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(54) METHOD AND COMPOSITIONS FOR REDUCING OIL POLLUTION ON WATER

(71) We, SNAMPROGETTI S.p.A., an Italian company, of Corso Venezia, 16, Milan, Italy, 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 compositions for, and methods of, reducing oil pollution on water, which can be fresh water or sea water. The oil pollution can be, for example, crude oil or a petroleum product comprising hydrocarbon(s).

In the Complete Specification of our British Patent Application No. 15778/74 (Serial No. 1 461 577) there are described and claimed a particle capable of floating on fresh water and comprising a body consisting of or containing one or more water-soluble compounds containing nitrogen and phosphorus in a form assimilable by aqueous microbes, and, on surface regions of the body, a film of a paraffin which is solid at room temperature; and a method of reducing oil pollution on the surface of water which contains microbes capable of assimilating the oil, which method comprises distributing over the polluted surface particles as specified above. The Complete Specification of Application No. 15778/74 (Serial No. 1 461 577) gives various examples of said water-soluble compound, one such compound being magnesium ammonium phosphate, which obviously contains both nitrogen and phosphorous.

In the Complete Specification of our British Patent Application No. 50375/76 (Serial No. 1 542 154) there is described and claimed a composition comprising particles capable of floating on fresh water and comprising a body consisting of or containing one or more water-soluble compounds containing nitrogen and phosphorus in a form assimilable by aqueous microbes capable of metabolizing hydrocarbons, and, on surface regions of the body, a film of a paraffin which is solid at room temperature, the composition also including at least one lypophilic material comprising a compound which has a poor solubility in water and contains nitrogen assimiliable by aqueous microbes, the poorly soluble compound being capable by itself of floating on fresh water or being treated with a paraffin which is solid at room temperature to make that compound capable of floating on fresh water.

The term "lypophilic" means the ability to increase the affinity between the surface of the particle and the crude oil.

In one embodiment of the invention described and claimed in British Patent Application No. 50375/76 (Serial No. 1542154), the lypophilic material is intimately mixed with said water-soluble compound(s) containing nitrogen and phosphorus, and both or all of the material and compound(s) coated with the paraffin film, whereas, in an alternative embodiment, the lypophilic material can be separate from said water soluble compound(s) which is/are coated with paraffin film, the lypophilic material being uncoated or coated with paraffin depending on its ability to float on fresh water.

The inclusion of the compounds which contain nitrogen and are poorly soluble in water, which can be regarded as slow-release nitrogen, which nitrogen can be assimilated by microbes, can give (P + N)-to-oil ratios which are more favourable to biological degradation, and a considerable saving of magnesium ammonium phosphate can be achieved. Such

compounds, which are referred to herein as lypophilic compounds, and which are well suited to this purpose, belong to the chemical class of the ureidic derivatives of aldehydes, which are easy to prepare and, moreover, are generally capable of floating on fresh water by themselves whenever the aldehydes concerned contain four or more carbon atoms.

As indicated in British Patent Application No. 50375/76 (Serial No. 1542154), the use of a material having a low specific gravity in the slow-release- nitrogen-containing formulation, can make possible a considerable reduction in the quantity of paraffin required.

Paraffins act in such a case as bonding agents and lypophilic agents, at least part of the floatability being provided by the low-specific-gravity materials. Examples of low-specific-gravity materials include cork granulates, cork dust, sawdust, maize cob dust (common scraps), or expanded silicates such as pumice, expanded riolite, and vermiculate. In addition, in the case of the use of ureidic derivatives of high aldehydes (i.e. those with at least four carbon atoms), such derivatives can replace partially or even totally the paraffins and also the low-specific-gravity materials, inasmuch as, owing to their physical properties, they can be used to coat a slightly water-soluble phosphate, such as for example magnesium ammonium phosphate or calcium phosphate, to obtain a floating and lypophilic granulate.

