United States Patent Office 3,296,135 Patented Jan. 3, 1967

3,296,135

LUBRICANT COMPOSITIONS HAVING IMPROVED CORROSION AND OXIDATION PROPERTIES

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No Drawing. Filed Feb. 8, 1963, Ser. No. 257,079
7 Claims. (Cl. 252—47.5)

This invention relates to synthetic lubricating oil compositions which exhibit improved resistance to corrosion and oxidation and which, in addition, possess a high degree of thermal stability. More particularly, this invention is directed to synthetic lubricant compositions which meet the critical test standards required of turboprop and turbojet lubricants.

This application is a continuation-in-part of United States applications Serial No. 76,162, now U.S. Patent Nos. 3,197,408, and 101,688, now abandoned, filed December 16, 1960 and April 10, 1961 in the name of R. A. Cupper and C. A. Wolf, Jr.

A wide variety of synthetic lubricants are known in the art as being useful lubricants for aircraft turbine engines which have high power outputs, such as turbojet and turboprop engines. The use of synthetic esters, has made it possible to extend the range of ambient temperature encountered and allow increased engine operating temperatures. However, the synthetic ester fluids employed as lubricants may be corrosive toward metal, particularly at high temperatures. It is immediately apparent that serious damage can occur to vital parts of an engine by permitting such corrosion to continue unchecked.

Turboprop and turbojet lubricants are required to meet certain critical viscosity requirements as set forth in specific test specifications. The critical turbojet standards for the military are set forth in U.S. Military Specification Mil–7808D. Turboprop lubricant specifications for commercial aircraft are described by the Allison Division Specification EMS–35F and the British Ministry of Supply Specification D. Eng. R.D. 2487, Issue 3. The British specification demands a very high degree of thermal stability as it requires the lubricant to exhibit satisfactory viscosity stability after exposure to temperatures as high as 536° F.

In the adaptation of synthetic lubricants such as those hereinbefore discussed, it has often been found necessary to modify the fluids with additives to improve their properties. For this purpose, amine-type antioxidants and various phenolic antioxidants have been employed, depending upon the particular use intended for the lubricant. The aforesaid vigorous standards demanded of turbojet and turboprop lubricants have been a barrier to the entry of uninhibited ester fluids into this field. This has prompted investigations to find a suitable additive system which will enable these synthetic base fluids to comply with the turboprop and turbojet lubricant specifications.

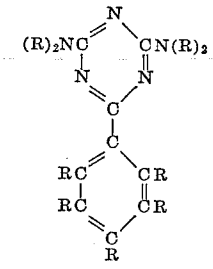
Accordingly it is an object of the present invention to provide a novel additive system which will impart to synthetic base fluids, high resistance to corrosion and oxidation.

It is a further object of the invention to modify said novel additive system to prevent thermal degradation of synthetic base fluids even in oxygen-free environments.

It is another object of this invention to provide a novel lubricating composition which may be used in turbojet and turboprop engines.

It is a still further object of the invention to provide such a novel lubricant which will meet the requirements demanded of advanced high temperature synthetic lubricants as exemplified by the Allison Test and the British Ministry of Supply Test.

In accordance with the present invention, it has been found that improved non-oxidative and non-corrosive properties may be achieved in synthetic lubricating compositions, by the addition thereto of a novel additive system which system comprises at least one amine antioxidant, at least one phenolic antioxidant, and a metal corrosion inhibitor of the formula:



wherein R represents hydrogen or an alkyl group having from 1 to 4 carbon atoms.

In a particular embodiment of this invention the said novel additives system may be modified to obtain thermal stability at temperatures in excess of 500° F. by addition thereto of certain condensed ring compounds as thermolysis additives. These thermolysis additives are particularly useful in lubricating compositions which must pass the high temperature requirements of the British Ministry of Supply Test. At such high temperatures as are present in this test, it has been found that molecular degradation of the synthetic lubricant fluid will take place even under non-oxidizing conditions, e.g., in oxygen-free environments. The compounds herein designated as thermolysis additives have been found to be extremely effective to prevent such high temperature degradation.

