70201/91

COMMONWEALTH OF AUSTRALIA

Patents Act 1952

632900

FORM 4 REGULATION 10

CONVENTION APPLICATION FOR A PATENT

We, MONSANTO EUROPE S.A., of Avenue de Tervuren 270-272, B-1150 Brussels, Belgium, hereby apply for the grant of a Patent for an invention entitled

GLYPHOSATE COMPOSITIONS AND THEIR USE

which is described in the accompanying complete specification. This application is made under the provision of Part XVI of the Patents Act 1952 and is based on an application for a Patent of similar protection made in Great Britain on 5 February 1990 under GB-9002495.1 by MONSANTO EUROPE S.A.

Our address for service is care of E.F. WELLINGTON & CO., Trade Mark and Patent Attorneys, 312 St. Kilda Road, Melbourne, 3004 in the State of Victoria, Commonwealth of Australia.

Dated this ..4th..... day of .February.. 1991

For and on behalf of
MONSANTO EUROPE S.A.
By:

B.S. WELLINGTON

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COMMONWEALTH OF AUSTRALIA

PATENTS ACT 1952-1969

FORM 7 REGULATION 11 (2)

DECLARATION IN SUPPORT OF A CONVENTION APPLICATION FOR A PATENT

In support of the Convention Application made by Monsanto Europe S.A. for a Patent for an invention entitled:

GLYPHOSATE COMPOSITIONS AND THEIR USE

I, BRIAN WALTER NASH of Monsanto Europe S.A., Avenue de Tervuren 270-272, B-1150 Brussels, Belgium do solemnly and sincerely declare as follows:

1. I am authorised by Monsanto Europe S.A. the applicant for the Patent to make this declaration on its behalf.

2. The basic application as defined by Section 141 of the Act was made at the British Patent Office in London, England on the fifth day of February 1990 by Monsanto Europe S.A.

3. Jean Pierre CLAUDE, of Poelweg 10, B-1900 Overijse, Belgium, Shuaib Ahmad KHAN, of Rue Philippe Baucq 100, B-1040, Brussels, Belgium, Robert William MITCHELL, of Robijnlaan 1, B-1900 Overijse, Belgium are the actual inventors of the invention and the facts upon which Monsanto Europe S.A. is entitled to make the application are as follows:

Monsanto Europe S.A. is the assignee of the actual inventors.

4. The basic application referred to in paragraph 2 of this Declaration was the first application made in a Convention country in respect of the invention the subject of the application.

Declared at Brussels, Belgium

This twenty-eighth day of January 1991

B.W. Nash

. . . . . .

BRIAN WALTER NASH

AU9170201

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(19) AUSTRALIAN PATENT OFFICE (10) Acceptance No. 632900

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GLYPHOSATE COMPOSITIONS AND THEIR USE

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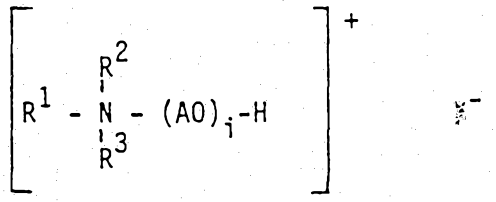
(56) Prior Art Documents
AU 40893/89 A01N 25/30 43/70

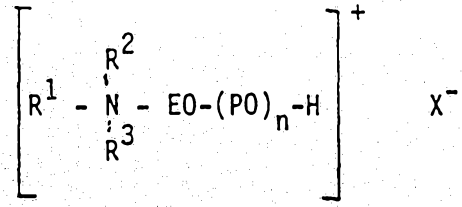
(57) Claim

1. A composition comprising a glyphosate herbicide and a quaternary ammonium compound having the formula:



wherein A or each A represents an alkylene group having 2 or 3 carbon atoms, R^1 and R^2 are each independently an alkyl group having from 1 to 5 carbon atoms; R^3 is an alkyl group having from 1 to 5 carbon atoms or a group having the formula $(AO)_j$-H; i (in a compound in which R^3 is an alkyl group) or i + j (in a compound in which R^3 is a group having the formula $(AO)_j$-H) has a value of from 2 to 20; and X^- is a suitable anion.

2. A composition comprising a glyphosate herbicide and a quaternary ammonium compound the latter having the formula



.../2

wherein -EO- means an ethylene oxide radical and -PO- means a propylene oxide radical, R^1 and R^2 are each independently an alkyl group having from 1 to 3 carbon atoms; R^3 is an alkyl group having from 1 to 3 carbon atoms, a group having the formula $(EO)_m$-H or a group having the formula EO-$(PO)_m$-H; n (in a compound in which R^3 is an alkyl group) or n + m (in a compound in which R^3 is a group having the formula $(EO)_m$-H or the formula EO-$(PO)_m$-H), has a value of from about 2 to about 20; and X^- is a suitable anion.

