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3,544,467

ACID-AMIDE POUR POINT DEPRESSANTS

George J. Kautsky, El Cerrito, Calif., assignor to Chevron Research Company, San Francisco, Calif., a corporation of Delaware

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The portion of the term of the patent subsequent to May 13, 1986, has been disclaimed
Int. Cl. C10I 1/22; C10m 1/32

U.S. Cl. 252—51.5

4 Claims

ABSTRACT OF THE DISCLOSURE

Pour point depressants for waxy hydrocarbonaceous fuels and oils having as the pour point depressant an alkenyl succinamic acid disubstituted on the nitrogen and the amine salts, the succinamic acid having a total of at least 50 carbon atoms. The succinamic acid or salt is preferably used in combination with a relatively low molecular weight ethylene-olefin copolymer.

CROSS-REFERENCE TO RELATED APPLICATIONS

This application is a divisional of application Ser. No. 719,701, filed Apr. 8, 1968, and now U.S. 3,444,082, which is a continuation-in-part of application Ser. No. 525,375, filed Feb. 7, 1966, and application Ser. No. 579,201, filed Sept. 14, 1966, the latter two applications both now abandoned.

BACKGROUND OF THE INVENTION

Field of the invention

When using liquid hydrocarbons as lubricating oils or fuels, it is necessary that the hydrocarbon fluids flow readily at low temperatures; that is, temperatures below the freezing point of water (0° C.). The flow of these fluids, particularly those with high wax content, is very sensitive to low temperatures. The crystallization results in the fluid setting up as a waxy material which does not pour. The pour point depressant additives do not reduce the amount of wax which crystallizes from the fluid, but modify the surface by absorption or cocrystallization and reduce fluid occlusion by the crystals. This changes the wax crystal structure and permits the fluid to flow.

Description of the prior art

Two major types of materials have found wide acceptance as pour point depressants: naphthalene alkylated with chlorinated waxes and homo- or copolymers of hydrocarbon olefins, methacrylates, vinyl esters, and alkyl styrenes. Carboxamides have found some mention in the patent literature, although they have not found particular commercial acceptance. See for example U.S. Pat. Nos. 1,870,074, 2,291,396, 2,342,114 and 2,727,862.

During the previous prosecution, the following patents were cited: U.S. Pat. Nos. 2,604,451; 2,699,427; 2,982,630; 2,982,634; 3,031,282; 3,148,960; 3,280,033; 3,219,666.

SUMMARY OF THE INVENTION

Pour point depressants for waxy hydrocarbonaceous fuels and oils are provided which are C-(n-aliphatic hydrocarbyl) succinamic acids of at least about a total of 50 carbon atoms, wherein the nitrogen has two aliphatic hydrocarbon substituents, each of which has at least 14 carbon atoms and the aliphatic hydrocarbyl group is of at least 14 carbon atoms and bonded to a carbon atom of the succinic acid radical at other than the terminal carbon

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atom of the aliphatic hydrocarbyl group (a secondary carbon atom). (Hydrocarbyl is an organic radical composed solely of carbon and hydrogen, which may be aliphatically saturated or unsaturated.)

The succinamic acid may be used as the acid or as the amine salt. The amine forming the salt may be a primary or secondary amine of from 0 to 50 carbon atoms.

Preferably, the succinamic acid or its salt or combination thereof is used with an ethylene-olefin copolymer wherein the ratio of the ethylene to the other olefin is in the range of 6-12:1.

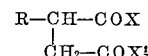
DESCRIPTION OF THE PREFERRED EMBODIMENTS

The pour point depressants of this invention are the C-(n-aliphatic hydrocarbyl) succinamic acids having 2 aliphatic hydrocarbon substituents on the nitrogen, the amine salt of the succinamic acid and, preferably, the combinations thereof. The succinamic acids, the salt or the combination thereof may be used by themselves or, preferably, with an ethylene-olefin copolymer wherein the ethylene to other olefin mole ratio is in the range of 6-12:1.

The polymers will be of a relatively low molecular weight, being of from about 1,000 to 100,000 molecular weight, more usually of from 1,000 to 20,000 molecular weight.