The amine antioxidants which are suitable for use in the novel additive package of this invention include phenothiazine, N-methylphenothiazine, N-phenyl-α-naphthylamine, N-phenyl-β-naphthylamine, p-phenylenediamine, o-phenylenediamine, 2,4′-diaminodiphenylamine, α,β-dinaphthylamine, 3,7-dioctylphenothiazine, p,p′dioctyldiphenylamine and the like. Preferred amine antioxidants are phenothiazine, N-methylphenothiazine, N-phenyl-α-naphthylamine, and N-phenyl-β-naphthylamine. Eminently preferred are phenothiazine and N-phenyl-α-naphthylamine.

The phenolic antioxidants which are useful in the novel additive package are 2,6-di-t-butyl-4-methylphenol, 2,6-di-t-butyl-4-benzylphenol, 3-t-butyltetrahydro-2-naphthol, 6-butyl-5-indanol, diphenylol propane, 2-2′-methylene-bis (4-ethyl-6-t-butylphenol), 4,4′-methylene-bis(6-t-butyl-5-indanol) 4,4′-methylene-bis(2,6-di-t-butylphenol), 2,2′-methylene-bis(4-methyl-6-t-butylphenol). Preferred phenolic antioxidants are 2,6-di-t-butyl-4-methylphenol, 4,4′-methylene-bis(6-t-butyl-5-indanol), and 2,2′-methylene-bis(4-ethyl-6-t-butylphenol). Highly preferred are 2,6-di-t-butyl-4-methylphenol and 2,2′-methylene-bis-(4-ethyl-6-t-butylphenol).

Metal corrosion inhibitors which may be used in the novel additive systems of this invention have been hereinbefore characterized by formula. These compounds include benzoguanamine and the alkyl substituted benzoguanamines. Benzoguanamine, itself, is preferably employed.

Although not wishing to be limited by any theory or mechanism by which stabilization is effected, it appears that the antioxidants employed in the novel additive system of the invention exert a complementary effect when used as herein prescribed. Amine antioxidants appear to be effective stabilizers against oxidation at temperatures

in excess of about 150° C., whereas the phenolic antioxidants seem to be effective at temperatures below about 150° C. At temperatures in excess of about 150° C., in the presence of conventional antioxidants, thermal breakdown of many base fluids has been found to occur even in oxygen-free environments. It has now been found that certain compounds, herein referred to as thermolysis additives, effectively prevent such high temperature breakdown of the fluid.

Compounds which have been found to operate effectively as thermolysis additives are condensed ring compounds such as acridine, phenazine, quinizarin, alizarin, 2-benzylpyridine, and N,N'-dibenzylpiperazine. Acridine is preferred.

The novel additive system of this invention contains from about 10 to 50 parts of amine antioxidant, from about 10 to 50 parts of phenolic antioxidant and from about 1 to 10 parts of the described metal corrosion inhibitor. In accordance with a preferred embodiment of the invention, the system also contains from about 1 to about 10 parts of thermolysis additive.

The additives are desirably employed in small amounts sufficient to stabilize the fluids, i.e., the amine antioxidant and the phenolic antioxidant are desirably present in the fluid in small amounts sufficient to lend oxidative stability and the metal corrosion inhibitor is utilized in quantities sufficient to impart non-corrosive qualities. These desirable effects will be admirably achieved by utilizing the additive system containing the components in the above proportions, in small amounts sufficient to impart superior resistance to oxidation and corrosion. Generally the additive system may be employed in amounts ranging from about 0.2 to about 12.0 percent by weight based on the base fluid. It is preferred that the said additive system be employed in amounts ranging from about 1.0 to about 6.0 percent by weight.

It is advantageous to heat the mixture in order to aid in the dissolving of the various components in the base fluid. It is pointed out that the novel lubricating compositions of this invention are formulated by mixing the components in the above defined proportions. The exact mechanism by which each of the additives functions is not precisely known. Therefore, upon mixing, the additives may in some measure, react with the base fluid resulting in a stable lubricant.

