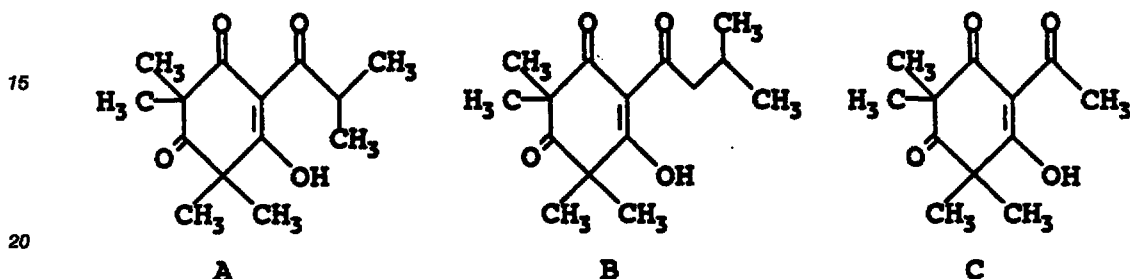


Description

The present invention concerns novel 2-glyoxyl-cyclohex-1-en-3,5-diones, a method for their preparation and their use as herbicides.

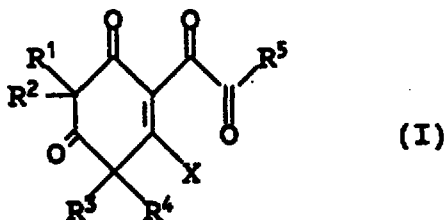
- 5 The compounds A (1-hydroxy-2-isobutyryl-4,4,6,6-tetramethyl-cyclohex-1-en-3,5-dione) and B (1-hydroxy-2-isovaleryl-4,4,6,6-tetramethyl-cyclohex-1-en-3,5-dione) have been isolated from plants of the family Myrtaceae, plant genus Leptospermum and Xanthostemon (R. O. Hellyer, Aust. J. Chem. 21, 2825 (1968)) but no use is disclosed for the compounds. Also, no use is described for compound C (1-hydroxy-2-acetyl-4,4,6,6-tetramethyl-cyclohex-1-en-3,5-dione) which has been disclosed by A.C. Jain and T.R. Seshadri, in Proc. Indian Acad. Sci. 42A, 279 (1955).



- 25 Additionally, it is known from U.S. Patent Specification No. 4,202,840, European Patent Specification No. 249,151A and British Patent Specification No. 2,215,333A that certain compounds of this class show herbicidal activity. However, the dosages of these herbicides required to give satisfactory herbicidal activity are quite high and, additionally, no selectivity can be observed.

- We have found now that, surprisingly, certain compounds distantly related to the natural products mentioned above, show an excellent herbicidal activity at low dosages and a good selectivity of action against broad-leaf weeds.

Accordingly, the present invention provides a compound of the general formula I



characterised in that

- 45 X represents a group OH, OR⁷, NH₂, NHR⁷, NR⁷R⁸, NOH, NOR⁷, OCOR⁷, OCONHR⁷, OCONHCOR⁷, OCONHSO₂R⁷, NHCONHCOR⁷, OCOSR⁷, NHCOSR⁷, SCOR⁷, SR⁷, SCONHR⁷, SCONHCOR⁷, SCONHSO₂R⁷ or SCOSR⁷;
- 50 R¹ to R⁴ each independently represents a hydrogen atom or a straight-chain or branched-chain C₁₋₆ alkyl, C₂₋₆ alkenyl or C₂₋₆ alkynyl group which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;
- or
- 55 R¹ and R² and/or R³ and R⁴ together form a straight-chain or branched-chain, saturated or unsaturated, C₂₋₆-methylene bridge, optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;
- R⁵ is a straight-chain or branched-chain C₁₋₂₀ alkyl, C₂₋₂₀ alkenyl or C₂₋₂₀ alkynyl group, a C₃₋₂₀ cycloalkyl, phenyl or naphthyl group, or a saturated or unsaturated heterocyclic residue having one or more

heteroatoms,

each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

5 R⁶ is a straight-chain or branched-chain C₁₋₂₀ alkyl group, a C₃₋₂₀ cycloalkyl, phenyl or naphthyl group, or a saturated or unsaturated heterocyclic residue having one or more heteroatoms,

10 each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, COOH, NO₂, NH₂, SO₃H and SH groups;

and

R⁷ and R⁸

each independently has the same meaning as R⁵.

Alkyl as a substituent or as part of other substituents, such as alkoxy, may be, for example, one of the following straight-chain or branched groups depending on the number of carbon atoms indicated: methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl, decyl, undecyl and dodecyl, including their isomers, 15 for example, isopropyl, isobutyl, tert-butyl and isopentyl. Halogen may be fluorine, chlorine, bromine or iodine. Alkenyl, for example, may be prop-1-enyl, allyl, but-1-enyl, but-2-enyl or but-3-enyl, as well as a group having more than one double bond. Alkynyl may be, for example, prop-2-ynyl, propargyl, but-1-ynyl or but-2-ynyl. Cycloalkyl includes the following groups depending on the number of carbon atoms indicated: 20 cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl, cyclooctyl and cyclononyl. A saturated or unsaturated heterocyclic residue is preferably derived from a saturated or unsaturated 5- or 6-membered heterocycle containing from one to four of the same or different heteroatoms, for example, nitrogen, oxygen, or sulphur. Suitable examples of such heterocycles are tetrahydrofuran, furan, tetrahydrothiophene, thiophene, pyrrolidine, pyrrole, pyrroline, pyrazole, imidazole, triazole, tetrazole, pyrazoline, oxazole, 25 thiazole, isoxazole, isothiazole, pyran, dihydropyran, tetrahydropyran, thiopyran, dihydrothiopyran, tetrahydrothiopyran, pyridine, piperidine, pyridazine, dihydropyridazine, tetrahydropyridazine, pyrimidine, dihydropyrimidine, tetrahydropyrimidine, pyrazine, dihydropyrazine, tetrahydropyrazine, morpholine, thiazine, dihydrothiazine, tetrahydrothiazine, piperazine and triazine.

30 The compounds according to general formula I are oils, gums, or, predominantly, crystalline solid materials at ambient temperature (~20 °C). They are distinguished by their valuable biocidal properties and can be used in agriculture or related fields for the control of undesired plants, and find especial application in the selective control of broad-leaf weeds in, for example, cereal crops.

Good control of undesired plants may be achieved with preferred compounds of general formula I in which

35 X represents a group OH, OR⁷, OCOR⁷, NH₂, NHR⁷, NR⁷R⁸, or SR⁷;

R¹ to R⁴ each independently represents a hydrogen atom or a straight-chain or branched-chain C₁₋₆ alkyl group optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ group,

40 or R¹/R² and/or R³/R⁴ together form a straight-chain or branched-chain, saturated or unsaturated, C₃₋₅ methylene bridge, optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

45 R⁵ is straight-chain or branched-chain C₁₋₈ alkyl, C₂₋₈ alkenyl or a C₂₋₈ alkynyl group, or a C₃₋₈ cycloalkyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, $\frac{R^6}{3}$, SO₂ $\frac{R^6}{2}$ and SR⁶ groups, or a phenyl or naphthyl group, or a saturated or unsaturated 5- or 6-membered heterocyclic residue having from one to four heteroatoms selected from nitrogen, oxygen and sulphur, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

50 R⁶ is a straight-chain or branched-chain C₁₋₆ alkyl group, a C₃₋₆-cycloalkyl, phenyl, or naphthyl group, or a saturated or unsaturated 5- or 6-membered heterocyclic residue having from one to four heteroatoms selected from nitrogen, oxygen and sulphur, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, COOH, NO₂, 55

NH₂, SO₃H and SH groups; and
 R⁷ and R⁸ each independently has the same meaning as R⁵.

Preferred compounds of general formula I for control of various broad-leaf weeds are those in which

X represents a group OH, OR⁷, OCOR⁷, NH₂, NR⁷R⁸ or SR⁷;

R¹ to R⁴ each independently represents a hydrogen atom or a straight-chain or branched-chain C₁₋₄ alkyl group optionally substituted by one or more of the same or different substituents selected from halogen atoms and OH, OR⁶, COOR⁶, NH₂ and NR⁶ groups;

R⁵, R⁷ and R⁸ each independently represents a straight-chain or branched-chain C₁₋₆ alkyl group, optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, OR⁶, COOR⁶, NO₂, NH₂, NR⁶, SO₃H, SO₂R⁶ and SR⁶ groups, or a phenyl, naphthyl or thienyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶, SO₃H, SO₂R⁶ and SR⁶ groups; and

R⁶ represents a straight-chain or branched-chain C₁₋₄ alkyl group or a phenyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, COOH, NO₂, NH₂, SO₃H and SH groups.

Broad-leaf weed control may be achieved both pre-and post-emergence by especially preferred compounds of general formula I in which

X represents a group OH, OR⁷, OCOR⁷ or NH₂;

R¹ to R⁴ each independently represents a hydrogen atom or a C₁₋₂ alkyl group;

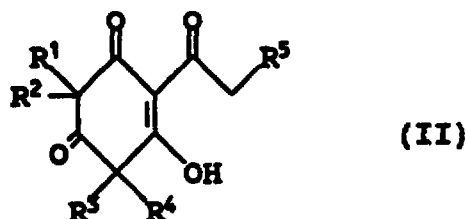
R⁵, R⁷ and R⁸ each independently represents a straight-chained or branched-chain C₁₋₄ alkyl group optionally substituted by one or more of the same or different substituents selected from halogen atoms and OR⁶ groups, or a phenyl, naphthyl or thienyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and R⁵, OR⁶ and SO₂R⁶ groups; and

R⁶ represents a methyl or trifluoromethyl group,

It will be appreciated that certain substituents will give rise to tautomeric forms of the compounds of general formula I. The present invention is to be understood to extend to all the various forms of the compounds of general formula I and mixtures thereof in whatever proportion. Thus the compounds of general formula I, their tautomers and mixtures thereof are included within the present invention, and the recitation of a compounds that exist in tautomeric form is to be understood as a recitation of their tautomers or tautomeric mixtures containing the recited compounds.

The compounds of general formula I wherein

R¹ to R⁶ are as defined above and X is an OH group, may be prepared by oxidising a compound of general formula II



wherein R¹ to R⁶ are as defined above and X is OH, which may be synthesised by the process disclosed in US Patent Specification No. 4,202,840. These compounds may be used as isolated materials, but it is advantageous to prepare them *in situ* and to oxidise them subsequently in a one-pot reaction.

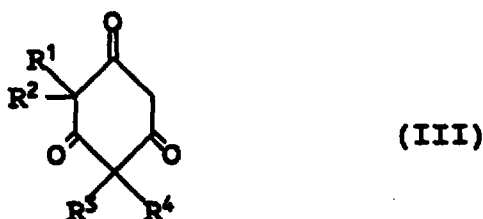
Preferably, the reaction is carried out as follows: Phlorphenacetophenone is used as such or alkylated with, for example, an alkyl halide, e.g. iodomethane, to yield the starting material of formula II. The subsequent oxidation may be carried out with any mild oxidising agent, such as iodine, permanganates, e.g. potassium permanganate, selenium dioxide and osmium tetroxide; however, the use of iodine or a permanganate has been found to be especially advantageous. The use of a particular solvent does not appear to be critical and both polar and aprotic solvents may be used, as well as mixtures of such solvents.

However, when iodine is used for oxidation it has been found that good results may be achieved by the use of polar solvents such as lower (C_{1-6} , preferably C_{1-4}) alcohols, for example methanol and ethanol.

The reaction is performed conveniently at a temperature in the range of from 0°C to 100°C , suitably in the range of from 5°C to 70°C . A reaction temperature of in the range of from 10°C to 50°C is a suitable reaction temperature for this preparation process.

After completion of the oxidation, the resulting compounds general formula I may be isolated by any suitable conventional procedure. For example, the solvent, such as methanol, and any excess of alkyl halide used for the preparation of the intermediate of general formula II in the first reaction step may be removed by distillation, the residue then being dissolved in water, acidified and extracted with an water-immiscible organic solvent. Any excess of oxidising agent may be removed by washing the organic layer with a mild reducing agent such as aqueous sodium bisulphite. The residue obtained after evaporation of the solvent may then be further purified by crystallisation or used directly for further derivatisation, for example esterification of the 1-hydroxyl group or replacement of this group by an amine.

