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3,493,510

LIQUID ESTER BASE OIL LUBRICATING COMPOSITIONS OF HIGH TEMPERATURE OXIDATIVE RESISTANCE

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No Drawing. Filed Feb. 18, 1966, Ser. No. 528,402

Int. Cl. C10m 3/28

U.S. Cl. 252—51.5

5 Claims

ABSTRACT OF THE DISCLOSURE

Normally liquid ester base oil lubricating compositions of high temperature oxidative resistance are provided containing minor amounts, effective to retard oxidation, of an ester base oil-soluble aromatic amine, such as N-phenyl-1-naphthylamine or p,p'-dioctyl-diphenylamine, and particular aromatic ketones having at least three hydroxy groups on the aromatic ring, such as 2,2',4,4'-tetrahydroxy benzophenone or 2,4,4'-trihydroxy benzophenone. These synthetic ester base oil lubricating compositions exhibit increased resistance to oxidation under high temperatures, e.g. in excess of 400° F. They have the further advantages of being non-corrosive to metals, possess excellent thermal stability and have compatibility with seal materials used in jet engines.

This invention relates to a synthetic lubricant composition containing a novel combination of base oil-soluble additive agents. More particularly this invention comprises a synthetic lubricant of superior high temperature oxidation resistance.

Organic compositions such as lubricating oils undergo oxidation upon exposure to air. This process is accelerated by elevated temperatures such as those which occur in engines and other operating machinery. When organic compositions are used as engine or machinery lubricants, their stability is still further drastically reduced by their contact with metal surfaces which give up metallic particles to the lubricant. Such abraded or dissolved metals or metal salts appear to act as oxidation catalysts in the lubricant causing the formation of oxidation products which in turn cause further degradation of the organic compounds present in the lubricant composition. In addition, water also causes corrosion of metallic surfaces and accelerates oxidation of the lubricant. Problems of this nature are encountered in mineral oils and are also troublesome in synthetic oleaginous fluids exemplified by esters. These synthetic fluids are usually not sufficiently resistant to oxidation to be useful alone. They can, however, be protected by the use of small amounts of additives. Aromatic amine antioxidants, e.g., N-phenyl-1-naphthylamine and p,p'-dioctyl-diphenylamine have been employed extensively to protect synthetic fluids against oxidation. Their activity, however, is not great enough to protect fluids for long periods of time above about 400° F. Aromatic hydroxy compounds, e.g., bis(3,5-di-tert-butyl-4-hydroxyphenyl) methane and 3,3',5,5'-tetra-tert-butyl-4,4'-dihydroxybiphenyl, have also been used as antioxidants, but their usefulness is generally limited to low temperature applications, e.g., in preventing the rancidity and auto-oxidation of fats and oils during storage, and in inhibiting the yellowing and deterioration of polymers during everyday usage. Their usefulness in synthetic lubricants which are exposed to high temperature is extremely limited.

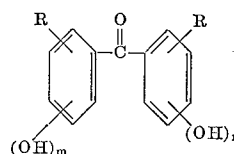
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The present invention provides a synthetic ester lubricant of lubricating viscosity which exhibits increased resistance to oxidation under high temperatures, e.g., in excess of 400° F. Advantageously, the present composition is more resistant to oxidation than similar esters containing amines such as N-phenyl-1-naphthylamine or p,p'-dioctyl-diphenylamine in reducing oxidation in synthetic lubricants.

It has been discovered by the present invention that particular aromatic ketones, for example, benzophenone, when substituted with at least three hydroxy groups on the aromatic ring and combined with an aromatic amine, provide an oil-soluble composition which is particularly effective in imparting to synthetic lubricants increased resistance to oxidation.

The particular ketones are not by themselves efficient antioxidants for synthetic lubricants as can be readily seen below in Table I, Example 5, where it is shown that 2500 ml. of oxygen is consumed in 69 minutes when testing a synthetic ester fluid containing only the ketone. However, when a minor amount of the particular ketones is added to synthetic ester lubricants containing one or more aromatic amine antioxidants, such as N-phenyl-1-naphthylamine and p,p'-dioctyl-diphenylamine, the oxidation resistance of the lubricants as measured by the rate of oxygen absorption, is substantially enhanced. The lubricants of the present invention have the further advantages of being non-corrosive to metals, excellent thermal stability, and compatibility with seal materials used in jet engines.

