

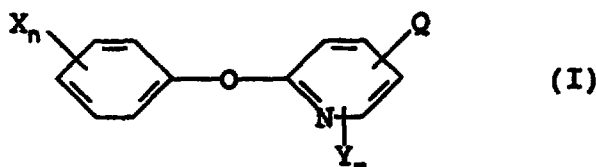
Description

This invention relates to certain new herbicidal acrylonitrile derivatives, their preparation, herbicidal compositions containing such derivatives and to a method of combating undesired plant growth using such derivatives and compositions.

EP-A-272 533, EP-A-200 677, PATENT ABSTRACTS OF JAPAN, vol.4, no. 178 (c-34)(660), 10 December 1980 & JP-A-55 118 450, 11 September 1980 and EP-A-0 348 002 disclose phenoxy pyridine derivatives having herbicidal activity.

The present invention relates to a novel class of phenoxy nicotinoyl/isonicotinoyl/picolinoyl acrylonitrile compounds.

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



in which Q represents a group of general formula $-C(O)-C(CN)=CH-NR^1R^2$, R^1 and R^2 each independently represents a C_{1-12} alkyl group, or together represent $-(CH_2)_4-$ or $-(CH_2)_2-O(CH_2)_2-$, m is 0 or an integer from 1 to 3, the or each Y independently represents a halogen atom or an alkyl group, n is 0 or an integer from 1 to 5, and the or each X independently represents a halogen atom or a group selected from alkyl, haloalkyl, cyanoalkyl, alkenyl, alkynyl, alkoxy, haloalkoxy, haloalkylthio, alkenyloxy, alkynyloxy, alkylthio, alkenylthio, alkynylthio, cyano, nitro, alkylsulphonyl, alkylsulphinyl and sulphonamido, or a phenyl group optionally substituted by one or more moieties independently selected from alkyl, haloalkyl and alkoxy groups and halogen atoms.

As used herein the term alkyl, alkenyl or alkynyl in respect of a radical or moiety refers to a straight or branched chain radical or moiety. Suitably an alkyl radical or moiety has from 1 to 12 carbon atoms, preferably from 1 to 6 and especially from 1 to 4 carbon atoms. Alkenyl and alkynyl radicals or moieties suitably have from 2 to 12 carbon atoms, preferably from 2 to 6, especially from 2 to 4, carbon atoms. Preferred halogen atoms are bromine, chlorine and fluorine atoms, especially chlorine and fluorine atoms.

Preferably R^1 and R^2 each independently represents an alkyl group of 1 to 4 carbon atoms. Preferably, at least one of the groups R^1 and R^2 represents a methyl group. Most preferably R^1 and R^2 are the same and each represents a methyl group.

When m is 1, the substituent Y is preferably a C_1-C_4 alkyl, especially methyl, group.

When m is greater than 1, the substituents Y may be the same or different and may suitably be selected from chlorine, bromine and fluorine atoms, and methyl and ethyl groups. However, good activity is shown by compounds in which m is 0 and it is most preferred that no Y substituent is present.

When n is greater than 1, the substituents X may be the same or different and may suitably be selected from halogen atoms, especially fluorine, chlorine and bromine atoms, C_{1-6} alkyl groups, C_{1-6} haloalkyl groups, C_{1-6} alkoxy groups, and cyano groups. Preferably n is 1 or 2, most preferably 1, and the or each substituent X independently represents a chlorine or fluorine atom, a C_{1-4} alkyl group, especially methyl, a C_{1-4} haloalkyl group, especially trifluoromethyl, a C_{1-4} alkoxy group, especially methoxy or ethoxy, or a cyano group.

Preferably a group X is located at the 3-position. Such compounds have been found to have particularly high activity.

Most preferably a phenyl group substituted by X_n is 3-trifluoromethylphenyl or 3-cyanophenyl.

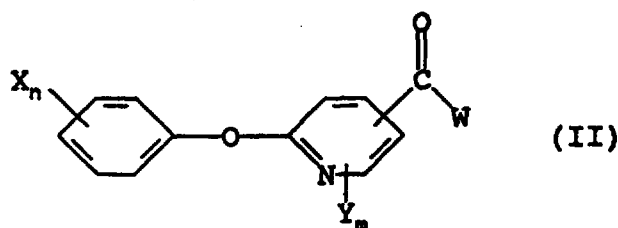
The group Q may be located at any of the available positions on the pyridyl ring, namely at the 3-, 4-, 5- or 6- position.

A particularly high level of activity is shown by compounds of general formula I in which the group Q is located at the 3- position on the pyridyl ring. Accordingly such compounds constitute a preferred class.

An especially high level of activity is shown by compounds of general formula I in which the group Q is located at the 6- position on the pyridyl ring. Accordingly such compounds constitute the most preferred class of compounds.

The compounds of general formula I may either be prepared from a corresponding phenoxy pyridine carboxylate derivative by reaction in one or more steps to introduce the dialkylaminoacrylonitrile moiety.

According to a further aspect of the present invention, a process for the preparation of a compound of general formula I is therefore provided, which process comprises reacting a compound of general formula II



wherein x , y , n and m are as defined above and W represents a halogen atom or a cyanomethyl group, with, in the case where W is halogen, a dialkylaminoacrylonitrile under basic conditions, or, in the case where W is cyanomethyl, either with a dialkylformamide dialkyl acetal or with a trialkylorthoformate followed by reaction with a dialkylamine, and, if desired or required, converting a resulting compound of general formula I into another of general formula I.

When W in a compound of general formula II represents a cyanomethyl group, the reaction to form the compound of formula I is carried out suitably using a di(C_1 -4alkyl)formamide di(C_1 -4alkyl) acetal, preferably dimethyl formamide dimethyl acetal. Suitably the reaction is carried out in the presence of an inert organic solvent, suitable solvents being halogenated hydrocarbons, for example dichloromethane, ethers, such as diethyl ether, or esters, such as ethyl acetate; mixtures of solvents may also be utilised. Preferably the reaction is carried out at a temperature in the range of from 0 to 50 °C, suitably at ambient temperature.

Alternatively, when W in a compound of general formula II represents a cyanomethyl group, the reaction may be carried out in a two-stage procedure by reacting the cyanomethyl carbonyl compound II with a trialkylorthoformate, preferably trimethylorthoformate, followed, optionally after isolation of the alkoxy pyridoyl acrylonitrile so formed, by reaction with excess dialkylamine, preferably dimethylamine in a suitable solvent. Suitable solvents are, for example, halogenated hydrocarbons, such as dichloromethane, alcohols, such as ethanol, ethers, such as diethyl ether, and esters; mixtures of solvents are also suitable. The reaction may be carried out at a temperature in the range of from -10 to 100 °C. Preferably the first stage is carried out at elevated temperature, for example at a temperature in the range of from 60 to 100 °C, suitably at approximately 80 °C, while the second stage is carried out at a lower temperature, suitably a temperature below 60 °C and conveniently ambient temperature. Preferably the reaction is carried out without isolation of the intermediate acrylonitrile.

When W in a compound of general formula II represents a halogen atom, preferably chlorine, the compound of formula II is reacted with a dialkylaminoacrylonitrile, preferably dimethylaminoacrylonitrile, preferably in the presence of a solvent, for example an ether solvent, and in the presence of a base, for example, a tertiary amine. A preferred ether solvent is dioxan and a preferred tertiary amine base is triethylamine. The reaction is preferably carried out at a temperature in the range of from 50 to 120 °C, conveniently at the reflux temperature of the solvent employed.

A preferred process for the preparation of a compound of general formula I employs a compound of general formula II wherein W represents a cyanomethyl group. Preferably, the reaction of such a compound of general formula II is with a dialkylformamide dialkyl acetal.

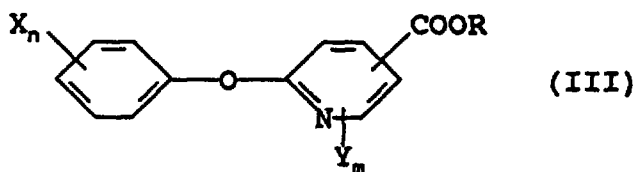
Compounds of general formula I may be converted into other compounds of general formula I using various standard methods. For example, a compound of formula I having certain groups R^1 and R^2 may be converted to another compound of general formula I having at least one different group R^1 or R^2 , by reaction with a secondary amine R^1R^2NH , suitably in the presence of an inert solvent, for example ethanol, at an elevated temperature, suitably under reflux.

Following its preparation the resulting compound of general formula I may be isolated and purified using conventional techniques, for example by solvent extraction, evaporation and recrystallisation or by chromatography on silica.

Compounds of general formula II are believed to be novel intermediates and the present invention further includes such compounds and their preparation.

Compounds of general formula II in which W represents a cyanomethyl group may be prepared from the alkyl ester of the corresponding phenyl pyridyl ether.

Thus the invention further provides a process for the preparation of a compound of formula II as defined above which comprises reacting a compound of formula III:



wherein x, Y, n and m are as defined above and R represents an alkyl group, suitably a C₁₋₄ alkyl group, with acetonitrile under basic conditions.