An alternative method of preparing compounds of general formula I in which X is an OH group is to react a compound of general formula III



wherein R^1 to R^4 and R^5 are as defined above, with an acid chloride of general formula IV



wherein R^5 and R^6 are as defined above.

Preferably, the reaction is carried out as follows: The cyclohexane derivative of general formula III is reacted with the acid chloride of general formula IV, preferably in the presence of an acid scavenger, especially pyridine. The reaction is performed conveniently at a temperature in the range of from 0°C to 100°C , suitably in the range of from 5°C to 70°C . A temperature of in the range of from 10°C and 50°C is a suitable reaction temperature for this method of preparation. After removal of the solvent, the residue is then dissolved in a tertiary amine and treated with a catalytic amount of acetone cyanohydrin to yield a hydroxy compound of general formula I.

After completion of the reaction the compounds of general formula I may be isolated by any suitable conventional technique. For example, the solvent may be evaporated *in vacuo*, the residue dissolved in water and extracted after acidification with ethyl acetate. Final purification may also be achieved by procedures such as column chromatography on silica gel.

Compounds of general formulae III and IV are either known compounds or are preparable by conventional procedures.

Compounds of general formula I in which X is other than OH may be prepared from the corresponding hydroxy compounds, directly *in situ* or from the isolated compounds, by methods well known to those skilled in the art, for example by esterification of the hydroxy group, or replacement of that group by an amine substituent.

The present invention also provides a herbicidal composition which comprises a compound of general formula I in association with a carrier. Preferably there are at least two carriers in a composition of the invention, at least one of which is a surface-active agent.

The present invention further provides a method of combating undesired plant growth at a locus, which may, for example, be the soil or plants in a crop area, by treating the locus with a compound or composition of the present invention. Application may be pre- or post-emergence. The dosage of active ingredient used may, for example, be in the range of from 0.01 to 10 kg of active ingredient per hectare.

Also provided by the present invention is the use of a compound of the invention as a herbicide. The compounds are particularly useful in the control of broad-leaf weeds.

The compounds of general formula I may be used as such; however, they are preferably used in composition form comprising, in addition to the compounds of the invention, one or more adjuvants and auxiliaries, suitably solid and/or liquid compounds which are known for formulation purposes, and are conveniently in the form of, for example, emulsion concentrates, solutions which may be sprayed directly or diluted, emulsions, wettable powders, soluble powders, dusts, granulates and microcapsulates by conventional procedures. The form of application such as spraying, atomizing, dispersing and pouring and the composition form, may be selected according to the locus to be treated, the prevailing atmospheric conditions, etc..

The herbicidal compositions of the invention may be prepared by conventional procedures, e.g. intensive mixing and/or grinding of the active ingredients with other substances, such as fillers, solvents, solid carriers, and preferably surface-active compounds (tensides).

Solvents may be aromatic hydrocarbons, preferably the fractions C_8-12 , e.g. xylenes or xylene mixtures, substituted naphthalenes, phthalic acid esters, such as dibutyl or dioctyl phthalate, aliphatic hydrocarbons, e.g. cyclohexane or paraffins, alcohols and glycols as well as their ethers and esters, e.g. ethanol, ethylene glycol mono- and di-methyl ether, ketones such as cyclohexanone, strongly polar solvents such as N-methyl 2 pyrrolidone, dimethyl sulfoxide, alkyl formamides, epoxidised vegetable oils, e.g. epoxidised coconut or soybean oil, and water.

Solid carriers, which may be used for dusts or dispersible powders, may be mineral fillers, such as calcite, talc, kaolin, montmorillonite and attapulgite. The physical properties may be improved by addition of highly dispersed silica gel or highly dispersed polymers. Carriers for granulates may be porous material, e.g. pumice, broken brick, sepiolite, bentonite; non-sorptive carriers may be calcite or sand. Additionally, a multitude of pre-granulated inorganic or organic materials may be used, such as dolomite or crushed plant residues.

Suitable surface-active substances may be non-ionogenic, anionic or cationic tensides with good dispersing, emulsifying and wetting properties depending on the nature of the compound of general formula I to be formulated. Mixtures of tensides may also be used.

Suitable tensides may be so-called water-soluble soaps as well as water-soluble synthetic surface-active compounds.

Soaps usually are alkali metal, alkaline earth metal or optionally-substituted ammonium salts of higher fatty acids (C_{10-20}), e.g. the sodium or potassium salts of oleic or stearic acid or of mixtures of natural fatty acids which are prepared, for example, from coconut or tallow oil. Furthermore, methyl taurine salts of fatty acids may be used.

However, so-called synthetic tensides are preferably used, especially fatty sulphonates, fatty sulphates, sulphonated benzimidazole derivatives or alkyl aryl sulphonates.

The fatty sulphates or fatty sulphonates are normally used as alkali metal, alkaline earth metal or optionally substituted ammonium salts, and have an alkyl moiety of from 8 to 22 carbon atoms, whereby alkyl also means the alkyl moiety of acyl residues, such as the sodium or calcium salt of lignin sulphonic acid, of sulphuric acid dodecylate or of a mixture of fatty alcohols prepared from natural fatty acids. This also includes the salts of sulphuric acid esters, sulphonic acids and adducts of fatty alcohols and ethylene oxide. The sulphonated benzimidazole derivatives preferably contain 2 sulphonic acid residues and a fatty acid residue with from 8 to 22 carbon atoms. Alkyl aryl sulphonates are, for example, the sodium, calcium or triethyl ammonium salts of dodecyl benzene sulphonic acid, dibutyl naphthalene sulphonic acid or of a condensate of naphthalene sulphonic acid and formaldehyde.

Furthermore, phosphates, such as the salts of the phosphoric acid ester of a p-nonylphenol-(4-14)-ethylene oxide adduct or phospholipids, may be used.

Non-ionic tensides are preferably polyglycol ether derivatives of aliphatic or cycloaliphatic alcohols, saturated or non-saturated fatty acids and alkylphenols, which have from 3 to 10 glycol ether groups and from 8 to 20 carbon atoms in the (aliphatic) hydrocarbon residue and from 6 to 18 carbon atoms in the alkyl residue of the alkyl phenols.

Other suitable non-ionic tensides are the water-soluble, 20 to 250 ethylene glycol ether groups and 10 to 100 polypropylene glycol ether groups containing polyadducts of ethylene oxide and polypropylene glycol, ethylene diamino polypropylene glycol and alkyl polypropylene glycol with from 1 to 10 carbon atoms in the alkyl moiety; the substances suitably contain 1 to 5 ethylene glycol units per propylene glycol unit.

Examples of non-ionic tensides are nonylphenol polyethoxy ethanol, castor oil polyglycol ether, polyadducts of ethylene oxide and polypropylene, tributyl phenoxy polyethoxy ethanol, polyethylene glycol and octyl phenoxy polyethoxy ethanol.

Furthermore, fatty acid esters of polyoxyethylene sorbitan, such as polyoxyethylene sorbitan trioleate may be used.

Cationic tensides preferably are quaternary ammonium salts, which have at least one alkyl residue with from 8 to 22 carbon atoms and, furthermore, low, optionally halogenated alkyl, benzyl or hydroxyalkyl residues. The salts are preferably halides, methyl sulphates or alkyl sulphates, e.g. stearyl trimethyl ammonium chloride or benzyl bis(2-chlorethyl) ethyl ammonium bromide.

The tensides suitably used for the herbicidal compositions are described in the publications: "McCutcheon's Detergents and Emulsifiers Annual", MC Publishing Corp., Ridgewood, NJ, USA 1981; H. Stache, "Tensid-Taschenbuch", 2nd ed., C. Hanser, Munich, Vienna, 1981; M. and J. Ash, "Encyclopedia of Surfactants", vol. I-III, Chemical Publishing Co., New York, NY, USA 1980-1981.

The herbicidal compositions of the present convention suitably comprise from 0.1% to 95%, preferably from 0.1% to 80%, by weight of at least one compound of general formula I, from 1% to 99.9% of a solid or liquid adjuvant and from 0% to 25%, preferably 0.1% to 25%, of a tenside.

Preferred composition forms and components are as follows (all percentages are by weight):

Emulsion Concentrates:	
Active ingredient:	1% to 20%, preferably 5% to 10%
Surface-active substance:	5% to 30%, preferably 10% to 20%
Liquid carrier:	50% to 94%, preferably 70% to 85%

Suspension Concentrates:	
Active ingredient:	5% to 75%, preferably 10% to 50%
Water:	94% to 24%, preferably 88% to 30%
Surface-active substance:	1% to 40%, preferably 2% to 30%

Wettable Powder:	
Active ingredient:	0.5% to 90%, preferably 1% to 80%
Surface-active substance:	0.5% to 20%, preferably 1% to 15%
Solid carrier:	5% to 95%, preferably 15% to 90%

Dusts:	
Active ingredient:	0.1% to 10%, preferably 0.1% to 1%
Solid carrier:	99.9% to 90%, preferably 99.9% to 99%

Granulates:	
Active ingredient:	0.5% to 30%, preferably 3% to 15%
Solid carrier:	99.5% to 70%, preferably 97% to 85%

Preferably the compositions are formulated in a concentrated form which is subsequently diluted by the user before application. The compositions may be diluted to a concentration of 0.001% of active ingredient (a.i.) for use. As noted above, application dosages are suitably in the range of from 0.01 to 10 kg a.i./ha.

The compositions may also comprise other auxiliaries such as, stabilizers, defoamers, viscosity controlling agents, thickeners, adhesives, fertilisers or other active ingredients, for example compounds having fungicidal or insecticidal properties, or other herbicides.

The following Example illustrate the invention.

Example 11-Hydroxy-2-phenylglyoxyl-4,4,6,6-tetramethyl cyclohex-1-en-3,5-dione

5 Method A:

Phlorphenacetophenone (27.0 g, 0.11 mol) was added to a solution of sodium (12.7 g, 0.55 mol) in methanol (400 ml). Subsequently, iodomethane (125 g, 0.88 mol) was added dropwise and the temperature maintained at 20 °C; the resulting mixture was kept at room temperature for 5 days. Then iodine (25.3 g, 0.1 mol) was added in several portions whilst the pH-value was kept between pH 7.5 and pH 8.0 by continuous addition of sodium methylate. After removal of excess methanol and iodomethane by distillation, the residue was dissolved in water. The solution was carefully acidified by addition of 2N hydrochloric acid and then extracted with diethyl ether (200 ml) three times. The collected organic layers were washed once with 10% aqueous sodium bisulphite and once with water, dried over anhydrous magnesium sulphate, and then evaporated. The residue was crystallised from isopropanol to yield a micro-crystalline material (11.4 g, 33% of theoretical) of melting point 150 °C-152 °C, identified as the title compound.

Method B

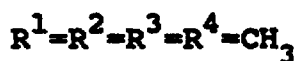
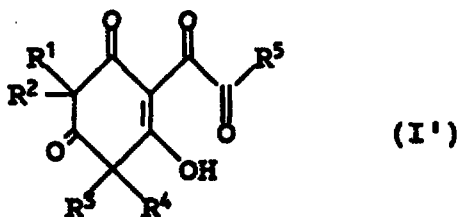
4,4,6,6-Tetramethyl-1-1,3,5-cyclohexantrione (100 mg, 0.55 mmol) was dissolved in pyridine (5 ml). Phenylglyoxylic acid chloride (93 mg, 0.55 mmol) was added and the mixture was kept at 50 °C for 2 days. Then the solvent was evaporated and the residue dissolved in triethylamine (10 ml). A catalytic amount of acetone cyanohydrin (2 drops, c. 0.1 ml) was added and the reaction mixture stirred for 24 hours. Subsequently, the solvent was evaporated in vacuo, the remaining residue dissolved in water (10 ml), acidified with 2N aqueous hydrogen chloride and the solution extracted with ethyl acetate. The separated organic layer was washed with water, dried, concentrated and applied onto a column packed with silica gel, eluting with dichloromethane/acetone (9:1, v/v). 1-Hydroxy-2-phenyl-glyoxyl-4,4,6,6-tetramethyl cyclohex-1-en-3,5-dione (93 mg, 54% of theoretical; melting point: 150 °C-152 °C) was obtained.