3. A composition according to Claim 2 wherein, in the formula of the quaternary ammonium compound, -PO- means isopropylene oxide and n (in a compound in which R^3 is a methyl or ethyl group) has a value of from 4 to 15, and n + m (in a compound in which R^3 is a group having the formula EO-$(PO)_m$-H) has a value of from 3 to 14 and X^- is halide, ethylsulfate, methylsulfate, dimethylphosphate, polyalkoxyphosphate, lactate or acetate.

13. A composition according to any one of Claims 1-12 which further contains anti-freeze agents and/or dyes and/or thickening agents and/or anti-foam agents and/or co-surfactants.

16. A composition according to any one of Claims 1 to 6, 12 and 13 which is a spray solution having a concentration such that when sprayed at 100 - 600 litres per hectare, 0.125 to 1.5 kg of glyphosate (expressed as glyphosate acid equivalent) per hectare is applied.

COMMONWEALTH OF AUSTRALIA

PATENTS ACT 1952

632900

FORM 10

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Class: Int. Class

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Related Art:

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Melbourne, 3004, Victoria.

Complete Specification for the invention entitled:

"GLYPHOSATE COMPOSITIONS AND THEIR USE"

The following statement is a full description of this invention including the best method of performing it known to us.

This invention relates to an agriculturally acceptable glyphosate composition, more particularly to such compositions containing a glyphosate herbicide and a surfactant which enhances the herbicidal activity of the glyphosate herbicide. This invention also relates to the use of such compositions to kill or control unwanted and undesired vegetation. More particularly, such compositions contain a herbicidally effective amount of glyphosate.

Numerous studies have been made on the effect of additives on the herbicidal action of glyphosate. For example Wyrill and Burnside, Weed Science, Vol. 25 (1977), 275-287, examined solutions containing different classes of surfactant, including polyoxyethylene stearyl methyl ammonium chlorides containing respectively 2 and 15 oxyethylene units. Some classes of surfactant were more effective than others in enhancing the herbicidal effect of glyphosate (used as a solution of the isopropylamine salt), but Wyrill and Burnside concluded that an effective surfactant is a critical component of any glyphosate spray mixture.

Commercially sold glyphosate formulations generally contain a surfactant. These formulations are safe herbicidal formulations from the point of view of lack of toxicity and environmental acceptability. Moreover, the level of toxicity or irritancy which is shown in certain situations, for example commercial glyphosate formulations containing an ethoxylated tallow amine surfactant, which have some toxicity to aquatic life, is essentially due to the presence of the ethoxylated tallow amine surfactant and not due merely to the glyphosate itself.

This invention satisfies the need for an improved glyphosate formulation. In this invention currently employed surfactants are advantageously replaced by other surfactants which provide a similar efficacy enhancing effect to the herbicidal activity of glyphosate but also provide lower irritancy and toxicity than currently employed surfactants.

Ammonium salts such as ammonium sulphate (Turner and Loader, Weed Research, Vol. 20 (1980), 139-146) and ammonium thiocyanate (US-A-4 612 034) have also been shown to further enhance the herbicidal activity of glyphosate salt formulations containing surfactants.

EP-A-0 290 416 allegedly discloses improved glyphosate formulations containing alkoxylated amine surfactants, more particularly alkoxylated tallow amine surfactants, and possibly ammonium sulfate. The additives disclosed are said to enhance visibly the glyphosate activity. The formulations disclosed are said to show low toxicity characteristics against the organisms, partially due to lower content of surfactant.

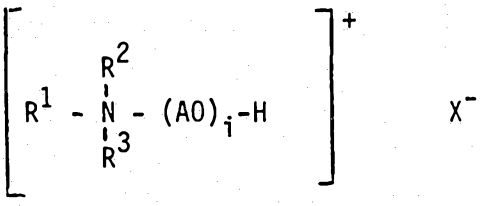
We have discovered that certain polyoxyalkylene quaternary ammonium compounds (disclosed hereinafter) are very effective in enhancing the herbicidal activity of glyphosate, and that improved glyphosate formulations containing such quaternary ammonium compounds are relatively non-toxic and non-irritant. We have also found that our new glyphosate formulations provide reduced foaming in comparison with known glyphosate formulations.

US-A-3 141 905, US-A-3 123 641 and US-A-3 123 640 disclose some particular polyoxyalkylated quaternary ammonium surfactants. The use of these types of surfactants in glyphosate compositions is however not disclosed nor suggested. Further, no toxicity or irritancy data in herbicide compositions are disclosed.

