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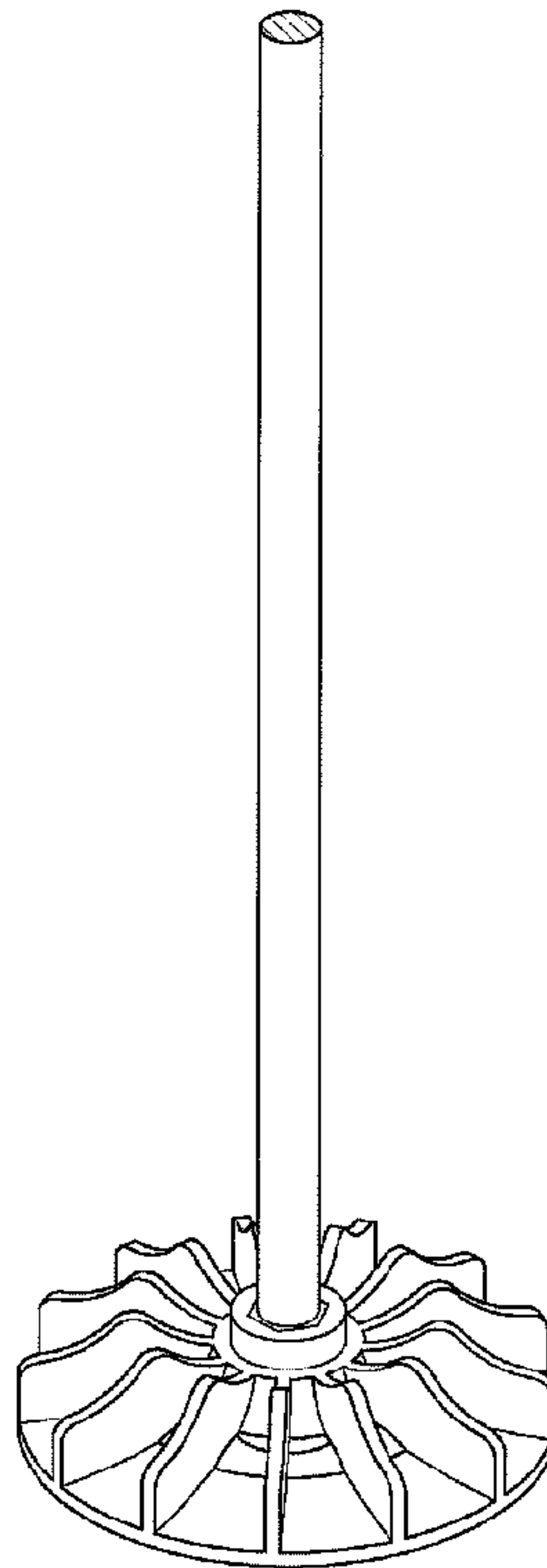
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(54) Titre : COPOLYMERES DE REDUCTION DE TRAINEE POUR APPLICATIONS A FLUIDE FROID

(54) Title: DRAG REDUCING COPOLYMERS FOR COLD FLUID APPLICATIONS



(57) Abrégé/Abstract:

Drag reducing compositions comprising polymer particles, where the polymer particles include copolymers comprising the residues of at least one methacrylate monomer and at least one comonomer having a polymerizable vinyl group, where the comonomer has no more than one pendant substituent per vinyl carbon. The drag reducing compositions can have improved dissolution rates in hydrocarbon-containing fluid to decrease the pressure drop associated with the turbulent flow of the hydrocarbon-containing fluid through a conduit.

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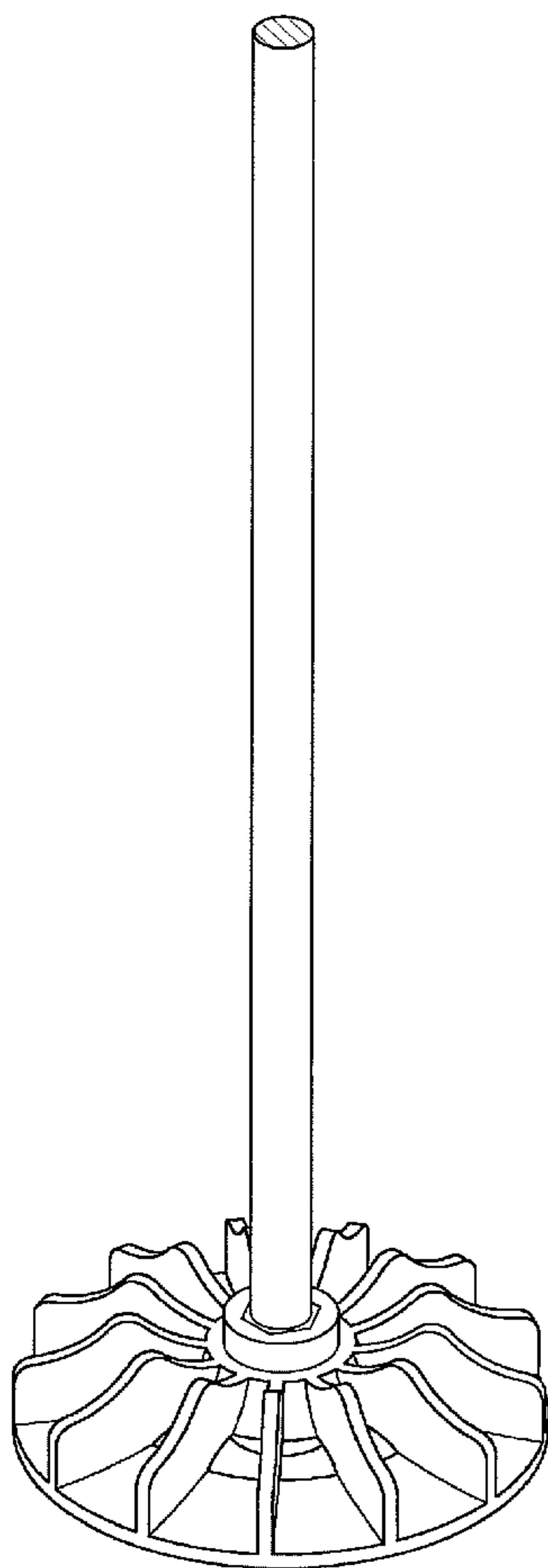


FIG. 2

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DRAG REDUCING COPOLYMERS FOR COLD FLUID APPLICATIONS

BACKGROUND OF THE INVENTION

5 1. Field of the Invention

The present invention relates generally to drag reducing compositions comprising copolymers. More specifically, the present invention relates to drag reducing compositions having copolymers comprising the residues of at least one methacrylate monomer and at least one comonomer having no more than one pendant substituent per
10 vinyl carbon.

2. Description of the Prior Art

When fluids are transported by a pipeline, a drop in fluid pressure typically occurs due to friction between the wall of the pipeline and the fluid. Due to this pressure drop, for a given pipeline, fluid must be transported with sufficient pressure to achieve a
15 desired throughput. When higher flow rates are desired through the pipeline, more pressure must be applied due to the fact that as flow rates are increased the difference in pressure caused by the pressure drop also increases. However, design limitations on pipelines limit the amount of pressure that can be employed. The problems associated with pressure drop are most acute when fluids are transported over long distances. Such
20 pressure drops can result in inefficiencies that increase equipment and operation costs.

To alleviate the problems associated with pressure drop, many in the industry utilize drag reducing additives in the flowing fluid. When the flow of fluid in a pipeline is turbulent, high molecular weight polymeric drag reducers can be employed to enhance the flow. A drag reducer is a composition capable of substantially reducing friction loss
25 associated with the turbulent flow of fluid through a pipeline. The role of these additives is to suppress the growth of turbulent eddies, which results in higher flow rate at a constant pumping pressure. Ultra-high molecular weight polymers are known to function well as drag reducers, particularly in hydrocarbon liquids. In general, drag reduction depends in part upon the molecular weight of the polymer additive and its
30 ability to dissolve in the hydrocarbon under turbulent flow. Effective drag reducing polymers typically have molecular weights in excess of five million. However, many

conventional drag reducers do not perform well in colder temperature fluids, which may be due to lower dissolution rates caused by lower temperatures.

SUMMARY OF THE INVENTION

5 In one embodiment of the present invention, there is provided a drag reducing composition comprising: a continuous phase and a plurality of drag reducing polymer particles. At least a portion of the polymer particles comprise copolymers containing the residues of at least one methacrylate monomer and the residues of at least one comonomer, and the comonomer comprises a polymerizable vinyl group having no more
10 than one pendant substituent per vinyl carbon.

 In another embodiment of the present invention, there is provided a method of making a drag reducer. The method of this embodiment comprises: (a) combining water, at least one surfactant, at least one methacrylate monomer, and at least one comonomer to thereby form a reaction mixture; and (b) subjecting the reaction mixture
15 to emulsion polymerization or suspension polymerization to thereby form a latex comprising a continuous phase and a plurality of drag reducing copolymer particles comprising residues of the methacrylate monomer and residues of the comonomer, where the comonomer comprises a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon.

20 In yet another embodiment of the present invention, there is provided a method for reducing the pressure drop associated with the turbulent flow of a hydrocarbon-containing fluid through a pipeline. The method of this embodiment comprises: (a) introducing a drag reducing composition into the hydrocarbon-containing fluid, where the drag reducing composition is in the form of a latex or suspension comprising a
25 continuous phase and a plurality of drag reducing polymer particles comprising copolymers containing the residues of at least one methacrylate monomer and the residues of at least one comonomer, where the comonomer comprises a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon; and (b) flowing the resulting treated hydrocarbon-containing fluid through said pipeline. The
30 hydrocarbon-containing fluid of this embodiment has an average temperature of 25°C or less.

In accordance with another embodiment of the present invention, there is provided a mixture comprising a hydrocarbon-containing fluid and a drag reducing composition, the drag reducing composition comprising: a continuous phase and a plurality of drag reducing polymer particles, wherein at least a portion of said polymer particles comprise
5 copolymers containing from about 40 wt% to about 95 wt% of at least one methacrylate monomer and from about 5 wt% to about 60 wt% of at least one comonomer, wherein said comonomer comprises a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon, wherein said drag reducing composition reduces a pressure drop associated with a turbulent flow of said hydrocarbon-containing fluid through a
10 conduit, and the hydrocarbon-containing fluid has an average temperature of 25°C or less.

In accordance with another embodiment of the present invention, there is provided a method of forming a mixture, said method comprising: (a) combining water, at least one surfactant, at least one methacrylate monomer, and at least one comonomer to thereby form a reaction mixture; (b) subjecting said reaction mixture to polymerization to thereby form
15 a composition comprising a continuous phase and a plurality of drag reducing copolymer particles comprising from about 40 wt% to about 95 wt% of said methacrylate monomer and from about 5 wt% to about 60 wt% of said comonomer, wherein said comonomer comprises a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon; and (c) adding the drag reducing composition to a hydrocarbon-containing
20 fluid having an average temperature of 20°C or less.

In accordance with another embodiment of the present invention, there is provided a method for reducing the pressure drop associated with the turbulent flow of a hydrocarbon-containing fluid through a pipeline, said method comprising: (a) introducing a drag reducing composition into said hydrocarbon-containing fluid, wherein said drag
25 reducing composition comprises a continuous phase and a plurality of drag reducing polymer particles comprising copolymers containing from about 40 wt% to about 95 wt% of at least one methacrylate monomer and from about 5 wt% to about 60 wt% of at least one comonomer, wherein said comonomer comprises a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon; and (b) flowing the resulting
30 treated hydrocarbon-containing fluid through said pipeline, wherein said hydrocarbon-containing fluid has an average temperature of 20°C or less.

In accordance with another embodiment of the present invention, there is provided a drag reducing composition, comprising: a continuous phase; and a plurality of drag reducing polymer particles, wherein at least a portion of the polymer particles comprises copolymers containing from about 10 wt% to about 99 wt% of at least one methacrylate monomer and from about 1 wt% to about 90 wt% of at least one comonomer, wherein the comonomer comprises a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon, wherein the drag reducing composition reduces a pressure drop associated with a turbulent flow of a hydrocarbon fluid having an average temperature of 20°C or less.

BRIEF DESCRIPTION OF THE DRAWING FIGURES

A preferred embodiment of the present invention is described in detail below with reference to the attached drawing figures, wherein:

5 FIG. 1 is a schematic illustration of a test apparatus used to perform dissolution rate tests on various drag reducers;

FIG. 2 is an isometric view of the stirrer employed in the dissolution rate tests;

FIG. 3 is a top view of the stirrer employed in the dissolution rate tests; and

FIG. 4 is a side view of the stirrer employed in the dissolution rate tests.

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DETAILED DESCRIPTION

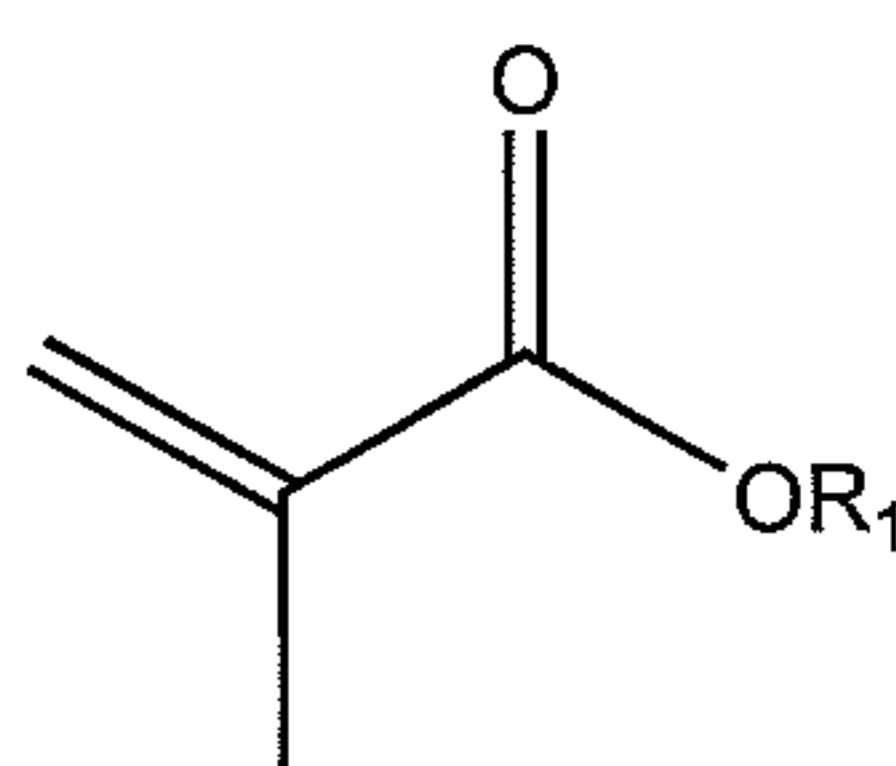
In accordance with one embodiment of the present invention, a drag reducing composition (i.e., a drag reducer) is provided comprising a plurality of drag reducing polymer particles comprising copolymers containing the residues of at least two different
15 monomers. The drag reducer of the present invention can be employed to at least partially reduce the pressure drop associated with the turbulent flow of a hydrocarbon-containing fluid through a conduit (e.g., a pipeline).

