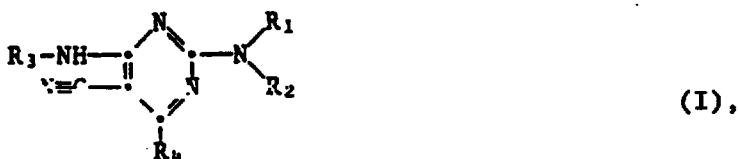


## Description

The present invention relates to novel stable, substituted 2,4-diamino-5-cyanopyrimidines and salts and intermediates thereof, to processes for their preparation and to their use for controlling noxious insects and ectoparasites.

The novel stable compounds according to the invention have the general formula I



wherein

$\text{R}_1$  is hydrogen,  $\text{C}_1\text{-C}_6$ alkyl,  $\text{C}_2\text{-C}_6$ alkenyl or  $\text{C}_2\text{-C}_6$ alkynyl,

$\text{R}_2$  is hydrogen,  $\text{C}_1\text{-C}_{10}$ alkyl or  $\text{C}_3\text{-C}_6$ cycloalkyl, or

$\text{R}_1$  and  $\text{R}_2$ , when taken together, are one of the radicals  $\text{-(CH}_2\text{)}_3\text{-}$ ,  $\text{-(CH}_2\text{)}_4\text{-}$  or  $\text{-(CH}_2\text{)}_5\text{-}$ ,

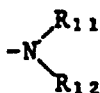
$\text{R}_3$  is hydrogen or one of the radicals  $\text{-CO-R}_5$  or  $\text{-SO}_2\text{-R}_6$ ,

$\text{R}_4$  is one of the radicals

$\text{-NH}_2$ ,  $\text{-NH-CO-R}_5$ ,  $\text{-NH-SO}_2\text{-R}_6$ ,



$\text{R}_5$  is hydrogen,  $\text{C}_1\text{-C}_{12}$ alkyl,  $\text{C}_3\text{-C}_6$ cycloalkyl,  $\text{C}_1\text{-C}_6$ alkoxy, perhalogenated  $\text{C}_1\text{-C}_3$ alkyl or the radical

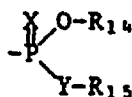


$\text{R}_6$  is  $\text{C}_1\text{-C}_6$ alkyl,

$\text{R}_7$  is hydrogen or  $\text{C}_1\text{-C}_6$ alkyl,

$\text{R}_8$  and  $\text{R}_9$  are each independently of the other hydrogen or  $\text{C}_1\text{-C}_6$ alkyl or, when taken together, are one of the radicals  $\text{-(CH}_2\text{)}_3\text{-}$ ,  $\text{-(CH}_2\text{)}_4\text{-}$  or  $\text{-(CH}_2\text{)}_5\text{-}$ ,

$\text{R}_{10}$  is one of the radicals  $\text{-SO}_2\text{-R}_{13}$  or



$R_{11}$  and  $R_{12}$  are each independently of the other hydrogen or  $C_1$ - $C_6$ alkyl or, when taken together, are one of the radicals  $-(CH_2)_3$ -,  $-(CH_2)_4$ - or  $-(CH_2)_5$ -,

$R_{13}$  is  $C_1$ - $C_{10}$ alkyl,  $C_1$ - $C_{10}$ alkyl which is substituted by up to 10 halogen atoms, or is  $C_3$ - $C_6$ cycloalkyl,

$R_{14}$  and  $R_{15}$  are each independently of the other  $C_1$ - $C_{10}$ alkyl; and

X and Y are each independently of the other oxygen or sulfur, and salts thereof.

Within the scope of this invention, halogen atoms and halogen substituents will be understood as meaning fluorine, chlorine, bromine or iodine, with fluorine and chlorine being preferred.

On account of their advantageous action, preferred compounds of the formula I are those wherein

$R_1$  is hydrogen or  $C_1$ - $C_4$ alkyl,

$R_2$  is  $C_1$ - $C_4$ alkyl or  $C_3$ - $C_6$ cycloalkyl, or

$R_1$  and  $R_2$ , when taken together, are one of the radicals  $-(CH_2)_4$ - or  $-(CH_2)_5$ -,

$R_3$  is hydrogen,

$R_4$  is one of the radicals  $-NH_2$  or  $-NH-CO-R_5$ , and

$R_5$  is  $C_1$ - $C_4$ alkyl or perhalogenated  $C_1$ - $C_3$ alkyl, and salts thereof, as well as those compounds of the formula I, wherein

$R_1$  is hydrogen, methyl or ethyl,

$R_2$  is  $C_1$ - $C_4$ alkyl or cyclopropyl,

$R_3$  is hydrogen,

$R_4$  is one of the radicals  $-NH_2$  or  $-NH-CO-R_5$ , and

$R_5$  is  $C_1$ - $C_3$ alkyl,

and salts thereof.

On account of their high pesticidal activity, particularly noteworthy compounds of the formula I are those wherein

$R_1$  is hydrogen, methyl or ethyl,

$R_2$  is cyclopropyl,

$R_3$  is hydrogen, and

$R_4$  is one of the radicals  $-NH_2$  or  $-NH-CO-C_2H_5$ ,

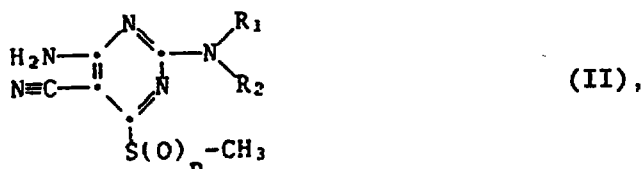
and salts thereof.

By salts of compounds of the formula I are meant the physiologically tolerable addition salts or inorganic and organic acids. Examples of inorganic acids are hydrochloric acid, hydrobromic acid, hydriodic acid, sulfuric acid, phosphoric acid, phosphorous acid and nitric acid.

Examples of organic acids are trifluoroacetic acid, trichloroacetic acid, formic acid, oxalic acid, succinic acid, maleic acid, lactic acid, glycolic acid, aconitic acid, citric acid, benzoic acid, benzenesulfonic acid and methanesulfonic acid.

The compounds of the formula I can be prepared by

(a) reacting a compound of the formula II



with ammonia, or

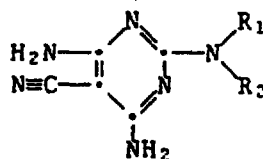
(b) the compound of the formula III



with an amine of the formula IV

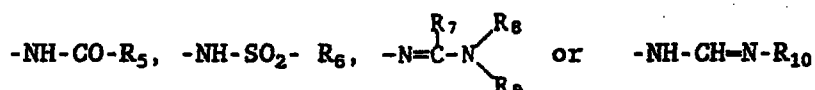


and, if desired, converting the particular resultant compound of the formula I, in which  $R_3$  is hydrogen and  $R_4$  is the radical  $-NH_2$  (compound Ia)



(IIa [sic]),

in a manner known per se, into a compound of the formula I, in which  $R_3$  has the meanings  $-CO-R_5$  or  $-SO_2-R_6$  and/or  $R_4$  has the meanings



where  $R_1$  to  $R_{15}$ , X and Y are as defined in claim 1 and n is 0, 1 or 2, and, if desired, converting a resultant compound of the formula I into a salt thereof.

Within the above process variant a), the reaction of a sulfonyl compound ( $n = 2$ ) of the formula II with ammonia is generally carried out in the temperature range from about 10 to 100°C in an organic solvent such as acetonitrile, tetrahydrofuran, dioxane or a solvent/water mixture. The same reaction conditions apply to process variant b), where 2-chloro-4,6-di-amino-5-cyanopyrimidine of the formula III is reacted with an amine of the formula IV. When in process variant a), methylthio compounds ( $n = 0$ ) of the formula (II) are used, they are reacted with ammonia, preferably in an autoclave under elevated pressure, in the temperature range from 100 to 170°C, usually at about 160°C.

The said conversion of a compound of the formula I, wherein  $R_4$  is  $-NH_2$ , into a compound in which  $R_4$  has another of the meanings indicated according to the invention can be effected by reaction of the amino group in the 6-position of the pyrimidine ring with appropriate reactants which are known per se. Thus, for example, a 6-aminopyrimidine of the formula I can be reacted with compounds of the  $Hal-CO-R_5$ ,  $Hal-SO_2-R_6$ ,  $(R_5-CO)_2O$  or  $R_6O-CH=N-R_{10}$  type to give compounds of the formula I which are suitably substituted in the 6-position, where Hal is a halogen atom, preferably chlorine, and  $R_5$ ,  $R_6$  and  $R_{10}$  have the meanings given above. If it is desired to prepare a compound of the formula I, wherein  $R_4$  is the

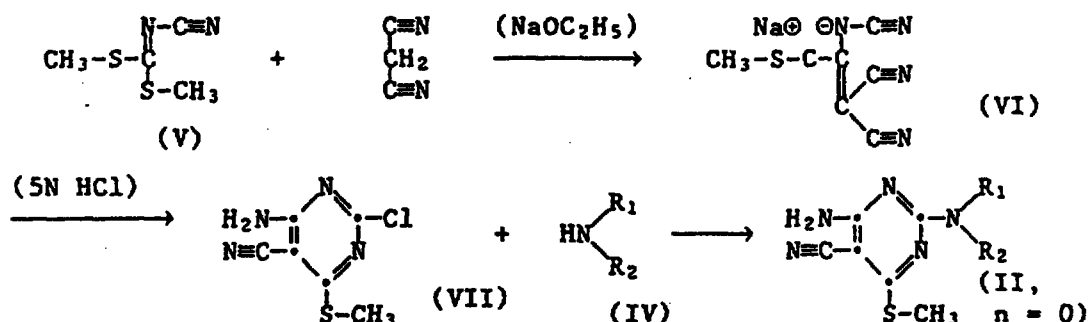
radical  $\begin{array}{c} R_7 \quad R_8 \\ \diagdown \quad / \\ -N=C-N \\ \diagup \quad \diagdown \\ R_9 \end{array}$ , the 6-NH<sub>2</sub> group is reacted, for example, with an acetal

of the formula  $\begin{array}{c} R_6O \quad R_7 \quad R_8 \\ \diagdown \quad / \quad \diagup \\ C-N \\ \diagup \quad \diagdown \\ R_6O \quad R_9 \end{array}$

where R<sub>6</sub>, R<sub>7</sub>, R<sub>8</sub> and R<sub>9</sub> have the meanings given above. In analogous manner and using appropriate reactants it is possible to prepare compounds of the formula I, wherein R<sub>3</sub> is one of the radicals -CO-R<sub>5</sub> or -SO<sub>2</sub>-R<sub>6</sub>, from 4-aminopyrimidines of the formula I, wherein R<sub>3</sub> is hydrogen. The above-described acylation reactions are carried out under normal pressure in inert solvents or diluents and in the presence of a base at temperatures from 0 to 120°C, preferably from 40 to 80°C. Examples of suitable solvents and diluents are: alkanes such as n-pentane as well as homologues thereof, including isomers up to n-heptadecane; ethers such as diethyl ether, dipropyl ether, dibutyl ether, dimethoxyethane, dioxane or tetrahydrofuran; chlorinated hydrocarbons such as chloroform, methylene chloride, carbon tetrachloride or chlorobenzene; aromatic hydrocarbons such as benzene, toluene or xylenes. Further inert solvents and diluents may also be used in the process. Possible bases which may be used in these reactions are, for example, alkylamines such as triethylamine or diisopropylethylamine, as well as pyridine or N-methylpyrrolidone.

