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3,362,801

HYDROCARBON OIL STABILIZATION

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17 Claims. (Cl. 44-63)

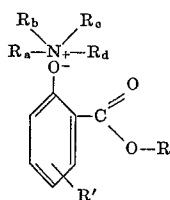
ABSTRACT OF THE DISCLOSURE

A blend containing alkyl quaternary ammonium salts of esters of salicylic acid or alkoxylated quaternary ammonium salts of esters of salicylic acid together with metal salts of esters of salicylic acid exhibits high stabilizing activity when present in a minor concentration in an unstable hydrocarbon oil.

This invention relates to blends of chemical compounds and to hydrocarbon oil compositions containing said blends.

The blend of chemical compounds of this invention includes alkyl quaternary ammonium salts of esters of salicylic acid or alkoxylated quaternary ammonium salts of esters of salicylic acid together with metal salts of esters of salicylic acid. These blends have been found to exhibit high stabilizing activity when present in a minor concentration in an unstable hydrocarbon oil, such as a fuel oil. The additive blend of this invention has high utility in fuel oil blends comprising both straight run and catalytically cracked components since such fuel oil blends have a high tendency to form deposits.

The alkyl quaternary ammonium salicylates of this invention can be selected from compounds having the general structure



wherein

R is a normal or iso alkyl, a normal or iso alkenyl, aralkyl or cycloalkyl substituent containing 1 to 22 carbon atoms and can be substituted with substituents such as nitrogen, phosphorus, sulfur or oxygen,

R' may be absent or is a normal or iso alkyl, a normal or iso alkenyl, aralkyl or cycloalkyl substituent containing 1 to 22 carbon atoms and can be substituted with substituents such as nitrogen, phosphorus, sulfur or oxygen,

Ra, Rb, Rc and Rd are each an oxygen-free normal or iso alkyl, a normal or iso alkenyl, aralkyl or cycloalkyl substituent containing 1 to 22 carbon atoms which can be substituted with substituents such as nitrogen, phos-

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phorus, or sulfur, at least one of said groups containing at least four carbon atoms,

N is nitrogen and can be the nitrogen of a heterocyclic ring such as imidazoline, pyridine, thiazine or piperazine, in which case Rc and Rd are members of the heterocyclic ring and at least one of the substituents Ra and Rb contains at least 4 carbon atoms,

C is carbon, and

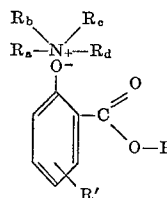
O is oxygen.

Some advantageous compounds of the above class of compounds include quaternary ammonium salts of octyl, dodecyl, tridecyl or hexadecyl salicylates where the quaternary ammonium group is selected from the following:

Alkyltrimethylammonium wherein the alkyl group is a C8- to C18- substituent; dialkyldimethylammonium wherein the alkyl groups are C8- to C8- substituents; or mixtures of said monoalkyltrimethylammonium and said dialkyldimethylammonium groups.

Some commercial starting materials which can be utilized as the quaternary ammonium base in the preparation of the above class of compounds are Arquad C, Arquad S, Arquad T, Arquad 12, Arquad 18, Arquad 2C, Arquad 2S, Arquad 2HT, Arquad T-2C, and Arquad S-2C, all of which are commercial products. Arquad C, Arquad T, Arquad S, Arquad 12 and Arquad 18 are alkyltrimethylammonium chlorides wherein the alkyl substituent comes from coco, tallow, or soya bean oils or fats, respectively, while in Arquad 12 and Arquad 18 the alkyl substituent is essentially pure dodecyl or octadecyl, respectively. Arquad 2C, Arquad 2S and Arquad 2HT are dialkyldimethylammonium chlorides wherein the alkyl substituent is coco, soya and hydrogenated tallow, respectively, and Arquad T-2C and Arquad S-2C are mixtures of the indicated alkyltrimethylammonium chlorides and dialkyldimethylammonium chlorides.