British patents No. 1 421 133, 1 462 043, 1 470 618, 1 450 531, 1 453 443, EP-A-0 066 946, WO-87/04595, GB-A-2 113 093, GB-A-2 059 773 and GB-A-2 047 098 disclose quaternary ammonium surfactants in pesticide formulations, but none of them discloses or suggests applicant's composition of glyphosate herbicide with a quaternary ammonium surfactant(s) disclosed herein.

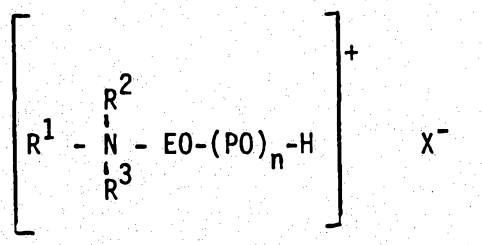
European patent No. 0 206 537 discloses the use of Emcol-CC 57, a polypropoxylated quaternary ammonium surfactant, in solid phytoactive glyphosate compositions.

A composition of the invention comprises a glyphosate herbicide and a quaternary ammonium compound having the formula:



wherein A or each A represents an alkylene group having 2 or 3 carbon atoms, R^1 and R^2 are each independently an alkyl group having from 1 to 5 carbon atoms; R^3 is an alkyl group having from 1 to 5 carbon atoms or a group having the formula $(AO)_j$-H; i (in a compound in which R^3 is an alkyl group) or i + j (in a compound in which R^3 is a group having the formula $(AO)_j$-H) has a value of from 2 to 20; and X^- is a suitable anion.

A preferred composition of the invention comprises a glyphosate herbicide and a quaternary ammonium compound the latter having a structure represented by the formula:



(I)

wherein -EO- is an ethylene oxide radical and -PO- is a propylene oxide radical, R^1 and R^2 are each independently an alkyl group having from 1 to 3 carbon atoms; R^3 is an alkyl group having from 1 to 3 carbon atoms, a group having the formula $(EO)_m$-H or a group having the formula EO-$(PO)_m$-H; n (in a compound in which R^3 is an alkyl group) or n + m (in a compound in which R^3 is a group having the formula $(EO)_m$-H or the formula EO-$(PO)_m$-H), has a value of from 2 to 20; and X^- is a suitable anion and n and m are each independently varying integers.

By a "suitable" anion is meant an anion such that applicant's composition is classified both as "non-irritant" when evaluated by a standard test procedure involving dermal and ocular tissue of the rabbit, and as "relatively non-toxic" to fish when evaluated by standard test procedures both hereinafter identified.

The compositions of this invention containing quaternary ammonium compounds as disclosed herein are non irritant and have relatively low toxicity. For example, they are less irritant and less toxic than compositions containing a glyphosate herbicide and a comparable amount of an ethoxylated fatty amine surfactant, one of the classes of surfactant(s) previously considered optimum for glyphosate enhancement.

Compared to alkyl polyglycoside surfactants known for their low eye irritancy characteristics in cosmetics for instance, and previously disclosed in liquid glyphosate compositions (see EP-A-0220 902 and EP-A-0 364 202) the formulations according to our present invention provide comparatively much reduced foaming. Our invention reduces or eliminates the need for an antifoam ingredient such as a silicone antifoam agent. Without antifoam in the formulation, the need to shake the formulation of this invention before use is reduced or eliminated. This is of especial advantage when the formulation is to be sold in large containers.

Furthermore the compositions of our invention provide stable tank mixing with other pesticides and particularly herbicides. More especially the compositions according to the invention show good compatibility with triazines.

The number of propylene oxide units of the quaternary amine symbolized in Formula (I) is preferably in the range from 2 to about 20 as the advantages of this invention are more pronounced in that range.

Herein, the term "glyphosate herbicide" means N-phonomethyl-glycine (glyphosate) and any form or derivative of glyphosate which in aqueous solution provides glyphosate anions. Suitable cations may also be present.

Examples of such suitable cations are alkali metal cations, for instance sodium and potassium, and ammonium and substituted ammonium cations. The latter include cations derived from primary or secondary amines such as isopropylamine or dimethylamine, and from diamines such as ethylenediamine.

Especially preferred as a glyphosate herbicide are water soluble salts of glyphosate such as the isopropylamine salt of glyphosate and the trimethylsulfonium salt of glyphosate. US Patent 3,799,758 dicloses salts of glyphosate useful herein. Other examples of agriculturally acceptable salts of glyphosate are aminoguanidine salts as disclosed in EP-A-0 088 180. Because glyphosate has more than one replaceable hydrogen atom, mono-and di- alkali metal salts are possible, as well as mixtures of such salts.

Especially preferred quaternary ammonium compounds for use in the present invention are those having a structure of Formula (I) wherein n has a value from 4 to 15 or n + m has a value from 3 to 14 and -PO- is isopropylene oxide.

