United States Patent Office 3,321,399
Patented May 23, 1967

3,321,399

PREPARATION OF OIL DISPERSIONS OF METAL CARBONATES

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No Drawing. Filed Oct. 20, 1961, Ser. No. 146,418
5 Claims. (Cl. 252—32.7)

This invention relates to the preparation of stable colloidal dispersions of metal carbonates in hydrocarbon oils including lubricating oils, fuel oils, motor fuels and the like. Such dispersions are useful for imparting corrosion inhibiting properties to lubricating oils and hydrocarbon fuels. The colloidal suspensions are preferably prepared in a hydrocarbon oil medium containing at least one oil-soluble surfactant, thus imparting both corrosion-inhibiting and detergent properties to the composition and making them particularly suitable for use in the crankcases of internal combustion engines.

In the development of modern piston-type internal combustion engines, there has been recognized a continuing need for improved lubricating oils that will have high detergency and that will at the same time possess satisfactory resistance to oxidation and freedom from corrosive tendencies. High detergency oxidation resistant lubricating oils serve to keep the engine free of varnish, sludge and coke-like deposits and thereby promote longer engine life through reduced wear. Heavy duty detergent type lubricating oil compositions must have the ability to maintain a high degree of engine cleanliness. To accomplish this, the compositions must be able to disperse insoluble material that is formed by combustion of the fuel or that results from oxidation of the oil or from both sources together. The lubricating oil composition should be capable of neutralizing acidic materials that may originate either from the oxidation of the lubricating oil or from the combustion products of the fuel or from both sources. Such acidic materials are objectionable not only because they tend to be corrosive but also because they tend to produce lacquer or varnish on the pistons.

In the development of heavy duty lubricants for internal combustion engines it has been established by experience that compositions that possess a high reserve of alkalinity are quite valuable for this type of service in that they will neutralize acidic materials which form during the operation of the engine being lubricated. Colloidal dispersions of alkali metal or alkaline earth metal carbonates in mineral oil base stocks containing oil-soluble surfactants have been recognized as one of the particularly useful types of detergent additives for heavy duty crankcase lubricants.

Previous workers in the art of lubricant additive manufacture have found that colloidal dispersions of metal carbonates in hydrocarbon oils can be prepared by reaction of alkali metal or alkaline earth metal oxides and/or hydroxides with carbon dioxide in the presence of oleophilic surfactants in a hydrocarbon oil medium. For the purposes of the present invention it is intended that magnesium be classed as an alkaline earth metal.

In systems of this type it has been found that, in general, when it is desired to increase the metal content it is necessary to increase the proportion of surfactant also in

order to ensure a stable system. As the surfactant represents a considerable item in the overall cost of the additive composition, there has been an incentive to develop methods for attaining high ratios of metal to surfactant in stable compositions of this type. Stability depends largely upon the degree of fineness of the particles in the colloidal dispersion. This in turn is easily determined by the relative absence of haze in the finished product. A haze rating of more than 100 nephelos on a mixture of 5 weight percent of the additive in white oil is considered unacceptable. A haze rating no greater than about 60 is even more desirable.

It is one object of the present invention to provide a process for preparing stable colloidal dispersions of metal carbonates having relatively high metal-to-surfactant ratios.

It is another object of the present invention to provide a process for the preparation of colloidal dispersions of metal carbonates that is more easily practiced than the previous processes.

It has now been found that stable colloidal dispersions of metal carbonates can be more conveniently produced by hydrolyzing urea in the presence of a metal oxide and/or hydroxide slurried in an oil solution of a suitable surface-active material. This process has a number of advantages over previously known processes, including the avoiding of the storage and transportation of compressed gases in pressure vessels and the critical metering of compressed gases during the reaction. In the process of the present invention, only solids and liquids that are easily handled and measured are required.

In general, the process of the present invention is carried out as follows. One or more oil-soluble or oil-dispersible surfactants are dissolved in a suitable hydrocarbon oil medium. Water is then dispersed in this solution. Following this, the amount of dry urea needed for the process is added and thoroughly dispersed in the mixture. Subsequently, the required amount of dry metal oxide or hydroxide is added to the mixture and thoroughly dispersed, after which the mixture is heated to the reaction temperature. Then water or steam is added until the mixture becomes substantially clear, after which the product is filtered.