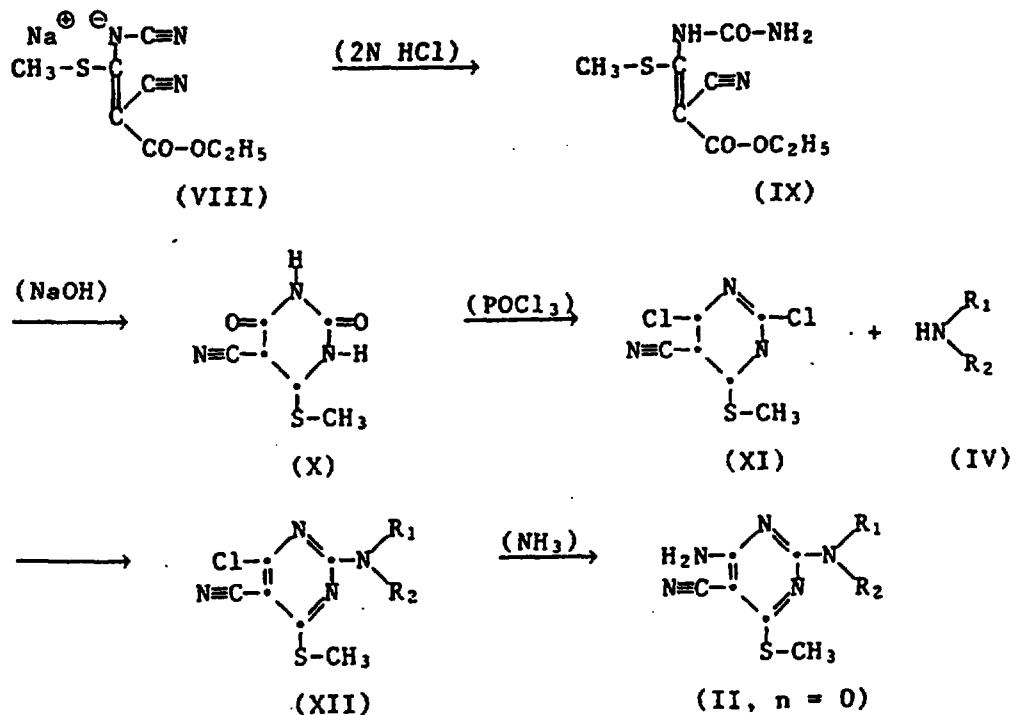
The starting compounds of the formula II are novel and likewise constitute a subject of the present invention, especially because they also have good pesticidal, in particular insecticidal, properties. They can be prepared as follows:

a) For example, a compound of the formula II in which n = 0 can be obtained as indicated schematically below:



The dimethyl N-cyanodithiocarboximide of the formula V used as starting material for the above procedure and the salt of the formula VI formed by reaction with malonitrile are known (cf. Rec. Trav. Ch. 90/1971, 463; J. Chem. Soc., Chem. Comm. 1974, 350). A compound of the formula II in which  $n = 0$  can be obtained from the salt VI by cyclization with 5N HCl and subsequent reaction of the resultant pyrimidine of the formula VII with a corresponding amine of the formula IV.

b) In addition, compounds of the formula II in which  $n = 0$  can be obtained as follows:

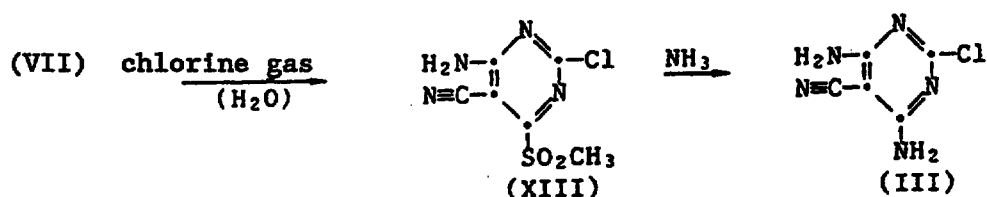


The sodium salt of the formula VIII indicated in the above reaction

scheme, the urea of the formula IX and the uracil of the formula X are known (cf. J. Chem. Soc., Chem. Comm. 1974, 350; Helv. Chim. Act. 1985, 1155). The preparation of the novel substituted pyrimidines of the formulae XI and XII is carried out in a manner known per se. Reaction of XII with ammonia gives a compound of the formula II in which  $n = 0$ .

c) Starting compounds of the formula II, wherein  $n = 1$  or  $2$ , can be prepared in a manner known per se by oxidation of compounds of the formula II, wherein  $n = 0$  (cf. "The Chemistry of Heterocyclic Compounds", Vol. 16: Pyrimidines, Intersc. Publ. Inc., N.Y. 1959).

The starting compound of the formula III, i.e. 2-chloro-4,6-di-amino-5-cyanopyrimidine, is known from Chem. Ber. 1968, 1244, but can be prepared from the above compound of the formula VII in accordance with the following scheme by oxidation to the sulfone and replacement of  $-SO_2-CH_3$  by  $-NH_2$  using ammonia:



Pesticidally, in particular insecticidally, active 2,4,6-triamino-5-nitro-pyrimidines, the amino groups of which may optionally be substituted, are already known from European patent application 0 084 758. The active substances of the formulae I and II according to the invention differ from these known compounds in terms of structure essentially by the presence of the cyano group in the 5-position.

Surprisingly, it has been found that the present compounds of the formulae I and II and salts thereof exhibit excellent activity as pesticides while being well tolerated by plants and having low toxicity to warm-blooded animals. They are particularly suitable for controlling insects and representatives of the order Acarina which infest plants and animals.

In particular, the compounds of the formulae I and II are suitable for

controlling insects of the orders: Lepidoptera, Coleoptera, Homoptera, Heteroptera, Diptera, Thysanoptera, Orthoptera, Anoplura, Siphonaptera, Mallophaga, Thysanura, Isoptera, Psocoptera and Hymenoptera, as well as representatives of the order Acarina of the families: Ixodidae, Argasidae, Tetranychidae and Dermanyssidae.

In addition to their action against flies, e.g. *Musca domestica*, and mosquito larvae, the active compounds of this invention are also suitable for controlling plant-destructive feeding insects in ornamentals and crops of useful plants, especially in cotton (e.g. against *Spodoptera littoralis* and *Heliothis virescens*) and in fruit and vegetables (e.g. against *Plutella xylostella*, *Laspeyresia pomonella*, *Leptinotarsa decemlineata* and *Epilachna varivestis*). The novel compounds of the formula I also possess a good activity against pests in rice. The compounds of the formula I also have a pronounced larvicidal action against insects, especially against larvae of noxious feeding insects. If the active compounds are ingested by adult insect stages with the feed, then a diminished oviposition and/or reduced hatching rate is observed in many insects, especially in Coleopterae, e.g. *Anthonomus grandis*.

The active compounds of the formulae I and II can also be used for controlling ectoparasites such as *Lucilia sericata*, in domestic animals and productive livestock, e.g. by treating animals, livestock buildings and pastures, with very good results.

In the treatment of grazing animals with the compounds of this invention, for example by means of cattle dips, pour-on methods or spray procedures, the surprising adhesive action of the active substances provides a long-lasting toxic effect against ectoparasites, for example harmful Diptera, on the skin and fur of the animals. This prevents the active compounds which have been applied to the skin or fur of productive livestock from being prematurely washed out or washed off by rainwater as it runs off.

A particular advantage of the active compounds of the invention resides in their oral administration to productive livestock. In this method of application the active compounds exhibit an effective and prolonged

insecticidal activity, in particular in the faeces excreted from the alimentary canal. Consequently, infestation by harmful insects, in particular Diptera, can be prevented before the pests occur in the vicinity of the animals, e.g. in livestock buildings, in enclosures and on grazing land, as the Diptera larvae hatching from the deposited eggs are killed immediately. A particularly important feature of this special form of application is that, by virtue of their structural properties, the compounds are physiologically harmless to warm-blooded animals. This method of selectively controlling the proliferation of insects is considerably more efficient and at the same time more economical than the customary large-scale disinfection of livestock buildings and enclosures.

The good pesticidal action of the compounds of the formulae I and II of the invention corresponds to a mortality of at least 50-60 % of the abovementioned pests.

The action of the compounds of the invention and of the agents containing them can be substantially broadened and adapted to prevailing circumstances by addition of other insecticides and/or acaricides. Examples of suitable additives include representatives from the following classes of active compounds: organophosphorus compounds, nitrophenols and derivatives thereof, formamidines, ureas, carbamates, pyrethroids, chlorinated hydrocarbons, and *Bacillus thuringiensis* preparations.

The compounds of the invention are used in unmodified form, or preferably together with the adjuvants conventionally employed in the art of formulation, and can therefore be formulated in known manner, for example, to emulsifiable concentrates, directly sprayable or dilutable solutions, dilute emulsions, wettable powders, soluble powders, dusts, granules, and also encapsulations in e.g. polymeric substances. As with the nature of the agents, the methods of application, such as spraying, atomising, dusting, scattering or pouring, are chosen in accordance with the intended objectives and the prevailing circumstances.

The formulation, i.e. the agents, preparations or compositions

containing the active compound of the formula I or II, or combinations of these active compounds with other insecticides or acaricides, and, where appropriate, a solid or liquid adjuvant, are prepared in a known manner, e.g. by intimately mixing and/or grinding the active compounds with extenders, such as, for example, solvents, solid carriers and, where appropriate, surface-active compounds (surfactants).

Suitable solvents may be: aromatic hydrocarbons, preferably the C<sub>8</sub> to C<sub>12</sub> fractions, such as, for example, xylene mixtures or substituted naphthalenes, phthalic acid esters such as dibutyl phthalate or dioctyl phthalate, aliphatic hydrocarbons such as cyclohexane or paraffins, alcohols and glycols and their ethers and esters, such as ethanol, ethylene glycol, ethylene glycol monomethyl or monoethyl ether, ketones such as cyclohexanone, strongly polar solvents such as N-methyl-2-pyrrolidone, dimethyl sulfoxide or dimethylformamide, as well as vegetable oils or epoxidised vegetable oils such as epoxidised coconut oil or soybean oil; or water.

The solid carriers used e.g. for dusts and dispersible powders are normally natural mineral fillers such as calcite, talcum, kaolin, montmorillonite or attapulgite. In order to improve the physical properties it is also possible to add highly disperse silicas or highly disperse absorbent polymers. Suitable granulated adsorptive carriers are porous types, for example pumice, broken brick, sepiolite or bentonite; and suitable nonsorbent carriers are materials such as calcite or sand. In addition, a great number of granulated materials of inorganic or organic nature can be used, e.g. especially dolomite or pulverised plant residues.

Depending on the nature of the active compound to be formulated, or of combinations of these active compounds with other insecticides or acaricides, suitable surface-active compounds are non-ionic, cationic and/or anionic surfactants having good emulsifying, dispersing and wetting properties. The term "surfactants" will also be understood as comprising mixtures of surfactants.

Suitable anionic surfactants can be both water-soluble soaps and

water-soluble synthetic surface-active compounds.

Suitable soaps are the alkali metal salts, alkaline earth metal salts or unsubstituted or substituted ammonium salts of higher fatty acids ( $C_{10}$ - $C_{22}$ ), e.g. the Na salts or K salts of oleic or stearic acid, or of natural fatty acid mixtures which can be obtained, e.g. from coconut oil or tallow oil. Furthermore, the fatty acid methyltaurine salts as well as modified and unmodified phospholipids should also be mentioned as surfactants.

More frequently, however, so-called synthetic surfactants are used, especially fatty sulfonates, fatty sulfates, sulfonated benzimidazole derivatives or alkylarylsulfonates.

The fatty sulfonates or sulfates are usually in the form of alkali metal salts, alkaline earth metal salts or unsubstituted or substituted ammonium salts and contain an alkyl radical having 8 to 22 carbon atoms, alkyl also including the alkyl moiety of acyl radicals, e.g. the Na or Ca salt of lignosulfonic acid, of dodecyl sulfate, or of a mixture of fatty alcohol sulfates obtained from natural fatty acids. These also comprise the salts of sulfuric acid esters and sulfonic acids of fatty alcohol/ethylene oxide adducts.

The sulfonated benzimidazole derivatives preferably contain 2 sulfonic acid groups and one fatty acid radical having about 8 to 22 carbon atoms. Examples of alkylarylsulfonates are the Na, Ca or triethanolamine salts of dodecylbenzenesulfonic acid, dibutyl-naphthalenesulfonic acid, or of a condensate of naphthalenesulfonic acid and formaldehyde. Also suitable are corresponding phosphates, e.g. salts of the phosphoric acid ester of an adduct of p-nonylphenol with 4 to 14 moles of ethylene oxide.

Non-ionic surfactants are preferably polyglycol ether derivatives of aliphatic or cycloaliphatic alcohols, saturated or unsaturated fatty acids and alkylphenols, said derivatives containing 3 to 30 glycol ether groups and 8 to 20 carbon atoms in the (aliphatic) hydrocarbon moiety and 6 to 18 carbon atoms in the alkyl moiety of the alkylphenols.