Preferably the basic conditions are provided by an alkali metal amide, such as sodamide or potassium amide, suitably in liquid ammonia, or sodium or potassium hydride, in a dry solvent such as dimethylformamide or diethyl ether. Alternatively the basic conditions may be provided by an alkoxide such as sodium or potassium methoxide with excess acetonitrile as solvent. Suitable temperatures for reaction are in the range of from -70°C to 100°C and are usually determined by the basic conditions used.

The compounds of general formula III may suitably be prepared by condensation of an optionally substituted phenol or phenolate with an optionally substituted halonicotinic/isonicotinic/picolinic acid ester, for example in the presence of an alkali metal hydride, for example sodium hydride, in a dry solvent, such as dimethylformamide. Alternatively the reaction may be carried out in the presence of an alkali metal alkoxide, such as sodium methoxide, followed by treatment with a copper catalyst such as cuprous chloride in pyridine and an aromatic hydrocarbon, such as xylene, as described in U.K. patent specification No. 2050168. The reaction may alternatively be carried out in the presence of an alkali metal carbonate, for example sodium or potassium carbonate, followed by treatment with cuprous oxide and/or copper powder in dimethylformamide or quinoline. The reaction may suitably be carried out at a temperature in the range of from 20 to 150°C and conveniently the reaction is carried out at the reflux temperature of the reaction mixture. Alternatively the phenoxy pyridine carboxylates may be prepared by condensation of an optionally substituted hydroxypyridine carboxylate and an optionally substituted halobenzene, suitably under the conditions of UK specification No. 2,050,168 described above.

The starting pyridine carboxylates are either known or can be prepared by conventional techniques, see, for example, Pesticide Science 1987, 18, pages 15 to 28.

Compounds of formula III may also be prepared by esterification of the corresponding acid with an alcohol, such as methanol, in the presence of an acidic catalyst, for example concentrated sulphuric acid, dry hydrogen chloride gas or p-toluene sulphonic acid.

Compounds of general formula II in which W represents a halogen atom may be derived from the corresponding ether carboxylic acid, for example by reaction of the acid with thionyl chloride. The acid may be derived as described above from a phenolic compound and a halopyridine carboxylic acid ester.

The compounds of the invention have been found to have surprisingly high herbicidal activity with a wide spectrum of activity against grasses and, especially, broadleaved weeds, when applied pre- and post-emergence. Some examples have been found to show selectivity to small grain cereals and rice, indicating that they may be useful in combating weeds growing in such crops.

Accordingly, the invention further provides a herbicidal composition comprising a compound of formula I as defined above in association with at least one carrier, and a method of making such a composition which comprises bringing a compound of formula I into association with at least one carrier.

The invention also provides the use of such a compound or composition according to the invention as a herbicide. Further, in accordance with the invention there is provided a method of combating undesired plant growth at a locus by treating the locus with a compound or composition according to the invention. Application to the locus may be pre-emergence or post-emergence. The dosage of active ingredient used, may, for example, be in the range of from 0.01 to 10 kg/ha, suitably 0.05 to 4 kg/ha. The locus may, for example, be the soil or plants in a crop area, typical crops being cereals such as wheat, barley and rice.

A carrier in a composition according to the invention is any material with which the active ingredient is formulated to facilitate application to the locus to be treated, or to facilitate storage, transport or handling. A carrier may be a solid or a liquid, including a material which is normally gaseous but which has been compressed to form a liquid, and any of the carriers normally used in formulating herbicidal compositions may be used. Preferably compositions according to the invention contain 0.5 to 95% by weight of active

ingredient.

Suitable solid carriers include natural and synthetic clays and silicates, for example natural silicas such as diatomaceous earths; magnesium silicates, for example talcs; magnesium aluminium silicates, for example attapulgites and vermiculites; aluminium silicates, for example kaolinites, montmorillonites and micas; calcium carbonate; calcium sulphate; ammonium sulphate; synthetic hydrated silicon oxides and synthetic calcium or aluminium silicates; elements, for example carbon and sulphur; natural and synthetic resins, for example coumarone resins, polyvinyl chloride, and styrene polymers and copolymers; solid polychlorophenols; bitumen; waxes; and solid fertilisers, for example superphosphates.

Suitable liquid carriers include water; alcohols, for example isopropanol and glycols; ketones, for example acetone, methyl ethyl ketone, methyl isobutyl ketone and cyclohexanone; ethers; aromatic or aliphatic hydrocarbons, for example benzene, toluene and xylene; petroleum fractions, for example kerosene and light mineral oils; chlorinated hydrocarbons, for example carbon tetrachloride, perchloroethylene and trichloroethane. Mixtures of different liquids are often suitable.

Agricultural compositions are often formulated and transported in a concentrated form which is subsequently diluted by the user before application. The presence of small amounts of a carrier which is a surface-active agent facilitates this process of dilution. Thus preferably at least one carrier in a composition according to the invention is a surface-active agent. For example the composition may contain at least two carriers, at least one of which is a surface-active agent.

A surface-active agent may be an emulsifying agent, a dispersing agent or a wetting agent; it may be nonionic or ionic. Examples of suitable surface-active agents include the sodium or calcium salts of polyacrylic acids and lignin sulphonc acids; the condensation of fatty acids or aliphatic amines or amides containing at least 12 carbon atoms in the molecule with ethylene oxide and/or propylene oxide; fatty acid esters of glycerol, sorbitol, sucrose or pentaerythritol; condensates of these with ethylene oxide and/or propylene oxide; condensation products of fatty alcohol or alkyl phenols, for example *p*-octylphenol or *p*-octylcresol, with ethylene oxide and/or propylene oxide; sulphates or sulphonates of these condensation products; alkali or alkaline earth metal salts, preferably sodium salts, of sulphuric or sulphonic acid esters containing at least 10 carbon atoms in the molecule, for example sodium lauryl sulphate, sodium secondary alkyl sulphates, sodium salts of sulphonated castor oil, and sodium alkylaryl sulphonates such as dodecylbenzene sulphonate; and polymers of ethylene oxide and copolymers of ethylene oxide and propylene oxide.

The compositions of the invention may for example be formulated as wettable powders, dusts, granules, solutions, emulsifiable concentrates, emulsions, suspension concentrates and aerosols. Wettable powders usually contain 25, 50 or 75% w of active ingredient and usually contain in addition to solid inert carrier, 3-10% w of a dispersing agent and, where necessary, 0-10% w of stabiliser(s) and/or other additives such as penetrants or stickers. Dusts are usually formulated as a dust concentrate having a similar composition to that of a wettable powder but without a dispersant, and are diluted in the field with further solid carrier to give a composition usually containing 0.5-10% w of active ingredient. Granules are usually prepared to have a size between 10 and 100 BS mesh (1.676 - 0.152 mm), and may be manufactured by agglomeration or impregnation techniques. Generally, granules will contain 0.5-75% w active ingredient and 0-10% w of additives such as stabilisers, surfactants, slow release modifiers and binding agents. The so-called "dry flowable powders" consist of relatively small granules having a relatively high concentration of active ingredient. Emulsifiable concentrates usually contain, in addition to a solvent and, when necessary, co-solvent, 10-50% w/v active ingredient, 2-20% w/v emulsifiers and 0-20% w/v of other additives such as stabilisers, penetrants and corrosion inhibitors. Suspension concentrates are usually compounded so as to obtain a stable, non-sedimenting flowable product and usually contain 10-75% w active ingredient, 0.5-15% w of dispersing agents, 0.1-10% w of suspending agents such as protective colloids and thixotropic agents, 0-10% w of other additives such as defoamers, corrosion inhibitors, stabilisers, penetrants and stickers, and water or an organic liquid in which the active ingredient is substantially insoluble; certain organic solids or inorganic salts may be present dissolved in the formulation to assist in preventing sedimentation or as anti-freeze agents for water.

Aqueous dispersions and emulsions, for example compositions obtained by diluting a wettable powder or a concentrate according to the invention with water, also lie within the scope of the invention. The said emulsions may be of the water-in-oil or of the oil-in-water type, and may have a thick "mayonnaise" - like consistency.

The compositions of the invention may also contain other active ingredients, for example compounds possessing insecticidal or fungicidal properties or other herbicides.

The following Examples illustrate the invention. Examples 1 to 23 relate to the preparation of starting materials, Examples 24 to 46 to the preparation of novel intermediates of general formula II and Examples

47 to 72 to the preparation of compounds of general formula I. All structures were confirmed by mass spectroscopy and ^{300}H nmr.