The compositions disclosed in British Patent Application No. 50375/76 (Serial No. 1 542 154) can be stored for a long time, are not toxic, can easily be shipped to the place of their intended use, and can easily spread over a polluted zone so that pollution caused by hydrocarbons and crude petroleum may be destroyed.

The nutrients in the compositions disclosed in British Patent Application No. 50375/76 (Serial No. 1 542 154) accelerate a natural biological degradation of the pollutants without disturbing the ecological equilibrium. On completion of their use no residues need remain; when porous bodies are present, the residues are generally materials which already exist in the natural environment.

The composition disclosed in British Patent Application No. 50375/76 (Serial No. 1 542 154) can also include a non-ionic dispersant. The latter can display an action which is synergistic with the action of the nutrients, inasmuch as they improve the distribution of the nutrients in the oily phase. It has been observed, moreover, that, in a few cases, improved biological degradation can be obtained by including in the composition a freeze-dried hydrocarbon-oxidizing microorganism.

It has now been ascertained, and this is the basis of part of one aspect of the present invention, that lecithin, a phosphatide ester occurring in many natural products of animal or vegetable natural, can efficiently replace both the phosphorus source and the dispersant of the compositions described in British Patent Application No. 50375/76 (Serial No. 1 542 154). Lecithin can be assimilated by the hydrocarbon-oxidizing microbes which occur naturally in sea water, as a phosphorus source.

In addition, inasmuch as lecithin is endowed of prominent emulsifying properties which are attributable to the presence of hydrophilic and lypophilic functional groups, it can replace, either wholly or in part, the dispersants.

In addition, it has also been found, and this is the basis of another part of the same aspect of the present invention, that the phosphatides in general (which include lecithin), both the synthetic phosphatides and those naturally occurring in many products of both animal and vegetable origin, can constitute both the phosphorus source and the dispersant.

It has also been found, and this is the basis of an aspect of the invention claimed in our British Patent Application No. 7901890 (Serial No. 1 582 966), which is a divisional application of the present application, that hydantoins and amides, in addition to the ureidic derivatives of aldehyde mentioned above, can advantageously be used as nitrogen sources which can be assimilated by the hydrocarbon-oxidizing microbes.

It has also been found, and this is the basis of another aspect of the invention claimed in our divisional British Patent Application No. 7901890 (Serial No. 1 582 966), that there can be used with advantage as nitrogen sources assimilable by the hydrocarbon-oxidizing microbes, in addition to hydantoins and amides, compounds selected from the allophanates, polyamines, acyl ureas and the esters of hydantoic and allantoic acids.

Thus one aspect of the invention claimed in our divisional British Patent Application No. 7901890 (Serial No. 1 582 966) provides, in a method of reducing pollution caused by hydrocarbonaceous material on water which contains microbes capable of metabolizing hydrocarbons, which method comprises contacting the polluted water with a compound containing phosphorus and a compound having poor solubility in water and containing nitrogen in a form assimilable by the aqueous microbes, the use as the nitrogen-containing compound of at least one compound selected from hydantoins, amides, allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids.

One aspect of the present invention provides, in a method of reducing pollution caused by hydrocarbonaceous material on water which contains microbes capable of metabolizing hydrocarbons, which method comprises contacting the polluted water with a compoud

containing phosphorus and a compound having a poor solubility in water and containing nitrogen in a form assimilable by the aqueous microbes, the use as the phosphorus-containing compound of at least one compound selected from lecithin and other phosphatides, both synthetic and naturally occurring, and the use as the nitrogen-containing compound of (i) at least one compound selected from hydantoins, amides, allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids, or (ii) a mixture of (a) at least one compound selected from hydantoins, amides, allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids, and (b) at least one ureidic derivative of an aldehyde.