Further suitable non-ionic surfactants are the water-soluble adducts of polyethylene oxide with polypropylene glycol, ethylenediaminopolypropylene glycol and alkylpolypropylene glycol having 1 to 10 carbon atoms in the alkyl chain, which adducts contain 20 to 250 ethylene glycol ether groups and 10 to 100 propylene glycol ether groups. The compounds stated usually contain 1 to 5 ethylene glycol units per propylene glycol unit.

Examples of non-ionic surfactants which may be mentioned are nonylphenolpolyethoxyethanols, castor oil polyglycol ethers, polypropylene/polyethylene oxide adducts, tributylphenoxypolyethoxyethanol, polyethylene glycol and octylphenoxypolyethoxyethanol. Fatty acid esters of polyoxyethylene sorbitan, e.g. polyoxyethylene sorbitan trioleate, are also suitable.

Cationic surfactants are in particular quaternary ammonium salts which contain, as N-substituent, at least one alkyl radical having 8 to 22 carbon atoms and, as further substituents, lower alkyl, which may be halogenated alkyl, benzyl or lower hydroxyalkyl radicals. The salts are preferably in the form of halides, methylsulfates or ethylsulfates, e.g. stearyltrimethylammonium chloride or benzyldi(2-chlorethyl)ethylammonium bromide.

The surfactants customarily employed in the art of formulation are described, inter alia, in the following publications:

"McCutcheon's Detergents and Emulsifiers Annual", MC Publishing Corp., Ridgewood, New Jersey, 1979; Dr. Helmut Stache, "Tensid Taschenbuch" (Handbook of Surfactants), Carl Hanser Verlag, Munich/Vienna 1981.

The pesticidal preparations usually contain, as a rule, 0.1 to 99 %, in particular 0.1 to 95 %, of active compound according to the invention or combinations of this active compound with other insecticides or acaricides, 1 to 99.9 % of a solid or liquid adjuvant, and 0 to 25 %, in particular 0.1 to 20 %, of a surfactant. Whereas commercial products are preferably concentrated agents, the end user will normally employ

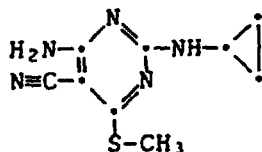
diluted formulations with substantially lower concentrations of active compound.

The agents may also contain further auxiliaries, such as stabilisers, antifoams, viscosity regulators, binders, tackifiers as well as fertilisers or other active compounds for obtaining special effects.

Example 1

a) Preparation of the starting compound 2-cyclopropylamino-4-amino-5-cyano-6-methylthiopyrimidine:

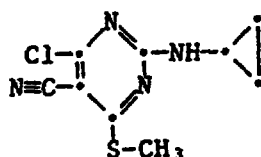
- i) While cooling with ice water, 650 ml of concentrated hydrochloric acid are added dropwise to a solution of 93 g of 2-cyano-3-cyanoamino-3-methylthioacrylonitrile, in the form of its sodium salt, in 425 ml of water. The batch is then stirred for about 12 hours at room temperature. The precipitate formed is isolated by filtration and treated with aqueous sodium carbonate solution. 20 g of the resultant 2-chloro-4-amino-5-cyano-6-methylthiopyrimidine (m.p. 268°C) are stirred in 250 ml of acetonitrile to give a suspension to which, with vigorous stirring, 11.4 g of cyclopropylamine are added dropwise at reflux temperature. The batch is stirred for about a further 12 hours and then water is added. The precipitate formed is filtered off. The title compound of the formula



with a melting point of 215-218°C is thus obtained (compound No. 3.1).

- ii) 20 g of N,N-diethylaniline are slowly added dropwise to a mixture of 18.3 g of 5-cyano-6-methylthiouracil and 80 ml of phosphorus oxychloride. The batch is then refluxed for 1.5 hours and subsequently concentrated by evaporation. The residue is mixed with 200 ml of ice-water, stirred, collected by filtration and washed with water. After drying, the crude product is chromatographed using

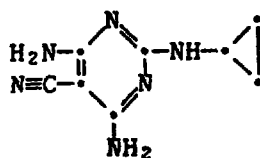
silica gel (6:3:1 toluene/chloroform/ethyl acetate). 22 g of the resultant 2,4-dichloro-5-cyano-6-methylthio-pyrimidine (m.p. 118-120°C) are dissolved in 200 ml of acetonitrile, and a solution of 11.4 g of cyclopropylamine in 40 ml of acetonitrile is added dropwise at -10°C. The reaction mixture is stirred for 2 hours at room temperature and then poured into 2 litres of ice-water. The residue formed is filtered off with suction. 26.1 g of the 2-cyclopropylamino-4-chloro-5-cyano-6-methylthio-pyrimidine obtained by the above procedure, of the formula



with a melting point of 139-141°C, are stirred in 100 ml of acetonitrile. 300 ml of 30 % aqueous ammonia are added to the suspension thus formed, which is then stirred for about 10 hours at room temperature and thereafter for 5 hours under reflux. The solid residue obtained from the cooled batch is filtered off with suction and washed with water. After recrystallisation from 150 ml of methyl cellusolve[sic], the title compound of the formula indicated under i) with a melting point of 215-218°C is obtained.

b) Preparation of 2-cyclopropylamino-4,6-diamino-5-cyano-pyrimidine:

33.7 g of the 2-cyclopropylamino-4-amino-5-cyano-6-methylthiopyrimidine prepared in accordance with a) are treated with 150 g of ammonia in an autoclave at 150°C for 15-20 hours. The reaction product formed is stirred repeatedly in water and then isolated by filtration. The title compound of the formula

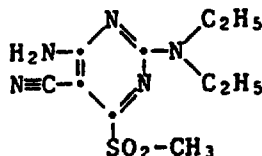


with a melting point of 249-251°C is obtained in this way (compound No. 1.1).

Example 2

a) Preparation of the starting compound 2-diethylamino-4-amino-5-cyano-6-methylsulfonylpyrimidine:

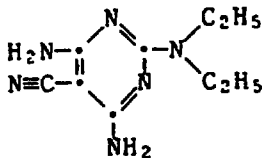
47.5 g of 2-diethylamino-4-amino-5-cyano-6-methylthio-pyrimidine are added to 650 ml of methylene chloride. Without cooling, 88.7 g of 3-chloroperbenzoic acid are added to the batch. The reaction mixture is stirred for 2 hours and then filtered. The filtrate is concentrated by evaporation and the residue is suspended in diethyl ether and filtered with suction. The title compound of the formula



with a melting point of 170-172°C is obtained (compound No. 3.2).

b) Preparation of 2-diethylamino-4,6-diamino-5-cyano-pyrimidine:

26.9 g of the 2-diethylamino-4-amino-5-cyano-6-methylsulfonylpyrimidine prepared in accordance with a) are added to a mixture consisting of 300 ml of 30 % aqueous ammonia and 100 ml of acetonitrile. The batch is then kept under reflux for 12 hours, cooled, and the precipitate is isolated by filtration. The precipitate is treated with water and filtered off with suction. The title compound of the formula

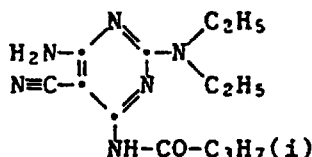


with a melting point of 222-224°C is thus obtained (compound No. 1.2).

Example 3

Preparation of 2-diethylamino-4-amino-5-cyano-6-isobutyryl-aminopyrimidine:

7 g of triethylamine are added to a solution of 10.3 g of 2-diethylamino-4,6-diamino-5-cyanopyrimidine (prepared in accordance with Example 2) in 90 ml of tetrahydrofuran. 8.5 g of isobutyric anhydride are added dropwise to this solution at 60°C. The reaction mixture is heated for 48 hours under reflux, concentrated by evaporation, and the residue formed is washed with diethyl ether. The title compound thus obtained of the formula

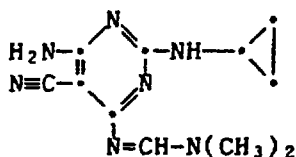


has a melting point of 151-152°C (compound No. 1.3).

Example 4

Preparation of N,N-dimethyl-N'-(2-cyanopropylamino-4-amino-5-cyanopyrimidin-6-yl)formamide

3.7 g of dimethylformamide diethyl acetal are added dropwise at 45-50°C to a solution of 3.8 g of 2-cyclopropylamino-4,6-diamino-5-cyanopyrimidine in 100 ml of dioxane. The reaction mixture is stirred for 8 hours at 45-50°C. After distilling off the solvent and recrystallizing the crude product from ethanol, the title compound of the formula



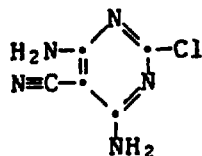
with a melting point of 192-195°C is obtained (compound No. 1.4).

Example 5

Preparation of the starting compound 2-chloro-4,6-diamino-5-cyano-pyrimidine:

A suspension of 50 g of finely powdered 2-chloro-4-amino-5-cyano-6-methylthiopyrimidine in 750 ml of dioxane and 250 ml of water is taken as the initial charge. A vigorous stream of chlorine is then passed into this suspension with stirring and cooling. A clear solution is formed after about 20 minutes. 30 minutes after the introduction of chlorine is terminated and the batch is stirred to complete reaction for 1/2 hour. The reaction mixture is concentrated by evaporation, the residue is suspended in 600 ml of ice-water and the suspension is filtered. 46.4 g of the filtration residue, 2-chloro-4-amino-5-cyano-6-methylsulfonyl-pyrimidine (m.p. 253°C, with decomposition) are stirred in 25 g of 30 % aqueous ammonia and 1000 ml of acetonitrile for 2 hours at room temperature. After concentrating the batch by evaporation, the residue is suspended and filtered.

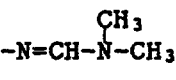
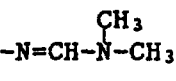
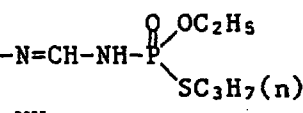
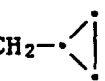
In this way the title compound of the formula



with a melting point of >260°C is obtained (compound No. 5.1).

The following compounds of the formula I are also prepared in analogy with the procedures described above:

| Compound No. | R <sub>1</sub>                     | R <sub>2</sub>                                      | R <sub>3</sub> | R <sub>4</sub>                           | m.p. [°C] |
|--------------|------------------------------------|-----------------------------------------------------|----------------|------------------------------------------|-----------|
| 1.5          | H                                  | -C <sub>3</sub> H <sub>7</sub> (i)                  | H              | -NH <sub>2</sub>                         | 230-232   |
| 1.6          | H                                  | -CH(CH <sub>3</sub> )-C <sub>2</sub> H <sub>5</sub> | H              | -NH <sub>2</sub>                         | 195-197   |
| 1.7          | -CH <sub>3</sub>                   | -CH <sub>3</sub>                                    | H              | -NH <sub>2</sub>                         | > 260     |
| 1.8          | -C <sub>3</sub> H <sub>7</sub> (n) | -C <sub>3</sub> H <sub>7</sub> (n)                  | H              | -NH <sub>2</sub>                         | 204-206   |
| 1.9          | -C <sub>4</sub> H <sub>9</sub> (n) | -C <sub>4</sub> H <sub>9</sub> (n)                  | H              | -NH <sub>2</sub>                         | 182-185   |
| 1.10         | -CH <sub>3</sub>                   | -C <sub>2</sub> H <sub>5</sub>                      | H              | -NH <sub>2</sub>                         | 250-251   |
| 1.11         |                                    | -(CH <sub>2</sub> ) <sub>4</sub> -                  | H              | -NH <sub>2</sub>                         | > 260     |
| 1.12         |                                    | -(CH <sub>2</sub> ) <sub>5</sub>                    | H              | -NH <sub>2</sub>                         | 234-236   |
| 1.13         | H                                  | -C(CH <sub>3</sub> ) <sub>3</sub>                   | H              | -NH <sub>2</sub>                         | 180-183   |
| 1.14         | -CH <sub>3</sub>                   | -C <sub>4</sub> H <sub>9</sub> (n)                  | H              | -NH <sub>2</sub>                         | 194-196   |
| 1.15         | H                                  |                                                     | H              | -NH-CO-CH(CH <sub>3</sub> ) <sub>2</sub> | 215-218   |
| 1.16         | H                                  |                                                     | H              | -NH-CO-C <sub>2</sub> H <sub>5</sub>     | 180-181   |
| 1.17         | H                                  |                                                     | H              | -NH-CO-CH <sub>3</sub>                   | 203-205   |
| 1.18         | -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                      | H              | -NH-CO-C <sub>2</sub> H <sub>5</sub>     | 158-160   |
| 1.19         | -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                      | H              | -NH-CO-C(CH <sub>3</sub> ) <sub>3</sub>  | 113-115   |
| 1.20         | -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                      | H              | -NH-CO-CH <sub>3</sub>                   | 168-171   |
| 1.21         | -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                      | H              | -NH-CO-C <sub>3</sub> H <sub>7</sub> (n) | 149-150   |
| 1.22         | H                                  |                                                     | H              | -NH-CO-C(CH <sub>3</sub> ) <sub>3</sub>  | 110-113   |
| 1.23         | -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                      | H              | -NH-CO-C <sub>4</sub> H <sub>9</sub> (n) | 119-120   |
| 1.24         | H                                  |                                                     | H              | -NH-CO-C <sub>3</sub> H <sub>7</sub> (n) | 175-176   |
| 1.25         | H                                  |                                                     | H              | -NH-CO-C <sub>4</sub> H <sub>9</sub> (n) | 187-188   |
| 1.26         | -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                      | H              | -NH-CO-CF <sub>3</sub>                   | 190-191   |

