1,035,094

PATENT SPECIFICATION

NO DRAWINGS

1,035,094

Date of Application and filing Complete Specification: Dec. 7, 1962.
No. 6193/66.
Application made in United States of America (No.217,521) on Aug. 17, 1962.
Application made in United States of America (No. 221,890) on Sept. 6, 1962.
Application made in United States of America (No. 232,311) on Oct. 22, 1962.
Application made in United States of America (No. 233,952) on Oct. 29, 1962.
(Divided out of No. 1,035,091.)
(Patent of Addition to No. 968,661 dated Aug. 15, 1960.)
Complete Specification Published: July 6, 1966.
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Index at acceptance:—C2 C(2A1, 2A5, 2B3A1, 2B3B, 2B3E, 2B3F, 2B3G1, 2B31, 2B33, 2R15, 2R16, 2R17, 2R18, 2T21, B4A1, B4A2, B4A4, B4M); A5 E(1C2, 1C3B1, 1C3B3, 1C4A2, 1C4A3, 1C4A4, 1C4B2, 1C4B3, 1C4B4, 1C5A, 1C5D, 1C7A, 1C7B, 1C12, 1C14B, 1C14C, 1C15)

Int. Cl.:—C 07 d 51/30 // A 01 n

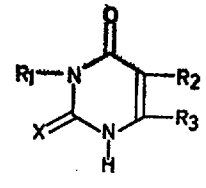
COMPLETE SPECIFICATION

Improvements relating to Herbicides and novel 3-Substituted Uracils

We, E. I. DU PONT DE NEMOURS AND COMPANY, a Corporation organised and existing under the laws of the State of Delaware, United States of America, of Wilmington 98, Delaware, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

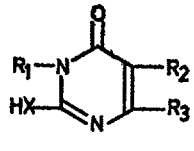
This invention relates to 3-substituted uracils and is an improvement in or modification of the invention described in our Patent No. 968,661.

Our Specification No. 968,661 discloses that certain 3-substituted uracils have herbicidal activity. Those 3-substituted uracils are compounds of the general formula:



I

or the corresponding isomeric formula:



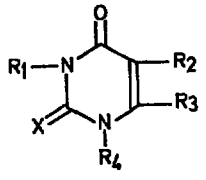
II

wherein R_1 is a covalent organic radical of molecular weight 15 to 250 which is an alkyl group of up to 10 carbon atoms, a substituted alkyl group in which the alkyl group contains up to 3 carbon atoms, a phenyl group, a substituted phenyl group, an aralkyl group of 7 or 8 carbon atoms, a substituted aralkyl group of 7 to 12 carbon atoms, an alkenyl group of 3 to 6 carbon atoms, a propynyl group, a butynyl group, a cycloalkyl group of 5 to 8 carbon atoms, a cycloalkenyl group of 5 to 8 carbon atoms or a cyano group; R_2 is a hydrogen or halogen atom or an alkyl group of up to 4 carbon atoms, an alkoxy group of up to 3 carbon atoms, a nitro group, an alkoxymethyl group of 2 to 4 carbon atoms, a hydroxymethyl group, a hydroxyethyl group, an allyl group, a thiocyanato group, a cyano group, a halogenomethyl group or a methyl thiomethyl group; R_3 is a hydrogen or chlorine atom or an alkyl group of up to 5 carbon atoms, a methoxy group or an ethoxy group; or R_2 and R_3 together form a divalent alkylene bridge of formula $(CH_2)_n$, wherein n is 3, 4 or 5; and X is oxygen or sulphur, preferably oxygen; with the proviso that if R_2 is a hydrogen atom, then either R_3 is an alkyl group of 1 to 5 carbon atoms and R_1 is an alkyl group of 3 to 6 carbon atoms, an allyl group, a propynyl group or a cycloalkyl or cycloalkenyl group of 5 to 8 carbon atoms, or R_3 is a hydrogen atom and R_1 is a cycloalkyl or cycloalkenyl group of 5 to 8 carbon atoms; and metal salts of such uracils.

Within the broad class of 3-substituted uracils defined in Specification No. 968,661 are four particular classes which are the subject

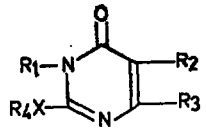
[*Price 4s. 6d.*]

of our Patents Nos. 968,662; 968,663; 968,664 and 968,665, which are patents of addition to Patent No. 968,661. Certain other 3-substituted uracils having herbicidal properties are disclosed in our Patent No. 968,666, which is also a patent of addition to our Patent No. 968,661. These uracils, all of which have a substituent in the 1- or 2-position are compounds of the general formula:



I

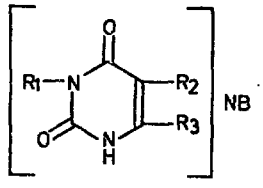
or the corresponding isomeric formula:



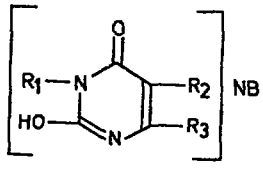
II

wherein R_1 is a covalent organic radical of molecular weight 15 to 250 which is an alkyl group of up to 10 carbon atoms, a substituted alkyl group in which the alkyl group contains up to 3 carbon atoms, a phenyl group, a substituted phenyl group, an aralkyl group of 7 or 8 carbon atoms, a substituted aralkyl group of 7 to 12 carbon atoms, an alkenyl group of 3 to 6 carbon atoms, a propynyl group, a butynyl group, a cycloalkyl group of 5 to 8 carbon atoms, a cycloalkenyl group of 5 to 8 carbon atoms or a cyano group; R_2 is a hydrogen or halogen atom or an alkyl group of up to 4 carbon atoms, an alkoxy group of up to 3 carbon atoms, a nitro group, an alkoxymethyl group of 2 to 4 carbon atoms, a hydroxymethyl group, a hydroxyethyl group, an allyl group, a thiocyanato group, a cyano group, a halogenomethyl group, or a methyl thiomethyl group; R_3 is a hydrogen or chlorine atom or an alkyl group of up to 5 carbon atoms, a methoxy group, or an ethoxy group; or R_2 and R_3 together form a divalent alkylene bridge of formula $(CH_2)_n$, wherein n is 3, 4 or 5; R_4 is a covalent radical of molecular weight up to 250 which is an alkyl group of up to 3 carbon atoms, a hydroxymethyl group, a hydroxyethyl group, a carboxyethyl group, a substituted carbamyl group, a substituted thiocarbamyl group, a diethylphosphoryl group, a chloromethylthio group, a phenylthio group, a halogenophenylthio group, a nitrophenylthio group, a dinitrophenylthio group, a tolylthio group or an acyl group; and X is oxygen or sulphur; with the proviso that if R_2 is a hydrogen atom and R_3 is a methyl group, then R_1 is a cycloalkyl or cycloalkenyl group of 5 to 8 carbon atoms.

The present invention relates to complexes of certain 3-substituted uracils and certain nitrogenous bases which have herbicidal activity and are believed to be new compounds, namely addition compounds of the general formula



or the corresponding isomeric formula



wherein NB represents a nitrogenous base having an ionisation constant K_b in water of at least 10^{-9}; and R_1, R_2 and R_3 are as defined in one of paragraphs (A) to (D) below:—

(A) R_1 represents an alkyl radical containing 1 to 10 carbon atoms, a substituted alkyl radical containing 1 to 8 carbon atoms, an aryl radical containing 5 to 10 carbon atoms, a substituted phenyl radical, an aralkyl radical containing 5 to 13 carbon atoms, a substituted aralkyl radical containing 5 to 13 carbon atoms, an alkenyl radical containing 3 to 8 carbon atoms, an alkynyl radical containing 3 to 8 carbon atoms, a cycloalkyl radical containing 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms, a cyano radical; a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms or a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms;

R_2 represents a chlorine, fluorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 6 carbon atoms, a hydroxy alkyl

radical containing 1 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a thiocyanato radical, a cyano radical, a mercaptomethyl (alternatively named methylthiol) radical, a methylthiomethyl radical, a phenylthiomethyl radical, an alkylthio radical containing 1 to 4 carbon atoms, a bromomethyl radical, a fluoromethyl radical, a chloromethyl radical or a carboxy-methylthiomethyl radical; and

R_3 represents a methyl radical;

(B) R_1 represents a cycloalkyl radical containing 3 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms; a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, a cycloalkenyl alkyl radical containing 4 to 13 carbon atoms or a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms;

R_2 represents a hydrogen atom; and

R_3 represents an alkyl radical containing 1 to 5 carbon atoms;