A variety of oil-soluble or oil-dispersible surfactants may be used as dispersing agents in practicing this invention. Among the oleophilic surfactants which may be employed are included metal aryl alkyl sulfonates, metal alkyl sulfates, hydrocarbon sulfonic acids, phosphosulfurized hydrocarbons, metal alkyl phenates, metal alkyl phenol sulfates, phosphate esters of high molecular weight alcohols such as those prepared by reaction of phosphorus pentoxide with the oxo alcohols, alkyl thiophosphates, alkyl phosphonates, alkyl thiophosphonates, high molecular weight saturated and unsaturated fatty acids and/or their metal salts, high molecular weight amines and multifunctional polymers such as vinyl acetate/alkyl fumarate/maleic anhydride copolymers.

In many cases it has been found preferable to use a mixture of two or more oil-soluble polar surfactants, or to use a mixture of one or more surfactants and a promoter. A particularly preferred combination is that of a phosphosulfurized polyolefin and an alkylated phenol. Phosphosulfurized hydrocarbons for use in this invention

can be prepared by reacting a sulfide of phosphorus, for example P_2S_5, with a suitable hydrocarbon base stock which, of course, should be one that results in materials that are completely oil soluble after phosphosulfurization. The preferred hydrocarbon starting materials used in this invention are (1) terpenes such as alpha-pinene, (2) heavy petroleum fractions, distillates or residue containing less than 5% of aromatics and having a viscosity at 210° F. of 140 to 250 SSU; and (3) polyolefins having a Staudinger molecular weight in the range of 500 to 200,000 and containing 2 to 6 carbon atoms per olefin monomer. Polybutenes that have Staudinger molecular weights in the range of 700 to 100,000 are particularly preferred.

Preferably the phosphosulfurized hydrocarbon is prepared by reacting approximately four moles of hydrocarbon base stock (e.g. a polyolefin) per mole of phosphorus pentasulfide. A slight excess of phosphorus pentasulfide over the 1 to 4 mole ratio can be used to insure complete phosphosulfurization. The phosphosulfurization reaction is conducted under anhydrous conditions at temperatures of 150° to 600° F. for a period in the range of 0.5 to 15 hours. A very slight amount of an alkyl phenol or alkyl phenol sulfide, preferably in the range of 0.001 to 1.0 percent by weight, can be added as a catalyst in the phosphosulfurization reaction. It has also proven very useful to treat or blow the phosphosulfurized product with an inert gas such as nitrogen for a period of 10 min. to 2 hours to aid in reducing hydrogen sulfide evolution and its corresponding odor. The preparation of phosphosulfurized hydrocarbons and the use of catalysts in the phosphosulfurization reaction are more fully described in U.S. Patent 2,875,188.

The alkyl phenol component of the reaction mixture is preferably one having a molecular weight in the range of from about 200 to about 700. For example, the alkylation product of phenol with diisobutylene or with tripropylene, i.e. nonyl phenol, may be used. In some cases it is possible to substitute for a portion or all of the alkyl phenol an alkyl phenol sulfide, which is the thioether or an alkyl phenol, i.e. a compound in which the alkylated phenol groups are joined by a divalent sulfur atom. The alkyl phenols are conveniently converted to alkyl phenol sulfides by reaction with sulfur dichloride.

The sulfonates used are also well known in the art. The sulfonic acids can be obtained through the sulfonation of either synthetic or natural hydrocarbons. The preferred sulfonic acids have molecular weights in the range of 300 to 700 (as the sodium soap). The synthetic acids preferably have molecular weights in the narrower range of 400 to 600. The acids can contain more than one sulfonyl group in the molecule. Suitable sulfonic acids are produced by sulfonating alkyl aromatic hydrocarbons such as didodecyl benzene. They can also be obtained by treatment of lubricating oil base stocks with concentrated or fuming sulfuric acid in a conventional manner to produce oil-soluble "mahogany" acids.

Specific examples of suitable sulfonates include calcium petroleum sulfonate, barium petroleum sulfonate, calcium di-C_9 alkyl benzene sulfonate (C_9 group from tripropylene), and barium C_{16} alkyl benzene sulfonate (C_{16} group from tetraisobutylene). The sulfonates may be of either the neutral type or of the "over-based" or "high alkalinity" type, containing metal base in excess of that required for simple neutralization, wherein the excess metal base has been neutralized with carbon dioxide.