Another aspect of the present invention provides a composition containing phosphorus and nitrogen in a form assimilable by aqueous microbes capable of metabolizing hydrocarbons which cause pollution on water, the composition being intended to reduce pollution and comprising, as a phosphorus-containing compound, lecithin and/or at least one other phosphatide; and, as a nitrogen-containing compound, either (i) at least one compound selected from hydantoins, amides, allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids, or (ii) a mixture of (a) at least one compound selected from hydantoins, amides, allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids, and (b) at least one ureidic derivative of an aldehyde.

While the fertilizing action of the ureidic derivatives of aldehydes is subordinate to a chemical hydrolysis, the fertilizing action of hydantoins and amides acts by enzymic hydrolysis as brought about by the hydrocarbon-oxidizing microbes themselves.

Now, while the chemical hydrolysis of the ureidic derivatives of aldehydes is governed by the physico-chemical factors of water, especially the temperature and pH value, environmental situations may arise, in which urea is set free either in excess of, or insufficient for, the nutritional requirements of the hydrocarbon-oxidizing micro-organisms: in the former case a slowdown of the biological degradation is brought about and, in the latter case, urea is inadequately utilized and this is economically wasteful.

This drawback is not experienced when using hydantoins and amides since the nitrogen contained therein is generally set free, and consumed directly by the microbes consistently with their actual demand.

Another aspect of the invention claimed in our divisional British Patent Application No. 7901890 (Serial No. 1582966) provides, in a method of reducing pollution caused by hydrocarbonaceous material on water which contains microbes capable of metabolizing hydrocarbons, which method comprises contacting the polluted water with a compound containing phosphorus and compounds having poor solubility in water and containing nitrogen in a form assimilable by the aqueous microbes, the use as the nitrogen-containing compounds of a mixture of (a) at least one compound selected from hydantoins, amides, allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids and (b) at least one ureidic derivative of an aldehyde.

It has been found, moreover, that a considerable increase of the biological degradation velocity of the hydrocarbons is obtained when a hydantoin or amide is employed concurrently with lecithin. Thus one embodiment of the present invention provides, in a method of reducing pollution caused by hydrocarbonaceous material on water which contains microbes capable of metabolizing hydrocarbons, which method comprises contacting the polluted water with a compound containing phosphorus and a compound having poor solubility in water and containing nitrogen in a form assimilable by the aqueous microbes, the use of lecithin as the phosphorus-containing compound and the use as the nitrogen-containing compound(s) of at least one compound selected from hydantoins and amides.

A further embodiment of the present invention provides a composition containing phosphorus and nitrogen in a form assimilable by aqueous microbes capable of metabolizing hydrocarbons which cause pollution on water, the composition being intended to reduce pollution and comprising lecithin and at least one compound selected from hydantoins and amides. In this composition, as well as the corresponding method, the nitrogen source can be supplemented with a ureidic derivative of an aldehyde.

It has been found, and this is the basis of the present invention, that a good increase of the biological degradation velocity of hydrocarbons is achieved when one or more of hydantoins and amides, or one or more of allophantes, polyamines, acyl ureas and esters of hydantoic and allantoic acids, is employed together with a phosphatide, either synthetic or naturally occurring.

Thus further embodiments of the present invention provide, in a method of reducing pollution caused by hydrocarbonaceous material on water which contains microbes capable of metabolizing hydrocarbons, which method comprises contacting the polluted water with a compound containing phosphorus and a compound having poor solubility in water and containing nitrogen in a form assimilable by the aqueous microbes, the use, as a phosphorus-containing compound, of at least one phosphatide which may be synthetic or naturally occurring, and the use, as the nitrogen-containing compound, of at least one compound

selected from allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids; as well as a composition containing phosphorus and nitrogen in a form assimilable by aqueous microbes capable of metabolizing hydrocarbons which cause pollution on water, the composition being intended to reduce pollution and comprising at least one phosphatide and at least one compound selected from allophanates, polyamines, acyl ureas and esters of the hydantoic and allantoic acids. Preferably the nitrogen source is supplemented by a ureidic derivative of an aldehyde.