(C) R_1 represents an alkyl radical containing 1 to 10 carbon atoms, a substituted alkyl radical containing 1 to 10 carbon atoms, an aryl radical containing 6 to 14 carbon atoms, a substituted aryl radical containing 6 to 14 carbon atoms, an aralkyl radical containing 7 to 15 carbon atoms, a substituted aralkyl radical containing 7 to 15 carbon atoms, an alkenyl radical containing 3 to 10 carbon atoms, an alkynyl radical containing 3 to 10 carbon atoms, a cycloalkyl radical containing 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contain 3 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms, a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 4 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms, a cyano radical, a dialkylamino radical containing 2 to 10 carbon atoms or an alkoxy radical containing 1 to 10 carbon atoms; and

R_2 and R_3 together represent a divalent alkylene radical of formula $(CH_2)_n$ where n is 3, 4 or 5;

(D) R_1 represents an alkyl radical containing 1 to 10 carbon atoms, a substituted alkyl radical containing 1 to 10 carbon atoms, an aryl radical containing 5 to 14 carbon atoms, a substituted aryl radical containing 6 to 14 carbon atoms, an aralkyl radical containing 5 to 15 carbon atoms, a substituted aralkyl radical containing 5 to 15 carbon atoms, an alkenyl radical containing 3 to 10 carbon atoms, an alkynyl radical containing 3 to 10 carbon atoms, a cycloalkyl radical containing 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms, a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms, a cyano radical, a dialkylamino radical containing 2 to 10 carbon atoms, or an alkoxy radical containing 1 to 10 carbon atoms;

R_2 represents a hydrogen, chlorine, fluorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 6 carbon atoms, a hydroxy alkyl radical containing 1 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a thiocyanato radical, a cyano radical, a mercaptomethyl radical, a methylthiomethyl radical, a phenylthiomethyl radical, an alkylthio radical containing 1 to 4 carbon atoms, a bromomethyl radical, a fluoromethyl radical, a chloromethyl radical or a carboxymethylthiomethyl radical; with the proviso that when R_2 represents a hydrogen atom, R_1 represents a cycloalkyl radical containing 3 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms, a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms or a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms; and

R_3 represents a hydrogen atom.

Suitable nitrogenous bases are cyclic and acyclic amines, amidines and guanidines. The amines may be primary, secondary or tertiary amines, polyamines, arylamines or heterocyclic

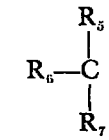
amines. Suitable nitrogenous bases include ethanolamine, dodecylamine, ethylenediamine, hexamethylenediamine, cocoadiamine, tallowdiamine, hexamethyleneimine, cyclohexylamine, methoxypropylamine, methylamine, dimethylamine, trimethylamine, ethylamine, propylamine, butylamine, octylamine, pyridine, piperidine, tetramethylguanidine, acetamidine, benzylamine, piperazine, 2-aminobutanol-1, 2-aminooctanol-1, *sec.*-butylamine, 2-amino-2-methyl-1,3-propanediol, trimethylenediamine or oleylamine. Ammonia can also be used in some cases, but it will not form complexes with all the uracils defined above, particularly when R_1, R_2, R_3 and R_4 are as defined in paragraph (B) or (C) above.

If R_1 represents a substituted alkyl radical, the or each substituent is preferably a bromine or chlorine atom, a hydroxy radical, an alkoxy radical, an alkoxycarbonyl radical, a dialkylamino radical or a cyano radical; if R_1 represents a substituted phenyl or aryl radical the or each substituent is preferably a chlorine, bromine or fluorine atom, an alkoxy radical, an alkyl radical, a nitro radical, a chloroalkyl radical, a chloroalkoxy radical or a trifluoromethyl radical; if R_1 represents a substituted benzyl radical the or each substituent is preferably a chlorine atom, an alkyl radical, an alkoxy radical or a nitro radical, and if R_1 represents a substituted cycloalkyl, substituted cycloalkenyl, (substituted cycloalkyl) alkyl or (substituted cycloalkenyl) alkyl radical the or each substituent is preferably an alkyl radical containing 1 to 4 carbon atoms, a methoxy radical or a chlorine or bromine atom.

In a first class of preferred compounds R_2 represents a chlorine, bromine or iodine atom or a methyl, hydroxymethyl, methoxymethyl or nitro radical, R_3 represents a hydrogen atom or methyl radical and R_1 represents an alkyl radical of 2 to 8 carbon atoms, a substituted alkyl radical of 2 to 8 carbon atoms, a phenyl radical, a substituted phenyl radical, an aralkyl radical of 5 to 10 carbon atoms, a substituted aralkyl radical of 5 to 10 carbon atoms, a cycloalkenyl radical of 5 to 12 carbon atoms or a cycloalkyl alkyl radical of 4 to 13 carbon atoms; and when R_3 represents a methyl radical, R_1 may also be a (substituted cycloalkyl) alkyl radical of 5 to 14 carbon atoms, and when R_3 represents a hydrogen atom, R_1 may also be a substituted cycloalkyl radical of 4 to 12 carbon atoms, a substituted cycloalkenyl radical of 5 to 12 carbon atoms or a (substituted cycloalkyl) alkyl radical of 4 to 13 carbon atoms.

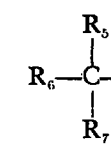
In a second class of preferred compounds R_2 and R_3 together represent a divalent alkylene radical of formula $(CH_2)_n$ where n is 3, 4 or 5 and R_1 represents an alkyl radical containing 3 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a phenyl radical, a substituted phenyl radical, a cycloalkyl radical of 3 to 10 carbon atoms or a cycloalkenyl radical of 4 to 10 carbon atoms.

In a third class of compounds X represents an oxygen atom, R_1 represents a radical of formula



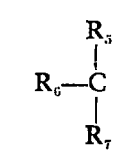
wherein R_5 represents a hydrogen atom or methyl radical, R_6 represents a methyl or ethyl radical and R_7 represents an alkyl radical containing 1 to 5 carbon atoms; R_2 represents a chlorine, fluorine, bromine or iodine atom; and R_3 represents a methyl radical.

In a fourth class of compounds X represents an oxygen atom, R_1 represents a phenyl radical, a substituted phenyl radical, a cycloalkyl radical containing 3 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a cycloalkenyl radical containing 5 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 5 to 12 carbon atoms, a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, or a radical of formula



wherein R_5, R_6 and R_7 are as defined above; R_2 represents a halogen atom or a methyl, hydroxymethyl, methoxymethyl or nitro radical; and R_3 represents a hydrogen atom.

In a fifth class, X represents an oxygen atom, R_2 and R_3 together form a divalent alkylene radical of formula $(CH_2)_n$ wherein n is 3, 4 or 5; and R_1 represents a cycloalkyl radical containing 3 to 10 carbon atoms; a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 10 carbon atoms; a cycloalkenyl radical containing 4 to 10 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 10 carbon atoms, or a radical of formula



wherein R_5, R_6 and R_7 are as defined above.

Specific radicals within the scope of the term "substituted alkyl" as used herein include bromoalkyl, chloroalkyl, hydroxyalkyl, alkoxyalkyl, alkoxycarbonyl alkyl and cyanoalkyl and also in compounds as defined in paragraphs (C) and (D), and dialkylamino alkyl.

Specific radicals within the terms "aryl" and "substituted aryl" as used herein include phenyl, naphthyl, *o*-biphenyl, pyridyl, chlorophenyl, bromophenyl, alkoxyphenyl, dichlorophenyl, dibromophenyl, fluorophenyl, trichlorophenyl, alkylphenyl of 7 to 11 carbon atoms, dialkylphenyl of 8 to 12 carbon atoms, chloroalkylphenyl of 7 to 10 carbon atoms, nitrochlorophenyl, nitrophenyl, dichloronitrophenyl, chloroalkoxyphenyl of 7 to 11 carbon atoms, trifluoromethylphenyl, alkylnaphthyl of 11 to 15 carbon atoms, chloronaphthyl, tetrahydronaphthyl and indenyl.

Specific radicals within the terms "aralkyl" and "substituted aralkyl" as used herein include furfuryl, benzyl, phenylalkyl of 8 to 11 carbon atoms (total), chlorobenzyl, dichlorobenzyl, alkylbenzyl of 8 to 11 carbon atoms (total), dialkylbenzyl of 9 to 13 carbon atoms (total), nitrobenzyl, alkoxybenzyl of 8 to 11 carbon atoms (total) and naphthylmethyl.