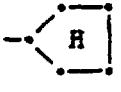
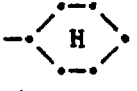
| Compound No. | R <sub>1</sub>                       | R <sub>2</sub>                                                                                         | R <sub>3</sub>      | R <sub>4</sub>                                                                       | m.p. [°C] |
|--------------|--------------------------------------|--------------------------------------------------------------------------------------------------------|---------------------|--------------------------------------------------------------------------------------|-----------|
| 1.27         | H                                    |                       | H                   | -NH-CO-CF <sub>3</sub>                                                               | 268       |
| 1.28         | -CH <sub>3</sub>                     | -C <sub>4</sub> H <sub>9</sub> (n)                                                                     | H                   | -NH-CO-CH(CH <sub>3</sub> ) <sub>2</sub>                                             | 139-140   |
| 1.29         | -CH <sub>3</sub>                     | -C <sub>2</sub> H <sub>5</sub>                                                                         | H                   | -NH-CO-CH(CH <sub>3</sub> ) <sub>2</sub>                                             | 146-149   |
| 1.30         | H                                    |                       | H                   | -NH-CO-CH(C <sub>2</sub> H <sub>5</sub> ) <sub>2</sub>                               | 196-197   |
| 1.31         | -C <sub>2</sub> H <sub>5</sub>       | -C <sub>2</sub> H <sub>5</sub>                                                                         | H                   | -NH-CO-(CH <sub>2</sub> ) <sub>4</sub> CH <sub>3</sub>                               | 133-135   |
| 1.32         | H                                    |                       | H                   | -NH-CO-(CH <sub>2</sub> ) <sub>4</sub> CH <sub>3</sub>                               | 170-173   |
| 1.33         | H                                    |                      | H                   |   | 192-195   |
| 1.34         | -C <sub>2</sub> H <sub>5</sub>       | -C <sub>2</sub> H <sub>5</sub>                                                                         | H                   |  | 160-165   |
| 1.35         | H                                    |                     | H                   | -N=CH-NH-SO <sub>2</sub> CH <sub>3</sub>                                             | 203-205   |
| 1.36         | -C <sub>2</sub> H <sub>5</sub>       | -C <sub>2</sub> H <sub>5</sub>                                                                         | H                   | -N=CH-NH-SO <sub>2</sub> CH <sub>3</sub>                                             | 182-185   |
| 1.37         | -C <sub>2</sub> H <sub>5</sub>       | -C <sub>2</sub> H <sub>5</sub>                                                                         | H                   |  | 127-128   |
| 1.38         | -CH <sub>2</sub> -CH=CH <sub>2</sub> | -CH <sub>3</sub>                                                                                       | H                   | -NH <sub>2</sub>                                                                     | 199-201   |
| 1.39         | -CH <sub>2</sub> -CH=CH <sub>2</sub> | H                                                                                                      | H                   | -NH <sub>2</sub>                                                                     | 191-193   |
| 1.40         | -CH <sub>2</sub> -C≡CH               | H                                                                                                      | H                   | -NH <sub>2</sub>                                                                     | >260      |
| 1.41         | H                                    | -CH <sub>2</sub> -  | H                   | -NH <sub>2</sub>                                                                     | 228-229   |
| 1.42         | H                                    |                     | -CO-CH <sub>3</sub> | -NH-CO-CH <sub>3</sub>                                                               | ~ 260     |
| 1.43         | -C <sub>2</sub> H <sub>5</sub>       | -C <sub>2</sub> H <sub>5</sub>                                                                         | -CO-CH <sub>3</sub> | -NH-CO-CH <sub>3</sub>                                                               | 263-265   |

The following salts of compounds of the formula I are prepared by reaction with the respective acids indicated:

| Compound No. | R <sub>1</sub>                 | R <sub>2</sub>                                                                      | NHR <sub>3</sub> | R <sub>4</sub>                           | Acid                              | m.p. [°C]  |
|--------------|--------------------------------|-------------------------------------------------------------------------------------|------------------|------------------------------------------|-----------------------------------|------------|
| 2.1          | H                              |    | -NH <sub>2</sub> | -NH <sub>2</sub>                         | HCl                               | 210 (dec.) |
| 2.2          | H                              |    | -NH <sub>2</sub> | -NH <sub>2</sub>                         | F <sub>3</sub> C-COOH             | 209-210    |
| 2.3          | H                              |    | -NH <sub>2</sub> | -NH <sub>2</sub>                         | (COOH) <sub>2</sub>               | 198 (dec.) |
| 2.4          | -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                      | -NH <sub>2</sub> | -NH <sub>2</sub>                         | HCl                               | 167-169    |
| 2.5          | -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                      | -NH <sub>2</sub> | -NH <sub>2</sub>                         | F <sub>3</sub> C-COOH             | 175-178    |
| 2.6          | -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                      | -NH <sub>2</sub> | -NH <sub>2</sub>                         | (COOH) <sub>2</sub>               | 178-188    |
| 2.7          | H                              |  | -NH <sub>2</sub> | -NH-CO-CH(CH <sub>3</sub> ) <sub>2</sub> | HCl                               | ~140       |
| 2.8          | H                              |  | -NH <sub>2</sub> | -NH-CO-CH(CH <sub>3</sub> ) <sub>2</sub> | CF <sub>3</sub> COOH              | 150-153    |
| 2.9          | H                              |  | -NH <sub>2</sub> | -NH <sub>2</sub>                         | H <sub>3</sub> PO <sub>4</sub>    | 208        |
| 2.10         | H                              |  | -NH <sub>2</sub> | -NH <sub>2</sub>                         | H <sub>2</sub> SO <sub>4</sub>    | 188        |
| 2.11         | -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                      | -NH <sub>2</sub> | -NH <sub>2</sub>                         | H <sub>2</sub> SO <sub>4</sub>    | 199-201    |
| 2.12         | -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                      | -NH <sub>2</sub> | -NH <sub>2</sub>                         | H <sub>3</sub> PO <sub>4</sub>    | 185        |
| 2.13         | -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                      | -NH <sub>2</sub> | -NH <sub>2</sub>                         | CH <sub>3</sub> SO <sub>3</sub> H | 205-207    |
| 2.14         | H                              |  | -NH <sub>2</sub> | -NH <sub>2</sub>                         | CH <sub>3</sub> SO <sub>3</sub> H | 250        |
| 2.15         | -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                      | -NH <sub>2</sub> | -NH <sub>2</sub>                         | maleic acid                       | 150        |
| 2.16         | H                              |  | -NH <sub>2</sub> | -NH <sub>2</sub>                         | maleic acid                       | 185        |
| 2.17         | -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                      | -NH <sub>2</sub> | -NH <sub>2</sub>                         | malonic acid                      | 216-220    |

| Compound No. | R <sub>1</sub>                 | R <sub>2</sub>                                                                    | NHR <sub>3</sub> | R <sub>4</sub>   | Acid             | m.p. [°C] |
|--------------|--------------------------------|-----------------------------------------------------------------------------------|------------------|------------------|------------------|-----------|
| 2.18         | H                              |  | -NH <sub>2</sub> | -NH <sub>2</sub> | HNO <sub>3</sub> | 163       |
| 2.19         | -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                    | -NH <sub>2</sub> | -NH <sub>2</sub> | HNO <sub>3</sub> | 179       |
| 2.20         | H                              |  | -NH <sub>2</sub> | -NH <sub>2</sub> | malonic acid     | >230      |

The following compounds of the formula I can also be prepared as described above:

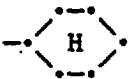
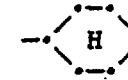
| R <sub>1</sub>                     | R <sub>2</sub>                                                                      | R <sub>3</sub>                        | R <sub>4</sub>                                                                      |
|------------------------------------|-------------------------------------------------------------------------------------|---------------------------------------|-------------------------------------------------------------------------------------|
| H                                  | -CH <sub>3</sub>                                                                    | H                                     | -NH <sub>2</sub>                                                                    |
| H                                  | -C <sub>2</sub> H <sub>5</sub>                                                      | H                                     | -NH <sub>2</sub>                                                                    |
| H                                  | -C <sub>3</sub> H <sub>7</sub> (n)                                                  | H                                     | -NH <sub>2</sub>                                                                    |
| H                                  |    | H                                     | -NH <sub>2</sub>                                                                    |
| H                                  |    | H                                     | -NH <sub>2</sub>                                                                    |
| H                                  |    | H                                     | -NH <sub>2</sub>                                                                    |
| H                                  | -CH(C <sub>2</sub> H <sub>5</sub> ) <sub>2</sub>                                    | H                                     | -NH <sub>2</sub>                                                                    |
| -C <sub>3</sub> H <sub>7</sub> (i) | -C <sub>3</sub> H <sub>7</sub> (i)                                                  | H                                     | -NH <sub>2</sub>                                                                    |
| H                                  |  | H                                     | -N=CH-NH-P(=O)(OC <sub>2</sub> H <sub>5</sub> )(SC <sub>3</sub> H <sub>7</sub> (n)) |
| H                                  |  | H                                     | -NH-CO-O-C <sub>2</sub> H <sub>5</sub>                                              |
| -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                                                      | H                                     | -NH-CO-O-C <sub>2</sub> H <sub>5</sub>                                              |
| -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                                                      | H                                     | -NH-CO-CH <sub>2</sub> Cl                                                           |
| H                                  |  | H                                     | -NH-CO-C <sub>2</sub> F <sub>5</sub>                                                |
| -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                                                      | H                                     | -NH-CO-C <sub>2</sub> F <sub>5</sub>                                                |
| H                                  |  | H                                     | -NH-CO-C <sub>3</sub> F <sub>7</sub> (n)                                            |
| -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                                                      | H                                     | -NH-CO-C <sub>3</sub> F <sub>7</sub> (n)                                            |
| H                                  |  | -CO-C <sub>2</sub> H <sub>5</sub>     | -NH-CO-C <sub>2</sub> H <sub>5</sub>                                                |
| H                                  |  | -CO-C <sub>3</sub> H <sub>7</sub> (n) | -NH-CO-C <sub>3</sub> H <sub>7</sub> (n)                                            |
| H                                  |  | -CO-CH(CH <sub>3</sub> ) <sub>2</sub> | -NH-CO-CH(CH <sub>3</sub> ) <sub>2</sub>                                            |

| R <sub>1</sub>                 | R <sub>2</sub>                                                                    | R <sub>3</sub>                        | R <sub>4</sub>                           |
|--------------------------------|-----------------------------------------------------------------------------------|---------------------------------------|------------------------------------------|
| -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                    | -CO-C <sub>2</sub> H <sub>5</sub>     | -NH-CO-C <sub>2</sub> H <sub>5</sub>     |
| -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                    | -CO-C <sub>3</sub> H <sub>7</sub> (n) | -NH-CO-C <sub>3</sub> H <sub>7</sub> (n) |
| -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                    | -CO-CH(CH <sub>3</sub> ) <sub>2</sub> | -NH-CO-CH(CH <sub>3</sub> ) <sub>2</sub> |
| H                              |  | -CO-CF <sub>3</sub>                   | -NH-CO-CF <sub>3</sub>                   |
| -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                    | -CO-CF <sub>3</sub>                   | -NH-CO-CF <sub>3</sub>                   |
| H                              |  | -CO-CH <sub>3</sub>                   | -NH-CO-CH(CH <sub>3</sub> ) <sub>2</sub> |
| -C <sub>2</sub> H <sub>5</sub> | -C <sub>2</sub> H <sub>5</sub>                                                    | -CO-C <sub>2</sub> H <sub>5</sub>     | -NH-CO-CH(CH <sub>3</sub> ) <sub>2</sub> |

