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ANTIKNOCK COMPOUND AND MOTOR FUEL CONTAINING SAME

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This invention relates to high octane motor fuel, to a method of preparing the s ame, and to a new composition of matter having the ability to increase octane number of hydrocarbon motor fuel.

Although it is known that various metal carbonyls and carbonyl compounds such as iron pentacarbonyl, iron tetracarbonyl, nitrosyl carbonyls of various metals and metal tricarbonyl mercaptides will impart improved antiknock properties to hydrocarbon motor fuels, these compounds all have a serious drawback in that they are relatively unstable in the presence of light and/or air at ordinary atmospheric temperatures and as a result cause heavy precipitation with consequent loss in antiknock value of the motor fuel. Although the metal carbonyl mercaptides exhibit a fair degree of stability under normal storage conditions, in a series of storage tests run on motor fuel samples the octane of which had been improved by the addition of iron tricarbonyl mercaptides, it was found that after 56 days in storage in the presence of light and air at room temperature, not only was a large amount of flocculent precipitate formed, but from 50% to 100% of the octane increase caused by the addition of the iron compound had been lost.

The object of this invention is to provide a carbonyl compound which has a high degree of stability in the presence of light and/or air.

Another object of this invention is to provide a motor fuel of high octane value which does not readily depreciate during storage.

Other objects of the invention will become apparent from the following description.

I have discovered that when phosphatides such as lecithin and cephalin are reacted with polymeric metal carbonyls such as iron tetracarbonyl, iron nonacarbonyl, cobalt tetracarbonyl and cobalt tricarbonyl, a reaction product is formed which is highly stable to light and/or air and which when added to hydrocarbon motor fuel in relatively small amounts increases the octane rating of the motor fuel and the motor fuel will remain stable over long periods of time without formation of precipitates and without deterioration in octane rating.

In order to prepare the novel compositions in accordance with my invention, the polymeric metal carbonyl—for example, iron tetracarbonyl—is added to an oxygen-free solution of lecithin or other phosphatide in a hydrocarbon solvent such as naphtha, or other inert solvent, and the mixture agitated or stirred at a temperature of approximately 170–180° F. in the absence of oxygen for a period of thirty to sixty minutes. A complete color change takes place from dark green to dark yellow or deep red, depending upon the concentration of the solution, indicating that a reaction takes place.

In preparing the reaction product between the carbonyl and the phosphatide the ratio of carbonyl to phosphatide may vary within rather wide limits. I have found that a mole ratio of polymeric metal carbonyl to phosphatide ranging from 1 to 1, to a ratio of 10 to 1 to be satisfactory.

A series of blends of motor fuel were made up from straight run naphtha and various amounts of iron pentacarbonyl, iron tetracarbonyl and the reaction product of iron tetracarbonyl and lecithin, the reaction product being designated in the following table as Fe X, and the increased octane obtained in each case was determined. The results are tabulated in Table I:

Table I

Sample	Naphtha	Oct. No.	Compound present	Iron	Lecithin	Octane No. increase	Oct. No. increase calculated in direct proportion for 1 g. iron per gal.
				G./gal.	*G./gal.*		
1	Blend "A"	41.5					
2	Blend "B"	50.2					
3	Blend "A"		$Fe(CO)_5$ lecithin	1.08	3.78	5.7	5.3
4	do		Fe X lecithin	0.76	3.78	3.0	3.9
5	do		$Fe(CO)_4$ lecithin	0.76	3.78	1.5	2.0
6	Blend "B"		Fe X lecithin	3.0	31.78	10.6	3.5

In the case of the naphthas containing the iron pentacarbonyl and the iron tetracarbonyl, the solutions were made by dissolving these two iron compounds in naphtha containing lecithin in the amount indicated in the table. In the case of the Fe X reaction product, it was dissolved in naphtha which did not contain lecithin, but the table indicates the amount of lecithin present in the naphtha in the form of the reaction product. As shown by the table, the ability of Fe X to increase octane is better than iron tetracarbonyl but not as good as iron penta-

carbonyl. However, the reaction product has the advantage of being fairly stable in the presence of light and air, whereas iron pentacarbonyl rapidly deteriorates unless lecithin is present, and even in the case of lecithin it is not as stable as the Fe X reaction product.