Specific radicals within the terms "cycloalkyl," "cycloalkenyl," "cycloalkyl alkyl," and "cycloalkenyl alkyl" include cyclododecyl, cyclohexyl, cyclohexenyl, cyclohexylalkyl, cyclohexenyl alkyl, cyclopentyl, cyclopentenyl, cyclopentyl alkyl, cyclopentenyl alkyl, cycloheptyl, cycloheptenyl, cyclooctyl, norbornyl, norbornenyl, norbornyl alkyl, norbornenyl alkyl, bicyclo(2,2,2) octyl, bicyclo(2,2,2) octenyl, bicyclo(2,2,2) octyl alkyl, bicyclo(2,2,2) octenyl alkyl, cyclopropyl, cyclobutyl, cyclobutylalkyl, cyclobutenyl, cyclobutenylalkyl, indanyl, hexahydroindenyl, tetrahydroindanyl, hexahydroindenyl alkyl, hexahydroindanyl, tetrahydroindanyl alkyl, fenchyl, bornyl, hexahydroindanyl alkyl, hexahydro-4,7-methanoindenyl, hexahydro - 4,7 - methanoindenyl alkyl, hexahydro - 4,7 - methanoindanyl, tetrahydro-4,7 - methanoindanyl, hexahydro - 4,7 - methanoindanyl alkyl, tetrahydro-4,7-methanoindanyl alkyl, decahydronaphthyl, decahydronaphthyl alkyl, tetrahydronaphthyl, tetrahydronaphthyl alkyl, decahydro - 1,4 - methanonaphthyl, decahydro - 1,4 - methanonaphthyl alkyl, octahydro - 1,4 - methanonaphthyl, octahydro - 1,4 - methanonaphthyl alkyl, decahydro-1,4,5,8-dimethanonaphthyl, decahydro - 1,4,5,8 - dimethanonaphthyl alkyl, octahydro - 1,4,5,8 - dimethanonaphthyl and octahydro-1,4,5,8-dimethanonaphthyl alkyl.

The addition compounds may be prepared generally by mixing together equimolar quantities of an appropriate uracil and nitrogenous base. The mixture is gradually heated, with stirring, until a clear melt is formed. On cooling, the addition compound crystallises. This product can then be recrystallised from a solvent such as benzene, cyclohexane, nitromethane or acetonitrile.

It is sometimes advantageous to use an inert solvent medium to carry out the reaction. Such a solvent moderates the reaction by acting as a heat sink, and allows better control of the reaction, especially if it is being carried out on a large scale. Suitable inert solvents are benzene, cyclohexane, nitromethane, acetonitrile and dioxane.

When an inert solvent is used, the addition compounds are prepared by dissolving the amine in the solvent and then adding the uracil gradually, with stirring. Stirring is continued for from ten minutes to two hours. Mild heating may be necessary. Some addition compounds precipitate and can be removed by filtration. Other addition compounds are isolated by evaporating the solvent. The addition compounds prepared in this way are suitable for use without further purification, but can be purified by recrystallisation if desired.

The nitrogenous base-uracil addition compounds as defined in paragraphs (B) and (C) are generally prepared by mixing an appropriate uracil with a 4 to 20-fold weight excess of nitrogenous base. The mixture is heated gently until a clear solution forms.

The addition compounds thus formed are stable in the presence of excess amine and can be diluted with suitable solvents for herbicidal applications.

It will be noted that the definition above refers to the isomeric enol form of the uracil, as well as the usual keto form. It is thought that there is a tautomeric equilibrium between the two forms. However, for the sake of convenience the keto form is generally referred to herein. It may also be noted that since "uracil" is the commonly accepted name for 2,4-dihydroxypyrimidine, or, in its keto form, 1,2,3,4 - tetra - hydropyrimidine - 2,4 - dione, the compounds can also be named as pyrimidines.

The compounds to which the present invention relates (referred to herein as "the said ~~compound~~ compounds") can be formulated as herbicidal compositions comprising at last one of the said compounds associated with a compatible diluent or surface-active agent, or both. The term "herbicidal composition" is used herein to include both compositions which are ready for use as herbicides and concentrates which require dilution before use as herbicides. The compositions may contain for example 0.5 to 95% by weight of the 3-substituted uracil. (Parts, proportions and percentages are by weight throughout this specification except where otherwise noted).

The diluent may be a liquid other than an oil, especially water, or an oil, or a solid carrier. The term "oil" is used herein to denote a liquid which is immiscible with water and soluble in ether, and which has a boiling point not lower than 125°C. Two or more different types of diluent may be present, as for example in compositions obtained by dispersing a wettable powder in a liquid diluent. Liquid compositions may be in the form of solutions or dispersions.

Powder and dust preparations can be made by mixing the said compounds with finely-

divided solids, preferably talcs, natural clays, pyrophyllite, diatomaceous earth, flours such as walnut shell, wheat, redwood, soya bean and cotton seed flours, or inorganic substances such as magnesium carbonate, calcium carbonate, calcium phosphate, sulphur and lime. These preparations are made by thoroughly blending the active ingredient and the solid. The particles in such preparations are preferably less than 50 microns in average diameter.

Water-soluble preparations can be prepared by mixing a said compound with an alkaline solubilising agent. Solid bases having a pH of at least 9.5 in a 1% aqueous solution, such as sodium or potassium phosphates, silicates, ~~carbonate~~, borates, oxides or hydroxides are suitable. The preparations can contain from 0.5 to 80% of the said compound and from 5 to 99.5% of the solubilising agent. Water-soluble diluents such as urea and dextrose may also be used.

Granules and pellets can be made by mixing a said compound in finely-divided form with a suitable clay, moistening this mixture with from 15 to 20% by weight of water, and for example extruding the mass through a suitable die under pressure. The extrusions are cut into pellets of pre-determined lengths and then dried. These pellets can be granulated if desired.

Granules or pellets can also be prepared by spraying a suspension or solution of a said compound onto the surface of a preformed granule of clay, vermiculite or other suitable granular material. If the said compound is in solution, it will penetrate into the pores of the granule and so will adhere without the aid of a binding agent. When the said compound is insoluble in the liquid and is carried as a suspension, it is preferable that a binding agent such as goulac, dextrin, swollen starch, glue or polyvinyl alcohol be added. In either case, the granule is then dried and ready for use.

The said compounds can also be formulated with non-aqueous liquids. Aliphatic and aromatic hydrocarbons, especially those derived from petroleum and having boiling points of from 125°C. to 400°C. are preferred. Hydrocarbons having lower boiling points should not be used because of their undesirable volatilisation characteristics and inflammability. In the case of those compounds having inadequate solubility in the carrier, it is convenient to prepare liquid compositions by milling the components in a mill such as a pebble mill until the particles have average diameters of from 1 to 50 microns, preferably 5 to 20 microns.

The herbicidal compositions, whatever physical form they take, can also contain a surface-active agent. The surfactant renders the preparations readily ~~disperible~~ in liquids and improves their action on waxy leaves and the like. For general application, surface-active agents are used in the compositions at concentrations of from 1 to 10% by weight. Levels of from 0.5 to 6 parts of surfactant for each part of uracil, however, sometimes give unusual and unexpected results. Preparations having these higher levels of surfactants sometimes show greater herbicidal effectiveness than can be expected from a consideration of activity of the components used separately.

The term "surface-active agent" is used herein to include wetting agents, dispersing agents, suspending agents and emulsifying agents. Surface-active agents suitable for use are set forth in "Detergents and Emulsifiers . . . up-to-date," 1962, John W. McCutcheon, Inc., Morristown, New Jersey. Other surface-active agents which can be used in these preparations are listed in U.S. Patent Specifications 2,412,510; 2,426,417 and 2,655,447, British Specification No. 492,742 and Bulletin E—607 of the Bureau of Entomology and Plant Quarantine of the U.S. Department of Agriculture.

The compositions can also optionally contain adhesives such as gelatin, blood albumin and such resins as rosin alkyd resins. These increase retention and tenacity of deposits following application.

The preparations can also contain corrosion inhibitors, antifoam agents, dyes and pigments, anticaking agents, and hard water stabilisers.

The herbicidal compositions can be formulated to contain two or more of the said compounds. They can also be formulated to contain other herbicides in addition to the said compounds to give compositions which have advantages over compositions containing only the individual herbicides. The other herbicide may be a 3-substituted uracil or salt or nitrogenous base or phenol addition compound thereof as disclosed in our copending applications Nos. 1,035,091, 46342/62; 1,035,092, 6191/66; 1,035,093, 6192/66; 1,035,095, 6194/66; 1,035,096, 6195/66; 1,035,097, 6196/66; and 1,035,098, 8987/66.

Among the known herbicides which can be combined with the said compounds are:

Substituted Ureas

3-(3,4-dichlorophenyl)-1,1-dimethylurea
3-(4-chlorophenyl)-1,1-dimethylurea
3-phenyl-1,1-dimethylurea
3-(3,4-dichlorophenyl)-3-methoxy-1,1-dimethylurea
3-(4-chlorophenyl)-3-methoxy-1,1-dimethylurea
3-(3,4-dichlorophenyl)-1-*n*-butyl-1-methylurea
3-(3,4-dichlorophenyl)-1-methoxy-1-methylurea
3-(4-chlorophenyl)-1-methoxy-1-methylurea
3-(3,4-dichlorophenyl)-1,1,3-trimethylurea

3 - (3,4 - dichlorophenyl) - 1,1 - diethylurea
3 - (*p* - chlorophenoxyphenyl) - 1,1 - dimethylurea.