The compositions according to the present invention are easy to use, and do not require any special implementation, beyond what has been used hitherto when using dispersants in cases of hydrocarbon pollution.

The substances which can be employed according to the method of the present invention can be stored indefinitely, are not toxic, can easily be shipped to the place of use, can readily be spread over the polluted areas and reduce the pollution of fresh and sea water bodies by crude oil petroleum products and their derivatives.

The employed substances accelerate a natural process of biological degradation of the pollutants, without interfering adversely with the ecological equilibria.

As with the earlier compositions, there can be employed paraffination, low specific gravity materials or freeze-dried microbes. The following Examples illustrate the present invention.

EXAMPLE 1

Four sets of 250 ml Erlenmeyer flasks with ground-glass necks which each contained 100 ml of unsterilized sea water and 100 milligrams of Basra crude oil were treated as follows. To each flask of the first set (a) were added 5 milligrams of soybean lecithin and 5 milligrams of urea; to each flask of the second set (b) were added 0.84 milligram of K_2HPO_4 and 5 milligrams of urea; to each flask of the third set (c) were added 5 milligrams of urea; and no addition was made to any of the flasks of the fourth set (d) which served as a control. The sets (b) and (c) were employed for comparative reasons.

The flasks were incubated at 25°C with rotary stirring (100 rpm, and an eccentricity of 5 centimetres). At the start and every 5 days the residue of crude oil was determined by extraction with CCl_4 according to the method described in British Patent Application No. 50375/76 (Serial No. 1 542 154).

The results which were obtained are illustrated graphically in Figure 1 of the accompanying drawings, in which the ordinates represent the residual crude oil expressed as a percentage of the original value, and the abscissae represent the time in days.

EXAMPLE 2

To the flask of five sets, (a) to (e), of 250 ml Erlenmeyer flasks having ground-glass necks and each containing 100 ml of unsterilized sea water and 100 milligrams of Basra crude oil, the following additions were made:-

to set (a), 5 milligrams of soybean lecithin and 6.5 milligrams of oxamide;

to set (b), 5 milligrams of soybean lecithin and 13 milligrams of D-L-5-phenylhydantoin;

to set (c), 5 milligrams of soybean lecithin and 10 milligrams of the condensation product of isovaleric aldehyde and urea (obtained as described in the following Example 3);

to set (d), 5 milligrams of soybean lecithin and 15 milligrams of the condensation product of 3-phenylpropionic aldehyde and urea, as obtained under the same conditions adopted for isovaleric aldehyde; and

to set (e), which served as a control, no addition.

The flasks were incubated and the residual hydrocarbons were extracted as described in Example 1.

The results which were obtained are set out in the following Table 1, which specifies the residual oil expressed as a percentage of the original value.

TABLE 1

Flask set	Incubation time, days 7	*12*	*19*	*25*
(a)	63.99	60.47	42.26	36.62
(b)	67.23	61.65	47.81	38.28
(c)	63.74	54.43	44.91	40.06
(d)	59.78	54.85	47.79	43.17
(e)	69.12	67.55	66.79	65.92

EXAMPLE 3

To the flasks of three sets, (a) to (c), of 250 ml Erlenmeyer flasks with ground-glass necks and each containing 100 ml of unsterilized sea water and 100 milligrams of Basra crude oil, the following additions were made:-

to set (a), 5 milligrams of soybean lecithin, 5 milligrams of an emulsifier (2,2'-hydroxydiethyloleylamide) and 10 milligrams of the condensation product of isovaleric aldehyde and urea;

to set (b), same additives as for set (a) but without the emulsifier; and

to set (c), which served as a control, no addition.

The results which were obtained are illustrated graphically in Figure 2 of the accompanying drawings, in which the ordinates represent the residual crude oil (as a percentage of the original value) and the abscissae represent the time in days.

