91	Column
30	3,5-$(CH_3)_2$-C_6H_3-	7.01-6.89(m, 3H), 5.20(dd, 1H, *J* = 26Hz, 4Hz), 2.24(s, 6H)	168(82), 153(46), 84(100), 62(59)	98	Column
31	3-CH_3O-C_6H_4-	7.50-6.69(m, 4H), 5.19(dd, 1H, *J* = 26Hz, 4Hz), 3.79(s, 3H)	170(100), 140(36), 127(42), 77(24)	98	33-34(1)
32	4-CH_3O-C_6H_4-	7.51-6.69(m, 4H), 5.29(dd, 1H, *J* = 26Hz, 4Hz), 3.86(s, 3H)	170(100), 155(68), 127(92), 84(21)	80	Column
33	4-C_2H_5O-C_6H_4-	7.49-6.81(m, 4H), 5.26(dd, 1H, *J* = 26Hz, 4Hz), 4.10(q, 2H),	184(82), 127(100), 53(40)	79	Column

Table 5 (Continued)

Prep. No.	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e)(m/e) (M, int)	Yield (%)	Boiling Point (mmHg)
34	3,4-OCH_2O-C_6H_3-	7.81-7.28(m, 3H), 6.08(s, 2H), 5.27(dd, 1H, *J* = 26Hz, 4Hz)	184(48), 165(100), 146(39)	83	column
35	3-Cl-C_6H_4-	7.36-7.12(m, 4H), 5.26(dd, 1H, *J* = 26Hz, 4Hz)	174(94), 119(38), 84(100), 48(93)	95	column
36	4-Cl-C_6H_4-	7.48-7.29(m, 4H), 5.23(dd, 1H, *J* = 26Hz, 4Hz)	174(58), 139(36), 119(29), 84(100), 49(56)	81	column
37	3-F-C_6H_4-	7.59-6.78(m, 4H), 5.21(dd, 1H, *J* = 26Hz, 4Hz)	158(21), 84(100), 47(42)	75	column
38	4-F-C_6H_4-	7.76-6.91(m, 4H), 5.25(dd, 1H, *J* = 26Hz, 4Hz)	158(100), 39(27)	88	column
39	3-CH_3-4-Cl-C_6H_3-	7.34-7.02(m, 3H), 5.24(dd, 1H, *J* = 26Hz, 4Hz), 2.34(s, 3H)	188(100), 153(71), 133(30)	87	column
40	(naphthalen-2-yl) $C_{10}H_7$-2-yl-	8.13-7.45(m, 7H), 5.81(dd, 1H, *J* = 26Hz, 4Hz)	190(89), 170(100), 138(28), 85(44)	90	column

Preparation 41: Preparation of 2,2-difluoro-1-trifluoromethylstyrene (compound of formula (VI-b))

26.2 g (0.1 mol) of triphenylphosphine was dissolved in 100 ml of dry tetrahydrofuran and 25.2 g (0.12 mol) of dibromodifluoro- methane was added dropwise thereto under a nitrogen atmosphere at a temperature below 10 ℃. The resulting solution was stirred for 30 minutes and thereto was added 8.71 g (0.05 mol) of the compound obtained in Step 1 of Preparation 24. The resultant solution was refluxed with heating for 48 hours, cooled and distilled under a reduced pressure. The obtained oil was redistilled at a temperature of 51 to 52 ℃/44 mmHg to obtain 7.07 g (yield 68%) of the title compound as a colorless liquid.

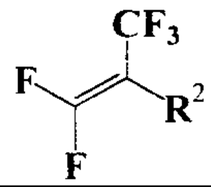
^{1}H-NMR ($CDCl_3$, TMS) δ (ppm) 7.59-7.31(m, 5H) 3.79(s, 3H)

MS (m/e): 208(M^+, 48), 84(83), 43(100)

Preparations 42 to 59

The procedure of Preparation 41 was repeated using corresponding halides in place of 4-bromobenzene to obtain various compounds of formula (VI-b). The ^{1}H -NMR and MS data of these compounds are listed in Table 6.

Table 6: Substituted 2,2-difluoro-1-trifluoromethylstyrene



(VI-b)

Prep. No.	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e)(m/e) (M, int)	Yield (%)	Boiling Point (mmHg)
41	C_6H_5-	7.59-7.31(m, 5H)	208(48), 84(83), 43(100)	68	51-52 (44)
42	3-CH_3-C_6H_4-	7.46-6.98(m, 4H), 2.43(s, 3H)	222(20), 203(70), 134(100)	45	column
43	4-CH_3-C_6H_4-	7.32-7.18(m, 4H), 2.45(s, 3H)	222(64), 203(23), 134(100)	62	column
44	4-C_2H_5-C_6H_4-	7.38-7.25(m, 4H), 2.68(q, 2H), 1.19(t, 3H)	236(20), 145(100), 90(54)	62	column
45	4-*n*-C_4H_9-C_6H_4-	7.32-7.25(m, 4H), 2.69(t, 2H), 2.01-1.23(m, 4H), 1.09(t, 3H)	264(30), 221(37), 151(36), 84(100), 57(50)	58	column
46	3,4-$(CH_3)_2$-C_6H_3-	7.28-7.02(m, 3H), 2.38(s, 3H), 2.32(s, 3H)	236(18), 84(33), 45(100)	78	column
47	3,5-$(CH_3)_2$-C_6H_3-	7.32-7.12(m, 3H), 2.41(s, 6H)	236(29), 217(68), 148(100), 45(92)	98	column
48	3-CF_3-C_6H_4-	7.82-7.18(m, 4H)	276(52), 257(92), 188(100)	52	column
49	4-CF_3-C_6H_4-	7.81-7.42(m, 4H)	276(42), 257(78), 188(95), 107(100)	64	column
50	3-CH_3O-C_6H_4-	7.48-6.87(m, 4H), 3.81(s, 3H)	238(42), 207(45), 139(100), 69(94)	54	75(10)

Table 6 (Continued)

Prep. No.	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e)(m/e) (M, int)	Yield(%)	Boiling Point (mmHg)
51	4-CH_3O-C_6H_4-	7.48-6.79(m, 4H), 3.79(s, 3H)	238(69), 195(14), 145(35), 74(33), 59(100)	79	72-74 (10)
52	4-C_2H_5O-C_6H_4-	7.51-6.85(m, 4H), 4.12(q, 2H), 1.29(t, 3H)	252(47), 233(100), 84(64)	73	column
53	3,4-OCH_2O-C_6H_3-	7.01-6.79(m, 3H), 6.01(s, 2H)	252(46), 233(63), 164(82), 69(100)	72	column
54	3-Cl-C_6H_4-	7.54-7.23(m, 4H)	242(26), 223(72), 188(49), 69(100)	63	column
55	4-Cl-C_6H_4-	7.56-7.21(m, 4H)	242(35), 174(70), 139(100), 69(79)	45	58(10)
56	3-F-C_6H_4-	7.53-6.96(m, 4H)	226(52), 207(25), 84(100)	54	column
57	4-F-C_6H_4-	7.52-6.83(m, 4H)	226(20), 84(100)	63	column
58	4-Br-C_6H_4-	7.81-7.19(m, 4H)	286(100), 207(86), 138(66), 69(57)	43	column
59	3,5-Cl_2-C_6H_3-	7.57-7.19(m, 3H)	276(100), 241(45)	84	85(10)

Preparation 60: Preparation of 1-[3-(1-fluoro-2-phenyl)ethenyloxy]phenylmethyl oxime (compound of formula (VII-a))

Step 1: Preparation of 1-[3-(1-fluoro-2-phenyl)ethenyloxy]phenyl-2-ethanone

In a dried vessel, 68 g (0.5 mol) of 3-hydroxyacetophenone was added to 400 ml of methylethylketone and thereto was added 83 g of potassium carbonate (0.6 mol). The resulting solution was stirred for 30 minutes and 70 g (0.5 mol) of 2,2-difluorostyrene obtained in Preparation 24 was added slowly thereto. The resultant solution was stirred for 24 hours with heating and filtered to remove solids. The filtrate was concentrated, and the residue was washed with water and extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure. The residue was distilled at 188 to 190 ℃/1 mmHg to obtain 118.5 g (yield 92%) of the title compound as a colorless liquid.

^{1}H-NMR($CDCl_3$, TMS)δ (ppm) 7.77-7.15(m, 9H), 5.75(d, 0.8H(*E*)), 5.38(d, 0.2H(*Z*)), 2.59(s, 3H)

MS (m/e): 256(M^+, 100), 165(27), 109(62), 91(32), 43(72)

Step 2: Preparation of 1-[3-(1-fluoro-2-phenyl)-ethenyloxy]phenylmethyloxime

76.8 g (0.3 mol) of the compound obtained in Step 1 above and 22.2 g (0.32 mol) of hydroxylamine hydrochloride were added to 500 ml of methanol, and thereto was added 25.9 ml (0.32 mol) of pyridine. The resulting solution was stirred at room temperature for 30 minutes and concentrated to remove the solvent. The residue was mixed with water and extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure. The residue was was subjected to silica gel column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 76.4 g (yield 94%) of the title compound as a colorless liquid.

^{1}H-NMR($CDCl_3$, TMS)δ (ppm) 8.26(br.s, 1H), 7.46-7.16(m, 9H), 5.74(d, 0.8H(*E*)), 5.37(d, 0.2H(*Z*)), 2.29(s, 3H)

MS (m/e): 271(M^+, 100), 118(42), 109(41), 90(22)

Preparation 61: Preparation of 1-[3-(1,3,3,3-tetrafluoro-2-phenyl)-1-propenyloxy] phenylmethyloxime (compound of formula (VII-b))

The procedure of Preparation 60 was repeated using 2,2-difluoro-1-trifluoromethylstyrene obtained in Preparation 41, in place of 2,2-difluorostyrene, to obtain the title compound as a colorless liquid.

^{1}H-NMR($CDCl_3$, TMS)δ (ppm) 8.44(br.s, 1H), 7.51-6.99(m, 9H), 2.25(s, 3H)

MS (m/e): 339(M^+, 100), 186(37), 134(40), 117(26), 89(36)

Example 1: Preparation of methyl (2*E*)-3-methoxy-2-{2'-[[[3"-(1'"-fluoro-2'"-phenyl-1'"-ethenyloxy)phenyl]imino]oxy]methylphenyl}propenoate (Compound 1)

341 mg (1 mmol) of the compound obtained in Preparation 12 was added to 10 ml of acetonitrile and 40 mg of sodium hydride (1 mmol) dispersed in mineral oil(60%) was added thereto under a nitrogen atmosphere. The resulting solution was stirred for 30 minutes and 140 mg (1 mmol) of the compound obtained in Preparation 24 was added slowly thereto. The resultant solution was stirred for 4 hours with heating, mixed with water and extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure. The residue was subjected to silica gel column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 420 mg (yield 91%) of the title compound as a colorless liquid.

^{1}H-NMR($CDCl_3$, TMS)δ (ppm) 8.04(s, 1H), 7.58(s, 1H), 7.50-7.08(m, 13H), 5.68(d, 1H), 5.09(s, 2H), 3.79(s, 3H), 3.65(s, 3H)

^{19}F-NMR($CDCl_3$, $CFCl_3$)δ (ppm): -83.066(d, 1F, *J*=28.614Hz, *Z* isomer), -83.344(d, 1F, *J*=5.55Hz, *E* -isomer)

MS(m/e): 461(M^+, 48), 205(33), 189(63), 145(100), 103(15)

Example 16: Preparation of *N*-methyl (2 *E*)-3-methoxy-2-{2'-[[[3"-(1'"-fluoro-2'"-phenyl-1'"-ethenyloxy)phenyl]imino]oxy]methylphenyl}propenamide (Compound 16)

170 mg (0.5 mmol) of the compound obtained in Preparation 18 was added to 10 ml of acetonitrile and 40 mg of sodium hydride (1 mmol) dispersed in mineral oil (60%) was added thereto under a nitrogen atmosphere. The resulting solution was stirred for 30 minutes and 70 mg (0.5 mmol) of the compound obtained in Preparation 24 was added slowly thereto. The resultant solution was stirred for 4 hours with heating, mixed with water and extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure. The residue was subjected to silica gel column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 221 mg (yield 96%) of the title compound as a colorless liquid.

^{1}H-NMR($CDCl_3$, TMS)δ (ppm) 8.07(s, 1H), 7.61(s, 1H), 7.59-6.89(m, 13H), 5.71(d, 1H), 5.17(s, 2H), 4.14(br, 1H), 3.64(s, 3H), 2.96(d, 3H)

MS(m/e): 460(M^+, 28), 188(100), 149(53)

Example 40: Preparation of methyl (2 *E*)-3-methoxy-2-{2'-[[[3"-(1'"-fluoro-2'"-phenyl-1'"-ethenyloxy)phenyl]methylimino]oxy]methylphenyl}propenoate (Compound 40)

28.4 g (0.1 mol) of methyl (2*E*)-3-methoxy-2-(2'-bromomethyl) phenyl-2-propenoate obtained in Preparation 1 (the compound of formula (II-a)) and 27.1 g (0.1 mol) of the compound obtained in Preparation 60 (the compound of formula (VII-a)) were added to 200 ml of acetone, and thereto was added 13.8 g (0.1 mol) of potassium carbonate. The resulting solution was stirred for 24 hours with heating, and cooled to room temperature. The solvent was removed under a reduced pressure, and the residue was mixed with water and extracted three times with 50 ml of ethyl acetate. The organic layer was dried over magnesium sulfate and filtered. The filtrate was concentrated and the residue was subjected to silica gel column chromatography using a mixture of *n*-hexane and ethyl acetate (9:1) as an eluent to obtain 33.8 g (yield 71%) of the title compound as a colorless liquid.

^{1}H-NMR($CDCl_3$, TMS)δ (ppm) 7.48(s, 1H), 7.42-6.87(m, 13H), 5.67(d, 1H), 5.20(s, 2H), 3.78(s, 3H), 3.65(s, 3H), 2.24(s, 3H)

MS (m/e): 475(M^+, 11), 205(35), 189(17), 145(100), 109(31)

Example 64: Preparation of methyl (2 *E*)-2-methoxyimino-2-{2'-[[[3"-(1'"-fluoro-2'"-phenyl-1'"-ethenyloxy)phenyl]methylimino]oxy]methylphenylacetate (Compound 64)

The procedure of Example 40 was repeated using methyl (2*E*)-2-methoxyimino-2-(2'-bromomethyl)phenylacetate obtained in Preparation 2, in place of the compound obtained in Preparation 1, to obtain the title compound as a colorless liquid.

^{1}H-NMR($CDCl_3$, TMS)δ (ppm) 7.49-7.16(m, 13H), 5.73(d, 1H), 5.15(s, 2H), 4.04(s, 3H), 3.82(s, 3H), 2.21(s, 3H)

MS (m/e): 476(M^+, 11), 131(68), 116(100), 59(44)

Example 77: Preparation of *N*-methyl (2 *E*)-2-methoxyimino-2-{2'-[[[3"-(1'"-fluoro-2'"-phenyl-1'"-ethenyloxy)phenyl]methylimino]oxy]methylphenylacetamide (Compound 77)

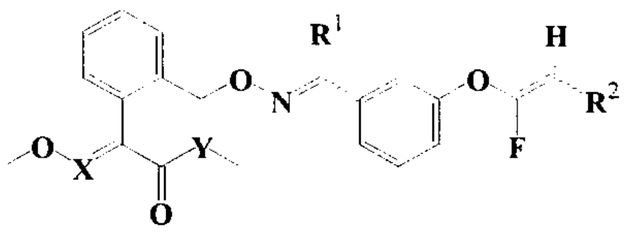
The procedure of Preparation 18 was repeated using Ccompound 64 obtained in Example 64, in place of the propenoate compound, to obtain the title compound as a colorless liquid.