In one embodiment of the present invention, the above-mentioned drag reducing composition can comprise polymer particles containing copolymers formed via emulsion
20 polymerization or suspension polymerization of a reaction mixture comprising at least two monomers, a continuous phase, at least one surfactant, and an initiation system. As used herein, the terms “emulsion polymer” and “emulsion drag reducing polymer” shall denote any polymer prepared via emulsion polymerization. As discussed in greater detail below, the resulting reaction product of the emulsion polymerization can be in the
25 form of a latex drag reducer composition. As used herein, the terms “suspension polymer” and “suspension drag reducing polymer” shall denote any polymer prepared via suspension polymerization. As discussed below, the resulting reaction product of the suspension polymerization can be in the form of a suspension drag reducer composition.

The continuous phase of the drag reducer composition can generally comprise at
30 least one component selected from the group consisting of water, one or more alcohols, one or more polyols, and mixtures thereof. When water is the selected constituent of the

continuous phase, the reaction mixture can also comprise a buffer. Additionally, as described in more detail below, the continuous phase can optionally comprise a hydrate inhibitor. As described in more detail below, a stabilizer optionally can be added.

As mentioned above, the copolymer of the drag reducing compositions of the present invention can comprise the residues of at least two different monomers. In one embodiment, at least one of the monomers employed in producing the drag reducing composition can be a methacrylate monomer. Methacrylate monomers suitable for use in the present invention can have the following structure:



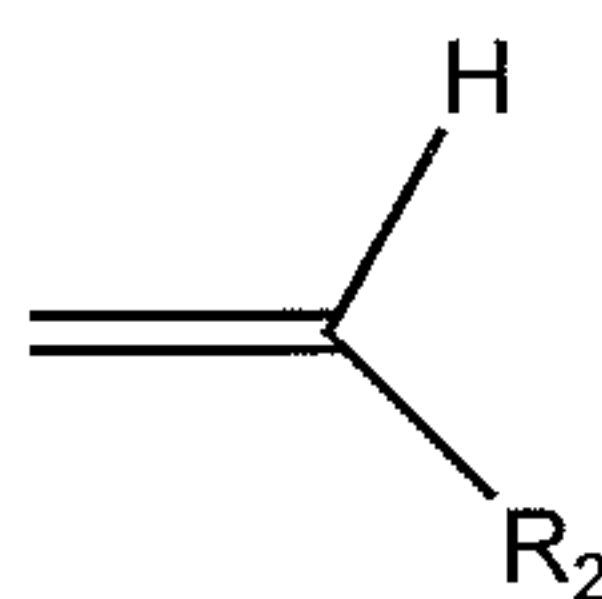
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In one embodiment, R_1 of the above structure can comprise a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl. In another embodiment, methacrylate monomers useful in the present invention can comprise C_4 - C_{20} alkyl, isoalkyl, cycloalkyl, C_6 - C_{20} substituted or unsubstituted aryl, or aryl-substituted C_1 - C_{10} alkyl esters of methacrylic acid. In still another embodiment, the methacrylate monomer employed in the present invention can comprise 2-ethylhexyl methacrylate.

The second monomer (a.k.a., comonomer) employed in making the drag reducing compositions of the present invention can comprise any monomer having a polymerizable vinyl group that has no more than one pendant substituent per vinyl carbon. As used herein, the term “polymerizable vinyl group” shall denote the carbon/carbon double bond of the monomer where polymerization of that monomer occurs. As used herein, the term “pendant substituent” shall denote any non-hydrogen atom or moiety covalently bonded to the vinyl carbon atom.

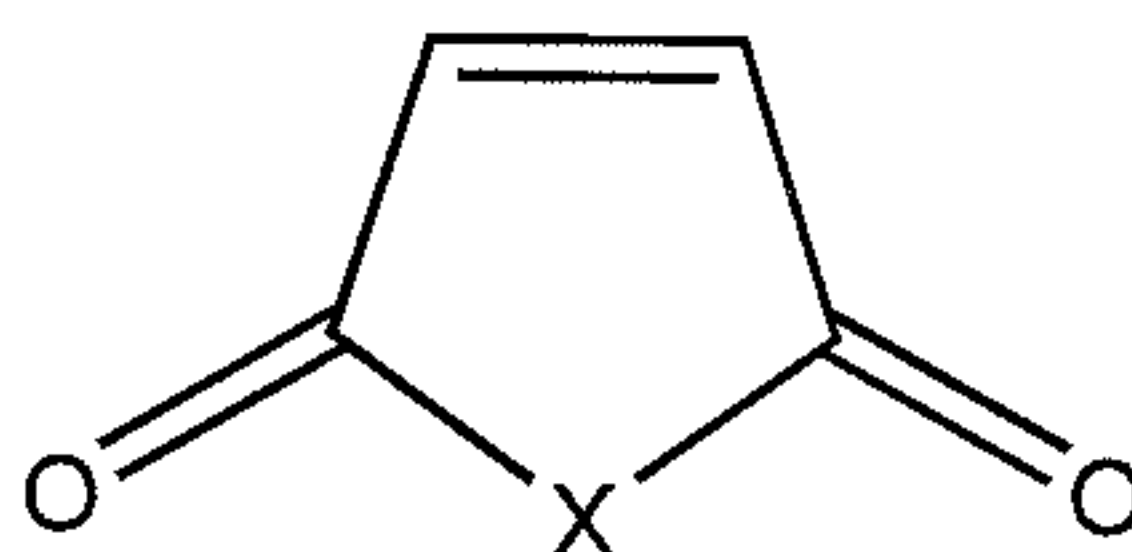
In one embodiment, the comonomer of the present invention can comprise one or more of the following structures:

a)



where R₂ can be -C(O)OR₃, phenyl or aryl, heterocyclic radical, such as, for example, a pyridinyl or pyridyl, -OR₄, or -O(O)CR₅; where R₃ can be a C₁ to C₂₀ alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; where R₄ can be a C₁ to C₂₀ alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; where R₅ can be a C₁ to C₂₀ alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; and/or

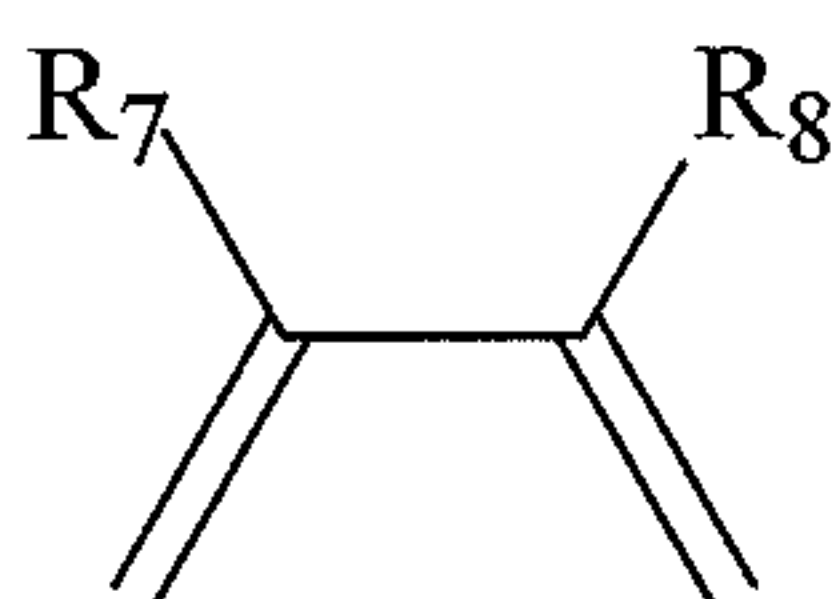
b)



10

where X can be O, or N-R₆, wherein R₆ is H, C₁ to C₂₀ alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl, or ring-opened alkyl diesters; and/or

c)

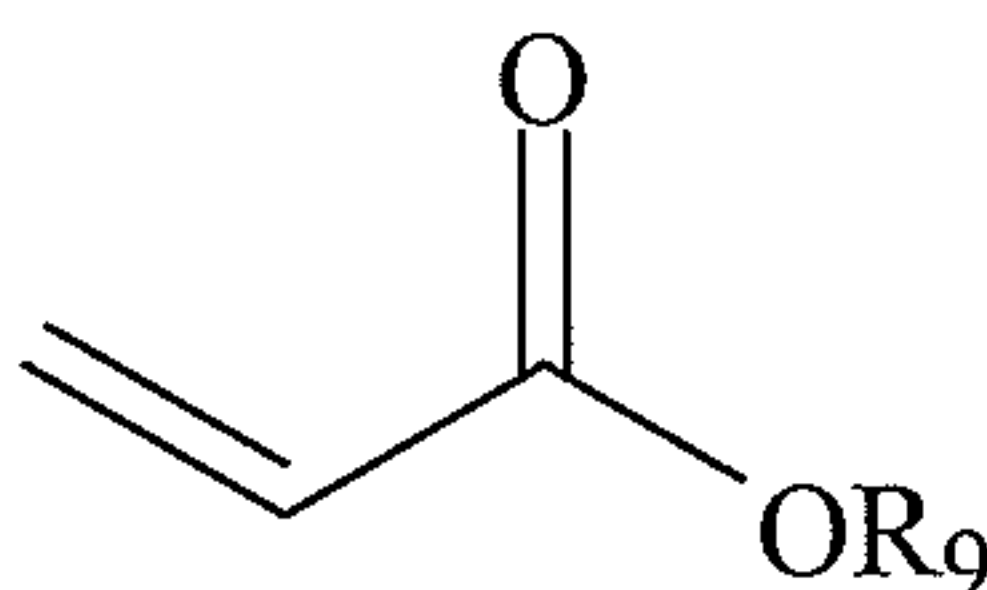


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where R₇ and R₈ can independently be H, C₁ to C₂₀ alkyl, isoalkyl, cycloalkyl, aryl, aryl-substituted alkyl, or heterocyclic radical.

20

In another embodiment, comonomers useful in the present invention can have the following structure:



The R₉ moiety in the above structure can be a straight or branched aliphatic alkyl group having 8 or fewer carbon atoms, 6 or fewer carbon atoms, or 4 or fewer carbon atoms. Additionally, the comonomer can be selected from the group consisting of n-butyl
 5 acrylate; 2-ethylhexyl acrylate; 3,5,5-trimethylhexyl acrylate; tert-butyl acrylate; 2-phenylethyl acrylate; iso-butyl acrylate; isobornyl acrylate; n-hexyl acrylate; sec-butyl acrylate; ethyl acrylate; and lauryl acrylate. Furthermore, in one embodiment the comonomer can comprise n-butyl acrylate.

The surfactant used in the above-mentioned reaction mixture can include at least
 10 one high HLB anionic or nonionic surfactant. The term “HLB number” refers to the hydrophile-lipophile balance of a surfactant in an emulsion. The HLB number is determined by the methods described by W.C. Griffin in *J. Soc. Cosmet. Chem.*, 1, 311 (1949) and *J. Soc. Cosmet. Chem.*, 5, 249 (1954).

As used herein, the term “high HLB” shall denote an HLB number of 7 or
 15 more. The HLB number of surfactants for use with forming the reaction mixture can be at least about 8, at least about 10, or at least 12.

Exemplary high HLB anionic surfactants include, but are not limited to, high HLB alkyl sulfates, alkyl ether sulfates, dialkyl sulfosuccinates, alkyl phosphates, alkyl aryl sulfonates, and sarcosinates. Suitable examples of commercially available high
 20 HLB anionic surfactants include, but are not limited to, sodium lauryl sulfate (available as RHODAPON[®] LSB from Rhodia Incorporated, Cranbury, NJ), dioctyl sodium sulfosuccinate (available as AEROSOL[®] OT from Cytec Industries, Inc., West Paterson, NJ), 2-ethylhexyl polyphosphate sodium salt (available from Jarchem Industries Inc., Newark, NJ), sodium dodecylbenzene sulfonate (available as NORFOX 40 from
 25 Norman, Fox & Co., Vernon, CA), and sodium lauroylsarcosinic (available as HAMPOSYL[®] L-30 from Hampshire Chemical Corp., Lexington, MA).

Exemplary high HLB nonionic surfactants include, but are not limited to, high HLB sorbitan esters, PEG fatty acid esters, ethoxylated glycerine esters, ethoxylated fatty amines, ethoxylated sorbitan esters, block ethylene oxide/propylene oxide
 30 surfactants, alcohol/fatty acid esters, ethoxylated alcohols, ethoxylated fatty acids, alkoxylated castor oils, glycerine esters, linear alcohol ethoxylates, and alkyl phenol

ethoxylates. Suitable examples of commercially available high HLB nonionic surfactants include, but are not limited to, nonylphenoxy and octylphenoxy poly(ethyleneoxy) ethanols (available as the IGEPAL[®] CA and CO series, respectively from Rhodia, Cranbury, NJ), C8 to C18 ethoxylated primary alcohols (such as
5 RHODASURF[®] LA-9 from Rhodia Inc., Cranbury, NJ), C11 to C15 secondary-alcohol ethoxylates (available as the TERGITOL[®] 15-S series, including 15-S-7, 15-S-9, 15-S-12, from Dow Chemical Company, Midland, MI), polyoxyethylene sorbitan fatty acid esters (available as the TWEEN[®] series of surfactants from Uniquema, Wilmington, DE), polyethylene oxide (25) oleyl ether (available as SIPONIC[™] Y-500-70 from
10 Americal Alcolac Chemical Co., Baltimore, MD), alkylaryl polyether alcohols (available as the TRITON[®] X series, including X-100, X-165, X-305, and X-405, from Dow Chemical Company, Midland, MI).