The novel additive system of this invention may be widely employed in a variety of synthetic lubricant base fluids. Generally the synthetic base fluids are those fluids of lubricating viscosity comprising compounds which consist essentially of carbon, hydrogen and oxygen, i.e., the essential nuclear structure is formed by these elements alone. Particularly contemplated are ester-type lubricants. For varying applications these ester lubricants may be utilized independently or together in blends i.e. with polyalkylene glycol derivatives depending on the characteristics desired in the finished lubricant. It has been found, in particular, that when base fluids comprising organic acid diesters and polyalkylene glycols or ethers of such polyglycols are utilized in conjunction with the additive system of this invention, lubricants may be provided which meet the rigid turboprop and turbojet lubricant requirements.

The ester-type lubricants contemplated can be simple diesters formed by the reaction of a dibasic acid with a monohydric alcohol, or polyesters prepared from polyhydric alcohols and polybasic acids in which reactive hydroxyl and/or carboxyl groups are not terminated, or polyesters prepared from polyhydric alcohols and polybasic acids in which reactive hydroxyl and carboxyl groups are terminated by reaction with monofunctional alcohols and/or acids. The latter type, i.e., terminated polyesters, are frequently referred to as complex esters. In general, ester type synthetic lubricants are characterized as having viscosity properties of between 1 and 60 centistokes at 210° F. and an ASTM pour point of below about 0° F.

Included among the suitable ester base fluids which may be used in the practice of this invention are:

(A) The dibasic acid esters which are represented by the formula

$$ROOC(C_nH_{2n})COOR$$

wherein n is an integer from 2 to 18 and preferably from 2 to 12, and wherein each R represents aliphatic or cycloaliphatic radicals, which have from 5 to 18 carbon atoms.

Mixtures of different dibasic acids may be employed in the preparation of these diesters. Physical mixtures of the diesters may also be used. Illustrative examples or organic dibasic acids which the diesters may be derived include, for example, succinic, glutaric, adipic, pimelic, suberic, azelaic, sebacic, hendecanedioic, dodecanedioic, brassylic, thapsic, octadecanedioic acids, and the like. The preferred acids are azelaic, adipic, sebacic, and suberic acids. The dibasic acids may be alkyl substituted to obtain lower viscosities at lower temperatures, so long as the substitution does not raise the freezing point of the diester to over 0° F.

The alcohols used to form the diester of the acid may be branched or straight chain, saturated or unsaturated aliphatic or cycloaliphatic alcohols. The saturated alcohols are preferred. Typical alcohols which may be employed include 2-ethylhexanol, 2-ethylbutanol, cetyl alcohol, n-octanol, amyl alcohol, oleyl alcohol, 2-butyl-octanol, methylcyclohexanol, dimethylcyclohexanol, and the like. Also useful are the "Oxo" alcohols such as isooctanol, isodecanol, tridecyl, alcohol, and the mixed branched chain nonanols which are prepared from the reaction of carbon monoxide and hydrogen upon olefins having a branched chain structure. Mixtures of alcohols may also be employed in the preparation of the diesters.

Representative synthetic diester lubricants which can be employed in conjunction with this invention include

di(2-ethylhexyl)sebacate,
di(2-ethylhexyl)azelate,
di[mixed-(2-ethylhexyl, isodecyl)]azelate,
di(isooctyl)azelate,
di(1-methylcyclohexylmethyl)sebacate,
di[mixed-(2-ethylhexyl, 2-propylheptyl)]azelate,
di(2,2,4-trimethylpentyl)sebacate,
di[mixed(2-methylpentyl, decyl)]azelate,
di(1-ethylpropyl)adipate,
di(3-methylbutyl)adipate,
di(1,3-dimethylbutyl)adipate,
di(2-ethylbutyl)adipate,
di(isooctyl)adipate,
di(undecyl)adipate,
di(tridecyl)adipate,
di(isodecyl)adipate,
di-[mixed(isooctyl, isodecyl)]adipate,
di(tetradecyl)adipate,
di(heptadecyl)adipate,
di(2,2,4-trimethyl-pentyl)adipate,
di(1-methylcyclohexylmethyl)adipate,
di(1-ethylpropyl)azelate,
di(3-methylbutyl)azelate,
di(2-ethylbutyl)azelate,
di(1-ethylpropyl)sebacate,
di(3-methylbutyl)sebacate,
di(1,3-dimethylbutyl)sebacate,
di(2-ethylbutyl)sebacate,
di(2-ethylhexyl)sebacate,
di[2-(2'-ethylbutoxy)ethyl]sebacate,
di(undecyl)sebacate,
di(tridecyl)segacate,
di(tetradecyl)sebacate,
di(heptadecyl)sebacate, di(2-ethylhexyl)glutarate,
di(undecyl)glutarate, and
di(tetradecyl)glutarate.