^{1}H-NMR($CDCl_3$, TMS)δ (ppm) 7.47-7.15(m, 13H), 6.71(b, 1H), 5.70(d, 1H), 5.13(s, 2H), 3.93(s, 3H), 2.84(s, 3H), 2.18(s, 3H)

MS (m/e): 475(M^+, 11), 132(50), 116(68), 58(100)

The procedure of Example 1 or 16 was repeated using corresponding phenolic ester compounds or phenolic amide comounds of formula (V) and fluorinated vinyl comounds of formula (VI-a), or, alternatively, the procedure of Example 40, 64 or 77 was repeated using corresponding bromide compounds of formula (II) and olefin substituted oxime compounds of formula (VII-a), to obtain various compounds of formula (I), as listed in Table 7.

Table 7



Ex.No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
1	CH	O	H	C_6H_5	8.04(s, 1H), 7.58(s, 1H), 7.50~7.08(m, 13H), 5.68(d, 1H), 5.09(s, 2H), 3.79(s, 3H), 3.65(s, 3H)	461(48), 205(33), 189(63), 145(100), 103(15)
2	CH	O	H	3-CH_3-C_6H_4	8.04(s, 1H), 7.58(s, 1H), 7.52~6.97(m, 12H), 5.68(d, 1H), 5.10(s, 2H), 3.78(s, 3H), 3.64(s, 3H), 2.25(s, 3H)	475(30), 205(27), 189(51), 145(100), 123(22)
3	CH	O	H	4-CH_3-C_6H_4	8.05(s, 1H), 7.59(s, 1H), 7.52~7.01(m, 12H), 5.70(d, 1H), 5.11(s, 2H), 3.80(s, 3H), 3.66(s, 3H), 2.28(s, 3H)	475(42), 205(21), 189(32), 145(100)
4	CH	O	H	3,4-$(CH_3)_2$-C_6H_3	8.01(s, 1H), 7.60(s, 1H), 7.59~6.92(m, 11H), 5.65(d, 1H), 5.15(s, 2H), 3.77(s, 3H), 3.62(s, 3H), 2.20(s, 6H)	489(12), 205(20), 189(22), 145(100), 118(59)
5	CH	O	H	3,5-$(CH_3)_2$-C_6H_3	8.02(s, 1H), 7.55(s, 1H), 7.51~6.84(m, 11H), 5.64(d, 1H), 5.12(s, 2H), 3.77(s, 3H), 3.65(s, 3H), 2.26(s, 6H)	489(26), 205(22), 145(100), 137(25)
6	CH	O	H	4-C_2H_5-C_6H_4	8.06(s, 1H), 7.59(s, 1H), 7.55~7.03(m, 12H), 5.70(d, 1H), 5.16(s, 2H), 3.76(s, 3H), 3.67(s, 3H), 2.61(q, 2H), 1.24(t, 3H)	489(44), 205(62), 168(14), 145(100), 86(26)
7	CH	O	H	4-*n*-C_4H_9-C_6H_4	8.02(s, 1H), 7.59(s, 1H), 7.57~7.01(m, 12H), 5.70(d, 1H), 5.17(s, 2H), 3.79(s, 3H), 3.66(s, 3H), 2.58(t, 2H), 1.64~1.21(m, 4H), 0.92(t, 3H)	517(21), 149(54), 84(78), 84(78), 55(45), 43(100)
8	CH	O	H	3-CH_3O-C_6H_4	8.01(s, 1H), 7.60(s, 1H), 7.57~6.70(m, 12H), 5.65(d, 1H), 5.15(s, 2H), 3.81(s, 3H), 3.79(s, 3H), 3.64(s, 3H)	491(42), 205(26), 189(29), 145(100)

Table 7 (Continued)

Ex.No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M. int)
9	CH	O	H	4-CH_3O-C_6H_4	8.06(s, 1H), 7.60(s, 1H), 7.53~6.78(m, 12H), 5.69(d, 1H), 5.10(s, 2H), 3.81(s, 3H), 3.79(s, 3H), 3.67(s, 3H)	491(56), 205(19), 189(25), 145(100), 139(24)
10	CH	O	H	4-C_2H_5O-C_6H_4	8.02(s, 1H), 7.61(s, 1H), 7.59~6.78(m, 12H), 5.64(d, 1H), 5.16(s, 2H), 4.00(q, 2H), 3.79(s, 3H), 3.63(s, 3H), 1.40(t, 3H)	505(19), 145(100), 125(37)
11	CH	O	H	3-F-C_6H_4	8.00(s, 1H), 7.60(s, 1H), 7.58~6.80(m, 12H), 5.63(d, 1H), 5.19(s, 2H), 3.78(s, 3H), 3.62(s, 3H)	479(35), 205(32), 189(42), 144(100)
12	CH	O	H	4-F-C_6H_4	8.05(s, 1H), 7.58(s, 1H), 7.56~6.89(m, 12H), 5.68(d, 1H), 5.16(s, 2H), 3.79(s, 3H), 3.66(s, 3H)	479(21), 205(95), 190(23), 145(100)
13	CH	O	H	3-Cl-C_6H_4	8.06(s, 1H), 7.57(s, 1H), 7.55~7.00(m, 12H), 5.67(d, 1H), 5.14(s, 2H), 3.76(s, 3H), 3.64(s, 3H)	495(41), 205(97), 189(49), 145(100)
14	CH	O	H	4-Cl-C_6H_4	8.05(s, 1H), 7.58(s, 1H), 7.54~7.02(m, 12H), 5.65(d, 1H), 5.13(s, 2H), 3.78(s, 3H), 3.65(s, 3H)	495(43), 205(59), 189(98), 145(100), 131(27), 103(21)
15	CH	O	H	3,4-OCH_2O-C_6H_3	8.01(s, 1H), 7.61(s, 1H), 7.59~6.64(m, 11H), 5.86(s, 2H), 5.56(d, 1H), 5.15(s, 2H), 3.77(s, 3H), 3.64(s, 3H)	505(36), 205(20), 188(23), 144(100)
16	CH	NH	H	C_6H_5	8.07(s, 1H), 7.61(s, 1H), 7.59~6.89(m, 13H), 5.71(d, 1H), 5.17(s, 2H), 4.14(b, 1H), 3.64(s, 3H), 2.96(d, 3H)	460(28), 188(100), 149(53)
17	CH	NH	H	3-CH_3-C_6H_4	8.09(s, 1H), 7.62(s, 1H), 7.61~6.97(m, 12H), 5.67(d, 1H), 5.17(s, 2H), 4.13(b, 1H), 3.63(s, 3H), 2.97(d, 3H), 2.29(s, 3H)	474(40), 188(100), 144(46), 42(21)
18	CH	NH	H	3,5-$(CH_3)_2$-C_6H_3	8.09(s, 1H), 7.61(s, 1H), 7.59~6.81(m, 11H), 5.71(d, 1H), 5.16(s, 2H), 4.16(b, 1H), 3.61(s, 3H), 2.95(d, 3H), 2.23(s, 6H)	488(52), 188(100), 144(58), 137(17), 42(26)

Table 7 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
19	CH	NH	H	4-C_2H_5-C_6H_4	8.07(s, 1H), 7.62(s, 1H), 7.61~7.09(m, 12H), 5.68(d, 1H), 5.17(s, 2H), 4.17(b, 1H), 3.63(s, 3H), 2.98(d, 3H), 2.59(q, 2H), 1.24(t, 3H)	488(52), 188(100), 144(42), 44(37)
20	CH	NH	H	4-CH_3O-C_6H_4	8.08(s, 1H), 7.63(s, 1H), 7.61~6.72(m, 12H), 5.70(d, 1H), 5.16(s, 2H), 4.17(b, 1H), 3.78(s, 3H), 3.63(s, 3H), 2.97(d, 3H)	490(45), 188(100), 144(58), 44(25)
21	CH	NH	H	4-F-C_6H_4	8.07(s, 1H), 7.62(s, 1H), 7.60~6.87(m, 12H), 5.67(d, 1H), 5.17(s, 2H), 4.14(b, 1H), 3.64(s, 3H), 2.96(d, 3H)	478(62), 188(100), 144(61), 103(23)
22	CH	NH	H	3-Cl-C_6H_4	8.10(s, 1H), 7.62(s, 1H), 7.60~6.97(m, 12H), 5.63(d, 1H), 5.16(s, 2H), 4.15(b, 1H), 3.61(s, 3H), 2.95(d, 3H)	494(42), 188(100), 144(46)
23	CH	NH	H	3,4-OCH_2O-C_6H_3	8.08(s, 1H), 7.62(s, 1H), 7.61~6.68(m, 11H), 5.91(s, 2H), 5.62(d, 1H), 5.18(s, 2H), 4.12(b, 1H), 3.61(s, 3H), 2.97(d, 3H)	504(28), 445(34), 188(94), 144(100), 103(21)
24	N	O	H	C_6H_5	8.01(s, 1H), 7.53~7.01(m, 13H), 5.67(d, 1H), 5.11(s, 2H), 4.00(s, 3H), 3.83(s, 3H)	462(42), 131(66), 116(100), 59(61)
25	N	O	H	4-CH_3-C_6H_4	8.01(s, 1H), 7.50~7.06(m, 12H), 5.68(d, 1H), 5.10(s, 2H), 4.01(s, 3H), 3.82(s, 3H), 2.30(s, 3H)	476(53), 131(67), 116(100), 59(52)
26	N	O	H	4-CH_3O-C_6H_4	8.00(s, 1H), 7.62~6.81(m, 12H), 5.65(d, 1H), 5.09(s, 2H), 4.02(s, 3H), 3.82(s, 3H), 3.78(s, 3H)	492(98), 139(83), 131(100), 116(77), 59(64)
27	N	O	H	4-F-C_6H_4	8.01(s, 1H), 7.46~6.93(m, 12H), 5.67(d, 1H), 5.09(s, 2H), 4.02(s, 3H), 3.83(s, 3H)	480(42), 131(64), 116(100), 59(61)
28	N	O	H	4-Cl-C_6H_4	8.00(s, 1H), 7.46~7.13(m, 12H), 5.66(d, 1H), 5.10(s, 2H), 4.01(s, 3H), 3.82(s, 3H)	496(17), 131(61), 116(100), 59(52)

Table 7 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
29	N	O	H	3,4-OCH_2 O-C_6H_3	8.00(s, 1H), 7.50~6.71(m, 11H), 6.97(s, 2H), 5.65(d, 1H), 5.10(s, 2H), 4.02(s, 3H), 3.82(s, 3H)	506(23), 153(55), 131(52), 116(80), 59(100)
30	N	NH	H	C_6H_5	8.01(s, 1H), 7.69~6.90(m, 13H), 6.74(b, 1H), 5.65(d, 1H), 5.11(s, 2H), 3.94(s, 3H), 2.86(d, 3H)	461(38), 221(81), 204(64), 132(73), 116(100), 58(61)
31	N	NH	H	3-CH_3-C_6H_4	8.00(s, 1H), 7.67~6.94(m, 12H), 6.76(b, 1H), 5.68(d, 1H), 5.14(s, 2H), 3.92(s, 3H), 2.87(d, 3H), 2.30(s, 3H)	475(31), 221(86), 204(53), 132(99), 116(100), 58(73)
32	N	NH	H	3,4-$(CH_3)_2$-C_6H_3	8.01(s, 1H), 7.72~7.00(m, 11H), 6.76(b, 1H), 5.63(d, 1H), 5.14(s, 2H), 3.85(s, 3H), 2.87(d, 3H), 2.23(s, 6H)	489(42), 307(47), 149(100)
33	N	NH	H	4-C_2H_5-C_6H_4	8.01(s, 1H), 7.55~7.09(m, 12H), 6.75(b, 1H), 5.70(d, 1H), 5.09(s, 2H), 3.93(s, 3H), 2.87(d, 3H), 2.60(q, 2H), 1.20(t, 3H)	489(55), 221(21), 132(100), 116(88)
34	N	NH	H	4-*n*-C_4H_9-C_6H_4	8.01(s, 1H), 7.53~7.02(m, 12H), 6.76(b, 1H), 5.68(d, 1H), 5.12(s, 2H), 3.94(s, 3H), 2.87(d, 3H), 2.56(t, 2H), 1.67-1.21(m, 4H), 0.91(t, 3H)	517(35), 205(37), 132(89), 116(73), 58(100)
35	N	NH	H	3-CH_3O-C_6H_4	8.00(s, 1H), 7.55~6.96(m, 12H), 6.75(b, 1H), 5.64(d, 1H), 5.13(s, 2H), 3.86(s, 3H), 3.78(s, 3H), 2.87(d, 3H)	491(15), 221(57), 205(35), 132(85), 116(86), 58(100)
36	N	NH	H	4-C_2H_5O-C_6H_4	8.00(s, 1H), 7.57~6.84(m, 12H), 6.75(b, 1H), 5.65(d, 1H), 5.15(s, 2H), 4.00(q, 2H), 3.87(s, 3H), 2.86(d, 3H)	505(82), 205(39), 181(23), 132(100), 116(84), 58(88)
37	N	NH	H	4-F-C_6H_4	8.01(s, 1H), 7.58~6.87(m, 12H), 6.77(b, 1H), 5.64(d, 1H), 5.11(s, 2H), 3.90(s, 3H), 2.88(d, 3H)	479(43), 221(44), 132(100), 116(76), 58(46)
38	N	NH	H	3-Cl-C_6H_4	8.01(s, 1H), 7.58~7.02(m, 12H), 6.74(b, 1H), 5.63(d, 1H), 5.12(s, 2H), 3.88(s, 3H), 2.88(d, 3H)	495(27), 221(99), 205(39), 132(90), 116(100), 58(79)