In one embodiment, the initiation system for use in the above-mentioned reaction mixture can be any suitable system for generating free radicals necessary to facilitate
15 emulsion polymerization or suspension polymerization. Possible initiators include, but are not limited to, persulfates (e.g., ammonium persulfate, sodium persulfate, potassium persulfate), peroxy persulfates, and peroxides (e.g., tert-butyl hydroperoxide) used alone or in combination with one or more reducing components and/or accelerators. Possible
20 reducing components include, but are not limited to, bisulfites, metabisulfites, ascorbic acid, erythorbic acid, and sodium formaldehyde sulfoxylate. Possible accelerators include, but are not limited to, any composition containing a transition metal having two oxidation states such as, for example, ferrous sulfate and ferrous ammonium sulfate. Alternatively, known thermal and radiation initiation techniques can be employed to generate the free radicals. An example is polymerization initiated using azo compounds
25 such as azobisisobutyronitrile (available from Aldrich Chemical Co.). In another embodiment, any polymerization and corresponding initiation or catalytic methods known by those skilled in the art may be used in the present invention. For example, when polymerization is performed by methods such as addition or condensation polymerization, the polymerization can be initiated or catalyzed by methods such as
30 cationic, anionic, or coordination methods.

When water is used to form the above-mentioned reaction mixture, the water can be purified water such as distilled or deionized water. However, the continuous phase of the emulsion can also comprise polar organic liquids or aqueous solutions of polar organic liquids, such as those listed below with regards to suitable hydrate inhibitors.

5 As previously noted, the reaction mixture can optionally include a buffer. The buffer can comprise any known buffer that is compatible with the initiation system. Examples of buffers suitable for use in the present invention include, but are not limited to, carbonate, phosphate, and/or borate buffers.

As previously noted, the reaction mixture can optionally include at least one
10 hydrate inhibitor. The hydrate inhibitor can be a thermodynamic hydrate inhibitor such as, for example, an alcohol and/or a polyol. In one embodiment, the hydrate inhibitor can comprise one or more polyhydric alcohols and/or one or more ethers of polyhydric alcohols. Suitable polyhydric alcohols include, but are not limited to, monoethylene glycol, diethylene glycol, triethylene glycol, monopropylene glycol, and/or dipropylene
15 glycol. Suitable ethers of polyhydric alcohols include, but are not limited to, ethylene glycol monomethyl ether, diethylene glycol monomethyl ether, propylene glycol monomethyl ether, and dipropylene glycol monomethyl ether.

Generally, the hydrate inhibitor can be any composition that when mixed with distilled water at a 1:1 weight ratio produces a hydrate inhibited liquid mixture having a
20 gas hydrate formation temperature at 2,000 psia that is lower than the gas hydrate formation temperature of distilled water at 2,000 psia by an amount in the range of from about 10 to about 150°F, in the range of from about 20 to about 80°F, or in the range of from 30 to 60°F. For example, monoethylene glycol qualifies as a hydrate inhibitor because the gas hydrate formation temperature of distilled water at 2,000 psia is about
25 70°F, while the gas hydrate formation temperature of a 1:1 mixture of distilled water and monoethylene glycol at 2,000 psia is about 28°F. Thus, monoethylene glycol lowers the gas hydrate formation temperature of distilled water at 2,000 psia by about 42°F when added to the distilled water at a 1:1 weight ratio. It should be noted that the gas hydrate formation temperature of a particular liquid may vary depending on the compositional
30 make-up of the natural gas used to determine the gas hydrate formation temperature. Therefore, when gas hydrate formation temperature is used herein to define what

constitutes a "hydrate inhibitor," such gas hydrate temperature is presumed to be determined using a natural gas composition containing 92 mole percent methane, 5 mole percent ethane, and 3 mole percent propane.

As previously discussed, the reaction mixture optionally can include at least one
5 stabilizer, also known as a suspending agent. The suspending agent can be of the form of one or more finely divided inorganic solids and/or water-soluble polymers; such suspending agents also can be called a protective colloid. Examples of finely divided inorganic solids include, but are not limited to, talc or fumed silica (both available from Aldrich Chemical Co.). An example of a protective colloid is poly(vinyl alcohol), 87-
10 89% hydrolyzed (available from Aldrich Chemical Co.).

In forming the reaction mixture, the at least two monomers, water, the at least one surfactant, and optionally the hydrate inhibitor, can be combined under a substantially oxygen-free atmosphere that is maintained at less than about 1,000 ppmw oxygen or less than 100 ppmw oxygen. The substantially oxygen-free atmosphere can be maintained by
15 continuously purging the reaction vessel with an inert gas such as nitrogen and/or argon. The temperature of the system can be kept at a level in the range of from the freezing point of the continuous phase up to about 60°C, in the range of from about 0 to about 45°C, or in the range of from 0 to 30°C. The system pressure can be maintained in the range of from about 5 to about 100 psia, in the range of from about 10 to about 25 psia,
20 or about atmospheric pressure. However, higher pressures up to about 300 psia can be useful to polymerize certain monomers, such as diolefins.

Next, a buffer can be added, if required, followed by addition of the initiation system, either all at once or over time. The polymerization reaction is carried out for a sufficient amount of time to achieve at least about 90 percent conversion by weight of
25 the monomers. Typically, this time period is in the range of from between about 1 to about 10 hours, or in the range of from 3 to 5 hours. During polymerization, the reaction mixture can be continuously agitated.

The following table sets forth approximate broad and narrow ranges for the amounts of the ingredients present in the reaction mixture.

Ingredient	Broad Range	Narrow Range
Total Monomer (wt. % of reaction mixture)	10 - 60%	30 - 50%
Water (wt. % of reaction mixture)	20 - 80%	50 - 70%
Surfactant (wt. % of reaction mixture)	0.1 - 10%	0.25 - 6%
Initiation system		
Monomer:Initiator (molar ratio)	$1 \times 10^3:1 - 5 \times 10^6:1$	$5 \times 10^3:1 - 2 \times 10^6:1$
Monomer:Reducing Comp. (molar ratio)	$1 \times 10^3:1 - 5 \times 10^6:1$	$1 \times 10^4:1 - 2 \times 10^6:1$
Accelerator: Initiator (molar ratio)	0.001:1 - 10:1	0.005:1 - 1:1
Buffer	0 to amount necessary to reach pH of initiation (initiator dependent, typically between about 6.5-10)	
Optional hydrate inhibitor	If present, the hydrate inhibitor can have a hydrate inhibitor-to-water weight ratio from about 1:10 to about 10:1, about 1:5 to about 5:1, or 2:3 to 3:2.	

The emulsion polymerization reaction achieved in the above-described reaction mixture can yield a latex composition comprising a dispersed phase of solid particles and a liquid continuous phase. The latex can be a stable colloidal dispersion comprising a dispersed phase of high molecular weight polymer particles and a continuous phase comprising water. The colloidal particles can make up in the range of from about 10 to about 60 percent by weight of the latex, or in the range of from 40 to 50 percent by weight of the latex. The continuous phase can comprise water, the high HLB surfactant, the hydrate inhibitor (if present), and buffer as needed. Water can be present in the range of from about 20 to about 80 percent by weight of the latex, or in the range of from about 40 to about 60 percent by weight of the latex. The high HLB surfactant can make up in the range of from about 0.1 to about 10 percent by weight of the latex, or in the range of from 0.25 to 6 percent by weight of the latex. As noted in the table above, the buffer can be present in an amount necessary to reach the pH required for initiation of the

polymerization reaction and is initiator dependent. Typically, the pH required to initiate a reaction is in the range of from 6.5 to 10.

The suspension polymerization reaction achieved in the above-described reaction mixture can yield a suspended polymer composition comprising a dispersed phase of solid particles and a liquid continuous phase. The suspension can be a stable colloidal dispersion comprising a dispersed phase of high molecular weight polymer particles and a continuous phase comprising water. The colloidal particles can make up in the range of from about 10 to about 60 percent by weight of the suspension, or in the range of from 40 to 50 percent by weight of the suspension. The continuous phase can comprise water, the high HLB surfactant, the hydrate inhibitor (if present), and buffer as needed. Water can be present in the range of from about 20 to about 80 percent by weight of the suspension, or in the range of from about 40 to about 60 percent by weight of the suspension. The high HLB surfactant can make up in the range of from about 0.1 to about 10 percent by weight of the suspension, or in the range of from 0.25 to 6 percent by weight of the suspension. As noted in the table above, the buffer can be present in an amount necessary to reach the pH required for initiation of the polymerization reaction and is initiator dependent. Typically, the pH required to initiate a reaction is in the range of from 6.5 to 10.

When a hydrate inhibitor is employed in the reaction mixture, it can be present in the resulting latex or suspension in an amount that yields a hydrate inhibitor-to-water weight ratio in the range of from about 1:10 to about 10:1, in the range of from about 1:5 to about 5:1, or in the range of from 2:3 to 3:2. Alternatively, all or part of the hydrate inhibitor can be added to the latex or suspension after polymerization to provide the desired amount of hydrate inhibitor in the continuous phase of the latex or suspension.

As mentioned above, at least two monomers can be employed when preparing the drag reducing composition of the present invention, thus resulting in a drag reducer comprising a plurality of polymer particles comprising copolymers. In one embodiment, the resulting copolymers can comprise residues of the above-mentioned methacrylate monomer in an amount in the range of from about 10 to about 99 weight percent, in the range of from about 20 to about 97 weight percent, or in the range of from 40 to 95 weight percent, based on the total weight of the copolymer. Additionally, the

copolymers can comprise residues of the above-described comonomer in an amount in the range of from about 1 to about 90 weight percent, in the range of from about 3 to about 80 weight percent, or in the range of from 5 to 60 weight percent, based on the total weight of the copolymer.

5 In one embodiment, the copolymers of the resulting drag reducing composition can comprise at least partially saturated hydrocarbon backbones comprising a plurality of saturated carbon atoms. The hydrocarbon backbones of the copolymers can be at least 90 percent saturated, at least 95 percent saturated, at least 99 percent saturated, or substantially fully saturated. In one embodiment of the present invention, fewer than 50
10 percent, fewer than 45 percent, or fewer than 40 percent of the saturated carbon atoms in the hydrocarbon backbone of the copolymer have more than one pendant substituent.

Though not wishing to be bound by theory, the inventors have unexpectedly discovered that employing a methacrylate monomer with a comonomer comprising a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon
15 appears to produce a copolymer having improved rates of dissolution in hydrocarbon-containing fluids at lower temperatures. Similarly, copolymers having fewer saturated backbone carbon atoms comprising more than one pendant substituent seem to exhibit improved rates of dissolution in hydrocarbon-containing fluids at lower temperatures. Thus, in one embodiment of the present invention, the polymer particles of the drag
20 reducing composition can have a hydrocarbon dissolution rate constant of at least 0.01 min^{-1} , at least 0.05 min^{-1} , or at least 0.1 min^{-1} in a mixture of kerosene and isopropyl alcohol ("IPA") in a volume ratio of 95:5 kerosene:IPA. Hydrocarbon dissolution rate constants are determined in accordance with the procedure outlined below in the "Dissolution Rate Testing" portion of the Examples section.

25 In one embodiment of the present invention, the emulsion drag reducing polymer of the dispersed phase of the latex can have a weight average molecular weight (M_w) of at least about $1 \times 10^6 \text{ g/mol}$, at least about $2 \times 10^6 \text{ g/mol}$, or at least $5 \times 10^6 \text{ g/mol}$. The colloidal particles of the emulsion drag reducing polymer can have a mean particle size of less than about 10 micrometers, less than about 1,000 nm (1 micrometer), in the range
30 of from about 10 to about 500 nm, or in the range of from 50 to 250 nm. At least about 95 percent by weight of the colloidal particles can be larger than about 10 nm and

smaller than about 500 nm. At least about 95 percent by weight of the particles can be larger than about 25 nm and smaller than about 250 nm. The continuous phase can have a pH in the range of from about 4 to about 10, or in the range of from about 6 to about 8, and contains few if any multi-valent cations.

5 In one embodiment of the present invention, the suspended drag reducing polymer of the dispersed phase of the suspension drag reducing polymer can have a weight average molecular weight (M_w) of at least about 1×10^6 g/mol, at least about 2×10^6 g/mol, or at least 5×10^6 g/mol. The colloidal particles of the suspended drag reducing polymer can have a mean particle size of less than about 1000 micrometers, less
10 than about 500 micrometers, in the range of from about 1 to about 500 micrometers, or in the range of from 10 to 400 micrometers. At least about 95 percent by weight of the colloidal particles can be larger than about 1 micrometer and smaller than about 500 micrometers. At least about 95 percent by weight of the particles can be larger than about 10 micrometers and smaller than about 400 micrometers. The continuous phase
15 can have a pH in the range of from about 4 to about 10, or in the range of from about 6 to about 8, and contains few if any multi-valent cations.

In one embodiment of the present invention, the above-described drag reducing composition can be added to a hydrocarbon-containing fluid. The resulting treated hydrocarbon-containing fluid can then be transported through a pipeline. The
20 hydrocarbon-containing fluid can comprise a liquid phase hydrocarbon, a non-liquid phase hydrocarbon, and/or a non-hydrocarbon fluid. In one embodiment, the hydrocarbon-containing fluid can comprise at least about 50 weight percent of a liquid phase hydrocarbon. Additionally, the hydrocarbon-containing fluid can comprise crude oil. Furthermore, the hydrocarbon-containing fluid of the present invention can have an
25 average temperature of 25°C or less, 22°C or less, or 20°C or less.

The resulting treated hydrocarbon-containing fluid can comprise a cumulative amount of the drag reducing copolymers sufficient to achieve a reduction in drag associated with the turbulent flow of the hydrocarbon-containing fluid through the pipeline. In one embodiment, the treated hydrocarbon-containing fluid can have a
30 cumulative concentration of drag reducing copolymers in the range of from about 0.1 to about 500 ppmw, in the range of from about 0.5 to about 200 ppmw, in the range of from

about 1 to about 100 ppmw, or in the range of from 2 to 50 ppmw. Typically, at least 50 weight percent, at least 75 weight percent, or at least 95 weight percent of the drag reducing copolymer particles can be dissolved by the hydrocarbon-containing fluid. Additionally, the drag reducers employed in the present invention can provide significant
5 percent drag reduction during transport of the hydrocarbon-containing fluid through a pipeline. For example, the drag reducers can provide at least about 5 percent drag reduction or at least 10 percent drag reduction.

The following examples are intended to be illustrative of the present invention in order to teach one of ordinary skill in the art to make and use the invention and are not
10 intended to limit the scope of the invention in any way.

EXAMPLES

Drag Reducer Preparation Procedure

15 Each of the polymer samples employed in the following examples were prepared according to the following method. Polymerization was performed in a 300 mL jacketed reaction kettle fitted with a condenser, mechanical stirrer, thermocouple, septum ports, and nitrogen inlets / outlets. The kettle was charged with 0.230g of sodium hydrogen phosphate, 0.230g potassium dihydrogen phosphate, 4.47g sodium dodecyl sulfate and
20 purged with nitrogen for at least one hour. The kettle was then charged with 120 mL of deoxygenated Type I water. 2-Ethylhexyl methacrylate and a comonomer of interest were separately stirred with inhibitor remover (available from Aldrich Chemical Co., designed to remove the polymerization inhibitor 4-methoxy phenol) under a nitrogen atmosphere for at least one hour. After the desired time, the inhibitor remover was
25 separated from the respective monomers by filtration under a nitrogen atmosphere. The desired amounts of the respective monomers were added to the reaction kettle using syringe techniques. Stirring was initiated at about 300 rpm and the kettle jacket was set to the desired polymerization temperature by using a circulating bath.