Quaternary ammonium compounds available in practice are not necessarily pure compounds in the sense of consisting of a single molecular species. In any particular instance, several different compounds where n or n and m vary over a narrow range will normally be present, so that the values of n and n + m mentioned above are understood as average values.

The anion symbolized as X^- in Formula (I) symbolizing a quaternary ammonium compound used in a composition of the invention can be for example a halide ion, for instance chloride or bromide, phosphate, or methylsulphate, or ethylsulphate, or a a glyphosate ion. Other suitable anions include acetate, lactate, dimethyl phosphate or polyalkoxylated phosphate and the like.

Particularly good results have been obtained with quaternary ammonium compounds having the above general Formula (I) wherein R^1 and R^2 are each methyl, X^- is Cl^-, and where (i) R^3 is $EO-(PO)_m-H$ and n + m has an average value of about 6; (ii) R^3 is methyl, and n has an average value of 6.3; or (iii) R^3 is methyl and n has a value of about 8.

In the compositions of this invention, the weight ratio of glyphosate (expressed as glyphosate acid equivalent) to the quaternary ammonium compound can vary over a considerable range, for example from about 1:5 to about 10:1. The optimum ratio will vary according to the manner in which the herbicidal composition is applied, the weed species to be treated, and the particular quaternary ammonium compound selected, but is normally within the range from about 1:2 to about 4:1, for example about 1:1 or about 2:1.

A composition of the invention can be a liquid aqueous concentrate intended to be diluted with water to form a spray solution for the actual herbicidal application. A liquid concentrate will normally contain at least 50 grams glyphosate

acid equivalent per litre, and preferably at least 100 g/l. Compositions of the invention may include a significant amount of an agriculturally-acceptable inorganic ammonium salt such as ammonium sulphate, in addition to the glyphosate and quaternary ammonium compound. Liquid concentrates without such an inorganic ammonium salt can contain up to 450 g/l or more, for example 300-450 g/l, glyphosate acid equivalent when the glyphosate is present as a salt having a high solubility, for example the isopropylamine salt. In liquid concentrates containing an inorganic ammonium salt (for example ammonium sulphate in an amount of from 100 to 500 g/l), the maximum amount of glyphosate which can be accommodated is less, and may be, for example, about 150 g/l glyphosate acid equivalent.

In another form, a composition of the invention is a solid, for example a free-flowing particulate, granular solid or compressed into tablets or briquets of any desired size and shape. The term "solid" as employed herein includes granular, particular, wettable powder, water soluble and water dispersible, mixtures thereof and the like. Typically such solids are dry. Such dry compositions will usually contain not more than 5% and preferably not more than 1% by weight of water.

In a solid composition, glyphosate is preferably present as the isopropylamine salt or an alkali metal salt such as a sodium or potassium salt, or as an ammonium salt. The surfactant used in the compositions of the invention is particularly suitable for dry formulations.

Optionally solid compositions may be formulated to include a water-soluble inert carrier, and for this purpose ammonium sulphate is particularly suitable. The weight ratio of glyphosate (expressed as glyphosate acid equivalent) to quaternary ammonium compound in such compositions will be within the general range mentioned above. The amount of water-soluble inert carrier is not

critical, and in the case of ammonium sulphate, the amount may, for example, range from 20% to 80% of the total weight of the composition. Solid compositions can be made, for example by spray drying an aqueous solution of the components, by dry-blending the ingredients in conventional blending apparatus, or by extrusion blending whereby a granular product is obtained in an essentially single operation. In a preferred method, an alkali metal salt of glyphosate is prepared in situ by the method of European patent application 8790 1321.7. It has been found that by radial extrusion, a product is obtained with better dissolution characteristics in water compared to a product with frontal extrusion.

Compositions of the invention also include solutions which may be applied by spraying for example. In these solutions, the concentration of glyphosate is selected according to the volume per unit area of spray solution to be used and the desired rate of application of glyphosate per unit area. For example, conventional spraying is done at 100-600 litres of spray solution per hectare, and the rate of application of glyphosate is typically 0.125 to 1.5 kg of glyphosate acid equivalent per hectare. In controlled drop spraying, the rate of application of glyphosate per hectare will normally be in the same range, but the volume of spray solution per hectare will be considerably less, perhaps 15-50 litres per hectare. Spray solutions for controlled drop spraying are therefore more concentrated than those used in conventional spraying. In spray solutions containing inorganic ammonium salts in addition to the glyphosate and quaternary ammonium compound, the amount of inorganic ammonium salt which can be included may be, for example, up to 10 times the weight of glyphosate acid equivalent.

Spray solution compositions can be prepared by diluting liquid concentrates or dissolving solid compositions as described above, or by tank mixing the separate components of the compositions.