Table 7 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
39	N	NH	H	3,4-OCH_2O-C_6H_3	8.02(s, 1H), 7.58~6.81(m, 11H), 6.76(b, 1H), 5.93(s, 2H), 5.66(d, 1H), 5.10(s, 2H), 3.94(s, 3H), 2.87(d, 3H)	505(63), 284(25), 153(39), 132(99), 116(100), 58(43)
40	CH	O	CH_3	C_6H_5	7.48(s, 1H), 7.42~6.87(m, 13H), 5.67(d, 1H), 5.20(s, 2H), 3.78(s, 3H), 3.65(s, 3H), 2.24(s, 3H)	475(11), 205(35), 189(17), 145(100), 109(31)
41	CH	O	CH_3	3-CH_3-C_6H_4	7.50(s, 1H), 7.48~6.92(m, 12H), 5.66(d, 1H), 5.19(s, 2H), 3.77(s, 3H), 3.67(s, 3H), 2.31(s, 3H), 2.23(s, 3H)	489(20), 206(40), 189(17), 145(100), 123(29)
42	CH	O	CH_3	4-CH_3-C_6H_4	7.50(s, 1H), 7.48~6.97(m, 12H), 5.64(d, 1H), 5.19(s, 2H), 3.75(s, 3H), 3.64(s, 3H), 2.31(s, 3H), 2.25(s, 3H)	489(31), 399(31), 206(25), 145(100), 103(18)
43	CH	O	CH_3	3,4-$(CH_3)_2$-C_6H_3	7.58(s, 1H), 7.57~6.98(m, 11H), 5.63(d, 1H), 5.18(s, 2H), 3.78(s, 3H), 3.63(s, 3H), 2.20(s, 3H), 2.18(s, 6H)	503(20), 205(44), 189(18), 145(100)
44	CH	O	CH_3	3,5-$(CH_3)_2$-C_6H_3	7.48(s, 1H), 7.45~6.67(m, 11H), 5.50(d, 1H), 5.10(s, 2H), 3.58(s, 3H), 3.52(s, 3H), 2.18(s, 3H), 2.15(s, 3H), 2.09(s, 3H)	503(26), 205(35), 145(100)
45	CH	O	CH_3	4-C_2H_5-C_6H_4	7.48(s, 1H), 7.44~6.93(m, 12H), 5.60(d, 1H), 5.11(s, 2H), 3.67(s, 3H), 3.57(s, 3H), 3.36(q, 2H), 2.11(s, 3H), 1.12(t, 3H)	503(11), 282(19), 205(40), 145(100)
46	CH	O	CH_3	4-*n*-C_4H_9-C_6H_4	7.59(s, 1H), 7.58~7.00(m, 12H), 5.64(d, 1H), 5.19(s, 2H), 3.79(s, 3H), 3.64(s, 3H), 2.57(t, 2H), 2.20(s, 3H), 1.44-1.19(m, 4H), 0.91(t, 3H)	531(17), 301(24), 205(55), 189(21), 145(100)
47	CH	O	CH_3	3-CH_3O-C_6H_4	7.59(s, 1H), 7.58~6.78(m, 12H), 5.65(d, 1H), 5.17(s, 2H), 3.80(s, 3H), 3.78(s, 3H), 3.62(s, 3H), 2.18(s, 3H)	505(51), 284(27), 205(44), 145(100), 131(38)

Table 7 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
48	CH	O	CH_3	4-CH_3O-C_6H_4	7.49(s, 1H), 7.46~6.68(m, 12H), 5.58(d, 1H), 5.08(s, 2H), 3.67(s, 3H), 3.65(s, 3H), 3.54(s, 3H), 2.11(s, 3H)	505(13), 205(24), 145(100), 139(32), 84(32)
49	CH	O	CH_3	4-C_2H_5O-C_6H_4	7.60(s, 1H), 7.58~6.81(m, 12H), 5.66(d, 1H), 5.19(s, 2H), 4.00(q, 2H), 3.76(s, 3H), 3.65(s, 3H), 2.20(s, 3H), 1.38(t, 3H)	519(49), 298(37), 159(46), 145(100), 131(59)
50	CH	O	CH_3	3-F-C_6H_4	7.58(s, 1H), 7.55~6.80(m, 12H), 5.64(d, 1H), 5.18(s, 2H), 3.77(s, 3H), 3.68(s, 3H), 2.18(s, 3H)	493(28), 205(33), 189(26), 145(100), 131(42)
51	CH	O	CH_3	4-F-C_6H_4	7.61(s, 1H), 7.58~6.94(m, 12H), 5.72(d, 1H), 5.20(s, 2H), 3.80(s, 3H), 3.68(s, 3H), 2.20(s, 3H)	493(71), 272(33), 205(69), 189(60), 145(100), 127(64)
52	CH	O	CH_3	3-CH_3-4-Cl-C_6H_3	7.61(s, 1H), 7.52~6.97(m, 11H), 5.57(d, 1H), 5.12(s, 2H), 3.78(s, 3H), 3.64(s, 3H), 2.21(s, 3H), 2.16(s, 3H)	523(44), 302(34), 205(23), 145(100)
53	CH	O	CH_3	3-Cl-C_6H_4	7.53(s, 1H), 7.52~7.00(m, 12H), 5.66(d, 1H), 5.15(s, 2H), 3.77(s, 3H), 3.67(s, 3H), 2.25(s, 3H)	509(42), 205(31), 189(26), 145(100)
54	CH	O	CH_3	4-Cl-C_6H_4	7.52(s, 1H), 7.50~7.01(m, 12H), 5.67(d, 1H), 5.18(s, 2H), 3.78(s, 3H), 3.66(s, 3H), 2.24(s, 3H)	509(37), 206(30), 189(24), 145(100), 103(25)
55	CH	O	CH_3	3,4-OCH_2O-C_6H_3	7.60(s, 1H), 7.58~6.75(m, 11H), 5.88(s, 2H), 5.63(d, 1H), 5.20(s, 2H), 3.79(s, 3H), 3.62(s, 3H), 2.19(s, 3H)	519(39), 205(31), 189(16), 145(100), 131(33)
56	CH	O	CH_3	$C_{10}H_7$-2-yl-(naphthalen-2-yl)	7.56(s, 1H), 8.08~7.05(m, 15H), 6.34(d, 1H), 5.16(s, 2H), 3.65(s, 3H), 3.61(s, 3H), 2.16(s, 3H)	525(27), 205(33), 159(28), 145(100)
57	CH	NH	CH_3	4-CH_3-C_6H_4	7.60(s, 1H), 7.58~7.02(m, 12H), 5.71(d, 1H), 5.18(s, 2H), 4.23(b, 1H), 3.61(s, 3H), 2.85(d, 3H), 2.37(s, 3H), 2.23(s, 3H)	488(23), 188(100), 144(56)

Table 7 (Continued)

Ex.No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M. int)
58	CH	NH	CH_3	3,5-$(CH_3)_2$-C_6H_3	7.61(s, 1H), 7.59~6.81(m, 11H), 5.66(d, 1H), 5.18(s, 2H), 4.23(b, 1H), 3.62(s, 3H), 2.91(d, 3H), 2.22(s, 6H)	502(48), 188(100), 144(76)
59	CH	NH	CH_3	4-C_2H_5-C_6H_4	7.61(s, 1H), 7.60~7.01(m, 12H), 5.70(d, 1H), 5.19(s, 2H), 4.22(b, 1H), 3.62(s, 3H), 2.88(d, 3H), 2.61(q, 2H), 2.22(s, 3H), 1.22(t, 3H)	502(13), 188(100), 144(94)
60	CH	NH	CH_3	4-CH_3O-C_6H_4	7.60(s, 1H), 7.59~6.75(m, 12H), 5.68(d, 1H), 5.18(s, 2H), 4.21(b, 1H), 3.78(s, 3H), 3.61(s, 3H), 2.92(d, 3H), 2.21(s, 3H)	504(31), 188(100), 144(85)
61	CH	NH	CH_3	3-CH_3-4-Cl-C_6H_3	7.62(s, 1H), 7.60~7.01(m, 11H), 5.64(d, 1H), 5.17(s, 2H), 4.21(b, 1H), 3.60(s, 3H), 2.90(d, 3H), 2.32(s, 6H), 2.21(s, 3H)	522(13), 188(100), 144(49)
62	CH	NH	CH_3	3-Cl-C_6H_4	7.61(s, 1H), 7.60~7.03(m, 12H), 5.67(d, 1H), 5.19(s, 2H), 4.21(b, 1H), 3.63(s, 3H), 2.92(d, 3H), 2.23(s, 3H)	508(32), 188(100), 144(60)
63	CH	NH	CH_3	3,4-OCH_2O-C_6H_3	7.60(s, 1H), 7.59~6.78(m, 11H), 5.91(s, 2H), 5.65(d, 1H), 5.18(s, 2H), 4.23(b, 1H), 3.61(s, 3H), 2.91(d, 3H), 2.21(s, 3H)	518(41), 188(100), 144(46)
64	N	O	CH_3	C_6H_5	7.49~7.16(m, 13H), 5.73(d, 1H), 5.15(s, 2H), 4.04(s, 3H), 3.82(s, 3H), 2.21(s, 3H)	476(14), 131(68), 116(100), 59(44)
65	N	O	CH_3	3-CH_3-C_6H_4	7.50~6.98(m, 12H), 5.68(d, 1H), 5.15(s, 2H), 4.03(s, 3H), 3.82(s, 3H), 2.31(s, 3H), 2.21(s, 3H)	490(27), 131(67), 116(100), 59(45)
66	N	O	CH_3	4-CH_3-C_6H_4	7.49~7.06(m, 12H), 5.67(d, 1H), 5.13(s, 2H), 4.02(s, 3H), 3.80(s, 3H), 2.29(s, 3H), 2.18(s, 3H)	490(28), 131(65), 123(66), 116(100), 59(64)
67	N	O	CH_3	3,4-$(CH_3)_2$-C_6H_3	7.45~7.00(m, 11H), 5.64(d, 1H), 5.12(s, 2H), 4.00(s, 3H), 3.79(s, 3H), 2.20(s, 6H), 2.17(s, 3H)	504(51), 137(62), 131(60), 116(100), 59(77)

Table 7 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
68	N	O	CH_3	3,5-$(CH_3)_2$-C_6H_3	7.46~6.82(m, 11H), 5.62(d, 1H), 5.13(s, 2H), 4.01(s, 3H), 3.79(s, 3H), 2.24(s, 6H), 2.18(s, 3H)	504(56), 131(62), 115(100), 59(48)
69	N	O	CH_3	4-C_2H_5-C_6H_4	7.50~7.08(m, 12H), 5.69(d, 1H), 5.12(s, 2H), 4.01(s, 3H), 3.81(s, 3H), 2.60(q, 2H), 2.18(s, 3H), 1.19(t, 3H)	504(31), 131(70), 116(100), 59(61)
70	N	O	CH_3	4-*n*-C_4H_9-C_6H_4	7.52~7.07(m, 12H), 5.70(d, 1H), 5.15(s, 2H), 4.04(s, 3H), 3.82(s, 3H), 2.58(t, 2H), 2.20(s, 3H), 1.67~1.24(m, 4H), 0.93(t, 3H)	532(35), 131(66), 116(100), 59(48)
71	N	O	CH_3	3-CH_3O-C_6H_4	7.58~6.77(m, 12H), 5.69(d, 1H), 5.13(s, 2H), 4.03(s, 3H), 3.83(s, 3H), 3.76(s, 3H), 2.19(s, 3H)	506(30), 131(69), 116(100), 59(69)
72	N	O	CH_3	4-CH_3O-C_6H_4	7.56~6.79(m, 12H), 5.66(d, 1H), 5.12(s, 2H), 4.01(s, 3H), 3.80(s, 3H), 3.76(s, 3H), 2.18(s, 3H)	506(46), 139(80), 131(79), 116(100), 59(79)
73	N	O	CH_3	4-C_2H_5O-C_6H_4	7.57~6.78(m, 12H), 5.65(d, 1H), 5.13(s, 2H), 4.02(s, 3H), 4.01(q, 2H), 3.80(s, 3H), 2.18(s, 3H), 1.38(t, 3H)	520(83), 131(70), 125(72), 116(100), 59(68)
74	N	O	CH_3	4-F-C_6H_4	7.50~6.91(m, 12H), 5.65(d, 1H), 5.13(s, 2H), 4.00(s, 3H), 3.79(s, 3H), 2.17(s, 3H)	494(25), 131(63), 116(100), 59(49)
75	N	O	CH_3	3-Cl-C_6H_4	7.49~7.08(m, 12H), 5.62(d, 1H), 5.13(s, 2H), 4.01(s, 3H), 3.80(s, 3H), 2.18(s, 3H)	510(43), 206(44), 131(71), 116(100), 59(65)
76	N	O	CH_3	4-Cl-C_6H_4	7.50~7.14(m, 12H), 5.67(d, 1H), 5.16(s, 2H), 4.04(s, 3H), 3.83(s, 3H), 2.21(s, 3H)	510(21), 206(61), 131(54), 116(100), 59(74)
77	N	NH	CH_3	C_6H_5	7.47~7.15(m, 13H), 6.71(b, 1H), 5.70(d, 1H), 5.13(s, 2H), 3.93(s, 3H), 2.84(d, 3H), 2.18(s, 3H)	475(24), 132(50), 116(68), 58(100)
78	N	NH	CH_3	3-CH_3-C_6H_4	7.46~7.16(m, 12H), 6.70(b, 1H), 5.66(d, 1H), 5.12(s, 2H), 3.93(s, 3H), 2.85(d, 3H), 2.30(s, 3H), 2.17(s, 3H)	489(22), 131(56), 116(73), 58(100)

Table 7 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
79	N	NH	CH_3	4-C_2H_5-C_6H_4	7.46~7.08(m, 12H), 6.68(b, 1H), 5.68(d, 1H), 5.11(s, 2H), 3.92(s, 3H), 2.84(d, 3H), 2.58(q, 2H), 2.16(s, 3H), 1.19(t, 3H)	503(54), 31(57), 115(78), 58(100)
80	N	NH	CH_3	3-CH_3O-C_6H_4	7.49~6.97(m, 12H), 6.72(b, 1H), 5.65(d, 1H), 5.13(s, 2H), 3.94(s, 3H), 3.78(s, 3H), 2.84(d, 3H), 2.18(s, 3H)	505(27), 32(47), 116(59), 58(100)
81	N	NH	CH_3	4-C_2H_5O-C_6H_4	7.50~6.81(m, 12H), 6.70(b, 1H), 5.67(d, 1H), 5.12(s, 2H), 4.00(q, 2H), 3.93(s, 3H), 2.83(d, 3H), 2.17(s, 3H), 1.38(t, 3H)	519(61), 32(52), 116(75), 58(100)
82	N	NH	CH_3	4-F-C_6H_4	7.47~6.93(m, 12H), 6.71(b, 1H), 5.68(d, 1H), 5.13(s, 2H), 3.94(s, 3H), 2.86(d, 3H), 2.16(s, 3H)	493(21), 32(43), 116(61), 58(100)
83	CH	O	CF_3	C_6H_5	7.52(s, 1H), 7.48~7.06(m, 13H), 5.68(d, 1H), 5.14(s, 2H), 3.67(s, 3H), 3.58(s, 3H)	529(12), 89(41), 145(100),
84	CH	O	CF_3	3-CH_3-C_6H_4	7.53(s, 1H), 7.44~6.97(m, 12H), 5.68(d, 1H), 5.16(s, 2H), 3.65(s, 3H), 3.57(s, 3H), 2.32(s, 3H)	543(48), 53(48), 189(48), 45(100), 123(30)
85	CH	O	CF_3	4-CH_3-C_6H_4	7.54(s, 1H), 7.49~7.05(m, 12H), 5.69(d, 1H), 5.17(s, 2H), 3.72(s, 3H), 3.61(s, 3H), 2.31(s, 3H)	543(65), 89(30), 145(100), 23(25)
86	CH	O	CF_3	3,5-$(CH_3)_2$-C_6H_3	7.54(s, 1H), 7.46~6.87(m, 11H), 5.67(d, 1H), 5.14(s, 2H), 3.64(s, 3H), 3.56(s, 3H), 2.33(s, 6H)	557(23), 05(27), 189(39), 45(100)
87	CH	O	CF_3	4-*n*-C_4H_9-C_6H_4	7.53(s, 1H), 7.43~7.05(m, 12H), 5.64(d, 1H), 5.18(s, 2H), 3.68(s, 3H), 3.61(s, 3H), 2.58(t, 2H), 1.64~1.24(m, 4H), 0.92(t, 3H)	585(41), 88(28), 144(100)
88	CH	O	CF_3	3-CH_3O-C_6H_4	7.54(s, 1H), 7.45~6.78(m, 12H), 5.68(d, 1H), 5.18(s, 2H), 3.77(s, 3H), 3.66(s, 3H), 3.54(s, 3H)	559(87), 89(48), 145(100), 39(72)