When ready to initiate polymerization, to the polymerization mixture in the kettle
30 was added 1.0mL of FeSO₄ solution (0.0021 g/mL). Following this, 1.0 mL of ammonium persulfate solution (0.0026 g/mL) and 1.0 mL of sodium formaldehyde

sulfoxylate (SFS) solution (0.0018 g/mL) were each added over four hours. At the end of the four-hour period, to the polymerization mixture 1.0 mL of tert-butyl hydroperoxide solution (0.0075 g/mL) and 1.0 mL SFS solution (0.0018 g/mL) were each added over two hours.

5 The resulting drag reducer was a latex containing a copolymer of 2-ethylhexyl methacrylate and a comonomer. The samples had solids contents of about 45 percent by mass and a nominal polymer content of approximately 40 percent. The densities of the samples were approximately 1.0005 g/mL. The carrier fluid for each sample was 100% water.

10 Dissolution Rate Testing

The dissolution rate constants of several copolymers prepared via the above method were determined according to the following procedure. The rate at which a polymer dissolves can be determined by a vortex inhibition test in a kerosene-containing medium at various temperatures. At a constant stirring speed, the depth of the vortex is
15 proportional to the amount of dissolved polymer in the kerosene-containing medium. The dissolution rate is a first order function:

$$d/dt (\text{Conc}_{\text{undissolved}}) = -k \times \text{Conc}_{\text{undissolved}}$$

20 wherein k is the dissolution rate constant. The time, T, for a certain fraction of the polymer to be dissolved is a function of k as follows:

$$T_{\% \text{ dissolved}} = [\ln 100/(100 - \% \text{ dissolved})]/k$$

25 FIG. 1 schematically illustrates the dissolution rate test apparatus used to determine the dissolution rate constant. The dissolution rate test apparatus included a rotating stirrer that was placed in a jacketed graduated 250 mL cylinder having an internal diameter of 48 mm. The upper end of the rotating stirrer was connected to a variable-speed motor (not shown). The specific configuration of the rotating stirrer is
30 illustrated in detail in FIGS. 2-4. The rotating stirrer employed in the dissolution rate tests was a Black & Decker paint stirrer made from a casting of oil resistant plastic. The

stirrer head was formed of a 45 mm diameter disk made up of a central disk and an outer ring. The central disk was 20 mm in diameter and 1.5 mm thick and was centered on a hub that was 12 mm in diameter and 12 mm thick. The hub was drilled in the center for attachment of the stirring head to a 4 mm diameter shaft. The shaft was threaded for 27
5 mm so that two small nuts held the stirring head to the shaft. The outer ring was 45 mm in diameter, 9 mm wide, and 1.5 mm thick. The outer ring was attached to the inner disk by 13 evenly spaced arcs 13 mm long and 1 mm thick. The outer ring resided 6 mm below the level of the inner disk. The arcs that attached the outer ring to the inner disk acted as paddles to stir the fluid in the test cylinder. The shaft that attached the stirring
10 head to the stirring motor (not shown) was 300 mm long. It should be noted that dissolution rate test results may vary somewhat if different stirrer configurations are used.

To conduct the dissolution rate test, the stirrer was positioned inside the cylinder and adjusted so that the bottom of stirrer head was about 5 millimeters from the bottom
15 of the cylinder. The cylinder jacket was then filled with water recirculated from a recirculating water bath with controlled heating and cooling capability. The desired temperature was selected and the bath was allowed to reach that temperature. With the stirrer in place, the jacketed graduated cylinder was filled to the 200 mL line with a mixture comprising 95 volume percent kerosene and 5 volume percent isopropyl alcohol
20 ("IPA"). The circulation of cooling fluid through the graduated cylinder jacket was initiated. The kerosene mixture inside the graduated cylinder was stirred for sufficient time to allow the temperature to equilibrate at the set temperature, usually 10-15 minutes. The kerosene mixture temperature was checked with a thermometer to insure that the kerosene mixture was at the desired test temperature. The speed of the motor was
25 adjusted to stir rapidly enough to form a vortex in the kerosene mixture that reached to the 125 mL graduation mark in the cylinder.

A 0.5 mL aliquot of the polymer latex was added to the stirring kerosene mixture with the vortex established at the 125 mL mark. The aliquot of the latex was added to the kerosene at the desired temperature, as indicated in Tables 1 and 2 below. A timer
30 was used to monitor and record the time required for the vortex to recede to each of the 5 mL increments on the cylinder: 130, 135, 140, and so on. However, the determination

was stopped when the time exceeded 30 minutes. The position of the vortex at the end of 30 minutes is designated as V_f .

The dissolution rate constant, k , was calculated from the slope of a plot of the log of the relative vortex against time. The relative vortex is the decimal fraction of the
5 relationship

$$[V_f - V_t] / [V_f - V_i]$$

where V_f is the final reading at the maximum vortex suppression within the 30 minute timeframe of the experiment, V_i is the initial vortex reading prior to addition of drag reducing polymer (which is routinely set at the 125 mL mark), and V_t is the vortex
10 reading at the specified marks 130, 135, 140, and so on up to the reading at the maximum vortex suppression. A linear regression analysis was performed on the plot of the log of the relative vortex against time. The resulting slope of the data gave the dissolution rate constant, k , for a given temperature and concentration of active polymer when multiplied by -2.303.

15

Example 1 – Comonomers with One or Less Pendant Substituents per Vinyl

Carbon

Copolymer latex drag reducers prepared from 2-ethylhexyl methacrylate monomer and a variety of comonomers comprising a polymerizable vinyl group having
20 no more than one pendant substituent per vinyl carbon were prepared according to the above-described procedure. Thereafter, the dissolution rate of each sample was tested and the dissolution rate constant was calculated as described above. The results from these procedures are listed in Table 1.

Table 1

**Dissolution Rates of Copolymers Comprising Comonomers Having One or Less
Pendant Substituents per Vinyl Carbon**

Copolymer	Monomer Ratio (molar)	Dissolution Rate Constant In 95:5 Kerosene:IPA		
		40°C	30°C	20°C
2-ethylhexyl methacrylate / ethylhexyl acrylate	100:0	N/A	0.361	0.273
	84:16	N/A	0.714	0.417
	84:16	N/A	0.796	0.563
	84:16	N/A	0.552	0.344
	100:0	N/A	No Dissolution	No Dissolution
	95:5	N/A	0.118	0.017
	89:11	N/A	0.206	0.089
	79:21	N/A	0.533	0.359
	100:0	0.144	0.013	No Dissolution
	95:5	0.316	0.080	No Dissolution
	90:10	0.266	0.197	0.074
	84:16	0.472	0.719	0.248
	79:21	0.759	0.486	0.316
2-ethylhexyl methacrylate / 3,5,5- trimethylhexyl acrylate	100:0	0.403	0.110	No Dissolution
	98.5:1.5	0.353	0.128	No Dissolution
	95:5	0.355	0.230	0.065
	100:0	0.373	0.044	No Dissolution
	98.5:1.5	0.246	0.064	No Dissolution
	95:5	0.500	0.102	No Dissolution
2-ethylhexyl methacrylate / n-hexyl acrylate	100:0	N/A	No Dissolution	No Dissolution
	94:6	N/A	0.093	No Dissolution
	88:12	N/A	0.368	0.135
	77:23	N/A	0.508	0.472
	100:0	N/A	N/A	No Dissolution
	94:6	N/A	N/A	No Dissolution
	88:12	N/A	N/A	0.111
	76:24	N/A	N/A	0.540
	100:0	No Dissolution	No Dissolution	No Dissolution
	94:6	0.253	0.099	0.160
	88:12	0.390	0.362	0.166
	76:24	0.177	0.754	0.383
	100:0	0.213	No Dissolution	No Dissolution
	99.4:0.6	0.277	0.070	No Dissolution
	98:2	No Dissolution	No Dissolution	No Dissolution
	94:6	0.396	0.176	0.120
	94:6	0.507	0.163	0.058

Copolymer	Monomer Ratio (molar)	Dissolution Rate Constant In 95:5 Kerosene:IPA		
		40°C	30°C	20°C
2-ethylhexyl methacrylate / benzyl acrylate	100:0	0.482	0.145	No Dissolution
	99.3:0.7	0.350	0.112	No Dissolution
	97.8:2.2	0.582	0.309	No Dissolution
2-ethylhexyl methacrylate / t-butyl acrylate	100:0	N/A	0.073	No Dissolution
	93:7	N/A	0.141	No Dissolution
	86:14	N/A	0.410	0.227
	73:27	N/A	0.504	0.292
	100:0	N/A	No Dissolution	No Dissolution
	93:7	N/A	0.213	0.075
	86:14	N/A	0.256	0.407
	73:27	N/A	0.367	0.174
	100:0	N/A	No Dissolution	No Dissolution
	93:7	N/A	0.250	0.092
	86:14	N/A	0.277	0.114
	73:27	N/A	0.339	0.446
	100:0	N/A	No Dissolution	No Dissolution
	93:7	N/A	0.250	0.092
	86:14	N/A	0.277	0.114
	73:27	N/A	0.339	0.446
2-ethylhexyl methacrylate / 2-phenylethyl acrylate	100:0	0.411	0.120	No Dissolution
	99.4:0.6	0.327	0.126	No Dissolution
	98:2	0.203	0.088	0.078
	94:6	0.269	0.143	0.066
	94:6	0.238	0.286	0.036
2-ethylhexyl methacrylate / n-butyl acrylate	100:0	N/A	0.155	0.016
	93:7	N/A	0.158	0.089
	86:14	N/A	0.638	0.163
	72:28	N/A	0.997	0.529
	100:0	N/A	N/A	N/A
	80:20	N/A	0.676	0.554
	60:40	N/A	0.506	0.513
	40:60	N/A	0.534	0.508
	100:0	N/A	0.175	0.137
	80:20	N/A	0.461	0.554
	40:60	N/A	0.462	0.616
	20:80	N/A	0.420	0.454
	72:28	N/A	0.586	0.270
	72:28	N/A	0.601	0.274
	72:28	N/A	0.625	0.464
	73:27	N/A	0.500	0.328
	72:28	N/A	0.720	0.359
	72:28	N/A	0.578	0.491
	72:28	N/A	0.546	0.340
	73:27	N/A	0.439	0.331

Copolymer	Monomer Ratio (molar)	Dissolution Rate Constant In 95:5 Kerosene:IPA		
		40°C	30°C	20°C
	100:0	N/A	N/A	No Dissolution
	93:7	N/A	N/A	0.016
	85:15	N/A	N/A	0.182
	73:27	N/A	N/A	0.394
	100:0	N/A	N/A	No Dissolution
	93:7	N/A	N/A	0.019
	85:15	N/A	N/A	0.202
	72:28	N/A	N/A	0.539
	100:0	N/A	N/A	No Dissolution
	93:7	N/A	N/A	No Dissolution
	86:14	N/A	N/A	0.210
	72:28	N/A	N/A	0.453
	100:0	0.182	No Dissolution	No Dissolution
	98:2	0.585	0.220	No Dissolution
	93:7	0.546	0.183	0.073
2-ethylhexyl methacrylate / isodecyl acrylate	100:0	0.287	0.085	No Dissolution
	99.5:0.5	0.421	0.094	No Dissolution
	98.6:1.4	0.526	0.068	No Dissolution
	96:4	0.259	0.121	No Dissolution
	96:4	0.232	0.135	No Dissolution
2-ethylhexyl methacrylate / isobutyl acrylate	100:0	N/A	0.019	No Dissolution
	93:7	N/A	0.143	0.065
	85:15	N/A	0.190	0.106
	72:28	N/A	0.414	0.335
	100:0	0.077	No Dissolution	No Dissolution
	93:7	0.162	0.082	0.017
	85:15	0.567	0.408	0.251
	72:28	1.000	0.838	0.614
2-ethylhexyl methacrylate / isobornyl acrylate	100:0	0.070	No Dissolution	No Dissolution
	99.5:0.5	0.082	No Dissolution	No Dissolution
	98.4:1.6	0.193	0.016	No Dissolution
	95:5	0.315	0.205	0.082
	95:5	0.175	0.022	No Dissolution
2-ethylhexyl methacrylate / sec-butyl acrylate	100:0	0.024	0.023	No Dissolution
	99.2:0.8	0.016	No Dissolution	No Dissolution
	98:2	0.028	No Dissolution	No Dissolution
	93:7	0.256	0.099	0.094
	93:7	0.224	0.106	No Dissolution
2-ethylhexyl methacrylate / ethyl acrylate	100:0	0.032	No Dissolution	No Dissolution
	99:1	0.278	0.085	No Dissolution
	98:2	0.072	No Dissolution	No Dissolution
	90:10	0.436	0.331	0.086

Copolymer	Monomer Ratio (molar)	Dissolution Rate Constant In 95:5 Kerosene:IPA		
		40°C	30°C	20°C
	90:10	0.379	0.155	0.016
2-ethylhexyl methacrylate / styrene	100:0	0.101	No Dissolution	No Dissolution
	99:1	0.281	0.077	No Dissolution
	97:3	0.190	No Dissolution	No Dissolution
2-ethylhexyl methacrylate / bis(2-ethylhexyl) maleate	100:0	0.194	0.013	No Dissolution
	99.7:0.3	0.081	No Dissolution	No Dissolution
	99:1	0.341	0.016	No Dissolution
	97:3	No Dissolution	No Dissolution	No Dissolution
	97:3	No Dissolution	No Dissolution	No Dissolution
2-ethylhexyl methacrylate / bis(2-ethylhexyl) fumarate	100:0	No Dissolution	No Dissolution	No Dissolution
	99.7:0.3	0.227	0.010	No Dissolution
	99:1	0.155	0.013	No Dissolution
	97:3	0.189	No Dissolution	No Dissolution
	97:3	0.075	No Dissolution	No Dissolution

In addition to the above, one sample drag reducer comprising an 80:20 molar ratio blend of 2-ethylhexyl methacrylate to n-butyl acrylate was tested at 10°C. This sample yielded a dissolution rate constant of 0.191.