Metal salts of alkyl phenols and of alkyl phenol sulfides are also well known in the art. Metal salts of alkyl phenols having alkyl groups of from 5 to 20 carbon atoms are preferred, and the metal used to form the phenate is preferably an alkaline earth metal, e.g. calcium or barium although the salts such as those of aluminum, cobalt, lead or tin are not excluded. A specific example is the barium salt of the alkylation product of phenol with tripropylene. Metal salts of the corresponding alkyl

phenol sulfides may also be used. The latter are the thioethers and polysulfides of alkyl phenols, i.e. compounds in which the alkyl groups are joined by one or more divalent sulfur atoms. The alkyl phenols can be converted to phenol sulfides by reaction with sulfur dichloride. If sulfur monochloride is used, the resulting products are primarily alkyl phenol disulfides.

High molecular weight amines that may be used as surfactants include those having from 12 to 20 carbon atoms such as the commercial product sold under the trade name Duomeen–T and the product known as Primene 81R, which is a mixture of tertiary alkyl primary amines of from 15 to 18 carbon atoms.

Oxo alcohols are well known to the art, being prepared by reaction of olefins with carbon monoxide and hydrogen in the presence of a cobalt or similar catalyst to form aldehydes of one more carbon atom than the starting olefins, followed by catalytic hydrogenation of the corresponding alcohols. Mixed phosphate esters of these and related alcohols, which may be used as surfactants in the practice of this invention, may be prepared by reacting one mole of P_2O_5 with from 2 to 6 moles, and preferably 2 to 4 moles of a suitable alcohol of say from 6 to 16 atoms per molecule at temperatures of from 50° to 200° F. for from ¼ to 4 hours. Depending upon the reaction conditions and the particular alcohols used, the reaction products will vary in composition but will usually comprise chiefly a mixture of the orthophosphoric acid mono and di-esters of the alcohols, along with minor amounts of unreacted alcohols, orthophosphoric acid, and condensed polyphosphates.

Other surfactants that may be used include various copolymers such as one derived by reacting mixed fumarates (e.g. tallow fumarates and C_8 oxo fumarates averaging about 420 molecular weight) with vinyl acetate (3 to 1 acetate to fumarate ratio) and a small proportion of maleic anhydride (e.g. 3 weight percent) and subsequent removing excess vinyl acetate. Other copolymer surfactants includes copolymers of mixed C_4–C_{18} methyl methacrylates with 5 to 15 weight percent of N-vinyl pyrrolidone and copolymers of 65 to 85 weight percent of mixed C_9 to C_{12} fumarates, 10 to 20 weight percent of vinyl acetate and 5 to 15 weight percent of N-vinyl pyrrolidone.

The procedure employed in the process of the present invention involves as the first step preparing a solution or dispersion of the oleophilic surfactant in a hydrocarbon oil. The hydrocarbon oil may comprise any fraction that has a sufficiently high boiling point so that it will not vaporize under the conditions of reaction or during the finishing steps in the preparation of the additive. Any hydrocarbon in the lubricating oil range may be used as the hydrocarbon medium. Light lubricating oil fractions are particularly suitable. The concentration of surfactants in the hydrocarbon oil will depend somewhat on its nature but will generally be in the range of about 5 to 70 weight percent, and preferably about 20 to 70 weight percent. As noted above, it is preferred to use either a mixture of surfactants or a surfactant in conjunction with a promoter such as an alkyl phenol of 200 to 700 molecular weight.

As stated previously, water and the required amounts of dry urea and of dry metal oxides and/or hydroxides are then sequentially dispersed in the oil medium. The reaction mixture is then heated to the desired temperature and water or steam is added until a clear solution is obtained.

Reaction temperatures may range from about 150° to about 300° F. but the preferred range is from about 220° to about 270° F. in order to accelerate the rate of hydrolysis of the urea. During this hydrolysis a slow controlled release of carbonic acid occurs so that the carbonates that are formed are of the desired degree of fineness and thus will be colloidally dispersed.

The broad and preferred temperature ranges for each of the steps of the process are given in Table I.

TABLE I

	Process Step	Temperature Range, ° F.	
		Broad	Preferred
1	Blend mineral oil and surfactants and promoters.	60–160	100–120
2	Disperse water in blend	60–160	100–120
3	Disperse dry urea	60–160	100–130
4	Add metal oxide and/or hydroxide and disperse.	60–200	170–190
5	Heat to reaction temperature	150–300	220–270
6	Add water and/or steam	150–300	220–270
7	Dehydrate by heating at least 10 minutes.	[1] 150–400	300–370
8	Filter	250–370	300–360

[1] With air or nitrogen blowing if temperatures are in the low range.