Table 7 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
89	CH	O	CF_3	4-CH_3O-C_6H_4	7.55(s, 1H), 7.43~6.79(m, 12H), 5.69(d, 1H), 5.19(s, 2H), 3.76(s, 3H), 3.65(s, 3H),	559(92), 89(24), 145(100), 39(26)
90	CH	O	CF_3	3-F-C_6H_4	7.54(s, 1H), 7.53~6.81(m, 12H), 5.64(d, 1H), 5.17(s, 2H), 3.74(s, 3H), 3.62(s, 3H)	547(42), 188(50), 144(100)
91	CH	O	CF_3	3-CH_3-4-Cl-C_6H_3	7.56(s, 1H), 7.47~7.03(m, 11H), 5.61(d, 1H), 5.20(s, 2H), 3.70(s, 3H), 3.61(s, 3H), 2.28(s, 3H)	577(41), 189(94), 145(100), 84(28)
92	CH	O	CF_3	4-Cl-C_6H_4	7.52(s, 1H), 7.48~7.04(m, 12H), 5.65(d, 1H), 5.16(s, 2H), 3.68(s, 3H), 3.59(s, 3H)	563(35), 189(48), 145(100),
93	CH	O	CF_3	$C_{10}H_7$-2-yl-(naphthalen-2-yl)	7.56(s, 1H), 8.06~7.10(m, 15H), 6.02(d, 1H), 5.19(s, 2H), 3.66(s, 3H), 3.57(s, 3H)	579(67), 189(28), 159(37), 145(100)
94	CH	O	CF_3	3,4-OCH_2O-C_6H_3	7.54(s, 1H), 8.01~7.11(m, 11H), 6.78(s, 2H), 5.68(d, 1H), 5.17(s, 2H), 3.63(s, 3H), 3.54(s, 3H)	573(61), 189(52), 159(100), 145(58)
95	CH	NH	CF_3	C_6H_5	7.58(s, 1H), 7.57~7.08(m, 13H), 5.71(d, 1H), 5.21(s, 2H), 4.09(b, 1H), 3.58(s, 3H), 2.82(d, 3H)	528(21), 188(100), 144(67), 109(34)
96	CH	NH	CF_3	3-CH_3-C_6H_4	7.57(s, 1H), 7.49~6.94(m, 12H), 5.68(d, 1H), 5.17(s, 2H), 4.08(b, 1H), 3.60(s, 3H), 2.71(d, 3H), 2.28(s, 3H)	542(61), 188(100), 144(54)
97	CH	NH	CF_3	3,5-$(CH_3)_2$-C_6H_3	7.60(s, 1H), 7.58~6.80(m, 11H), 5.67(d, 1H), 5.20(s, 2H), 4.17(b, 1H), 3.60(s, 3H), 2.83(d, 3H), 2.23(s, 6H)	556(22), 188(100), 144(50)
98	CH	NH	CF_3	3-CH_3O-C_6H_4	8.01(b, 1H), 7.57(s, 1H), 7.56~6.78(m, 12H), 5.69(d, 1H), 5.20(s, 2H), 3.78(s, 3H), 3.58(s, 3H), 2.86(d, 3H)	558(23), 188(100), 144(65)
99	CH	NH	CF_3	4-CH_3O-C_6H_4	7.63(s, 1H), 7.61~6.81(m, 12H), 5.70(d, 1H), 5.21(s, 2H), 4.18(b, 1H), 3.78(s, 3H), 3.61(s, 3H), 2.91(d, 3H)	558(19), 188(100), 144(64)

Table 7 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
100	CH	NH	CF_3	4-Cl-C_6H_4	7.59(s, 1H), 7.51~7.09(m, 12H), 5.69(d, 1H), 5.18(s, 2H), 4.10(b, 1H), 3.58(s,	562(30), 188(100), 144(62), 42(30)
101	CH	NH	CF_3	$C_{10}H_7$-2-yl-(naphthalen-2-yl)	7.55(s, 1H), 8.01~7.08(m, 15H), 6.33(d, 1H), 5.17(s, 2H), 4.11(b, 1H), 3.56(s, 3H), 2.74(d, 3H)	578(42), 188(44), 144(54), 145(100)
102	N	O	CF_3	C_6H_5	7.51~7.08(m, 13H), 5.70(d, 1H), 5.17(s, 2H), 3.98(s, 3H), 3.73(s, 3H)	530(25), 206(18), 131(60), 116(100), 59(68)

Example 103: Preparation of methyl (2*E*)-3-methoxy-2-{2'-[[[3"-(1'"-fluoro-3'",3'",3'"-trifluoro-2'"-phenyl-1'"-propenyloxy)phenyl]imino]oxy]methylphenyl} propenoate (Compound 103)

341 mg (1 mmol) of the compound obtained in Preparation 12 was added to 10 ml of acetonitrile and 40 mg of sodium hydride (1 mmol) dispersed in mineral oil (60%) was added thereto under a nitrogen atmosphere. The resulting solution was stirred for 30 minutes and 208 mg (1 mmol) of the compound obtained in Preparation 41 was added slowly thereto. The resultant solution was stirred for 4 hours with heating, mixed with water and extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure. The residue was subjected to silica gel column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 470 mg (yield 91%) of the title compound as a colorless liquid.

^{1}H-NMR($CDCl_3$, TMS)δ (ppm): 8.01(s, 1H), 7.59(s, 1H), 7.58-6.92(m, 13H), 5.18(s, 2H), 3.78(s, 3H), 3.61(s, 3H)

^{19}F-NMR($CDCl_3$, $CFCl_3$)δ (ppm): -75.916(q, 3F, *J*=12.22Hz), -58.714(d, 1F, *J*=12.267Hz) *E,E*-isomer, -76.313(q, 3F, *J*=23.93Hz), -58.518(d, 1F, *J*=24.676Hz) *E,Z*-isomer

MS(m/e): 529(M^+, 18), 205(59), 189(75), 145(100), 131(25)

Example 120: Preparation of *N*-methyl (2*E*)-3-methoxy-2-{2'-[[[3"-(1'"-fluoro-3'",3'",3'"-trifluoro-2'"-phenyl-1'"-propenyloxy)phenyl]imino]oxy] methylphenyl}propenamide (Compound 120)

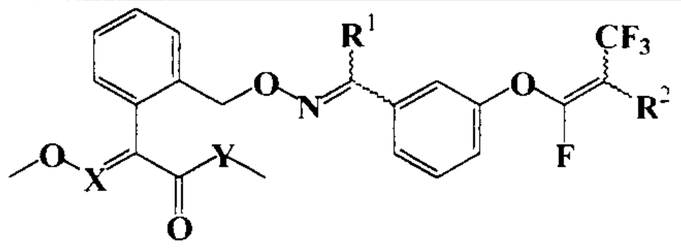
170 mg (0.5 mmol) of the compound obtained in Preparation 18 was added to 10 ml of acetonitrile and 40 mg of sodium hydride (1 mmol) dispersed in mineral oil (60%) was added thereto under a nitrogen atmosphere. The resulting solution was stirred for 30 minutes and 104 mg (0.5 mmol) of the compound obtained in Preparation 41 was added slowly thereto. The resultant solution was stirred for 2 hours at room temperature, mixed with water and extracted with ethyl acetate. The organic layer was dried over magnesium sulfate and the solvent was removed under a reduced pressure. The residue was subjected to silica gel column chromatography using a mixture of *n*-hexane and ethyl acetate (4:1) as an eluent to obtain 243 mg (yield 92%) of the title compound as a colorless liquid.

^{1}H-NMR($CDCl_3$, TMS)δ (ppm): 8.01(s, 1H), 7.62(s, 1H), 7.61-7.00(m, 13H), 5.18(s, 2H), 4.17(b, 1H), 3.63(s, 3H), 2.98(d, 3H)

MS(m/e): 528(M^+, 54), 188(53), 144(100), 103(36), 76(46)

The procedure of Example 103 or 120 was repeated using corresponding phenolic ester compounds or phenolic amide comounds of formula (V) and fluorinated vinyl compounds of formula (VI-b); or, alternatively, the procedure of Example 40, 64 or 77 was repeated using corresponding bromide compounds of formula (II) and the olefin substituted oxime compounds of formula (VII-b), to obtain various compounds of formula (I), as listed in Table 8.

Table 8



Ex.No	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
103	CH	O	H	C_6H_5	8.01(s, 1H), 7.59(s, 1H), 7.58~6.92(m, 13H), 5.18(s, 2H), 3.78(s, 3H), 3.61(s, 3H)	529(18), 205(59), 189(75), 145(100), 131(25)
104	CH	O	H	3-CH_3-C_6H_4	8.01(s, 1H), 7.60(s, 1H), 7.59~6.92(m, 12H), 5.16(s, 2H), 3.78(s, 3H), 3.67(s, 3H), 2.32(s, 3H)	543(21), 221(55), 205(91), 189(84), 145(100)
105	CH	O	H	4-CH_3-C_6H_4	8.00(s, 1H), 7.54(s, 1H), 7.52~6.97(m, 12H), 5.10(s, 2H), 3.71(s, 3H), 3.63(s, 3H), 2.28(s, 3H)	543(31), 205(24), 189(52), 145(100), 103(17)
106	CH	O	H	3,4-$(CH_3)_2$-C_6H_3	8.01(s, 1H), 7.60(s, 1H), 7.59~6.90(m, 11H), 5.19(s, 2H), 3.77(s, 3H), 3.63(s, 3H), 2.25(s, 6H)	557(42), 205(58), 189(70), 145(100), 76(20)
107	CH	O	H	3,5-$(CH_3)_2$-C_6H_3	8.02(s, 1H), 7.60(s, 1H), 7.58~7.01(m, 11H), 5.20(s, 2H), 3.77(s, 3H), 3.74(s, 3H), 2.38(s, 3H), 2.36(s, 3H)	557(11), 221(21), 205(87), 190(33), 145(100)
108	CH	O	H	4-C_2H_5-C_6H_4	8.00(s, 1H), 7.60(s, 1H), 7.59~6.98(m, 12H), 5.19(s, 2H), 3.79(s, 3H), 3.63(s, 3H), 2.65(q, 2H), 1.22(t, 3H)	557(23), 539(26), 205(100), 189(93), 146(62)
109	CH	O	H	4-*n*-C_4H_9-C_6H_4	8.01(s, 1H), 7.61(s, 1H), 7.58~6.97(m, 12H), 5.18(s, 2H), 3.80(s, 3H), 3.64(s, 3H), 2.61(t, 2H), 1.74~1.28(m, 4H), 0.94(t, 3H)	585(28), 205(52), 145(100), 131(20)
110	CH	O	H	3-CH_3O-C_6H_4	8.00(s, 1H), 7.59(s, 1H), 7.58~6.86(m, 12H), 5.16(s, 2H), 3.81(s, 3H), 3.78(s, 3H), 3.62(s, 3H)	559(45), 205(42), 189(62), 145(100), 86(39)
111	CH	O	H	4-CH_3O-C_6H_4	8.01(s, 1H), 7.56(s, 1H), 7.54~6.81(m, 12H), 5.18(s, 2H), 3.78(s, 3H), 3.75(s, 3H), 3.68(s, 3H)	559(47), 205(31), 189(47), 145(100), 137(17), 103(14)

Table 8 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
112	CH	O	H	3-F-C_6H_4	8.00(s, 1H), 7.59(s, 1H), 7.58~6.91(m, 12H), 5.20(s, 2H), 3.78(s, 3H), 3.62(s, 3H)	547(32), 279(13), 167(34), 145(100), 71(55), 57(94)
113	CH	O	H	4-F-C_6H_4	8.03(s, 1H), 7.61(s, 1H), 7.58~6.94(m, 12H), 5.17(s, 2H), 3.81(s, 3H), 3.71(s, 3H)	547(31), 205(85), 190(26), 145(100)
114	CH	O	H	3-CF_3-C_6H_4	8.00(s, 1H), 7.58(s, 1H), 7.56~6.89(m, 12H), 5.18(s, 2H), 3.77(s, 3H), 3.62(s, 3H)	597(31), 205(33), 189(69), 145(100), 76(29)
115	CH	O	H	4-CF_3-C_6H_4	8.02(s, 1H), 7.59(s, 1H), 7.57~6.94(m, 12H), 5.20(s, 2H), 3.79(s, 3H), 3.63(s, 3H)	597(43), 279(21), 189(39), 167(66), 149(100), 71(46)
116	CH	O	H	3-Cl-C_6H_4	8.03(s, 1H), 7.60(s, 1H), 7.57~7.02(m, 12H), 5.20(s, 2H), 3.81(s, 3H), 3.72(s, 3H)	563(42), 221(69), 205(94), 190(87), 145(100)
117	CH	O	H	4-Cl-C_6H_4	8.01(s, 1H), 7.60(s, 1H), 7.59~6.92(m, 12H), 5.19(s, 2H), 3.79(s, 3H), 3.64(s, 3H)	563(26), 285(25), 149(60), 111(49), 57(100)
118	CH	O	H	4-Br-C_6H_4	8.06(s, 1H), 7.61(s, 1H), 7.60~7.00(m, 12H), 5.18(s, 2H), 3.80(s, 3H), 3.70(s, 3H)	607(11), 221(21), 205(89), 190(38), 145(100)
119	CH	O	H	3,4-OCH_2O-C_6H_3	8.00(s, 1H), 7.61(s, 1H), 7.60~6.81(m, 11H), 5.86(s, 2H), 5.19(s, 2H), 3.80(s, 3H), 3.64(s, 3H)	573(20), 189(73), 145(100)
120	CH	NH	H	C_6H_5	8.01(s, 1H), 7.62(s, 1H), 7.61~7.00(m, 13H), 5.18(s, 2H), 4.17(b, 1H), 3.63(s, 3H), 2.98(d, 3H)	528(54), 188(53), 144(100), 103(36), 76(46)
121	CH	NH	H	4-CH_3-C_6H_4	8.00(s, 1H), 7.61(s, 1H), 7.59~7.02(m, 12H), 5.18(s, 2H), 4.16(b, 1H), 3.64(s, 3H), 2.97(d, 3H), 2.37(s, 3H)	542(49), 188(51), 84(100), 47(66)