30 EXAMPLE 21-Amino-2-phenylglyoxyl-4,4,6,6-tetramethyl cyclo-hex-1-en-3,5-dione

1-Hydroxy-2-phenylglyoxyl-4,4,6,6-tetramethyl cyclohex-1-en-3,5-dione (7.1 g, 0.23 mol), prepared as in Example 1, was dissolved in methyl tert-butyl ether (70 ml). In an autoclave liquid ammonia (70 ml) was added to the solution and the mixture was stirred for 15 hours. Then the ammonia was evaporated off and the resulting solution was washed with a solution of concentrated hydrochloric acid (8 ml) in water (70 ml). The aqueous layer was separated and extracted twice with diethyl ether. The collected organic layers were washed twice with aqueous sodium hydroxide and once with water, dried over anhydrous magnesium sulphate and then evaporated. The colourless micro-crystalline product (4.0 g, 57% of theoretical) melted at 139 °C with decomposition and was identified as the title compound.

The identification data for the compounds of Examples 1 and 2 above, and for further compounds of general formula I prepared by analogous procedures to those described in Examples 1 to 3, are given in Table 1 below.

Further examples of compounds of general formula I encompassed by the present invention, and preparable by analogous procedures to those described in Examples 1 to 3 above, are given in Table 2 below.

Table 1

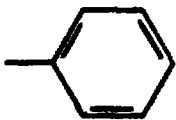
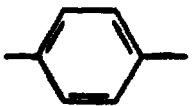
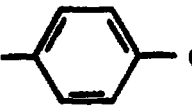
Ex. No.	R ⁵	Melting Point (°C)	¹ H-NMR
1		150-152	1.32 (s, 6H), 1.56 (s, 6H), 7.49 (dd, 2H) 7.60 (t, 1H), 7.89 (d, 2H), 15.9 (br s, 1H)
3	 Me	153-155	1.35 (s, 6H), 1.78 (s, 6H), 2.44 (s, 3H), 7.33 (d, 2H), 7.89 (d, 2H), 15.7 (br s, 1H)
4	 OMe	137-138	1.37 (s, 6H), 1.58 (s, 6H), 3.90 (s, 3H), 6.99 (d, 2H), 7.84 (d, 2H)

Table 1 (continued)

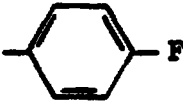
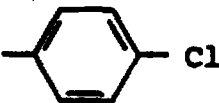
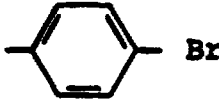
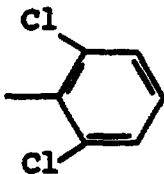
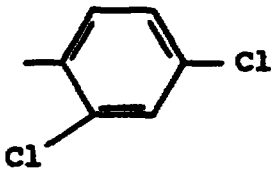
Ex. No.	R ⁵	Melting Point (°C)	¹ H-NMR
5		135-136	1.38 (s, 6H), 1.60 (s, 6H), 7.21 (d, 2H), 7.94 (m, 2H)
6		152-155	1.45 (s, 12H), 7.50 (s, 4H)
7		161-164	1.37 (s, 6H), 1.78 (s, 6H), 7.70 (t, 2H), 7.74 (t, 2H)
8		oil	1.38 (s, 6H), 1.43 (s, 6H), 7.08-7.35 (m, 3H), 13.80 (s, 1H)
9		152-155	1.35 (s, 6H), 1.60 (s, 6H), 7.43 (m, 3H), 8.12 (d, 1H)

Table 1 (continued)

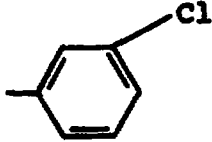
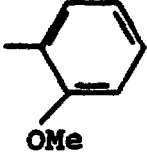
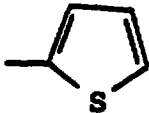
Ex. No.	R ⁵	Melting Point (°C)	¹ H-NMR
10		138-140	1.37 (s, 6H), 1.59 (s, 6H), 7.46 (t, 1H), 7.60 (d, 1H), 7.75 (d, 1H), 7.83 (d, 1H)
11		152-154	1.37 (s, 6H), 1.59 (s, 6H), 3.74 (s, 3H), 6.98 (d, 1H), 7.12 (t, 1H), 7.59 (t, 1H), 8.13 (d, 1H), 15.75 (br s, 1H)
12		158-161	1.38 (s, 6H), 1.58 (s, 6H), 7.22 (dd, 1H), 7.67 (d, 1H), 7.83 (d, 1H)
13	-n-Pr	oil	0.97 (t, 3H), 1.39 (s, 6H), 1.45 (s, 6H), 1.64 (quint, 2H), 2.99 (dd, 2H), 13.31 (s, 1H)

Table 1 (continued)

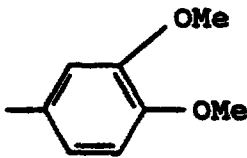
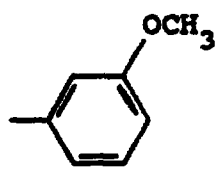
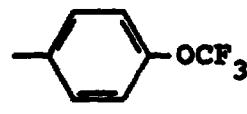
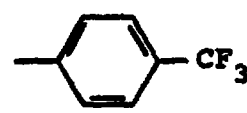
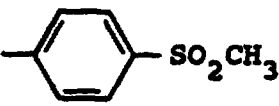
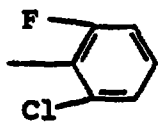
Ex. No.	R ⁵	Melting Point (°C)	¹ H-NMR
14	-n-Bu	oil	0.89 (t, 3H), 1.38 (m, 9H), 1.43 (s, 6H), 1.64 (m, 2H), 2.98 (t, 2H), 13.31 (s, 1H)
15		oil	1.31 (s, 6H), 1.43 (s, 6H), 3.86 (s, 3H), 3.89 (s, 3H), 6.82 (m, 3H), 13.63 (s, 1H)
16		109-112	1.33 (s, 6H), 1.55 (s, 6H), 3.84 (s, 3H), 7.15 (d, 1H), 7.36 (m, 2H), 7.46 (m, 1H)
17		133-136	1.32 (s, 6H), 1.56 (s, 6H), 7.33 (d, 2H), 7.92 (d, 2H)
18			1.74 (br.s, 12H) 7.97 (m, 4H)

Table 1 (continued)

Ex. No.	R ⁵	Melting Point (°C)	¹ H-NMR
19			1.70 (br.s, 12H), 3.36 (s, 3H), 8.02 (d, 2H), 8.21 (d, 2H)
20		139-141	1.43 (s, 12H), 7.22 (dd, 1H), 7.41 (d, 1H), 7.57 (dt, 1H)

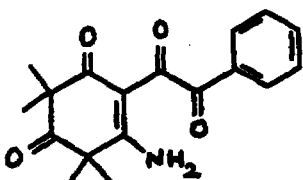
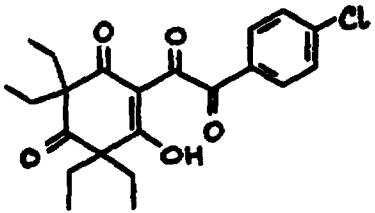
Ex. No.	Compound	Melting Point (°C)	¹ H-NMR
2		139 (d.)	1.35 (m, 12H), 7.20 (m, 5H)
21		oil	0.78 (2t, 6H), 0.87 (t, 3H), 0.98 (t, 3H), 1.87 (m, 4H), 2.00 (dt, 2H), 2.29 (dt, 2H), 7.42 (d, 2H), 7.54 (d, 2H)

Table 1 (continued)

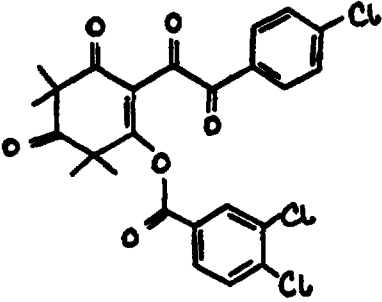
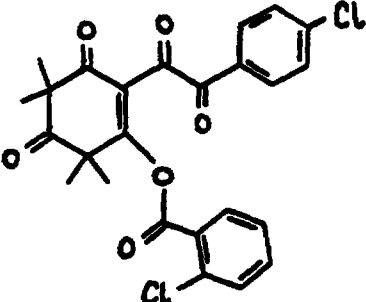
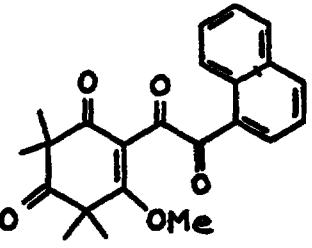
Ex. No.	Compound	Melting Point (°C)	¹ H-NMR
22		204-206	
23		178-180	
24		166-168	1.38 (s, 3H), 1.42 (s, 3H), 1.62 (s, 3H), 1.72 (s, 3H), 3.53 (s, 3H), 7.39 (t, 1H), 7.77 (m, 3H), 7.90 (2d, 2H), 8.79 (d, 1H)

Table 1 (continued)

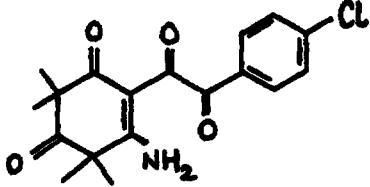
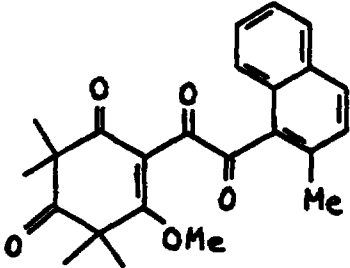
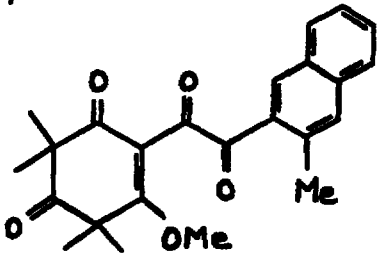
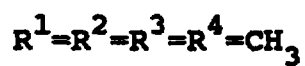
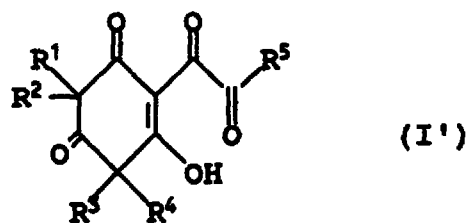
Ex. No.	Compound	Melting Point (°C)	¹ H-NMR
25		>200 (d.)	1.23 (s, 6H), 1.54 (s, 3H), 1.60 (s, 3H), 2.49 (s, 1H), 7.43 (m, 4H), 7.53 (s, 1H), 10.58 (s, 1H)
26		gum	1.44 (s, 3H), 1.45 (s, 3H), 1.67 (s, 3H), 1.67 (s, 3H), 2.55 (s, 3H), 3.40 (s, 3H), 7.22 (d, 1H), 7.46 (m, 2H), 7.77 (m, 2H), 8.61 (d, 1H)
27		150-151	1.37 (s, 3H), 1.43 (s, 3H), 1.59 (s, 3H), 1.72 (s, 3H), 2.71 (s, 3H), 3.54 (s, 3H), 7.24 (d, 1H), 7.44 (d, 1H), 7.82 (m, 2H), 8.03 (d, 1H), 8.81 (d, 1H)

Table 2

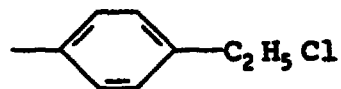
Compound No.

 R^5

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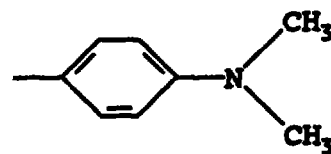
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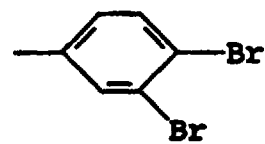
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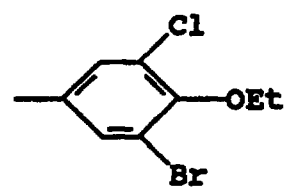
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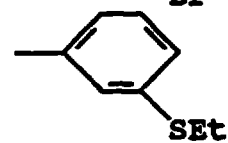
5.



6.