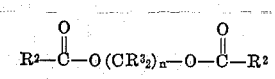
(B) Esters of aromatic acids which correspond to the general formula:

$$Z(COOR)_n$$

wherein n is an integer from 2 to 4, Z is a benzene or naphthylene nucleus substituted to the degree indicated by n, the remaining substitution being hydrogen substitution, and each R has the same meaning as previously described under (A) above.

Examples of acids from which the aromatic esters can be prepared include, phthalic, terephthalic, pyromellitic and naphthalene-1,4-dicarboxylic acid. The alcohols which are reacted with the aromatic acids are the same as those mentioned under (A) above.

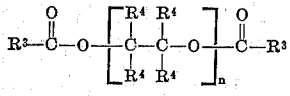
(C) Complex glycol centered esters having the general formula:



wherein n represent a number from 4 to 100, each R^2 represents alkyl or cycloalkyl radicals having from 4 to 24 preferably 4 to 12 carbon atoms, and each R^3 represents a hydrogen atom, a methyl group or an ethyl group.

Examples of the above esters include 3-methyl-1,5-pentanediol diisodecanoate, and hexamethylene 1,6-glycol-diisooctanoate, 2-ethylhexane-1,3-diol-di-valerate, 2,2,4-trimethyl-1,3-pentandiol-dipelargonate.

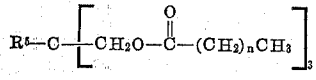
(D) Complex glycol centered esters containing recurring ether linkages represented by the formula:



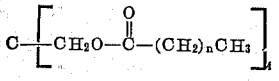
wherein each R^3 individually represents an alkyl or cycloalkyl radical having from 4 to 24 carbon atoms, each R^4, individually represents hydrogen atoms, methyl groups, or ethyl groups, and n represents an integer from 2 to 70.

Examples of these glycol centered esters include dipropylene glycol diperlargonate, the diester of valeric acid and polyethylene glycol in which the latter has an average molecular weight of 200, and the diester of 2-ethylhexanoic acid and polypropylene glycol in which the latter has an average molecular weight of 425.

(E) Esters prepared from a monobasic and an alkanepolyol corresponding to the formulae:



or



wherein R^5 represents an alkyl group having from 1 to 8 carbon atoms, and n is an integer from 0 to 18.

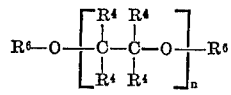
Examples of alcohols which are reacted with aliphatic monocarboxylic acids include trimethylolethane, trimethylolpropane, and pentaerythritol.

It is to be understood that the above classes of base fluids are representative of fluids recognized in the art and are merely illustrative of some of the base fluids useful in this invention. The complex esters have been described in the art as well as in U.S. Patents Nos. 2,575,-195; 2,575,196; and 2,703,811.

(F) Alkyl terminated polyesters.—These are complex polyesters prepared from polyhydric alcohols and polybasic acids wherein reactive carboxyl or hydroxyl groups are terminated by reaction with either monofunctional alcohols or acids. These esters are known in the art and are described in U.S. Patent No. 2,705,724.

(G) Hydroxyl terminated polyesters.—This class of compounds refers to those polyesters having free hydroxyl groups in the polymer chain which are not blocked or terminated by reaction with monofunctional or polyfunctional compounds. These compounds are known in the art and can be prepared by reacting a dibasic acid with a glycol or alkane diol as disclosed in U.S. Patent No. 2,929,786, or by reacting a dibasic acid with mixtures of polypropylene glycol and a 1,3-alkanediol.