A group of quaternary ammonium salicylates that can be utilized either in place of or together with the quaternary ammonium salicylates described above are alkoxylated quaternary ammonium salicylates selected from compounds having the general structure



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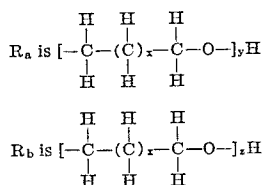
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wherein

R is a normal or iso alkyl, a normal or iso alkenyl, aralkyl or cycloalkyl substituent containing 1 to 22 carbon atoms and can be substituted with nitrogen, halogen, phosphorus, sulfur or oxygen,

R' may be absent or is a normal or iso alkyl, a normal or iso alkenyl, aralkyl, or cycloalkyl substituent containing 1 to 22 carbon atoms and can be substituted with nitrogen, halogen, phosphorus, sulfur or oxygen,

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where $x=0$ to 2, and $y+z=2$ to 15,

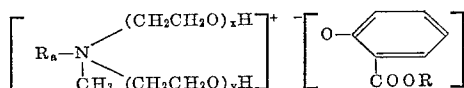
R_c and R_d are each a normal or iso alkyl, a normal or iso alkenyl, aralkyl, or cycloalkyl substituent containing 1 to 22 carbon atoms which can be substituted with nitrogen, halogen, phosphorus, sulfur or oxygen, at least one of said substituents containing at least 4 carbon atoms,

N is nitrogen and can be the nitrogen in a heterocyclic nitrogen-containing ring such as imidazoline, pyridine, thiazine or piperazine, in which case R_c and R_d are members of the heterocyclic ring,

C is carbon, and

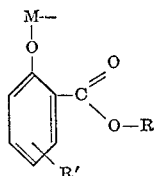
O is oxygen.

Compounds of the above group include



where R_a is a C_8 - to C_{18} -alkyl substituent such as oleyl; R can be octyl, isooctyl, dodecyl, tridecyl or hexadecyl; and $x+y=2$.

The metal salts of alkyl esters of salicylic acid of this invention can be selected from compounds having the general structure



wherein

R is a normal or iso alkyl, a normal or iso alkenyl, aralkyl or cycloalkyl substituent containing 4 to 22 carbon atoms and can be substituted with substituents such as nitrogen, phosphorus, sulfur or oxygen,

R' may be absent or is a normal or iso alkyl, a normal or iso alkenyl, aralkyl or cycloalkyl substituent containing 1 to 22 carbon atoms and can be substituted with substituents such as nitrogen, phosphorus, sulfur or oxygen,

M is a metal of Group I-A or Group II-A of the periodic table, particularly barium or calcium, or can be a metal from another group such as, for example, aluminum, zinc, tin, antimony, silver, mercury, vanadium, or a rare earth metal,

C is carbon, and

O is oxygen.

The dashed line attached to M indicates that M can be a polyvalent metal. Where M is a polyvalent metal, the plurality of groups attached to the metal can all be identical or nearly so or can differ from each other. For example, in addition to a salicylic ester radical attached to the metal, an hydroxyl radical could be attached to the second bond of a divalent metal to produce a basic salt of the metal.

Salicylic acid is a convenient starting material in the preparation of the quaternary ammonium salicylates of this invention. Salicylic acid contains an hydroxyl radical and a carboxylic acid radical at ortho positions on a benzene ring. In preparing the quaternary ammonium

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salts of this invention, the reactant containing the quaternary ammonium cation is reacted with the hydroxyl radical of the salicylic acid rather than with the carboxylic radical. However, the carboxylic acid radical is considerably more

acidic and therefore more reactive with the quaternary ammonium cation than is the hydroxyl radical and in order for the quaternary ammonium cation to react with the hydroxyl radical rather than with the carboxylic acid radical the salicylic acid must be esterified prior to the reaction thereof with the quaternary ammonium compound. Therefore, the preparation of the quaternary ammonium salt of this invention is a two-step process in which the order of performance of the steps is critical, i.e. the salicylic acid starting material is first esterified with a compound such as an alcohol and only after esterification is it converted to a quaternary salt.