C-(n-aliphatic hydrocarbyl) succinamic acids

The succinamic acids will, for the most part, have the following formula:



wherein R is a straight chain aliphatic hydrocarbon group having from 0 to 1 site of olefinic unsaturation (alkyl or alkenyl) attached at a secondary carbon atom to the succinyl group and is of at least 14 carbon atoms, generally in the range of 15 to 28 carbon atoms and more usually in the range of 15 to 22 carbon atoms. One of X and X¹ is hydroxyl and the other is:



wherein N has its normal meaning of nitrogen and Y and Y¹ are aliphatic hydrocarbyl groups of from 14 to 28 carbon atoms, more usually of from 15 to 22 carbon atoms, having a total of from about 30 to 52 carbon atoms, more usually of from 32 to 48 carbon atoms, and, preferably, of from 32 to 40 carbon atoms.

Y and Y¹ may be aliphatically saturated or aliphatically unsaturated, generally free of acetylenic unsaturation (alkyl or alkenyl). There may be from 1 to 2 sites of olefinic unsaturation. Y and Y¹ may be the same or different and may be straight chain or branched chain, preferably straight chain. The branches will normally be not greater than 1 carbon atom, i.e., methyl. The position of attachment to nitrogen may be at a terminal or internal carbon atom.

As is evidenced from the above formula, it is not important which position the alkyl or alkenyl group has in relation to the carboxamide or carboxyl group. Because of the bulky nature of the amine, the usual method of preparation through the succinic anhydride will provide the alkenyl group β to the carboxamide as the major product. To the extent that this is the more easily accessible derivative, this derivative is preferred. However, as far as operability is concerned, either isomer or a mixture of the two isomers may be used.

Individual compounds or mixtures of compounds may be used as pour point depressants. Mixtures of different C- and/or N-substituents, both as to homologs and isomers, will frequently be employed when the individual

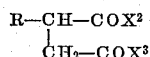
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precursors to the succinamic acid product are not readily available.

Illustrative succinamic acids include N,N-dihexadecyl hexadecylsuccinamic acid, N-hexadecyl, N-octadecyl octadecylsuccinamic acid, N,N-dihexadecenyl C₁₅₋₂₀-alkenylsuccinamic acid, N-hexadecenyl N-eicosenyl octadecylsuccinamic acid, N,N-diocetadecenyl C₁₆₋₁₈-alkenylsuccinamic acid, etc.

As indicated previously, the succinamic acid may be used as its amine salt, preferably as a mixture of acid and amine salt.

The amine salt or acid or mixtures thereof can be represented by the following formula:



wherein R is as previously defined, one of the X² and X³ is —NYY¹ wherein Y and Y¹ have been previously defined. The other of X² and X³ is of the formula:



wherein Y² and Y³ may be hydrogen, aliphatic hydrocarbon of from 1 to 30 carbon atoms or oxaliphatic hydrocarbon (there being 1 etheral oxygen atom present in the radical bonded to nitrogen at least β to the nitrogen atom) of from 3 to 30 carbon atoms. Y² and Y³ may be taken together to form a heterocyclic ring of from 5 to 7 members having nitrogen and oxygen as the only heteromembers. n varies from 0 to 1, preferably from 0.1 to 0.9. That is, from 10 to 90 mole percent of the succinamic acid present is in the form of its salt.

The aliphatic hydrocarbon groups may be saturated or unsaturated usually having not more than 2 sites of ethylenic unsaturation. The total number of carbon atoms for HNY²Y³ will be from 0 to 60, usually 1 to 40.

The groups indicated for Y and Y¹ may also be used for Y² and Y³. However, as already indicated, primary amines may be used as well as secondary amines to form the salt. Usually, where an amine other than the one used to prepare the succinamic acid is used to form the salt, as will be explained subsequently, there will be a mixture of salts; both the added amine and the secondary amine employed to prepare the succinamic acid will be involved in salt formation.

Illustrative amines which may be used to form salts are di-sec.-butyl amine, heptyl amine, dodecyl amine, octadecyl amine, tert.-butyl amine, morpholine, diethyl amine, methoxybutylamine, methoxyhexylamine, etc.

The alkenyl succinamic acids of this invention are readily prepared by reacting an alkyl or alkenyl succinic anhydride with the desired secondary amine at a temperature in the range of about 150° to 250° F. in approximately equimolar amounts, either neat or in the presence of an inert solvent. The time for the reaction is generally in the range of 15 minutes to 1 hour. This reaction is well known in the art and does not require extensive discussion here.