Example 1

Preparation of methyl-2-(3'- α,α,α -trifluoromethylphenoxy)nicotinate

3-Hydroxybenzotrifluoride (16.2 g) was added dropwise to a suspension of sodium hydride (4 g of 60% dispersion in oil) in dry dimethylformamide (100 ml). After sodium salt formation was complete, methyl-2-chloronicotinate (17.1 g) in dimethylformamide (25 ml) was added. The reaction mixture was heated at 100°C for 3 hours. After cooling to ambient temperature, the mixture was poured into water (300 ml) and the aqueous mixture extracted with diethyl ether. After drying the extracts over anhydrous magnesium sulphate, the solvent was evaporated and the residue purified on a silica gel column using 10% diethyl ether-dichloromethane (v/v) as eluant. The title compound was obtained (9 g) as a white solid of m.p. $83-85^\circ\text{C}$ m/e Theory: Found 297 : 297

Analysis:			
Calculated for $\text{C}_{14}\text{H}_{10}\text{NO}_3\text{F}_3$	C 56.5	H 3.4	N 4.7%
Found	C 56.0	H 3.7	N 4.1%

Example 2

Preparation of methyl-2-(3'- α,α,α -trifluoromethylphenoxy) isonicotinate

A solution of sodium methoxide (from 2.65 g sodium in 45 ml methanol) was added to a solution of 3-hydroxybenzotrifluoride (17.8 g) in xylene (100 ml). The solvents were evaporated *in vacuo* to give the dry sodium phenolate salt. Pyridine (50 ml) and xylene (100 ml) were added followed by cuprous chloride (3 g) and the mixture heated to reflux. A solution of methyl-2-chloro isonicotinate (17.1 g) in xylene (25ml) was added dropwise and the mixture refluxed a further 6 hours. After cooling, the mixture was poured into water (250 ml) and acidified with dilute hydrochloric acid. The aqueous solution was extracted with diethyl ether. The ethereal extracts were dried over anhydrous magnesium sulphate and evaporated. The residual oil was purified on a silica gel column using dichloromethane as eluant to give the title compound (22 g) as a colourless oil. m/e Theory : Found 297 : 297

Analysis			
Calculated for $\text{C}_{14}\text{H}_{10}\text{O}_3\text{NF}_3$	C 56.5	H 3.4	N 4.7%
Found	C 55.8	H 3.5	N 4.6%

Example 3

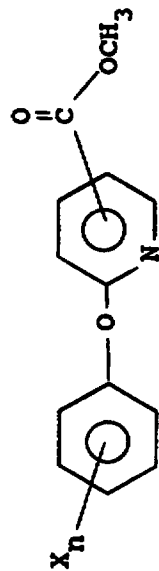
Preparation of methyl 5-methyl-2-(3'- α,α,α -trifluoromethylphenoxy)-3-pyridine carboxylate

3- α,α,α -trifluoromethylphenol (4.3g) in dry dimethylformamide (25ml) was added dropwise to a stirred suspension of sodium hydride (1.1g, 60% dispersion in oil) in dry dimethylformamide (100ml). When the solution became clear, a solution of methyl 2-bromo-5-methyl-3-pyridine carboxylate (6g) in dimethylformamide (25ml) was added dropwise. The reaction mixture was then heated to 100°C for 4 hours. After cooling, the reaction mixture was poured onto water, and extracted with diethyl ether. After drying over anhydrous magnesium sulphate, the ether was evaporated off. The residue was purified on a silica gel column using 50% (v/v) 40-60 petroleum ether-diethyl ether as eluant to give the title compound (1.3g) of mp. $49-51^\circ\text{C}$. m/e Theory : Found 311:311

Analysis			
Calculated for $C_{15}H_{12}NO_3F_3$	C 57.9	H 3.9	N 4.5%
Found	C 57.9	H 4.1	N 4.0%

The compounds of general formula III listed in Table 1 below, were prepared by methods similar to that described in Examples 1 and 2 above. More specifically, Examples 17 to 22 were made by the method of Example 2 whilst the remaining Examples of Table 1 were made by the method of Example 1.

Table 1



(General Formula III)
Q is $C(O)OCH_3$; m is 0)

Example No.	X_n	Ester group position (to -N-)	Elemental Analysis (%)				Melting Point ($^{\circ}C$)	m/e (Theory Found)
			C	H	N			
			(Calc. Found)	(Calc. Found)	(Calc. Found)			
4	3-Cl	3	59.3	3.8	5.3		89-92	263
			59.4	4.1	4.1			263
5	4-Cl	3	59.3	3.8	5.3		113-114	263
			59.6	4.2	5.0			263
6	3-CH ₃	3	69.1	5.4	5.8		72-75	243
			70.8	6.1	5.3			243
7	3-CH ₃ O	3	64.9	5.0	5.4		59-62	259
			65.0	5.0	4.9			259

Table 1 (continued)

Example No.	X_n	Ester group position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
			C	H	N			
			(Calc. Found)	(Calc. Found)	(Calc. Found)			
8	3-CN	3	66.1	3.9	11.0		124-125	254
			68.8	5.1	10.8			254
9	3-CF ₃ ,4-Cl	3	50.7	2.7	4.6		79-81	331
			50.1	3.1	4.6			331
10	-	3	68.1	4.8	6.1		67-69	229
			70.5	5.8	6.3			229
11	2,4-F ₂	3	58.9	3.4	5.3		89-92	265
			61.1	4.4	5.3			265
12	2-CH ₃	3	69.1	5.4	5.8		77-78	243
			68.3	5.4	5.8			243
13	3-F	3	63.2	4.1	5.7		75-77	247
			66.4	5.3	5.5			247

Table 1 (continued)

Example No.	X_n	Ester group position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
			C	H	N			
			(Calc. Found)	(Calc. Found)	(Calc. Found)			
14	3,5-Cl ₂	3	52.5	3.0	4.7		111-113	297
			53.1	3.3	4.6			297
15	2-Cl	3	59.3	3.8	5.3		97-98	263
			63.6	3.8	5.3			263
16	3-C ₂ H ₅ O	3	65.9	5.5	5.1		86-87	273
			67.1	6.6	4.7			273
17	3-Cl	4	59.2	3.8	5.3		58-60	263
			60.1	3.9	5.3			263
18	2,5-Cl ₂	4	52.3	3.0	4.7		89-91	298
			52.3	3.0	4.9			298
19	3-CF ₃	6	56.6	3.4	4.7		43-44	297
			55.5	3.3	4.8			297

Table 1 (continued)

Example No.	X_h	Ester group position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
			C	H	N			
			(Calc. Found)	(Calc. Found)	(Calc. Found)			
20	3-CH ₃ O	6	64.9	5.0	5.4		oil	259
			64.7	4.7	5.1			259
21	3-Cl	6	59.3	3.8	5.3		60-61	263
			59.7	3.8	5.2			263
22	-	6	68.1	4.8	6.1		71-72	229
			66.8	4.6	5.8			229
23	3-CF ₃	5	56.5	3.4	4.7		80-82	297
			60.8	3.5	4.7			297

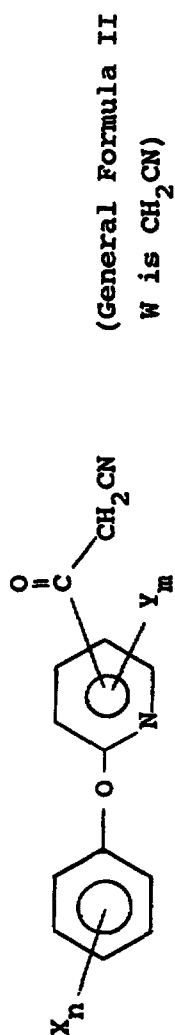
Example 24Preparation of 2-(3'- α,α,α -trifluoromethylphenoxy)nicotinoylacetonitrile

5 To a suspension of sodamide (prepared from 1.2 g of sodium metal in 60 ml of liquid ammonia with 0.1 g ferric nitrate) was added a solution of dry acetonitrile (2.1 g) in dry diethyl ether (5 ml) over 5 minutes. After a further 5 minutes, methyl-2-(3'- α,α,α -trifluoromethylphenoxy) nicotinate (7.4 g) in diethyl ether (25 ml) was added as quickly as possible. After stirring for a further 1 hour at below -30°C , the ammonia was removed by warming on a water bath, whilst at the same time, adding diethyl ether to maintain the reaction mixture volume of approximately 100 ml. The ethereal solution was then cautiously poured onto ice (50 g).
 10 The aqueous layer was separated, washed with diethyl ether and acidified with 6M hydrochloric acid. The aqueous solution was then extracted with diethyl ether, the extracts were dried over anhydrous magnesium sulphate and the diethyl ether removed by evaporation. Purification of the residual solid on a silica gel column using 5% diethyl ether-dichloromethane (v/v) as eluant gave the title compound (3.5 g) as a white
 15 solid of m.p. $66-68^{\circ}\text{C}$. m/e Theory : Found 306 : 306

Analysis			
Calculated for $\text{C}_{15}\text{H}_9\text{N}_2\text{O}_2\text{F}_3$	C 58.8	H 2.9	N 9.1%
Found	C 59.6	H 3.3	N 9.1%

20 The compounds of general formula II listed in Table 2 below, were prepared by methods similar to that described in Example 24 above, from the intermediates of Examples 2 to 23.