The condensation product of aldehyde and urea was obtained in the following way: to 20 ml of an aqueous solution of urea containing 25 grams of urea per decilitre of solution, was added 1 ml glacial acetic acid and 2 ml of isovaleric aldehyde; the mixture was stirred at room temperature for three hours. The precipitate which resulted was collected on a filter paper, washed with water and dried in a vacuum oven at 40°C for 24 hours.

EXAMPLE 4

To the flasks of five sets, (a) to (e), of 250 ml Erlenmeyer flasks having ground-glass necks and each containing 100 ml of unsterilized sea water and 100 ml of Basra crude oil, the following additions were made:-

to set (a), 5 milligrams of soybean lecithin and 5 milligrams of urea;

to set (b), 5 milligrams of dihexadecanoylphosphatidyl ethanolamine and 5 milligrams of urea;

to set (c), 0.84 milligram of K_2HPO_4 and 5 milligrams of urea;

to set (d), 5 milligrams of urea; and

to set (e), which served as a control, no addition.

Sets (c) and (d) were employed for comparative reasons.

The flasks were incubated at 25°C with rotary stirring (100 rpm, with an eccentricity of 5 centimetres).

At the start and every 5 days thereafter the residue of crude oil was determined by extraction with CCl_4 according to the method disclosed in British Patent Application No. 50375/76 (Serial No. 1 542 154).

The results which were obtained are shown graphically in Figure 3 of the accompanying drawing in which the ordinates represent the residual crude oil as a percentage of the original value, and the abscissae represent the time in days.

EXAMPLE 5

To the flasks of nine sets, (a) to (i), of 250 ml Erlenmeyer flasks having ground-glass necks and each containing 100 ml of unsterilized sea water and 100 milligrams of Basra crude oil, the following additions were made:-

to set (a), the same additives as for set (a) of Example 2;

to set (b), the same additives as for set (b) of Example 2;

to set (c), the same additives as for set (c) of Example 2;

to set (d), the same additives as for set (d) of Example 2;

to set (e), 5 milligrams of soybean lecithin and 10 milligrams of ethyl allophanate;

to set (f), 5 milligrams of soybean lecithin and 8 milligrams of spermidin;

to set (g), 5 milligrams of soybean lecithin and 12 milligrams of phenylacetyl urea;
to set (h), 5 milligrams of soybean lecithin and 12 milligrams of butyl hydantoinate; and
to set (i), 5 milligrams of soybean lecithin and 8 milligrams of butyl allantoinate.

The flasks were incubated and the residual hydrocarbons extracted as described in Example 1.

The results which were obtained are set out in the following Table 2, in which the residual oil content is expressed as a percentage of the initial value.

TABLE 2

	Incubation time, days			
Flask set	*7*	*12*	*19*	*25*
(a)	63.99	60.47	42.26	36.62
(b)	67.23	61.65	47.81	38.28
(c)	63.74	54.43	44.91	40.06
(d)	59.78	54.85	47.79	43.17
(e)	66.12	59.77	46.00	36.12
(f)	67.24	60.44	47.32	39.39
(g)	65.66	56.11	45.28	37.21
(h)	61.88	56.19	47.29	39.18
(i)	60.03	54.28	45.14	37.13

As can be seen, the results of Table 1 of Example 2 have fully been confirmed for sets (a), (b), (c) and (d) in Example 5.

WHAT WE CLAIM IS:-

1. In a method of reducing pollution caused by hydrocarbonaceous material on water which contains microbes capable of metabolizing hydrocarbons, which method comprises contacting the polluted water with a compound phosphorus and a compound having a poor solubility in water and containing nitrogen in a form assimilable by the aqueous microbes, the use as the phosphorus-containing compound of at least one compound selected from lecithin and other phosphatides, both synthetic and naturally occurring, and the use as the nitrogen-containing compound of (i) at least one compound selected from hydantoins, amides, allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids, or (ii) a mixture of (a) at least one compound selected from hydantoins, amides, allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids, and (b) at least one ureidic derivative of an aldehyde.