The alkyl or alkenyl succinic anhydride which is used may be individual compounds or mixtures of compounds. That is, various alkyl or alkenyl groups of differing number of carbon atoms or different positions of attachment to the succinic anhydride group may be used. Alternatively, a single isomer may be used. Since mixtures are generally more readily available, to that degree they are preferred. Frequently, mixtures will be used of aliphatic hydrocarbyl substituted succinic anhydrides wherein no single homolog is present in amount greater than 25 mole percent, each homolog being present in at least 5 mole percent.

Various secondary amines may be used, both those having the same aliphatic hydrocarbon groups and those having different aliphatic hydrocarbon groups. Either alkyl or alkenyl substituents may be present on the nitrogen, each having at least 14 carbon atoms. The range of difference between the two aliphatic hydrocarbon

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groups bonded at the nitrogen is not critical, but will generally be fewer than 8 carbon atoms, more usually fewer than 6 carbon atoms. For the most part, the aliphatic hydrocarbon groups will be straight chain, i.e., normal, with the amino nitrogen bonded either to internal or terminal carbon atoms.

It is found that when using approximately a 1:1 mole ratio of amine to succinic anhydride, depending on the reaction conditions, a significant amount of amine may be unreacted and remain to form the salt of the succinamic acid which is formed. In some instances, as much as 30 percent of the amine may remain unreacted, forming a significant amount of salt. Thus, the salt will frequently be from 10 to 30 mole percent of the total succinamic acid present.

Also, in situations where significant amounts of water are present during the course of the reaction, the water may react with a succinic anhydride to form succinic acid. If the temperature is not high enough to regenerate the succinic anhydride, the succinic acid will probably remain unreacted or form the amine salt with available unreacted amine. Therefore, the mixtures of amic acid salts may be conveniently prepared merely by using a 1:1 mole ratio of amine to succinic anhydride, and not attempting to drive the reaction to completion, or up to a mole excess of amine.

The amine salts are readily prepared by adding the amine to the succinamic acid, conveniently as prepared, or in an inert solvent. Mild heating may facilitate the reaction.

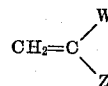
Ethylene copolymers

A preferred aspect of this invention is to use ethylene-olefin copolymers of from about 1,000 to 100,000 molecular weight, preferably from about 1,500 to 20,000 molecular weight wherein the mole ratio of ethylene to its comonomer is from about 6–12:1.

The polymers employed in this invention should have polyethylene segments in the polymer approximating the chain length of the wax. That is, the polyethylene segments should have from about 6 to 12 monomers on the average.

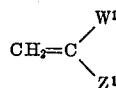
The major function of the other monomer, therefore, is to act as a divider between the polyethylene segments. For this reason, various monomers may be used which can be conveniently copolymerized with the ethylene. These olefins include hydrocarbon terminal olefins of from about 3 to 12 carbon atoms, more usually of from about 3 to 6 carbon atoms and various heteroatom containing addition polymerizable terminal olefins such as the acrylates, methacrylates, vinyl ethers, vinyl ketones, vinyl esters, etc.

The hydrocarbon olefins which find use will have the following formula:



wherein W is hydrogen or methyl and Z is hydrocarbon of from 1 to 10 carbon atoms, more usually alkyl of from 1 to 4 carbon atoms. Z is free of aliphatic unsaturation.

For the most part, the heteroatom containing olefins will have the following formula:



wherein W¹ is hydrogen, alkyl of from 1 to 3 carbon atoms or Z¹ and Z¹ is hydrocarboxycarbonyl



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wherein Q is aliphatically saturated hydrocarbyl), hydrocarbyloxy, acyloxy (QCO_2 —), and hydrocarbyl carbonyl. Z¹ is free of aliphatic unsaturation.

The preferred Z¹ is acyloxy and hydrocarbyloxy-carbonyl. The heteroatom containing monomer will generally be of from 4 to 24 carbon atoms, more usually of from 4 to 20 carbon atoms, have from 1 to 2 oxygen heteroatoms, and have only one site of olefinic unsaturation as its only aliphatic unsaturation.

The method of preparation of the polymer is not critical to this invention. Any convenient method for obtaining polymers of the desired molecular weight may be used. In preparing the hydrocarbon copolymers, usually nonstereospecific catalysts will be employed. Illustrative of such catalysts are triethylaluminum with vanadium oxychloride or titanium tetrachloride. These catalysts are in the category known as "Ziegler-type catalysts. Alternatively, free radical high pressure polymerizations may also be used.