The following intermediates of the formula II are prepared as described above:

| Compound No. | R <sub>1</sub>                     | R <sub>2</sub>                                                                      | R <sub>3</sub> | n | m.p. [°C]   |
|--------------|------------------------------------|-------------------------------------------------------------------------------------|----------------|---|-------------|
| 3.3          | H                                  | -CH <sub>3</sub>                                                                    | H              | 0 | 270-274     |
| 3.4          | H                                  | -C <sub>3</sub> H <sub>7</sub> (i)                                                  | H              | 0 | 171-173     |
| 3.5          |                                    | -(CH <sub>2</sub> ) <sub>4</sub> -                                                  | H              | 0 | 198-200     |
| 3.6          |                                    | -(CH <sub>2</sub> ) <sub>5</sub> -                                                  | H              | 0 | 204-206     |
| 3.7          | H                                  | -C(CH <sub>3</sub> ) <sub>3</sub>                                                   | H              | 0 | 194-196     |
| 3.8          | H                                  | -CH(CH <sub>3</sub> )-C <sub>2</sub> H <sub>5</sub>                                 | H              | 0 | 143-145     |
| 3.9          | H                                  | -CH(C <sub>2</sub> H <sub>5</sub> ) <sub>2</sub>                                    | H              | 0 | 151-153     |
| 3.10         | -CH <sub>3</sub>                   | -CH <sub>3</sub>                                                                    | H              | 0 | 216-218     |
| 3.11         | -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                                                      | H              | 0 | 131         |
| 3.12         | -CH <sub>3</sub>                   | -C <sub>2</sub> H <sub>5</sub>                                                      | H              | 0 | 158-160     |
| 3.13         | -CH <sub>3</sub>                   | -C <sub>4</sub> H <sub>9</sub> (n)                                                  | H              | 0 | 158-160     |
| 3.14         | -C <sub>3</sub> H <sub>7</sub> (n) | -C <sub>3</sub> H <sub>7</sub> (n)                                                  | H              | 0 | 160-162     |
| 3.15         | -C <sub>4</sub> H <sub>9</sub> (n) | -C <sub>4</sub> H <sub>9</sub> (n)                                                  | H              | 0 | 120-122     |
| 3.16         | H                                  | H                                                                                   | H              | 0 | ~270 (dec.) |
| 3.17         | H                                  |  | H              | 2 | 167-170     |
| 3.18         | -CH <sub>3</sub>                   | -CH <sub>3</sub>                                                                    | H              | 2 | 237-239     |
| 3.19         | -C <sub>3</sub> H <sub>7</sub> (n) | -C <sub>3</sub> H <sub>7</sub> (n)                                                  | H              | 2 | 222-225     |
| 3.20         | -C <sub>4</sub> H <sub>9</sub> (n) | -C <sub>4</sub> H <sub>9</sub> (n)                                                  | H              | 2 | 165-167     |
| 3.21         | -CH <sub>3</sub>                   | -C <sub>2</sub> H <sub>5</sub>                                                      | H              | 2 | 203-205     |
| 3.22         |                                    | -(CH <sub>2</sub> ) <sub>4</sub> -                                                  | H              | 2 | 235-237     |
| 3.23         |                                    | -(CH <sub>2</sub> ) <sub>5</sub> -                                                  | H              | 2 | 204-207     |
| 3.24         | -CH <sub>3</sub>                   | -C <sub>4</sub> H <sub>9</sub> (n)                                                  | H              | 2 | 190-191     |

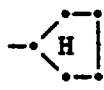
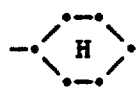
The following intermediates of the formula II can also be prepared as described above:

| R <sub>1</sub>                     | R <sub>2</sub>                                                                      | R <sub>3</sub> | n |
|------------------------------------|-------------------------------------------------------------------------------------|----------------|---|
| H                                  | -C <sub>2</sub> H <sub>5</sub>                                                      | H              | 0 |
| H                                  | -C <sub>3</sub> H <sub>7</sub> (n)                                                  | H              | 0 |
| H                                  | -C <sub>3</sub> H <sub>7</sub> (i)                                                  | H              | 0 |
| H                                  | -CH(C <sub>2</sub> H <sub>5</sub> ) <sub>2</sub>                                    | H              | 0 |
| H                                  |    | H              | 0 |
| H                                  |    | H              | 0 |
| H                                  |    | H              | 0 |
| H                                  | -CH(CH <sub>3</sub> )-C <sub>2</sub> H <sub>5</sub>                                 | H              | 0 |
| -CH <sub>3</sub>                   | -CH <sub>3</sub>                                                                    | H              | 0 |
| -C <sub>3</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                                                      | H              | 0 |
| H                                  | -CH <sub>3</sub>                                                                    | H              | 2 |
| H                                  | -C <sub>2</sub> H <sub>5</sub>                                                      | H              | 2 |
| H                                  | -C <sub>3</sub> H <sub>7</sub> (n)                                                  | H              | 2 |
| -C <sub>3</sub> H <sub>7</sub> (i) | -C <sub>3</sub> H <sub>7</sub> (i)                                                  | H              | 2 |
| H                                  |  | H              | 2 |
| H                                  |  | H              | 2 |
| H                                  |  | H              | 2 |
| H                                  | -CH(CH <sub>3</sub> )-C <sub>2</sub> H <sub>5</sub>                                 | H              | 2 |
| H                                  | -CH(C <sub>2</sub> H <sub>5</sub> ) <sub>2</sub>                                    | H              | 2 |

The following intermediates of the formula XI are prepared as described above:

| Compound No. | R <sub>1</sub>                     | R <sub>2</sub>                                                                     | m.p. [°C]     |
|--------------|------------------------------------|------------------------------------------------------------------------------------|---------------|
| 4.1          | H                                  | -CH <sub>3</sub>                                                                   | 270-274       |
| 4.2          | H                                  | -C <sub>3</sub> H <sub>7</sub> (i)                                                 | 171-173       |
| 4.3          | H                                  |  | 215-218       |
| 4.4          | H                                  | -CH(CH <sub>3</sub> )-C <sub>2</sub> H <sub>5</sub>                                | 143-145       |
| 4.5          | H                                  | -CH(C <sub>2</sub> H <sub>5</sub> ) <sub>2</sub>                                   | 151-153       |
| 4.6          | -CH <sub>3</sub>                   | -CH <sub>3</sub>                                                                   | 216-218       |
| 4.7          | -C <sub>2</sub> H <sub>5</sub>     | -C <sub>2</sub> H <sub>5</sub>                                                     | 131           |
| 4.8          | -C <sub>3</sub> H <sub>7</sub> (n) | -C <sub>3</sub> H <sub>7</sub> (n)                                                 | 160-162       |
| 4.9          | -C <sub>4</sub> H <sub>9</sub> (n) | -C <sub>4</sub> H <sub>9</sub> (n)                                                 | 120-122       |
| 4.10         | H                                  | H                                                                                  | 270<br>(dec.) |
| 4.11         | -CH <sub>3</sub>                   | -C <sub>2</sub> H <sub>5</sub>                                                     | 158-160       |
| 4.12         | H                                  | -C(CH <sub>3</sub> ) <sub>3</sub>                                                  | 194-196       |
| 4.13         | -(CH <sub>2</sub> ) <sub>4</sub> - |                                                                                    | 198-200       |
| 4.14         | -(CH <sub>2</sub> ) <sub>5</sub> - |                                                                                    | 204-206       |
| 4.15         | -CH <sub>3</sub>                   | -C <sub>4</sub> H <sub>9</sub> (n)                                                 | 158-160       |

The following intermediates of the formula XI can also be prepared as described above:

| R <sub>1</sub>                     | R <sub>2</sub>                                                                        |
|------------------------------------|---------------------------------------------------------------------------------------|
| H                                  | -C <sub>2</sub> H <sub>5</sub>                                                        |
| H                                  | -C <sub>3</sub> H <sub>7</sub> (n)                                                    |
| H                                  |  |
| H                                  |  |
| H                                  |  |
| -C <sub>3</sub> H <sub>7</sub> (i) | -C <sub>3</sub> H <sub>7</sub> (i)                                                    |

**Example 6:**

**Formulations for active compounds according to Examples 1 to 3 or combinations of these active compounds with other insecticides or acaricides (% - percent by weight)**

| <b>1. Wettable powders</b>                                             | <b>a)</b> | <b>b)</b> | <b>c)</b> |
|------------------------------------------------------------------------|-----------|-----------|-----------|
| active compounds or combination of                                     |           |           |           |
| active compounds                                                       | 25 %      | 50 %      | 75 %      |
| Na lignosulfonate                                                      | 5 %       | 5 %       | -         |
| Na lauryl sulfate                                                      | 3 %       | -         | 5 %       |
| Na diisobutyl-naphthalenesulfonate                                     | -         | 6 %       | 10 %      |
| octylphenol polyethylene glycol ether<br>(7-8 moles of ethylene oxide) | -         | 2 %       | -         |
| highly disperse silica                                                 | 5 %       | 10 %      | 10 %      |
| kaolin                                                                 | 62 %      | 27 %      | -         |

The active compound or combination of active compounds is thoroughly mixed with the adjuvants and the mixture is thoroughly ground in a suitable mill. Wettable powders are obtained which can be diluted with water to give suspensions of any desired concentration.

| <b>2. Emulsifiable concentrate</b>                                     |      |
|------------------------------------------------------------------------|------|
| active compound or combination of active compounds                     | 10 % |
| octylphenol polyethylene glycol ether<br>(4-5 moles of ethylene oxide) | 3 %  |
| Ca dodecylbenzenesulfonate                                             | 3 %  |
| castor oil polyglycol ether<br>(36 moles of ethylene oxide)            | 4 %  |
| cyclohexanone                                                          | 30 % |
| xylene mixture                                                         | 50 % |

Emulsions of any desired concentration can be obtained from this concentrate by dilution with water.

3. Dusts

active compound or combination of

active compounds

talcum

kaolin

a)

b)

5 %

8 %

95 %

-

-

92 %

Ready-to-use dusts are obtained by mixing the active compound or combination of active compounds with the carrier, and grinding the mixture in a suitable mill.

|                                                     |      |
|-----------------------------------------------------|------|
| 4. Extruder granules                                |      |
| active compounds or combination of active compounds | 10 % |
| Na lignosulfonate                                   | 2 %  |
| carboxymethylcellulose                              | 1 %  |
| kaolin                                              | 87 % |

The active compound or combination of active compounds is mixed with the adjuvants, ground and moistened with water. This mixture is extruded, granulated and then dried in a stream of air.

|                                                    |      |
|----------------------------------------------------|------|
| 5. Coated granules                                 |      |
| active compound or combination of active compounds | 3 %  |
| polyethylene glycol (mol. mass 200)                | 3 %  |
| kaolin                                             | 94 % |

The finely ground active compound or combination of active compounds is uniformly applied, in a mixer, to the kaolin moistened with polyethylene glycol. Non-dusty coated granules are obtained in this manner.

| 6. Suspension concentrate                                             |       |
|-----------------------------------------------------------------------|-------|
| active compound or combination of active compounds                    | 40 %  |
| ethylene glycol                                                       | 10 %  |
| nonylphenol polyethylene glycol ether<br>(15 moles of ethylene oxide) | 6 %   |
| Na lignosulfonate                                                     | 10 %  |
| carboxymethylcellulose                                                | 1 %   |
| 37 % aqueous formaldehyde solution                                    | 0.2 % |
| silicone oil in the form of a 75 % aqueous emulsion                   | 0.8 % |
| water                                                                 | 32 %  |

The finely ground active compound or combination of active compounds is intimately mixed with the adjuvants. A suspension concentrate is obtained from which suspensions of any desired concentration can be obtained by dilution with water.

7. Pour-on solution

|                                  |         |
|----------------------------------|---------|
| active substance                 | 30.00 g |
| sodium dioctyl sulfosuccinate    | 3.00 g  |
| benzyl alcohol                   | 35.46 g |
| ethylene glycol monomethyl ether | 35.46 g |
| <hr/>                            |         |
| 103.92 g = 100 ml                |         |

With vigorous stirring, the active substance is dissolved in the bulk of the mixture of the two solvents. The sodium dioctyl sulfosuccinate is subsequently dissolved in the resultant solution, with heating if necessary, and the solution then made up with the rest of the solvent mixture.