7.



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Table 2 (continued)

5

Compound No.

 R^5

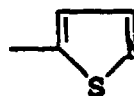
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8.



9.



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10.



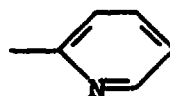
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11.



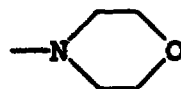
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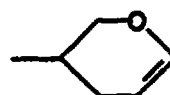
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13.



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14.



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Table 2 (continued)

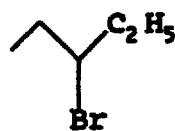
Compound No.	R ⁵
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5

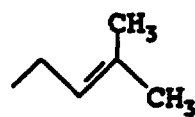
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17.



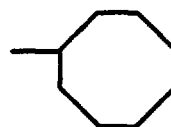
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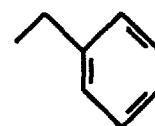
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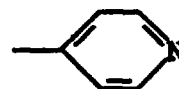
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20.



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21.



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Table 2 (continued)

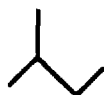
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Compound No.

 R^5

10

22.



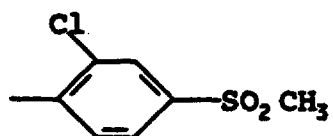
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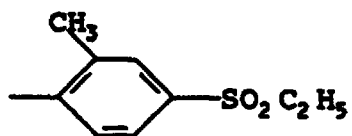
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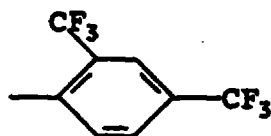
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25.



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Table 2 (continued)

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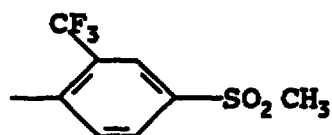
Compound No.

 R^5

10

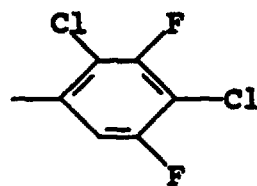
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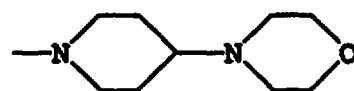
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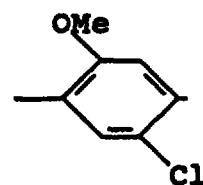
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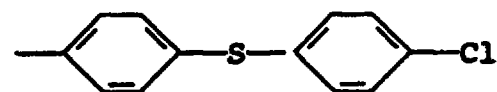
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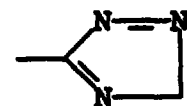
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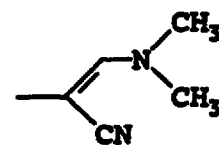
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32.



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33.



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Table 2 (continued)

Compound No. R^5

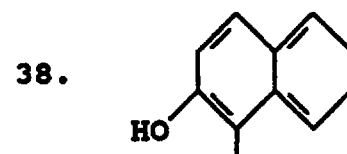
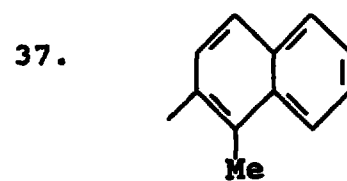
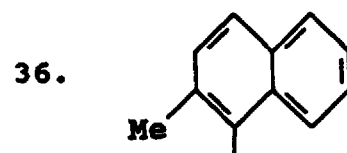
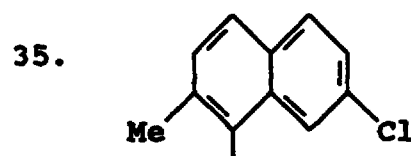
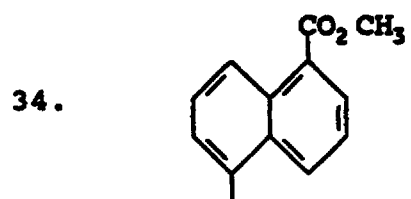
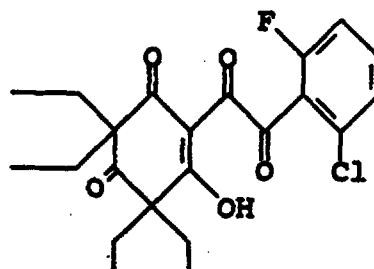
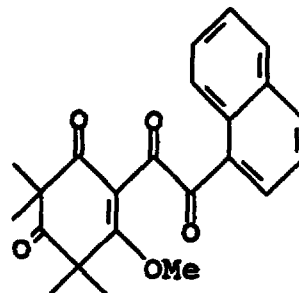


Table 2 (continued)
Compound No. R^5

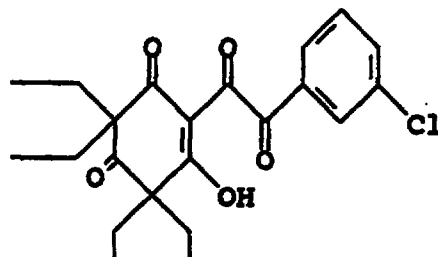
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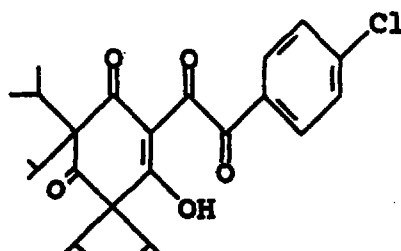
40.



41.



42.



43.

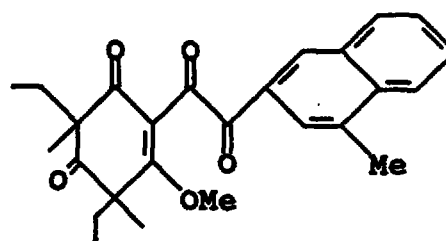
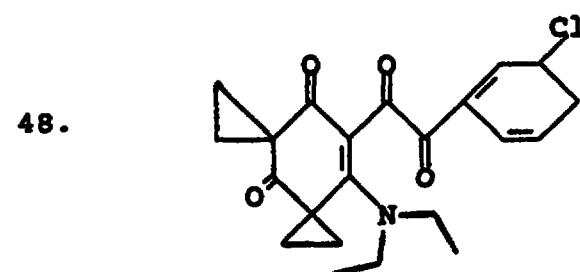
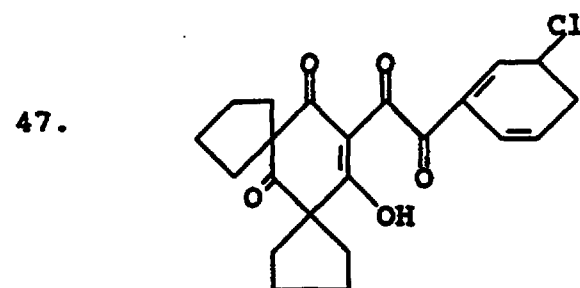
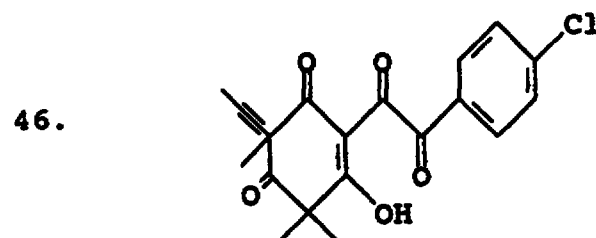
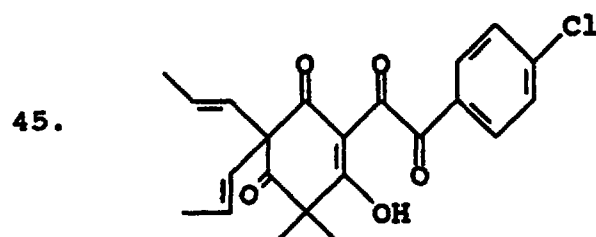
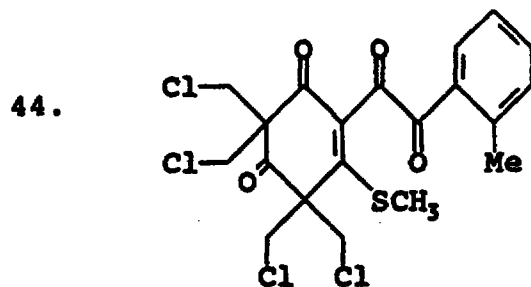


Table 2 (continued)

Compound No.

 R^5 

Biological ActivityHerbicidal effect of pre-emergence treatment ("pre-em")

- 5 The test plants were sown in pots at a depth of 2 cm, and on the same day the surface of the covering soil was sprayed using a spray volume of 800 l/ha and a dose rate corresponding to 2 kg/ha, using a band sprayer. The treated pots were then placed into a greenhouse. The herbicidal effect was determined after three weeks in comparison with untreated controls, using an evaluation scale of 1 to 9, where 1 represents a 100% effect and 9 no effect at all.
- 10 In this scale

1	indicates 100%
2	indicates 100 - 97.5%
3	indicates 97.5 - 95%
4	indicates 95 - 90%
5	indicates 90 - 85%
6	indicates 85 - 75%
7	indicates 75 - 65%
8	indicates 65 - 32.5%
9	indicates 32.5 - 0% activity

Herbicidal effect of post-emergence treatment ("post-em")

- 25 The test plants were sown in pots at a depth of 2 cm and precultivated to the 2.5 leaf stage (monocotyledons/grasses) or to the 1.5 metaphyll stage. The leaves were then sprayed with a band sprayer at a spray volume of 800 l/ha and a dose rate corresponding to 2 kg/ha, and the treated pots were placed into a greenhouse. The herbicidal effect was determined after three weeks in comparison to untreated controls. The evaluation method was the same as that used for the pre-emergence treatment.
- 30 The tests were carried out on the following plants:

AVEFA	<u>Avena fatua</u> L.
ALOMY	<u>Alopecurus myosuroides</u> Huds.
ECHCG	<u>Echinochloa crus-galli</u> (L.) P. Beauv.
35 SINAL	<u>Sinapis alba</u> L.
LYPES	<u>Lycopersicon esculentum</u> Mill.
BEAVA	<u>Beta vulgaris</u> L. var. <u>altissima</u>
CYPES	<u>Cyperus esculentus</u> L.

The results of these tests are shown in the following table, Table 3:

Table 3

Compound of Example No.	PRE-EM							POST-EM						
	AVE FA	ALO MY	ECH CG	SIN AL	LYP ES	BEA VA	CYP ES	AVE FA	ALO MY	ECH CG	SIN AL	LYP ES	BEA VA	CYP ES
1	4	4	2	2	2	2	2	6	6	2	2	4	6	2
3	6	6	6	2	6	6	6	6	6	6	2	4	6	6
4	4	4	2	2	2	2	4	6	6	2	2	2	6	2
5	6	6	1	1	1	2	2	4	6	1	2	2	2	2
6	6	6	2	2	2	2	6	6	6	1	2	2	2	4
7	6	6	2	2	2	2	4	6	6	2	2	2	4	2
8	6	6	6	6	4	6	6	6	6	2	2	4	1	6

Table 3 (continued)

Compound of Example No.	PRE-EM							POST-EM						
	AVE FA	ALO MY	ECH CG	SIN AL	LYP ES	BEA VA	CYP ES	AVE FA	ALO MY	ECH CG	SIN AL	LYP ES	BEA VA	CYP ES
9	6	6	6	2	4	2	4	6	6	6	2	6	6	2
10	6	6	4	2	2	4	6	6	6	4	2	2	4	4
11	6	6	4	2	4	4	6	6	6	4	2	4	2	2
12	6	6	6	2	4	4	4	6	6	6	4	2	4	6
13								6	6	4	4	2	1	4
14								4	4	4	1	1	1	4
17	6	6	2	2	6	4	6	4	6	1	2	2	4	4

Table 3 (continued)

Compound of Example No.	PRE-EM							POST-EM						
	AVE FA	ALO MY	ECH CG	SIN AL	LYP ES	BEA VA	CYP ES	AVE FA	ALO MY	ECH CG	SIN AL	LYP ES	BEA VA	CYP ES
18	6	6	2	2	2	4	6	4	6	1	2	2	2	4
19								6	6	6	4	4	6	6
20	6	6	4	2	2	4	4	6	6	2	2	4	1	6
23	6		6	4	4			6		6	6	4		
24	6		4	2	4	4		6		6	2	4		
25	6	6	4	2	2	2	4	6	6	4	2	2	4	6
26	6	6	6	6	4	6	6	6	6	6	4	4	6	6