These ureas can be mixed with the said compounds in proportions of from 1:4 to 4:1 respectively, the preferred ratio being 1:2 to 2:1.

Substituted Triazines

2-chloro-4,6-bis(ethylamino)-*s*-triazine
2 - chloro - 4 - ethylamino - 6 - isopropylamino-*s*-triazine
2 - chloro - 4,6 - bis(methoxypropylamino)-*s*-triazine
2 - methoxy - 4,6 - bis(isopropylamino)-*s*-triazine
2 - diethylamino - 4 - isopropylacetamido-6-methoxy-*s*-triazine
2 - isopropylamino - 4 - methoxyethylamino-6-methylmercapto-*s*-triazine
2 - methylmercapto - 4,6 - bis(isopropylamino)-*s*-triazine
2 - methylmercapto - 4,6 - (bis(ethylamino)-*s*-triazine
2 - methylmercapto - 4 - ethylamino - 6-isopropylamino-*s*-triazine
2 - methoxy - 4,6 - bis(ethylamino) - *s*-triazine
2 - methoxy - 4 - ethylamino - 6 - isopropylamino-*s*-triazine
2 - chloro - 4,6 - bis(isopropylamino)-*s*-triazine

These triazines can be mixed with the said compounds in proportions of from 1:4 to 4:1 respectively, the preferred ratio being 1:2 to 2:1.

Phenols

Dinitro-o-*sec.*-butylphenol and its salts
Pentachlorophenol and its salts.

These phenols can be mixed with the said compounds in the proportions of 1:10 to 20:1, respectively, the preferred ratio being 1:5 to 5:1.

Carboxylic Acids and Derivatives

The following carboxylic acids and derivatives can be mixed with the said compounds in the listed proportions:

A. 2,3,6-trichlorobenzoic acid and its salts
2,3,5,6-tetrachlorobenzoic acid and its salts
2-methoxy-3,5,6-trichlorobenzoic acid and its salts
2-methoxy-3,6-dichlorobenzoic acid and its salts
3-amino-2,5-dichlorobenzoic acid and its salts
3-nitro-2,5-dichlorobenzoic acid and its salts
2-methyl-3,6-dichlorobenzoic acid and its salts
2,4-dichlorophenoxyacetic acid and its salts and esters
2,4,5-trichlorophenoxyacetic acid and its salts and esters
(2-methyl-4-chlorophenoxy)acetic acid and its salts and esters
2-(2,4,5-trichlorophenoxy)propionic acid and its salts and esters
2 - (2,4,5 - trichlorophenoxy)ethyl 2,2-dichloropropionate
4-(2,4-dichlorophenoxy)butyric acid and its salts and esters
4-(2-methyl-4-chlorophenoxy)butyric acid and its salts and esters
2,3,6-trichlorobenzoyloxypropanol
Mixed in a 1:16 to 8:1 ratio, preferably a 1:4 to 4:1 ratio.

B. 2,6-dichlorobenzonitrile.
Mixed in a 1:4 to 4:1 ratio, preferably a 1:3 to 3:1 ratio.

C. Trichloroacetic acid and its salts
Mixed in a 1:2 to 25:1 ratio, preferably a 1:1 to 8:1 ratio.

D. 2,2-dichloropropionic acid and its salts
Mixed in a 1:4 to 8:1 ratio, preferably a 1:2 to 4:1 ratio.

E. N,N-di(*n*-propyl)thiolcarbamic acid, ethyl ester
N,N-di(*n*-propyl)thiolcarbamic acid, *n*-propyl ester
N-ethyl-N-(*n*-butyl)thiolcarbamic acid, ethyl ester
N-ethyl-N-(*n*-butyl)thiolcarbamic acid, *n*-propyl ester
Mixed in a 1:2 to 24:1 ratio, preferably a 1:1 to 12:1 ratio.

F. N-phenylcarbamic acid, isopropyl ester
N-(*m*-chlorophenyl)carbamic acid, isopropyl ester
N-(*m*-chlorophenyl)carbamic acid, 4-chloro-2-butynyl ester
Mixed in a 1:2 to 24:1 ratio, preferably a 1:1 to 12:1 ratio.

G. 2,3,6-trichlorophenylacetic acid and its salts
Mixed in a 1:12 to 8:1 ratio, preferably a 1:4 to 4:1 ratio.

H. 2-chloro-N,N-diallylacetamide
Maleic hydrazide
Mixed in a 1:2 to 10:1 ratio, preferably a 1:1 to 5:1 ratio.

Inorganic and Mixed Inorganic-Organic Salts

The following salts can be mixed with the said compounds in the listed proportions:

A. Calcium propylarsonate
Disodium monomethylarsonate
Octyl-dodecylammoniummethylarsonate
Dimethylarsinic acid
Mixed in a 1:4 to 4:1 ratio, preferably a 1:2 to 2:1 ratio.

B. Sodium arsenite
Mixed in a 1:5 to 40:1 ratio, preferably a 1:4 to 25:1 ratio.

C. Lead arsenate
Calcium arsenate
Mixed in a 100:1 to 600:1 ratio, preferably a 150:1 to 400:1 ratio.

D. Sodium tetraborate hydrated, granulated
Sodium metaborate
Sodium pentaborate
Polyborchlorate
Unrefined borate ore such as borascu
Mixed in a 3:1 to 1500:1 ratio, preferably a 6:1 to 1000:1 ratio.

E. Ammonium thiocyanate
Mixed in a 1:10 to 20:1 ratio, preferably a 1:5 to 5:1 ratio.

F. Sodium chlorate
Mixed in a 1:1 to 40:1 ratio, preferably a 2:1 to 20:1 ratio.

G. Ammonium sulphamate
Mixed in a 1:1 to 100:1 ratio, preferably a 1:1 to 50:1 ratio.

Other Organic Herbicides

These organic herbicides can be mixed with the said compounds in the listed proportions:

A. 5,6 - dihydro - (4A,6A) - dipyrido - (1,2-A,2',1'-C) pyrazinium dibromide
Mixed in a 1:20 to 16:1 ratio, preferably a 1:5 to 5:1 ratio.

B. 3-amino-1,2,4-triazole
Mixed in a 1:20 to 20:1 ratio, preferably a 1:5 to 5:1 ratio.

C. 3,6-endoxohexahydrophthalic acid
Mixed in a 1:3 to 20:1 ratio, preferably a 1:2 to 10:1 ratio.

D. Hexachloroacetone
Mixed in a 1:2 to 16:1 ratio, preferably a 1:1 to 8:1 ratio.

E. Diphenylacetonitrile
N,N-dimethyl-α,α-diphenylacetamide
N,N - di - *n* - propyl - 2,6 - dinitro - 4-trifluoromethylaniline
N,N - di - *n* - propyl - 2,6 - dinitro - 4-methylaniline
Mixed in a 1:10 to 30:1 ratio, preferably a 1:5 to 20:1 ratio.

F. O - (2,4 - dichlorophenyl) - O - methyl-isopropylphosphoramidothioate
2,3,5,6 - tetrachloroterephthalic acid, dimethyl ester
Mixed in a 1:4 to 20:1 ratio, preferably a 1:3 to 15:1 ratio.

G. 2,4-dichloro-4'-nitrodiphenyl ether
Mixed in a 1:10 to 30:1 ratio, preferably a 1:5 to 20:1 ratio.

In a further aspect the invention provides a method of controlling the growth of weeds at a locus which comprises applying to the locus a herbicidally effective quantity of at least one of the said compounds. The amount of active compound used will depend on the weeds to be controlled, the formulation used, the mode of application, the climate, season of the year, rainfall and other variables, and because of this it is not possible to state generally a particular rate of application which will be suitable for all situations.

Some of the said compounds are especially useful in controlling the growth of weeds on uncultivated areas. Thus in suitable circumstances some of the said compounds can be used as soil sterilants and/or in soil-foliage applications.

It has been found that strikingly similar herbicidal activity can be obtained with seemingly unrelated substituents round the uracil ring. It has also been found that changes in useful selective phytotoxicity (applicable in certain weed-crop combinations) may result when a relatively minor structural modification of R_1 (for example a normal alkyl chain altered to a branch chain) has been made. The 5-substituents (described for R_2 above) for example, have been found to have a minimum range of herbicidal effectiveness and/or species selectivity dependent on the nature of the substituent (if any) in the 3- or 6-position. There has been found little continuity of herbicidal effect which could be ascribed to a particular substituent independently of the other two substituents in the 3,5,6-positions.