Reaction times, i.e. in step 6, may range from about 2 to 6 hours and are preferably in the range of 3 to 4 hours. Addition of the steam and/or water is preferably conducted slowly, i.e. dropwise over the reaction period. While the initial charge of water, i.e. step 2, may be dispensed with, it is preferably to employ it, because the added urea dissolves in the dispersed water (complete solution occurs at 165° F.) and thus promotes a finer dispersion of urea in the reaction medium than is attained when merely adding the dry urea without the prior addition of water. This in turn enhances the fineness of the particles in the finished colloidal dispersion. It should be noted that this initial charge of water is relatively small in comparison to the total amount of water used in the process, as sufficient additional water or steam is needed to hydrolyze the urea completely. Preferably this amounts to at least 80 weight percent of the reacting materials. The initial water charge is preferably in the range of 5 to 8 weight percent on the same basis.

The proportions of reacting materials may vary within the ranges shown in Table II.

TABLE II

Reactants	Weight Percent	
	Broad	Preferred
Surfactant	14–24	19–22
Promoter (e.g. Alkyl Phenol)	5–12	9–11
Diluent Oil	25–45	35–42
Water (Initial Charge)	0–8	3–5
Urea	5–25	8–16
Metal Oxide	8–30	12–17

One-half to one and one-half chemical equivalents of urea to metal ion may be used, but 0.85 to 1.0 chemical equivalent of urea to metal ion is preferred.

The following examples illustrate the manner in which this invention may be practiced.

EXAMPLE 1

A phosphosulfurized hydrocarbon was prepared by reacting 100 parts by weight of a polybutene having an average Staudinger molecular weight of about 780 with 15 parts by weight of P_2S_5 for about 10 hours at about 450° F. The product was then diluted with 30 weight percent of light mineral oil for ease of handling. The 70 weight percent concentrate analyzed 2.36 weight percent phosphorus and 4.70 weight percent sulfur.

EXAMPLE 2

Following the general procedure outlined in Table I, a detergent additive was prepared using as the feed 20.7 weight percent (active ingredient basis) of the phosphosulfurized polybutene of Example 1, 10.0 weight percent of nonyl phenol (derived by alkylation of phenol with tripropylene), 16.8 weight percent of calcium hydroxide, 12.6 weight percent of urea, and 39.9 weight percent of mineral oil diluent (surfactant diluent plus neutral mineral oil of 155 SSU viscosity at 100° F.). The initial water charge was 6.2 percent by weight based on the other components, and the water added during the reaction amounted to an additional 109 weight percent on the same basis. The general procedure was varied to the extent that ⅔ of the initial charge, and then the urea, were added at 110° F., and the remaining ⅓ of the initial water charge and the calcium hydroxide were added at 170–200° F. The remaining water for steaming was added dropwise over a 3 to 4 hour period, using a reaction temperature of 245° F. The product was dehydrated by heating to 370° F. and holding it at that temperature for 10 minutes. Filtration was then conducted in the temperature range of 360–260° F. The resulting additive concentrate had the composition shown in Table III. The ratio of calcium equivalents to surfactants (calculated on the basis that all of the phosphorus in the phosphosulfurized hydrocarbon is available as a monobasic acid) in the product was about 6.3.

The haze reading of the product in 5 weight percent concentration in white oil was 54 nephelos.

Table III

	Product composition weight percent
Calcium oxide	12.2
Carbon dioxide	8.8
Phosphosulfurized hydrocarbon	23.8
Nonyl phenol	11.3
Diluent mineral oil	43.8

EXAMPLE 3

A solution of 300 grams of the P_2S_5-treated polybutene concentrate of Example 1 (70 weight percent concentrate), 67 grams of nonyl phenol, 31.2 grams of ammonium sulfate concentrate of 450 molecular weight (50 weight percent concentrate in oil) and 318 grams of a light mineral lubricating oil was prepared by blending the ingredients at room temperature. Then 42 grams of water was thoroughly dispersed in this mixture. Following this, 132 grams of dry powdered urea was dispersed in the mixture and then 165 grams of hydrated lime was similarly dispersed. The temperature was raised to 155° F. and then slowly raised over a period of 4 hours to a final temperature of 300° F. At about 155° F. the dispersion became noticeably darker, indicating that the urea had gone into solution. At this temperature also the first evolution of gas was noted, indicating the start of urea decomposition. The product was filtered and gave a clear fluid product analyzing 23.4 weight percent sulfated ash and 6.88 weight percent calcium.