Example 7: Action against Musca domestica:

50 g of freshly prepared nutrient substrate for maggots are weighed into each of a number of beakers. A specific amount of an acetonetic solution containing 1 % by weight of the respective active compound is pipetted onto the nutrient substrate present in the beakers to give an active compound concentration of 800 ppm. After thoroughly mixing the substrate

the acetone is subsequently allowed to evaporate over a period of at least 20 hours.

Then for each active compound and concentration 25 one-day-old maggots of *Musca domestica* are put into each of the beakers containing the treated nutrient substrate. After the maggots have pupated, the pupae formed are separated from the substrate by flushing them out with water and deposited in containers closed with a perforated top. The pupae flushed out of each batch are counted (toxic influence of the active compound on maggot development). A count is then made after 10 days of the number of flies which have hatched out of the pupae.

Compounds of the formulae I and II according to Examples 1 to 3 exhibit good action in this test.

Example 8: Action against *Lucilia sericata*:

1 ml of an aqueous formulation containing 0.5 % by weight of active substance is added at 50°C to 9 ml of a culture medium. Then about 30 freshly hatched *Lucilia sericata* larvae are added to the culture medium, and the insecticidal action is determined after 48 and 96 hours by evaluating the mortality rate.

In this test, compounds of the formulae I and II according to Examples 1 to 3 exhibit good action against *Lucilia sericata*.

Example 9: Action against *Lucilia cuprina*

Freshly deposited eggs of the blowfly *L. cuprina* are put in small portions (30-50 eggs) into test tubes, in which 4 ml of nutrient medium have previously been mixed with 1 ml of active compound solution. After inoculation of the culture medium, the test tubes are closed with cotton-wool plugs and are then incubated in an incubator at 30°C for 4 days.

In the untreated medium serving as control, larvae about 1 cm in length (stage 3) have developed by this time. When a substance is active, by the end of this period the larvae are either dead or moribund. The test is carried out at an active compound concentration of 400 ppm.

Repellency is also taken into account, if the larvae migrate from the medium and starve to death.

In this test, compounds of the formulae I and II according to Examples 1 to 3 exhibit good action against *Lucilia cuprina*.

Example 10: Action against *Aedes aegypti*:

A concentration of 800 ppm is obtained by pipetting a sufficient amount of a 0.1 % by weight solution of the test compound in acetone onto the surface of 150 ml of water in a container. After the acetone has evaporated, 30 to 40 two-day-old *Aedes* larvae are put into the container. Mortality counts are made after 1, 2 and 5 days.

In this test, compounds of the formulae I and II according to Examples 1 to 3 exhibit good action against *Aedes aegypti*.

Example 11: Insecticidal stomach poison action

Cotton plants about 25 cm high, in pots, are sprayed with aqueous emulsions which contain the active compound in concentrations of 100, 200 and 400 ppm.

After the spray coating has dried, the cotton plants are populated with *Spodoptera littoralis* and *Heliothis virescens* larvae in the third larval stage. The test is carried out at 24°C and 60 % relative humidity. The percentage mortality of the larvae is determined after 2 days.

Compounds No. 1.1 and 1.2 according to the invention exhibit an action of (mortality) 80-100 % against *Spodoptera* larvae at 200 and 400 ppm respectively. Compounds No. 1.1 and 1.2 exhibit 80-100 % action (mortality) against *Heliothis* larvae at 100 ppm and 400 ppm respectively.

Example 12: Action against *Anthonomus grandis* (adults)

Two cotton plants in the 6-leaf stage, in pots, are each sprayed with a wettable aqueous emulsion formulation containing 400 ppm of the active compound to be tested. After the spray coating has dried (after about 1.5 hours), each plant is populated with 10 adult beetles (*Anthonomus*

grandis). Plastic cylinders, covered at the top with gauze, are then slipped over the treated plants populated with the test animals to prevent the beetles from migrating. The treated plants are then kept at 25°C and about 60 % relative humidity. Evaluation is made after 2, 3, 4 and 5 days to determine the percentage mortality of the test animals used (in dorsal position) as well as the anti-feeding action as compared with untreated control batches.

Compounds of the formulae I and II according to Examples 1 to 3 exhibit good action in the above test.

Example 13: Action against ticks

A) Amblyomma hebraeum

50 nymphs are counted into a test tube and immersed for 1 to 2 minutes in 2 ml of an aqueous emulsion containing 400 ppm of test substance. The test tube is then closed with a standard cotton wool plug and placed on its head to enable the cotton wool to absorb the active-substance emulsion.

Evaluation is made 1 week later. Two replicates are carried out for each test.

B) Boophilus microplus (larvae)

Tests are carried out with 20 OP-sensitive and 20 OP-resistant larvae using a dilution series similar to that used in Test A). (The resistance refers to the tolerance towards diazinone).

In this test, compounds of the formulae I and II according to Examples 1 to 3 exhibit good action against nymphs and larvae of the ticks *Amblyomma hebraeum* and *Boophilus microplus*.

Example 14: Insecticidal action: Nilaparvata lugens

Rice plants are sprayed with a test solution containing 400 ppm of the compound to be tested. After the coating has dried, the plants are populated with nymphs of *Nilaparvata lugens* (N<sub>2</sub> or N<sub>3</sub> stage). Two plants are used per test compound and per test species. Evaluation of the mortality rate achieved is made 6 days later. The test is carried out at

26°C and 60 % relative humidity.

In the above test, compound No. 1.1 exhibits 80-100 % action against nymphs of *Nilaparvata lugens*.

Example 15: Action against soil insects (*Diabrotica balteata*)

5 maize seedlings 1 to 3 cm in height and a filter paper disc are immersed in an aqueous solution of the active compound containing about 4 % by volume of acetone. The immersed filter paper disc is placed on the bottom of a plastic beaker (200 ml capacity) and a dry filter paper disc together with the maize seedlings and 10 *Diabrotica balteata* larvae in the second or third larval stage are then placed on it. The batch is kept at about 24°C and at 40-60 % relative humidity and in daylight. Evaluation is made 6 days later in comparison with untreated control batches.

In this test, compound No. 1.15 exhibits an action of 80-100 % (mortality) at 400 ppm.

Example 16: Action against *Nephotettix cincticeps* (nymphs)

The test is carried out with growing plants. For this purpose rice plants about 20 days old and about 15 cm in height are planted in pots (diameter: 5.5 cm).

The plants are sprayed on a turntable with 100 ml in each case of an acetic solution containing 400 ppm of the active compound to be tested. After the spray coating has dried, each plant is populated with 20 nymphs of the test animals in the second or third stage. To prevent the cicadas from escaping, a plexiglass cylinder is slipped over each of the populated plants and this is covered over with a gauze lid. The nymphs are kept for 5 days on the treated plants, which have to be resprayed at least once. The test is carried out at a temperature of about 23°C and at 55 % relative humidity and with a period of illumination of 16 hours.

In this test, compound No. 1.1 according to the invention and according to Example 1 exhibits 80-100 % action.

Example 17: Insecticidal stomach poison action against *Plutella xylostella*:

Potted Chinese cabbage plants (pot size: 10 cm diameter) in the 4-leaf stage are sprayed with aqueous emulsions of the active compound which contain the active compound in concentrations of 3 to 400 ppm and which dry on the plants.

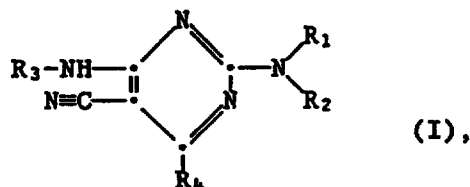
After 2 days, each treated Chinese cabbage plant is populated with 10 *Platella* [sic] *xylostella* larvae in the second larval stage. The test is carried out at 24°C and at 60 % relative humidity in dim light. After 2 and 5 days evaluation is made to determine the percentage mortality of the larvae.

Compounds of the formula I according to Examples 1 to 4 exhibit an action of 80-100 % (morality) in this test.



## Patent Claims

1. A stable compound of the formula



wherein

$\text{R}_1$  is hydrogen,  $\text{C}_1\text{-C}_6$ alkyl,  $\text{C}_2\text{-C}_6$ alkenyl or  $\text{C}_2\text{-C}_6$ alkynyl,

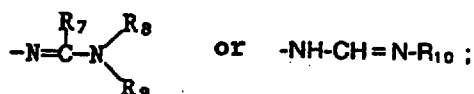
$\text{R}_2$  is hydrogen,  $\text{C}_1\text{-C}_{10}$ alkyl or  $\text{C}_3\text{-C}_6$ cycloalkyl, or

$\text{R}_1$  and  $\text{R}_2$ , when taken together, are one of the radicals  $-(\text{CH}_2)_3-$ ,  $-(\text{CH}_2)_4-$  or  $-(\text{CH}_2)_5-$ ,

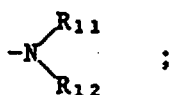
$\text{R}_3$  is hydrogen or one of the radicals  $-\text{CO-R}_5$  or  $-\text{SO}_2\text{-R}_6$ ,

$\text{R}_4$  is one of the radicals

$-\text{NH}_2$ ,  $-\text{NH-CO-R}_5$ ,  $-\text{NH-SO}_2\text{-R}_6$ ,



$\text{R}_5$  is hydrogen,  $\text{C}_1\text{-C}_{12}$ alkyl,  $\text{C}_3\text{-C}_6$ cycloalkyl,  $\text{C}_1\text{-C}_6$ alkoxy, perhalogenated  $\text{C}_1\text{-C}_3$ alkyl or the radical

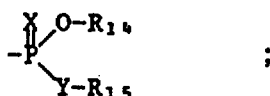


$\text{R}_6$  is  $\text{C}_1\text{-C}_6$ alkyl,

$\text{R}_7$  is hydrogen or  $\text{C}_1\text{-C}_6$ alkyl,

$\text{R}_8$  and  $\text{R}_9$  are each independently of the other hydrogen or  $\text{C}_1\text{-C}_6$ alkyl or, when taken together, are one of the radicals  $-(\text{CH}_2)_3-$ ,  $-(\text{CH}_2)_4-$  or  $-(\text{CH}_2)_5-$ ,

$\text{R}_{10}$  is one of the radicals  $-\text{SO}_2\text{-R}_{13}$  or



$\text{R}_{11}$  and  $\text{R}_{12}$  are each independently of the other hydrogen or  $\text{C}_1\text{-C}_6$ alkyl or, when taken together, are one of the radicals  $-(\text{CH}_2)_3-$ ,  $-(\text{CH}_2)_4-$  or



$-(CH_2)_5-$ ,

$R_{13}$  is  $C_1-C_{10}$ alkyl,  $C_1-C_{10}$ alkyl which is substituted by up to 10 halogen atoms, or is  $C_3-C_6$ cycloalkyl,

$R_{14}$  and  $R_{15}$  are each independently of the other  $C_1-C_{10}$ alkyl; and

X and Y are each independently of the other oxygen or sulfur, and salts thereof.

2. A compound according to claim 1 of the formula I,  
wherein

$R_1$  is hydrogen or  $C_1-C_4$ alkyl,

$R_2$  is  $C_1-C_4$ alkyl or  $C_3-C_6$ cycloalkyl, or

$R_1$  and  $R_2$ , when taken together, are one of the radicals  $-(CH_2)_4-$  or  $-(CH_2)_5-$ ,

$R_3$  is hydrogen,

$R_4$  is one of the radicals  $-NH_2$  or  $-NH-CO-R_5$ , and

$R_5$  is  $C_1-C_4$ alkyl or perhalogenated  $C_1-C_3$ alkyl, and salts thereof.

3. A compound according to claim 2 of the formula I,  
wherein

$R_1$  is hydrogen, methyl or ethyl,

$R_2$  is  $C_1-C_4$ alkyl or cyclopropyl,

$R_3$  is hydrogen,

$R_4$  is one of the radicals  $-NH_2$  or  $-NH-CO-R_5$ , and

$R_5$  is  $C_1-C_3$ alkyl, and salts thereof.

