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(54) Title: HERBICIDAL COMPOSITIONS

(57) Abstract: The invention provides an effective, sustainable, post-emergent, non-selective, contact, systemic herbicidal formula-
tion containing a surfactant, such as sodium lauryl sulfate, an oil, such as a vegetable oil, and an acid, such as dilute acetic acid or
citric acid.



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HERBICIDAL COMPOSITIONS

Field of the Invention

The invention provides an effective, sustainable, post-emergent, non-selective, contact, systemic herbicidal formulation containing a surfactant, such as sodium lauryl sulfate, an oil, such as a vegetable oil, and an organic acid, such as dilute acetic acid or citric acid.

Background of the Invention

Non-selective pre- and post-emerge herbicides containing glyphosate (RoundUp[®]), pyrimidinedione, 2-(4-chloro-2-methylphenoxy) propionic acid (Mecoprop), 1,1'-dimethyl-4,4'-bipyridylium (Paraquat), and 2-(1-methyl-*n*-propyl)-4,6-dinitrophenol (Dinoseb), glufosinate and an alkyl polyglycoside, ammonium sulfate, and alkyl polysaccharide, trifluralin (Treflan), sethoxydim (Poast), and other complex chemistry products are commonly used to eliminate unwanted broadleaf and grass species for example on fields, in gardens, in between hard or loose stone road and walkway surfaces. Though effective in killing the annual and perennial weeds after one or a few applications, many of the popular herbicides bring with proper use unwanted side effects including, but not limited to: adverse health effects in humans and animals, adverse environmental effects to surface waterways, adverse effects in invertebrates, persistence in the soil, and the development of specific chemical resistant weeds. Many of the aforementioned contact herbicides utilize a systemic mechanism to kill the undesirable plants.

To counteract the adverse health and environmental effects of some herbicides, manufacturers catering to the rising demand for herbicide treatments capable of compliance with federal and local requirements, which are generally regarded as safe, and some of which have attained certified organic status, have brought to market a variety of dilutions of

“chemical” formulations in isolation or in concert with “natural” herbicide formulations. Many such alternative formulations consist of vinegar (acetic acid), herbicidal soap, surfactants, essential oils, citric acid, and combinations thereof (see, e.g., US Patents: 7,226,892; 7,094,735). It is the aim of alternative herbicide producers to earn USEPA exempt status for their product preparation and labeling (Federal Register: March 6, 1996 (Volume 61, Number 45); Rules and Regulations page 8876-8879), or registered Reduced Risk status. EPA-exemption is of special interest to small and large organic farmers, landscapers, and nurserymen whose customer base demand holistic landscape treatment. Some Reduced Risk products are also eligible for Certified Organic registration.

Reports via fact sheet and articles on vinegar-based herbicides by various State Extension Service faculties conclude that the effective concentration for acetic acid or vinegar in such herbicide is at or above 20%, which is available to the professional agricultural market (North Dakota Pesticide Quarterly, Vol. 21, No. 1, Jan. 2003 [<http://www.ext.nodak.edu/extnews/pestqtrly/pq21-1-c.htm>]). Application of a 20% acetic acid solution, however, carries with it increased risk of contact irritation of eyes, skin, and respiratory mucus membranes. In order to comply with the USEPA exemption list of active and inert ingredients for herbicides, manufacturers have had to keep their concentration of inert ingredients such as acetic acid below 8%. Acetic acid at 8% and lower is only listed by the EPA as an inert ingredient, and is not on the active ingredient list. However, acetic acid, at any concentration, has been deemed an active herbicidal ingredient. It is therefore ineligible as a herbicide ingredient for EPA-exempt status. Acetic acid-based herbicides, regardless of how dilute, must be registered with the EPA. These less or non toxic formulations work on contact with the plant, acting as a desiccant to wither the top growth, depriving the plant of access to nutrients including water and its photosynthetic capability.

On their own vinegar, citric acid, or soaps do not employ a systemic mode of action, however, annual plants and young perennials not too entrenched with deep root systems will die after one or two applications due to top growth kill. These products have limited appeal for general weed reduction maintenance. To enhance the effectiveness of eradicating herbal vegetation regardless of the product seasonality plays a part in the elimination success rate. Scheduling the application of herbicide to specific species needs to coincide with the appropriate season and conditions for its optimal susceptibility (<http://www.extension.umn.edu/distribution/horticulture/dg1137.html>).

Thus, existing herbicides suffer from a variety of disadvantages, including environmental harm and irritation risks associated with exposure thereto. The art is in need of improved herbicides which overcome the disadvantages of current herbicides, whether FIFRA 25b eligible, or as a registered Reduced Risk product.