EXAMPLE 4

The same composition as in Example 3 was used. The temperature was raised to 240° F. and maintained between 240 and 260° F. while 790 grams of water was added over a 3-hour period. The clear solution was heated to 300° F. and was filtered to give an oily product, which had a sulfated ash of 30.5 wt. percent and 8.96 wt. percent calcium.

EXAMPLE 5

The same composition as in Example 3 was again used. The temperature was raised to 255° F. and maintained between 255 and 260° F. while 800 grams of water was added dropwise over a 2.5 hour period. The temperature was then raised to 300° F. and filtered to give a fluid product, which had a sulfated ash of 30.1 wt. percent and a calcium content of 8.85 wt. percent.

EXAMPLE 6

The same composition as in Example 3 was used, except the quantity of urea was reduced to 114 grams. The temperature was raised to 240° F. and maintained be-

tween 240 and 260° F. for 4.5 hours while 900 grams of water was added slowly. The temperature was raised to 300° F. and the product was then filtered to a fluid product of 27.8 wt. percent sulfated ash and 8.2 wt. percent calcium.

EXAMPLE 7

In an oil solution of 350 grams of nonyl phenol in 360 grams of a light lubricating oil, 52 grams of water was dispersed, followed by 214 grams of calcium hydroxide and an additional 25 grams of water. The temperature was then raised to 245° F. and maintained at that point for 2.5 hours while an additional 1300 grams of water was added slowly. The product obtained was opaque and could not be filtered. This indicates that nonyl phenol alone is not a satisfactory surfactant for the process.

EXAMPLE 8

To study the effect of the amount of water used in the reaction, Example 2 was repeated, but instead of the initial water charge of 6.2 weight percent and the additional water charge of 109 weight percent, there were used 4.5 weight percent initial charge and 55 weight percent additional water charge, and the reaction time was increased 40 percent in order to make more efficient use of the hydrolyzing steam. The product had a somewhat lower calcium content (11.7% vs. 12.2 wt. percent CaO) than the product of Example 2 and was not satisfactory from the standpoint of haze, giving a nephelometer reading of more than 130 nephelos when tested in 5 weight percent concentration in white oil.

EXAMPLE 9

A further study of the effect of total water used in the reaction was made using the general reaction conditions of Example 2 and employing the same quantities of phosphosulfurized polybutene, nonyl phenol, ammonium sulfonate and light mineral oil as in Example 3. In reaction A there were used 42 grams of water initially, followed by 134 grams of urea, and then 55 grams of additional water during the reaction. In reaction B 61 grams of water was used initially, then 124 grams of urea, and 800 grams of additional water during the reaction. The two runs are compared in Table IV, which lists the water percentages used, the haze ratings of the respective products and the rates of filtration of the reaction mixtures in the respective preparations. The filtration rates are based on the time required to filter 1000 grams of the material through a 12.5 cm. heated Buchner funnel using 5 weight percent admixed Dicalite Speedflow filter aid and precoating the filter with 2.5 weight percent of the filter aid. The high haze reading and the slower filtration rate are both indications of an unsatisfactory product in the case of reaction A.

From the results of a number of tests it was concluded that the use of at least 80 weight percent water based on the reactants is necessary during the steaming reaction to ensure satisfactory products.

TABLE IV.—EFFECT OF WATER ON PRODUCT QUALITY

Reaction	Water Used as Percent of Other Componebts		Product Haze Nephelos (1)	Filter Rate, gal./hr./sq. ft.
	Initially	During Steaming		
A	4.5	5.1	<130	1.7
B	6.7	88.0	45	4.1

[1] In 5 wt. percent concentration in white oil.

The colloidal dispersions prepared in accordance with this invention are in the nature of additive concentrates containing up to about 70 weight percent of surfactant material and up to about 10 weight percent of metal. These concentrates may be added to any of several types of hydrocarbons ranging from fuel oils through lubricating oils. The magnesium compounds are of particular value for addition to residual fuel oils containing vanadium compounds. The latter are objectionable because the ash tends to be corrosive to metal parts exposed to high temperatures. Magnesium compounds combat this corrosion. For this use sufficient of the colloidal dispersion of $MgCO_3$, for example, will be added to furnish from 0.5 to 4.5 parts of magnesium per part of vanadium. The total amount needed will of course depend on the vanadium content of the fuel oil, which may range anywhere from 5 to 1000 parts per million, for example. Magnesium carbonate dispersions may also be useful in fuel oils to reduce corrosion caused by sulfur, as by preventing or suppressing sulfur trioxide formation.