Fuel and oil compositions

The succinamic acids of this invention (when referring to succinamic acid it is intended to include the salts or combinations of acids and salts), either by themselves or in combination with the ethylene-olefin copolymers, may be used with a wide variety of hydrocarbon fluids, either fuels or lubricating oils which require the lowering of their pour points. The compositions of this invention are particularly useful with mid-range distillate fuels.

Both naturally derived and synthetic hydrocarbon fuels or lubricating oils may be used in conjunction with the pour point depressing compositions of this invention. Naturally derived oils include naphthenic, paraffinic, asphaltic or mixed base oils, which may be waxy or partially dewaxed. Synthetic oils may be derived by polymerization of olefins, generally in the range of from C₆ to C₁₂, using any convenient catalyst.

The combination of the succinamic acid and the ethyleneolefin copolymer is particularly useful with diesel fuels obtained from cracked light cycle oils. Cracked light cycle oils generally have boiling ranges in the range of 300° to 700° F. (ASTM D 158-54).

Usually, at least 100 parts per million (p.p.m.) or more of the pour point depressing composition will be used. Generally, the amount of pour point depressant used will be less than about 2 weight percent and generally less than about 1 weight percent of the hydrocarbon fluid, usually in the range of 150 p.p.m. to 1,000 p.p.m.

The ratio of succinamic acid to ethylene-olefin copolymer will generally be about 0.25 to 10 parts of the succinamic acid to 1 part of the polymer, more usually from about 2 to 8 parts of the succinamic acid per part of polymer.

The pour point depressing compositions may be used in the presence of various other additives which are common to compounded fuels and lubricating oils. In addition to the pour point depressants, there may be present rust inhibitors, oiliness agents, dyes, detergents, extreme pressure additives, etc. Usually, these other additive will be present in amounts of from about 0.1 to 10 weight percent.

EXAMPLES

The following examples are offered by way of illustration 608.01(g).

Example A.—Isomerization of C₁₅₋₂₀ (number of carbon atoms) cracked wax olefins to internal unsaturation

Twenty pounds of C₁₅₋₂₀ 1-olefins are charged into a kettle and mixed with 2½ pounds of a silica-alumina catalyst (known under the trade name of Aero Cat.). Stirring is continued for ½ hour to disperse the catalyst, and then 800 pounds of C₁₅₋₂₀ 1-olefins are added. Heating is started, and the mixture is kept at 400° F. with stirring for 3 hours. Initially, the temperature of the re-

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action mixture rises to 425° F. due to the heat of reaction. The completion of isomerization is followed by the disappearance of an infrared band at 910 cm.⁻¹. After completion of the reaction, the mixture is filtered, and the isomerized olefin is distilled under 100 mm. pressure at 370° F. The distillation is complete when the kettle temperature reaches 575° F.

Example B.—Adduction of the isomerized olefin and maleic anhydride

A kettle is charged with 510 pounds of isomerized C₁₅₋₂₀ 1-olefins and 100 pounds of maleic anhydride (a ratio of olefin to maleic anhydride of 2:1 moles). The mixture is purged with nitrogen, and the system is sealed. Heating and stirring are started, and the mixture is kept at 450° F. The pressure in the system is about 25 p.s.i.g. due to vapors of maleic anhydride. The reaction is complete in about 3 hours, and the completion is determined by the disappearance of an infrared band at 840 cm.⁻¹. The excess olefin is thereupon removed by vacuum distillation.

Example I.—Exemplary preparation of succinamic acid

Into a reaction vessel was introduced 520 g. (1 mole) of di(hydrogenated tallow amine) (supplied by Foremost Chemical Co. as Formonyte 703) and 343 g. (1 mole) of alkenyl succinic anhydride (prepared as described in Example B) and the mixture heated to 150° F. The product was then isolated and characterized by titration and its infrared spectrum.

A number of other succinamic acids were formed, varying the alkenyl group and the secondary amine. These succinamic acids were tested according to ASTM D 97-57 with a variety of fuels. The following table indicates the results.

TABLE I

Alkenylsuccinamic acid	Alkenyl ¹	Amine ²	p.p.m.	Pour point ° F. ³			
				Light cycle oil		Diesel fuel	
				A	B	A	B
Example:							
I..... A	A ¹	500	B-80 ¹	B-80	-35	-35	
II..... B	A ¹	500	B-80	B-80	-35	-45	
III..... A	B ¹	500	B-80	-50	-25	-15	
IV..... B	B ¹	500	-55	-35	-20	-25	
			+10	0	0	+15	

¹ Alkenyl=A—C₁₈, internally unsaturated alkenyl bonded to the succinyl radical at other than a terminal carbon atom. B=C₁₅₋₂₀, internally unsaturated alkenyl bonded to the succinyl radical at other than a terminal carbon atom.