Brief Summary of the Invention

The inventor has improved upon previous herbicide formulations to prevent environmental perturbation and/or hazards caused when using strong chemicals to kill unwanted herbaceous and ligneous plants. The present invention successfully addresses several aspects of sustainable improvement.

In one aspect all ingredients both active and inert are consistent with the parameters prescribed by the approved US EPA FIFRA 25b exemption list, such as in a citric acid formulation and others eligible as registered Reduced Risk, containing herbicidal substances generally regarded as safe (GRAS) when used repeatedly, as directed, and exposed to the environment.

Surprisingly, the inventor hereof has found that the acid component of an herbicidal formulation need not be as high as current formulations use. In another aspect of the invention, then, the present formulation reduces the ready-to-use concentration of the inert ingredient such as acetic acid from the 8-20% used by other brands deemed necessary to get a rapid plant kill. One aspect of the invention uses less acetic acid, such as 5% or even less. This reduction complies with EPA-Reduced Risk guidelines for EPA-registered herbicide formulations. In another aspect of the invention, the herbicidal formulation uses a solution based on citric acid at between about 20%, up to a saturated citric acid solution (60%). These concentrations comply with FIFRA 25b EPA-exempt active ingredient restrictions.

In another aspect of the invention a systemic mode of action is employed. As part of an emulsion of the ingredients the oil component acts to ease the highly polar formula past the protective lipid outer tissue layer of the plant leaf. The oil also acts to facilitate persistence of the product on the leaf surface. Once past the exterior often waxy layer of the plant tissue the highly aqueous product containing surfactant in the form of sodium lauryl sulfate utilizes the plant's nutrient transport system to deliver the destructive agent deep into the plant where it denatures the proteins and disables protein synthesis (Plant Science letters, 6 [1976] 237-242; Plant Physiol. [1986] 81, 802-8060032-0889/86/8 1/0802/05/\$0 1.00/0). Further, the organic acid continues the desiccation of the plant as it takes up acidic water from the formula in its effort to recover. This mode of action does not only rely on top growth wither, but rather capitalizes on subsequent protein synthesis disruption for total plant kill. As with other herbicide preparations deeply rooted perennial grass and broadleaf species might require follow up applications after a week as the plant sends up weak new shoots in an effort to recover its nutrient supply.

Another aspect of the invention is that, due to its low concentration of potentially irritating components in ready to use form, application of a composition of the invention in the field requires no extraordinary OSHA-certified protection for individuals handling the herbicide in most states in the US.

Thus, in one aspect, the invention provides an herbicidal composition comprising an oil, an acid, and a surfactant, wherein the acid, such as acetic acid, is present at less than 7%, or less than 6%, or less than 5% by weight. In an embodiment employing citric acid, the citric acid may be present at saturation concentration for best effectiveness, that is, at 60%, while lower concentrations down to about 20% still provide effective herbicidal characteristics. The oil may be a vegetable oil, such as castor oil or others as described below, and the acid may be acetic acid or citric acid or other organic acids. In one aspect, the surfactant is sodium dodecyl sulfate, otherwise known as sodium lauryl sulfate (SLS).

In another aspect, the oil is castor oil, the acid is acetic acid, and the surfactant is sodium lauryl sulfate.

In another aspect, the oil is present at between 0.5-1.5% by weight, the acetic acid is present at between 4.0-6.0% by weight, and the sodium lauryl sulfate is present at between 0.01-1.0% by weight.

In another aspect, the oil is present at 1.0% by weight, the acetic acid is present at 5.0% by weight, and the sodium lauryl sulfate is present at 0.02% by weight.

In yet another aspect, the invention provides a concentrated herbicidal composition which upon dilution with water yields an herbicidal composition as described above.

In another aspect, the invention provides a method of controlling or eliminating weeds, in which an effective amount of any previously described aspect is applied to such weeds.

These and additional features of the invention are exemplified and further described in the Detailed Description of the Invention below.

Detailed Description of the Invention

The invention generally provides an herbicidal formulation comprising an oil, a dilute organic acid, and a surfactant. An herbicide of the invention utilizes the synergistic effects of dilute organic acid such as acetic or higher concentrations of citric acid, and an oil such as a vegetable or mineral oil, with the protein disruptive ability of sodium dodecyl sulfate, also known as sodium lauryl sulfate, to formulate a sustainable, systemic, contact herbicide capable of controlling broadleaf and grass weeds. No additional herbicidal components are necessary, that is, the herbicide of the invention may be free of other herbicidal components.