The aromatic ketones which can be used in the present invention include the hydroxy benzophenones which are represented by the following general formula:



wherein R is hydrogen or a hydrocarbon, alkoxy, aryloxy or acyloxy group of 1-22 carbon atoms; m and n are numbers from 0 to 4; and m plus n is a number from 3 to 6. Preferably both m and n are at least one. R may be substituted or unsubstituted and saturated or unsaturated, and is preferably of 1 to 12 carbon atoms, especially lower alkyl.

Benzophenones which are especially useful as antioxidants in the composition of the present invention are 2,2',4,4'-tetrahydroxy benzophenone and 2,4,4'-trihydroxy benzophenone. Illustrative of other suitable benzophenones are 2,2',4-trihydroxy-5'-octyl-benzophenone, 2,2',4-trihydroxy-4'-lauryloxy-benzophenone, 2,2',4-trihydroxy-4'-butoxy-benzophenone, 2,2',4-trihydroxy-5'-(p-hydroxyphenyl) benzophenone, 2,2',4-trihydroxy-5'-phenoxy-benzophenone, 2,2',4,4'-tetrahydroxy-5'-hexyl-benzophenone, 2,2',3,4-tetrahydroxy-benzophenone, 2,2',3,4-tetrahydroxy-4'-stearyloxy-benzophenone, 2,2',4,4'-tetrahydroxy-1-naphthophenone, 2,2',3,4,4'-pentahydroxy-benzophenone, and 2,2',3,3',4,4'-hexahydroxy-benzophenone. The benzophenones are added to synthetic ester lubricants containing aromatic amines in minor amounts effective to impart to the lubricant increased resistance to oxidation. The amount of hydroxy benzophenone added should be soluble in the base oil into which they are introduced and these amounts may vary with the particular base oil utilized. The benzophenones may often be present in the base oil in amounts of about 0.02% to 5% by weight, preferably about 0.1% to 1%.

The aromatic amine component of the invention is soluble in the ester fluid at least to the extent used and can be represented by the following general formula:



wherein Q is a monovalent hydrocarbon radical of 1 to 20 carbons, preferably 6 to 12 carbon atoms, whose adjacent carbon atoms are no closer than 1.40 Å. (i.e., a non-olefinic, non-acetylenic, monovalent hydrocarbon), and Q' is a non-olefinic, non-acetylenic aromatic hydrocarbon radical of 6 to 12 or 16 carbon atoms. Thus Q can be an alkyl group, including cycloalkyl, or an aromatic group. Preferably, both Q and Q' are aromatic, and often at least one is a fused ring aromatic, e.g., naphthyl. Q and Q' can be substituted with non-interfering substituents such as alkyl, aryl, hydroxyl and amine groups, preferably alkyl or aromatic amines, and Q and Q' can be linked together by means of a non-interfering element such as carbon, sulfur and oxygen. Illustrative of suitable amines are phenothiazine, N-phenyl- α -naphthyl amine; di(α -naphthyl amine); N,N'-diphenyl para-phenylene diamine; N, N' - dioctyl para - phenylene diamine, N,N' - diheptyl-para-phenylene diamine; diphenyl amine, p-octyl diphenylamine; p,p'-dioctyl diphenylamine; etc. The aromatic amine antioxidants are usually present in the lubricant composition in amounts from about 0.01 to 5 percent by weight of the final composition with a preferred amount being from about 0.1 to 2 percent and the amine may be substituted as with the alkyl groups, for instance, in the naphthyl or phenyl ring. For best results the relative concentrations of the additive may vary with the particular lubricant employed and may also be dependent on the characteristics of the final lubricant composition desired. Often it is preferred that the benzophenone to aromatic amine weight ratio be about 0.5 to 1 to 2 to 1, advantageously about 0.75 to 1 to 1.5 to 1. Increasing the amounts of the hydroxy benzophenone will generally be beneficial.

The lubricant composition of the invention includes as the major component a base oil which is an ester of lubricating viscosity which may be, for instance, a simple ester or compounds having multiple ester groupings such as complex esters, di- or other polyesters, and polymer esters. These esters are usually made from mono- and polyfunctional aliphatic alcohols or alkanols, and aliphatic mono- and polycarboxylic or alkanolic acids. Frequently, the alcohols and acids have about 4 to 12 carbon atoms. The reaction product of a mono-functional alcohol and a monocarboxylic acid is usually considered to be a simple ester. A diester is usually considered to be the reaction product of 1 mole of a dicarboxylic acid, say of 6 to 10 carbon atoms, with 2 moles of a monohydric alcohol or of 1 mole of a glycol, for instance, of 4 to 10 carbon atoms, with two moles of a monocarboxylic acid, e.g., of 4 to 10 carbon atoms. The diesters frequently contain from 16 to 40 carbon atoms.