In the following specific example of the preparation of alkyl quaternary ammonium salts of this invention from salicylic acid, the more reactive carboxylic acid radical is first reacted with an alcohol to form an ester. Thereupon, a commercial quaternary ammonium chloride is converted to the corresponding quaternary ammonium hydroxide. The quaternary ammonium hydroxide is then reacted with the hydroxyl radical of the ester of salicylic acid to form a quaternary ammonium salt of salicylic acid ester.

EXAMPLE 1

Step 1.—Preparation of isooctyl salicylate from salicylic acid

A mixture containing 900 grams of salicylic acid, 1300 grams (10 moles) of isooctyl alcohol, 30 grams of p-toluenesulfonic acid and 1000 milliliters of toluene was heated at reflux in a 5-liter flask fitted with a Dean and Stark trap. Reflux was continued until no more water collected in the trap. The solution was cooled to room temperature and extracted with one liter of water, then with one liter of 10 percent sodium bicarbonate solution followed by a washing with one liter of water. The extracted solution was dried over anhydrous sodium sulfate, filtered and vacuum distilled. After distillation of toluene and excess isooctyl alcohol, 1568 grams of a clear oil was collected at 150° - 160° C. and 3.2 millimeters pressure. Analysis of the product showed 72.21 percent carbon, 8.90 percent hydrogen, 18.21 percent oxygen and a molecular weight of 265. Theoretically, isooctyl salicylate contains 71.9 percent carbon, 8.8 percent hydrogen, and 19.2 percent oxygen, and has a molecular weight of 250.

Step 2.—Preparation of di(hydrogenated tallow)dimethylammonium isooctyl salicylate from isooctyl salicylate

Ninety grams (0.154 mole) of di(hydrogenated tallow)dimethylammonium chloride as a 75 weight percent paste containing isopropanol and a small amount of water, was dissolved in 200 milliliters of benzene. The composition of the 75 weight percent of the paste which consisted of di(hydrogenated tallow)dimethylammonium chloride was 75 percent by weight of distearyldimethylammonium chloride, one percent by weight of dioctadecenyldimethylammonium chloride and 24 percent by weight of dihexadecyldimethylammonium chloride. With stirring, a warm solution containing 10.1 grams (0.153 mole) of potassium hydroxide (assay 85%) in absolute ethanol was added to the benzene solution. The precipitated potassium chloride was removed by filtration. To the filtrate was added 38.4 grams (0.153 mole) of isooctyl salicylate. Solvent was removed. The product, a brown oil which solidified on cooling, contained 1.81 percent of nitrogen. The calculated nitrogen content of di(hydrogenated tallow)dimethylammonium octyl salicylate is 1.75 percent.

Following is a specific example of a process for the preparation of an alkoxyated quaternary ammonium salt of an ester of salicylic acid.

EXAMPLE 2

Step 1.—Preparation of isooctyl salicylate from salicylic acid

Same procedure as Step 1 of Example 1.

Step 2.—Preparation of dihydroxyethylmethyleylammonium isooctyl salicylate from isooctyl salicylate

To 12.5 grams (0.05 mole) of isooctyl salicylate in 30 milliliters of benzene was added 29.3 grams (0.05 mole based on neutralization equivalent) of dihydroxyethylmethyleylammonium hydroxide in isopropanol solution. The mixture was stirred. Solvents, including water of reaction, were removed at reduced pressure with slight heating. The product, dihydroxyethylmethyleylammonium isooctyl salicylate, a viscous clear brown liquid, weighed 29.1 grams and had a nitrogen content of 2.17 percent by weight, the theoretical nitrogen content being 2.3 percent by weight.