2. A composition containing phosphorus and nitrogen in a form assimilable by aqueous microbes capable of metabolizing hydrocarbons which cause pollution on water, the composition being intended to reduce pollution and comprising, as a phosphorus-containing compound, lecithin and/or at least one other phosphatide; and, as a nitrogen-containing compound, either (i) at least one compound selected from hydantoins, amides, allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids, or (ii) a mixture of (a) at least one compound selected from hydantoins, amides, allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids, and (b) at least one ureidic derivative of an aldehyde.

3. In a method of reducing pollution caused by hydrocarbonaceous material on water which contains microbes capable of metabolizing hydrocarbons, which method comprising contacting the polluted water with a compound containing phosphorus and a compound containing phosphorus and a compound having poor solubility in water and containing nitrogen in a form assimilable by the aqueous microbes, the use of lecithin as the phosphorus-containing compound and the use as the nitrogen-containing compound(s) of at least one compound selected from hydantoins and amides.

4. In a method of reducing pollution caused by hydrocarbonaceous material on water which contains microbes capable of metabolizing hydrocarbons, which method comprises contacting the polluted water with a compound containing phosphorus and compounds having poor solubility in water and containing nitrogen in a form assimilable by the aqueous microbes, the use as a phosphorus-containing compound of lecithin, and the use as nitrogen-containing compounds of a mixture of (a) at least one compound selected from hydantoins and amides, and (b) at least one ureidic derivative of an aldehyde.

5. A composition containing phosphorus and nitrogen in a form assimilable by aqueous

microbes capable of metabolizing hydrocarbons which cause pollution on water, the composition being intended to reduce pollution and comprising lecithin and at least one compound selected from hydantoins and amides.

6. A composition containing phosphorus and nitrogen in a form assimilable by aqueous microbes capable of metabolizing hydrocarbons which cause pollution on water, the composition being intended to reduce pollution and comprising lecithin and a mixture of (a) at least one compound selected from hydantoins and amides and (b) at least one ureidic derivative of an aldehyde.

7. In a method of reducing pollution caused by hydrocarbonaceous material on water which contains microbes capable of metabolizing hydrocarbons, which method comprises contacting the polluted water with a compound containing phosphorus and a compound having poor solubility in water and containing nitrogen in a form assimilable by the aqueous microbes, the use, as a phosphorus-containing compound, of at least one phosphatide which may be synthetic or naturally occurring, and the use, as the nitrogen-containing compound, of at least one compound selected from allophanates, polyamines, acyl ureas and esters of hydantoic and allantoic acids.

8. In a method of reducing pollution caused by hydrocarbonaceous material on water which contains microbes capable of metabolizing hydrocarbons, which method comprises contacting the polluted water with a compound containing phosphorus and compounds having poor solubility in water and containing nitrogen in a form assimilable by the aqueous microbes, the use, as a phosphorus-containing compound, of at least one phosphatide, and the use, as nitrogen-containing compounds, of a mixture of (a) at least one compound selected from allophanates, polyamines, acyl ureas, and esters of hydantoic and allantoic acids, and (b) at least one ureidic derivative of an aldehyde.

9. A composition containing phosphorus and nitrogen in a form assimilable by aqueous microbes capable of metabolizing hydrocarbons which cause pollution on water, the composition being intended to reduce pollution and comprising at least one phosphatide and at least one compound selected from allophanates, polyamines, acyl ureas and esters of the hydantoic and allantoic acids.

10. A composition containing phosphorus and nitrogen in a form assimilable by aqueous microbes capable of metabolizing hydrocarbons which cause pollution on water, the composition being intended to reduce pollution and comprising at least one phosphatide and a mixture of (a) at least one compound selected from allophanates, polyamides, acyl ureas, and esters of hydantoic and allantoic acids, and (b) at least one ureidic derivatives of an aldehyde.