5

Example 2 – Comonomers with More Than One Pendant Substituent on a Vinyl Carbon

Copolymer latex drag reducers prepared from 2-ethylhexyl methacrylate monomer and a variety of comonomers comprising a polymerizable vinyl group with at least one vinyl carbon having more than one pendant substituent were prepared according to the above-described procedure. Thereafter, the dissolution rate of each sample was tested and the dissolution rate constant was calculated as described above. The results from these procedures are listed in Table 2.

10

Table 2

**Dissolution Rates of Copolymers Comprising Comonomers Having More Than One
Pendant Substituent on a Vinyl Carbon**

Copolymer	Monomer Ratio (molar)	Dissolution Rate Constant In 95:5 Kerosene:IPA		
		40°C	30°C	20°C
2-ethylhexyl methacrylate / lauryl methacrylate	100:0	No Dissolution	No Dissolution	No Dissolution
	96:4	No Dissolution	No Dissolution	No Dissolution
2-ethylhexyl methacrylate / n-octyl methacrylate	100:0	0.051	No Dissolution	No Dissolution
	98.5:1.5	0.245	No Dissolution	No Dissolution
2-ethylhexyl methacrylate / benzyl methacrylate	100:0	No Dissolution	No Dissolution	No Dissolution
	94:6	No Dissolution	No Dissolution	No Dissolution
2-ethylhexyl methacrylate / t-butyl methacrylate	100:0	0.028	No Dissolution	No Dissolution
	93:7	No Dissolution	No Dissolution	No Dissolution
2-ethylhexyl methacrylate / 2-phenylethyl methacrylate	100:0	0.216	No Dissolution	No Dissolution
	96:4	0.035	0.017	No Dissolution
2-ethylhexyl methacrylate / n-butyl methacrylate	100:0	N/A	0.214	0.155
	80:20	N/A	0.053	No Dissolution
	40:60	N/A	No Dissolution	No Dissolution
	100:0	0.110	No Dissolution	No Dissolution
	98:2	0.117	No Dissolution	No Dissolution
	93:7	0.251	No Dissolution	No Dissolution
2-ethylhexyl methacrylate / cyclohexyl methacrylate	100:0	0.329	0.055	0.014
	98:2	0.089	0.013	No Dissolution
	94:6	0.146	0.011	No Dissolution
2-ethylhexyl methacrylate / isodecyl methacrylate	100:0	No Dissolution	No Dissolution	No Dissolution
	98.7:1.3	No Dissolution	No Dissolution	No Dissolution
2-ethylhexyl methacrylate / isobutyl methacrylate	100:0	No Dissolution	No Dissolution	No Dissolution
	93.5:6.5	No Dissolution	No Dissolution	No Dissolution

Copolymer	Monomer Ratio (molar)	Dissolution Rate Constant In 95:5 Kerosene:IPA		
		40°C	30°C	20°C
2-ethylhexyl methacrylate / hexadecyl methacrylate	100:0	0.186	0.020	No Dissolution
	99.7:0.3	0.389	0.098	No Dissolution
	97:3	0.025	No Dissolution	No Dissolution
2-ethylhexyl methacrylate / isobornyl methacrylate	100:0	0.140	No Dissolution	No Dissolution
	99.5:0.5	0.393	0.072	No Dissolution
	95:5	0.334	0.112	No Dissolution
2-ethylhexyl methacrylate / tridecyl methacrylate	100:0	0.417	0.118	No Dissolution
	99.6:0.4	0.047	No Dissolution	No Dissolution
	99:1	No Dissolution	No Dissolution	No Dissolution
	96:4	No Dissolution	No Dissolution	No Dissolution
	96:4	No Dissolution	No Dissolution	No Dissolution
2-ethylhexyl methacrylate / dibutyl itaconate	100:0	0.022	No Dissolution	No Dissolution
	99.5:0.5	0.026	No Dissolution	No Dissolution
	98.7:1.3	0.456	0.085	No Dissolution
	96:4	0.101	0.012	No Dissolution
	95:5	0.354	0.120	0.085

As can be seen by comparing Tables 1 and 2, preparing a copolymer from a 2-ethylhexyl methacrylate monomer and a comonomer comprising a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon produces a drag reducer having better dissolution rate constants at lower temperatures than when a comonomer having more than one substituent on a vinyl carbon is employed. For example, only one of the copolymers containing a comonomer having more than one substituent on a vinyl carbon (i.e., dibutyl itaconate) prepared in Example 2 was able to be dissolved according to the above-described procedure at a temperature of 20°C.¹ By comparison, of the 15 copolymer samples tested containing a comonomer having no more than one pendant substituent per vinyl carbon, 10 were able to be dissolved at 20°C in at least one of the sample ratios prepared. Additionally, at least one sample prepared employing a comonomer having no more than one pendant substituent per vinyl carbon (i.e., n-butyl acrylate) was able to be dissolved at 10°C. Accordingly, comonomers

¹ Note that the entries for n-butyl methacrylate and cyclohexyl methacrylate comonomers do indicate some dissolution at 20°C; however, no comonomer was present in the samples exhibiting dissolution (i.e., the monomer-to-comonomer ratio was 100:0).

comprising a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon appear to produce drag reducing copolymers having improved dissolution rate constants at lower temperatures over comonomers having more than one pendant substituent on a vinyl carbon. This is even more evident by comparing polymers that
5 differ only with respect to the presence or absence of the second pendant group. As an example, the dissolution rate constant, k , for 80:20 EHMA/n-butyl acrylate is about 0.5 min^{-1} at 20°C and dissolves much more rapidly compared to the control EHMA homopolymer while the corresponding EHMA/n-butyl methacrylate polymer does not even dissolve at 20°C under the test conditions.

10

NUMERICAL RANGES

The present description uses numerical ranges to quantify certain parameters relating to the invention. It should be understood that when numerical ranges are provided, such ranges are to be construed as providing literal support for claim
15 limitations that only recite the lower value of the range as well as claims limitation that only recite the upper value of the range. For example, a disclosed numerical range of 10 to 100 provides literal support for a claim reciting "greater than 10" (with no upper bounds) and a claim reciting "less than 100" (with no lower bounds).

20 DEFINITIONS

As used herein, the terms "comprising," "comprises," and "comprise" are open-ended transition terms used to transition from a subject recited before the term to one or more elements recited after the term, where the element or elements listed after the transition term are not necessarily the only elements that make up the subject.

25 As used herein, the terms "including," "includes," and "include" have the same open-ended meaning as "comprising," "comprises," and "comprise."

As used herein, the terms "having," "has," and "have" have the same open-ended meaning as "comprising," "comprises," and "comprise."

30 As used herein, the terms "containing," "contains," and "contain" have the same open-ended meaning as "comprising," "comprises," and "comprise."

As used herein, the terms "a," "an," "the," and "said" mean one or more.

As used herein, the term “and/or,” when used in a list of two or more items, means that any one of the listed items can be employed by itself or any combination of two or more of the listed items can be employed. For example, if a composition is described as containing components A, B, and/or C, the composition can contain A
5 alone; B alone; C alone; A and B in combination; A and C in combination; B and C in combination; or A, B, and C in combination.

The preferred forms of the invention described above are to be used as illustration
10 only, and should not be used in a limiting sense to interpret the scope of the present invention. Obvious modifications to the exemplary embodiments, set forth above, could be readily made by those skilled in the art.

15

Claims

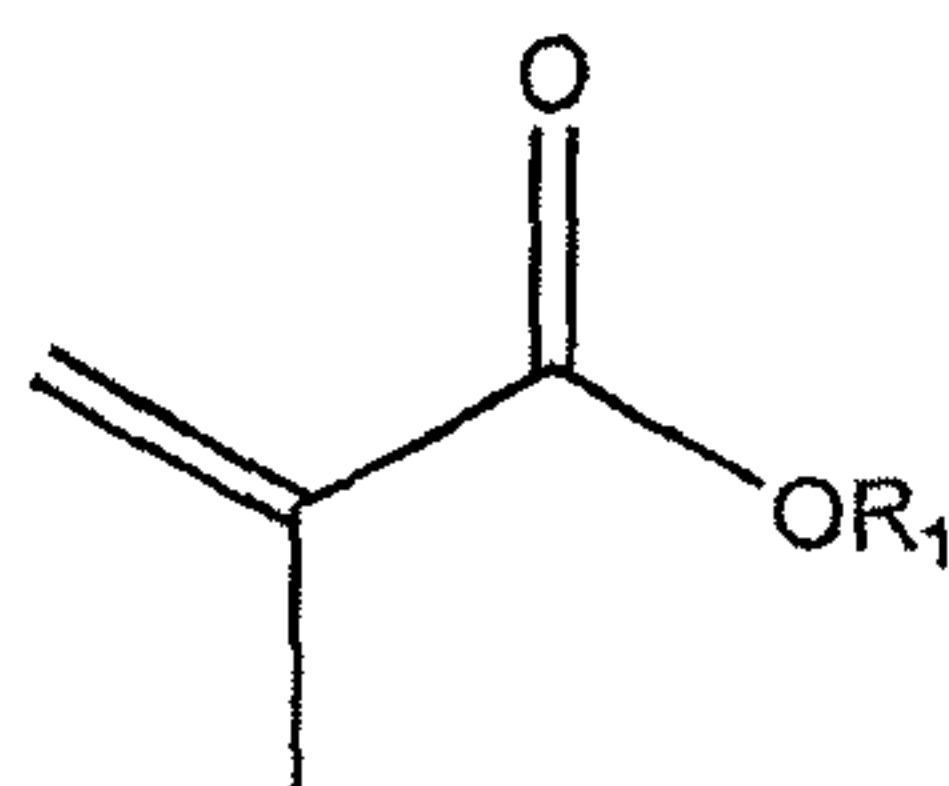
1. A mixture comprising a hydrocarbon-containing fluid and a drag reducing composition, the drag reducing composition comprising:

a continuous phase and a plurality of drag reducing polymer particles, wherein at least a portion of said polymer particles comprise copolymers containing from about 40 wt% to about 95 wt% of at least one methacrylate monomer and from about 5 wt% to about 60 wt% of at least one comonomer, wherein said comonomer comprises a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon, wherein said drag reducing composition reduces a pressure drop associated with a turbulent flow of said hydrocarbon-containing fluid through a conduit, and the hydrocarbon-containing fluid has an average temperature of 25°C or less.

2. The mixture of claim 1, wherein said copolymers comprise saturated hydrocarbon backbones comprising a plurality of carbon atoms, wherein fewer than 50 percent of said carbon atoms have more than one pendant substituent.

3. The mixture of claim 1 or 2, wherein said copolymers comprise saturated hydrocarbon backbones comprising a plurality of carbon atoms, wherein fewer than 45 percent of said carbon atoms have more than one pendant substituent.

4. The mixture of any one of claims 1 to 3, wherein said methacrylate monomer has the following structure:



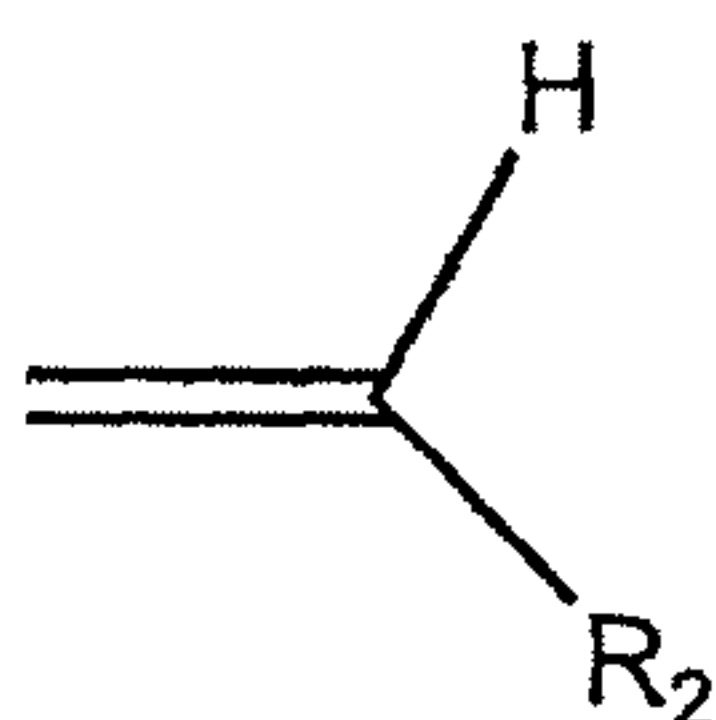
wherein R₁ is a C₁ to C₂₀ alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl.

5. The mixture of any one of claims 1 to 3, wherein said methacrylate monomer comprises C₄-C₂₀ alkyl, C₆-C₂₀ substituted or unsubstituted aryl, or aryl-substituted C₁-C₁₀ alkyl esters of methacrylic acid.