The novel knock inhibitor composition in accordance with my invention may be prepared in concentrated solution in a solvent such as naphtha or toluene and the concentrated solution used as the knock inhibitor additive or the phosphatide and carbonyl may be added in the desired amounts to the gasoline whose knock rating it is desired to improve and the mixture heated to the desired temperature, namely, 170°–180° F., to form the reaction product in situ. In preparing the desired reaction product the temperature should be sufficiently high to cause fairly rapid reaction, but not so high as to favor the formation of decomposition products. At temperatures below 150° F. reaction takes place very slowly, whereas at temperatures above 240° F. decomposition products form with appreciable amounts of precipitate. At atmospheric temperatures of 70°–80° F. no visible reaction appears to take place. At temperatures of 170°–180° F. reaction is completed in from 15 to 20 minutes without any noticeable decomposition.

It has previously been pointed out that the reaction takes place only between polymeric metal carbonyls and phosphatides. An attempt to react iron pentacarbonyl, a monomeric carbonyl, with lecithin produced no color change in the solution when heated and stirred under the conditions hereinbefore set forth for the reaction between lecithin and iron tetracarbonyl.

The color of the reaction product between lecithin and iron tetracarbonyl varies from dark yellow through brown to dark red, depending upon the concentration of the solution in which it is prepared. The more dilute solutions are yellow and the most concentrated solutions are red. If a small amount of a concentrated solution of the reaction product is added to motor fuel or naphtha, the color of the naphtha at first turns red, but upon standing from 24 to 72 hours the color changes to dark yellow or brown, depending on the amount of concentrate.

In order to determine the stability of various solutions of the reaction product, a number of samples were prepared with varying ratios of iron tetracarbonyl to lecithin and the samples were permitted to stand in glass bottles in daylight at atmospheric temperatures with air in the top of the bottles. The results of the storage tests are given in Table II:

Table II

Sample	Mol. $Fe(CO)_4$ / Mol. phosphatide	Iron	Solvent or fuel used	Color after 10 days	Extent of precipitate formation, days		
					Trace after—	Slight after—	Appreciable after—
		G./gal.					
1	1.4/1	0.28	Petroleum naphtha.	Yellow	31		
2	4.4/1	0.76	do	Deep yellow	31		
3	7.0/1	1.26	do	Amber	15	31	
4	2.0/1	3.00	do	Dark brown	10	31	
5	Sample 1+isobutyl p-aminophenol (16#/1,000 bbls.)			Amber	31		
6	Sample 3+isobutyl p-aminophenol (16#/1,000 bbls.)			Deep amber	15	31	
C—1	2.0/1	11.4	do	Dark brown	1	2	21
C—1—1	2.0/1	0.76	Gasoline		21		
C—2	2.0/1	99.9	Petroleum naphtha.	Deep red	½	1	21
C—2—1	2.0/1	0.5	Gasoline	Yellow	[1] 14		
C—2—2	2.0/1	1.0	do	Dark yellow	14		
C—2—3	2.0/1	2.0	do	Amber	6	14	
C—2—4	2.0/1	4.0	do	Brown	2	14	
C—2—5	2.0/1	0.5	Ethyl gasoline	Light red	[1] 14		
C—3	1.3/1	67.2	Toluene	Deep red	14		
C—3—1	1.3/1	0.5	Gasoline	Yellow	[1] 14		
C—3—2	1.3/1	1.0	do	Dark yellow	14		
C—3—3	1.3/1	2.0	do	Amber	6	14	
C—3—4	1.3/1	4.0	do	Brown	2	14	
C—3—5	1.3/1	0.5	do	Light red	[1] 14		
C—4	4.0/1	113.6	Toluene	Deep red	11		
C—4—4	4.0/1	4.0	Gasoline	Brown	10		
C—5	8.0/1	113.6	Toluene	Deep red	2	5	10
C—5—4	8.0/1	4.0	Gasoline	Brown	1	10	

[1] None.