11. A method according to any one of claims 1, 3, 4, 7 and 8, which also includes introducing into or onto the water, a freeze-dried microbe capable of metabolizing hydrocarbons on the water.

12. A composition according to any one of claims 5, 6, 9 and 10, which also includes a freeze-dried microbe capable of metabolizing hydrocarbons on the water.

13. A method according to any one of claims 1, 3, 4, 7, 8 and 11, wherein the compounds containing phosphorus and nitrogen are in the form of granules coated with a film of paraffin.

14. A composition according to any one of claims 5, 6, 9, 10 and 12, wherein the compounds containing phosphorus and nitrogen are in the form of granules coated with a film of paraffin.

15. A composition according to any one of claims 5, 6, 9, 10, 12 and 14, wherein the compounds containing phosphorus and nitrogen are in the form of granules and the granules also include a material of low specific gravity to assist buoyancy of the granules.

16. A method according to claim 1 of reducing pollution caused by hydrocarbonaceous material on water, substantially as described in relation to the flasks of set (a) of the foregoing Example 1.

17. A method according to claim 1 of reducing pollution caused by hydrocarbonaceous material on water, substantially as described in relation to the flasks of set (a), (b), (c) or (d) of the foregoing Example 2.

18. A method according to claim 1 of reducing pollution caused by hydrocarbonaceous material on water, substantially as described in relation to the flasks of set (a) or (b) of the foregoing Example 3.

19. A method according to claim 1 of reducing pollution caused by hydrocarbonaceous material on water, substantially as described in relation to the flasks of set (a) or (b) of the foregoing Example 4.

20. A method according to claim 1 of reducing pollution caused by hydrocarbonaceous material on water, substantially as described in relation to the flasks of set (e), (f), (g), (h) or (i) of the foregoing Example 5.

21. A compositon intended for reducing pollution, substantially as described in relation to the flasks of set (a) of the foregoing Example 1.

22. A composition intended for reducing pollution substantially as described in relation

to the flasks of set (a), (b), (c) or (d) of the foregoing Example 2.

23. A composition intended for reducing pollution substantially as described in relation to the flasks of set (a) or (b) of the foregoing Example 3.

24. A composition intended for reducing pollution substantially as described in relation to the flasks of set (a) or (b) of the foregoing Example 4.

25. A composition intended for reducing pollution substantially as described in relation to the flasks of set (e), (f), (g), (h) or (i) of the foregoing Example 5.

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1582965 COMPLETE SPECIFICATION
3 SHEETS This drawing is a reproduction of the Original on a reduced scale
Sheet 1