Compositions of the invention can optionally contain other components, preferably those which do not have an adverse effect on the resulting composition as to irritancy and toxicity. These additional components may include anti-freeze agents such as ethylene glycol, polyethylene glycols and glycerol. Other examples of additional components are dyes, thickening agents, anti-foam agents, for instance silicone-based anti-foam agents, agents suitable for pH adjustment to optimize herbicidal efficacy and stability of the composition, and certain surfactants, for instance non-ionic surfactants such as polyoxy-ethylene ethers or esters, and sugar ethers. Concentrates and solutions of the invention may also be mixed with one or more herbicides, themselves preferably having low or no irritancy and relatively low toxicity.

Some of the quaternary ammonium compounds shown in Table 1 following were used to prepare compositions in turn used in investigating their potentiating effect on the herbicidal action of glyphosate.

TABLE 1

Quaternary Amine No.	R^1	R^2	R^3	X^-	Average n + m
1	C_2H_5	C_2H_5	CH_3	Cl^-	8
2	CH_3	CH_3	-EO-$(PO)_m$-H	Cl^-	6
3	CH_3	CH_3	-EO-$(PO)_m$-H	Cl^-	8
4	CH_3	CH_3	-EO-$(PO)_m$-H	Cl^-	9.7
5	CH_3	CH_3	CH_3	Cl^-	6.3
6	CH_3	CH_3	CH_3	Cl^-	8
7	CH_3	CH_3	CH_3	CH_3SO_4-	8.4
8	CH_3	CH_3	CH_3	Cl^-	3
9	CH_3	CH_3	CH_3	Cl^-	14.2
10	C_3H_7	C_3H_7	CH_3	CH_3SO_4-	8
11	CH_3	CH_3	CH_3	CH_3COO^-	8
12	CH_3	CH_3	CH_3	$PO_4(CH_3)_2^-$	8
13	CH_3	CH_3	CH_3	Lactate	8
14	CH_3	CH_3	CH_3	Cl^-	20
15	CH_3	CH_3	CH_3	Cl^-	8
16	CH_3	C_2H_5	C_2H_5	Cl^-	7

Table 2 below shows the components and their concentrations in examples of prepared compositions of the invention which are liquid concentrates. The glyphosate is employed in the compositions as the isopropylamine salt of glyphosate, and the concentration of the isopropylamine salt glyphosate is expressed as glyphosate acid equivalent (a.e.).

TABLE 2

COMPOSITION No.	QUATERNARY AMINE No.	QUATERNARY AMINE Conc. (g/l)	GLYPHOSATE A.E. Conc. (g/l)
1	2	180	360
2	2	200	400
3	2	225	450
4	5	180	360
5	5	200	400
6	5	225	450
7	6	180	360
8	6	200	400
9	6	225	450

g/l = grams per litre.

Examples of two solid compositions of this invention are shown in Table 3 below.

TABLE 3

Components	Composition No. 10 (% by weight)	Composition No. 11 (% by weight)
Glyphosate sodium salt	24.03	39.48
Quaternary amine No. 6 (TABLE I)	10.63	17.50
Ammonium sulphate	63.79	40.80
Silicone defoamer	0.08	0.12
Water	0.50	0.50
Inert(s)	0.97	1.60

The herbicidal effectiveness of spray solutions prepared by the dilution of some of the liquid concentrates exemplified above was evaluated as follows:

For greenhouse tests, plants were grown from seed in 13 cm pots containing a natural sandy loam soil enriched with a mixture of fertilizer. All irrigation was supplied automatically from below. The environment was controlled at a temperature regime of 18°C (day), 12°C (night), a relative humidity of 65% day and 75% night, and an illuminance of 1500 microeinsteins $m^{-2}s^{-1}$ (with an artificial shading if natural light gave more, and an artificial supply if the natural light was less than 1300 microeinstein $m^{-2}s^{-1}$).

Before spraying, pots were selected for uniformity as far as possible, and atypical examples were discarded. Spray solutions were applied with a Mardrive precision laboratory sprayer, calibrated to deliver spray solution in one pass at a rate equivalent to 200 l/ha. All replicate pots (3-5 replicates per species, per treatment) were sprayed with one pass of the sprayer.

After treatment, control pots were placed at random among treated pots. Assessments of "% phytotoxicity" were made by comparison with untreated controls and with Controls* sprayed with a glyphosate solution containing an ethoxylated tallow amine surfactant in a weight ratio of 2:1 glyphosate a.e.:surfactant, on an arbitrary scale from 0 to 100%, where 0 means no visible effect and 100% means death of all plants. For any one assessment, all pots were rated by the same individual, assessments being performed "blind", without knowledge of the treatment.

Results were as follows. The results in Table 4 for % phytotoxicity represent the average of 3 rates (200, 400 and 600 g glyphosate a.e./ha) x 3 replicates, 34 days after treatment for each quaternary amine (designation from Table 1).