For lubricating oil compositions the colloidal dispersions prepared by this invention may be added in sufficient quantity to furnish from 0.01 to about 3 weight percent of metal, depending upon the particular use. In the case of calcium-containing materials, concentrations providing from about 0.02 to about 0.5 weight percent calcium are advantageous in crankcase lubricants for example, while for marine diesel lubrication concentrations providing as much as 2 weight percent calcium might be used. The oil compositions may contain other additives such as other detergents, sludge dispersers, viscosity index improvers, e.g. polymethacrylates, polybutenes, etc., antioxidants such as phenyl-alpha-naphthylamine, alkyl phenols, bis phenols and the like, pour point depressants, dyes, anti-wear agents such as zinc dialkyldithiophosphates, and other additives for improving the properties of the compositions.

As a specific example, a crankcase lubricant incorporating an additive of the present invention may consist of 98 volume percent of an SAE 30 grade highly refined base stock, 0.3 volume percent of a viscosity index improver, 0.7 volume percent of a zinc dialkyl dithiophosphate (e.g. one having C_4 and C_5 alkyl groups) and 1 volume percent of an additive concentrate made as described in Example 2.

While lubricants for use in internal combustion engine crankcases are particularly contemplated, transmission lubricants, gear oils, flushing oils, hydraulic oils, greases, industrial lubricants and the like are also within the purvue of the invention. The lubricating oil base stock may be of any desired type, i.e. those derived from the ordinary paraffinic, naphthenic, asphaltic or mixed base crude oils by suitable refining methods.

It will be understood that the foregoing examples are merely illustrative of the invention and are not intended to limit its scope.

What is claimed is:

1. An improved process for preparing a colloidal dispersion of a metal carbonate in a mineral oil which comprises:

(1) preparing a blend of from 14 to 24 parts by weight of an oleophilic surfactant, from 5 to 12 parts by weight of an alkyl phenol of from 200 to 700 molecular weight, and from 25 to 45 parts by weight of mineral oil;

(2) dispersing in said blend, at a temperature in the range of from about 60° to about 160° F., from 0 to 8 parts by weight of water, from 5 to 25 parts by weight of dry urea and from 8 to 30 parts by weight of a metal compound selected from the group consisting of the oxides and hydroxides of alkali metals and of alkaline earth metals, the quantity of urea thereupon being present being in the range of from 0.5 to 1.5 chemical equivalents per chemical equivalent of said metal;

(3) heating said mixture to a temperature in the range of about 150° to about 300° F.;

(4) adding to the heated mixture at said last named temperature gradually over a period of from 2 to 6 hours a hydrolyzing agent, selected from the group

consisting of water and steam, in the proportion of at least 80 parts of hydrolyzing agent per 100 parts of the other components of the reaction mixture;

(5) and thereafter dehydrating said mixture at a temperature in the range of about 150° to 400° F.,

said surfactant consisting of at least one material selected from the group consisting of phosphosulfurized hydrocarbons, metal salts of hydrocarbon sulfonic acids of 300 to 700 molecular weight, metal salts of C_5 to C_{20} alkyl phenols, metal salts of C_5 to C_{20} alkyl phenol sulfides, C_{12} to C_{20} alkyl amines, C_6 to C_{16} alcohol phosphates, alkyl fumarate-vinyl acetate-maleic acid copolymers, fumarate-vinyl acetate-vinyl pyrrolidone copolymers, and alkyl methacrylate-vinyl pyrrolidone copolymers.

2. Process as defined by claim **1** wherein said metal compound comprises calcium hydroxide and said surfactant comprises a phosphosulfurized hydrocarbon.

3. Process as defined by claim **4** wherein said surfactant comprises a phosphosulfurized hydrocarbon.

4. Process as defined by claim **1** wherein from 5 to 8

weight percent of water, based on the total weight of the components of the reacting mixture other than water, is incorporated prior to the addition of urea and metal compound, thereby promoting the efficiency of dispersion of the urea in said reaction mixture.

5. Process as defined by claim **1** wherein said alkyl phenol is nonyl phenol.

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