In the preparation of the preferred metal salts of an ester of salicylic acid of this invention, salicylic acid is first esterified. Thereupon, the sodium salt, or phenolate, is prepared. The desired metal salt is then precipitated from an aqueous solution of the sodium salt by reaction of the sodium salt of the ester of salicylic acid with a chloride or other soluble salt of the desired cationic metal, such as calcium or barium.

Following is a specific example of a process for the preparation of a metal salt of an ester of salicylic acid.

EXAMPLE 3

Step 1.—Preparation of isooctyl salicylate from salicylic acid

Same procedure as Step 1 of Example 1.

Step 2.—Preparation of calcium isooctyl salicylate from isooctyl salicylate

With stirring, a mixture containing 50.0 grams (0.2 mole) of isooctyl salicylate in 100 milliliters of 95 percent ethyl alcohol and 100 milliliters of water was treated with 8.0 grams (0.2 mole) of sodium hydroxide dissolved in 100 milliliters of water. The resulting sodium salt was dissolved by the addition of more ethyl alcohol-water. To this solution, with stirring, then was added 14.7 grams (0.1 mole) of calcium chloride dihydrate dissolved in 100 milliliters of water. The white precipitate which formed was filtered off, water-washed and dissolved in hexane. The resulting hexane solution was water-washed. Hexane was removed and the product was dried to constant weight. The yield was 53 grams and the calcium analysis was 7.0 percent. The theoretical calcium content of calcium isooctyl salicylate is 7.43 percent.

Tests were conducted to illustrate the highly cooperative stabilization effect which occurs when a metal salt of an ester of salicylic acid and a quaternary ammonium salt of an ester of salicylic acid are utilized together in a hydrocarbon oil. Each stability test was performed by heating 600 gram samples of a control fuel oil or a control fuel oil plus an additive or additives for a period of 64 hours at 210° F. in loosely stoppered, one-quart clear glass bottles. Following the heating period each test sample was cooled to room temperature and filtered by suction through a tared, medium porosity, fritted glass Gooch-type crucible. The sludge in each crucible was washed with heptane. Complete removal of the sludge adhering to the inside of the bottles was obtained by means of a rubber policeman and heptane. The respective crucibles were dried in an oven maintained at 210° F. for 1 hour, cooled in a desiccator and reweighed. The increase in weight was recorded as milligrams of sludge per 600 grams of oil.

In the first series of tests a control fuel oil sample was tested by itself to determine its stability and three comparative stability tests were conducted. The first comparative test utilized the control fuel oil plus the calcium

salt of isooctyl salicylate, the second comparative test utilized the control fuel oil plus the di(hydrogenated tallow)dimethylammonium salt of isooctyl salicylate, and the third comparative test utilized the control plus both the calcium salt of isooctyl salicylate and the di(hydrogenated tallow)dimethylammonium salt of isooctyl salicylate. The results of these tests are shown in Table 1. Table 1 also indicates for each test the results of a visual determination of the initial color of the oil and the color of the oil filtrate after completion of the test.

TABLE 1

Control fuel oil: A mixture of 50 percent by volume of FCC Light Catalytic Gas Oil and 50 percent by volume of Straight Run No. 2 Fuel Oil Distillate.	Control Fuel Oil	Control Fuel Oil	Control Fuel Oil	Control Fuel Oil
Additive:				
Calcium salt of isooctyl salicylate, pounds per 1,000 barrels.....		15		15
Di(hydrogenated tallow) dimethylammonium salt of isooctyl salicylate, pounds per 1,000 barrels.....			15	15
Stability Test:				
Potential Insolubles, milligrams per 600 grams of oil after heating 64 hours at 210° F.....	81.4	8.3	49.9	0.7
Color, ASTM D1500:				
Initial.....	L1.0	L1.0	L1.0	L1.0
At 64 hours.....	L5.0	L2.5	L4.0	L3.0