TABLE 4

Quaternary No.	Lolium multiflorum	Geranium molle
1	32	36
2	45	47
3	29	35
4	32	36
5	46	51
6	44	45
Control*	34	41

The results in Table 5 for the % phytotoxicity are the average of 3 rates (200, 400 and 600 g glyphosate e.e./ha) x 3 replicates, 30 days after treatment.

TABLE 5

Quaternary No.	Elymus repens
2	43
3	39
4	52
6	55
Control *	46

A field test was carried out in a sown 90 m² pasture containing a mixture of perennial ryegrass and blackgrass for the narrow leaved species (grasses) and a mixture of Sinapis arvensis Matricaria sp. and Trifolium repens broadleaves. This was sprayed at a spray volume application rate equivalent to about 400 l/ha, and at an air temperature of about 18°C. No rain was noted for 5 days after spraying. The test design was a complete randomized block design with 3 replicates of each treatment and one untreated plot per 4 treated plots. The results in Table 6 below for the % phytotoxicity are the average of 3 rates (800, 1200 and 1600 g glyphodate a.e./ha) x 3 replicates, 30 days after treatment.

TABLE 6

Quaternary No.	Grasses	Broadleaves
2	45	49
6	40	45
Control*	42	47

Compositions containing the quaternary Nos. 2, 5 and 6 of this invention in these tests (Table 4, 5 and 5) show comparable efficacy to the Control* composition therein. Thus in considering these data, these phytotoxicity tests show that Quaternaries Nos. 2, 5 and 6 enhanced the performance of glyphosate to a similar degree as did the ethoxylated fatty amine surfactant of the Control.

Standard skin and eye irritancy tests were carried out using compositions Nos. 2, 5 and 8 on New Zealand white rabbits by the procedures of OECD Guidelines for Testing of Chemicals, Tests No. 404 "Acute Dermal Irritancy/Corrosion", 12 May 1981, and 405 "Acute Eye Irritancy/Corrosion" 24 February 1987, respectively. When the results were assessed according to the criteria of European Commission Directive 83/467/EEC, all the compositions were classified as non-irritant to both dermal and ocular tissue.

The same compositions were examined for toxicity to fish by the procedure of OECD Guidelines for Testing of Chemicals, Test No. 203, 4 April 1984, "Fish Acute Toxicity Test". When the results were assessed on the Proposed Toxicity Rating Scale for Use at CNFRL (Columbia National Fisheries Research Laboratory, U.S.A.) the compositions were classified as "relatively non-toxic".

Quaternary amines designated in Table 1 as numbers 6-10 were tested in another field test in Summer. The rates of glyphosate (as glyphosate isopropylamine salt) application were equivalent to about 300, 600 and 900g a.e./ha. The application was made at about 250 l/ha using standard field spraying equipment. The ratio of glyphosate acid equivalent surfactant was about 2/1. The weed assessed is Trifolium repens (TRFRE).

The weather at the spraying was clear and hot, no wind. Temperature was around 30°C and no rain was recorded in six hours following spraying.

Assessment of per cent phytotoxicity was made by comparison with untreated control plots on an arbitrary scale from 0 to 100%, where 0 means no visible effect and 100 means death of all plants.

Randomised block design with 3 replicates was used (1 check per 5-6 treatment plots). Unit plot size = 1.5m x 3.5m.

TABLE 7

Quaternary Amine No	Glyphosate g. a.e./ha	TRFRE 32 DAT^{XXX}
Control*	300	35
	600	47
	900	53
6	300	25
	600	38
	900	55
7	300	27
	600	38
	900	50
10	300	23
	600	35
	900	47
9	300	25
	600	48
	900	50
8	300	18
	600	33
	900	45

DAT^{XXX} = days after treatment.

The quaternary amines numbered 6-10 in TABLE 2 in a series of 2/1 glyphosate acid equivalent/surfactant compositions were also tested in a further greenhouse test. The conditions were the same as previously. The rates of application were 360, 540 and 1080 g a.e./ha (glyphosate in the form of the isopropylamine salt).

Results are shown in table 8.

TABLE 8

Quaternary No.	Rate g a.e. /ha	BRSNW (DAT) 14	20	36	GERMO (DAT) 14	20	36	LOLRI (DAT) 20	36	AGGRE (DAT) 20	36
7	360	17	27	39	15	60	85	47	77	70	90
	540	23	38	58	35	70	93	92	100	75	82
	1080	47	63	78	40	75	98	83	100	77	100
10	360	30	42	42	27	65	80	55	85	60	82
	540	47	55	65	43	73	88	67	92	75	87
	1080	45	62	75	55	78	95	99	100	78	100
9	360	30	23	35	15	33	60	58	82	63	75
	540	37	55	58	50	60	75	60	93	73	93
	1080	53	78	95	53	65	78	88	100	83	100
8	360	10	20	8	10	35	75	50	85	47	73
	540	25	20	12	33	63	95	63	97	70	87
	1080	28	45	60	48	73	100	80	98	82	97
6	360	12	20	33	15	70	80	52	87	77	93
	540	30	40	52	40	65	85	83	95	85	100
	1080	37	68	83	50	70	90	83	98	82	98
Control*											
	360	17	20	37	8	35	65	62	92	65	80
	540	42	57	65	40	68	100	87	100	85	100
	1080	50	67	75	58	80	100	93	100	73	100