Table 8 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
122	CH	NH	H	3,5-$(CH_3)_2$-C_6H_3	8.06(s, 1H), 7.62(s, 1H), 7.60~6.81(m, 11H), 5.18(s, 2H), 4.18(b, 1H), 3.61(s, 3H), 2.37(s, 3H)	556(81), 405(25), 188(49), 111(52), 83(46), 57(100)
123	CH	NH	H	4-C_2H_5-C_6H_4	8.02(s, 1H), 7.62(s, 1H), 7.60~7.08(m, 12H), 5.20(s, 2H), 4.15(b, 1H), 3.61(s, 3H), 3.00(d, 3H), 2.61(q, 2H), 1.21(t, 3H)	556(38), 188(100), 144(70)
124	CH	NH	H	4-*n*-C_4H_9-C_6H_4	8.00(s, 1H), 7.62(s, 1H), 7.61~7.01(m, 12H), 5.19(s, 2H), 4.14(b, 1H), 3.60(s, 3H), 2.97(d, 3H), 2.60(t, 2H), 1.64~1.25(m, 4H), 0.93(t, 3H)	584(48), 188(100), 144(49)
125	CH	NH	H	3-CH_3O-C_6H_4	8.05(s, 1H), 7.62(s, 1H), 7.61~7.01(m, 12H), 5.17(s, 2H), 4.16(b, 1H), 3.76(s, 3H), 3.62(s, 3H), 2.94(d, 3H)	558(48), 188(100), 144(45), 115(30)
126	CH	NH	H	4-CH_3O-C_6H_4	8.01(s, 1H), 7.61(s, 1H), 7.60~7.05(m, 12H), 5.18(s, 2H), 4.17(b, 1H), 3.78(s, 3H), 3.61(s, 3H), 2.98(d, 3H)	558(47), 188(100), 144(71)
127	CH	NH	H	3-F-C_6H_4	8.00(s, 1H), 7.62(s, 1H), 7.60~7.06(m, 12H), 5.19(s, 2H), 4.17(b, 1H), 3.61(s, 3H), 2.91(d, 3H)	546(38), 188(100), 144(42), 76(22)
128	CH	NH	H	3-CF_3-C_6H_4	8.00(s, 1H), 7.62(s, 1H), 7.61~7.08(m, 12H), 5.20(s, 2H), 4.19(b, 1H), 3.62(s, 3H), 2.90(d, 3H)	596(51), 188(100), 144(66), 76(25)
129	CH	NH	H	4-CF_3-C_6H_4	8.03(s, 1H), 7.61(s, 1H), 7.60~7.10(m, 12H), 5.17(s, 2H), 4.17(b, 1H), 3.61(s, 3H), 2.91(d, 3H)	596(42), 188(100), 144(49), 76(23)
130	CH	NH	H	3-Cl-C_6H_4	8.01(s, 1H), 7.60(s, 1H), 7.54~7.12(m, 12H), 5.18(s, 2H), 4.16(b, 1H), 3.62(s, 3H), 2.93(d, 3H)	562(48), 188(100), 144(55), 76(18)
131	N	O	H	C_6H_5	7.97(s, 1H), 7.58~6.97(m, 13H), 5.10(s, 2H), 4.01(s, 3H), 3.80(s, 3H)	530(27), 221(45), 131(46), 116(100), 59(85)

Table 8 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
132	N	O	H	4-CH_3-C_6H_4	7.99(s, 1H), 7.57~6.96(m, 12H), 5.12(s, 2H), 4.02(s, 3H), 3.82(s, 3H), 2.34(s, 3H)	544(46), 116(77), 59(100)
133	N	O	H	3,5-$(CH_3)_2$-C_6H_3	7.97(s, 1H), 7.56~6.87(m, 11H), 5.10(s, 2H), 4.01(s, 3H), 3.81(s, 3H), 2.27(s, 6H)	558(32), 116(78), 59(100)
134	N	O	H	3-CH_3O-C_6H_4	7.98(s, 1H), 7.58~6.81(m, 12H), 5.09(s, 2H), 4.00(s, 3H), 3.81(s, 3H), 3.74(s, 3H)	560(38), 131(54), 116(100), 59(49)
135	N	O	H	4-F-C_6H_4	7.97(s, 1H), 7.57~6.93(m, 12H), 5.10(s, 2H), 4.00(s, 3H), 3.80(s, 3H)	548(22), 116(44), 59(100)
136	N	O	H	4-CF_3-C_6H_4	7.98(s, 1H), 7.71~6.95(m, 12H), 5.10(s, 2H), 4.01(s, 3H), 3.81(s, 3H)	598(41), 131(52), 116(97), 59(100)
137	N	O	H	3-Cl-C_6H_4	7.98(s, 1H), 7.56~6.92(m, 12H), 5.11(s, 2H), 4.01(s, 3H), 3.81(s, 3H)	564(34), 131(45), 116(100), 59(67)
138	N	O	H	3,4-OCH_2O-C_6H_3	7.97(s, 1H), 7.57~6.78(m, 11H), 5.94(s, 2H), 5.09(s, 2H), 4.01(s, 3H), 3.80(s, 3H)	574(42), 116(41), 59(100)
139	N	NH	H	C_6H_5	7.97(s, 1H), 7.51~6.96(m, 13H), 6.76(b, 1H), 5.09(s, 2H), 3.92(s, 3H), 2.86(d, 3H)	529(29), 221(43), 132(79), 116(89), 58(100)
140	N	NH	H	3-CH_3-C_6H_4	7.98(s, 1H), 7.58~6.93(m, 12H), 6.78(b, 1H), 5.12(s, 2H), 3.94(s, 3H), 2.84(d, 3H), 2.32(s, 3H)	543(51), 221(96), 205(42), 132(94), 116(100), 58(71)
141	N	NH	H	3,5-$(CH_3)_2$-C_6H_3	7.99(s, 1H), 7.58~6.89(m, 11H), 6.73(b, 1H), 5.10(s, 2H), 3.95(s, 3H), 2.83(d, 3H), 2.32(s, 6H)	557(76), 221(88), 205(47), 132(97), 116(98), 58(100)
142	N	NH	H	4-*n*-C_4H_9-C_6H_4	7.98(s, 1H), 7.54~6.89(m, 12H), 6.76(b, 1H), 5.10(s, 2H), 3.95(s, 3H), 2.83(d, 3H), 2.61(t, 2H), 1.64~1.21(m, 4H), 0.91(t, 3H)	585(73), 221(98), 205(61), 132(100), 116(63), 58(88)

Table 8 (Continued)

Ex.No	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
143	N	NH	H	3-CH_3O-C_6H_4	7.97(s, 1H), 7.50~6.80(m, 12H), 6.76(b, 1H), 5.10(s, 2H), 3.93(s, 3H), 3.74(s, 3H), 2.84(d, 3H)	559(89), 338(41), 221(98), 205(45), 132(90), 58(100)
144	N	NH	H	4-C_2H_5O-C_6H_4	7.97(s, 1H), 7.51~6.80(m, 12H), 6.75(b, 1H), 5.10(s, 2H), 4.01(q, 2H), 3.93(s, 3H), 2.88(d, 3H), 1.41(t, 3H)	573(67), 221(79), 205(45), 132(93), 116(93), 58(100)
145	N	NH	H	4-F-C_6H_4	7.98(s, 1H), 7.57~6.94(m, 12H), 6.74(b, 1H), 5.11(s, 2H), 3.94(s, 3H), 2.84(d, 3H)	547(43), 221(85), 132(81), 116(86), 58(100)
146	N	NH	H	3-CF_3-C_6H_4	7.97(s, 1H), 7.72~6.92(m, 12H), 6.78(b, 1H), 5.11(s, 2H), 3.94(s, 3H), 2.84(d, 3H)	597(62), 221(59), 132(71), 116(85), 58(100)
147	N	NH	H	4-CF_3-C_6H_4	7.95(s, 1H), 7.74~6.90(m, 12H), 6.75(b, 1H), 5.11(s, 2H), 3.95(s, 3H), 2.83(d, 3H)	597(83), 221(81), 132(84), 116(83), 58(100)
148	N	NH	H	3-Cl-C_6H_4	7.96(s, 1H), 7.56~6.92(m, 12H), 6.74(b, 1H), 5.09(s, 2H), 3.96(s, 3H), 2.82(d, 3H)	563(71), 221(100), 132(71), 58(67)
149	N	NH	H	3,4-OCH_2O-C_6H_3	7.97(s, 1H), 7.58~6.80(m, 11H), 6.73(b, 1H), 5.94(s, 2H), 5.10(s, 2H), 3.96(s, 3H), 2.84(d, 3H)	573(36), 221(42), 205(45), 176(77), 132(86), 116(100)
150	CH	O	CH_3	C_6H_5	7.57(s, 1H), 7.54~6.92(m, 13H), 5.14(s, 2H), 3.74(s, 3H), 3.67(s, 3H), 2.18(s, 3H)	543(19), 205(43), 189(44), 145(100)
151	CH	O	CH_3	3-CH_3-C_6H_4	7.56(s, 1H), 7.54~7.00(m, 12H), 5.16(s, 2H), 3.77(s, 3H), 3.71(s, 3H), 2.28(s, 3H), 2.17(s, 3H)	557(31), 336(57), 205(39), 189(29), 145(100)
152	CH	O	CH_3	4-CH_3-C_6H_4	7.57(s, 1H), 7.54~6.98(m, 12H), 5.17(s, 2H), 3.77(s, 3H), 3.68(s, 3H), 2.26(s, 3H), 2.17(s, 3H)	557(47), 336(71), 205(37), 189(39), 145(100)

Table 8 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
153	CH	O	CH_3	3,4-$(CH_3)_2$-C_6H_3	7.59(s, 1H), 7.58~6.97(m, 11H), 5.18(s, 2H), 3.74(s, 3H), 3.67(s, 3H), 2.26(s, 3H), 2.24(s, 6H)	571(26), 553(55), 480(61), 350(34), 205(87), 145(100)
154	CH	O	CH_3	3,5-$(CH_3)_2$-C_6H_3	7.45(s, 1H), 7.42~6.79(m, 11H), 5.04(s, 2H), 3.61(s, 3H), 3.52(s, 3H), 2.20(s, 3H), 2.17(s, 3H), 2.08(s, 3H)	571(12), 205(28), 145(100), 117(14), 103(18)
155	CH	O	CH_3	4-C_2H_5-C_6H_4	7.61(s, 1H), 7.60~7.01(m, 12H), 5.21(s, 2H), 3.76(s, 3H), 3.71(s, 3H), 2.67(q, 2H), 2.21(s, 3H), 1.28(t, 3H)	571(11), 350(21), 205(99), 145(100)
156	CH	O	CH_3	4-*n*-C_4H_9-C_6H_4	7.61(s, 1H), 7.60~7.00(m, 12H), 5.21(s, 2H), 3.75(s, 3H), 3.70(s, 3H), 2.65(t, 2H), 2.21(s, 3H), 1.75~1.26(m, 4H), 0.98(t, 3H)	599(34), 582(36), 508(31), 205(100), 145(30)
157	CH	O	CH_3	3-CH_3O-C_6H_4	7.62(s, 1H), 7.61~6.86(m, 12H), 5.20(s, 2H), 3.79(s, 3H), 3.79(s, 3H), 3.71(s, 3H), 2.21(s, 3H)	573(16), 352(28), 205(96), 190(27), 145(100)
158	CH	O	CH_3	4-CH_3O-C_6H_4	7.60(s, 1H), 7.58~6.84(m, 12H), 5.21(s, 2H), 3.81(s, 3H), 3.75(s, 3H), 3.70(s, 3H), 2.21(s, 3H)	573(12), 352(12), 205(38), 145(100), 103(23)
159	CH	O	CH_3	3-F-C_6H_4	7.65(s, 1H), 7.63~6.94(m, 12H), 5.20(s, 2H), 3.79(s, 3H), 3.69(s, 3H), 2.21(s, 3H)	561(31), 543(54), 470(67), 340(24), 205(97), 145(100)
160	CH	O	CH_3	4-F-C_6H_4	7.63(s, 1H), 7.62~6.98(m, 12H), 5.23(s, 2H), 3.78(s, 3H), 3.70(s, 3H), 2.25(s, 3H)	561(13), 340(24), 205(87), 145(100)
161	CH	O	CH_3	4-CF_3-C_6H_4	7.60(s, 1H), 7.63~6.98(m, 12H), 5.18(s, 2H), 3.78(s, 3H), 3.68(s, 3H), 2.20(s, 3H)	611(51), 593(32), 391(27), 205(48), 189(43), 145(100)
162	CH	O	CH_3	3-Cl-C_6H_4	7.58(s, 1H), 7.56~6.97(m, 12H), 5.17(s, 2H), 3.77(s, 3H), 3.67(s, 3H), 2.16(s, 3H)	577(30), 356(10), 205(49), 189(42), 145(100)

Table 8 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
163	CH	O	CH_3	4-Cl-C_6H_4	7.59(s, 1H), 7.56~6.95(m, 12H), 5.16(s, 2H), 3.76(s, 3H), 3.66(s, 3H), 2.17(s, 3H)	577(38), 356(36), 205(24), 189(39), 145(100), 77(21)
164	CH	O	CH_3	3,5-Cl_2-C_6H_3	7.61(s, 1H), 7.60~7.00(m, 11H), 5.19(s, 2H), 3.80(s, 3H), 3.70(s, 3H), 2.23(s, 3H)	611(28), 520(30), 205(94), 190(32), 173(22), 145(100)
165	CH	O	CH_3	4-Br-C_6H_4	7.60(s, 1H), 7.59~6.97(m, 12H), 5.20(s, 2H), 3.79(s, 3H), 3.71(s, 3H), 2.24(s, 3H)	621(15), 400(12), 205(86), 189(46), 145(100)
166	CH	O	CH_3	3,4-OCH_2O-C_6H_3	7.59(s, 1H), 7.57~6.78(m, 11H), 5.87(s, 2H), 5.19(s, 2H), 3.79(s, 3H), 3.65(s, 3H), 2.20(s, 3H)	587(31), 205(75), 189(29), 145(100)
167	CH	NH	CH_3	C_6H_5	7.56(s, 1H), 7.53~6.78(m, 13H), 5.15(s, 2H), 4.31(b, 1H), 3.63(s, 3H), 2.81(d, 3H), 2.22(s, 3H)	542(34), 354(26), 188(100), 144(94)
168	CH	NH	CH_3	4-CH_3-C_6H_4	7.60(s, 1H), 7.57~6.94(m, 12H), 5.17(s, 2H), 4.28(b, 1H), 3.62(s, 3H), 2.83(d, 3H), 2.38(s, 3H), 2.22(s, 3H)	556(42), 336(43), 188(100), 144(56)
169	CH	NH	CH_3	4-C_2H_5-C_6H_4	7.61(s, 1H), 7.59~6.78(m, 12H), 5.18(s, 2H), 4.25(b, 1H), 3.61(s, 3H), 2.93(d, 3H), 2.67(q, 2H), 2.23(s, 3H), 1.24(t, 3H)	570(16), 188(100), 144(48)
170	CH	NH	CH_3	4-*n*-C_4H_9-C_6H_4	7.61(s, 1H), 7.60~6.92(m, 12H), 5.18(s, 2H), 4.27(b, 1H), 3.61(s, 3H), 2.87(d, 3H), 2.61(t, 2H), 2.34(s, 3H),	598(20), 188(100), 144(53)
171	CH	NH	CH_3	3-CH_3O-C_6H_4	7.62(s, 1H), 7.60~6.80(m, 12H), 5.20(s, 2H), 4.23(b, 1H), 3.80(s, 3H), 3.61(s, 3H), 2.93(d, 3H), 2.21(s, 3H)	572(39), 188(100), 144(70)
172	CH	NH	CH_3	4-CH_3O-C_6H_4	7.60(s, 1H), 7.58~6.81(m, 12H), 5.19(s, 2H), 4.24(b, 1H), 3.81(s, 3H), 3.62(s, 3H), 2.91(d, 3H), 2.22(s, 3H)	572(23), 188(45), 144(29), 84(100)