* Typical Control Fuel Oil Inspections:

Gravity, ° API, ASTM D287.....	34.0
Viscosity SUV, Sec., ASTM D446 100° F.....	34.3
Flash, P-M, ° F., ASTM D93.....	156
Pour, ° F., ASTM D97.....	-5
Carbon Residue, D524 on 10% Bottoms.....	0.2
Distillation, ASTM D158:	
Over Point, ° F.....	369
End Point, ° F.....	595
10% at, ° F.....	434
50% at, ° F.....	502
90% at, ° F.....	563
Residue, Percent.....	1.7

Table 1 shows that the combination of the calcium salt of isooctyl salicylate and the di(hydrogenated tallow) dimethylammonium salt of isooctyl salicylate reduced the potential insolubles in the control fuel oil to a level which is much lower than the potential insolubles level obtained when either of these additives was utilized alone in the control fuel oil.

A second series of tests was conducted which was similar to the tests reported in Table 1 and which utilized a portion of the same control fuel oil used in the tests of Table 1. The second series of tests differed from the series shown in Table 1 only in that the dihydroxyethylmethyleylammonium salt of isooctyl salicylate was utilized in place of the di(hydrogenated tallow)dimethylammonium salt of isooctyl salicylate of the tests of Table 1. The results of the second series of tests are shown in Table 2.

TABLE 2

Control fuel oil: A mixture of 50 percent by volume of FCC Light Catalytic Gas Oil and 50 percent by volume of Straight Run No. 2 Fuel Oil Distillate.	Control Fuel Oil	Control Fuel Oil	Control Fuel Oil	Control Fuel Oil
Additive:				
Calcium salt of isooctyl salicylate, pounds per 1,000 barrels.....		15		15
Dihydroxyethylmethyleylammonium salt of isooctyl salicylate, pounds per 1,000 barrels.....			15	15
Stability Test:				
Potential Insolubles, milligrams per 600 grams of oil after heating 64 hours at 210° F.....	81.4	8.3	98.6	1.0
Color, ASTM D1500:				
Initial.....	L1.0	L1.0	L1.0	L1.0
At 64 hours.....	L5.0	L2.5	L4.5	L2.5

Table 2 shows that the strong cooperative effect exerted by the additive compounds of this invention is a highly unexpected phenomenon. The data show that in the particular concentration used the dihydroxyethylmethyleylammonium salt of isooctyl salicylate, in the absence of the calcium salt of isooctyl salicylate, actually increased

the potential insolubles in the control fuel oil but when used together with the calcium salt resulted in a substantial reduction in the level of potential insolubles as compared to the potential insoluble level obtained when the calcium salt was utilized alone.

Additional data were taken to illustrate the effect of concentration and ratios of the additives of this invention to each other in hydrocarbon oil. The results of these tests are shown in Table 3.

TABLE 3

	Stability	
	Potential Insolubles ¹	Color ²
1. Control—50:50 Volume Blend of Straight Run No. 2 Fuel Oil Distillate and FCC Light Catalytic Gas Oil.....	127.4	L5.5
2. Control plus 30 pounds per 1,000 barrels of di(hydrogenated tallow)dimethylammonium isooctyl salicylate.....	14.8	L3.5
3. Control plus 30 pounds per 1,000 barrels of calcium isooctyl salicylate.....	9.3	L2.5
4. Control plus 30 pounds per 1,000 barrels of a 1:1 weight mixture of di(hydrogenated tallow)dimethylammonium isooctyl salicylate: calcium isooctyl salicylate.....	0.7	L3.0
5. Control plus 30 pounds per 1,000 barrels of a 2:1 weight mixture of di(hydrogenated tallow)dimethylammonium isooctyl salicylate: calcium isooctyl salicylate.....	0.5	L3.5
6. Control plus 30 pounds per 1,000 barrels of a 1:2 weight mixture of di(hydrogenated tallow)dimethylammonium isooctyl salicylate: calcium isooctyl salicylate.....	0.1	L3.5
7. Control plus 30 pounds per 1,000 barrels of dihydroxyethylmethylolammonium isooctyl salicylate.....	16.3	L3.0
8. Control plus 30 pounds per 1,000 barrels of 1:1 weight mixture of dihydroxyethylmethylolammonium isooctyl salicylate: calcium isooctyl salicylate.....	1.0	L2.5
9. Control plus 30 pounds per 1,000 barrels of a 2:1 weight mixture of dihydroxyethylmethylolammonium isooctyl salicylate: calcium isooctyl salicylate.....	3.4	L3.0
10. Control plus 30 pounds per 1,000 barrels of a 1:2 weight mixture of dihydroxyethylmethylolammonium isooctyl salicylate: calcium isooctyl salicylate.....	1.9	L2.5