In addition to the esters above, polyalkylene glycol derivatives are also stabilized by the novel additive system of the invention. These compounds may be represented by the formula:



wherein each R^6 represents hydrogen or an alkyl radical having from 1 to 12 carbon atoms, each R^4 represents hydrogen atoms, methyl groups or ethyl groups, and n represents a positive integer from 2 to 70. R^4 can be varied so as to provide homo- and hetero- polymers, i.e., polyethylene glycols, polypropylene glycols, mixed polyethylene-polypropylene glycols and also the mono alkyl ethers and dialkyl ethers of the above.

This class of compounds is well known in the art and the individual members of this class can be made in a variety of ways. Illustrative of the above class of compounds, as well as methods for the preparation of individual members can be found in U.S. Patents Nos. 2,293,-868; 2,425,755; 2,425,845; 2,448,664; 2,520,611 and 2,520,612.

Although this invention involves the use of the novel additives package in various synthetic lubricants set forth above, particularly preferred compositions have base fluids comprising diesters of dibasic acids and blends of these diesters with monoalkyl ethers of polyalkylene glycols, particularly polypropylene glycol. Such compositions may contain 25 to 80 percent by weight of a diester or of a mixture of diesters, 20 to 75 percent by weight of the said monoalkyl ether of a polyalkylene glycol, or mixtures of glycol ethers, and from about 1.0 to about 6.0 percent by weight of the novel additive system containing an amine antioxidant, a phenolic antioxidant and a metal corrosion inhibitor in the proportions hereinbefore set forth.

Highly preferred compositions contain 55 to 75 percent of the diesters, 45 to 25 percent of the glycol monoethers, and from about 1.25 to 4.5 percent of the said additive system.

In a preferred embodiment the above compositions containing the novel additive package of this invention may be advantageously complemented as hereinbefore disclosed, by the addition of a thermolysis additive which imparts to the lubricant compositions an ability to resist thermal breakdown at high temperatures. These thermolysis additives have been found to be necessary to meet requirements set forth in the British Ministry of Supply Specification for gas turbine lubricants.

It is to be understood that the novel lubricants of this invention can contain other well known additives. These additives can act as viscosity improvers, metal deactivators and anti-wear additives. For example, the polyacrylates may be employed as viscosity improvers. Disalicylidene-1,2-diaminopropane may be used as the metal deactivator and tricresyl phosphate may be used as a lubricity and anti-wear additive.

Commercial turboprop aircraft lubricant specifications are described by the Allison Division Specification EMS-34F and the British Ministry of Supply Specification, D. Eng. R.D. 2487, Issue 3. Of particular interest in evaluating the fluids of this invention is the 347° F. corrosion-oxidation test required by the Allison specification and the 536° F. thermal stability test of the British specification.

The corrosion-oxidation test used in evaluation of the subject fluids is simulative of the Federal Test Method 791-5308.4 that is employed in the Allison specification for turboprop lubricants. The test consists of placing the

test fluid in a long cylindrical cell in the presence of weighed steel, copper, and lead specimens and maintaining the fluid at a temperature of 352° F. for 72 hours while purging with clean, dry air at the rate of 5 liters (±0.5) per hour. The stability of the fluid is determined by measuring the viscosity and acid number change, observing the weight change of the metal specimens, and examining the fluid for the formation of insoluble material.

The British Ministry of Supply thermal stability test is described in Specification D, Eng. R.D. 2487, Issue 3. In this test, a 100 ml. sample of test oil is placed in a long test cell, and after sparging with nitrogen gas for 20 minutes, is exposed to a temperature of 280° C. for six hours in the absence of air. After cooling, a sample of the fluid is taken for viscosity measurement. This operation is repeated for four consecutive 6 hour cycles or until the maximum permissible viscosity change is reached. The resultant change in viscosity at 100° F. of the oil shall not at any time exceed the limits −10 to +10 percent. Thermal stabilities reported here were determined by the procedure described in the British specification except that test temperature was maintained at 282° C. rather than 280° C. as required in the specification test.

The following examples are illustrative.