Table 2



Example No.	X _n	Y _m and Acetonitrile position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
			C	H	N			
			(Calc. Found)	(Calc. Found)	(Calc. Found)			
25	3-Cl	-	61.6	3.3	10.3		62-63	272
26	3-CF ₃	5-CH ₃	61.8	3.5	10.0			272
			60.0	3.4	8.8		101-103	320
27	4-Cl	-	58.8	3.5	8.4			320
			61.6	3.3	10.3		not	272
28	3-CH ₃	-	60.7	3.3	9.7		recorded	272
			71.4	4.8	11.1		102-103	252
			72.1	4.9	11.1			252

Table 2 (continued)

Example No.	X_n	Y_m and position (to -N-)	Acetonitrile position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
				C	H	N			
				(Calc. Found)	(Calc. Found)	(Calc. Found)			
29	3-CH ₃ O	-	3	67.2	4.5	10.5		97-100	268
30	3-CN	-	3	66.9	4.6	9.8			268
31	3-CF ₃ , 4-Cl	-	3	68.4	3.4	15.9		82-84	263
32	-	-	3	67.8	3.9	15.4		84-85	263
				52.9	2.3	8.2			340
				53.5	2.5	8.2			340
				70.6	4.2	11.8		135-136	238
				70.2	4.4	11.8			238
33	2,4-F ₂	-	3	61.3	2.9	10.2		107-109	274
				59.7	3.0	10.1			274
34	2-CH ₃	-	3	71.4	4.8	11.1		125-126	252
				71.5	5.0	11.0			252

Table 2 (continued)

Example No.	X _n	Y _m position (to -N-)	Acetonitrile position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
				C	H	N			
				(Calc. Found)	(Calc. Found)	(Calc. Found)			
35	3-F	-	3	65.6 65.6	3.5 3.5	10.9 10.6		85-88	256 256
36	3,5-Cl ₂	-	3	54.9 54.7	2.6 2.6	9.2 9.2		119-120	306 306
37	2-Cl	-	3	61.8 62.0	3.3 3.5	10.3 10.3		155-157	272 272
38	3-C ₂ H ₅ O	-	3	68.1 69.6	5.0 5.0	9.9 10.0		82-84	282 282
39	3-CF ₃	-	4	58.8 57.5	2.9 3.2	9.1 9.8		oil	306 306
40	3-Cl	-	4	61.6 61.7	3.3 3.5	10.3 10.5		66-68	272 272

Table 2 (continued)

Example No.	X _n	Y _m position (to -N-)	Acetonitrile position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
				C	H	N			
				(Calc. Found)	(Calc. Found)	(Calc. Found)			
41	2,5-Cl ₂	-	4	54.7 54.0	2.6 2.8	9.2 8.8		118-119	306 306
42	3-CF ₃	-	6	58.8 57.7	2.9 3.0	9.2 9.3		75-76	306 306
43	3-CH ₃ O	-	6	67.2 67.3	4.5 4.4	10.4 10.9		71-72	268 268
44	3-Cl	-	6	61.8 61.8	3.3 3.3	10.3 10.3		90-91	272 272
45	-	-	6	70.6 70.1	4.2 4.2	11.8 12.0		96-97	238 238
46	3-CF ₃	-	5	58.8 58.8	2.9 3.0	9.1 9.0		oil	306 306

Example 47**Preparation of 2-[2-(3'- α,α,α -trifluoromethylphenoxy)nicotinoyl]-3-dimethylaminoacrylonitrile**

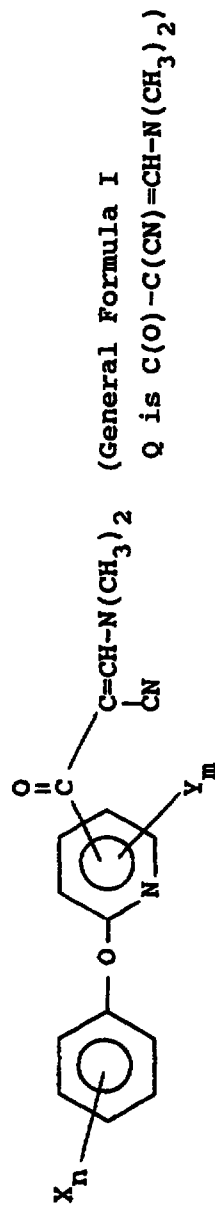
5 To a solution of 2-(3'- α,α,α -trifluoromethylphenoxy)nicotinoylacetonitrile (3.2g) in dichloromethane (70 ml) was added dimethylformamide dimethylacetal (5 ml). After stirring at ambient temperature for a half hour, the dichloromethane was removed by evaporation. The residue was purified on a silica gel column using 5% diethyl ether-dichloromethane (v/v) as eluant to give the title compound (3.5 g) as a white solid of m.p. 141-142 °C.

10 m/e Theory : Found 361 :361

15	Analysis			
	Calculated for C ₁₈ H ₁₄ N ₃ O ₂ F ₃	C 59.8	H 3.9	N 11.6%
	Found	C 59.9	H 3.9	N 11.5%

The compounds of general formula I listed in Table 3 below were prepared by methods similar to that described in Example 47 above, from the intermediates of Examples 25 to 46 above.

Table 3



Example No.	X _n	Y _m position (to -N-)	Acryloyl position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
				C	H	N			
				(Calc. Found)	(Calc. Found)	(Calc. Found)			
48	3-Cl	-	3	62.4	4.3	12.8		140-141	327
49	3-CF ₃	5-CH ₃	3	62.7	4.3	12.5			327
				60.8	4.3	11.2		44-47	375
				61.0	4.3	11.1			375
50	4-Cl	-	3	62.3	4.3	12.8		171-172	327
				61.7	4.3	12.7			327
51	3-CH ₃	-	3	70.4	5.5	13.7		147-148	307
				70.4	5.5	13.5			307

Table 3 (continued)

Example No.	X _n	Y _m position (to -N-)	Acryloyl position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
				C	H	N			
				(Calc. Found)	(Calc. Found)	(Calc. Found)			
52	3-CH ₃ O	-	3	66.9	5.3	13.0		106-107	323
				66.5	5.3	12.9			323
53	3-CN	-	3	67.9	4.4	17.6		140-141	318
				67.3	4.5	17.7			318
54	3-CF ₃ , 4-Cl	-	3	54.7	3.3	10.6		147-148	395
				55.0	3.6	10.6			395
55	-	-	3	69.6	5.1	14.3		160-161	293
				69.5	5.1	14.2			293
56	2,4-F ₂	-	3	62.0	4.0	12.8		112-113	329
				62.6	4.0	12.7			329
57	2-CH ₃	-	3	70.4	5.5	13.7		164-166	307
				70.6	5.7	13.7			307

Table 3 (continued)

Example No.	X_n	Y_m position (to -N-)	Acryloyl position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
				C	H	N			
				(Calc. Found)	(Calc. Found)	(Calc. Found)			
58	3-F	-	3	65.6	4.5	13.5		110-112	311
59	3,5-Cl ₂	-	3	65.4	4.6	13.2			311
				56.5	3.6	11.6		207-210	361
60	2-Cl	-	3	55.9	3.6	11.5			361
				62.4	4.3	12.8		138-140	327
61	3-C ₂ H ₅ O	-	3	62.5	4.3	12.8			327
				67.7	5.6	12.5		45-46	337
62	3-CF ₃	-	4	66.6	5.2	12.2			337
				59.8	3.9	11.6		114-116	361
63	3-Cl	-	4	59.2	3.8	11.5			361
				62.3	4.3	12.8		106-108	327
				62.2	4.5	13.0			327

Table 3 (continued)

Example No.	X_n	Y_m position (to -N-)	Acryloyl position (to -N-)	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)		
				C		H				N	
				(Calc. Found)	(Calc. Found)	(Calc. Found)	(Calc. Found)				
64	2,5-Cl ₂	-	4	56.3 55.6	3.6 3.7	11.6 11.3	150-151	361 361			
65	3-CF ₃	-	6	59.8 59.8	3.9 3.8	11.6 11.4	77-78	361 361			
66	3-CH ₃ O	-	6	66.9 66.5	5.3 5.3	13.0 14.0	66-67	323 323			
67	3-Cl	-	6	62.4 62.5	4.3 4.4	12.8 13.0	78-79	327 327			
68	-	-	6	69.6 69.4	5.1 5.6	14.3 14.5	70-71	293 293			
69	3-CF ₃	-	5	59.8 59.2	3.9 4.3	11.6 11.1	Gum	361 361			

Example 70Preparation of 2-[2-(3'-chlorophenoxy)nicotinoyl]-3-pyrrolidinoacrylonitrile

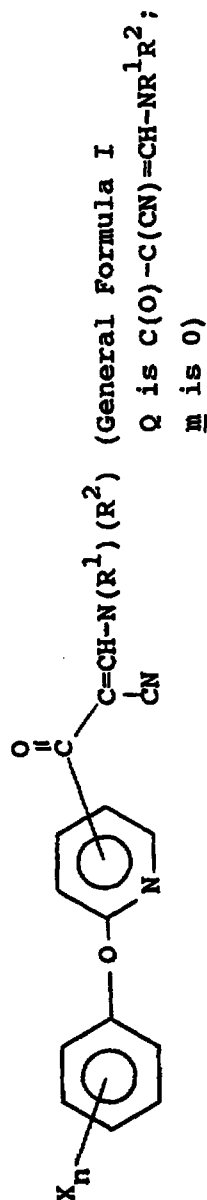
A solution of 2-[2-(3'-chlorophenoxy)-nicotinoyl]-3-dimethylaminoacrylonitrile (1 g) and pyrrolidine (2 ml) in ethanol was refluxed for 4 hours. The solution was then evaporated to dryness and the residue purified on a silica gel column, using 10% diethyl ether-dichloromethane (v/v) to give the title compound (0.8 g) as a white solid of m.p. 119-121 °C.

m/e Theory : Found 353 : 353

Analysis			
Calculated for $C_{19}H_{16}N_3O_2Cl$	C 64.6	H 4.5	N 11.9%
Found	C 64.4	H 4.5	N 12.3%

The further compounds of general formula I listed in Table 4 below were prepared by methods as described in Example 70, using appropriate compounds of formula I, as described above, and morpholine and methylethylamine respectively.