The assessed weeds are:

BRSNW	Brassica (oil seed rape)
GERMO	Geranium molle
LOLRI	Lolium rigidum
AGGRE	Agropyron repens (couch)

The results are shown in the previous Tables 7 and 8 indicate that the glyphosate phytotoxicity of compositions of the invention employed herein is enhanced to a level comparable to that of the Control* composition.

Compositions of the invention are particularly compatible with other herbicides and show good stability characteristics when mixed with one or more other herbicides. Also tank mix properties appeared very good.

In a compatibility test, the following formulations were used:

Composition	Ingredient	Amount of Composition Employed with Ingredient
(1)	Glyphosate + Quaternary amine (2)	$6L^{XX}$ in 400L
(7)	Glyphosate + Quaternary amine (6)	6L in 400L
GESATOPE	Simazine	6L in 400L
CENT - 7	Isoxaben	8L in 400L
SEXTAN	Isoxaben/Simazine	7.5L in 400L
VITIDOR	Oryzalin/Simazine	12L in 400L
FENICAN	Diuron/Terbuthylazine	6L in 400L
ZORIAL	Norflurazon	2.5kg in 400L
GOLTIX	70% Metamitron	5kg in 400L
PYRAMIN	65% Chloridazon	$5kg^{XXX}$ in 400L

The coherbicide was mixed with water and then composition (1) or (7) was added and the mixture thoroughly mixed. Observations were made at 1, 5, 10, 30 and 60 minutes after mixing. Final evaluation was done after two hours. No compatibility agent was added.

L^{XX} = liter

kg^{XXX} = Kilogram

COMPATIBILITY TEST

	MIXTURE No.	after 1 min.[X]	after 5 min.	after 10 min.	after 30 min.	after 60 min.	after 2 hours
5	(1) + Gesatope	OK	OK	OK	sed/red	Sed/red	Sed/red
	(1) + Cent - 7	OK	OK	OK	OK	Sed/red	Sed/red
	(1) + Sextan	OK	OK	OK	OK	Sed/red	Sed/red
	(1) + Vitidor	OK	OK	OK	OK	Sed/red	Sed/red
	(1) + Fenican	OK	OK	OK	OK	Sed/red	Sed/red
10	(1) + Zorial	OK	OK	OK	OK	Sed/red	Sed/red
	(1) + Goltix	OK	OK	OK	OK	Sed/red	Sed/red
	(1) + Pyramin	OK	OK	OK	OK	Sed/red	Sed/red
	(7) + Gesatope	OK	OK	OK	Sed/red	Sed/red	Sed/red
	(7) + Cent-7	OK	OK	OK	OK	Sed/red	Sed/red
15	(7) + Sextan	OK	OK	OK	OK	Sed/red	Sed/red
	(7) + Vitidor	OK	OK	OK	OK	Sed/red	Sed/red
	(7) + Fenican	OK	OK	OK	OK	Sed/red	Sed/red
	(7) + Zorial	OK			Tr.sed	Sed/red	Sed/red
	(7) + Goltix	OK	OK	OK	Sed/red	Sed/red	Sed/red
20	(7) + Pyramin	OK	OK	OK	OK	Sed/red	Sed/red

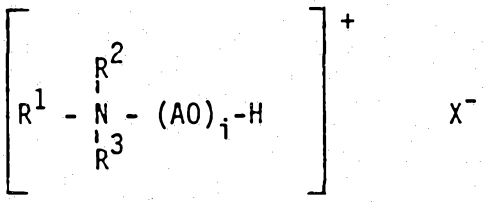
OK = No Compatibility Problems
Tr.sed = Trace of Sedimentation
Sed/red = Sedimentation but Easily Re-dispersible
min. = Minutes

25 As an advantage, compositions of this invention may contain suitable coherbicides including sulfonyl ureas depending on the intended use of the composition containing the coherbicide(s).

The matter contained in each of the following claims is to be read as part of the general description
30 of the present invention.