¹ Milligrams per 600 grams of fuel after heating 64 hours at 210° F.
² ASTM D1500 filtrate at 64 hours.

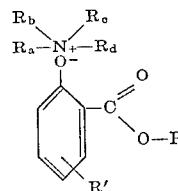
The data of Table 3 show that the advantageous cooperative effect of the metal salts and the quaternary ammonium salts of this invention prevails over wide ranges of ratios of these salts to each other in hydrocarbon oils.

The quaternary ammonium salicylates and the metal salicylates of this invention are soluble in and provide useful additive blends for hydrocarbon oils over a wide range of ratios of one compound to the other. For example, additive blends in which the weight ratio of quaternary ammonium salt to metal salt is generally between 1:9 and 9:1 and, preferably between 1:2 and 2:1 can be employed. These blends can be dissolved in hydrocarbon oils and other solvents and can be utilized in hydrocarbon oils over a wide range of concentrations. For example, in additive concentrates 10 percent by weight or more of the additive blends might be dissolved in oil. Usually, an additive blend would be present in hydrocarbon oils over a concentration range of about 0.002 to 1.0 percent by weight, generally, or, preferably, over a concentration range of about 0.005 to 0.1 percent by weight. Although useful in a wide variety of hydrocarbon oils such as gasoline, jet fuels, diesel fuels, etc., the additive blends of this invention are especially useful in fuel oils, particularly blends of straight run and catalytically cracked fuel oils, since such oil blends tend to be highly unstable and have a tendency toward deposit formation. Fuel oils containing the additive blend of this invention are commonly in contact with a water phase.

Various changes can be made without departing from the spirit of this invention or the scope thereof as defined in the following claims.

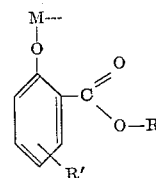
We claim:

1. A composition comprising constituents A and B in which A is a compound having the structure



wherein

R is a substituent containing 1 to 22 carbon atoms, R_a, R_b, R_c and R_d are each substituents containing 1 to 22 carbon atoms, at least one of said groups containing at least 4 carbon atoms, and R' is absent or is a substituent containing 1 to 22 carbon atoms; and B is a compound having the structure



wherein

R is a substituent containing 4 to 22 carbon atoms, M is a metal, and R' is absent or is a substituent containing 1 to 22 carbon atoms.

2. The composition of claim 1 in a hydrocarbon oil.

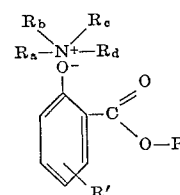
3. The composition of claim 1 wherein each of said substituents R, R_a, R_b, R_c and R_d is an alkyl, alkenyl, aralkyl or cycloalkyl substituent.

4. The composition of claim 1 wherein M is selected from the group consisting of calcium and barium.

5. The composition of claim 1 wherein the ratio of constituent A to constituent B is between about 9:1 and 1:9.

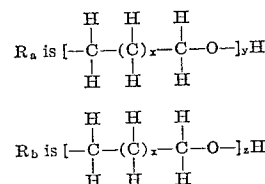
6. A composition comprising between about 0.002 and 1.0 percent by weight of the composition of claim 5 in a fuel oil.