Table 4



Example No.	X _n	R ¹	R ²	Elemental Analysis (%)				Melting Point (°C)	m/e (Theory Found)
				Acryloyl position (to -N-)	C	H	N		
71	-	-	-(CH ₂) ₂ -O-(CH ₂) ₂ -	3	68.1	5.1	12.5	182-184	335
					64.5	5.1	12.6		335
72	3-CF ₃	CH ₃	C ₂ H ₅	6	60.8	4.3	11.2	81-83	375
					62.2	4.5	11.4		375

Herbicidal Activity

To evaluate their herbicidal activity, compounds of formula I according to the invention were tested using as representative range of plants: maize, Zea mays (Mz); rice, Oryza sativa (R); barnyard grass, Echinochloa crusgalli (BG); oat, Avena sativa (O); linseed, Linum usitatissimum (L); mustard, Sinapsis alba (M); sugar beet, Beta vulgaris (SB) and soya bean, Glycine max (S).

The tests fall into two categories, pre-emergence and post-emergence. The pre-emergence tests involved spraying a liquid formulation of the compound onto the soil in which the seeds of the plant specified mentioned above had recently been sown. The post-emergence tests involved two types of test, viz., soil drench and foliar spray tests. In the soil drench tests the soil in which the seedling plants of the above species were growing was drenched with a liquid formulation containing a compound of the invention, and in the foliar spray tests the seedling plants were sprayed with such a formulation.

The soil used in the tests was a prepared horticultural loam.

The formulations used in the tests were prepared from solutions of the test compounds in acetone containing 0.4% by weight of an alkylphenol/ethylene oxide condensate available under the trade mark TRITON X-155. These acetone solutions were diluted with water and the resulting formulations applied at dosage levels corresponding to 5kg and 1kg of active material per hectare in a volume equivalent to 900 litres per hectare in the soil spray and foliar spray test, and at a dosage of level equivalent to 10 kilograms of active material per hectare in a volume equivalent to approximately 3,000 litres per hectare in the soil drench tests.

In the pre-emergence tests untreated sown soil and in the post-emergence tests untreated soil bearing seedling plants were used as controls.

The herbicidal effects of the test compounds were assessed visually twelve days after spraying the foliage and the soil, and thirteen days after drenching the soil and were recorded on a 0-9 scale. A rating 0 indicates growth as untreated control, a rating 9 indicates death. An increase of 1 unit on the linear scale approximates to a 10% increase in the level of effect.

The results of the tests are set out in Table 5 below. A blank space in Table 5 indicates a rating 0, and the symbol * indicates that no result was obtained.

Table 5

Compound of Ex. No.	Soil drench 10/kg/ha								Dosage kg/ha	Foliar spray								Pre-emergence								
	Mz	R	BG	O	L	M	SB	S		Mz	R	BG	O	L	M	SB	S	Mz	R	BG	O	L	M	SB	S	
47	7	5	/	/	/	/	8	6	5		6	4	8	5	6	8	8	7	5	4	8	6	4	8	9	4
									1		4	1	6	3	3	8	8	7	4	3	8	6	4	8	9	3
48	8	7	7	7	4	9	8	7	5		5	2	6	3	4	8	8	7	6	5	6	7	2	7	8	4
									1		3	4	1	3	8	7	7	5	3	5	6	1	4	8	2	
49									5		3			4	7	5	4									
									1					1	6	2	1									
50	7	7	7	7	2	7	7	5	5		4	4		3	7	6	6	6	4	3	6	3	1	3	4	
									1		1	1		3	5	5	5	5			2			2	2	
51	8	8	7	8	3	9	7	7	5		3	2	5	3	3	8	6	4	7	5	6	5		6	8	5
									1		3	1	2	2	7	5	4	4	4	1	3	1		2	3	1

Table 5 (continued)

Compound of Ex. No.	Soil drench 10/kg/ha								Dosage kg/ha	Foliar spray								Pre-emergence								
	Mz	R	BG	O	L	M	SB	S		Mz	R	BG	O	L	M	SB	S	Mz	R	BG	O	L	M	SB	S	
52	8	7	8	1	4	9	8	6	5		6	3	4	3	5	8	8	6	7	6	9	7	3	9	8	6
									1		3	2	2	2	3	7	6	5	6	3	7	3	1	8	6	3
53	9	8	8	8	6	9	8	7	5		6	5	6	4	5	9	7	5	8	8	8	8	3	8	9	5
									1		2	2	3	1	3	8	7	5	5	4	6	5	2	6	8	2
54	5	2	5	5		3	5	3	5		5	2	6	2	5	6	5	5	3	4	5	3	7	8	1	
									1		3	1	5	1	4	5	4	4	1	3	3	2	6	7	1	
55	8	8	8	8	5	9	7	8	5		3	1	4	2	3	8	6	7	8	5	6	4	2	7	9	6
									1		2				2	6	5	5	4	1	2		3	4	3	
56	7	6	6	5	4	9	6	5	5		2		3	1	4	6	5	5	5	3	2	3	1	5	7	3
									1		1		1		2	4	3	3					1	3	1	

Table 5 (continued)

Compound of Ex. No.	Soil drench 10/kg/ha						Dosage kg/ha	Foliar spray						Pre-emergence					
	Mz	R	B	G	O	L	M	SB	S	Mz	R	B	G	O	L	M	SB	S	S
57	4		2	1		4	5	3		2					2	4	3	3	
							1								1	3	1	3	
58	8	7	8	7	3	8	8	8		7	2	7	4	5	7	8	7		
							1			2		3		3	5	6	6		
59	6	4	6	6		3	5	4		*	*	*	*	*	*	*	*	*	*
							1			2		5	3	2	6	8	5		
60	2	1				1	2	1		2		1		1	3	3	3		
							1			1				1	2	3	2		
61	8	7	8	8	4	7	8	6		6	3	7	4	6	8	9	8		
							1			2	1	2	2	4	7	8	5		

Table 5 (continued)

Compound of Ex. No.	Soil drench 10/kg/ha								Dosage kg/ha	Foliar spray								Pre-emergence								
	Mz	R	BG	O	L	M	SB	S		Mz	R	BG	O	L	M	SB	S	Mz	R	BG	O	L	M	SB	S	
62	5	4	5	5	5	6	2	5	1	4	1	7	4	5	8	8	5	4	1	3	3	4	3	2		
63	6	3	5	7	3	8	2	5	1	2	1	7	4	5	9	9	4	4	2	3	2	1	4	1		
64	4	2	5	2	4	4	4	5	1	2	1	3	2	4	6	5	3	2	3	2	1	4	1	2		
65	7	5	7	8	5	8	9	4	5	2	5	2	8	4	7	9	9	7	5	5	7	6	4	7	9	4
66	5	5	6	7	5	9	9	4	5	1	2	7	3	5	8	9	7	2	4	5	4	2	9	8	4	

Table 5 (continued)

Compound of Ex. No.	Soil drench 10/kg/ha								Dosage kg/ha	Foliar spray								Pre-emergence							
	Mz	R	BG	O	L	M	SB	S		Mz	R	BG	O	L	M	SB	S	Mz	R	BG	O	L	M	SB	S
67	6	5	7	7	4	8	7	4	5	4	2	8	4	5	9	9	7	4	5	7	7	4	8	9	4
										1	3	1	7	3	5	8	8	4	2	3	5	1	1	6	6
68	6	5	8	6	4	9	9	3	5	4	3	8	4	6	8	8	7	4	4	7	4	3	8	9	4
										1	3	1	6	2	4	7	7	6	2	1	5	1	1	7	8
69									5	2	6	2	4	8	8	3									
										1				2	7	2									
70	5	3	5	5	2	4			5	3	2	2	4	6	5	5	3	1	3	1	2	2	1		
										1				1	1	2					1				
71									5	3	2	2	1	3	6	4	4				2				
										1	1	1		1	3	2	2								