Table 3 (continued)

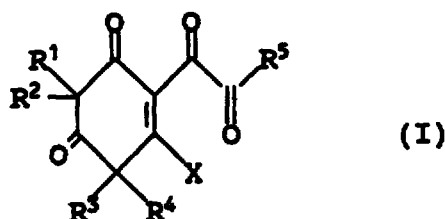
Compound of Example No.	PRE-EM								POST-EM							
	AVE	ALO	ECH	SIN	LYP	BEA	CYP		AVE	ALO	ECH	SIN	LYP	BEA	CYP	
27	FA	MY	CG	AL	ES	VA	ES		FA	MY	CG	AL	ES	VA	ES	
	6	6	6	6	4	6	6		6	6	6	4	4	6	4	



Claims

Claims for the following Contracting States : AT, BE, CH, DE, DK, FR, GB, GR, IT, LI, LU, NL, SE

1. A compound of the general formula I



characterised in that

X represents a group OH, OR⁷, NH₂, NHR⁷, NR⁷R⁸, NOH, NOR⁷, OCOR⁷, OCONHR⁷, OCONHCOR⁷, OCONHSO₂R⁷, NHCONHCOR⁷, OCOSR⁷, NHCOSR⁷, SCOR⁷, SR⁷, SCONHR⁷, SCONHCOR⁷, SCONHSO₂R⁷ or SCOSR⁷;

R¹ to R⁴ each independently represents a hydrogen atom or a straight-chain or branched-chain C₁₋₆ alkyl, C₂₋₆ alkenyl or C₂₋₆ alkynyl group which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

or

R¹ and R² and/or R³ and R⁴ together form a straight-chain or branched-chain, saturated or unsaturated, C₂₋₆-methylene bridge, optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

R⁵ is a straight-chain or branched-chain C₁₋₂₀ alkyl, C₂₋₂₀ alkenyl or C₂₋₂₀ alkynyl group, a C₃₋₂₀ cycloalkyl, phenyl or naphthyl group, or a saturated or unsaturated heterocyclic residue having one or more heteroatoms,

each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

R⁶ is a straight-chain or branched-chain C₁₋₂₀ alkyl group, a C₃₋₂₀ cycloalkyl, phenyl or naphthyl group, or a saturated or unsaturated heterocyclic residue having one or more heteroatoms,

each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, COOH, NO₂, NH₂, SO₃H and SH groups;

and

R⁷ and R⁸ each independently has the same meaning as R⁶.

2. A compound as claimed in claim 1 wherein

X represents a group OH, OR⁷, OCOR⁷, NH₂, NHR⁷, NR⁷R⁸, or SR⁷;

R¹ to R⁴ each independently represents a hydrogen atom or a straight-chain or branched-chain C₁₋₆ alkyl group optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ group,

or

R¹/R² and/or R³/R⁴ together form a straight-chain or branched-chain, saturated or unsaturated, C₃₋₅ methylene bridge, optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

R⁵ is straight-chain or branched-chain C₁₋₈ alkyl, C₂₋₈ alkenyl or a C₂₋₈ alkynyl group, or a C₃₋₈ cycloalkyl group, each of which is optionally substituted by



5 one or more of the same or different substituents selected from halogen atoms and CN, OH, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups, or a phenyl or naphthyl group, or a saturated or unsaturated 5- or 6-membered heterocyclic residue having from one to four heteroatoms selected from nitrogen, oxygen and sulphur, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

10 R⁶ is a straight-chain or branched-chain C₁₋₆ alkyl group, a C₃₋₆-cycloalkyl, phenyl, or naphthyl group, or a saturated or unsaturated 5- or 6-membered heterocyclic residue having from one to four heteroatoms selected from nitrogen, oxygen and sulphur, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, COOH, NO₂, NH₂, SO₃H and SH groups;

15 and
R⁷ and R⁸ each independently has the same meaning as R⁵.

3. A compound as claimed in claim 1 or claim 2 wherein

20 X represents a group OH, OR⁷, OCOR⁷, NH₂, NR⁷R⁸, or SR⁷;
R¹ to R⁴ each independently represents a hydrogen atom or a straight-chain or branched-chain C₁₋₄ alkyl group optionally substituted by one or more of the same or different substituents selected from halogen atoms and OH, OR⁶, COOR⁶, NH₂ and NR⁶₂ groups;

25 R⁵, R⁷ and R⁸ each independently represents a straight-chain or branched-chain C₁₋₆ alkyl group, optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups, or a phenyl, naphthyl or thienyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups; and

30 R⁶ represents a straight-chain or branched-chain C₁₋₄ alkyl group or a phenyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, COOH, NO₂, NH₂, SO₃H and SH groups.

35 4. A compound as claimed in any one of claims 1 to 3, wherein

X represents a group OH, OR⁷, OCOR⁷ or NH₂;
R¹ to R⁴ each independently represents a hydrogen atom or a C₁₋₂ alkyl group;
40 R⁵, R⁷ and R⁸ each independently represents a straight-chained or branched-chain C₁₋₄ alkyl group optionally substituted by one or more of the same or different substituents selected from halogen atoms and OR⁶ groups, or a phenyl, naphthyl or thienyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and R⁶, OR⁶ and SO₂R⁶ groups; and

45 R⁶ represents a methyl or trifluoromethyl group,

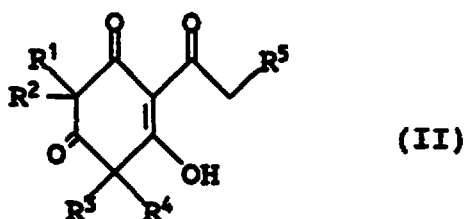
5. A process for the preparation of a compound of general formula I as claimed in claim 1, which comprises

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(a) oxidising a compound of general formula II

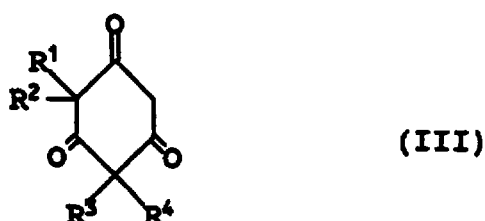


in which

R¹ to R⁶ are as defined in claim 1,

or

b) reacting a compound of general formula III



in which R¹ to R⁴, and R⁶, are as defined in claim 1, with an acid chloride of general formula IV

ClCO-CO-R⁵

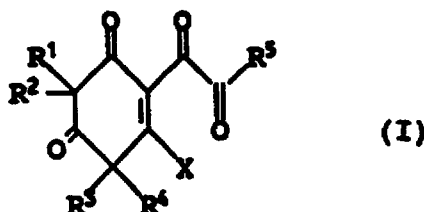
in which R⁵, and R⁶, are as defined in claim 1, and then, if desired, converting a resulting compound of general formula I in which X represents an OH group into a compound of general formula I in which X is other than an OH group.

- 35
6. A process as claimed in claim 5, wherein reaction a) is carried out using an oxidising agent which is iodine or a permanganate.
- 40
7. A process as claimed in claim 5 or claim 6, wherein the oxidation reaction is carried out in situ.
8. A herbicidal composition which comprises a compound as claimed in any one of claims 1 to 4, in association with a carrier.
- 45
9. A method of combating undesired plant growth at a locus which comprises treating the locus with a compound as claimed in any one of claims 1 to 4, or with a composition as claimed in claim 8.
10. The use of a compound as claimed in any one of claims 1 to 4, or of a composition as claimed in claim 8, in combating undesired plant growth.



Claims for the following Contracting State : ES

1. A process for the preparation of a compound of general formula I in which



X represents a group OH, OR⁷, NR₂, NHR⁷, NR⁷R⁸, NOH, NOR⁷, OCOR⁷, OCONHR⁷, OCONHCOR⁷, OCONHSO₂R⁷, NHCONHCOR⁷, OCOSR⁷, NHCOSR⁷, SCOR⁷, SR⁷, SCONHR⁷, SCONHCOR⁷, SCONHSO₂R⁷ or SCOSR⁷;

R¹ to R⁴ each independently represents a hydrogen atom or a straight-chain or branched-chain C₁₋₆ alkyl, C₂₋₆ alkenyl or C₂₋₅ alkynyl group which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

or

R¹ and R² and/or R³ and R⁴ together form a straight-chain or branched-chain, saturated or unsaturated, C₂₋₅-methylene bridge, optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

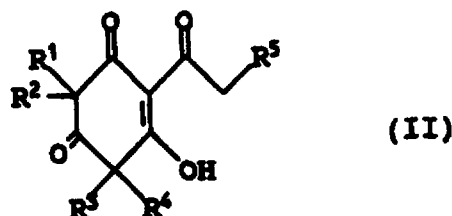
R⁵ is a straight-chain or branched-chain C₁₋₂₀ alkyl, C₂₋₂₀ alkenyl or C₂₋₂₀ alkynyl group, a C₃₋₂₀ cycloalkyl, phenyl or naphthyl group, or a saturated or unsaturated heterocyclic residue having one or more heteroatoms,

each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ and SR⁶ groups;

R⁶ is a straight-chain or branched-chain C₁₋₂₀ alkyl group, a C₃₋₂₀ cycloalkyl, phenyl or naphthyl group, or a saturated or unsaturated heterocyclic residue having one or more heteroatoms, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, COOH, NO₂, NH₂, SO₃H and SH groups; and

R⁷ and R⁸ each independently has the same meaning as R⁵, which comprises.

(a) oxidising a compound of general formula II



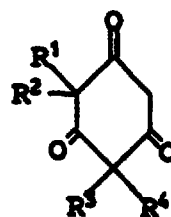
in which

R¹ to R⁶ are as defined as above,

or



b) reacting a compound of general formula III



(III)

in which R^1 to R^4 , and R^5 , are as defined above, with an acid chloride of general formula IV

ClCO-CO-R^5 (IV)

in which R^5 , and R^6 , are as defined as above, and then, if desired, converting a resulting compound of general formula I in which X represents an OH group into a compound of general formula I in which X is other than an OH group.

2. A process as claimed in claim 1, wherein X represents a group OH, OR^7 , OCOR^7 , NH_2 , NHR^7 , NR^7R^8 , or SR^7 .

R^1 to R^4 each independently represents a hydrogen atom or a straight-chain or branched-chain C_{1-6} alkyl group optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R^6 , OR^6 , COOR^6 , NO_2 , NH_2 , NR^6_2 , SO_3H , SO_2R^6 and SR^6 group,

or

R^1/R^2 and/or R^3/R^4 together form a straight-chain or branched-chain, saturated or unsaturated, C_{3-5} methylene bridge, optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R^6 , OR^6 , COOR^6 , NO_2 , NH_2 , NR^6_2 , SO_3H , SO_2R^6 and SR^6 groups;

R^5 is straight-chain or branched-chain C_{1-8} alkyl, C_{2-8} alkenyl or a C_{2-8} alkynyl group, or a C_{3-8} cycloalkyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, OR^6 , COOR^6 , NO_2 , NR_2 , NR^6_2 , SO_3H , SO_2R^6 and SR^6 groups, or a phenyl or naphthyl group, or a saturated or unsaturated 5- or 6-membered heterocyclic residue having from one to four heteroatoms selected from nitrogen, oxygen and sulphur, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R^6 , OR^6 , COOR^6 , NO_2 , NH_2 , NR^6_2 , SO_3H , SO_2R^6 and SR^6 groups;

R^6 is a straight-chain or branched-chain C_{1-6} alkyl group, a C_{3-6} -cycloalkyl, phenyl, or naphthyl group, or a saturated or unsaturated 5- or 6-membered heterocyclic residue having from one to four heteroatoms selected from nitrogen, oxygen and sulphur, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, COOH , NO_2 , NH_2 , SO_3H and SH groups;

and

R^7 and R^8 each independently has the same meaning as R^5 .