6. The mixture of any one of claims 1 to 3, wherein said methacrylate monomer comprises 2-ethylhexyl methacrylate.

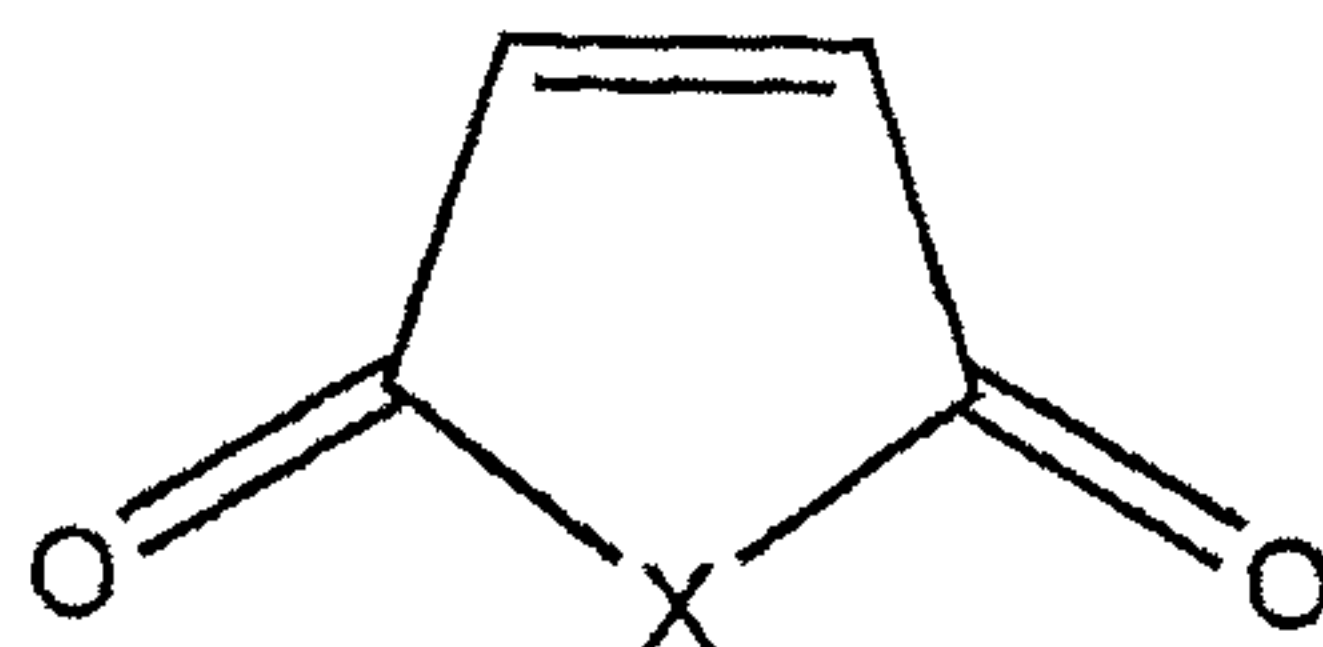
7. The mixture of any one of claims 1 to 6, wherein said comonomer comprises one or more of the following structures:

a)



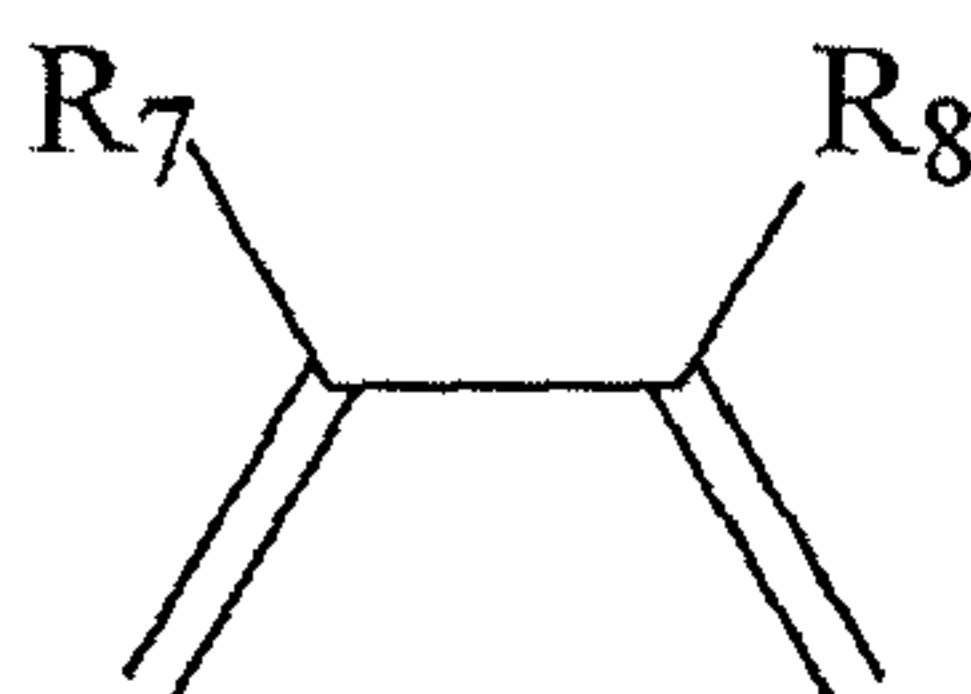
wherein R_2 is $-C(O)OR_3$, phenyl or aryl, heterocyclic, $-OR_4$, or $-O(O)CR_5$; wherein R_3 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; wherein R_4 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; wherein R_5 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; and/or

b)



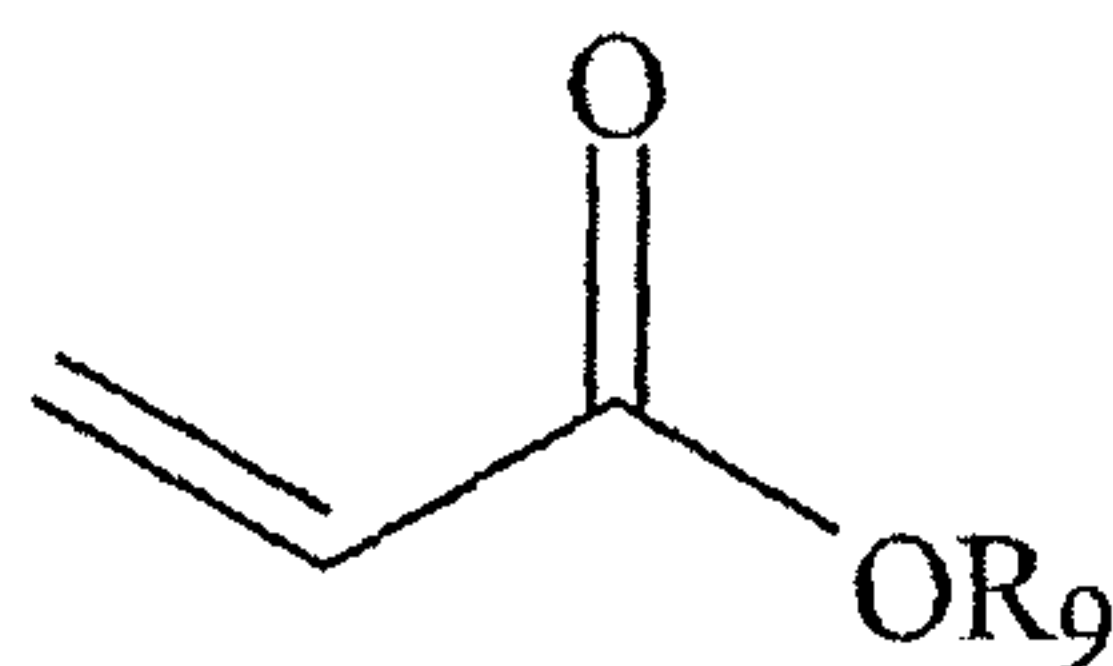
wherein X is O , or $N-R_6$, wherein R_6 is H , C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, aryl-substituted alkyl, or ring-opened alkyl diesters; and/or

c)



wherein R_7 and R_8 can independently be H , C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, aryl-substituted alkyl, or heterocyclic.

8. The mixture of any one of claims 1 to 6, wherein said comonomer is an acrylate having the following structure:



wherein R₉ is a straight or branched aliphatic alkyl group having 8 or fewer carbon atoms.

9. The mixture of claim 8, wherein R₉ is a straight or branched aliphatic alkyl group having 6 or fewer carbon atoms.

10. The mixture of any one of claims 1 to 6, wherein said comonomer is selected from the group consisting of n-butyl acrylate; 2-ethylhexyl acrylate; 3,5,5-trimethylhexyl acrylate; tert-butyl acrylate; 2-phenylethyl acrylate; iso-butyl acrylate; isobornyl acrylate; n-hexyl acrylate; sec-butyl acrylate; ethyl acrylate; and lauryl acrylate.

11. The mixture of any one of claims 1 to 6, wherein said comonomer comprises n-butyl acrylate.

12. The mixture of any one of claims 1 to 11, wherein said polymer particles have a hydrocarbon dissolution rate constant of at least about 0.01 min⁻¹ at 20°C in a mixture of kerosene and isopropyl alcohol ("IPA") in a volume ratio of 95:5 kerosene:IPA.

13. The mixture of any one of claims 1 to 12, wherein said continuous phase comprises water, one or more alcohols, and/or one or more polyols.

14. The mixture of any one of claims 1 to 13, wherein said drag reducing composition comprises said polymer particles in an amount in the range of from about 10 to about 60 weight percent based on the total weight of said drag reducing composition.

15. The mixture of any one of claims 1 to 14, wherein said polymer particles have a weight average molecular weight of at least 1x10⁶ g/mol.

16. The mixture of any one of claims 1 to 15, wherein said drag reducing composition comprises an emulsion drag reducing polymer wherein said polymer particles have an average particle size in the range of from about 10 to about 500 nm.

17. The mixture of any one of claims 1 to 16, wherein said drag reducing composition comprises a suspension drag reducing polymer wherein said polymer particles have an average particle size in the range of from about 1 to about 500 micrometers.

18. A method of forming a mixture, said method comprising:

(a) combining water, at least one surfactant, at least one methacrylate monomer, and at least one comonomer to thereby form a reaction mixture;

(b) subjecting said reaction mixture to polymerization to thereby form a composition comprising a continuous phase and a plurality of drag reducing copolymer particles comprising from about 40 wt% to about 95 wt% of said methacrylate monomer and from about 5 wt% to about 60 wt% of said comonomer,

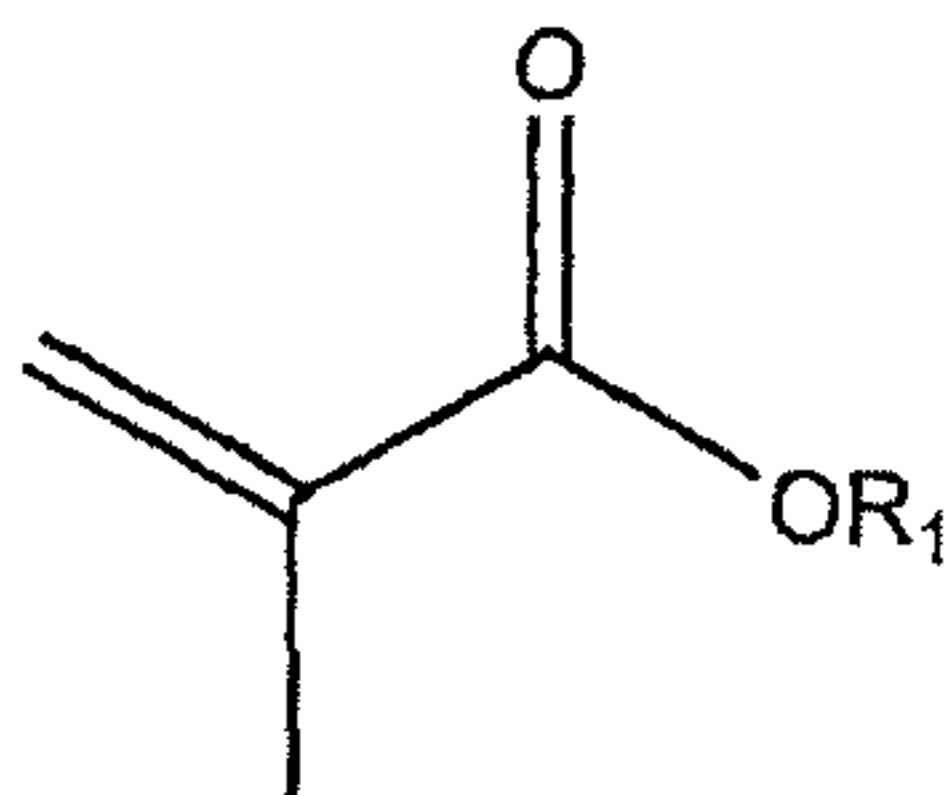
wherein said comonomer comprises a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon; and

(c) adding the drag reducing composition to a hydrocarbon-containing fluid having an average temperature of 20°C or less.

19. The method of claim 18, wherein said copolymers comprise saturated hydrocarbon backbones comprising a plurality of carbon atoms, wherein fewer than 50 percent of said carbon atoms have more than one pendant substituent.

20. The method of claim 18 or 19, wherein said copolymers comprise saturated hydrocarbon backbones comprising a plurality of carbon atoms, wherein fewer than 45 percent of said carbon atoms have more than one pendant substituent.

21. The method of any one of claims 18 to 20, wherein said methacrylate monomer has the following structure:

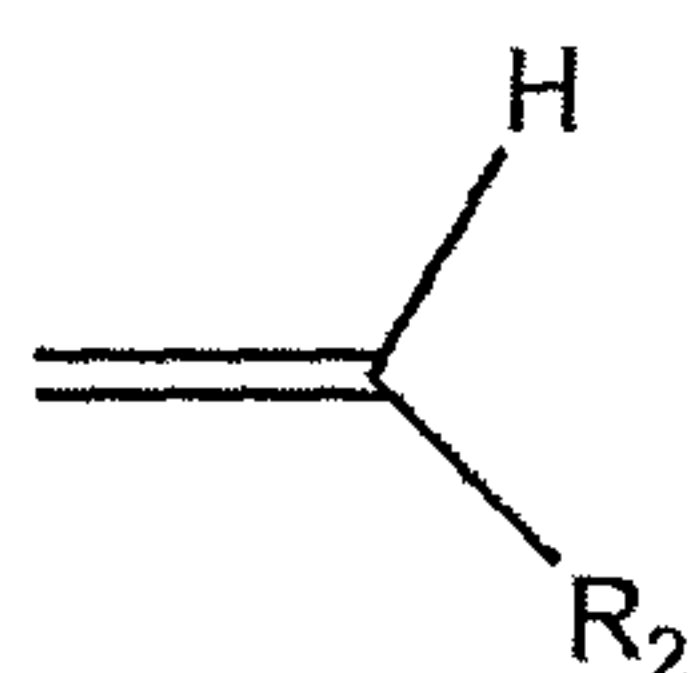


wherein R₁ is a C₁ to C₂₀ alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl.