Table 8 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
173	N	O	CH_3	C_6H_5	7.47~7.21(m, 13H), 5.12(s, 2H), 4.01(s, 3H), 3.78(s, 3H), 2.15(s, 3H)	544(24), 222(43), 131(60), 116(100), 59(49)
174	N	O	CH_3	3-CH_3-C_6H_4	7.49~6.97(m, 12H), 5.12(s, 2H), 4.00(s, 3H), 3.79(s, 3H), 2.31(s, 3H), 2.14(s, 3H)	558(36), 222(40), 131(64), 116(100), 59(41)
175	N	O	CH_3	4-CH_3-C_6H_4	7.47~6.99(m, 12H), 5.13(s, 2H), 4.01(s, 3H), 3.78(s, 3H), 2.32(s, 3H), 2.15(s, 3H)	558(31), 222(41), 131(62), 116(100), 59(42)
176	N	O	CH_3	3,4-$(CH_3)_2$-C_6H_3	7.45~6.92(m, 11H), 5.12(s, 2H), 3.99(s, 3H), 3.76(s, 3H), 2.32(s, 3H), 2.26(s, 3H), 2.14(s, 3H)	572(21), 206(46), 131(66), 115(100)
177	N	O	CH_3	3,5-$(CH_3)_2$-C_6H_3	7.47~6.93(m, 11H), 5.11(s, 2H), 4.00(s, 3H), 3.79(s, 3H), 2.33(s, 3H), 2.27(s, 3H), 2.15(s, 3H)	572(28), 206(52), 131(72), 115(100)
178	N	O	CH_3	4-C_2H_5-C_6H_4	7.50~6.97(m, 12H), 5.12(s, 2H), 4.00(s, 3H), 3.78(s, 3H), 2.62(q, 2H), 2.14(s, 3H), 1.22(t, 3H)	572(18), 222(49), 206(57), 131(69), 116(100), 83(94)
179	N	O	CH_3	4-*n*-C_4H_9-C_6H_4	7.48~6.95(m, 12H), 5.11(s, 2H), 3.99(s, 3H), 3.77(s, 3H), 2.61(t, 2H), 2.13(s, 3H), 1.61~1.24(m, 4H), 0.92(t, 3H)	600(15), 206(67), 131(61), 116(100), 83(76), 58(81)
180	N	O	CH_3	3-CH_3O-C_6H_4	7.52~6.86(m, 12H), 5.14(s, 2H), 4.02(s, 3H), 3.82(s, 3H), 3.80(s, 3H), 2.17(s, 3H)	574(34), 222(43), 131(63), 116(100)
181	N	O	CH_3	4-CH_3O-C_6H_4	7.48~6.80(m, 12H), 5.10(s, 2H), 3.97(s, 3H), 3.77(s, 3H), 3.72(s, 3H), 2.12(s, 3H)	574(16), 206(47), 131(69), 115(100), 59(53)
182	N	O	CH_3	3-F-C_6H_4	7.49~6.97(m, 12H), 5.14(s, 2H), 4.02(s, 3H), 3.80(s, 3H), 2.17(s, 3H)	562(24), 222(48), 131(67), 116(100), 59(51)

Table 8 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
183	N	O	CH_3	4-F-C_6H_4	7.50~6.98(m, 12H), 5.12(s, 2H), 4.00(s, 3H), 3.79(s, 3H), 2.16(s, 3H)	562(41), 222(51), 131(67), 116(100)
184	N	O	CH_3	3-Cl-C_6H_4	7.53~7.00(m, 12H), 5.13(s, 2H), 4.01(s, 3H), 3.78(s, 3H), 2.15(s, 3H)	578(53), 222(47), 131(64), 116(100)
185	N	O	CH_3	4-Cl-C_6H_4	7.50~6.96(m, 12H), 5.15(s, 2H), 4.00(s, 3H), 3.80(s, 3H), 2.17(s, 3H)	578(37), 131(60), 116(100), 59(60)
186	N	O	CH_3	3,5-Cl_2-C_6H_3	7.51~6.97(m, 11H), 5.14(s, 2H), 4.02(s, 3H), 3.81(s, 3H), 2.18(s, 3H)	612(31), 222(67), 131(76), 116(100), 59(60)
187	N	O	CH_3	4-Br-C_6H_4	7.59~6.96(m, 12H), 5.13(s, 2H), 4.01(s, 3H), 3.80(s, 3H), 2.16(s, 3H)	622(26), 222(79), 206(56), 130(100), 115(89)
188	N	O	CH_3	3-CF_3-C_6H_4	7.72~6.95(m, 12H), 5.14(s, 2H), 4.02(s, 3H), 3.80(s, 3H), 2.15(s, 3H)	612(24), 131(55), 116(100), 105(93), 83(97)
189	N	O	CH_3	4-CF_3-C_6H_4	7.66~7.00(m, 12H), 5.13(s, 2H), 4.01(s, 3H), 3.79(s, 3H), 2.16(s, 3H)	612(31), 131(58), 116(100), 59(64)
190	N	O	CH_3	3,4-OCH_2O-C_6H_3	7.49~6.78(m, 11H), 6.77(s, 2H), 5.14(s, 2H), 4.03(s, 3H), 3.80(s, 3H), 2.17(s, 3H)	588(43), 131(63), 116(100), 59(97)
191	N	NH	CH_3	C_6H_5	7.52~6.97(m, 13H), 6.73(b, 1H), 5.14(s, 2H), 3.94(s, 3H), 2.84(d, 3H), 2.16(s, 3H)	543(35), 221(83), 132(88), 116(100), 58(82)
192	N	NH	CH_3	4-CH_3-C_6H_4	7.53~6.96(m, 12H), 6.72(b, 1H), 5.13(s, 2H), 3.95(s, 3H), 2.85(d, 3H), 2.39(s, 3H), 2.17(s, 3H)	557(27), 221(88), 205(54), 116(100), 58(80)
193	N	NH	CH_3	3,4-$(CH_3)_2$-C_6H_3	7.51~6.98(m, 11H), 6.74(b, 1H), 5.14(s, 2H), 3.94(s, 3H), 2.84(d, 3H), 2.37(s, 6H), 2.15(s, 3H)	571(34), 221(60), 205(52), 132(86), 116(100), 58(76)

Table 8 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
194	N	NH	CH_3	3,5-$(CH_3)_2$-C_6H_3	7.59~6.94(m, 11H), 6.81(b, 1H), 5.18(s, 2H), 3.64(s, 3H), 3.80(d, 3H), 2.38(s, 3H), 2.33(s, 3H), 2.20(s, 3H)	571(43), 221(50), 205(38), 132(100), 116(95), 58(46)
195	N	NH	CH_3	4-C_2H_5-C_6H_4	7.50~6.97(m, 12H), 6.73(b, 1H), 5.12(s, 2H), 3.92(s, 3H), 2.83(d, 3H), 2.64(q, 2H), 2.14(s, 3H), 1.21(t, 3H)	571(27), 221(77), 205(55), 132(91), 116(100), 58(91)
196	N	NH	CH_3	4-*n*-C_4H_9-C_6H_4	7.53~7.02(m, 12H), 6.77(b, 1H), 5.18(s, 2H), 3.91(s, 3H), 2.80(d, 3H), 2.60(t, 2H), 2.18(s, 3H), 1.64~1.20(m, 4H), 0.89(t, 3H)	599(21), 221(55), 132(100), 116(81), 58(41)
197	N	NH	CH_3	3-CH_3O-C_6H_4	7.48~6.82(m, 12H), 6.74(b, 1H), 5.11(s, 2H). 3.91(s, 3H), 3.74(s, 3H), 2.82(d, 3H), 2.13(s, 3H)	573(31), 221(87), 205(50), 132(87), 116(100), 58(84)
198	N	NH	CH_3	4-CH_3O-C_6H_4	7.51~6.80(m, 12H), 6.68(b, 1H), 5.09(s, 2H), 3.89(s, 3H), 3.74(s, 3H), 2.81(d, 3H), 2.11(s, 3H)	573(14), 221(83), 205(63), 132(80), 116(100)
199	N	NH	CH_3	4-F-C_6H_4	7.47~6.93(m, 12H), 6.72(b, 1H), 5.12(s, 2H), 3.93(s, 3H), 2.87(d, 3H), 2.15(s, 3H)	561(42), 221(49), 132(77), 116(100), 58(97)
200	N	NH	CH_3	3-CF_3-C_6H_4	7.65~6.95(m, 12H), 6.73(b, 1H), 5.11(s, 2H), 3.93(s, 3H), 2.85(d, 3H), 2.14(s, 3H)	611(33), 221(94), 205(60), 132(97), 116(100), 58(65)
201	N	NH	CH_3	3-Cl-C_6H_4	7.48~6.93(m, 12H), 6.74(b, 1H), 5.12(s, 2H), 3.94(s, 3H), 2.84(d, 3H), 2.16(s, 3H)	577(37), 221(72), 205(49), 132(77), 116(100), 58(76)
202	CH	O	CF_3	C_6H_5	7.51(s, 1H), 7.45~7.00(m, 13H), 5.18(s, 2H), 3.68(s, 3H), 3.56(s, 3H)	597(51), 205(50), 189(87), 145(100), 131(34), 103(37)

Table 8 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
203	CH	O	CF_3	3-CH_3-C_6H_4	7.52(s, 1H), 7.51~7.03(m, 12H), 5.18(s, 2H), 3.68(s, 3H), 3.59(s, 3H), 2.38(s, 3H)	611(39), 205(31), 189(57), 145(100)
204	CH	O	CF_3	4-CH_3-C_6H_4	7.51(s, 1H), 7.37~6.86(m, 12H), 5.17(s, 2H), 3.67(s, 3H), 3.56(s, 3H), 2.36(s, 3H)	611(46), 205(41), 189(100), 145(93)
205	CH	O	CF_3	3,4-$(CH_3)_2$-C_6H_3	7.53(s, 1H), 7.52~6.98(m, 11H), 5.17(s, 2H), 3.69(s, 3H), 3.60(s, 3H), 2.28(s, 3H), 2.22(s, 3H)	625(21), 205(39), 188(75), 144(100)
206	CH	O	CF_3	3,5-$(CH_3)_2$-C_6H_3	7.54(s, 1H), 7.52~6.85(m, 11H), 5.19(s, 2H), 3.68(s, 3H), 3.58(s, 3H), 2.28(s, 6H)	625(13), 221(50), 205(97), 190(60), 173(20), 145(100)
207	CH	O	CF_3	4-C_2H_5-C_6H_4	7.50(s, 1H), 7.49~7.00(m, 12H), 5.20(s, 2H), 3.66(s, 3H), 3.52(s, 3H), 2.61(q, 2H), 1.13(t, 3H)	625(54), 222(21), 205(73), 190(32), 145(100)
208	CH	O	CF_3	4-*n*-C_4H_9-C_6H_4	7.55(s, 1H), 7.53~7.01(m, 12H), 5.21(s, 2H), 3.68(s, 3H), 3.57(s, 3H), 2.64(t, 2H), 1.76~1.19(m, 4H), 0.97(t, 3H)	653(30), 207(15), 189(98), 145(100)
209	CH	O	CF_3	3-CH_3O-C_6H_4	7.52(s, 1H), 7.51~6.79(m, 12H), 5.18(s, 2H), 3.79(s, 3H), 3.74(s, 3H), 3.58(s, 3H)	627(22), 188(79), 145(100)
210	CH	O	CF_3	4-CH_3O-C_6H_4	7.46(s, 1H), 7.44~6.78(m, 12H), 5.08(s, 2H), 3.57(s, 3H), 3.56(s, 3H), 3.48(s, 3H)	627(31), 337(61), 205(33), 189(56), 145(100)
211	CH	O	CF_3	3-F-C_6H_4	7.50(s, 1H), 7.49~7.91(m, 12H), 5.20(s, 2H), 3.66(s, 3H), 3.54(s, 3H)	615(53), 221(94), 206(40), 189(95), 173(47), 145(100)
212	CH	O	CF_3	4-F-C_6H_4	7.51(s, 1H), 7.48~6.86(m, 12H), 5.17(s, 2H), 3.68(s, 3H), 3.54(s, 3H)	615(24), 221(37), 205(98), 190(60), 145(100)

Table 8 (Continued)

Ex. No.	X	Y	R^1	R^2	^{1}H-NMR($CDCl_3$, TMS) δ (ppm)	MS(m/e) (M, int)
213	CH	O	CF_3	3-CF_3-C_6H_4	7.55(s, 1H), 7.54~7.04(m, 12H), 5.18(s, 2H), 3.69(s, 3H), 3.58(s, 3H)	665(19), 189(86), 145(100)
214	CH	O	CF_3	4-CF_3-C_6H_4	7.53(s, 1H), 7.52~7.07(m, 12H), 5.17(s, 2H), 3.70(s, 3H), 3.58(s, 3H)	665(12), 189(84), 145(100)
215	CH	O	CF_3	3-Cl-C_6H_4	7.54(s, 1H), 7.53~7.08(m, 12H), 5.19(s, 2H), 3.70(s, 3H), 3.59(s, 3H)	631(32), 205(46), 189(100), 145(72)
216	CH	O	CF_3	4-Cl-C_6H_4	7.43(s, 1H), 7.40~6.91(m, 12H), 5.07(s, 2H), 3.59(s, 3H), 3.47(s, 3H)	631(27), 273(26), 253(23), 84(100), 43(66)
217	CH	O	CF_3	3,5-Cl_2-C_6H_3	7.50(s, 1H), 7.49~6.94(m, 11H), 5.19(s, 2H), 3.69(s, 3H), 3.57(s, 3H)	665(29), 221(32), 189(89), 161(22), 145(100)
218	CH	O	CF_3	4-Br-C_6H_4	7.53(s, 1H), 7.52~6.97(m, 12H), 5.18(s, 2H), 3.67(s, 3H), 3.56(s, 3H)	675(21), 387(22), 221(32), 198(95), 173(26), 145(100)
219	CH	O	CF_3	3,4-OCH_2O-C_6H_3	7.53(s, 1H), 7.52~6.71(m, 11H), 5.91(s, 2H), 5.20(s, 2H), 3.69(s, 3H), 3.59(s, 3H)	641(18), 188(50), 144(100)
220	CH	NH	CF_3	4-Cl-C_6H_4	7.97(s, 1H), 7.67~6.78(m, 12H), 5.19(s, 2H), 4.10(b, 1H), 3.74(s, 3H), 2.81(d, 3H)	630(51), 408(45), 188(100), 144(57)
221	N	O	CF_3	C_6H_5	7.52~7.00(m, 13H), 5.15(s, 2H), 3.91(s, 3H), 3.71(s, 3H)	598(43), 376(45), 222(81), 206(45), 131(63), 116(100)
222	N	O	CF_3	3-CH_3-C_6H_4	7.54~6.97(m, 12H), 5.17(s, 2H), 3.98(s, 3H), 3.76(s, 3H), 2.32(s, 3H)	612(42), 222(20), 206(74), 131(67), 116(100), 59(49)
223	N	O	CF_3	4-Cl-C_6H_4	7.41~6.87(m, 12H), 5.05(s, 2H), 3.84(s, 3H), 3.61(s, 3H)	632(32), 222(56), 206(50), 131(76), 116(100), 59(53)
224	N	NH	CF_3	4-Cl-C_6H_4	7.84(b, 1H), 7.56~6.80(m, 12H), 5.12(s, 2H), 3.96(s, 3H), 2.87(s, 3H)	631(48), 205(28), 137(26), 116(75), 58(100)

Fungicidal Activity Test

To examine fungicidal activity of the compounds of the present invention, each of the compounds listed in Table 3 and 4 was dissolved in 10% acetone to a concentration of 250 ppm, and Tween-20 was added thereto to a concentration of 250 or 500 ppm. 50 ml of the resulting solution was sprayed on leaves of a host plant. The plant was kept at room temperature for 24 hours to let the solvent evaporate, and then, a pathogenic fungus was inoculated thereonto. The plant was held in a humidity chamber for 24 hours, transferred to an plant growth room kept at 20 to 27 ℃ and a relative humidity of 60 to 80% and kept to induce disease. Subsequently, the lesion area (L.A.) attacked by the pathogenic fungus was measured according to a method of Cho(Cho, K.Y., Search Report by Korea Research Institute of Chemical Technology(1989))". This procedure was repeated twice for each test. 10% Acetone solution containing 250 ppm of Tween-20 was used as a control.