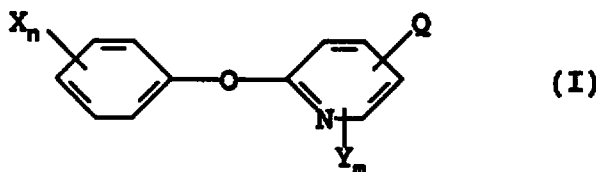
Table 5 (continued)

Compound of Ex. No.	Soil drench 10/kg/ha					Dosage kg/ha	Foliar spray					Pre-emergence					
	Mz	R	BG	O	L	M	SB	S	Mz	R	BG	O	L	M	SB	S	
72	2	1.					5	5	2	2	7	7	4	2	1	3	4
							3	1	2	5	4	3	1				

**Claims**

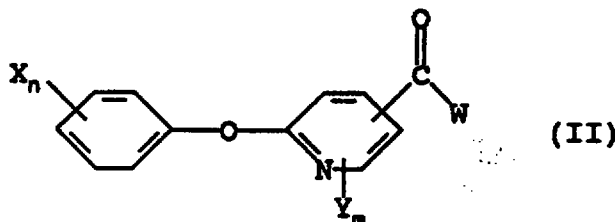
Claims for the following Contracting States : AT, BE, CH, LI, DE, FR, GB, GR, IT, LU, NL, SE

1. A compound of general formula I



in which Q represents a group of general formula $-C(O)-C(CN)=CH-NR^1R^2$, R^1 and R^2 each independently represents a C_{1-12} alkyl group, or together represent $-(CH_2)_4-$ or $-(CH_2)_2-O-(CH_2)_2-$; m is 0 or an integer from 1 to 3, the or each Y independently represents a halogen atom or an alkyl group, n is 0 or an integer from 1 to 5, and the or each X independently represents a halogen atom or a group selected from alkyl, haloalkyl, cyanoalkyl, alkenyl, alkynyl, alkoxy, haloalkoxy, haloalkylthio, alkenyloxy, alkynyloxy, alkylthio, alkenylthio, alkynylthio, cyano, nitro, alkylsulphonyl, and sulphonamido, or a phenyl group optionally substituted by one or more moieties independently selected from alkyl, haloalkyl and alkoxy groups and halogen atoms.

2. A compound as claimed in claim 1, wherein R^1 and R^2 are the same and each represents a methyl group.
3. A compound as claimed in claim 1 or claim 2, wherein the group Q is located at the 3- or 6-position on the pyridyl ring (relative to the nitrogen atom).
4. A compound as claimed in any one of claims 1 to 3, wherein n is 1 or 2, and the or each group X is independently selected from halogen atoms, C_{1-6} alkyl groups, C_{1-6} haloalkyl groups, C_{1-6} alkoxy groups, and cyano groups.
5. A compound as claimed in Claim 4, wherein the or each group X is independently selected from fluorine and chlorine atoms and methyl, trifluoromethyl, methoxy, ethoxy and cyano groups.
6. A compound as claimed in any preceding claim, wherein X_n represents a 3-trifluoromethyl or 3-cyano group.
7. A process for the preparation of a compound of general formula I as claimed in any preceding claim, which comprises reacting a compound of general formula II



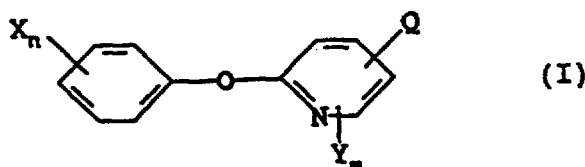
in which X, Y, n and m are as defined in any preceding claim, and W represents a halogen atom or a cyanomethyl group, with, in the case where W is halogen, a dialkylaminoacrylonitrile under basic conditions, or, in the case where W is cyanomethyl, either with a dialkylformamide dialkyl acetal, or with a trialkylorthoformate followed by reaction with a dialkylamine; and if desired or required, converting a resulting compound of general formula I into another compound of general formula I.



8. A process as claimed in claim 7, wherein W is cyanomethyl and the compound of general formula II is reacted with a di(C₁₋₄alkyl)formamide di(C₁₋₄alkyl)acetal in an inert organic solvent at a temperature in the range 0 to 50 °C.
9. A herbicidal composition which comprises a compound as claimed in any one of claims 1 to 6 together with at least one carrier.
10. 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 6, or a composition as claimed in claim 9.
11. The use of a compound as claimed in any one of claims 1 to 6, or a composition as claimed in claim 9, as a herbicide.

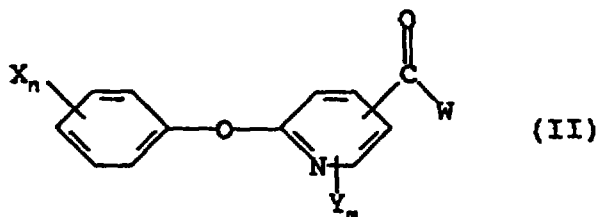
Claims for the following Contracting State : ES

1. A method of combating undesired plant growth at a locus, which comprises treating the locus with a compound of general formula I



in which Q represents a group of general formula $-C(O)-C(CN)=CH-NR^1R^2$, R¹ and R² each independently represents a C₁₋₁₂ alkyl group, or together represent $-(CH_2)_4-$ or $-(CH_2)_2-O-(CH_2)_2$; m is 0 or an integer from 1 to 3, the or each Y independently represents a halogen atom or an alkyl group, n is 0 or an integer from 1 to 5, and the or each X independently represents a halogen atom or a group selected from alkyl, haloalkyl, cyanoalkyl, alkenyl, alkynyl, alkoxy, haloalkoxy, haloalkylthio, alkenyloxy, alkynyloxy, alkylthio, alkenylthio, alkynylthio, cyano, nitro, alkylsulphonyl and sulphonamido, or a phenyl group optionally substituted by one or more moieties independently selected from alkyl, haloalkyl and alkoxy groups and halogen atoms.

2. A method as claimed in Claim 1, wherein R¹ and R² are the same and each represents a methyl group.
3. A method as claimed in Claim 1 or Claim 2, wherein the group Q is located at the 3- or 6- position on the pyridyl ring (relative to the nitrogen atom).
4. A method as claimed in any one of Claims 1 to 3, wherein n is 1 or 2, and the or each group X is independently selected from halogen atoms, C₁₋₆ alkyl groups, C₁₋₆ haloalkyl groups, C₁₋₆ alkoxy groups, and cyano groups.
5. A method as claimed in Claim 4, wherein the or each group X is independently selected from fluorine and chlorine atoms and methyl, trifluoromethyl, methoxy, ethoxy and cyano groups.
6. A method as claimed in any preceding claim, wherein X_n represents a 3-trifluoromethyl or 3-cyano group.
7. A process for the preparation of a compound of general formula I as defined in any preceding claim, which comprises reacting a compound of general formula II



in which X, Y_n and m are as defined in any preceding claim, and W represents a halogen atom or a cyanomethyl group, with, in the case where W is halogen, a dialkylaminoacrylonitrile under basic conditions, or, in the case where W is cyanomethyl, either with a dialkylformamide dialkyl acetal, or with a trialkylorthoformate followed by reaction with a dialkylamine; and if desired or required, converting a resulting compound of general formula I into another compound of general formula I.

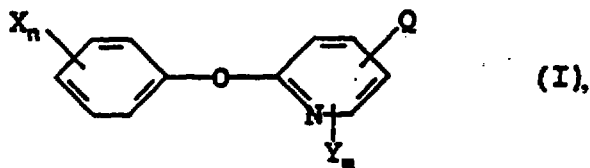
8. A process as claimed in Claim 7, wherein W is cyanomethyl and the compound of general formula II is reacted with a di(C₁-4 alkyl)formamide di(C₁-4 alkyl)acetal in an inert organic solvent at a temperature in the range 0 to 50 °C.

9. A herbicidal composition which comprises a compound as defined in any one of Claims 1 to 6 together with at least one carrier.

Patentansprüche

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

1. Eine Verbindung der allgemeinen Formel I



worin Q für eine Gruppe der allgemeinen Formel $-C(O)-C(CN)=CH-NR^1R^2$ steht, R¹ und R² jeweils unabhängig voneinander eine C₁-12-Alkylgruppe bedeuten oder zusammengekommen für $-(CH_2)_4-$ oder $-(CH_2)_2-O-(CH_2)_2-$ stehen, m den Wert 0 oder eine ganze Zahl von 1 bis 3 aufweist, der oder jeder Rest Y unabhängig voneinander ein Halogenatom oder eine Alkylgruppe darstellt, n den Wert 0 oder eine ganze Zahl von 1 bis 5 aufweist und der oder jeder Rest X unabhängig voneinander ein Halogenatom oder eine unter Alkyl, Halogenalkyl, Cyanoalkyl, Alkenyl, Alkynyl, Alkoxy, Halogenalkoxy, Halogenalkylthio, Alkenyloxy, Alkinyloxy, Alkylthio, Alkenylthio, Alkynylthio, Cyano, Nitro, Alkylsulfonyl und Sulfonamido ausgewählte Gruppe oder eine Phenylgruppe darstellt, die gegebenenfalls durch einen oder durch mehrere Substituenten, unabhängig voneinander ausgewählt unter Alkyl-, Halogenalkyl- und Alkoxygruppen und Halogenatomen, substituiert ist.