22. The method of any one of claims 18 to 20, wherein said methacrylate monomer comprises 2-ethylhexyl methacrylate.

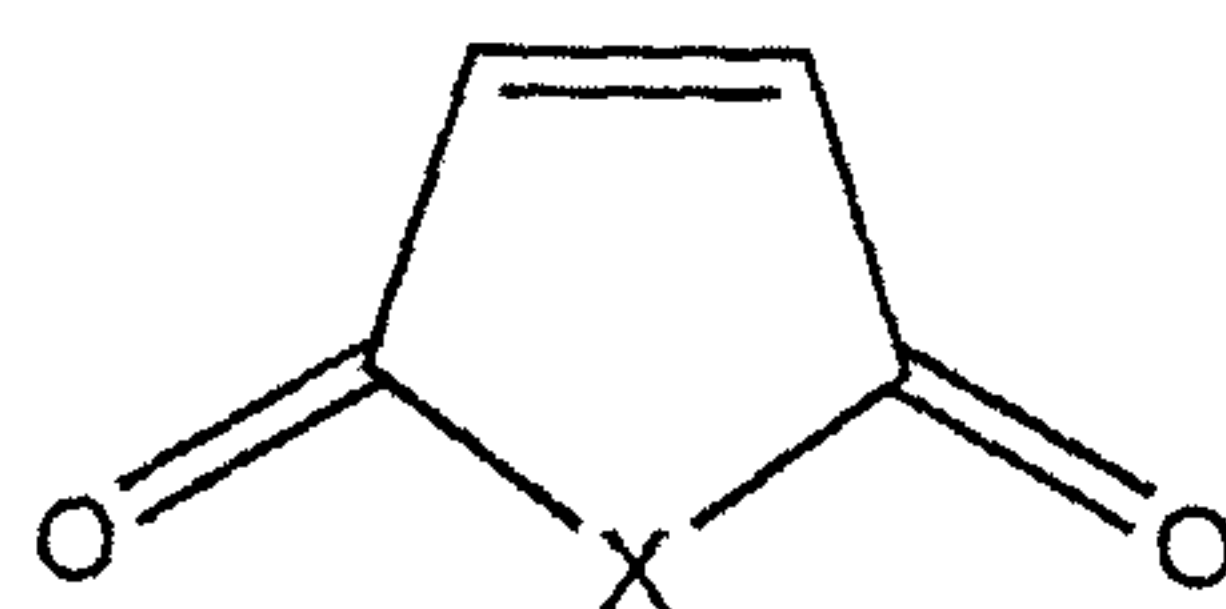
23. The method of any one of claims 18 to 22, wherein said comonomer comprises one or more of the following structures:

a)



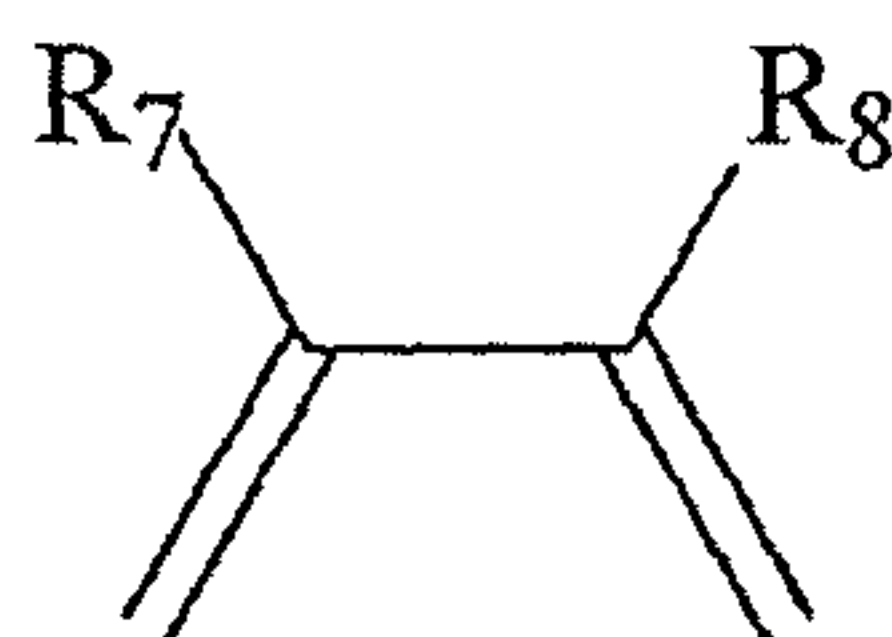
wherein R_2 is $-C(O)OR_3$, phenyl or aryl, heterocyclic, $-OR_4$, or $-O(O)CR_5$; wherein R_3 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; wherein R_4 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; wherein R_5 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; and/or

b)



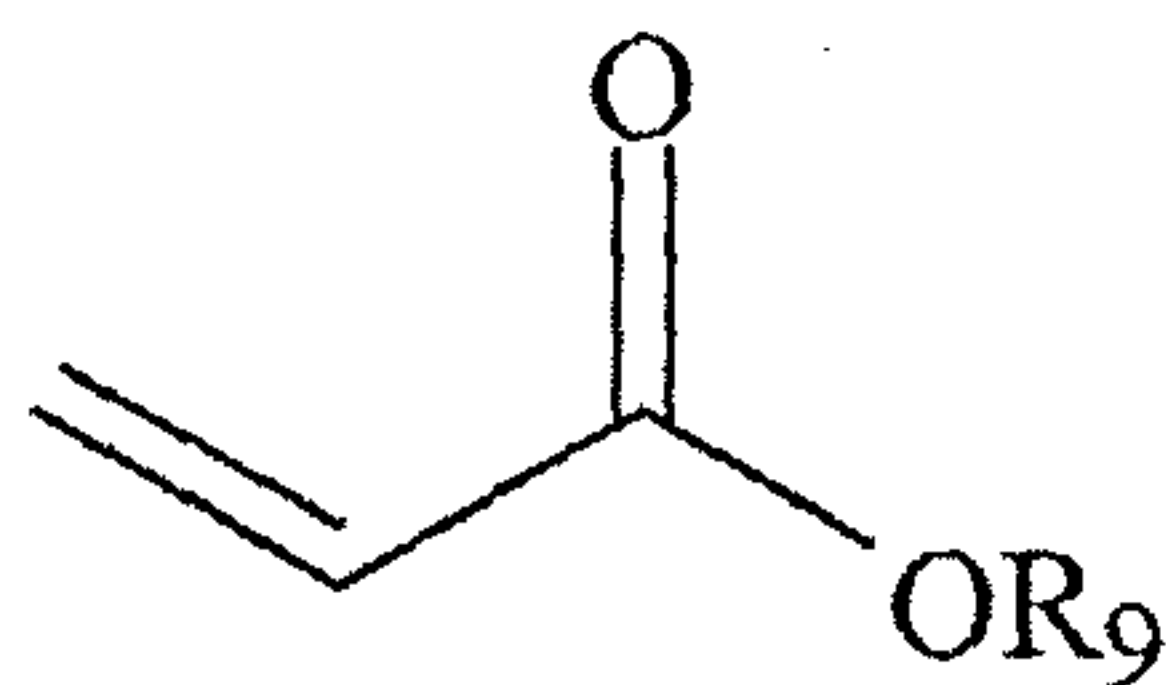
wherein X is O , or $N-R_6$, wherein R_6 is H , C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, aryl-substituted alkyl, or ring-opened alkyl diesters; and/or

c)



wherein R_7 and R_8 can independently be H , C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, aryl-substituted alkyl, or heterocyclic.

24. The method of any one of claims 18 to 22, wherein said comonomer is an acrylate having the following structure:



wherein R₉ is a straight or branched aliphatic alkyl group having 8 or fewer carbon atoms.

25. The method of any one of claims 18 to 22, wherein said comonomer comprises n-butyl acrylate.

26. The method of any one of claims 18 to 25, wherein said polymer particles have a hydrocarbon dissolution rate constant of at least about 0.01 min⁻¹ at 20°C in a mixture of kerosene and isopropyl alcohol ("IPA") in a volume ratio of 95:5 kerosene:IPA.

27. The method of any one of claims 18 to 26, wherein said drag reducing composition comprises said copolymer particles in an amount in the range of from about 10 to about 60 weight percent based on the total weight of said drag reducing composition.

28. The method of any one of claims 18 to 27, wherein said copolymer particles have a weight average molecular weight of at least 1x10⁶ g/mol.

29. The method of any one of claims 18 to 28, wherein said drag reducing copolymer comprises an emulsion drag reducing polymer wherein said copolymer particles have an average particle size in the range of from about 10 to about 500 nm.

30. The method of any one of claims 18 to 29, wherein said drag reducing copolymer comprises a suspension drag reducing polymer wherein said polymer particles have an average particle size in the range of from about 1 to about 500 micrometers.

31. A method for reducing the pressure drop associated with the turbulent flow of a hydrocarbon-containing fluid through a pipeline, said method comprising:

(a) introducing a drag reducing composition into said hydrocarbon-containing fluid, wherein said drag reducing composition comprises a continuous phase and a plurality of drag reducing polymer particles comprising copolymers containing from about 40 wt% to about 95 wt% of at least one methacrylate monomer and from about 5 wt% to

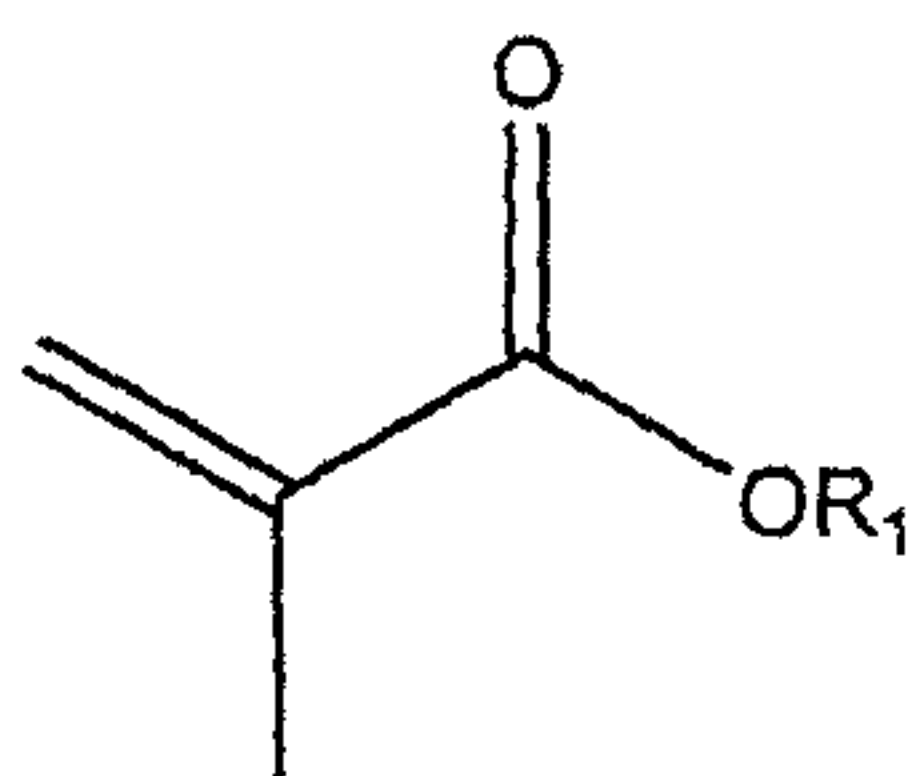
about 60 wt% of at least one comonomer, wherein said comonomer comprises a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon; and

(b) flowing the resulting treated hydrocarbon-containing fluid through said pipeline, wherein said hydrocarbon-containing fluid has an average temperature of 20°C or less.

32. The method of claim 31, wherein said copolymers comprise saturated hydrocarbon backbones comprising a plurality of carbon atoms, wherein fewer than 50 percent of said carbon atoms have more than one pendant substituent.

33. The method of claim 31 or 32, wherein said copolymers comprise saturated hydrocarbon backbones comprising a plurality of carbon atoms, wherein fewer than 45 percent of said carbon atoms have more than one pendant substituent.

34. The method of any one of claims 31 to 33 wherein said methacrylate monomer has the following structure:



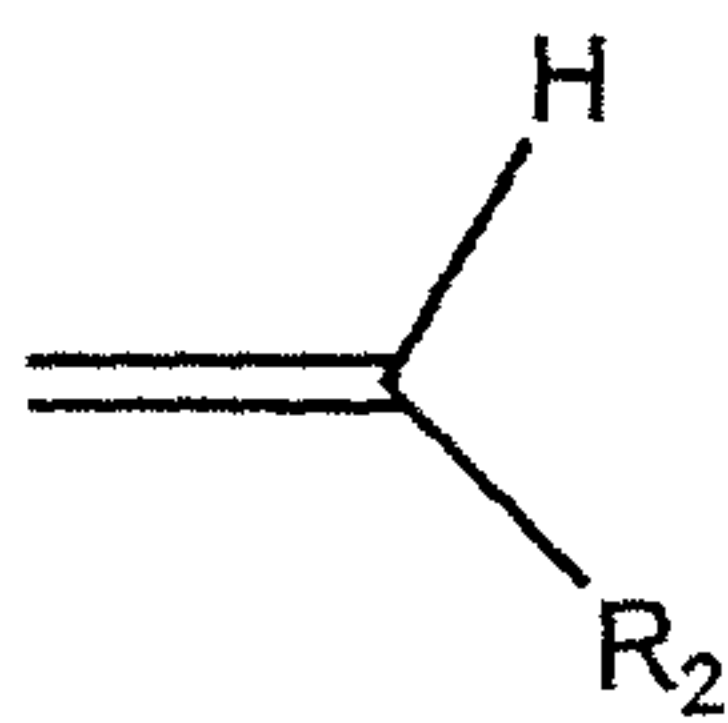
wherein R₁ is a C₁ to C₂₀ alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl.

35. The method of any one of claims 31 to 33, wherein said methacrylate monomer comprises C₄-C₂₀ alkyl, C₆-C₂₀ substituted or unsubstituted aryl, or aryl-substituted C₁-C₁₀ alkyl esters of methacrylic acid.