Some of the said compounds are particularly useful against both broadleaf and grass weeds. Others are particularly effective against hard-to-kill nutsedge and perennial grasses such as quack grass, Johnson grass, and Bermuda grass. Others are particularly effective on highly adsorptive substrates such as railroad ballast, heavy clay soil, and soils high in organic matter. For example, a pre-emergence treatment has been used to control such germinating broadleaf weeds as pigweed, lamb's quarter, mustard, chickweed and ragweed, and such grass weeds as crab grass and foxtails. Some of the said compounds are particularly effective in soil-foliage applications for the control of such broadleaves as pigweed, lamb's quarter, mustard, chickweed and ragweed and such grass weeds as crab grass, foxtails, quack grass and seedling Johnson grass.

Some of the said compounds are particularly useful as soil sterilants and for general weed control, e.g. on industrial areas, railroad rights-of-way, and areas adjacent to croplands in agricultural areas.

Certain of the said compounds also exhibit

selective herbicidal action in crops. By properly selecting a said compound and a rate and time of application, annual grass and broadleaf seedlings in such crops as asparagus, corn, flax, sugar cane, safflower, peanuts, citrus, alfalfa, strawberries, gladiolus, stone fruits, sorghum, sugar beets, red beets, mangels, spinach, potatoes, pineapple, cotton and cucurbits can be controlled.

By proper selection of rate and time of application, certain of the said compounds can also be used to control weeds growing in dormant crops.

This selective activity and activity on weeds growing in dormant crops is described in more detail in the Examples.

Many of the compounds have unusually high oil solubility, and so are useful for application in oils such as herbicidal oils, diesel oil, kerosene, xylene, and other commercially available spray oils. High oil solubility makes shipment of many uracil/oil "concentrates" practical. Such concentrates can be diluted with low-cost diesel oil and herbicidal oils at the site of application.

Broadly speaking, the compounds are used at levels of about $\frac{1}{4}$ pound per acre to about 80 pounds per acre for general weed control. For selective weed control in crops, rates of $\frac{1}{4}$ or $\frac{1}{2}$ to 3 or 8 pounds per acre will generally be used, and in pre-emergence treatments from $\frac{1}{4}$ to 5, preferably $\frac{1}{2}$ to 2 or 4 pounds per acre, may for example be used. More of the active material can be used to control difficult-to-kill species growing under adverse conditions. Economic factors, such as inaccessibility of the area to be treated, e.g. fire breaks in forests, may also favour higher rates, with less frequent treatments.

The invention is illustrated by the following Examples.

EXAMPLE 1

Preparation of the ethanolamine addition compound of 5-bromo-3-isopropyl-6-methyluracil.

A solution is prepared by mixing together 247 parts of 5-bromo-3-isopropyl-6-methyluracil, 392 parts of acetonitrile and 61 parts of ethanolamine. The solvent is distilled off at reduced pressure and the oil which remains gradually solidifies. The molar addition compound is recrystallized from nitromethane; it melts at 80—83°C.

EXAMPLE 2

Preparation of the octylamine addition compound of 5 - bromo - 3 - isopropyl - 6-methyluracil.

A mixture containing one part of octylamine and one part of 5 - bromo - 3 - isopropyl - 6-methyluracil is stirred and maintained at about 40°C. with gentle heating until the ingredients are in complete solution. The solution is cooled and the hard granular crystals which form are filtered off and washed three times with small portions of cold benzene. The resulting molar addition compound melts at 74—78°C.

EXAMPLE 3

Preparation of the ethylenediamine addition compound of 5 - bromo - 3 - isopropyl-6-methyluracil.

Two hundred and forty-seven parts of 5-bromo-3-isopropyl-6-methyluracil are dissolved in 300 parts of acetonitrile at ambient temperature. Sixty parts of ethylenediamine are added and the mixture is stirred vigorously. The solid white addition compound which gradually precipitates is filtered off and dried at room temperature under reduced pressure. The addition compound melts at 102—106°C. It can be further purified by recrystallization from ~~acetonitrile~~ if desired.

The following addition compounds are prepared according to any of the procedures of Examples 1, 2 and 3 by substituting molar equivalents of a propertly substituted uracil and an appropriate amine for 5-bromo-3-isopropyl-6-methyluracil, ethanolamine, octylamine or ethylenediamine:

5 - bromo - 3 - *sec.* - butyl - 6 - methyluracil . ethanolamine addition compound

3 - *sec.* - butyl - 5 - chloro - 6 - methyluracil . ethanolamine addition compound

5 - bromo - 3 - *sec.* - butyl - 6 - methyluracil. ethylenediamine addition compound

5 - bromo - 3 - *sec.* - butyl - 6 - methyluracil . dodecylamine addition compound

5 - bromo - 3 - *sec.* - butyl - 6 - methyluracil . cocoadiamine addition compound

5 - bromo - 3 - isopropyl - 6 - methyluracil . dodecylamine addition compound

5 - chloro - 3 - isopropyl - 6 - methyluracil . ethanolamine addition compound

5 - bromo - 3 - *tert.* - butyl - 6 - methyluracil . ethanolamine addition compound

5 - bromo - 3 - *tert.* - butyl - 6 - methyluracil . dodecylamine addition compound

5,6 - dimethyl - 3 - isopropyluracil . ethanolamine addition compound

5,6 - dimethyl - 3 - isopropyluracil . dodecylamine addition compound

5 - bromo - 6 - methyl - 3 - phenyluracil . ethylenediamine addition compound

5 - bromo - 6 - methyl - 3 - phenyluracil . dodecylamine addition compound

5 - chloro - 6 - methyl - 3 - phenyluracil . ethanolamine addition compound

3 - isopropyl - 6 - methyl - 5 - nitrouracil . ethanolamine addition compound

5 - bromo 6 - methyl - 3 - *sec.* - pentyluracil . ethanolamine addition compound

5 - bromo - 3 - (3 - chlorophenyl) - 6 - methyluracil . piperidine addition compound

5 - bromo - 6 - methyl - 3 - norbornyluracil . piperidine addition compound

3 - isopropyl - 6 - methyl - 5 - nitrouracil . ammonia addition compound

3 - isopropyl - 6 - methyl - 5 - nitrouracil . methylamine addition compound

acetonitrile

EXAMPLE 4

Preparation of the addition compound of 3-cyclohexyl-6-propyluracil with dodecylamine.

A mixture containing 15 parts of 3-cyclohexyl-6-propyluracil and 85 parts of dodecylamine is stirred and gently heated. A clear solution is obtained which can be extended for use to any desired spray concentration with aromatic heavy naphtha or Lion Herbicidal Oil No. 6.

The addition compounds in the following list can be similarly prepared by substituting proper amounts of suitable reactants for 3-cyclohexyl-6-propyluracil and dodecylamine.

6 - ethyl - 3 - (3 - methylcyclohexyl)uracil-dodecylamine
6 - ethyl - 3 - (3 - methoxycyclohexyl)uracil-dodecylamine
6 - ethyl - 3 - (2 - methylcyclohexyl)uracil-oleylamine
3 - norbornyl - 6 - propyluracil - dodecylamine
6 - methyl - 3 - norbornyluracil - dodecylamine
3 - norbornylmethyl - 6 - propyluracil-dodecylamine
3 - cycloheptyl - 6 - ethyluracil - triethylamine
3 - cyclooctyl - 6 - methyluracil - 2 - amino-octanol-1

EXAMPLE 5

Preparation of the addition compound of 3-cyclooctyl-6-propyluracil with piperidine.

A mixture of one part of 3-cyclooctyl-6-propyluracil and four parts of piperidine is stirred with gentle heating until a clear solution is obtained.

This solution can be diluted to spray concentrations with herbicidal oils or water.

The following addition compounds can be similarly prepared by substituting proper amounts of suitable reactants for 3-cyclooctyl-6-propyluracil and piperidine:

3 - (2 - methylcyclohexyl) - 6 - ethyluracil-piperidine
3 - norbornyl - 6 - propyluracil - piperidine
3 - cycloheptyl - 6 - ethyluracil - 2 - methyl-piperidine
3 - cyclooctyl - 6 - methyluracil - piperidine
3 - cyclodecyl - 6 - methyluracil - 4 - methyl-piperidine
3 - cyclohexylmethyl - 6 - propyluracil - piperidine
3 - norbornylmethyl - 6 - methyluracil - piperidine

EXAMPLE 6

Preparation of 3 - *sec.* - butyl - 5,6 - tetramethyleneuracil-piperidine addition compound.