3. A process as claimed in claim 1 or claim 2 wherein

X represents a group OH, OR^7 , OCOR^7 , NH_2 , NR^7R^8 , or SR^7 ;

R^1 to R^4 each independently represents a hydrogen atom or a straight-chain or branched-chain C_{1-4} alkyl group optionally substituted by one or more of the same or different substituents selected from halogen atoms and OH, OR^6 , COOR^6 , NR_2 and NR^6_2 groups;



5 R^5 , R^7 and R^8 each independently represents a straight-chain or branched-chain C_{1-6} alkyl group, optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, OR^6 , $COOR^6$, NO_2 , NH_2 , NR^6_2 , SO_3H , SO_2R^6 and SR^6 groups, or a phenyl, naphthyl or thienyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, R^6 , OR^6 , $COOR^6$, NO_2 , NH_2 , NR^6_2 , SO_3H , SO_2R^6 and SR^6 groups; and

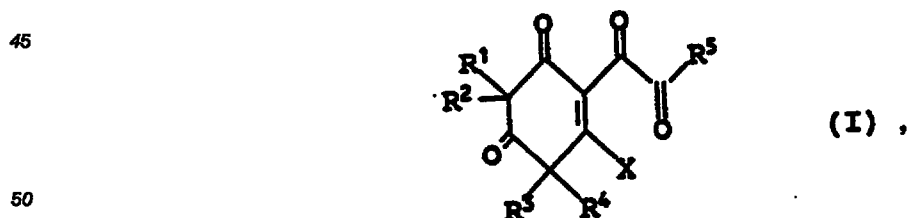
10 R^6 represents a straight-chain or branched-chain C_{1-4} alkyl group or a phenyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and CN, OH, COOH, NO_2 , NH_2 , SO_3H and SH groups.

4. A process as claimed in any one of claims 1 to 3, wherein
- 15 X represents a group OH, OR^7 , $OCOR^7$ or NH_2 ;
 R^1 to R^4 each independently represents a hydrogen atom or a C_{1-2} alkyl group;
 R^5 , R^7 and R^8 each independently represents a straight-chained or branched-chain C_{1-4} alkyl group optionally substituted by one or more of the same or different substituents selected from halogen atoms and OR^6 groups, or a phenyl, naphthyl or thienyl group, each of which is optionally substituted by one or more of the same or different substituents selected from halogen atoms and R^6 , OR^6 and SO_2R^6 groups; and
- 20 R^6 represents a methyl or trifluoromethyl group,
5. A process as claimed in any of claims 1-4, wherein reaction a) is carried out using an oxidising agent which is iodine or a permanganate.
- 25 6. A process as claimed in claim 5 wherein the oxidation reaction is carried out in situ.
7. A herbicidal composition which comprises a compound prepared according to the process as claimed in any one of claims 1 to 6, in association with a carrier.
- 30 8. A method of combating undesired plant growth at a locus which comprises treating the locus with a compound prepared according to the process as claimed in any one of claims 1 to 6, or with a composition as claimed in claim 7.
- 35 9. The use of a compound prepared according to the process as claimed in any one of claims 1 to 6, or of a composition as claimed in claim 7, in combating undesired plant growth.

Patentansprüche

40 Patentansprüche für folgende Vertragsstaaten : AT, BE, CH, DE, DK, FR, GB, GR, IT, LI, LU, NL, SE

1. Eine Verbindung mit der allgemeinen Formel I



dadurch gekennzeichnet, daß

X

55

R^1 bis R^4

eine Gruppe OH, OR^7 , NH_2 , NHR^7 , NR^7R^8 , NOH, NOR^7 , $OCOR^7$, $CONHR^7$, $CONHCOR^7$, $CONHSO_2R^7$, $NHCONHCOR^7$, $OCOSR^7$, $NHCOSR^7$, $SCOR^7$, SR^7 , $SCONHR^7$, $SCONHCOR^7$, $SCONHSO_2R^7$ oder $SCOSR^7$ bedeutet;
 unabhängig voneinander ein Wasserstoffatom oder eine geradket-



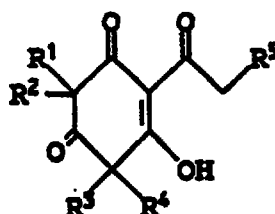
- 5 R^1 und R^2 und/oder R^3 und R^4 tige oder verzweigt-kettige C_{1-6} -Alkylgruppe, C_{2-6} -Alkenylgruppe oder C_{2-6} -Alkynylgruppe darstellen, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R^5 , OR^5 , $COOR^5$, NO_2 , NH_2 , NR^5_2 , SO_3H , SO_2R^5 und SR^5 , substituiert sind; oder zusammen eine geradkettige oder verzweigte, gesättigte oder ungesättigte C_{2-6} -Methylenbrücke bilden, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R^5 , OR^5 , $COOR^5$, NO_2 , NH_2 , NR^5_2 , SO_3H , SO_2R^5 und SR^5 , substituiert ist;
- 10 R^5 eine geradkettige oder verzweigt-kettige C_{1-20} -Alkylgruppe, C_{2-20} -Alkenylgruppe oder C_{2-20} -Alkynylgruppe, eine C_{3-20} -Cycloalkylgruppe, Phenyl- oder Naphthylgruppe, oder einen gesättigten oder ungesättigten heterocyclischen Rest mit einem oder mit mehreren Heteroatomen darstellt, wobei jede Gruppe gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R^5 , OR^5 , $COOR^5$, NO_2 , NH_2 , NR^5_2 , SO_3H , SO_2R^5 und SR^5 , substituiert ist;
- 15 R^6 eine geradkettige oder verzweigt-kettige C_{1-20} -Alkylgruppe, eine C_{3-20} -Cycloalkylgruppe, Phenyl- oder Naphthylgruppe oder einen gesättigten oder ungesättigten heterocyclischen Rest mit einem oder mit mehreren Heteroatomen bedeutet, wobei jede Gruppe gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, $COOH$, NO_2 , NH_2 , SO_3H und SH , substituiert ist; und unabhängig voneinander die gleiche Bedeutung wie R^5 aufweisen.
- 20 R^7 und R^8
- 25
2. Verbindung nach Anspruch 1, worin
- 30 X eine Gruppe OH, OR^7 , $OCOR^7$, NH_2 , NHR^7 , NR^7R^8 oder SR^7 darstellt; R^1 bis R^4 unabhängig voneinander ein Wasserstoffatom oder eine geradkettige oder verzweigt-kettige C_{1-6} -Alkylgruppe bedeutet, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R^5 , OR^5 , $COOR^5$, NO_2 , NH_2 , NR^5_2 , SO_3H , SO_2R^5 und SR^5 , substituiert ist; oder
- 35 R^1/R^2 und/oder R^3/R^4 gemeinsam eine geradkettige oder verzweigt-kettige, gesättigte oder ungesättigte C_{3-5} -Methylenbrücke ausbilden, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R^5 , OR^5 , $COOR^5$, NO_2 , NH_2 , NR^5_2 , SO_3H , SO_2R^5 und SR^5 , substituiert ist;
- 40 R^5 eine geradkettige oder verzweigt-kettige C_{1-8} -Alkylgruppe, C_{2-8} -Alkenylgruppe oder C_{2-8} -Alkynylgruppe oder eine C_{3-8} -Cycloalkylgruppe bedeutet, die jeweils gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R^5 , OR^5 , $COOR^5$, NO_2 , NH_2 , NR^5_2 , SO_3H , SO_2R^5 und SR^5 , substituiert ist, oder eine Phenyl- oder Naphthylgruppe oder einen gesättigten oder ungesättigten, 5- oder 6-gliedrigen heterocyclischen Rest bedeutet, der ein bis vier, unter Stickstoff, Sauerstoff und Schwefel ausgewählte Heteroatome aufweist, wobei jede dieser Gruppen gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R^5 , OR^5 , $COOR^5$, NO_2 , NH_2 , NR^5_2 , SO_3H , SO_2R^5 und SR^5 , substituiert ist;
- 45 R^6 eine geradkettige oder verzweigt-kettige C_{1-6} -Alkylgruppe, eine C_{3-6} -Cycloalkylgruppe, Phenyl- oder Naphthylgruppe oder einen gesättigten oder ungesättigten, 5- oder 6-gliedrigen heterocyclischen Rest mit ein bis vier, unter Stickstoff, Sauerstoff und Schwefel ausgewählten Heteroatomen bedeutet, wobei jede Gruppe gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen
- 50
- 55

R⁷ und R⁸

und CN, OH, COOH, NO₂, NH₂, SO₃H und SH, substituiert ist; und jeweils unabhängig die gleiche Bedeutung wie R⁵ aufweisen.

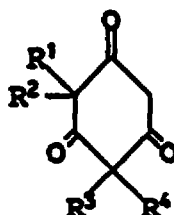
3. Verbindung nach Anspruch 1 oder 2, worin
- 5 X eine Gruppe OH, OR⁷, OCOR⁷, NH₂, NR⁷R⁸ oder SR⁷ bedeutet;
 R¹ bis R⁴ jeweils unabhängig ein Wasserstoffatom oder eine geradkettige oder verzweigt-kettige C₁₋₄-Alkylgruppe bedeutet, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und OH, OR⁵, COOR⁵, NH₂ und NR⁵₂, substituiert ist;
- 10 R⁵, R⁷ und R⁸ jeweils unabhängig eine geradkettige oder verzweigt-kettige C₁₋₆-Alkylgruppe, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, OR⁵, COOR⁵, NO₂, NH₂, NR⁵₂, SO₃H, SO₂R⁵ und SR⁵, substituiert ist, oder eine Phenyl-, Naphthyl- oder Thienylgruppe bedeuten, von denen jede gewünschtenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R⁵, OR⁵, COOR⁵, NO₂, NH₂, NR⁵₂, SO₃H, SO₂R⁵ und SR⁵, substituiert ist; und
- 15 R⁶ eine geradkettige oder verzweigt-kettige C₁₋₄-Alkylgruppe oder eine Phenylgruppe darstellt, von denen jede gewünschtenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, COOH, NO₂, NH₂, SO₃H und SH, substituiert ist.
- 20
4. Verbindung nach einem der Ansprüche 1 bis 3, worin
- 25 X eine Gruppe OH, OR⁷, OCOR⁷ oder NH₂ bedeutet;
 R¹ bis R⁴ jeweils unabhängig ein Wasserstoffatom oder eine C₁₋₂-Alkylgruppe bedeuten;
 R⁵, R⁷ und R⁸ jeweils unabhängig eine geradkettige oder verzweigt-kettige C₁₋₄-Alkylgruppe, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und OR⁵-Gruppen, substituiert ist, oder eine Phenyl-, Naphthyl- oder Thienylgruppe darstellen, von denen jede gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und R⁵-, OR⁵- und SO₂R⁵-Gruppen, substituiert ist; und
- 30 R⁶ eine Methyl- oder Trifluormethylgruppe bedeutet.

- 35 5. Verfahren zur Herstellung einer Verbindung der allgemeinen Formel I, wie in Anspruch 1 beansprucht, welches
- (a) ein Oxidieren einer Verbindung der allgemeinen Formel II



(II) ,

- worin R¹ bis R⁶ wie in Anspruch 1 definiert sind, oder
- (b) ein Umsetzen einer Verbindung der allgemeinen Formel III



(III) ,



worin R^1 bis R^4 und R^5 wie in Anspruch 1 definiert sind, mit einem Säurechlorid der allgemeinen Formel IV

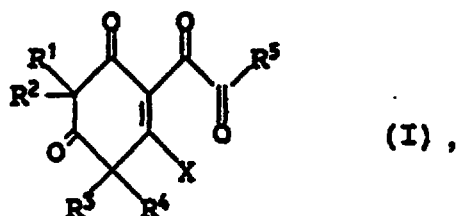


worin R^5 und R^6 wie in Anspruch 1 definiert sind, und gewünschtenfalls ein anschließendes Überführen einer erhaltenen Verbindung der allgemeinen Formel I, worin X für eine OH-Gruppe steht, in eine Verbindung der allgemeinen Formel I, worin X eine andere Bedeutung als eine OH-Gruppe aufweist, umfaßt.