In the foregoing table samples 1–6 were prepared by forming the reaction product in situ in the naphtha. Samples C—1, C—2, C—3, C—4 and C—5 represent concentrated solutions of the reaction product which were prepared and the remaining samples were prepared by blending these concentrated solutions of the reaction product with motor fuel. It is apparent from the results in the table that the most stable solutions are prepared by forming the reaction products in situ in motor fuel in proportions in which it is desired to use them and that the smaller the amount of reaction product in solution, the more stable is the resulting solution. It is further apparent that the most stable concentrate that was prepared was the concentrate prepared using toluene as the solvent. Aromatic hydrocarbon solvents are therefore preferred because of their stabilizing effect on the reaction product. The addition of isobutyl p-aminophenol had no effect on the stability of the solutions but did affect the color thereof, causing them to become considerably darker.

The reaction products formed by the reaction of the polymeric metal carbonyls and the phosphatides can be reduced to the solid form by evaporating concentrates thereof either at room temperature or by gentle heating. The solid product is clear, dark brown in color, is non-sticky and non-brittle, is stable in air indefinitely and resembles a soft resin or plastic. This product can be redissolved in gasoline or aromatic solvents to give the required motor fuel blend or concentrate.

The stabilization of motor fuel containing metal carbonyls by addition of phosphatides to the fuel is claimed in my co-pending application Serial No. 415,065.

I claim:

1. A motor fuel comprising a major portion of hydrocarbons boiling within gasoline boiling range and a minor portion of the reaction product obtained by reacting a phosphatide with a polymeric metal carbonyl within the temperature range of approximately 150°–240° F. in the absence of oxygen.

2. A motor fuel in accordance with claim 1 in which the phosphatide is lecithin.

3. A motor fuel in accordance with claim 1 in which the phosphatide is cephalin.

4. A motor fuel in accordance with claim 1 in which the polymeric metal carbonyl is iron tetracarbonyl.

5. A motor fuel in accordance with claim 1 in which the polymeric metal carbonyl is cobalt tetracarbonyl.

6. A motor fuel in accordance with claim 1 in which the polymeric metal carbonyl is cobalt tricarbonyl.

7. A motor fuel comprising a major portion of hydrocarbons boiling within gasoline boiling range and a minor portion of the reaction product obtained by reacting between approximately 1 to 10 moles of polymeric metal carbonyl with approximately 1 mol of a phosphatide at a temperature between approximately 150°–240° F. in the absence of oxygen.

8. A composition of matter useful as an additive for motor fuels comprising the reaction product of a polymeric metal carbonyl and a phosphatide at a temperature between approximately 150° and 240° F. in the absence of oxygen.

9. A composition in accordance with claim 8 in which the phosphatide is lecithin.

10. A composition in accordance with claim 8 in which the metal carbonyl is a polymeric iron carbonyl.

11. A composition in accordance with claim 8 in which the metal carbonyl is a polymeric cobalt carbonyl.

12. Method of preparing a motor fuel additive comprising reacting a polymeric metal carbonyl with a phosphatide at a temperature between approximately 150° and 240° F. in the absence of oxygen.

13. Method in accordance with claim 12 in which the proportion in moles of polymeric metal carbonyl to phosphatide ranges from approximately 10 to 1 of the former to 1 part of the latter.

14. Method in accordance with claim 12 in which the phosphatide is lecithin.

15. Method in accordance with claim 12 in which the metal carbonyl is a polymeric iron carbonyl.

16. Method in accordance with claim 12 in which the metal carbonyl is a polymeric cobalt carbonyl.

17. Method of preparing a motor fuel additive comprising reacting at temperatures of about 150° to 240° F., a polymeric metal carbonyl and a phosphatide in a mole ratio of 1 to 10 of the former to 1 of the latter, said phosphatide being present in the form of an oxygen-free solution in an inert solvent.

18. A motor fuel additive comprising the reaction product obtained by heating a polymeric iron carbonyl and a solution of a phosphatide in an inert aromatic solvent to a temperature of approximately 170–180° F. in the absence of oxygen until the reaction is completed, the molar ratio of carbonyl to phosphatide being from 1 to 10 of the former to 1 of the latter.

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