4. A compound according to claim 3 of the formula I,  
wherein

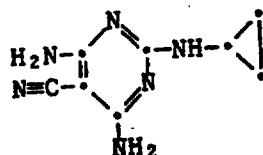
$R_1$  is hydrogen, methyl or ethyl,

$R_2$  is cyclopropyl,

$R_3$  is hydrogen, and

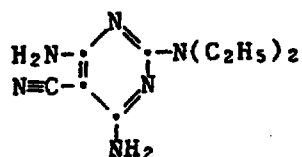
$R_4$  is one of the radicals  $-NH_2$  or  $-NH-CO-C_2-H_5$ , and salts thereof.

5. A compound according to claim 4 of the formula

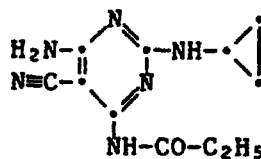




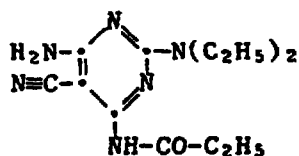
6. A compound according to claim 4 of the formula



7. A compound according to claim 4 of the formula

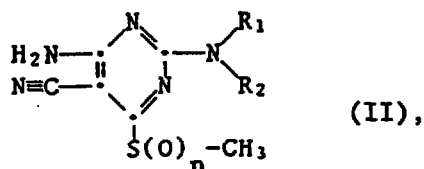


8. A compound according to claim 4 of the formula



9. A process for the preparation of a stable compound according to any one of claims 1 to 8, and salts thereof, which comprises

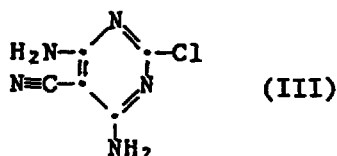
a) reacting a compound of the formula II



with ammonia, or



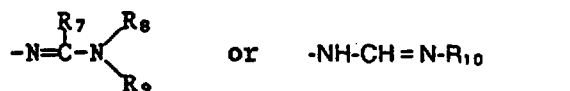
b) the compound of the formula III



with a compound of the formula IV

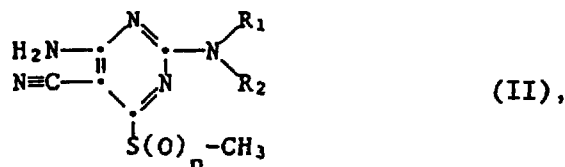


and, if desired, converting the particular resultant compound of the formula I, in which  $R_3$  is hydrogen and  $R_4$  is the radical  $-NH_2$ , in a manner known per se, into a compound of the formula I, in which  $R_3$  has the meanings  $-CO-R_5$  or  $-SO_2-R_6$  and/or  $R_4$  has the meanings



where  $R_1$  to  $R_{15}$ , X and Y are as defined in claim 1 and n is 0, 1 or 2, and, if desired, converting a resultant compound of the formula I into a salt thereof.

10. A stable compound of the formula II



wherein

$R_1$  is hydrogen,  $C_1$ - $C_6$ alkyl,  $C_2$ - $C_6$ alkenyl or  $C_2$ - $C_6$ alkynyl,

$R_2$  is hydrogen,  $C_1$ - $C_{10}$ alkyl or  $C_3$ - $C_6$ cycloalkyl, or

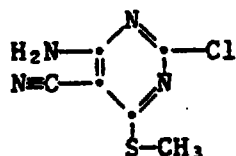
$R_1$  and  $R_2$ , when taken together, are one of the radicals  $-(CH_2)_4-$  or  $-(CH_2)_5-$ , and

n is 0, 1 or 2.

11. A process for the preparation of a compound of the formula III, which comprises oxidising the compound of the formula VII

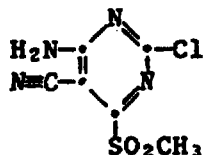


- 39 -



(VII)

with chlorine, to give the compound of the formula XIII



(XIII)

and reacting the compound of the formula XIII with ammonia.

12. A pesticide which contains, as active component, a compound of the formula I or II according to any one of claims 1 to 8 or 10, or a salt thereof, together with suitable carriers and/or other adjuvants.

13. Use of a compound according to any one of claims 1 to 8 or 10, or a salt thereof, for controlling insects and representatives of the order Acarina on animals or plants.

14. Use according to claim 13 for controlling larval stages of plant-destructive insects.

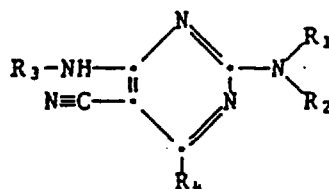
15. Use according to claim 13 for controlling ectoparasites in domestic animals and productive livestock.

16. A method of controlling insects and representatives of the order Acarina, which comprises treating or contacting these pests, their various development stages or the locus thereof with a pesticidally effective amount of a compound of the formula I according to any one of claims 1 to 8 or 10, or of a salt thereof, or with an agent comprising a pesticidally effective amount of this compound, together with adjuvants and carriers.



[As printed]  
Claims

1. A stable compound of the formula



(I),

wherein

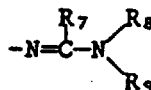
R<sub>1</sub> is hydrogen, C<sub>1</sub>-C<sub>6</sub> alkyl, C<sub>2</sub>-C<sub>6</sub> alkenyl or C<sub>2</sub>-C<sub>6</sub> alkynyl,

R<sub>2</sub> is hydrogen, C<sub>1</sub>-C<sub>10</sub> alkyl or C<sub>3</sub>-C<sub>6</sub> cycloalkyl, or

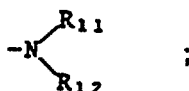
R<sub>1</sub> and R<sub>2</sub>, when taken together, are one of the radicals -(CH<sub>2</sub>)<sub>3</sub>-, -(CH<sub>2</sub>)<sub>4</sub>- or -(CH<sub>2</sub>)<sub>5</sub>-.



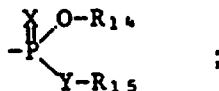
$R_3$  is hydrogen or one of the radicals  $-\text{CO}-R_5$  or  $-\text{SO}_2-R_6$ ,  
 $R_4$  is one of the radicals  
 $-\text{NH}_2$ ,  $-\text{NH}-\text{CO}-R_5$ ,  $-\text{NH}-\text{SO}_2-R_6$ ,



or  $-\text{NH}-\text{CH}=\text{N}-R_{10}$  ;  
 $R_5$  is hydrogen,  $\text{C}_1$ - $\text{C}_{12}$  alkyl,  $\text{C}_3$ - $\text{C}_6$  cycloalkyl,  $\text{C}_1$ - $\text{C}_6$  alkoxy, perhalogenated  $\text{C}_1$ - $\text{C}_3$  alkyl or the radical



$R_6$  is  $\text{C}_1$ - $\text{C}_6$  alkyl,  
 $R_7$  is hydrogen or  $\text{C}_1$ - $\text{C}_6$  alkyl,  
 $R_8$  and  $R_9$  are each independently of the other hydrogen or  $\text{C}_1$ - $\text{C}_6$  alkyl or, when taken together, are one of the radicals  $-(\text{CH}_2)_3-$ ,  $-(\text{CH}_2)_4-$  or  $-(\text{CH}_2)_5-$ ,  
 $R_{10}$  is one of the radicals  $-\text{SO}_2-R_{13}$  or



$R_{11}$  and  $R_{12}$  are each independently of the other hydrogen or  $\text{C}_1$ - $\text{C}_6$  alkyl or, when taken together, are one of the radicals  $-(\text{CH}_2)_3-$ ,  $-(\text{CH}_2)_4-$  or  $-(\text{CH}_2)_5-$ ,  
 $R_{13}$  is  $\text{C}_1$ - $\text{C}_{10}$  alkyl,  $\text{C}_1$ - $\text{C}_{10}$  alkyl which is substituted by up to 10 halogen atoms, or is  $\text{C}_3$ - $\text{C}_6$  cycloalkyl,  
 $R_{14}$  and  $R_{15}$  are each independently of the other  $\text{C}_1$ - $\text{C}_{10}$  alkyl; and  
 $\text{X}$  and  $\text{Y}$  are each independently of the other oxygen or sulfur, and salts thereof.

2. A compound according to claim 1 of the formula I,  
 wherein

$R_1$  is hydrogen or  $\text{C}_1$ - $\text{C}_4$  alkyl,  
 $R_2$  is  $\text{C}_1$ - $\text{C}_4$  alkyl or  $\text{C}_3$ - $\text{C}_6$  cycloalkyl, or  
 $R_1$  and  $R_2$ , when taken together, are one of the radicals  $-(\text{CH}_2)_4-$  or  $-(\text{CH}_2)_5-$ ,  
 $R_3$  is hydrogen,  
 $R_4$  is one of the radicals  $-\text{NH}_2$  or  $-\text{NH}-\text{CO}-R_5$ , and  
 $R_5$  is  $\text{C}_1$ - $\text{C}_4$  alkyl or perhalogenated  $\text{C}_1$ - $\text{C}_3$  alkyl, and salts thereof.

3. A compound according to claim 2 of the formula I,  
 wherein

$R_1$  is hydrogen, methyl or ethyl,  
 $R_2$  is  $\text{C}_1$ - $\text{C}_4$  alkyl or cyclopropyl,  
 $R_3$  is hydrogen,  
 $R_4$  is one of the radicals  $-\text{NH}_2$  or  $-\text{NH}-\text{CO}-R_5$ , and  
 $R_5$  is  $\text{C}_1$ - $\text{C}_3$  alkyl, and salts thereof.

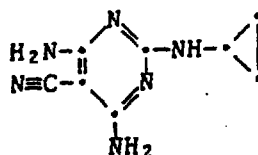
4. A compound according to claim 3 of the formula I,  
 wherein

$R_1$  is hydrogen, methyl or ethyl,

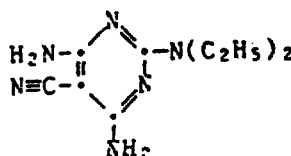


- $R_2$  is cyclopropyl,  
 $R_3$  is hydrogen, and  
 $R_4$  is one of the radicals  $-NH_2$  or  $-NH-CO-C_2H_5$ , and salts thereof.

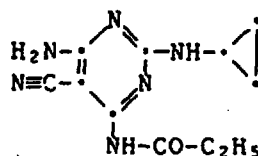
A compound according to claim 4 of the formula



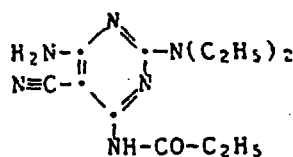
6. A compound according to claim 4 of the formula



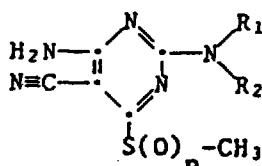
7. A compound according to claim 4 of the formula



8. A compound according to claim 4 of the formula



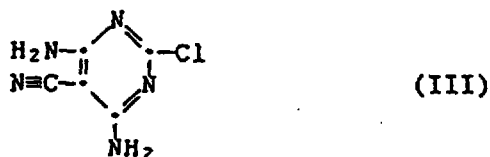
9. A process for the preparation of a stable compound according to any one of claims 1 to 8, and salts thereof, which comprises  
 a) reacting a compound of the formula II



(II),



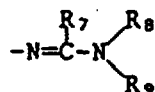
with ammonia, or  
b) the compound of the formula III



with a compound of the formula IV



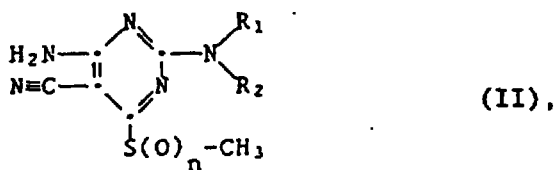
and, if desired, converting the particular resultant compound of the formula I, in which R<sub>3</sub> is hydrogen and R<sub>4</sub> is the radical -NH<sub>2</sub>, in a manner known per se, into a compound of the formula I, in which R<sub>3</sub> has the meanings -CO-R<sub>5</sub> or -SO<sub>2</sub>-R<sub>6</sub> and/or R<sub>4</sub> has the meanings -NH-CO-R<sub>5</sub>, -NH-SO<sub>2</sub>-R<sub>6</sub>,



or -NH-CH=N-R<sub>10</sub>,

where R<sub>1</sub> to R<sub>15</sub>, X and Y are as defined in claim 1 and n is 0, 1 or 2, and, if desired, converting a resultant compound of the formula I into a salt thereof.