Alone, the individual components of the invention formula, in specified concentration, do not generally exhibit more than top-kill herbicidal properties. However, when combined as taught herein into a dilute acetic acid or higher citric acid concentration, the components act synergistically as a systemic contact herbicide capable of eradicating herbaceous vegetation.

The Oil Component

One component of the composition of the invention is an oil, which may be any vegetable or mineral oil; however, corn, castor, or linseed oils are generally preferred in order to meet

EPA FIFRA 25b exemption. As the exemption status of other oils changes, such newly exempted oils are also amenable to use in the invention. The herbicide of the invention may contain a single oil or a combination of such oils, so long as the total oil in the ready-to-use formulation is between about 0.5% and about 5%, or between about 1.0% and about 3.0%, by volume. Higher concentrations of oil tend to interfere with a typical spray applicator's ability to effectively spray. Many other oils are suitably adapted for use in the invention, including such oils as: castor oil, cedar oil, cinnamon oil, citronella oil, clove oil, corn oil, cottonseed oil, garlic oil, geranium oil, lemongrass oil, linseed oil, mint oil, peppermint oil, rosemary oil, sesame oil, soybean oil, and thyme oil.

The Acid Component

An herbicide of the invention also contains an acid component, generally an organic acid component. While acetic acid and citric acid are preferred for their GRAS status, other organic acids with similar status are suitable for the invention. An acetic acid or citric acid inert ingredient can be derived from, for example, household table vinegar, which is commonly available at a concentration of 5% acetic acid, or it can be derived by diluting glacial acetic acid, concentrated citrus fruit juice, or dry citric acid to appropriate concentration in water; generally the acetic acid concentration prior to mixing in the final formulation is approximately 5% by weight (the overall weight percentage in the final formulation therefore being lowered as it is mixed with the other components of the formulation). Similarly, citric acid may be employed at saturation concentration prior to mixing into the final formulation, or may simply be added to the other ingredients. Alternatively, the acid may be added in higher strength to the other components of the

composition, in which case water is added as another component to bring the final concentration of the acid to working, ready-to-use strength. Thus, the particular concentration of the acid component prior to mixing into the final formulation is chosen to achieve the desired concentration in the final formulation, and water is added as necessary to achieve such concentration.

The final concentration of acetic acid in a ready-to-use formulation of the herbicide of the invention ranges from about 3.5 - 7.5%, or between about 4.5 - 6%, or about 5%. For embodiments of the invention using citric acid, the final concentration in a ready-to-use formulation ranges from about 20-60%. Those of skill in the art will appreciate the wide variety of organic acids suitable for use in the invention, along with their optimal concentrations. But notwithstanding the choice of organic acid, the remaining components are free of other herbicidal ingredients; rather, the herbicide of the invention relies on the synergistic effect of the organic acid, oil, and surfactant components. However, those of skill in the art will appreciate that other components, such as fragrances, emulsifiers, stabilizers, etc., may be included in the formulations.

The Surfactant Component

The herbicidal formulation of the invention also contains a surfactant component. The surfactant sodium dodecyl sulfate, also known as sodium lauryl sulfate (SLS), a common ingredient in household products such as soaps and toothpaste, is one preferred surfactant, for its GRAS status (and on the FIFRA 25b exemption list), as are others with similar status at desirable quantities. The surfactant ingredient in the herbicide formula is generally present in quantities of between 0.005 and 1 %, and generally less than 1% by weight, optionally less

than 0.5%, less than 0.1%, less than 0.05%, less than 0.03%, less than 0.02%, or between about 0.01-0.02%. However, other similarly structured surfactants can be substituted. Those of skill in the art will appreciate the wide variety of surfactants (anionic, cationic, zwitterionic, hydrophobic, hydrophilic, and amphiphilic) suitable for use in the invention.

In an embodiment using acetic acid, the three components (plus water, where necessary to achieve the concentrations as shown below) of the invention combined as described herein are not exempted from registration requirements by the USEPA for pesticide use, but are compliant with EPA-registered Reduced Risk status, are generally regarded as safe, and as such are considered safe when applied as a ready-to-use product around humans, animals, the soil, and surface water ways.

One example recipe utilizes 0.04 L of castor oil, 0.6 g of sodium lauryl sulfate (SLS), and 3.745 L of a 5% acetic acid solution, yielding 3.785 L of working, ready-to-use herbicide composition. As those of skill in the art will appreciate, the 3.745 L of 5% acetic acid solution may be added directly to the other components, or about 0.187 L of glacial acetic acid and 3.558 L of water could be combined to make 3.745 L of a 5% solution.

Alternatively, 0.187 L of glacial acetic acid can be combined with the other components of the composition, and 3.745 L of water added to the combination to bring the total volume to about 3.785 L.