A complex ester is usually considered to be of the type X—Y—Z—Y—X in which X represents a mono-alcohol residue. Y represents a dicarboxylic acid residue and Z represents a glycol residue and the linkages are ester linkages. Those esters, wherein X represents a monoacid residue, Y represents a glycol residue and Z represents a dibasic acid residue are also considered to be complex esters. The complex esters often have 30 to 50 carbon atoms.

Polymer esters or "polyester bright stocks" can be prepared by direct esterification of dicarboxylic acids with glycols in about equimolar quantities. The polyesterification reaction is usually continued until the product has a kinematic viscosity from about 15 to 200 centistokes at 210° F., and preferably 40 to 130 centistokes at 210° F.

Although each of these products in itself is useful as a lubricant, they are particularly useful when added or blended with each other in synthetic lubricant composi-

tions. These esters and blends have been found to be especially adaptable to the conditions to which turbine engines are exposed, since they can be formulated to give a desirable combination of high flash point, low pour point, and high viscosity at elevated temperatures. In addition, many complex esters have shown good stability to shear. Natural esters, such as castor oil may be employed and also be included in the blends, as may be small amounts of a foam inhibitor such as a methyl silicone polymer, or other additives of lubricant components to provide a particular characteristic, for instance, extreme pressure or load carrying agents, corrosion inhibitors, etc., can be added.

The monohydric alcohols employed in these esters usually contain about 4 to 20 carbon atoms and are generally aliphatic. Preferably the alcohol contains up to about 12 carbon atoms. Useful alkanols include butyl, hexyl, n-octyl, isooctyl and dodecyl alcohols, C₁₂ oxo alcohols and octadecyl alcohols. C₈ to C₁₀ branched chain primary alcohols are frequently used to improve the low temperature viscosity of the finished lubricant composition. Alcohols such as n-decanol, 2-ethylhexanol, "oxo" alcohols, prepared by the reaction of carbon monoxide and hydrogen upon the olefins obtainable from petroleum products such as diisobutylene and C₇ olefins, ether alcohols such as butyl Carbitol, tripropylene glycol monoisopropyl ether, dipropylene glycol monoisopropyl ether, and products such as "Tergitol 3A3" which has the formula C₁₃H₂₇O(CH₂CH₂O)₃H, are suitable alcohols for use to produce the desired lubricant. If the alcohol has no hydrogens on the beta carbon atoms, it is neo-structured; and esters of such alcohols are often preferred. In particular, the neo-C₈ alcohol—2,2,4 - trimethylpentanol-1—gives lubricating diesters or complex esters suitable for blending with diesters to produce lubricants which meet stringent viscosity requirements. Iso-octanol and iso-decanol are alcohol mixtures made by the oxo process from C₃—C₄ copolymer heptenes. The cut which makes up isooctanol usually contains about 17% 3,4-dimethylhexanol; 29% 3,5-dimethylhexanol; 25% 4,5-dimethylhexanol; 1.4% 5,5-dimethylhexanol; 16% of a mixture of 3-methylheptanol and 5-ethylheptanol; 2.3% 4-ethylhexanol; 4.3% α -alkyl alkanols and 5% other materials.

Generally, the glycols contain from about 4 to 12 carbon atoms; however, if desired they could contain a greater number. Among the specific glycols which can be employed are 2-ethyl-1, 3-hexanediol, 2-propyl-3, 3-heptanediol, 2 - methyl - 1, 3 - pentanediol, 2-butyl-1, 3-butanediol, 2,4 - diphenyl - 1, 3 - butanediol, and 2,4-dimethyl-1, 3-butanediol. In addition to these glycols, ether glycols may be used, for instance, where the alkylene radical contains 2 to 4 carbon atoms such as diethylene glycol, dipropylene glycol and ether glycols up to 1000 to 2000 molecular weight. The most popular glycols for the manufacture of ester lubricants appear to be polypropylene glycols having a molecular weight of about 100–300 and 2-ethyl hexanediol. The 2,2-dimethyl glycols, such as neopentyl glycol have been shown to impart heat stability to the final blends. Minor amounts of other glycols or other materials can be present as long as the desired properties of the product are not unduly deleteriously affected.