² Amine=A¹=di(hydrogenated tallow) amine (C₁₈₋₁₉) supplied by Foremost Chem. Co. as Formonyte 703. B¹=di(behenyl-arachidyl) amine (C₂₂₋₂₄) supplied by Humko Chem. Co. as Kemamine S-190.

³ See table below.

	Light cycle oil		Diesel fuel	
	A	B	A	B
ASTM distillate ° F.....	394-622	410-683	370-664	340-688
50% point ° F.....	493	554	511	532
Gravity (° API).....	25.7	19.5	39.0	37.4
Cloud point ° F.....			-10	0
Sulfur, wt. percent.....	1.3	1.3	0.81	1.0

⁴ B-80=below 80° F., the lower measured limit of the procedure.

The alkenyl succinamic acid of Example I was titrated and various amounts of different amines were added based on the titer obtained. The titration is carried out as follows: A sample of about 1.5 g. is weighed accurately in a 250 ml. beaker. The sample is dissolved in 100 ml. of chloroform and 10 ml. of methanol. The solution is stirred with a magnetic bar and 1 ml. increments of 0.1 normal ethanolic KON are added, the titration being followed by means of an electric pH meter. The titration is plotted and the end point determined.

Using fuels analogous to those in Table I, the pour

points were determined for the succinamic acid salt-acid mixtures.

TABLE II

Amine	Percent of succinamic acid neutralized with amine	Add. conc. p.p.m.	Pour point depression ° F. ¹			
			Light cycle oil		Diesel fuel	
			D ²	E ²	D ²	E ²
Di-(hydrogenated tallow)amine ²	25	300	>60	40	15	25
	50	300	10	20	45	40
	25	300	>70	60	25	40
	50	300	>70	>60	35	35
Di(sec-butyl)amine.....	100	1,000	>70	40	30	35
2-heptylamine ⁴	25	300	>70	50	25	30
	75	300	>70	35	30	45
2-nonylamine ⁴	25	300	>60	50	30	45
	75	300	>70	55	35	45
2-undecylamine ⁴	25	300	>70	40	25	40
	75	300	>70	50	25	50
2-pentadecylamine ⁴	25	1,000	>70	25	25	50
Morpholine.....	50	300	>70	25	25	50
tert.-butylamine.....	50	300	>70	25	25	50

¹ The degrees of depression are reported, subtracting the pour point of the fuel having the additive from the original pour point.

² See table below.

	Pour point ° F.
LCO.....	-10
LCO E.....	-10
DF D.....	+5
DF E.....	+10

³ Supplied by Foremost Dairy Co. as Pormonyte 703.

⁴ Supplied by Armour Industrial Chem. Co. as the Armeen L series.

In order to demonstrate the effect of a combination of an alkenyl succinamic acid and ethylene/propylene copolymer, the pour points of a light cycle oil, C, with varying combinations of the polymer and succinamic acid were determined. The same ASTM procedure as previously described was used.

TABLE III

Alkenyl ¹ succinamic acid, p.p.m.:	Ethylene/ ² propylene polymer, p.p.m.	ASTM pour point of light cycle oil C ³ ° F.
170.....		+10
225.....		+5
		0
	170	-15
	225	-30
85.....	85	-25
170.....	85	-50

¹ ° Alkenyl succinamic acid of Example I.

² Ethylene/propylene copolymer, ~1,500 mol wt.; mol ratio C²/C³~9/1.

³ API gravity—27.2 ASTM, Dist., ° F., 411-622 at 650 mm. Hg.

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In order to demonstrate the effectiveness of the succinamic acids in combination with other ethylene-comonomer copolymers, mixtures were prepared of the succinamic acid of Example I with 2 ethylene copolymers.