For commercial purposes, the composition of the invention may be provided as a prepared ready-to-use formula. Alternatively, a concentrate to be diluted by an end-user can be made available to customers. For industrial/commercial/agricultural customers such as landscapers, farmers, and small farm or suburban development customers, a concentrate form of the invention may be more desirable for transport and economy. A concentrated form of the

composition of the invention is therefore also contemplated. The concentrate comprises sufficient oil, acid, surfactant, and water components such that dilution of the concentrate with water yields the working, ready-to-use strength composition. For example, concentrated herbicide may be, for example, 2x, 5x, 10x, or 15x strengths. In some embodiments of the invention, the composition may be a 1x concentration (ready-to-use) or a concentrated strength from about 1.5x to about 18x.

A 10x concentrate, for example, may be prepared by combining 0.4 L castor oil, 6.0 g SLS, 1.87 L of glacial acetic acid, and 1.515 L of water, for a total volume of about 3.785 L. This 10x concentrate is capable of being diluted, 1 part concentrate plus 9 parts water, to yield as much as 37.85 L of ready-to-use formulation. Smaller quantities may be prepared as desired, for example, 0.25 L of 10x concentrate may be mixed with 2.25 L of water to prepare 2.5 L of ready-to-use composition, a comfortable quantity for application use in a gallon size spray bottle.

Table 1 below, in Example 3 below, illustrates several strengths and concentrates of the compositions of the invention.

In light of the foaming capacity of the surfactant it is recommended that where a concentrate is used the water diluents should first be filled into the dispensing mechanism, topped off by the appropriate aliquot of concentrate. Defoamers are not necessary where proper filling instructions are followed.

In an embodiment of the invention using citric acid as the organic acid component, the herbicidal composition in a ready-to-use formulation comprises citric acid at between 20-60%; the oil component at between 1.0 and 3.0%; and the surfactant component at between about 0.005 and 1 %, and generally less than 1% by weight, optionally less than 0.5%, less

than 0.1%, less than 0.05%, or between about 0.01-0.04%. Example 4 provides one example of such a citric acid based herbicidal composition of the invention, comprising in a total volume of 3.785 L the following components: 0.12 L castor oil; 1.8 g SLS; 2271 g citric acid; 2.612 L water.

The invention is further exemplified by the following illustrative and non-limiting Examples.

Examples

Example 1: A ready-to-use herbicidal composition

A ready-to-use herbicidal composition of the invention was prepared by combining the following components in a container, as measured by weight:

castor oil	0.040L
sodium lauryl sulfate	0.6g
5% acetic acid solution	3.745L
Total	3.785L

The final concentration of acetic acid is thus slightly less than 5.0%, while the oil component is slightly greater than 1.0%. The weight percent of the SLS surfactant is about 0.02%. The composition is easily applied to a variety of plant growths in a wide variety of terrains, using common application devices.

Example 2: Method of use of the composition of Example 1

The herbicidal composition of Example 1 was sprayed on a 10 x 100 ft. plot of combined and integrated grass and assorted broadleaf growth, in dry sunny conditions, where daytime temperatures ranged from 25-30°C, and ground moisture ranged between 20 and 27%. At 1100h growth was sprayed directly onto the leaves of the plants so as to wet the leaves but not saturate the ground. The plot was left to dry, which occurred within fifteen minutes of application. Within two hours, broadleaves curled, and all growth began to brown. By 1500h the next day (i.e., 28 hours post-application) growth was brown and dry showing no signs of green health.

Example 3: Concentrated formulations requiring dilution prior to use

A 10x strength concentrated herbicidal composition of the invention is prepared by combining the following components in a container:

castor oil	0.40L
sodium lauryl sulfate	6.0g
glacial acetic acid	1.87L
water	1.515L
Total	3.785L

Upon ten-fold dilution with water (one part concentrate plus 9 parts water), the composition is at ready-to-use strength, equivalent to the same strength as the composition prepared in Example 1. Table 1 below illustrates recipes for preparing several useful concentrate strengths, as well as the 1x ready-to-use strength.