A mixture containing one part of 3-*sec.*-butyl-5,6-tetramethyleneuracil and 4 parts of piperidine is stirred and gently heated until a clear solution is obtained

By similarly employing equivalent amounts of appropriate uracils and nitrogenous bases in place of the 3-*sec.*-butyl-5,6-tetramethyleneuracil and piperidine, the following uracil-nitrogenous-base-addition products can be prepared:

3 - *sec.* - butyl - 5,6 - pentamethyleneuracil, triethylenediamine addition compound
3 - *sec.* - butyl - 5,6 - pentamethyleneuracil, ethylenediamine addition compound
3 - *sec.* - butyl - 5,6 - pentamethyleneuracil, octylamine addition compound
3 - *sec.* - butyl - 5,6 - pentamethyleneuracil, dodecylamine addition compound
3 - *sec.* - butyl - 5,6 - pentamethyleneuracil, cocoadiamine addition compound
3 - (2 - pentyl) - 5,6 - tetramethyleneuracil, piperidine addition compound
3 - (3,4 - dimethylcyclohexyl) - 5,6 - trimethyleneuracil, piperidine addition compound.

EXAMPLE 7

Preparation of 3 - (2 - methylcyclohexyl)-5,6 - trimethyleneuracil—piperidine addition compound.

A mixture containing one part of 3 - (2-methylcyclohexyl) - 5,6 - trimethyleneuracil, and 20 parts of piperidine is stirred and gently heated until a clear solution is obtained.

The following can be prepared in the same fashion by susbtituting equivalent amounts of appropriate uracils and amines for the 3-(2-methylcyclohexyl)-5,6-trimethyleneuracil and piperidine:

3 - *sec.* - butyl - 5,6 - trimethyleneuracil, butylamine addition compound
3 - (3 - methylcyclohexyl) - 5,6 - trimethyleneuracil, piperidine addition compound
3 - (3,4 - dimethylcyclohexyl) - 5,6 - trimethyleneuracil, piperidine addition compound
3 - (3 - methylcyclohexyl) - 5,6 - tetramethyleneuracil, methoxypropylamine addition compound
3 - *tert.* - butyl - 5,6 - tetramethyleneuracil, hexamethylenediamine addition compound
3 - cyclohexyl - 5,6 - trimethyleneuracil, piperazine addition compound
3 - *sec.* - butyl - 5,6 - trimethyleneuracil, *sec.*-butylamine addition compound.

EXAMPLE 8

Preparation of the ethylenediamine addition compound of 5-bromo-3-isopropyluracil.

A mixture of 233 parts of 5-bromo-3-isopropyluracil in sufficient acetonitrile to form a concentrated solution at room temperature is stirred as 60 parts of ethylenediamine are gradually added.

The solid pure addition compound of 5-bromo-3-isopropyluracil and ethylenediamine which precipitates is filtered off.

EXAMPLE 9

Preparation of the ethanolamine addition compound of 3 - cyclohexyl - 5 - methyluracil.

A mixture of 208 parts of 3-cyclohexyl-5-methyluracil and 61 parts of ethanolamine is gradually heated. The resulting liquid is stirred to assure even distribution of the two components, and is then allowed to cool and solidify.

The resulting solid addition compound is sufficiently pure for incorporation into herbicidal formulations. If desired, it may be purified by recrystallization from such solvents as acetonitrile or nitromethane.

Oil Formulations

EXAMPLE 10

5-bromo-3-*sec.*-butyl-6-methyluracil, 1:1 addition compound with dodecylamine	25%
Dodecylamine	25%
Aromatic heavy naphtha	50%

These components are mixed until a clear solution results.

Sixty pounds of this formulation are mixed with 80 gallons of Lion Herbicidal Oil No. 6 and applied at the rate of 15 pounds of active ingredient per acre to a weed infestation of annual and perennial broadleaves and grasses growing along a railroad right-of-way. Excellent initial contact kill of weeds, followed by good residual weed control, is obtained. Such weed species as pigweed, ragweed, Johnson grass, broomsedge, foxtail, crab grass, dock and wild carrot are controlled.

EXAMPLE 11

A wettable powder is prepared by blending the following components in a ribbon blender and micropulverizing them until the particles are below 50 microns in diameter. The mixture is then reblended until homogeneous.

1:1 addition compound of 3-isopropyl-5-bromo-6-methyluracil with octylamine	25%
Attapulgite clay	70%
Dioctyl ester of sodium sulfosuccinic acid	1%
Sodium lignin sulfonate	4%

This wettable powder is dispersed in 60 gallons of water and applied at the rate of fifteen pounds of active ingredient per acre on an unsurfaced parking lot. Volunteer small grains, crab grass, ryegrass, wild mustard, chickweed, and dandelion are controlled.

EXAMPLE 12—*Solution.*

A mixture of 1 part 3-cyclohexyl-6-methyluracil and 9 parts ethanolamine is stirred and gently heated to form a clear solution.

This formulation is applied pre-emergence at the rate of 1 to 2 pounds (active) in 40 gallons of water to a field planted to sugar cane. Excellent control of seedling Johnson grass, ryegrass, crab grass, foxtail, water grass, lamb's quarters and wild mustard is obtained.

This formulation also gives post-emergence control of crab grass and foxtail in established stands of sugar cane. Two pounds (active) are mixed in 40 gallons water. To this mixture is added 2½ pounds of a surfactant such as trimethylnonyl ether of polyetheylene glycol. This mixture is applied as a directed spray to crab grass and foxtail grasses 4 inches high, growing in sugar cane 12—14 inches high. Quick kill of these weeds is obtained.

The following addition compounds can be similarly formulated and used, pre-emergence, with substantially equivalent results:

3 - cyclopentyl - 6 - propyluracil - methoxypropylamine
3 - cycloheptyl - 6 - ethyluracil - propanolamine
3 - cyclooctyl - 6 - methyluracil - butanolamine
3 - cyclohexyl - 6 - methyluracil - piperidine
3 - cyclohexylmethyl - 6 - methyluracil - ethanolamine
3 - (3 - methylcyclohexyl) - 6 - methyluracil-ethanolamine
3 - cyclopropyl - 6 - propyluracil - piperidine
3 - cyclodecyl - 6 - methyluracil - ethanolamine
3 - cyclopentenyl - 6 - ethyluracil - ethanolamine
3 - (2 - methylcyclohexyl) - 6 - methyluracil-ethanolamine
3 - (3 - methoxycyclohexyl) - 6 - methyluracil-propanolamine
3 - cyclohexyl - 6 - methyluracil - ethanolamine
3 - norbornyl - 6 - methyluracil - piperidine
3 - norbornylmethyl - 6 - methyluracil-ethanolamine
3 - norbornylmethyl - 6 - methyluracil - piperidine

EXAMPLE 13 — *Aqueous Solution*

3 - *sec.* - butyl - 5,6 - tetramethyleneuracil	20%
Piperidine	80%

The components are mixed together at room temperature until a clear solution is formed. This solution is extendable with water.

Twenty pounts (active) of this formulation are mixed with 60 gallons of water in a spray tank. Eight pounds of trimethylnonyl ether of polyethylene glycol are added. Only slight agitation is required for complete mixing.

The sixty gallons are sprayed on an area of one acre along roadsides. Excellent kill and residual control of such species as wild oats, cheatgrass, annual bluegrass, crab grass, foxtails, ryegrass, filaree, wild mustard, and beggar tick is obtained.

The following compounds can be substituted for 3 - *sec.* - butyl - 5,6 - tetramethyleneuracil-piperidine addition compound, in herbicidally equivalent amounts, and when used in a like fashion, will give good weed control:

3 - isopropyl - 5,6 - pentamethyleneuracil, piperidine addition compound
3 - *tert.* - butyl - 5,6 - tetramethyleneuracil, piperidine addition compound
3 - *tert.* - butyl - 5,6 - pentamethyleneuracil, ethylenediamine addition compound
3 - *n* - butyl - 5,6 - pentamethyleneuracil, piperidine addition compound
3 - *sec.* - butyl - 5,6 - tetramethyleneuracil, hexamethylenediamine addition compound
3 - *sec.* - butyl - 5,6 - pentamethyleneuracil, *sec.*-butylamine addition compound
3 - *sec.* - butyl - 5,6 - pentamethyleneuracil, cyclohexylamine addition compound
3 - *sec.* - butyl - 5,6 - tetramethyleneuracil, piperazine addition compound

EXAMPLE 14 — *Wetttable Powder.*

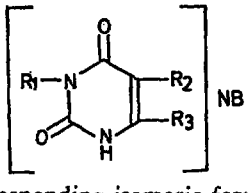
3-isopropyl-5-methyluracil, 1-1 addition compound with octyl amine	40.0%
Alkyl naphthalene sulfonic acid, sodium salt	1.0%
Low viscosity methyl cellulose	0.3%
Attapulgite clay	58.7%

These components are blended, micropulverized, and reblended to give a homogeneous powder.