The fungicidal activity of the compound of the present invention is repressed by a control value (C.V.) calculated as;

$$C.V.(\%) = \frac{\text{L.A. of control} - \text{L.A. of test}}{\text{L.A. of control}} \times 100$$

Test Example 1: Fungicidal activity against Rice Blast(RCB) disease

Pyricularia oryzae Carvara KA301 was inoculated on a rice bran agar medium (rice bran 20 g, dextrose 10 g, agar 15 g and distilled water 1 ℓ and cultured at 26 ℃ for 1 week. The surface of the medium was scratched using a rubber polishman to remove aerial mycelia, and cultured under a fluorescent light for 48 hours to form a spore. Spores were suspended in sterilized water at a concentration of 1x106 spore/ml. The spore suspension was sprayed enough to soak the leaves of a RBC disease-sensitive Nakdong rice plant having 3 or 4 leaves. The rice plant was held in a humidified dark room for 24 hours, transferred to an incubator kept at 24 to 28 ℃ and a relative humidity of more than 80% and kept for 5 days to induce RCB. L.A. on a fully grown leaf appearing underneath an uppermost leaf was measured to calculate an C.V.

Test Example 2: Fungicidal activity against Rice Sheath Blight(RSB) disease

Rhizoctonia solani AG-1 was cultured on a PDA medium (potato 200 g, dextrose

20 g, agar 20 g and distilled water 1 ? for 3 days and the agar disc(diameter 0.6 cm) was inoculated and cultured on sterilized wheat bran medium in a 1 L bottle at 26 to 28℃ for 7 days. A mycelial mass was ground using a homogenizer, inoculated uniformly on soil of a pot wherein a Nakdong rice plant having 2 or 3 leaves and an height of 5 cm grew, and kept in humidity polyvinyl chamber for 5 days to induce RSB. L.A. on a leaf sheath was measured to calculate an C.V.

Test Example 3: Fungicidal activity against Cucumber Gray Mold Rot (CGM) disease

Botrytis cinerae, which was isolated from cucumber infected thereby, was inoculated on a PDA agar medium and cultured under a 12L/12D cycle at 25 ℃ for 15 days to form spore. The spores were scraped, filtered through a gauze and then suspended in potato dextrose liquid medium at a concentration of 1x106 spore/ml. The spore suspension was sprayed on a cucumber plant having one leaf. The cucumber plant was held in a humidified room at 20 ℃ for 3 days. L.A. on a leaf was measured to calculate an C.V.

Test Example 4: Fungicidal activity on Tomato Late Blight(TLB) disease

Phytophthora infestans was cultured on a juice agar medium(V-8 juice 200 ml, $CaCO_3$ 4.5 g, agar 15 g and distilled water 800 ml) under a 16L/8D cycle at 20 ℃ for 14 days. Sterilized water was added thereto, the vessel was shaken to free zoospore sacs from the fungus mass and the zoospore sacs were collected using a four-layered gauze. A zoospore sac suspension having a concentration of 1x105 spore/ml was sprayed on a young tomato plant. The tomato plant was held in a humidified room at 20 ℃ for 24 hours, transferred to an incubator maintained at a temperature of 20 ℃ and a relative humidity of more than 80% for 4 days and cultured in to induce RBC. L.A. on primary and secondary leaves was measured to calculate an C.V.

Test Example 5: Fungicidal activity against Wheat Leaf Rust(WLR) disease

Puccinia recondita was subcultured on a wheat plant in a laboratory. 15 g of wheat seeds was sowed in a pot diameter 6.5 cm) and cultured in a greenhouse for 7 days to obtain a wheat plant having only a primary leaf. The wheat plant was inoculated with spores by shaking thereover another plant infected thereby. The inoculated wheat plant was held in a humidified room at 20 ℃ for 24 hours, transferred to an incubator maintained at a temperature of 20 ℃ and a relative humidity of 70%

and cultured for 10 days to induce WLR. L.A. on the primary leaf was measured to calculate an C.V.

Test Example 6: Fungicidal activity against Barley Powdery Mildew(BPM) disease

Erysiphae graminis was subcultured on a wheat plant in a laboratory. 15 g of barley seeds was sowed in a pot (diameter 6.5 cm) and cultured in a greenhouse for 7 days to obtain a barley plant having only a primary leaf. The barley plant was inoculated with spores by shaking thereover another plant infected by BPM. The inoculated barley plant was cultured in an incubator maintained at a temperature of 22 to 24 ℃ and a relative humidity of 50% for 7 days to induce BPM. L.A. on the leaf was measured to calculate an C.V.

The results of subjecting the compounds of the present invention in Test example 1 to 6 at a concentration level of 250 ppm was more than 90% in most cases. Accordingly, these compounds having a C.V. of more than 90% were subjected to another series of tests at reduced concentration levels of 50, 10 and 2 ppm. The test results are shown in Table 9.

Table 9: Fungicidal Activities

Compound No.	Concentration (ppm)	RCV	RSB	CGM	TLB	WLR	BPM
1	50	100	65	69	12	100	100
	10	86	-	-	-	100	100
	2	71	-	-	-	96	99
2	50	93	60	17	85	100	100
	10	86	-	-	-	100	99
	2	33	-	-	-	97	98
3	50	100	60	46	84	100	100
	10	80	-	-	-	98	99
	2	0	-	-	-	96	86
5	50	41	55	17	28	100	100
	10	-	-	-	-	100	100
	2	-	-	-	-	96	98
6	50	99	75	44	33	100	100
	10	58	-	-	-	100	99
	2	8	-	-	-	98	97
9	50	100	50	53	12	100	100
	10	83	-	-	-	98	99
	2	8	-	-	-	95	55
11	50	100	40	17	85	100	100
	10	86	-	-	-	100	99
	2	33	-	-	-	97	98
12	50	98	70	52	83	100	100
	10	93	-	-	-	100	100
	2	33	-	-	-	100	98
13	50	93	70	36	46	100	100
	10	86	-	-	-	100	100
	2	41	-	-	-	99	99
14	50	100	75	7	12	100	100
	10	86	-	-	-	99	100
	2	58	-	-	-	96	99
24	50	97	95	50	33	99	100
	10	88	72	-	-	97	100
	2	28	44	-	-	95	95

Table 9 (continued)

Compound No.	Concentration (ppm)	RCV	RSB	CGM	TLB	WLR	BPM
25	50	98	40	9	99	100	100
	2	66	15	-	75	95	93
26	50	95	77	77	90	93	100
27	50	100	25	0	69	86	91
28	50	99	50	9	38	100	95
	2	66	0	-	50	97	86
29	50	100	35	0	84	88	80
	2	93	20	-	78	96	41
31	50	100	52	25	8	100	100
	2	97	-	-	-	83	8
32	50	100	41	8	0	99	100
	2	90	-	-	-	3	0
33	50	96	17	0	0	96	100
	2	97	-	-	-	10	0
35	50	100	52	0	0	97	100
	2	97	-	-	-	83	0
36	50	100	0	0	0	93	100
	2	88	-	-	-	0	0
37	50	100	29	0	0	98	100
	2	98	-	-	-	37	42
38	50	100	64	0	0	100	100
	2	97	-	-	-	99	87
39	50	100	58	0	83	100	100
	2	93	-	-	-	0	0
40	50	100	95	75	97	100	100
	10	100	75	-	68	100	100
	2	99	70	-	12	99	100
41	50	100	95	51	96	100	100
	10	100	70	-	25	100	100
	2	98	55	-	6	99	100

Table 9 (continued)

Compound No.	Concentration(ppm)	RCV	RSB	CGM	TLB	WLR	BPM
42	50	100	70	64	97	100	100
	10	96	-	-	-	100	100
	2	58	-	-	-	98	100
44	50	100	75	5	71	100	100
	10	95	-	-	-	100	100
	2	85	-	-	-	96	100
45	50	97	65	17	85	100	100
	10	93	-	-	-	100	100
	2	41	-	-	-	95	99
48	50	100	75	64	95	100	100
	10	96	-	-	-	100	100
	2	41	-	-	-	99	100
51	50	100	90	52	97	100	100
	10	99	88	-	-	100	100
52	50	97	65	52	42	100	100
	10	80	-	-	-	96	100
54	50	100	90	51	84	100	100
	10	99	80	-	-	100	100
	2	97	50	-	-	97	99
56	50	95	70	5	57	100	100
	10	80	-	-	-	100	100
	2	25	-	-	-	95	100
57	50	99	47	8	0	98	98
	2	95	-	-	-	27	0
58	50	100	23	8	0	98	98
59	50	100	47	0	0	73	91
60	50	100	29	8	0	66	100
61	50	91	23	33	0	43	86

Table 9 (continued)

Compound No.	Concentration (ppm)	RCV	RSB	CGM	TLB	WLR	BPM
62	50	96	0	25	0	99	100
	2	92	-	-	-	83	0
63	50	96	11	33	0	90	97
64	50	100	85	75	95	100	100
	10	100	65	-	90	100	100
	2	100	45	-	87	99	100
65	50	100	100	71	0	100	100
	10	100	60	-	-	100	100
	2	100	30	-	-	99	99
66	50	100	100	75	0	100	100
	10	100	65	-	-	100	100
	2	100	20	-	-	99	100
67	50	100	100	37	0	100	100
	10	100	43	-	-	100	100
	2	100	10	-	-	99	99
68	50	100	100	56	20	100	100
	10	100	70	-	-	100	100
	2	100	30	-	-	99	100
69	50	100	100	25	60	100	100
	2	100	30	-	25	97	99
70	50	100	85	50	0	97	100
	2	99	20	-	12	93	83
71	50	100	100	25	50	100	100
	2	100	35	-	25	98	100
72	50	98	65	0	66	100	100
	2	100	25	-	0	98	100
73	50	95	100	0	60	100	100
	2	100	25	-	0	98	100
74	50	100	100	72	56	100	100
	2	100	25	-	12	99	100
75	50	100	100	0	66	100	100
	2	100	25	-	0	98	100

Table 9 (continued)

Compound No.	Concentration (ppm)	RCV	RSB	CGM	TLB	WLR	BPM
76	50	100	100	62	0	100	100
	2	100	35	-	0	100	100
77	50	100	100	85	80	100	100
	10	100	57	36	88	100	100
	2	100	40	21	50	100	100
78	50	100	90	82	94	100	100
	2	100	45	-	50	100	100
79	50	100	65	80	82	100	100
	2	99	35	-	43	98	100
80	50	100	100	77	72	100	100
	2	100	45	-	12	96	100
81	50	100	100	83	64	100	100
	2	100	30	-	12	99	100
82	50	96	85	82	84	100	100
	2	100	55	-	85	100	100
85	50	93	40	7	0	96	99
	10	16	-	-	-	93	92
89	50	80	50	0	12	96	100
	10	-	-	-	-	70	99
90	50	76	35	9	27	86	99
	10	-	-	-	-	-	97
94	50	100	60	54	68	100	100
	10	100	-	-	-	96	100
	2	99	-	-	-	93	100
102	50	99	50	23	0	99	100
	10	86	-	-	-	96	100
	2	71	-	-	-	94	97
104	50	96	45	0	0	97	100
	10	75	-	-	-	93	99
	2	25	-	-	-	66	80

Table 9 (continued)

Compound No.	Concentration (ppm)	RCV	RSB	CGM	TLB	WLR	BPM
105	50	0	20	0	0	100	100
	10	-	-	-	-	98	99
	2	-	-	-	-	86	98
111	50	0	0	17	0	98	100
	10	-	-	-	-	95	100
	2	-	-	-	-	80	95
113	50	0	20	0	21	100	100
	10	-	-	-	-	98	96
	2	-	-	-	-	86	86
116	50	0	15	0	14	96	100
	10	-	-	-	-	96	96
	2	-	-	-	-	53	33
118	50	33	30	36	0	100	100
	10	-	-	-	-	98	96
	2	-	-	-	-	91	88
131	50	80	55	42	4	100	100
	2	43	-	-	-	98	76
135	50	86	61	71	0	100	100
	2	43	20	-	-	94	93
139	50	100	20	0	46	98	98
	2	53	10	-	-	93	97
140	50	97	64	0	0	100	86
	2	86	-	-	-	0	-
143	50	100	58	0	0	90	93
	2	83	-	-	-	36	0
144	50	96	47	0	0	86	95
	2	88	-	-	-	-	0
145	50	100	64	0	0	100	100
	2	83	-	-	-	83	33
150	50	100	92	44	50	100	100
	10	100	65	-	-	100	100
	2	83	55	-	-	95	100

Table 9 (continued)

Compound No.	Concentration (ppm)	RCV	RSB	CGM	TLB	WLR	BPM
151	50	100	95	24	12	100	100
	10	99	55	-	-	95	100
	2	93	35	-	-	86	98
152	50	100	90	44	0	100	100
	10	97	60	-	-	98	100
	2	86	45	-	-	86	95
153	50	86	55	20	13	100	100
	10	25	-	-	-	100	99
	2	0	-	-	-	91	83
154	50	16	90	58	12	98	100
	10	-	35	-	-	96	98
	2	-	25	-	-	73	68
155	50	80	60	52	6	100	100
	10	-	-	-	-	99	99
	2	-	-	-	-	83	95
156	50	33	20	36	0	93	92
	10	-	-	-	-	95	91
157	50	93	60	68	6	100	100
	10	76	-	-	-	100	99
	2	33	-	-	-	98	97
158	50	41	70	44	0	100	100
	10	-	-	-	-	98	100
	2	-	-	-	-	91	98
159	50	96	65	52	20	100	100
	10	91	-	-	-	100	100
	2	25	-	-	-	94	99
160	50	100	70	36	92	100	100
	10	100	-	-	-	100	100
	2	86	-	-	-	99	99
163	50	90	92	51	12	100	100
	10	86	40	-	-	98	100
	2	58	40	-	-	86	94

Table 9 (continued)