One group of useful monocarboxylic acids includes those of 8 to 18 or even 24 carbon atoms such as stearic, lauric, etc. The carboxylic acids employed in making ester lubricants will often contain from about 4 to 12 carbon atoms. Suitable acids are described in U.S. Patent No. 2,575,195, and include the aliphatic dibasic acids of branched or straight chain structures which are saturated or unsaturated. The preferred acids are the saturated aliphatic carboxylic acids containing not more than about 12 carbon atoms, and mixtures of these acids. Such acids include succinic, adipic, suberic, azelaic, and sebacic acids and "isosebacic" acid which is a mixture of α -ethyl suberic

acid, α,α' -diethyl adipic acid and sebacic acid. This composite of acids is attractive from the viewpoint of economy and availability since it is made from petroleum hydrocarbons rather than the natural oils and fats which are used in the manufacture of many other dicarboxylic acids, which natural oils and fats are frequently in short supply. The preferred dibasic acids are sebacic and azelaic or mixtures thereof. Minor amounts of adipic used with a major amount of sebacic may also be used with advantage.

The ester base oils to which incorporation of the additive combination of the invention is particularly advantageous are the oils commonly referred to as neostuctured polyol polyesters, i.e., having more than one ester

made according to the present invention containing both the aromatic amine and the benzophenone were subjected to oxygen absorption tests. The tests were conducted at 450° F. using 12 p.p.m. Fe (as iron octoate) as a catalyst, by passing a stream of oxygen at the rate of one cubic foot per hour through 75 grams of the ester fluid containing the inhibitors and comparing the amount of oxygen absorbed vs. time. The induction period is signalled by a marked increase in the rate of oxygen adsorption. The results are shown in Table I.

In the tables, the notation *Ti* indicates time of induction, *Tt* indicates total time, *Vi* indicates volume absorbed during induction, and *Vt* indicates total volume absorbed.

TABLE I

Ex. No.	Basic Components	Aromatic Ketone	Wt., Percent	O ₂ Absorption Test Data			
				Ti (min.)	Tt (min.)	Vi (ml.)	Vt (ml.)
1.	A ¹			23	87	204	2,500
2.	A ¹	2,2',4,4'-tetrahydroxybenzophenone	+0.50	327	352	1,860	2,500
3.	A ¹	do	+0.30	230	264	1,150	2,500
4.	A ¹	2,4,4'-trihydroxybenzophenone	+0.50	238	272	1,530	2,500
5.	B ²	2,2',4,4'-tetrahydroxybenzophenone	+0.50	0	69		2,500
6.	C ³			46	93	410	2,500
7.	C ³	2,2',4,4'-tetrahydroxybenzophenone	+0.50	266	304	1,060	2,500

¹ Base formulation A contains 72.22% Hercolube A and 24.08% Hercolube F to which was added 1% N-phenyl-1-naphthylamine 1% dioctyldiphenyl amine, 1.5% tetrabutyl ester of ethylene diamine tetraacetic acid and 0.20% of a half amide of hydrogenated dimer of linoleic acid.

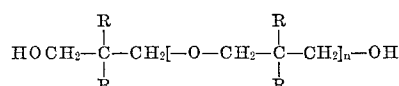
² Base formulation B contains 75% Hercolube A and 25% Hercolube F.

³ Base formulation C contains 75% Hercolube A and 25% Hercolube F, 1% N-phenyl-1-naphthylamine and 1% dioctyldiphenylamine.

Hercolube A is an essentially complete pentaerythritol ester of fatty acids having an average of 5-6 carbon atoms and with the following approximate inspection data: Acid No. 0.01; saponification No. 420-430; viscosity at 210° F., 4.25-4.35 cs.; viscosity at 100° F., 20.5-21.5 cs.

Hercolube F is an essentially complete ester of dipentaerythritol and a mixture of alkanolic acids containing an average of 5 to 6 carbon atoms per molecule and characterized by an acid number of about 0.01, a saponification number of about 390-398, 0.3% hydroxyl, and a kinematic viscosity at 210° F. of 8.7-8.9 cs. and at 100° F. of 55.5-57.5 cs.

group. These are the esters of aliphatic carboxylic acids, generally monoalkanoic acids, of about 4 to 12 carbon atoms, and a polyhydric alcohol free of beta hydrogen, i.e. containing no hydrogen on the beta carbon atoms, and including the di(polyhydric alcohol) ethers. The polyhydric alcohol generally contains about 2 to 6 hydroxy groups and about 5 to 20, preferably 5 to 12 carbon atoms. Illustrative of the alcohols are those having the general formula:



wherein *n* is 0 to 1 and R is a lower alkyl group, preferably of about 1 to 4 carbon atoms, which can be straight or branched chain, or a hydroxy lower alkyl, e.g. hydroxy methyl, group. These esters can be made by reacting a mole of the alcohol with about 2 moles up to the stoichiometric equivalent of the carboxylic acid.