Example 1

A blend was prepared having the following composition:

	Percent by wt.
Di-2-ethylhexyl azelate	58.8
"Ucon" lubricant LB–625 [1]	31.36
"Ucon" lubricant LB–1715 [2]	7.84
2,6-ditertiary-butyl-4-methylphenol	1.0
Benzoguanamine	0.50

[1] Monobutyl ether of polypropylene glycol having a S.U.S. viscosity of 625 at 100° F.
[2] Monobutyl ether of polypropylene glycol having a S.U.S. viscosity of 1715 at 100° F.

Properties of this blend are shown in Table 1.

Example 2

A blend was made having the following composition:

	Percent by wt.
Di-2-ethylhexyl azelate	54.2
"Ucon" lubricant LB–625	44.0
Phenothiazine	0.5
2,6-di-t-butyl-4-methyl phenol	1.0
Benzoguanamine	0.3

Properties of this product are shown in Table 1.

TABLE 1

	Allison EMS 35F Limits	Examples	
		1	2
Kinematic Viscosity, cs. at:			
210° F	7.5 (min.)	7.93	7.8
100° F		36.4	26.47
−40° F	13,000 (max.)	12,950	
Corrosion and Oxidation, 352° F./72 hrs., Weight Change, mg./sq. cm.:			
Fe	±0.2	±0.03	Nil
Cu	±0.4	−0.06	−0.06
Pb		−.06	−0.32
Viscosity Change at 100° F	% −5 to +12	−0.2	+0.19
Total Acid No. Increase	1.5 (max.)	1.3	1.07

The following table will summarize various lubricating oil compositions and the effect of omitting one or more of the critical components of the additive system of this invention.

TABLE II

Example	Base Fluid	Weight Percent Composition, Additives			Corrosion-Oxidation Test 72 hrs. at 352° F.			
		Amine	Phenolic	Corrosion Inhibitor	Percent Viscosity Change, 100° F.	Metal Weight Change, mg./cm.[3]		
						Fe	Cu	Pb
3	32% Polyglycol AU [1], 66.3% Di-2-ethylhexyl azelate.	0.5 PHTZ [2]	1.0 DBMP [3]	0.2 BG [4]	−4.1	0.05	Nil	Nil
4	32% Polyglycol AU, 66% Di-2-ethylhexyl azelate.	0.5 PHTZ	1.0 DBMP	0.5 BG	−0.99	−0.03	−0.03	−0.04
5	32% Polyglycol AU, 67.0% Di-2-ethylhexy azelate.			1.0 BG	−0.99	−1.4	−3.5	−69.9
7	32% Polyglycol AU, 67.5% Di-2-ethylhexyl azelate.	0.5 PHTZ			21.5	Nil	−0.16	−1.0
8	44% Polyglycol BU [5], 54.2% Di-2-ethylhexyl azelate.	0.5 PHTZ	1.0 DBMP	0.3 BG	−0.19	Nil	−0.06	−0.32
9	44% Polyglycol BU, 53.7% Di-2-ethylhexyl azelate.	1.0 PANA [6]	1.0 DBMP	0.3 BG	+4.9	Nil	−0.12	−7.1
10	44% Polyglycol BU, 55.5% Di-2-ethylhexyl azelate.	0.5 PHTZ			−11.7	0.03	−0.58	−7.3
11	44% Polyglycol BU, 55% 2-ethylhexyl azelate.	1.0 PANA			−8.2	0.02	0.18	−9.4
12	30% monobutyl ether of polypropylene glycol (S.U.S. Visc., 1,145 at 100° F.), 70% diethylhexyl sebacate.	1.0 PANA	1.0 MEBP [7]	0.5 BG	−7.2	−0.007	−0.299	−2.38
	Corrosion-Oxidation Test Limits				−5.0 to +12.0	±0.2	±0.4	

[1] 43.4 percent (weight) Ucon LB 625
56.6 percent (weight) Ucon LB 1715.
[2] Phenothiazine.
[3] 2, 6-ditertiarybutyl-4-methylphenol.
[4] Benzoguanamine.
[5] Ucon LB 625.
[6] N–phenylalphanaphthylamine.
[7] 2, 2′,-methylene-bis (4-ethyl-6-t-butylphenol).