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The compositions were tested at varying ratios and with fuels analogous to the fuels indicated in Example I. The following table indicates the results

TABLE IV

Copolymer ¹ :	Wt. ratio of Ex. I to copolymer ²	p.p.m.	Fuels, pour point, ° F.			
			A ¹	B ³	C ³	D ³
A.....	4:1.....	600	-25, -25	-20, -15	-25	-50, -50
	1:1.....	600	-15	0	-0 ²	
	1:4.....	600	-15	+5	-25	
	Only copolymer.....	600	-15, -15	0, -5	-25, -45	-30
	Only Ex. I.....	600	-10, -20	-15, -10	-15, -35	-30, -25
B.....	4:1.....	600	-30	-20	-30	
	1:1.....	600	-20	0	-30	
	1:4.....	600	-10	0	-25	
	Only copolymer.....	600	-15	0	-30	
	Only Ex. I.....	600	-10	-15	-15	
	No additive.....		+10	+20	+5	

¹ A.—Ethylene-vinyl acetate copolymer (9:1 mol ratio); ~1,500 molecular weight. B.—Ethylene-isobutyl acrylate copolymer (~7:1 mol ratio); ~2,100 molecular weight.

² The copolymer is used as a 50 wt. percent active solution; Ex. I is used as a 80 wt. percent active solution.

³ See table below.

	A	B	C	D
Distillation, ASTM D-86, ° F.:				
S.....	362	378	402	371
50.....	534	551	554	543
90.....	596	628	634	610
9 API gravity, 60/60.....	36.4	35.7	34.8	35.3
Vis. 100° F., SUS.....	37.64	38.64	39.8	38.1
Flash Pt. ° F. ASTM D-93.....	182	174	120	220
Percent sulfur, ASTM D-1266.....	0.37	.38	.37	.42

The composition of Example I was also tested for its effect on cloud point according to ASTM D 97-57. The following table indicates the results obtained.

TABLE V

	p.p.m.	Cloud point ° F. ¹			
		Light cycle oil		Diesel fuel	
		A	B	A	B
Alkenylsuccinamic acid:					
I.....	500	-12	-14		
Do.....	1,000			+6	+12
		-2	0	+6	+18

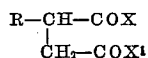
¹ Refer to Table I for a description of the various oils and fuels.

It is evident from the above data that the succinamic acids used in this invention either by themselves or in combination with the ethylene copolymers are excellent pour point depressants for a wide variety of hydrocarbonaceous media, for which pour point depression is only difficultly achieved. Moreover, pumpability is retained even below the cloud point of the hydrocarbonaceous media. Also, it is found that the succinamic acids do not interfere with other additives which may be present in the oils or fuels and do not add to or enhance undesirable qualities of the hydrocarbonaceous media. The pour point depressants of this invention provide compositions having good water tolerance, tend to enhance corrosion inhibition, both with mild steel and zinc, and do not significantly affect the stability of the hydrocarbonaceous media to oxidation.

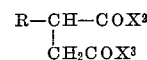
As will be evident to those skilled in the art, various modifications on this invention can be made or followed, in the light of the foregoing disclosure and discussion, without departing from the spirit or scope of the disclosure or from the scope of the following claims.

I claim:

1. A material useful as a pour point depressant consisting essentially of (I) or (II) or mixtures thereof, wherein I is of the formula:



and II is of the formula:



wherein R is a straight chain aliphatic hydrocarbon having from 0 to 1 site of olefinic unsaturation of from 14 to 28 carbon atoms and attached at a secondary carbon atom to the succinyl group;

wherein one of X and X¹ is —NYY¹ and the other is hydroxyl;

wherein one of X² and X³ is —NYY¹ and the other is —OH(NHY²Y³);

wherein Y and Y¹ are aliphatic hydrocarbyl groups each of from 14 to 28 carbon atoms and Y² and Y³ are hydrogen, aliphatic hydrocarbon of from 1 to 30 carbon atoms or oxyaliphatic hydrocarbon of from 1 to 30 carbon atoms, and may be taken together with the nitrogen to which they are attached to form a heterocyclic ring of from 5 to 7 annular members.

2. A material according to claim 1 wherein Y and Y¹ are of from 15 to 22 carbon atoms and have 0 to 2 sites of olefinic unsaturation.

3. A material according to claim 1 wherein Y² and Y³ are aliphatic hydrocarbon of from 15 to 22 carbon atoms, and II is present in from 10 to 30 mole percent.

4. A material according to claim 1 wherein the total number of carbon atoms of Y and Y¹ is in the range of 30 to 52 and the total number of carbon atoms of Y² and Y³ is in the range of 0 to 60.

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DANIEL E. WYMAN, Primary Examiner

W. J. SHINE, Assistant Examiner

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