Illustrative of polyhydric alcohols free of beta hydrogen

As it can be seen from the table, time required for the absorption of 2500 ml. of oxygen for a synthetic ester lubricant formulation (Example 1) is 87 minutes. When as in Example 2, 0.5% of 2,2',4,4'-tetrahydroxy benzophenone is added, the time required for the absorption of the same amount of oxygen is increased to 352 minutes. Example 3 differs from Example 2 in the amount of benzophenone added to the lubricant composition and Example 4 shows the effectiveness of using 2,4,4'-trihydroxy benzophenone. The same base oil containing 0.5% 2,2',4,4'-tetrahydroxy benzophenone as the sole antioxidant (Example 5) lasted only 69 minutes in the test. Examples 6 and 7 show similar results to Examples 1-4 in the absence of tetrabutyl ester of ethylene diamine tetraacetic acid and half amide of hydrogenated dimer acid, which serve respectively as corrosion inhibitor and anti-scuff and anti-fatigue additives in the formulation.

Table II shows a comparison between the benzophenones of the present invention with conventional phenolic antioxidants.

TABLE II

Ex. No.	Basic Components	Aromatic Ketone	Wt., Percent	O ₂ Absorption Test Data			
				Ti (min.)	Tt (min.)	Vi (ml.)	Vt (ml.)
1.	A ¹			23	87	204	2,500
2.	A ¹	2,2',4,4'-tetrahydroxybenzophenone	+0.50	327	352	1,860	2,500
8.	A ¹	3,3',5,5'-tetra-tert-butyl-4,4'-dihydroxybiphenyl	1.0	43	86	765	2,500
9.	A ¹	Bis(3,5-di-tert-butyl-4-hydroxyphenyl)-methane	1.0	61	101	950	2,500

¹ Hercolube A is an essentially complete pentaerythritol ester of fatty acids having an average of 5-6 carbon atoms and with the following approximate inspection data: Acid No. 0.01; saponification No. 420-430; viscosity at 210° F., 4.25-4.35 cs.; viscosity at 100° F., 20.5-21.5 cs.

are neopentyl glycol, trimethylolethane, trimethylolpropane, pentaerythritol, dipentaerythritol, 2-butyl-2-ethyl-1,3-propanediol, etc. Suitable aliphatic carboxylic acids with which the polyhydric alcohols free of beta hydrogen may be esterified are n-butyric acid, isobutyric acid, valeric acid, isopentanoic acid, caproic acid, isohexanoic acid, n-heptanoic acid, isoheptanoic acid, neopentanoic acid, n-octanoic acid, isooctanoic acid, pelargonic acid, n-decanoic acid, isodecanoic acid, neodecanoic acid, lauric acid, myristic acid, stearic acid, isostearic acid, etc.

Samples of ester-based fluids containing only the aromatic amine or the hydroxy benzophenone and samples

As can be seen from Table 2, the use of the conventional dihydroxy phenolic antioxidants in lubricants (Examples 8 and 9) compares very unfavorably with the benzophenones of the present invention (Example 2).

Table III shows the results of a 400° F., 72 hour Oxidation-Corrosion Test for a lubricant containing 0.3% 2,2',4,4'-tetrahydroxy benzophenone in addition to the components given in Example 1. No weight losses were shown for metal pieces made of copper, silver, steel, aluminum and magnesium. The slight weight increases are well within the limits of the specification. The excellent oxidation resistance of the lubricant is also reflected in

the low viscosity increase and low acid number of the need oil.

TABLE III

Material	Experimental Weight Change, mg./cm. ²	Specification Requirements Weight Change, mg./cm. ²
Copper.....	+0.10	±0.8
Silver.....	+0.031	±0.2
Steel.....	+0.038	±0.2
Aluminum.....	+0.015	±0.2
Magnesium.....	+0.007	±0.2
Used oil properties:		
Viscosity at 100° F., percent increase.....	19.65	-5 to 25
Acid No. change.....	0.96	3.0

Table IV shows the results of a number of other bench tests on the lubricant containing the additives of the present invention. As can be seen, the lubricant causes no excessive swelling of H-rubber and Viton A, causes no hardening of Viton A, and has a very low SOD lead corrosion. It passed the Navy seven-day, 221° F., oven storage test, the Allison -40° F., 72 hour stability test and the Seq. I, II, III foam tests.