2. Verbindung nach Anspruch 1, worin R¹ und R² die gleiche Bedeutung haben und jeweils eine Methylgruppe darstellen.

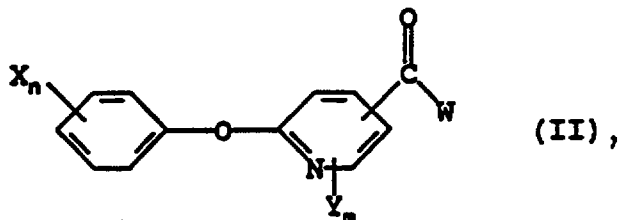
3. Verbindung nach Anspruch 1 oder 2, worin die Gruppe Q an der 3- oder 6-Stellung am Pyridylring (bezogen auf das Stickstoffatom) angeordnet ist.

4. Verbindung nach einem der Ansprüche 1 bis 3, worin n den Wert 1 oder 2 aufweist und die oder jede Gruppe x unabhängig voneinander unter Halogenatomen, C₁-6-Alkylgruppen, C₁-6-Halogenalkylgruppen, C₁-6-Alkoxygruppen und Cyanogruppen ausgewählt ist.

5. Verbindung nach Anspruch 4, worin die oder jede Gruppe X unabhängig voneinander unter Fluor- und Chloratomen und Methyl-, Trifluormethyl-, Methoxy-, Ethoxy- und Cyanogruppen ausgewählt ist.



6. Verbindung nach einem der vorstehenden Ansprüche, worin X_n eine 3-Trifluormethylgruppe oder eine 3-Cyanogruppe bedeutet.
7. Verfahren zur Herstellung einer Verbindung der allgemeinen Formel I, wie in einem der vorstehenden Ansprüche beansprucht, welches ein Umsetzen einer Verbindung der allgemeinen Formel II

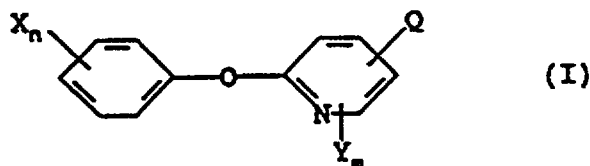


worin X, Y, n und m wie in einem der vorstehenden Ansprüche definiert sind und W ein Halogenatom oder eine Cyanomethylgruppe bedeutet, für den Fall, daß W für Halogen steht, mit einem Dialkylaminoacrylnitril unter basischen Bedingungen oder, für den Fall, daß W für Cyanomethyl steht, entweder mit einem Dialkylformamid-dialkylacetal oder mit einem Trialkylorthoformiat, gefolgt von einer Umsetzung mit einem Dialkylamin, umfaßt; und, falls gewünscht oder erforderlich, ein Überführen einer erhaltenen Verbindung der allgemeinen Formel I in eine andere Verbindung der allgemeinen Formel I umfaßt.

8. Verfahren nach Anspruch 7, worin W für Cyanomethyl steht und die Verbindung der allgemeinen Formel II mit einem $\text{Di}(\text{C}_{1-4}\text{-alkyl})\text{formamid-di}(\text{C}_{1-4}\text{-alkyl})\text{acetal}$ in einem inerten organischen Lösungsmittel bei einer Temperatur im Bereich von 0 bis 50°C umgesetzt wird.
9. Herbizide Zusammensetzung, welche eine Verbindung, wie in einem der Ansprüche 1 bis 6 beansprucht, zusammen mit wenigstens einem Träger umfaßt.
10. Verfahren zur Bekämpfung von unerwünschtem Pflanzenwuchs an einem Ort, welches ein Behandeln des Ortes mit einer Verbindung, wie in einem der Ansprüche 1 bis 6 beansprucht, oder mit einer Zusammensetzung, wie in Anspruch 9 beansprucht, umfaßt.
11. Verwendung einer Verbindung, wie in einem der Ansprüche 1 bis 6 beansprucht, oder einer Zusammensetzung, wie in Anspruch 9 beansprucht, als ein Herbizid.

Patentansprüche für folgenden Vertragsstaat : ES

1. Verfahren zur Bekämpfung von unerwünschtem Pflanzenwuchs an einem Ort, welches ein Behandeln des Ortes mit einer Verbindung der allgemeinen Formel I

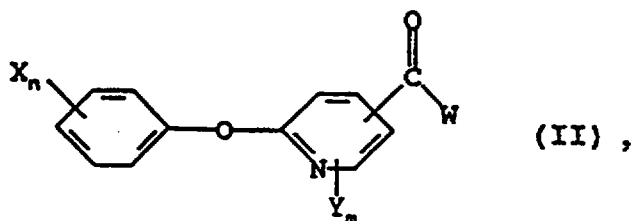


- umfaßt, worin Q für eine Gruppe der allgemeinen Formel $-\text{C}(\text{O})-\text{C}(\text{CN})=\text{CH}-\text{NR}^1\text{R}^2$ steht, R^1 und R^2 jeweils unabhängig voneinander eine C_{1-12} -Alkylgruppe bedeuten oder zusammengenommen für $-(\text{CH}_2)_4-$ oder $-(\text{CH}_2)_2-\text{O}-(\text{CH}_2)_2-$ stehen, m den Wert 0 oder eine ganze Zahl von 1 bis 3 aufweist, der oder jeder Rest Y unabhängig voneinander ein Halogenatom oder eine Alkylgruppe darstellt, n den Wert 0 oder eine ganze Zahl von 1 bis 5 aufweist und der oder jeder Rest X unabhängig voneinander ein Halogenatom oder eine unter Alkyl, Halogenalkyl, Cyanoalkyl, Alkenyl, Alkynyl, Alkoxy, Halogenalkoxy, Halogenalkylthio, Alkenyloxy, Alkinyloxy, Alkylthio, Alkenylthio, Alkinylthio, Cyano, Nitro, Alkylsulfonyl und Sulfonamido ausgewählte Gruppe oder eine Phenylgruppe darstellt, die gegebenenfalls durch einen oder durch mehrere Substituenten, unabhängig voneinander ausgewählt unter Alkyl-, Halogenal-



kyl- und Alkoxygruppen und Halogenatomen, substituiert ist.

2. Verfahren nach Anspruch 1, worin R^1 und R^2 die gleiche Bedeutung haben und jeweils eine Methylgruppe darstellen.
3. Verfahren nach Anspruch 1 oder 2, worin die Gruppe Q an der 3- oder 6-Stellung am Pyridylring (bezogen auf das Stickstoffatom) angeordnet ist.
4. Verfahren nach einem der Ansprüche 1 bis 3, worin n den Wert 1 oder 2 aufweist und die oder jede Gruppe X unabhängig voneinander unter Halogenatomen, C_1 - C_6 -Alkylgruppen, C_1 - C_6 -Halogenalkylgruppen, C_1 - C_6 -Alkoxygruppen und Cyanogruppen ausgewählt ist.
5. Verfahren nach Anspruch 4, worin die oder jede Gruppe X unabhängig voneinander unter Fluor- und Chloratomen und Methyl-, Trifluormethyl-, Methoxy-, Ethoxy- und Cyanogruppen ausgewählt ist.
6. Verfahren nach einem der vorstehenden Ansprüche, worin X_n eine 3-Trifluormethylgruppe oder eine 3-Cyanogruppe bedeutet.
7. Verfahren zur Herstellung einer Verbindung der allgemeinen Formel I, wie in einem der vorstehenden Ansprüche definiert, welches ein Umsetzen einer Verbindung der allgemeinen Formel II



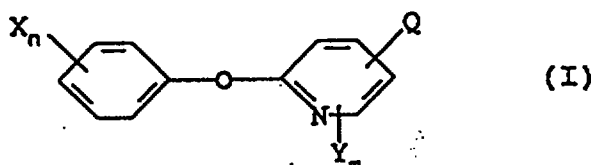
worin X, Y, n und m wie in einem der vorstehenden Ansprüche definiert sind und W ein Halogenatom oder eine Cyanomethylgruppe bedeutet, für den Fall, daß W für Halogen steht, mit einem Dialkylaminoacrylnitril unter basischen Bedingungen oder, für den Fall, daß W für Cyanomethyl steht, entweder mit einem Dialkylformamid-dialkylacetal oder mit einem Trialkylorthoformiat, gefolgt von einer Umsetzung mit einem Dialkylamin, umfaßt; und, falls gewünscht oder erforderlich, ein Überführen einer erhaltenen Verbindung der allgemeinen Formel I in eine andere Verbindung der allgemeinen Formel I umfaßt.