6. Verfahren nach Anspruch 5, worin die Umsetzung a) unter Anwendung eines Oxidationsmittels, bei dem es sich um Iod oder um ein Permanganat handelt, ausgeführt wird.
7. Verfahren nach Anspruch 5 oder 6, worin die Oxidationsreaktion in situ vorgenommen wird.
8. Herbizide Zusammensetzung, die eine Verbindung, wie in einem der Ansprüche 1 bis 4 beansprucht, in Assoziation mit einem Träger umfaßt.
9. Verfahren zur Bekämpfung von unerwünschtem Pflanzenwuchs an einem Ort, welches ein Behandeln des Ortes mit einer Verbindung, wie in einem der Ansprüche 1 bis 4 beansprucht, oder mit einer Zusammensetzung, wie in Anspruch 8 beansprucht, umfaßt.
10. Verwendung einer Verbindung, wie in einem der Ansprüche 1 bis 4 beansprucht, oder einer Zusammensetzung, wie in Anspruch 8 beansprucht, in der Bekämpfung von unerwünschtem Pflanzenwuchs.

Patentansprüche für folgenden Vertragsstaat : ES

1. Verfahren zur Herstellung einer Verbindung mit der allgemeinen Formel I



worin
X

R^1 bis R^4

R^1 und R^2 und/oder R^3 und R^4

R^5

eine Gruppe OH, OR^7 , NH_2 , NHR^7 , NR^7R^8 , NOH , NOR^7 , OCOR^7 , OCONHR^7 , OCONHCO^7 , $\text{OCONHSO}_2\text{R}^7$, NHCONHCO^7 , OCOSR^7 , NHCOSR^7 , SCOR^7 , SR^7 , SCONHR^7 , SCONHCO^7 , $\text{SCONHSO}_2\text{R}^7$ oder SCOSR^7 bedeutet;

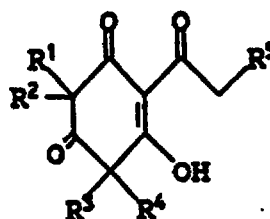
unabhängig voneinander ein Wasserstoffatom oder eine geradkettige oder verzweigt-kettige C_{1-6} -Alkylgruppe, C_{2-6} -Alkenylgruppe oder C_{2-6} -Alkynylgruppe darstellen, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R^6 , OR^6 , COOR^6 , NO_2 , NH_2 , NR^6_2 , SO_3H , SO_2R^6 und SR^6 , substituiert sind; oder zusammen eine geradkettige oder verzweigte, gesättigte oder ungesättigte C_{2-6} -Methylenbrücke bilden, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R^6 , OR^6 , COOR^6 , NO_2 , NH_2 , NR^6_2 , SO_3H , SO_2R^6 und SR^6 , substituiert ist; eine geradkettige oder verzweigt-kettige C_{1-20} -Alkylgruppe, C_{2-20} -Alkenylgruppe oder C_{2-20} -Alkynylgruppe, eine C_{3-20} -Cycloalkylgruppe, Phenyl- oder Naphthylgruppe, oder einen gesätt-

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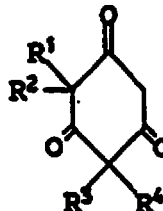
(a) ein Oxidieren einer Verbindung der allgemeinen Formel II



(II).

20

(b) ein Umsetzen einer Verbindung der allgemeinen Formel III



(III) .

worin R¹ bis R⁴ und R⁶ wie oben definiert sind, mit einem Säurechlorid der allgemeinen Formel IV



worin R⁵ und R⁶ wie oben definiert sind, und gewünschtenfalls ein anschließendes Überführen einer erhaltenen Verbindung der allgemeinen Formel I, worin X für eine OH-Gruppe steht, in eine Verbindung der allgemeinen Formel I, worin X eine andere Bedeutung als eine OH-Gruppe aufweist, umfaßt.

R¹ bis R⁴

x

R¹ bis R⁴

R¹/R² und/oder R³/R⁴

eine Gruppe OH, OR⁷, OCOR⁷, NH₂, NHR⁷, NR⁷R⁸ oder SR⁷ darstellt; unabhängig voneinander ein Wasserstoffatom oder eine geradkettige oder verzweigte C₁₋₆-Alkylgruppe bedeutet, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R⁶, OR⁶, COOR⁵, NO₂, NH₂, NR⁶, SO₃H, SO₂R⁶ und SR⁶, substituiert ist; oder gemeinsam eine geradkettige oder verzweigte, gesättigte oder ungesättigte C₃₋₅-Methylenbrücke ausbilden, die gegebenenfalls durch einen



- oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ und SR⁶, substituiert ist;
- 5 R⁵ eine geradkettige oder verzweigt-kettige C₁₋₈-Alkylgruppe, C₂₋₈-Alkenylgruppe oder C₂₋₈-Alkynylgruppe oder eine C₃₋₈-Cycloalkylgruppe bedeutet, die jeweils gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ und SR⁶, substituiert ist, oder eine Phenyl- oder Naphthylgruppe oder einen gesättigten oder ungesättigten, 5- oder 6-gliedrigen heterocyclischen Rest bedeutet, der ein bis vier, unter Stickstoff, Sauerstoff und Schwefel ausgewählte Heteroatome aufweist, wobei jede dieser Gruppen gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ und SR⁶, substituiert ist;
- 10 R⁶ eine geradkettige oder verzweigt-kettige C₁₋₆-Alkylgruppe, eine C₃₋₆-Cycloalkylgruppe, Phenyl- oder Naphthylgruppe oder einen gesättigten oder ungesättigten, 5- oder 6-gliedrigen heterocyclischen Rest mit ein bis vier, unter Stickstoff, Sauerstoff und Schwefel ausgewählten Heteroatomen bedeutet, wobei jede Gruppe gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, COOH, NO₂, NH₂, SO₃H und SH, substituiert ist; und
- 15 R⁷ und R⁸ jeweils unabhängig die gleiche Bedeutung wie R⁵ aufweisen.
- 25 3. Verfahren nach Anspruch 1 oder 2, worin
- X eine Gruppe OH, OR⁷, OCOR⁷, NH₂, NR⁷R⁸ oder SR⁷ bedeutet;
- R¹ bis R⁴ jeweils unabhängig ein Wasserstoffatom oder eine geradkettige oder verzweigt-kettige C₁₋₄-Alkylgruppe bedeutet, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und OH, OR⁶, COOR⁶, NH₂ und NR⁶₂, substituiert ist;
- 30 R⁵, R⁷ und R⁸ jeweils unabhängig eine geradkettige oder verzweigt-kettige C₁₋₆-Alkylgruppe, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ und SR⁶, substituiert ist, oder eine Phenyl-, Naphthyl- oder Thienylgruppe bedeuten, von denen jede gewünschtenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ und SR⁶, substituiert ist; und
- 35 R⁶ eine geradkettige oder verzweigt-kettige C₁₋₄-Alkylgruppe oder eine Phenylgruppe darstellt, von denen jede gewünschtenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und CH, OH, COOH, NO₂, NH₂, SO₃H und SH, substituiert ist.
- 40 4. Verfahren nach einem der Ansprüche 1 bis 3, worin
- X eine Gruppe OH, OR⁷, OCOR⁷ oder NH₂ bedeutet;
- R¹ bis R⁴ jeweils unabhängig ein Wasserstoffatom oder eine C₁₋₂-Alkylgruppe bedeuten;
- R⁵, R⁷ und R⁸ jeweils unabhängig eine geradkettige oder verzweigt-kettige C₁₋₄-Alkylgruppe, die gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und OR⁶-Gruppen, substituiert ist, oder eine Phenyl-, Naphthyl- oder Thienylgruppe darstellen, von denen jede gegebenenfalls durch einen oder mehrere, gleiche oder verschiedene Substituenten, ausgewählt unter Halogenatomen und R⁶-, OR⁶- und SO₂R⁶-Gruppen, substituiert ist; und
- 50 R⁶ eine Methyl- oder Trifluormethylgruppe bedeutet.
- 55 5. Verfahren nach einem der Ansprüche 1 bis 4, worin die Umsetzung a) unter Anwendung eines Oxidationsmittels, bei dem es sich um Iod oder um ein Permanganat handelt, ausgeführt wird.

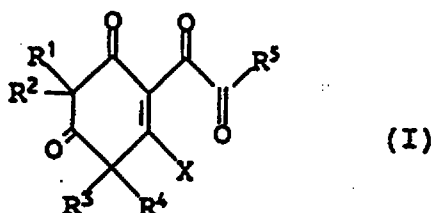


6. Verfahren nach Anspruch 5, worin die Oxidationsreaktion in situ vorgenommen wird.
7. Herbizide Zusammensetzung, die eine Verbindung, hergestellt nach dem Verfahren, wie in einem der Ansprüche 1 bis 6 beansprucht, in Assoziation mit einem Träger umfaßt.
- 5 8. Verfahren zur Bekämpfung von unerwünschtem Pflanzenwuchs an einem Ort, welches ein Behandeln des Ortes mit einer Verbindung, hergestellt nach dem Verfahren, wie in einem der Ansprüche 1 bis 6 beansprucht, oder mit einer Zusammensetzung, wie in Anspruch 7 beansprucht, umfaßt.
- 10 9. Verwendung einer Verbindung, hergestellt nach dem Verfahren, wie in einem der Ansprüche 1 bis 6 beansprucht, oder einer Zusammensetzung, wie in Anspruch 7 beansprucht, zur Bekämpfung von unerwünschtem Pflanzenwuchs.

Revendications

- 15 Revendications pour les Etats contractants suivants : AT, BE, CH, DE, DK, FR, GB, GR, IT, LI, LU, NL, SE

1. Un composé de la formule générale I



- 20 caractérisé en ce que

X représente un groupe OH, OR⁷, NH₂, NHR⁷, NR⁷R⁸, NOH, NOR⁷, OCOR⁷, OCONHR⁷, OCONH-COR⁷, OCONHSO₂R⁷, NHCONHCOR⁷, OCOSR⁷, NHCOSR⁷, SCOR⁷, SR⁷, SCONHR⁷, SCONHCOR⁷, SCONHSO₂R⁷, ou SCOSR⁷ ;

- 35 R¹ à R⁴ représentent chacun indépendamment un atome d'hydrogène ou un groupe C₁₋₆ alcoyle, C₂₋₆ alcényle ou C₂₋₆ alcynyle à chaîne droite ou à chaîne ramifiée qui est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶ ; ou

- 40 R¹ et R² et/ou R³ et R⁴ forment ensemble un pont de C₂₋₆-méthylène, saturé ou insaturé, à chaîne droite ou à chaîne ramifiée, éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶ ;

- 45 R⁵ est un groupe C₁₋₂₀ alcoyle, C₂₋₂₀ alcényle ou C₂₋₂₀ alcynyle à chaîne droite ou à chaîne ramifiée, un groupe C₃₋₂₀ cycloalcoyle, phényle ou naphthyle ou un résidu hétérocyclique saturé ou insaturé ayant un seul ou plusieurs hétéro-atomes, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶ ;

- 50 R⁶ est un groupe C₁₋₂₀ alcoyle à chaîne droite ou à chaîne ramifiée, un groupe C₃₋₂₀ cycloalcoyle, phényle ou naphthyle ou un résidu hétérocyclique saturé ou insaturé ayant un seul ou plusieurs hétéro-atomes, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, COOH, NO₂, NH₂, SO₃H et SH ; et

R⁷ et R⁸ ont chacun indépendamment la même Signification que R⁵.

2. Un composé selon la revendication 1, dans lequel

- 55 X représente un groupe OH, OCOR⁷, NH₂, NHR⁷, NR⁷R⁸ ou SR⁷ ;

R¹ à R⁴ représentent chacun indépendamment un atome d'hydrogène ou un groupe C₁₋₆ alcoyle à chaîne droite ou à chaîne ramifiée éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶,



COOR⁵, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶ ; ou

R¹ et R² et/ou R³ et R⁴ forment ensemble un pont de C₃₋₅ méthylène, saturé ou insaturé, à chaîne droite ou à chaîne ramifiée, éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶ ;

R⁵ est un groupe C₁₋₈ alcoyle, C₂₋₈ alcényle ou C₂₋₈ alcynyle à chaîne droite ou à chaîne ramifiée ou un groupe C₃₋₈ cycloalcoyle, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶, ou un groupe phényle ou naphthyle, ou un résidu hétérocyclique pentagonal ou hexagonal saturé ou insaturé ayant de 1 à 4 hétéro-atomes choisis parmi l'azote, l'oxygène et le soufre, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶ ;

R⁶ est un groupe C₁₋₆ alcoyle à chaîne droite ou à chaîne ramifiée, un groupe C₃₋₆ cycloalcoyle, phényle ou naphthyle ou un résidu hétérocyclique pentagonal ou hexagonal saturé ou insaturé ayant de 1 à 4 hétéro-atomes choisis parmi l'azote, l'oxygène et le soufre, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, COOH, NO₂, SO₃H et SH ; et

R⁷ et R⁸ ont chacun indépendamment la même signification que R⁵.