Fig.1

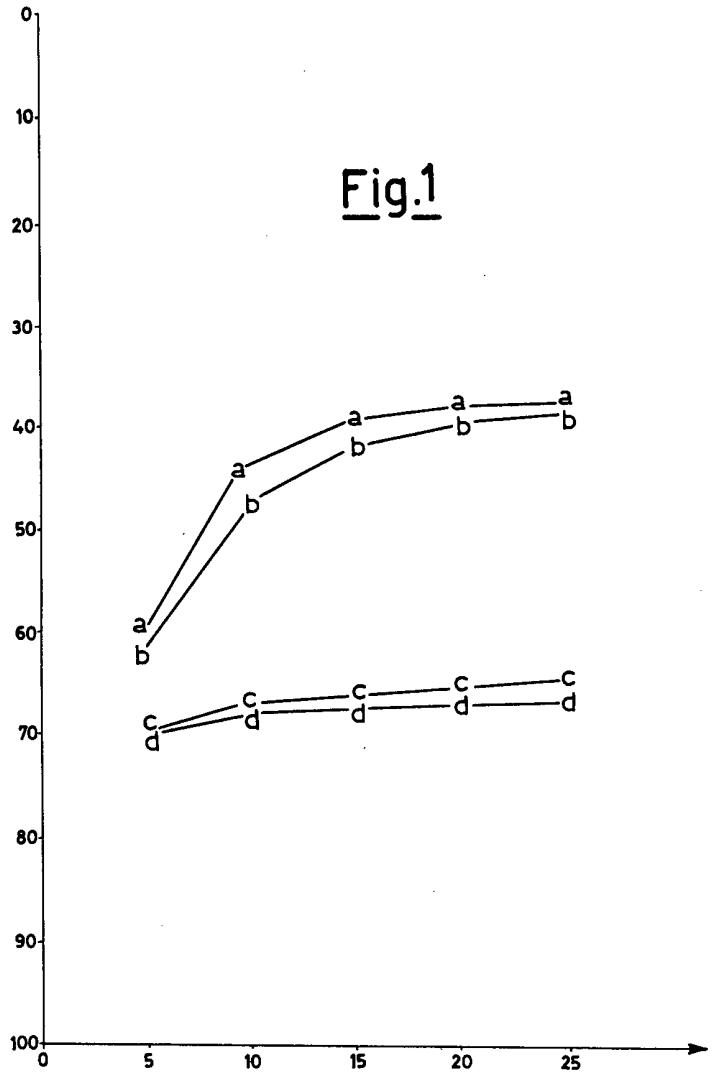

Fig.2

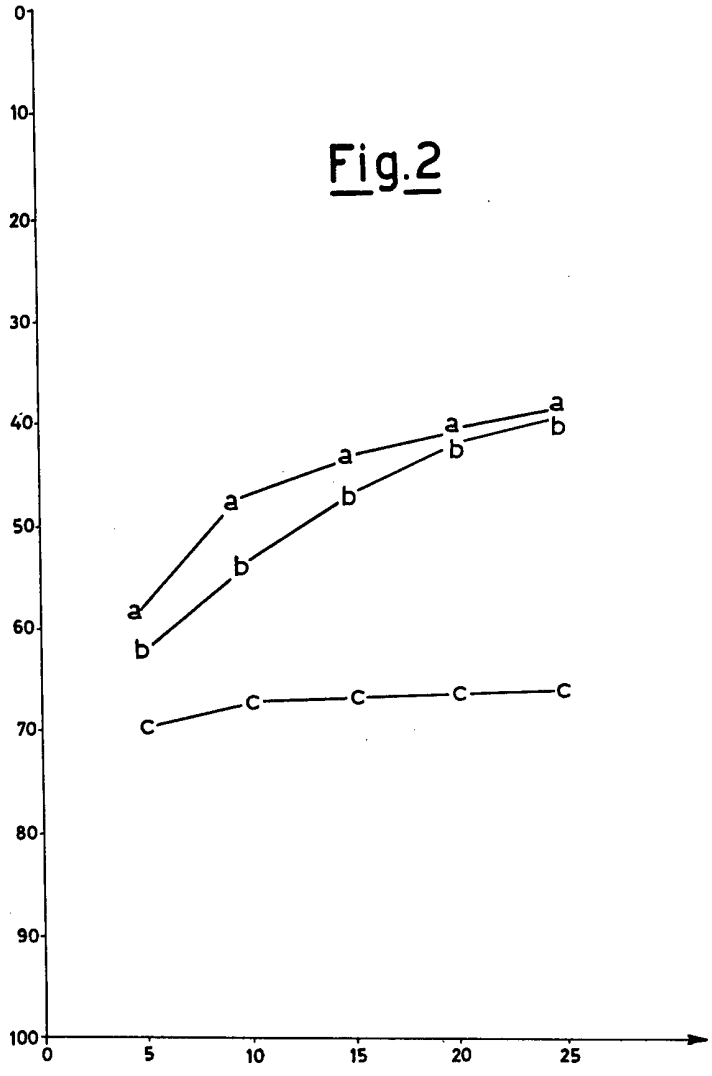

Fig.3