8. Verfahren nach Anspruch 7, worin W für Cyanomethyl steht und die Verbindung der allgemeinen Formel II mit einem Di(C_1 - C_4 -alkyl)formamid-di(C_1 - C_4 -alkyl)acetal in einem inerten organischen Lösungsmittel bei einer Temperatur im Bereich von 0 bis 50°C umgesetzt wird.
9. Herbizide Zusammensetzung, welche eine Verbindung, wie in einem der Ansprüche 1 bis 6 definiert, zusammen mit wenigstens einem Träger umfaßt.

45 Revendications

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

1. Un composé de formule générale I

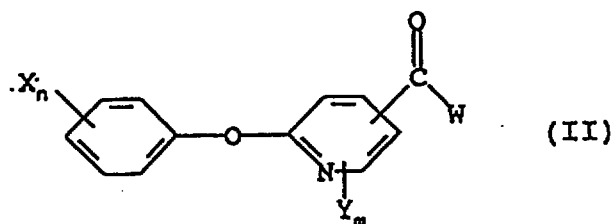


dans laquelle Q représente un groupe de formule générale $-\text{C}(\text{O})-\text{C}(\text{CN})=\text{CH}-\text{NR}^1\text{R}^2$, R^1 et R^2



représentent chacun indépendamment un groupe alcoyle en C_{1-12} ou représentent ensemble $-(CH_2)_4-$ ou $(CH_2)_2-O-(CH_2)_2-$; m est 0 ou un nombre entier de 1 à 3, Y ou chaque Y indépendamment représente un atome d'halogène ou un groupe alcoyle, n est 0 ou un nombre entier de 1 à 5 et X ou chaque X indépendamment représente un atome d'halogène ou un groupe choisi parmi les groupes alcoyle, haloalcoyle, cyanoalcoyle, alcényle, alcynyle, alcoxy, haloalcoxy, haloalcoylthio, alcényloxy, alcynyloxy, alcoylthio, alcynythio, cyano, nitro, alcoylsulfonyl et sulfonamido, ou un groupe phényle éventuellement substitué par une ou plusieurs portions choisies indépendamment parmi des groupes alcoyle, haloalcoyle et alcoxy et des atomes d'halogènes.

2. Un composé selon la revendication 1, dans lequel R^1 et R^2 sont identiques et représentent chacun un groupe méthyle.
3. Un composé selon la revendication 1 ou la revendication 2, dans lequel le groupe Q est situé à la position 3 ou 6 sur le noyau pyridyle (par rapport à l'atome d'azote).
4. Un composé selon l'une quelconque des revendications 1 à 3, dans lequel n est 1 ou 2 et X ou chaque x est choisi indépendamment parmi des atomes d'halogènes, des groupes alcoyle en C_{1-6} , des groupes haloalcoyle en C_{1-6} , des groupes alcoxy en C_{1-6} et des groupes cyano.
5. Un composé selon la revendication 4, dans lequel X ou chaque X est choisi indépendamment parmi des atomes de fluor et de chlore et des groupes méthyle, trifluorométhyle, méthoxy, éthoxy et cyano.
6. Un composé selon l'une quelconque des revendications précédentes, dans lequel X_n représente un groupe 3-trifluorométhyle ou cyano.
7. Un procédé pour la préparation d'un composé de formule générale I tel que revendiqué dans l'une quelconque des revendications précédentes, qui comprend la réaction d'un composé de formule générale II



où x, Y, n et m sont tels que définis dans l'une quelconque des revendications précédentes et W représente un atome d'halogène ou un groupe cyanométhyle, avec, dans le cas où W est un halogène, un dialcoylaminoacrylonitrile dans des conditions basiques, ou, dans le cas où W est un groupe cyanométhyle, soit avec un dialcoyl acétal de dialcoylformamide, soit avec un orthoformate de trialcoyle, cela étant suivi d'une réaction avec une dialcoylamine; et, si on le désire ou si c'est nécessaire, la conversion d'un composé résultant de formule générale I en un autre composé de formule générale I.

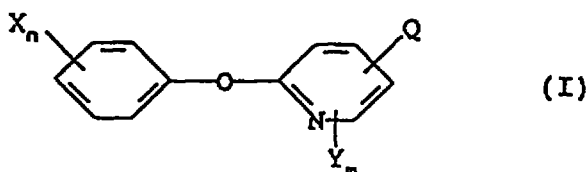
8. Un procédé selon la revendication 7, dans lequel W est un groupe cyanométhyle et le composé de formule générale II est mis à agir avec un $di(C_{1-4} \text{ alcoyl})acétal$ de $di(C_{1-4} \text{ alcoyl})formamide$ dans un solvant organique inerte à une température comprise entre 0 et 50 °C.
9. Une composition herbicide qui comprend un composé selon l'une quelconque des revendications 1 à 6 en même temps qu'au moins un véhicule.
10. 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 6 ou une composition selon la revendication 9.



11. L'utilisation d'un composé selon l'une quelconque des revendications 1 à 6 ou d'une composition selon la revendication 9 comme herbicide.

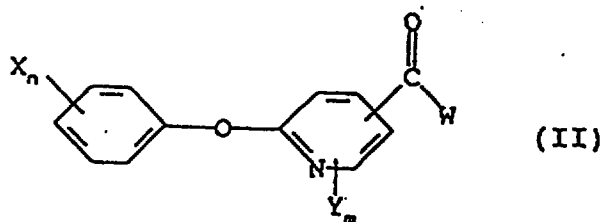
Revendications pour l'Etat contractant suivant : ES

1. Une méthode de lutte contre la croissance de plantes indésirables en un lieu, qui comprend le traitement du lieu avec un composé de formule générale I



dans laquelle Q représente un groupe de formule générale $-C(O)-C(CN)=CH-NR^1R^2$ R^1 et R^2 représentent chacun indépendamment un groupe alcoyle en C_{1-12} ou représentent ensemble $-(CH_2)_4-$ ou $-(CH_2)_2-O-(CH_2)_2-$; m est 0 ou un nombre entier de 1 à 3, Y ou chaque Y indépendamment représente un atome d'halogène ou un groupe alcoyle, n est 0 ou un nombre entier de 1 à 5 et x ou chaque X indépendamment représente un atome d'halogène ou un groupe choisi parmi des groupes alcoyle, haloalcoyle, cyanoalcoyle, alcényle, alcynyle, alcoxy, haloalcoxy, haloalcoylthio, alcényloxy, alcynyloxy, alcoylthio, alcénylthio, alcynylthio, cyano, nitro, alcoylsulfonyl et sulfonamide, ou un groupe phényle éventuellement substitué par une ou plusieurs portions choisies indépendamment parmi des groupes alcoyle, haloalcoyle et alcoxy et des atomes d'halogènes.

2. Une méthode selon la revendication 1, dans laquelle R^1 et R^2 sont identiques et représentent chacun un groupe méthyle.
3. Une méthode selon la revendication 1 ou la revendication 2, dans laquelle le groupe Q est situé à la position 3 ou 6 sur le noyau pyridyle (par rapport à l'atome d'azote).
4. Une méthode selon l'une quelconque des revendications 1 à 3, dans laquelle n est 1 ou 2 et le groupe X ou chaque groupe X est choisi indépendamment parmi des atomes d'halogènes, des groupes alcoyle en C_{1-6} , des groupes haloalcoyle en C_{1-6} , des groupes alcoxy en C_{1-6} , et des groupes cyano.
5. Une méthode selon la revendication 4, dans laquelle le groupe X ou chaque groupe X est choisi indépendamment parmi des atomes de fluor et de chlore et des groupes méthyle, trifluorométhyle, méthoxy, éthoxy et cyano.
6. Une méthode selon l'une quelconque des revendications précédentes, dans laquelle X_n représente un groupe 3-trifluorométhyle ou 3-cyano.
7. Un procédé pour la préparation d'un composé de formule générale I tel que défini dans l'une quelconque des revendications précédentes, qui comprend la réaction d'un composé de formule générale II



où X, Y, n et m sont tels que définis dans l'une quelconque des revendications précédentes, et W



représente un atome d'halogène ou un groupe cyanométhyle, avec, dans le cas où W est un halogène, un dialcoylaminoacrylonitrile dans des conditions basiques, ou, dans le cas où W est un groupe cyanométhyle, soit avec un dialcoyl acétal de dialcoylformamide, soit avec un orthoformiate de trialcoyle, cela étant suivi d'une réaction avec une dialcoylamine ; et, si on le désire ou si c'est nécessaire, la conversion d un composé résultant de formule générale I en un autre composé de formule générale I.

8. Un procédé selon la revendication 7, dans lequel W est un groupe cyanométhyle et le composé de formule générale II est mis à agir avec un di(C₁₋₄ alcoyl)acétal de di(C₁₋₄ alcoyl)formamide dans un solvant organique inerte à une température comprise entre 0 et 50 ° C.
9. Une composition herbicide qui comprend un composé tel que défini dans l'une quelconque des revendications 1 à 6 en même temps qu'au moins un véhicule.