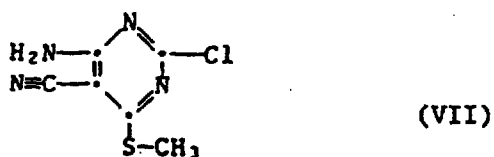
10. A stable compound of the formula II



wherein

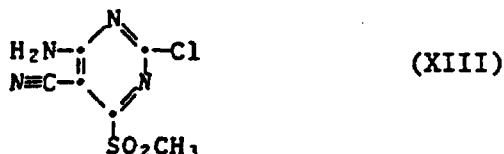
- R<sub>1</sub> is hydrogen, C<sub>1</sub>-C<sub>6</sub> alkyl, C<sub>2</sub>-C<sub>6</sub> alkenyl or C<sub>2</sub>-C<sub>6</sub> alkynyl,
- R<sub>2</sub> is hydrogen, C<sub>1</sub>-C<sub>10</sub> alkyl or C<sub>3</sub>-C<sub>6</sub> cycloalkyl, or
- R<sub>1</sub> and R<sub>2</sub>, when taken together, are one of the radicals -(CH<sub>2</sub>)<sub>4</sub>- or -(CH<sub>2</sub>)<sub>5</sub>-, and
- n is 0, 1 or 2.

11. A process for the preparation of a compound of the formula III, which comprises oxidising the compound of the formula VII





with chlorine, to give the compound of the formula XIII



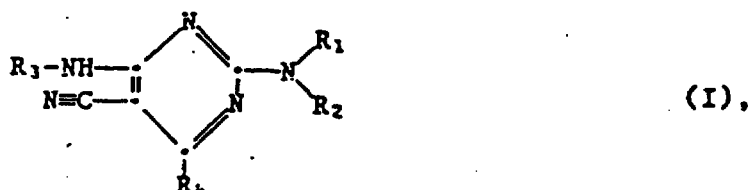
and reacting the compound of the formula XIII with ammonia.

12. A pesticide which contains, as active component, a compound of the formula I or II according to any one of claims 1 to 8 or 10, or a salt thereof, together with suitable carriers and/or other adjuvants.
13. Use of a compound according to any one of claims 1 to 8 or 10, or a salt thereof, for controlling insects and representatives of the order Acarina on animals or plants.
14. Use according to claim 13 for controlling larval stages of plant-destructive insects.
15. Use according to claim 13 for controlling ectoparasites in domestic animals and productive livestock.
16. Method of controlling insects and representatives of the order Acarina, which comprises treating or contacting these pests, their various development stages or the locus thereof with a pesticidally effective amount of a compound of the formula I according to any one of claims 1 to 8 or 10, or of a salt thereof, or with an agent comprising a pesticidally effective amount of this compound, together with adjuvants and carriers.

[As printed]

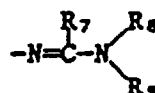
#### Revendications

1. Composé stable de formule



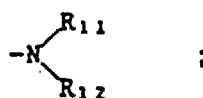
où

- R<sub>1</sub> représente un hydrogène, un alkyle en C<sub>1</sub>-C<sub>6</sub>, un alcényle en C<sub>2</sub>-C<sub>6</sub> ou un alcynyle en C<sub>2</sub>-C<sub>6</sub>;
- R<sub>2</sub> représente un hydrogène, un alkyle en C<sub>1</sub>-C<sub>10</sub> ou un cycloalkyle en C<sub>3</sub>-C<sub>6</sub>; ou
- R<sub>1</sub> et R<sub>2</sub> représentent ensemble un des restes -(CH<sub>2</sub>)<sub>3</sub>-, -(CH<sub>2</sub>)<sub>4</sub>- ou -(CH<sub>2</sub>)<sub>5</sub>;
- R<sub>3</sub> représente un hydrogène ou un des restes -CO-R<sub>5</sub> ou -SO<sub>2</sub>-R<sub>6</sub>,
- R<sub>4</sub> représente un des restes -NH<sub>2</sub>, -NH-CO-R<sub>5</sub>, -HH-SO<sub>2</sub>-R<sub>6</sub>,

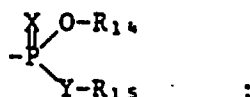


ou -NH-CH=N-R<sub>10</sub>;

- R<sub>5</sub> représente un hydrogène, un alkyle en C<sub>1</sub>-C<sub>12</sub>, un cycloalkyle en C<sub>3</sub>-C<sub>6</sub>, un alcoxy en C<sub>1</sub>-C<sub>6</sub>, un alkyle en C<sub>1</sub>-C<sub>3</sub> perhalogéné ou le reste



- $R_6$  représente un alkyle en  $C_1-C_6$ ;  
 $R_7$  représente un hydrogène ou un alkyle en  $C_1-C_6$ ;  
 $R_8$  et  $R_9$ , indépendamment l'un de l'autre, représentent un hydrogène ou un alkyle en  $C_1-C_6$  ou ensemble un des restes  $-(CH_2)_3-$ ,  $-(CH_2)_4-$  ou  $-(CH_2)_5-$ ;  
 $R_{10}$  représente un des restes  $-SO_2-R_{13}$  ou



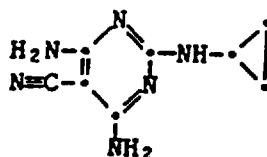
- $R_{11}$  et  $R_{12}$  indépendamment l'un de l'autre représentent un hydrogène ou un alkyle en  $C_1-C_6$  ou ensemble un des restes  $-(CH_2)_3-$ ,  $-(CH_2)_4-$  ou  $-(CH_2)_5-$ ;  
 $R_{13}$  représente un alkyle en  $C_1-C_{10}$ , un alkyle en  $C_1-C_{10}$  substitué avec jusqu'à 10 atomes d'halogène ou un cycloalkyle en  $C_3-C_6$ ;  
 $R_{14}$  et  $R_{15}$  indépendamment l'un de l'autre représentent un alkyle en  $C_1-C_{10}$ ;  
 $X$  et  $Y$  indépendamment l'un de l'autre représentent un oxygène ou un soufre, ainsi que ses sels.

2. Composé selon la revendication 1 de formule I, caractérisé en ce que
- $R_1$  représente un hydrogène ou un alkyle en  $C_1-C_4$ ;  
 $R_2$  représente un alkyle en  $C_1-C_4$  ou un cycloalkyle en  $C_3-C_6$  ou  
 $R_1$  et  $R_2$  ensemble représentent un des restes  $-(CH_2)_4-$  ou  $-(CH_2)_5-$ ;  
 $R_3$  représente un hydrogène;  
 $R_4$  représente un des restes  $-NH_2$  ou  $-NH-CO-R_5$ ; et  
 $R_5$  représente un alkyle en  $C_1-C_4$  ou un alkyle en  $C_1-C_3$  perhalogéné, ainsi que ses sels.

3. Composé selon la revendication 2 de formule I, caractérisé en ce que
- $R_1$  représente un hydrogène, un méthyle ou un éthyle;  
 $R_2$  représente un alkyle en  $C_1-C_4$  ou un cyclopropyle;  
 $R_3$  représente un hydrogène;  
 $R_4$  représente un des restes  $-NH_2$  ou  $-NH-CO-R_5$ ;  
 $R_5$  représente un alkyle en  $C_1-C_3$ , ainsi que ses sels.

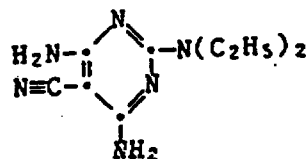
4. Composé selon la revendication 3 de formule I, caractérisé en ce que
- $R_1$  représente un hydrogène, un méthyle ou un éthyle;  
 $R_2$  représente un cyclopropyle;  
 $R_3$  représente un hydrogène; et  
 $R_4$  représente un des restes  $-NH_2$  ou  $-NH-CO-C_2H_5$ , ainsi que ses sels.

5. Composé selon la revendication 4 de formule

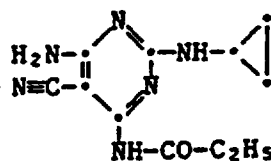




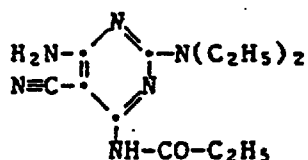
Composé selon la revendication 4 de formule



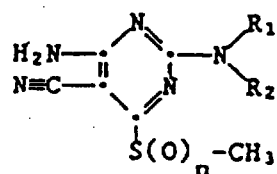
7. Composé selon la revendication 4 de formule



8. Composé selon la revendication 4 de formule

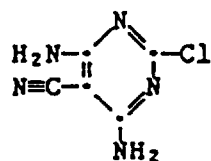


9. Procédé pour préparer un composé stable de formule I selon l'une des revendications 1 à 8, ainsi que ses sels, caractérisé en ce que l'on fait réagir  
a) un composé de formule II



(II),

avec l'ammoniac, ou on fait réagir  
b) le composé de formule III



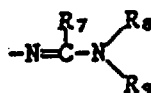
(III)

avec un composé de formule IV



(IV)

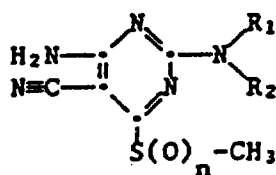
et en ce que l'on transforme le composé de formule I qui se forme à chaque fois, dans lequel  $R_3$  représente un hydrogène et  $R_4$  représente le reste  $-\text{NH}_2$ , éventuellement, de façon connue en soi, en un composé de formule I où  $R_3$  a les significations  $-\text{CO}-R_5$  ou  $-\text{SO}_2-R_6$  et/ou  $R_4$  a les significations  $-\text{NH}-\text{CO}-R_5$ ,  $-\text{NH}-\text{SO}_2-R_6$ ,



ou  $-\text{NH}-\text{CH}=\text{N}-R_{10}$ ,

où  $R_1$  à  $R_{15}$ , X et Y ont les significations données à la revendication 1, et n est un nombre égal à 0, 1 ou 2, et en ce que, si on le désire, on transforme un composé obtenu de formule I en un de ses sels.

#### 10. Composé stable de formule II

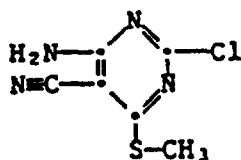


(II),

où

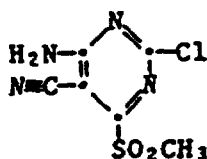
- $R_1$  représente un hydrogène, un alkyle en  $\text{C}_1-\text{C}_6$ , un alcényle en  $\text{C}_2-\text{C}_6$  ou un alcynyle en  $\text{C}_2-\text{C}_6$ ;  
 $R_2$  représente un hydrogène, un alkyle en  $\text{C}_1-\text{C}_{10}$  ou un cycloalkyle en  $\text{C}_3-\text{C}_6$ ; ou  
 $R_1$  et  $R_2$  représentent ensemble un des restes  $-(\text{CH}_2)_4$ - ou  $-(\text{CH}_2)_5$ ;-  
 $n$  représente un nombre égal à 0, 1 ou 2.

#### 11. Procédé pour préparer le composé de formule III, caractérisé en ce que l'on oxyde le composé de formule VII



(VII),

avec le chlore pour obtenir le composé de formule XIII



(XIII)

et en ce que l'on fait réagir le composé de formule XIII avec de l'ammoniac.



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12. Moyen pour lutter contre les parasites, qui contient comme composant actif un composé des formules I ou II conforme à l'une des revendications 1 à 8 ou 10, ou un de ses sels, en même temps que des supports appropriés et/ou d'autres additifs appropriés.
13. Utilisation d'un composé selon l'une des revendications 1 à 8 ou 10, ou un de ses sels, pour lutter contre les insectes et les représentants de l'ordre de l'acare sur les animaux et les plantes.
14. Utilisation selon la revendication 13 pour lutter contre les insectes au stade de larves qui sont nuisibles pour les plantes.
15. Utilisation selon la revendication 13 pour lutter contre les ectoparasites sur les animaux de compagnie et les animaux utiles.
16. Procédé pour lutter contre les insectes et les représentants de l'ordre de l'acare, caractérisé en ce que l'on traite ces parasites, ou leurs divers stades d'évolution ou leurs lieux de séjour, avec une quantité efficace comme pesticide d'un composé de formule I selon l'une des revendications 1 à 8 ou 10, ou d'un de ses sels, ou avec un moyen contenant, en plus des additifs et des supports, une quantité efficace comme pesticide, de ce composé, ou en ce qu'on les met au contact de ladite quantité ou dudit moyen.