Table 1

	ready to use (1x)	ready to use (1x)	2x	5x	10x	15x
castor oil	0.040 L	0.040 L	0.080 L	0.200 L	0.400 L	0.600 L
SLS	0.6 g	0.6 g	1.2 g	3.0 g	6.0 g	9.0 g
acetic acid	3.745 L (of 5% sol'n)	0.187 L (glacial)	0.375 L (glacial)	0.935 L (glacial)	1.87 L (glacial)	2.8 L (glacial)
water	-0-	3.558 L	3.33 L	2.65 L	1.515 L	0.385 L
total	3.785 L	3.785 L	3.785 L	3.785 L	3.785 L	3.785 L

Example 4: Herbicidal formulation with citric acid

A ready-to-use herbicidal composition of the invention was prepared by combining the following components in a container, as measured by weight or volume as appropriate:

castor oil	0.12L
sodium lauryl sulfate	1.8g
citric acid	2271g
water	2.612 L
Total	3.785L

The final concentration of citric acid is 60%, while the oil component is slightly greater than 3.0% by volume. The SLS component is about 0.04% by weight. The composition is easily applied as a ready-to-use formulation to a variety of plant growths in a wide variety of terrains, using common application devices. This formulation may also be diluted with water

at 1 part concentrate to between 1 and 2 parts water, which diluted formulations are also effective.

It will be apparent to persons skilled in the art that numerous enhancements and modifications can be made to the above described apparatus without departing from the basic inventive concepts. All such modifications and enhancements are considered to be within the scope of the present invention, the nature of which is to be determined from the foregoing description and the appended claims. Furthermore, the preceding Examples are provided for illustrative purposes only, and not intended to limit the scope of the invention. All references cited are incorporated herein in their entirety.

I claim:

1. An herbicidal composition comprising an oil, an organic acid, and a surfactant, wherein the acid is present at less than 7% by weight.
2. A composition of claim 1, wherein the oil is castor oil.
3. A composition of claim 1, wherein the acid is acetic acid.
4. A composition of claim 1, wherein the surfactant is sodium lauryl sulfate.
5. A composition of claim 1, wherein the oil is castor oil, the acid is acetic acid, and the surfactant is sodium lauryl sulfate.
6. A composition of claim 5, wherein the oil is present at between 0.5-1.5% by volume, the acetic acid is present at between 4.0-6.0% by weight, and the sodium lauryl sulfate is present at between 0.01-1.0% by weight.
7. A composition of claim 6, wherein the oil is present at 1.0% by volume, the acetic acid is present at 5.0% by weight, and the sodium lauryl sulfate is present at 0.02% by weight.
8. A concentrated herbicidal composition which upon dilution with water is an herbicidal composition of claim 1.
9. An herbicidal composition comprising an oil, citric acid, and a surfactant, wherein the citric acid is present at 20-60% by weight.
10. A composition of claim 9, wherein the oil is castor oil.

11. A composition of claim 9, wherein the surfactant is sodium lauryl sulfate.
12. A composition of claim 9, wherein the oil is castor oil, and the surfactant is sodium lauryl sulfate.
13. A composition of claim 12, wherein the oil is present at between 0.5-5.0% by volume, and the sodium lauryl sulfate is present at between 0.01-1.0% by weight.
14. A composition of claim 13 wherein the oil is present at 3.0% by volume, the citric acid is present at 60.0% by weight, and the sodium lauryl sulfate is present at 0.04% by weight.
15. A concentrated herbicidal composition which upon dilution with water is an herbicidal composition of claim 1.
16. A method of controlling or eliminating weeds, comprising the step of administering an effective amount of a composition of any of claims 1-8.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 12/24793

A. CLASSIFICATION OF SUBJECT MATTER

IPC(8) - A01N 37/00 (2012.01)

USPC - 504/320

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC(8) - A01N 37/00 (2012.01)

USPC - 504/320

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
USPC - 504/320,357 (see search terms below)

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

PubWEST (PGPB,USPT,USOC,EPAB,JPAB,DWPI,TDBD); Google

Search Terms: herbicidal, acid, acetic, citric, oil, castor, surfactant, sodium lauryl sulfate

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X --- Y	US 2009/0099022 A1 (Fernandez et al.) 16 April 2009 (16.04.2009) entire document, especially para [0011]-[0018], [0028]-[0033]	1-13 and 15-16 ----- 14
Y	US 2002/0039970 A1 (Roberts et al.) 04 April 2002 (04.04.2002) entire document, especially para [0017], [0036]	14
A	US 2006/0205596 A1 (Deckwer et al.) 14 September 2006 (14.09.2006) entire document.	1-16
A	US 2003/0211945 A1 (Lewis) 13 November 2003 (13.11.2003) entire document.	1-16
A	US 2003/0148889 A1 (Herold et al.) 07 August 2003 (07.08.2003) entire document.	1-16
A	US 5,888,938 A (Lojek) 30 March 1999 (30.03.1999) entire document.	1-16

☐ Further documents are listed in the continuation of Box C.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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Date of the actual completion of the international search

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Date of mailing of the international search report

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