3. Un composé selon la revendication 1 ou la revendication 2, dans lequel

X représente un groupe OH, OR⁷, OCOR⁷, NH₂, NR⁷R⁸ ou SR⁷ ;

R¹ à R⁴ représentent chacun indépendamment un atome d'hydrogène ou un groupe C₁₋₄ alcoyle à chaîne droite ou à chaîne ramifiée éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes OH, OR⁶, COOR⁶, NH₂ et NR⁶₂ ;

R⁵, R⁷ et R⁸ représentent chacun indépendamment un groupe C₁₋₆ alcoyle à chaîne droite ou à chaîne ramifiée, éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶, ou un groupe phényle, naphthyle ou thiényl, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₂R⁶ et SR⁶ ; et

R⁶ représente un groupe C₁₋₄ alcoyle à chaîne droite ou à chaîne ramifiée ou un groupe phényle, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, COOH, NO₂, NH₂, SO₃H et SH.

4. Un composé selon l'une quelconque des revendications 1 à 3, dans lequel

X représente un groupe OH, OR⁷, OCOR⁷ ou NH₂ ;

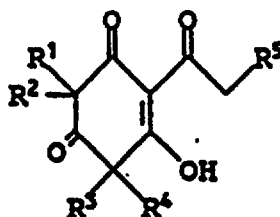
R¹ à R⁴ représentent chacun indépendamment un atome d'hydrogène ou un groupe C₁₋₂ alcoyle ;

R⁵, R⁷ et R⁸ représentent chacun indépendamment un groupe C₁₋₄ alcoyle à chaîne droite ou à chaîne ramifiée éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes OR⁶, ou un groupe phényle, naphthyle ou thiényl, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes R⁶, OR⁶ et SO₂R⁶ ; et

R⁶ représente un groupe méthyle ou trifluorométhyle.

5. Un procédé pour la préparation d'un composé de formule générale I tel que revendiqué dans la revendication 1, qui comprend

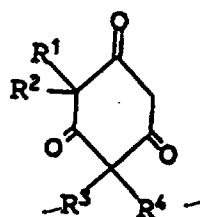
(a) l'oxydation d'un composé de formule générale II



(II)



où R¹ à R⁶ sont tels que définis dans la revendication 1, ou
(b) la réaction d'un composé de formule générale III



(III)

où R¹ à R⁴ et R⁶ sont tels que définis dans la revendication 1, avec un chlorure d'acide de formule générale IV

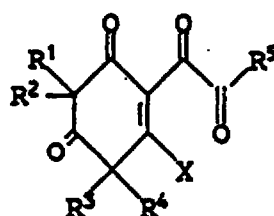


où R⁵ et R⁶ sont tels que définis dans la revendication 1, et ensuite, si on le désire, la conversion d'un composé résultant de la formule générale I dans lequel X représente un groupe OH en un composé de la formule générale I dans lequel X est autre qu'un groupe OH.

6. Un procédé selon la revendication 5, dans lequel on conduit la réaction (a) en utilisant un agent oxydant qui est de l'iode ou un permanganate.
7. Un procédé selon la revendication 5 ou la revendication 6, dans lequel la réaction d'oxydation est conduite in situ.
8. Une composition herbicide, qui comprend un composé selon l'une quelconque des revendications 1 à 4, en association avec un véhicule.
9. Une méthode de lutte contre la croissance de plantes indésirables en un lieu, qui comprend le traitement du lieu avec un composé selon l'une quelconque des revendications 1 à 4 ou avec une composition selon la revendication 8.
10. L'utilisation d'un composé selon l'une quelconque des revendications 1 à 4 ou d'une composition selon la revendication 8 dans la lutte contre la croissance de plantes indésirables.

Revendications pour l'Etat contractant suivant : ES

1. Un procédé pour la préparation d'un composé de la formule générale I



(I)

dans laquelle

X représente un groupe OH, OR⁷, NH₂, NHR⁷, NR⁷R⁸, NOH, NOR⁷, OCOR⁷, OCONHR⁷, OCONH-COR⁷, OCONHSO₂R⁷, NHCONHCOR⁷, OCOSR⁷, NHCOSR⁷, SCOR⁷, SR⁷, SCONHR⁷, SCONHCOR⁷, SCONHSO₂R⁷ ou SCOSR⁷ ;

R¹ à R⁴ représentent chacun indépendamment un atome d'hydrogène ou un groupe C₁₋₆ alcoyle, C₂₋₆ alcényle ou C₂₋₆ alcynyle à chaîne droite ou à chaîne ramifiée qui est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des



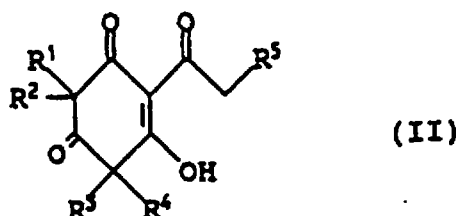
groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶ ;

R¹ et R² et/ou R³ et R⁴ forment ensemble un pont de C₂₋₆-méthylène saturé ou insaturé, à chaîne droite ou à chaîne ramifiée, éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶ ;

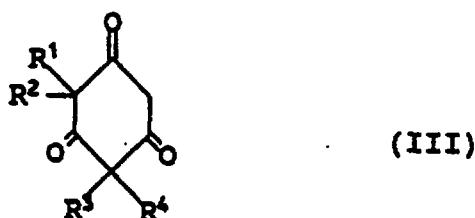
R⁵ est un groupe C₁₋₂₀ alcoyle, C₂₋₂₀ alcényle ou C₂₋₂₀ alcynyle à chaîne droite ou à chaîne ramifiée, un groupe C₃₋₂₀ cycloalcoyle, phényle ou naphthyle ou un résidu hétérocyclique saturé ou insaturé ayant un seul ou plusieurs hétéro-atomes, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶ ;

R⁶ est un groupe C₁₋₂₀ alcoyle à chaîne droite ou à chaîne ramifiée, un groupe C₃₋₂₀ cycloalcoyle, phényle ou naphthyle ou un résidu hétérocyclique saturé ou insaturé ayant un seul ou plusieurs hétéro-atomes, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, COOH, NO₂, NH₂, SO₃H et SH ; et

R⁷ et R⁸ ont chacun indépendamment la même signification que R⁵, qui comprend
(a) l'oxydation d'un composé de la formule générale II



dans laquelle R¹ à R⁶ sont tels que définis ci-dessus, ou
(b) la réaction d'un composé de la formule générale III



dans laquelle R¹ à R⁴ et R⁶ sont tels que définis ci-dessus, avec un chlorure d'acide de la formule générale IV



dans laquelle R⁵ et R⁶ sont tels que définis ci-dessus et ensuite, si on le désire, la conversion d'un composé résultant de la formule générale I dans lequel X représente un groupe OH en un composé de la formule générale I dans lequel X est autre qu'un groupe OH.

2. Un procédé selon la revendication 1, dans lequel

X représente un groupe OH, OR⁷, OCOR⁷, NH₂, NHR⁷, NR⁷R⁸ ou SR⁷,

R¹ à R⁴ représentent chacun indépendamment un atome d'hydrogène ou un groupe C₁₋₆ alcoyle à chaîne droite ou à chaîne ramifiée éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂, SO₃H, SO₂R⁶ et SR⁶, ou

R¹ et R² et/ou R³ et R⁴ forment ensemble un pont de C₃₋₅ méthylène saturé ou insaturé, à chaîne droite ou à chaîne ramifiée, éventuellement substitué par un ou plusieurs substituants identiques ou



différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁵, OR⁵, COOR⁵, NO₂, NH₂, NR⁵₂, SO₃H, SO₂R⁵ et SR⁵ ;

5 R⁵ est un groupe C₁₋₈ alcoyle, C₂₋₈ alcényle ou C₂₋₈ alcynyle à chaîne droite ou à chaîne ramifiée ou un groupe C₃₋₈ cycloalcoyle, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, OR⁵, COOR⁵, NO₂, NH₂, NR⁵₂, SO₃H, SO₂R⁵ et SR⁵ ou un groupe phényle ou naphthyle, ou un résidu hétérocyclique pentagonal ou hexagonal saturé ou insaturé ayant de 1 à 4 hétéro-atomes choisis parmi l'azote, l'oxygène et le soufre, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁵, OR⁵, COOR⁵, NO₂, NH₂, NR⁵₂, SO₃H, SO₂R⁵ et SR⁵ ;

10 R⁵ est un groupe C₁₋₆ alcoyle à chaîne droite ou à chaîne ramifiée, un groupe C₃₋₆ cycloalcoyle, phényle ou naphthyle ou un résidu hétérocyclique pentagonal ou hexagonal saturé ou insaturé ayant de 1 à 4 hétéro-atomes choisis parmi l'azote, l'oxygène et le soufre, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, COOH, NO₂, NH₂, SO₃H et SH ; et

15 R⁷ et R⁸ ont chacun indépendamment la même signification que R⁵.

3. Un procédé selon la revendication 1 ou la revendication 2, dans lequel

X représente un groupe OH, OR⁷, OCOR⁷, NH₂, NR⁷R⁸ ou SR⁷ ;

20 R¹ à R⁴ représentent chacun indépendamment un atome d'hydrogène ou un groupe C₁₋₄ alcoyle à chaîne droite ou à chaîne ramifiée éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes OH, OR⁵, COOR⁵, NH₂ et NR⁵₂ ;

25 R⁵, R⁷ et R⁸ représentent chacun indépendamment un groupe C₁₋₆ alcoyle à chaîne droite ou à chaîne ramifiée, éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, OR⁵, COOR⁵, NO₂, NH₂, NR⁵₂, SO₃H, SO₂R⁵ et SR⁵, ou un groupe phényle, naphthyle ou thiényl, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, R⁶, OR⁶, COOR⁶, NO₂, NH₂, NR⁶₂ et SA⁵ ; et 2

30 R⁶ représente un groupe C₁₋₄ alcoyle à chaîne droite ou à chaîne ramifiée ou un groupe phényle, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes CN, OH, COOH, NO₂, NH₂, SO₃H et SH.

4. Un procédé selon l'une quelconque des revendications 1 à 3, dans lequel

35 X représente un groupe OH, OR⁷, OCOR⁷ ou NH₂ ;

R¹ à R⁴ représentent chacun indépendamment un atome d'hydrogène ou un groupe C₁₋₂ alcoyle ;

40 R⁵, R⁷ et R⁸ représentent chacun indépendamment un groupe C₁₋₄ alcoyle à chaîne droite ou à chaîne ramifiée éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes OR⁶, ou un groupe phényle, naphthyle ou thiényl, dont chacun est éventuellement substitué par un ou plusieurs substituants identiques ou différents choisis parmi des atomes d'halogènes et des groupes R⁶, OR⁶ ou SO₂R⁶ ; et

R⁶ représente un groupe méthyle ou trifluorométhyle.

45 5. Un procédé selon l'une quelconque des revendications 1-4, dans lequel on conduit la réaction (a) en utilisant un agent oxydant qui est de l'iode ou un permanganate.

6. Un procédé selon la revendication 5, dans lequel la réaction d'oxydation est conduite in situ.

50 7. Une composition herbicide qui comprend un composé préparé par le procédé tel que revendiqué dans l'une quelconque des revendications 1 à 6, en association avec un véhicule.

8. Une méthode de lutte contre la croissance de plantes indésirables en un lieu, qui comprend le traitement du lieu avec un composé préparé par le procédé tel que revendiqué dans l'une quelconque des revendications 1 à 6; ou avec une composition selon la revendication 7.

55 9. L'utilisation d'un composé préparé selon le procédé tel que revendiqué dans l'une quelconque des revendications 1 à 6, ou d'une composition telle que revendiquée dans la revendication 7, pour lutter contre la croissance de plantes indésirables.