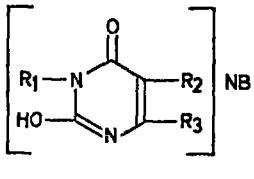
This wettable powder is applied as a pre-emergence treatment, at a rate of 5 pounds of active herbicide per acre in 40 gallons of water, to the area surrounding an outdoor electrical transformer and switching station. When so applied, it gives excellent control of seedling Bermuda grass, seedling bindweed, wild mustard, giant foxtail, crab grass, barnyard grass, and chickweed.

WHAT WE CLAIM IS:—

1. A herbicidal composition comprising a diluent which is at least in part an oil and in association therewith at least one 3-substituted uracil addition compound of the general formula



or the corresponding isomeric formula



wherein NB represents a nitrogenous base having an ionisation constant K_b in water of at least 10^{-9}; and R_1, R_2 and R_3 are as defined in one of paragraphs (A) to (D) below:—

(A) R_1 represents an alkyl radical containing 1 to 10 carbon atoms, a substituted alkyl radical containing 1 to 8 carbon atoms, an aryl radical containing 5 to 10 carbon atoms, a substituted phenyl radical, an aralkyl radical containing 5 to 13 carbon atoms, a substituted aralkyl radical containing 5 to 13 carbon atoms, an alkenyl radical containing 3 to 8 carbon atoms, an alkynyl radical containing 3 to 8 carbon atoms, a cycloalkyl radical containing 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms, a cyano radical; a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms or a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms;
R_2 represents a chlorine, fluorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 6 carbon atoms, a hydroxy alkyl radical containing 1 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a thiocyanato radical, a cyano radical, a mercaptomethyl radical, a methylthiomethyl radical, a phenylthiomethyl radical, an alkylthio radical containing 1 to 4 carbon atoms, a bromomethyl radical, a fluoromethyl radical, a chloromethyl radical or a carboxy-methylthiomethyl radical; and
R_3 represents a methyl radical;

(B) R_1 represents a cycloalkyl radical containing 3 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms; a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, a cycloalkenyl alkyl radical containing 4 to 13 carbon atoms or a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms;
R_2 represents a hydrogen atom; and
R_3 represents an alkyl radical containing 1 to 5 carbon atoms;

The following compounds can be substituted for 3 - *sec.* - butyl - 5,6 - tetramethyleneuracil-piperidine addition compound, in herbicidally equivalent amounts, and when used in a like fashion, will give good weed control:

3 - isopropyl - 5,6 - pentamethyleneuracil, piperidine addition compound
3 - *tert.* - butyl - 5,6 - tetramethyleneuracil, piperidine addition compound
3 - *tert.* - butyl - 5,6 - pentamethyleneuracil, ethylenediamine addition compound
3 - *n* - butyl - 5,6 - pentamethyleneuracil, piperidine addition compound
3 - *sec.* - butyl - 5,6 - tetramethyleneuracil, hexamethylenediamine addition compound
3 - *sec.* - butyl - 5,6 - pentamethyleneuracil, *sec.*-butylamine addition compound
3 - *sec.* - butyl - 5,6 - pentamethyleneuracil, cyclohexylamine addition compound
3 - *sec.* - butyl - 5,6 - tetramethyleneuracil, piperazine addition compound

EXAMPLE 14 — *Wetttable Powder.*

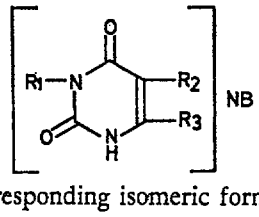
3-isopropyl-5-methyluracil, 1-1 addition compound with octyl amine	40.0%
Alkyl naphthalene sulfonic acid, sodium salt	1.0%
Low viscosity methyl cellulose	0.3%
Attapulgite clay	58.7%

These components are blended, micropulverized, and reblended to give a homogeneous powder.

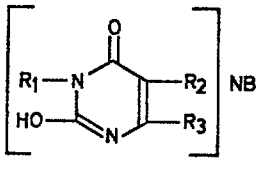
This wettable powder is applied as a pre-emergence treatment, at a rate of 5 pounds of active herbicide per acre in 40 gallons of water, to the area surrounding an outdoor electrical transformer and switching station. When so applied, it gives excellent control of seedling Bermuda grass, seedling bindweed, wild mustard, giant foxtail, crab grass, barnyard grass, and chickweed.

WHAT WE CLAIM IS:—

1. A herbicidal composition comprising a diluent which is at least in part an oil and in association therewith at least one 3-substituted uracil addition compound of the general formula



or the corresponding isomeric formula



wherein NB represents a nitrogenous base having an ionisation constant K_b in water of at least 10^{-9}; and R_1, R_2 and R_3 are as defined in one of paragraphs (A) to (D) below:—

(A) R_1 represents an alkyl radical containing 1 to 10 carbon atoms, a substituted alkyl radical containing 1 to 8 carbon atoms, an aryl radical containing 5 to 10 carbon atoms, a substituted phenyl radical, an aralkyl radical containing 5 to 13 carbon atoms, a substituted aralkyl radical containing 5 to 13 carbon atoms, an alkenyl radical containing 3 to 8 carbon atoms, an alkynyl radical containing 3 to 8 carbon atoms, a cycloalkyl radical containing 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms, a cyano radical; a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms or a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms;
R_2 represents a chlorine, fluorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 6 carbon atoms, a hydroxy alkyl radical containing 1 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a thiocyanato radical, a cyano radical, a mercaptomethyl radical, a methylthiomethyl radical, a phenylthiomethyl radical, an alkylthio radical containing 1 to 4 carbon atoms, a bromomethyl radical, a fluoromethyl radical, a chloromethyl radical or a carboxy-methylthiomethyl radical; and
R_3 represents a methyl radical;

(B) R_1 represents a cycloalkyl radical containing 3 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms; a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, a cycloalkenyl alkyl radical containing 4 to 13 carbon atoms or a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms;
R_2 represents a hydrogen atom; and
R_3 represents an alkyl radical containing 1 to 5 carbon atoms;

(C) R_1 represents an alkyl radical containing 1 to 10 carbon atoms, a substituted alkyl radical containing 1 to 10 carbon atoms, an aryl radical containing 6 to 14 carbon atoms, a substituted aryl radical containing 6 to 14 carbon atoms, an aralkyl radical containing 7 to 15 carbon atoms, a substituted aralkyl radical containing 7 to 15 carbon atoms, an alkenyl radical containing 3 to 10 carbon atoms, an alkynyl radical containing 3 to 10 carbon atoms, a cycloalkyl radical containing 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms, a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 4 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms, a cyano radical, a dialkylamino radical containing 2 to 10 carbon atoms or an alkoxy radical containing 1 to 10 carbon atoms; and

R_2 and R_3 together represent a divalent alkylene radical of formula $(CH_2)_n$ where n is 3, 4 or 5;

(D) R_1 represents an alkyl radical containing 1 to 10 carbon atoms, a substituted alkyl radical containing 1 to 10 carbon atoms, an aryl radical containing 5 to 14 carbon atoms, a substituted aryl radical containing 6 to 14 carbon atoms, an aralkyl radical containing 5 to 15 carbon atoms, a substituted aralkyl radical containing 5 to 15 carbon atoms, an alkenyl radical containing 3 to 10 carbon atoms, an alkynyl radical containing 3 to 10 carbon atoms, a cycloalkyl radical containing 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms, a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms, a cyano radical, a dialkylamino radical containing 2 to 10 carbon atoms, or an alkoxy radical containing 1 to 10 carbon atoms;

R_2 represents a hydrogen, chlorine, fluorine, bromine or iodine atom, a methyl radical, an ethyl radical, a propyl radical, a butyl radical, a methoxy radical, an ethoxy radical, a propoxy radical, a butoxy radical, a nitro radical, an alkoxymethyl radical containing 2 to 6 carbon atoms, a hydroxy alkyl radical containing 1 to 6 carbon atoms, an alkenyl radical containing 3 to 6 carbon atoms, a thiocyanato radical, a cyano radical, a mercaptomethyl radical, a methylthiomethyl radical, a phenylthiomethyl radical, an alkylthio radical containing 1 to 4 carbon atoms, a bromomethyl radical, a fluoromethyl radical, a chloromethyl radical or a carboxymethylthiomethyl radical; with the proviso that when R_2 represents a hydrogen atom, R_1 represents a cycloalkyl radical containing 3 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a cycloalkenyl radical containing 4 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 12 carbon atoms, a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, a cycloalkenyl alkyl radical containing 5 to 13 carbon atoms or a (substituted cycloalkenyl) alkyl radical containing 5 to 14 carbon atoms; and

R_3 represents a hydrogen atom.

2. A herbicidal composition comprising at least one 3-substituted uracil addition compound as defined in claim 1 associated with a diluent which is at least in part a solid.

3. A herbicidal composition according to claim 1 which contains a surface-active agent.

4. A herbicidal composition according to claim 2 which contains a surface-active agent.

5. A herbicidal composition comprising at least one 3-substituted uracil addition compound as defined in claim 1 associated with a surface-active agent and a diluent which is a liquid other than an oil.