TABLE IV

Formulation:	Weight percent
Hercolube A	72.69
Hercolube F	23.81
N-phenyl-1-naphthylamine	1.00
Diocetyl-diphenylamine	1.00
Tetrabutyl ester of EDTA	1.00
2,2',4,4' - tetrahydroxy benzophenone	0.30
Half amide of hydrogenated dimer acid ¹	0.20
Dow Corning fluid (200-60,000) ²	+0.001

¹ An extreme pressure agents.

² A Dow Corning silicone anti-foaming agent having a viscosity of 200 cs. at 250° C. and a molecular weight of 60,000.

The sample (formulation) was filtered and submitted to the followings tests:

		Required Specification
(1) Inspection tests:		
KV/210° F. cs.....	5.385.....	5.0-5.5.
KV/-40° cs.....	28.86.....	100 max.
KV/-40° F. cs.....	10,675.....	13,000 max.
Flash point, ° F.....	500.....	400 min.
Fire point, ° F.....	555.....	
Pour point, ° F.....	Below -80.....	-75 max.
Cloud point, ° F.....	Below -80.....	
Acid No. pH 11.....	0.45.....	0.50.
Sonic shear: Δ(KV/210), percent.....	<1.....	4 max.
Rubber swell, wt. percent:		
H-rubber.....	21.2.....	12-35.
Viton A.....	-1.1.....	
Durometer reading.....	65.....	
Foam test:		
Sequence I.....	0-0-0.....	25-0.
Sequence II.....	0-0-0.....	25-0.
Sequence III.....	0-0-0.....	25-0.
(2) SOD lead corrosion, 325° F., 1 hour mg./in. ²	-0.62.....	-6.0 max.
(3) Navy oven storage, 7 days 221° F.:		
Appearance.....	Clear.....	Clear.
Centrifuge deposit, ml./200 ml.....	Trace.....	0.005 max.

Table V shows the results of a Type 1½ bearing rig test. The viscosity rise at 100° F. at the end of 100 hours of test was only 20%, further indicating the excellent oxidation resistance provided by the antioxidants of the present invention.

TABLE V

Test conditions:

Duration, hrs.	100
Bearing speed, r.p.m.	10,000
Test oil sump temp., °F.	390
Test oil in temp., °F.	350
Bearing temp., °F.	500

Test results:

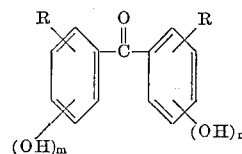
Oil viscosity increase at 100° F., percent	20.0
WADD demerit rating	96
Final acid No.	1.37
100 mesh filter deposit, gms.	1.902

It is claimed:

1. A normally liquid lubricating oil composition consisting essentially of a major amount of a normally liquid ester base oil of lubricating viscosity and minor amounts, effective to retard oxidation, of an ester base oil-soluble aromatic amine having the general formula:



wherein Q is a non-olefinic, non-acetylenic monovalent hydrocarbon radical of 1 to 20 carbon atoms and Q' is a non-olefinic, non-acetylenic aromatic hydrocarbon radical of 6 to 16 carbon atoms, and a hydroxy benzophenone of the formula:



wherein R is selected from the groups consisting of hydrogen and hydrocarbon, alkoxy, aryloxy, and acyloxy groups of 1-22 carbon atoms, and wherein m and n are numbers from 0 to 4 and m plus n is 3 to 6.

2. The composition of claim 1 wherein the aromatic amine is present in amounts of from about 0.1-3 percent, the benzophenone is present in amounts of from about 0.1 to 1 percent, and both m and n are at least 1.

3. The composition of claim 2 wherein the aromatic amine is selected from the group consisting of N-phenyl-1-naphthylamine and p,p' - dioctyl - diphenylamine and mixtures thereof.

4. The composition of claim 3 wherein the hydroxy benzophenone is 2,2',4,4'-tetrahydroxy benzophenone.

5. The composition of claim 3 wherein the hydroxy benzophenone is 2,4,4' - trihydroxy benzophenone.

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