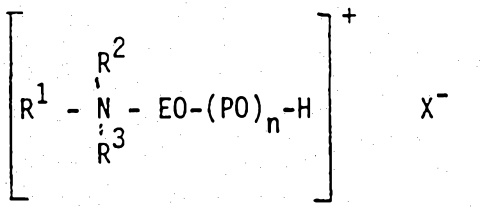
The claims defining the invention are as follows:

1. A composition comprising a glyphosate herbicide and a quaternary ammonium compound having the formula:



wherein A or each A represents an alkylene group having 2 or 3 carbon atoms, R^1 and R^2 are each independently an alkyl group having from 1 to 5 carbon atoms; R^3 is an alkyl group having from 1 to 5 carbon atoms or a group having the formula $(AO)_j$-H; i (in a compound in which R^3 is an alkyl group) or i + j (in a compound in which R^3 is a group having the formula $(AO)_j$-H) has a value of from 2 to 20; and X^- is a suitable anion.

2. A composition comprising a glyphosate herbicide and a quaternary ammonium compound the latter having the formula



wherein -EO- means an ethylene oxide radical and -PO- means a propylene oxide radical, R^1 and R^2 are each independently an alkyl group having from 1 to 3 carbon atoms; R^3 is an alkyl group having from 1 to 3 carbon atoms, a group having the formula $(EO)_m$-H or a group having the formula EO-$(PO)_m$-H; n (in a compound in which R^3 is an alkyl group) or n + m (in a compound in which R^3 is a group having the formula $(EO)_m$-H or the formula EO-$(PO)_m$-H), has a value of from about 2 to about 20; and X^- is a suitable anion.

3. A composition according to Claim 2 wherein, in the formula of the quaternary ammonium compound, -PO- means isopropylene oxide and n (in a compound in which R^3 is a methyl or ethyl group) has a value of from 4 to 15, and n + m (in a compound in which R^3 is a group having the formula $EO-(PO)_m-H$) has a value of from 3 to 14 and X^- is halide, ethylsulfate, methylsulfate, dimethylphosphate, polyalkoxyphosphate, lactate or acetate.

4. A composition according to Claim 3 wherein R^1 and R^2 are each methyl, X^- is Cl^-, and where (i) R^3 is $EO-(PO)_m-H$ and n + m has a value of about 6; or (ii) R^3 is methyl, and n has an average value of 6.3; or (iii) R^3 is methyl and n has a value of about 8.

5. A composition according to any one of Claims 1 to 4 in which the weight ratio of glyphosate (expressed as glyphosate acid equivalent) to the quaternary ammonium compound is in the range from about 1:2 to about 4:1.

6. A composition according to any one of Claims 1 to 5 in which the glyphosate is present as the isopropylamine salt or the trimethylsulfonium salt of glyphosate.

7. A composition according to any one of Claims 1 to 6 which is a liquid concentrate containing at least 100 g/l of glyphosate expressed as glyphosate acid equivalent.

8. A composition according to any one of Claims 1 to 5 in which the glyphosate is present as an alkali metal or ammonium salt.

9. A composition according to Claim 8 in which the glyphosate is present as a sodium salt.

10. A composition according to either of Claims 8 and 9 which is a solid composition.

11. A composition according to Claim 10 which includes a water-soluble inert support.

12. A composition according to Claim 11 in which the water-soluble inert support is ammonium sulphate in an amount of from 20% to 80% of the total weight of the composition.

13. A composition according to any one of Claims 1-12 which further contains anti-freeze agents and/or dyes and/or thickening agents and/or anti-foam agents and/or co-surfactants.

14. A composition according to any one of Claims 1-12 which further contains a co-herbicide selected from Simazine, Isoxaben, Oryzalin, Diuron, Terbuthylazine, Norflurazon, Metamitron, Chloridazon or sulfonyl urea in an agriculturally effective amount.

15. A composition according to any one of Claims 1-13 where an agriculturally effective amount of glyphosate is present along with an effective amount of said quaternary amine composed of said formula.

16. A composition according to any one of Claims 1 to 6, 12 and 13 which is a spray solution having a concentration such that when sprayed at 100 - 600 litres per hectare, 0.125 to 1.5 kg of glyphosate (expressed as glyphosate acid equivalent) per hectare is applied.

17. Use of a composition according to any one of Claims 1-15 as a growth regulating agent or herbicide for killing narrow leaved and/or broadleaved unwanted vegetation in a rate of 0.125 and 1.5 kg glyphosate acid equivalent per hectare.

18. A process for the preparation of a dry solid composition of Claim 1, comprising spray drying an aqueous solution of the components, or dry-blending the ingredients of the composition, or extruding the ingredients of the composition.

19. Use of a quaternary ammonium compound as shown in any one of Claims 1-16 as a tank mix adjuvant to form a herbicidal composition containing glyphosate.

20. A composition according to Claim 1, substantially as hereinbefore described.

21. A composition according to Claim 2, substantially as hereinbefore described.

22. A use according to Claim 17 or 19, substantially as hereinbefore described.

DATED this 4th day of February, 1991

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