It can be seen that only compositions which contained all three of the critical additives of this invention (Examples 3, 4, 8, 9 and 12) met the critical specification for a lubricant as defined by the Allison specification.

Other lubricant blends were prepared having varying compositions. The compositions of these lubricant blends are set out in Table III below.

TABLE III

	Blend A, percent	Blend B, percent	Blend C, percent	Blend D, percent	Blend E, percent
Di-2-ethylhexyl azelate	55.5	55.4	55.2	58.8	58.62
Ucon LB-625 [1]	44.0	44.0	44.0	31.36	31.26
Ucon LB-1715 [2]				7.84	7.82
Phenothiazine	0.5	0.5	0.5	0.50	0.5
2,6-Ditertiary-butyl-4-methyl phenol				1.0	1.0
Benzoguanamine				0.5	0.5
Acridine		0.1	0.3		0.3
	100.0	100.0	100.0	100.0	100.0

[1] Monobutyl ether of polypropylene glycol having a viscosity of 625 Saybolt Universal Seconds at 100° F.

[2] Monobutyl ether of polypropylene glycol having a viscosity of 1715 Saybolt Universal Seconds at 100° F.

The following Tables IV and V illustrate the results of the British Ministry of Supply thermal stability test and the corrosion-oxidation test made on each of the compositions prepared in Table I.

TABLE IV.—THERMAL STABILITY (282° C.)

[Percent Viscosity Change at 100° F.: Test Time, hours]

	6	12	18	24
Blend A	−6.1	−8.5	−17.9	
Blend B	−2.2	−6.8	−8.5	−11.1
Blend C	−2.9	−1.4	7.7	−8.8
Blend D	−12.6			
Blend E	−0.74	+0.49	−2.4	−5.0
British Specification D. Eng. RD 2487, Issue 3	(1)	(1)	(1)	(1)

[1] Limits: −10 to +20%.

TABLE V

[Corrosion-oxidation test, 72 hours at 352° F.]

	Percent Viscosity Change	Percent Metal Weight Change mg./cm.^{2}		
		Fe	Cu	Pb
Blend A	−11.7	−0.03	−0.58	−7.3
Blend B	−19.3	Nil	−0.68	−3.4
Blend C				
Blend D	−0.2	−0.06	−0.03	−0.06
Blend E	+1.1	−0.06	−0.02	−0.08
352° F. Corrosion Oxidation Test Limits	−5 to 12	±0.2	±0.4	±1.0

From the above tests it may be seen that Blend E containing antioxidants, metal corrosion inhibitor and acridine as a thermoylsis inhibitor was the only lubricant to successfully pass both the thermal stability test and the corrosion oxidation test.

Specification tests on other useful thermolysis additives appears in Table VI below.

TABLE VI

[Specification tests on potential thermolysis additives]

Fluid compositions:	Weight percent
Di-2-ethylhexyl azelate	66.9
UCON LB-1145	30.592
Phenylalphanaphthylamine	1.003
2,6-ditertiarybutyl-4-methylphenol	1.003
Benzoguanamine	0.502

Test Additive	Exposure Time, hours	Percent Viscosity Change
0.3 percent Acridine	24	−1.89
0.2 percent Phenazine (1% phenylbeta-naphthylamine inhibited	24	+1.05
0.3% Quinizarin	24	−7.27
0.3% percent 2-benzylpyridine	24	+3.08
0.3 percent Alizarin	24	+0.18
0.3 percent N,N′-dibenzyl-2,5-dibenzylpiperazine	24	+1.0

The following table demonstrates further how the addition of acridine to the additive system improves stability of various lubricants under the conditions of the British Ministry of Supply Test.