7. A composition comprising constituents A and B in which A is a compound having the structure



wherein

R is a substituent containing 1 to 22 carbon atoms, and



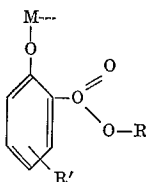
where x=0 to 2, and y+z=2 to 15,

R_c and R_d are each substituents containing 1 to 22 carbon atoms, at least one of which contains at least 4 carbon atoms, and

R' is absent or is a substituent containing 1 to 22 carbon atoms

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and B is a compound having the structure



wherein

R is a substituent containing 4 to 22 carbon atoms, M is a metal, and

R' is absent or is a substituent containing 1 to 22 carbon atoms.

8. The composition of claim 7 in a hydrocarbon oil.

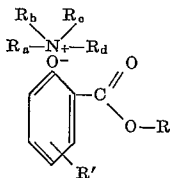
9. The composition of claim 7 wherein each of said substituents R_c and R_d is an alkyl, alkenyl, aralkyl or cycloalkyl substituent having 1 to 22 carbon atoms.

10. The composition of claim 7 wherein M is selected from the group consisting of calcium and barium.

11. The composition of claim 7 wherein the ratio of constituent A to constituent B is between about 9:1 and 1:9.

12. A composition comprising between about 0.002 and 1.0 percent by weight of the composition of claim 11 in a fuel oil.

13. A composition comprising constituents A and B in which A is a compound having the structure

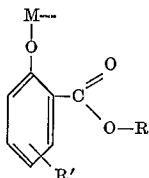


wherein

R is a substituent containing 1 to 22 carbon atoms, R_a and R_b are each substituents containing 1 to 22 carbon atoms, one of which contains at least 4 carbon atoms,

R_c and R_d are members of the same heterocyclic ring, and R' is absent or is a substituent containing 1 to 22 carbon atoms

and B is a compound having the structure



wherein

R is a substituent containing 4 to 22 carbon atoms, M is a metal, and

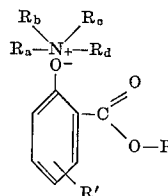
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R' is absent or is a substituent containing 1 to 22 carbon atoms.

14. The composition of claim 13 in a hydrocarbon oil.

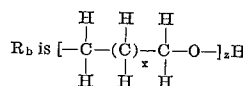
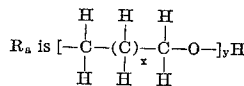
15. The composition of claim 13 wherein the ratio of constituent A to constituent B is between about 9:1 and 1:9.

16. A composition comprising constituents A and B in which A is a compound having the structure



wherein

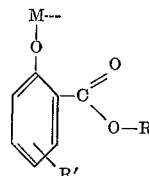
R is a substituent containing 1 to 22 carbon atoms,



where x=0 to 2, and y+z=2 to 15,

R_c and R_d are members of the same heterocyclic ring, and R' is absent or is a substituent containing 1 to 22 carbon atoms

and B is a compound having the structure



wherein

R is a substituent containing 4 to 22 carbon atoms, M is a metal, and

R' is absent or is a substituent containing 1 to 22 carbon atoms.

17. The composition of claim 16 in a hydrocarbon oil.

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55 DANIEL E. WYMAN, *Primary Examiner*.

Y. H. SMITH, *Assistant Examiner*.

UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,362,801

January 9, 1968

Elizabeth L. Fareri et al.

It is hereby certified that error appears in the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 2, line 16, for "C₈ to C₈-" read -- C₈ to C₁₈---

Signed and sealed this 11th day of February 1969.

(SEAL)

Attest:

Edward M. Fletcher, Jr.

Attesting Officer

EDWARD J. BRENNER

Commissioner of Patents