Compound No.	Concent-ration (ppm)	RCV	RSB	CGM	TLB	WLR	BPM
164	50	41	20	36	0	96	100
	10	-	-	-	-	95	99
	2	-	-	-	-	60	71
165	50	80	45	68	6	100	100
	10	-	-	-	-	96	99
	2	-	-	-	-	86	71
173	50	97	100	25	20	100	100
	2	93	30	-	-	99	99
174	50	96	60	0	0	100	100
	2	86	25	-	-	90	99
175	50	96	65	12	0	100	100
	2	76	10	-	-	86	83
176	50	86	60	12	70	100	100
	2	83	20	-	-	60	80
180	50	100	60	56	56	100	100
	2	86	25	-	-	97	98
181	50	95	45	77	30	99	100
	2	98	35	-	68	83	83
182	50	95	50	77	40	100	100
	2	95	30	-	62	95	96
183	50	100	100	56	50	100	100
	2	99	20	-	12	99	100
184	50	96	85	0	50	99	100
	2	90	20	-	0	98	99
185	50	96	75	0	50	100	100
	2	73	25	-	25	95	98
186	50	83	50	25	0	99	100
	2	73	15	-	0	66	86
187	50	83	85	0	0	100	98
	2	97	20	-	25	95	97

Table 9 (continued)

Compound No.	Concent-ration (ppm)	RCV	RSB	CGM	TLB	WLR	BPM
188	50	71	55	0	40	95	100
	2	76	0	-	0	66	91
190	50	93	65	0	0	100	100
	2	66	15	-	0	93	91
191	50	100	75	68	60	100	100
	2	100	35	-	0	99	100
192	50	100	100	75	40	100	100
	2	100	40	-	37	98	100
193	50	96	80	75	40	100	100
	2	93	30	-	12	98	90
194	50	100	64	16	0	98	98
	2	95	-	-	-	20	0
195	50	96	80	82	50	100	100
	2	100	25	-	25	98	99
197	50	100	80	85	50	100	100
	2	100	35	-	0	98	100
198	50	100	58	16	0	100	100
	2	95	-	-	-	53	41
199	50	100	100	80	60	100	100
	2	100	35	-	12	99	100
200	50	96	85	81	50	100	100
	2	95	15	-	12	93	99
201	50	97	85	85	50	100	100
	2	98	20	-	25	97	100
221	50	41	30	0	0	100	100
	10	-	-	-	-	91	96
ORIBRIGHT[1)]	10	90	70	7	40	60	83
	2	60	15	20	0	15	50
	0.40	8	0	7	0	15	0
	0.08	0	0	0	0	10	0
FENARIMOL[2)]	10	0	85	7	80	100	100
	2	0	40	7	75	80	100
	0.40	0	30	7	60	15	95
	0.08	0	14	0	0	15	50

[1]: a product of Japan, Shionogi

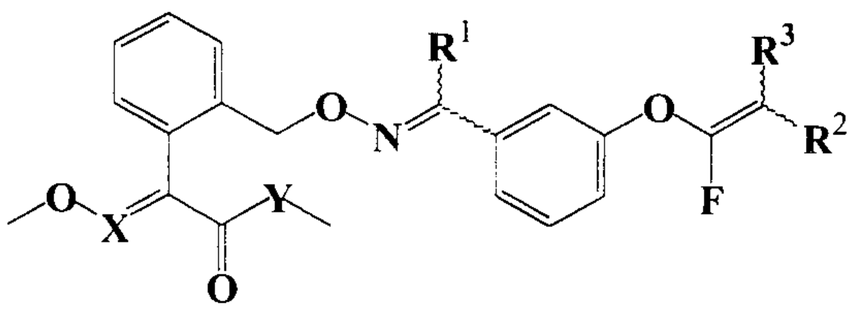
[2]: a product of U.S.A., Dow Elanco

As can be seen from Table 9, the compounds of the present invention have a broad fungicidal activity spectrum against the target fungi when compared with the control compounds such as ORIBRIGHT™ and FENARIMOL™. In particular, the inventive compounds have excellent fungicidal activity against RCB, RSB, WLR and BPM.

While the invention has been described with respect to the specific embodiments, it should be recognized that various modifications and changes may be made by those skilled in the art to the invention which also fall within the scope of the invention as defined by the appended claims.

What is claimed is:

1. A compound of formula (I) or a stereoisomer thereof:



(I)

wherein,

X is CH or N;

Y is O or NH;

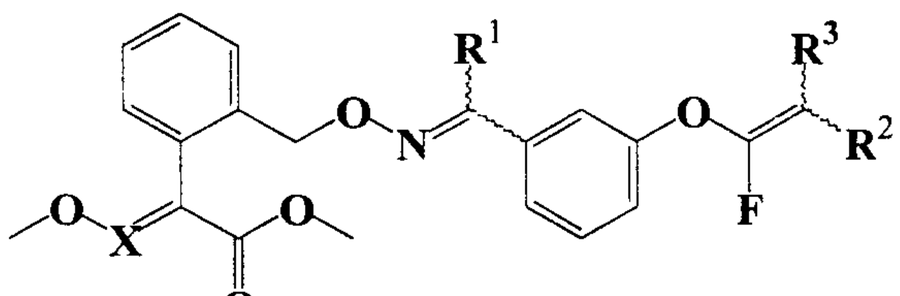
R^1 is hydrogen, C_{1-4} alkyl, or halogen-substituted C_{1-4} alkyl,

R^2 is a phenyl group optionally carrying one or more substituents selected from the group consisting of C_{1-4} alkyl, C_{1-4} alkoxy, methylenedioxy and halogen; or a naphthyl group; and

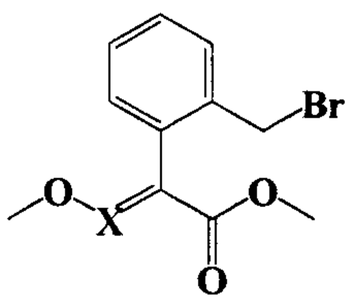
R^3 is hydrogen or CF_3.

2. The compound of claim 1, wherein R^1 is hydrogen or C_{1-4} alkyl, and R^2 is a phenyl group optionally carrying one or more halogen radicals.

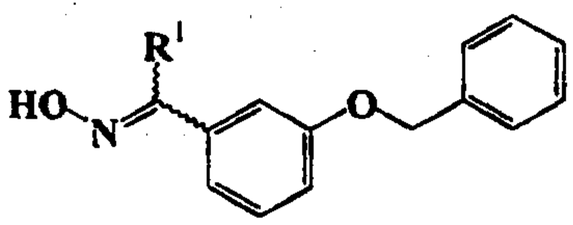
3. A process for the preparation of a compound of formula (I-a), which comprises: (a) reacting a compound of formula (II) with a compound of formula (III) in the presence of a base to obtain a compound of formula (IV); (b) debenzylating the compound of formula (IV) by hydrogenolysis in the presence of a Pd-based catalyst to obtain a compound of formula (V-a); and (c) reacting the compound of formula (V-a) with a compound of formula (VI) in the presence of a base:



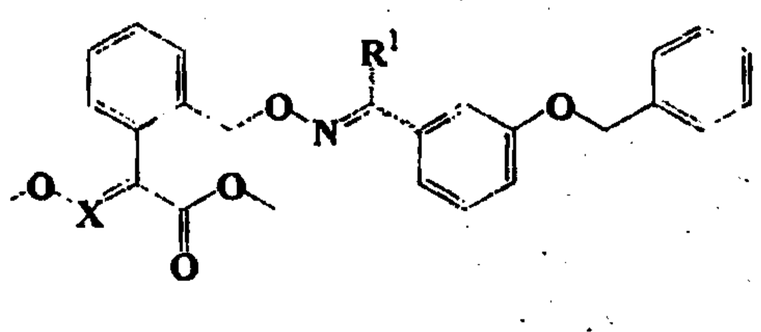
(I-a)



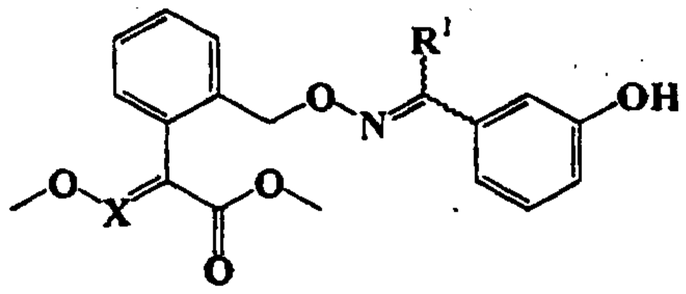
(II)



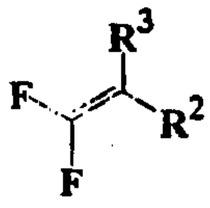
(III)



(IV)



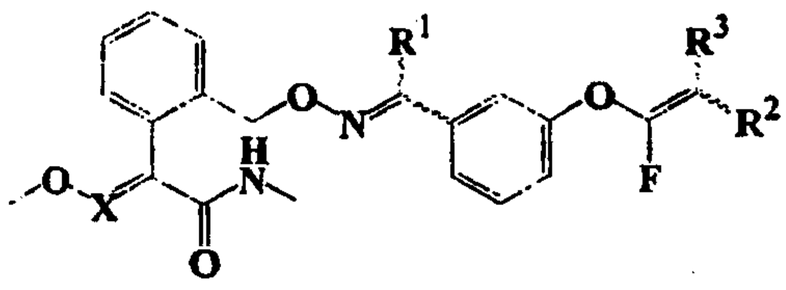
(V-a)



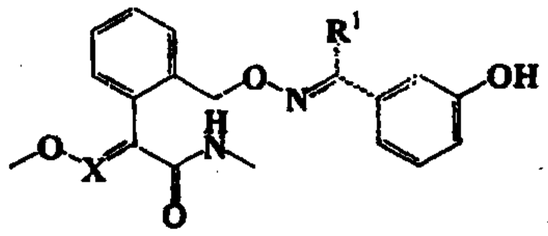
(VI)

wherein, X, R^1, R^2 and R^3 have the same meanings as defined in claim 1.

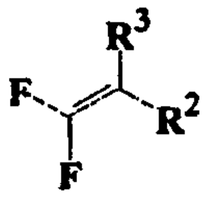
4. A process for the preparation of a compound of formula (I-b), which comprises (a) reacting the phenolic ester compound of formula (V-a) as defined in claim 3 with methylamine to obtain a compound of formula (V-b) and (b) reacting the compound of formula (V-b) with a compound of formula (VI) in the presence of a base:



(I-b)



(V-b)

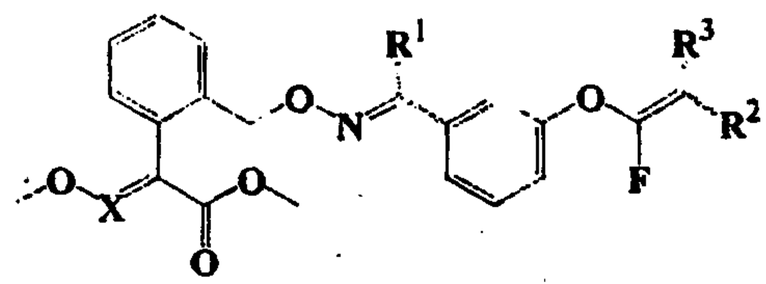


(VI)

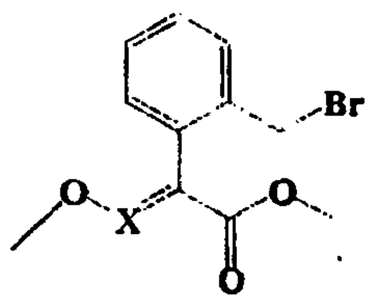
wherein,

X, R^1, R^2 and R^3 have the same meanings as defined in claim 1.

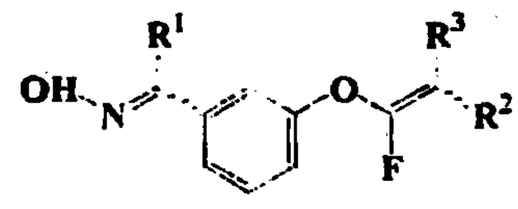
5. A process for the preparation of a compound of formula (I-a), which comprises reacting a compound of formula (II) with a compound of formula (VII) in the presence of a base:



(I-a)



(II)

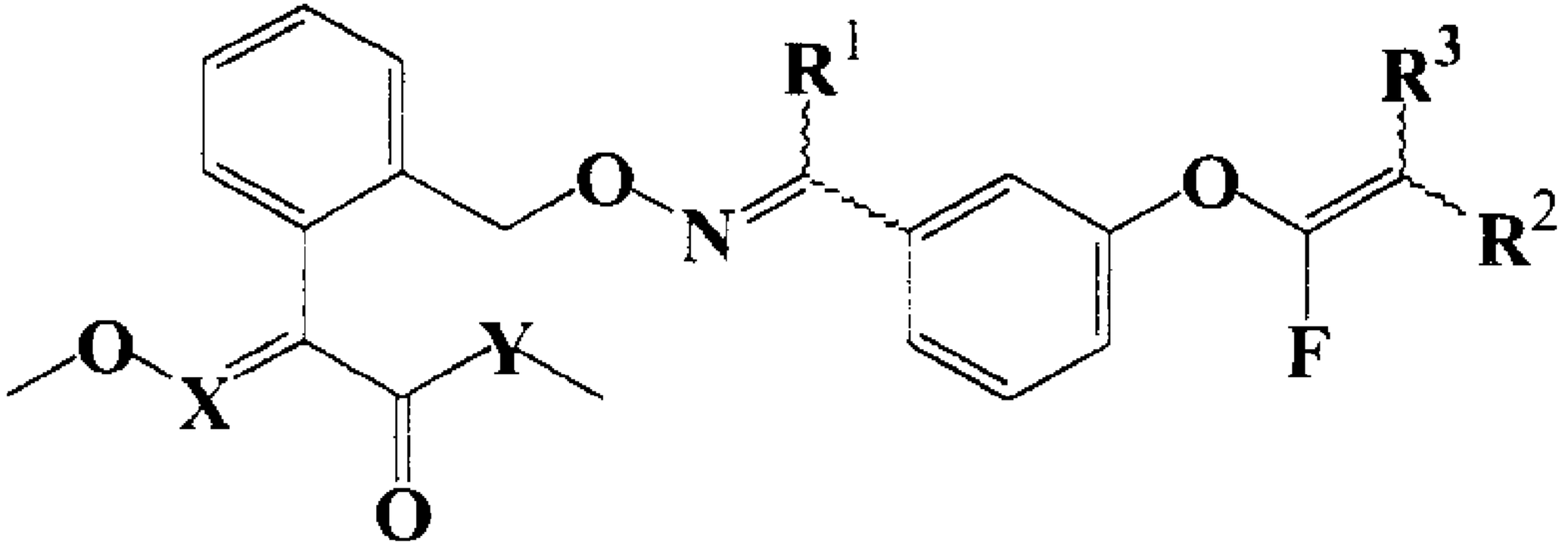


(VII)

wherein,

X, R^1, R^2 and R^3 have the same meanings as defined in claim 1.

6. A fungicidal composition comprising a fungicidally effective amount of the compound according to claim 1 or 2 as the active ingredient and an inert carrier.



(I)