6. A herbicidal composition according to claim 5 wherein the liquid is water.

7. A herbicidal composition comprising at least one 3-substituted uracil addition compound as defined in claim 1 associated with a surface-active agent.

8. A herbicidal composition according to any one of claims 3 to 7, which contains 1 to 10% by weight of the surface-active agent.

9. A herbicidal composition according to any one of the preceding claims which contains, in addition to the 3-substituted uracil addition compound, another compound having herbicidal properties.

10. A herbicidal composition comprising at least one 3-substituted uracil addition compound as defined in claim 1 in association with a diluent and another compound having herbicidal properties.

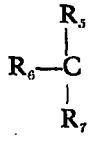
11. A herbicidal composition according to any one of the preceding claims wherein the nitrogenous base is ammonia, ethanolamine, dodecylamine, ethylenediamine, hexamethylene-

diamine, cocoadiamine, tallowdiamine, hexamethyleneimine, cyclohexylamine, methoxypropylamine, methylamine, dimethylamine, trimethylamine, ethylamine, propylamine, butylamine, octylamine, pyridine, piperidine, tetramethylguanidine, acetamidine, benzylamine, piperazine, 2-aminobutanol-1, 2-aminooctanol-1, *sec.*-butylamine, 2-amino-2-methyl-1,3-propanediol, trimethylenediamine or oleylamine.

12. A herbicidal composition according to any one of the preceding claims, wherein if R_1 represents a substituted alkyl radical, the or each substituent is a bromine or chlorine atom, a hydroxy radical, an alkoxy radical, an alkoxycarbonyl radical, a dialkylamino radical or a cyano radical; if R_1 represents a substituted phenyl or aryl radical the or each substituent is a chlorine, bromine or fluorine atom, an alkoxy radical, an alkyl radical, a nitro radical, a chloroalkyl radical, a chloroalkoxy radical or a trifluoromethyl radical; if R_1 represents a substituted benzyl radical the or each substituent is a chlorine atom, an alkyl radical, an alkoxy radical or a nitro radical; and if R_1 represents a substituted cycloalkyl, substituted cycloalkenyl, (substituted cycloalkyl) alkyl or (substituted cycoalkenyl) ~~alky~~ alkyl radical the or each substituent is an alkyl radical containing 1 to 4 carbon atoms, a methoxy radical or a chlorine or bromine atom.

13. A herbicidal composition according to any one of claims 1 to 12 wherein R_1, R_2 and R_3 are as defined in paragraph (A) of Claim 1.

14. A herbicidal composition according to Claim 13 wherein X represents an oxygen atom, R_1 represents a radical of formula

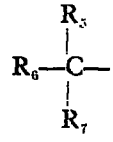


wherein R_5 represents a hydrogen atom or methyl radical, R_6 represents a methyl or ethyl radical and R_7 represents an alkyl radical containing 1 to 5 carbon atoms; and R_2 represents a chlorine, fluorine, bromine or iodine atom.

15. A herbicidal composition according to Claim 13 wherein the 3-substituted uracil addition compound is one of the compounds specifically named in Examples 1, 2, 3, 10 and 11.

16. A herbicidal composition according to any one of claims 1 to 12 wherein R_1, R_2 and R_3 are as defined in paragraph (D) of claim 1.

17. A herbicidal composition according to claim 16 wherein X represents an oxygen atom, R_1 represents a phenyl radical, a substituted phenyl radical, a cycloalkyl radical containing 3 to 12 carbon atoms, a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 12 carbon atoms, a cycloalkenyl radical containing 5 to 12 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 5 to 12 carbon atoms, a cycloalkyl alkyl radical containing 4 to 13 carbon atoms, a (substituted cycloalkyl) alkyl radical containing 5 to 14 carbon atoms, or a radical of formula



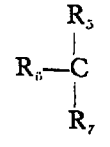
wherein R_5, R_6 and R_7 are as defined in Claim 14; and

R_2 represents a halogen atom or a methyl, hydroxymethyl, methoxymethyl or nitro radical.

18. A herbicidal composition according to Claim 16 wherein the 3-substituted uracil addition compound is one of the compounds specifically named in Examples 8, 9 and 14.

19. A herbicidal composition according to any one of claims 1 to 12 wherein R_1, R_2 and R_3 are as defined in paragraph (C) of Claim 1.

20. A herbicidal composition according to Claim 19 wherein X represents an oxygen atom, R_1 represents a cycloalkyl radical containing 3 to 10 carbon atoms; a substituted cycloalkyl radical in which the cycloalkyl radical contains 3 to 10 carbon atoms; a cycloalkenyl radical containing 4 to 10 carbon atoms, a substituted cycloalkenyl radical in which the cycloalkenyl radical contains 4 to 10 carbon atoms, or a radical of formula



wherein R_5, R_6 and R_7 are as defined in Claim 14.

21. A herbicidal composition according to Claim 19 wherein the 3-substituted uracil addition compound is one of the compounds specifically named in Examples 6, 7 and 13.

22. A herbicidal composition according to any one of Claims 1 to 12 wherein R_1, R_2 and R_3 are as defined in paragraph (B) of Claim 1.

23. A herbicidal composition according to Claim 22 wherein the 3-substituted uracil addition compound is one of the compounds specifically named in Examples 4, 5 and 12.

24. A herbicidal composition according to any one of Claims 1 to 12 which contains a 3-substituted uracil addition compound specifically hereinbefore named.

25. A herbicidal composition according to any one of the preceding claims containing one of the known herbicides specifically hereinbefore named.

26. A herbicidal composition according to any one of the preceding claims substantially as hereinbefore described.

* C L M S *

27. A herbicidal composition according to any one of the preceding claims substantially as described in any one of the foregoing Examples.

28. A method of controlling the growth of weeds at a locus which comprises applying to the locus a herbicidally effective quantity of at least one 3-substituted uracil addition compound as defined in any one of claims 1 to 24.

29. A method according to claim 28, wherein the 3-substituted uracil addition compound is applied in the form of a herbicidal composition as claimed in any one of Claims 1 to 27, after dilution where appropriate.

30. A method according to Claim 28 or 29 wherein the growth of the weeds specified in any one of Examples 10 to 14 is controlled by the application of the 3-substituted uracil addition compound specified in that Example.

31. A method according to Claim 28 substantially as hereinbefore described.

32. A method according to Claim 28 wherein the 3-substituted uracil addition compound is as defined in Claim 13, 14 or 15.

33. A method according to Claim 32 wherein the 3-substituted uracil addition compound is applied in the form of a herbicidal composition as claimed in Claim 13, 14 or 15.

34. A method according to Claim 32 wherein the growth of the weeds specified in Examples 10 and 11 is controlled by the application of the 3-substituted uracil addition compound specified in that Example.

35. A method according to Claim 28 wherein the 3-substituted uracil addition compound is as defined in Claim 16, 17 or 18.

36. A method according to Claim 35 wherein the 3-substituted uracil addition compound is applied in the form of a herbicidal composition as claimed in Claim 16, 17 or 18.

37. A method according to Claim 35 wherein the growth of the weeds specified in Example 14 is controlled by the application of the 3-substituted uracil addition compound specified in that Example.

38. A method according to Claim 28 wherein the 3-substituted uracil addition compound is as defined in Claim 19, 20 or 21.

39. A method according to Claim 38 wherein the 3-substituted uracil addition compound is applied in the form of a herbicidal composition as claimed in Claim 19, 20 or 21.

40. A method according to Claim 38 wherein the growth of the weeds specified in Example 13 is controlled by the application of the 3-substituted uracil addition compound specified in that Example.

41. A method according to Claim 28 wherein the 3-substituted uracil addition compound is as defined in Claim 22 or 23.

42. A method according to Claim 41 wherein the 3-substituted uracil addition compound is applied in the form of a herbicidal composition as claimed in Claim 22 or 23.

43. A method according to Claim 41 wherein the growth of the weeds specified in Example 12 is controlled by the application of the 3-substituted uracil addition compound specified in that Example.

44. As a new compound a 3-substituted uracil addition compound as defined in any one of claims 1 to 24.

45. As a new compound a 3-substituted uracil addition compound as defined in Claim 13, 14 or 15.

46. As a new compound a 3-substituted uracil addition compound as defined in Claim 16, 17 or 18.

47. As a new compound a 3-substituted uracil addition compound as defined in Claim 19, 20 or 21.

48. As a new compound a 3-substituted uracil addition compound as defined in Claim 22 or 23.

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Leamington Spa: Printed for Her Majesty's Stationery Office, by the Courier Press (Leamington) Ltd.—1966. Published by The Patent Office, 25 Southampton Buildings, London, W.C.2, from which copies may be obtained.