TABLE VII.—EFFECTIVENESS OF ACRIDINE IN IMPROVING THERMAL STABILITY

[Test Method; Turboprop lubricant specification as per British Ministry of Supply Specification D. Eng. R.D. 2487 Issue 3]

Fluid Composition	Additive	Hours at 280° C.	Percent Change in 100° F. Viscosity
74% di(isodecyl)adipate 26% UCON LB-625.	2.5% Additive Package A*	24	−6.63
74% di(isodecyl)adipate 26% UCON LB-625	2.5% AdditivePackage A 0.3% Acridine	24	−0.9
Di(2-ethylhexyl)sebacate	2.5% Additive Package A	24	+15.46
Di(2-ethylhexyl)sebacate	2.5% Additive Package A 0.3% Acridine.	24	+2.67
70% di(2-ethylhexyl)sebacate 30% UCON LB-1145	1% phenylalphanaphthylamine 1% 2,2′-methylene-bis(4-ethyl-6-ditertiary butyl phenol). 0.5% benzoguanamine.	24	−3.6
70% di(2-ethylhexyl)sebacate 30% UCON LB-1145	1% phenylalphanaphthylamine 1% 2,2′-methylene-bis(4-ethyl-6-ditertiary butyl phenol). 0.5% benzoguanamine. 0.3% Acridine.	24	−0.19
UCON LB-625	2.5% Additive Package A	6	−21
UCON LB-625	2.5% Additive Package A 0.3% Acridine.	18	−29.4

*Additive Package A: 50 parts phenylalphanaphthylamine; 50 parts 2,6-ditertiary-butyl-4-methyl phenol; 25 parts benzoguanamine.

What is claimed is:

1. A novel additive system for use in synthetic ester and glycol lubricants which comprises from about 10 to about 50 parts by weight of a compound selected from the group consisting of phenothiazine and N-phenylalphanaphthylamine, from about 10 to about 50 parts by weight of 2,6-di-t-butyl-4-methylphenol and from about 1 to about 10 parts by weight of benzoguanamine.

2. A non-corrosive, non-oxidative lubricant composition which comprises a lubricant base fluid which consists essentially of synthetic fluid of lubricating viscosity selected from the group of organic dibasic acid esters, the monoalkyl ethers of polypropylene glycol, and mixtures thereof, and from about 0.2 to about 12 percent by weight, based on the base fluid, of the additive system of claim 1.

3. The lubricant of claim 2 which contains from 1 to 10 parts by weight acridine based on the said additive system.

4. A non-corrosive, non-oxidative lubricant composition which comprises a lubricant base fluid which consists essentially of from about 25 to about 80 percent by weight of at least one diester of a dibasic carboxylic acid and from about 20 to 75 percent by weight of a monoalkyl ether of a polyalkylene glycol and from about 1.0 to about 6.0 percent by weight of an additive system which comprises from about 10 to 50 parts by weight of a compound selected from the group consisting of phenothiazine and N-phenyl-naphthylamine, from about 10 to 50 parts by weight of 2,6-di-t-butyl-4-methylphenol and from about 1 to 10 parts by weight benzoguanamine.

5. The non-corrosive, non-oxidative lubricants of claim 4 which contains from about 1 to 10 parts of acridine based on the novel additive system.

6. A non-corrosive, non-oxidative lubricant composition which comprises a synthetic lubricant base fluid which consists essentially of from about 25 to about 80 percent of at least one dialkyl ester of a dibasic carboxylic acid selected from the group of adipic, sebacic and azelaic acid, and from about 20 to about 75 percent of a monoalkyl ether of polypropylene glycol, and from about 1.0 to about 6.0 percent by weight of an additive system

which comprises from about 10 to about 50 parts by weight of a compound selected from the group consisting of phenothiazine and N-phenyl-naphthylamine, from about 10 to about 50 parts by weight of 2,6-di-t-butyl-4-methylphenol, from about 1 to 10 parts by weight of benzoguanamine and from about 1 to 10 parts by weight acridine.

7. A novel additive system for use in synthetic ester and glycol lubricants which comprises from about 10 to about 50 parts by weight of a compound selected from the group consisting of phenothiazine and N-phenylalphanaphthylamine, from about 10 to about 50 parts by weight of 2,6-di-t-butyl-4-methylphenol, from about 1 to about 10 parts by weight of benzoguanamine and from about 1 to about 10 parts by weight of a thermolysis additive selected from the group consisting of acridine, phenazine, quinizarin, alizarin, 2-benzylpyridine, and N,N'-dibenzylpiperazine.

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