36. The method of any one of claims 31 to 33, wherein said methacrylate monomer comprises 2-ethylhexyl methacrylate.

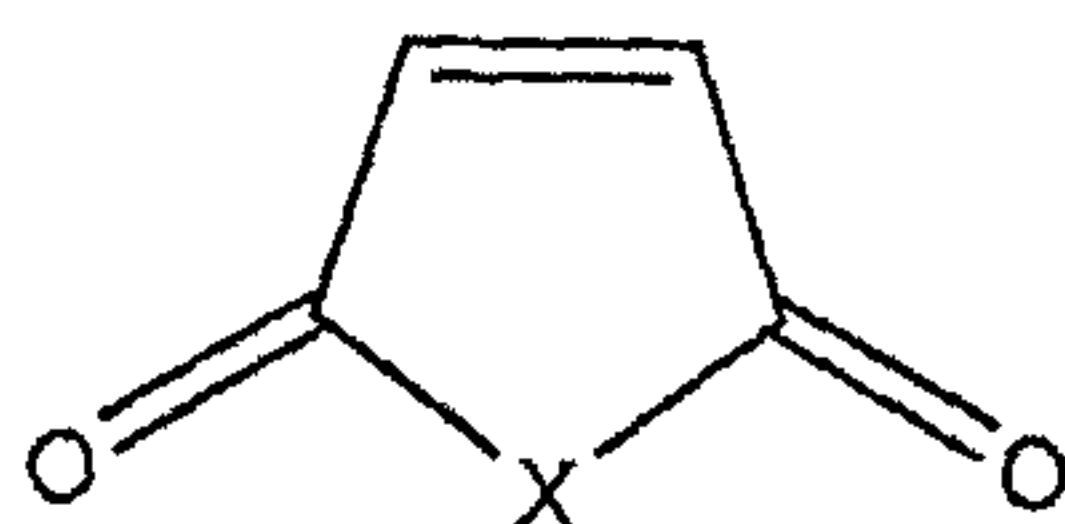
37. The method of any one of claims 31 to 36, wherein said comonomer comprises one or more of the following structures:

a)



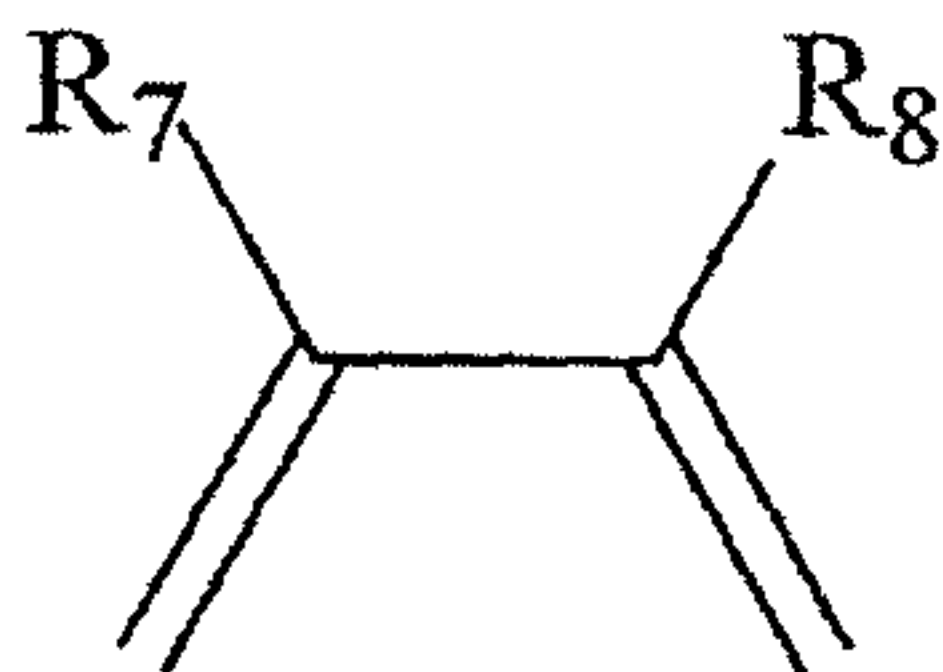
wherein R_2 is $-C(O)OR_3$, phenyl or aryl, heterocyclic, $-OR_4$, or $-O(O)CR_5$; wherein R_3 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; wherein R_4 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; wherein R_5 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; and/or

b)



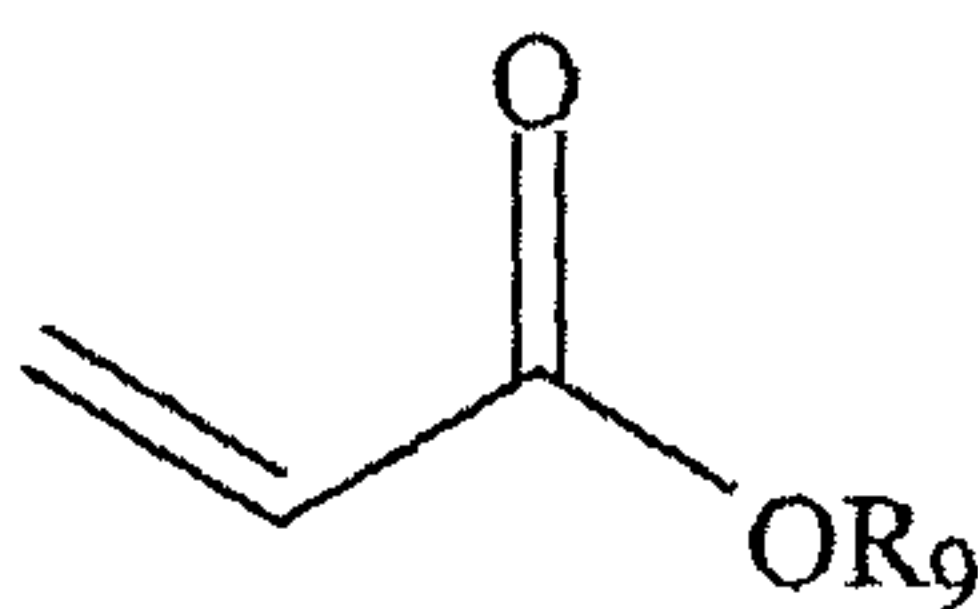
wherein X is O , or $N-R_6$, wherein R_6 is H , C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, aryl-substituted alkyl, or ring-opened alkyl diesters; and/or

c)



wherein R_7 and R_8 can independently be H , C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, aryl-substituted alkyl, or heterocyclic.

38. The method of any one of claims 31 to 36, wherein said comonomer is an acrylate having the following structure:



wherein R₉ is a straight or branched aliphatic alkyl group having 8 or fewer carbon atoms.

39. The method of any one of claims 31 to 36, wherein said comonomer is selected from the group consisting of n-butyl acrylate; 2-ethylhexyl acrylate, 3,5,5-trimethylhexyl acrylate; tert-butyl acrylate, 2-phenylethyl acrylate, iso-butyl acrylate; isobornyl acrylate; n-hexyl acrylate; sec-butyl acrylate; ethyl acrylate; and lauryl acrylate.

40. The method of any one of claims 31 to 36, wherein said comonomer comprises n-butyl acrylate.

41. The method of any one of claims 31 to 40, wherein said polymer particles have a hydrocarbon dissolution rate constant of at least about 0.01 min⁻¹ at 20°C in a mixture of kerosene and isopropyl alcohol ("IPA") in a volume ratio of 95:5 kerosene:IPA.

42. The method of any one of claims 31 to 41, wherein said hydrocarbon-containing fluid has an average temperature of 22°C or less.

43. The method of any one of claims 31 to 42, wherein said treated hydrocarbon-containing fluid has a cumulative concentration of said copolymers in the range of from about 0.1 to about 500 ppmw.

44. The method of any one of claims 31 to 43, wherein said hydrocarbon-containing fluid comprises a liquid phase hydrocarbon, a non-liquid phase hydrocarbon, and/or a non-hydrocarbon fluid, wherein said hydrocarbon-containing fluid comprises said liquid phase hydrocarbon in an amount of at least about 50 weight percent based on the total weight of the hydrocarbon-containing fluid.

45. The method of any one of claims 31 to 44, wherein said hydrocarbon-containing fluid comprises crude oil.

46. A drag reducing composition, comprising:

a continuous phase; and

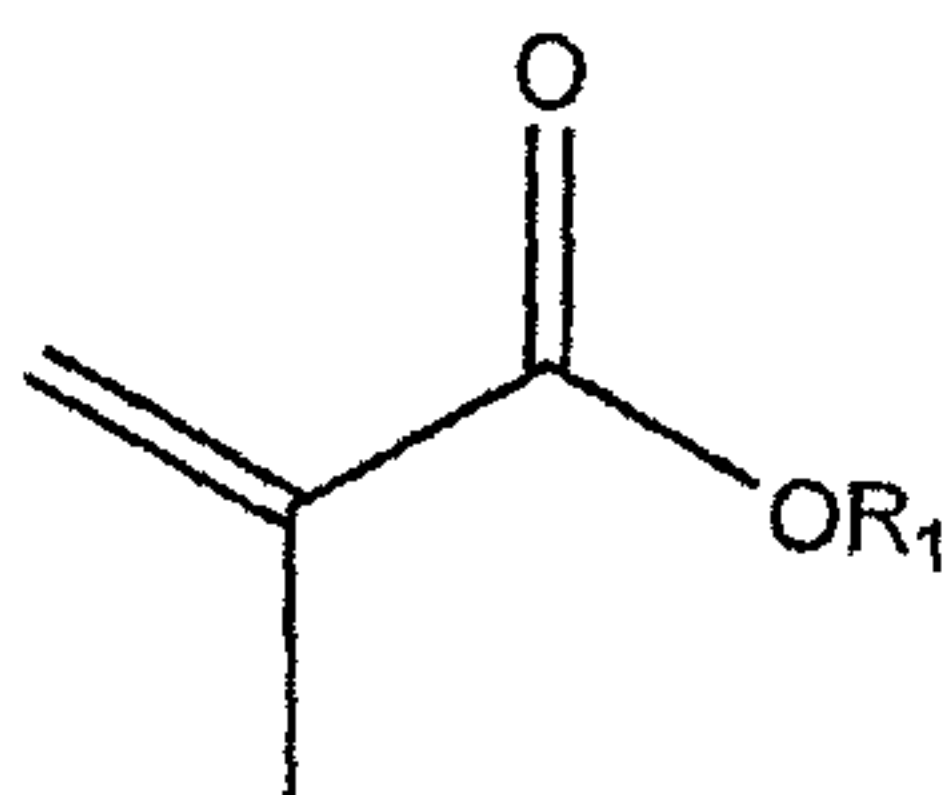
a plurality of drag reducing polymer particles, wherein at least a portion of the polymer particles comprises copolymers containing from about 10 wt% to about 99 wt%

of at least one methacrylate monomer and from about 1 wt% to about 90 wt% of at least one comonomer, wherein the comonomer comprises a polymerizable vinyl group having no more than one pendant substituent per vinyl carbon, wherein the drag reducing composition reduces a pressure drop associated with a turbulent flow of a hydrocarbon fluid having an average temperature of 20°C or less.

47. The drag reducing composition of claim 46, wherein the copolymers comprise saturated hydrocarbon backbones comprising a plurality of carbon atoms, wherein fewer than 50 percent of the carbon atoms have more than one pendant substituent.

48. The drag reducing composition of claim 46 or 47, wherein the copolymers comprise saturated hydrocarbon backbones comprising a plurality of carbon atoms, wherein fewer than 45 percent of the carbon atoms have more than one pendant substituent.

49. The drag reducing composition of any one of claims 46 to 48, wherein the methacrylate monomer has the following structure:



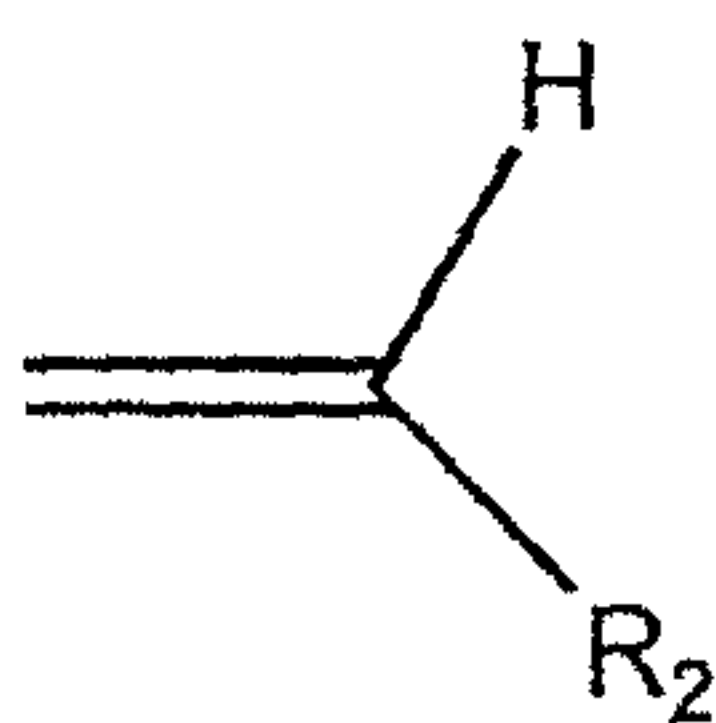
wherein R₁ is a C₁ to C₂₀ alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl.

50. The drag reducing composition of any one of claims 46 to 48, wherein the methacrylate monomer comprises C₄-C₂₀ alkyl, C₆-C₂₀ substituted or unsubstituted aryl, or aryl-substituted C₁-C₁₀ alkyl esters of methacrylic acid.

51. The drag reducing composition of any one of claims 46 to 48, wherein the methacrylate monomer comprises 2-ethylhexyl methacrylate.

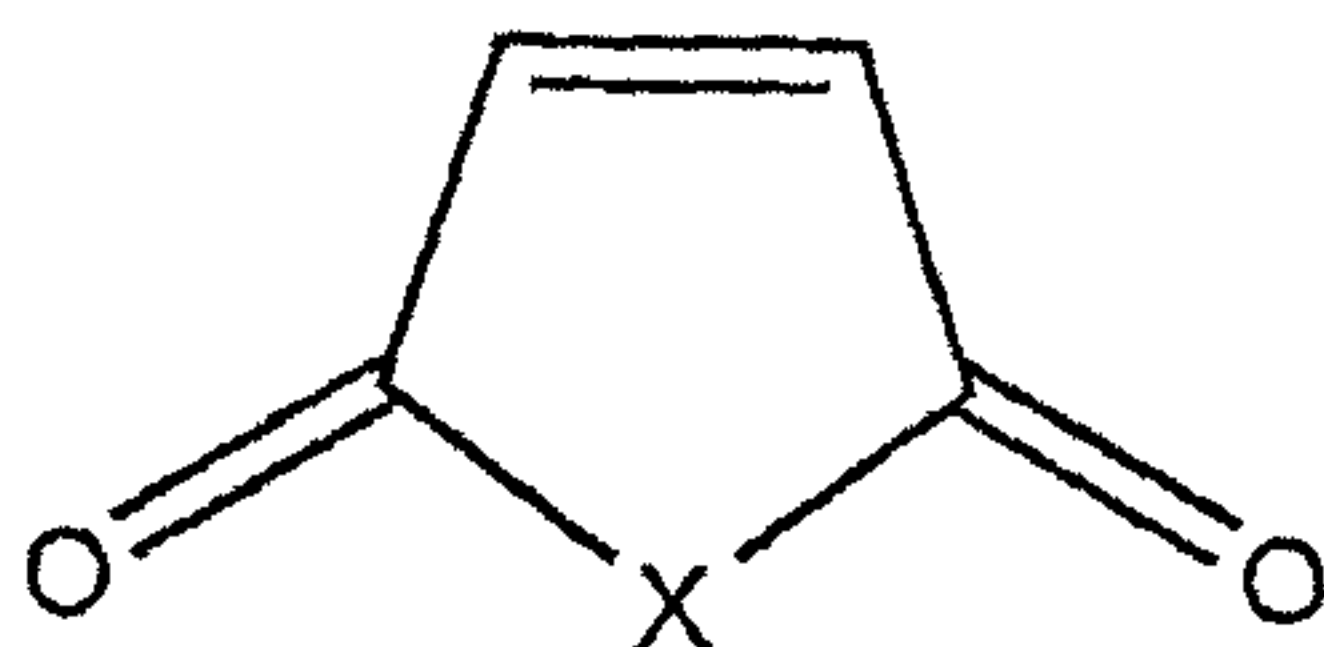
52. The drag reducing composition of any one of claims 46 to 51, wherein the comonomer comprises one or more of the following structures:

a)



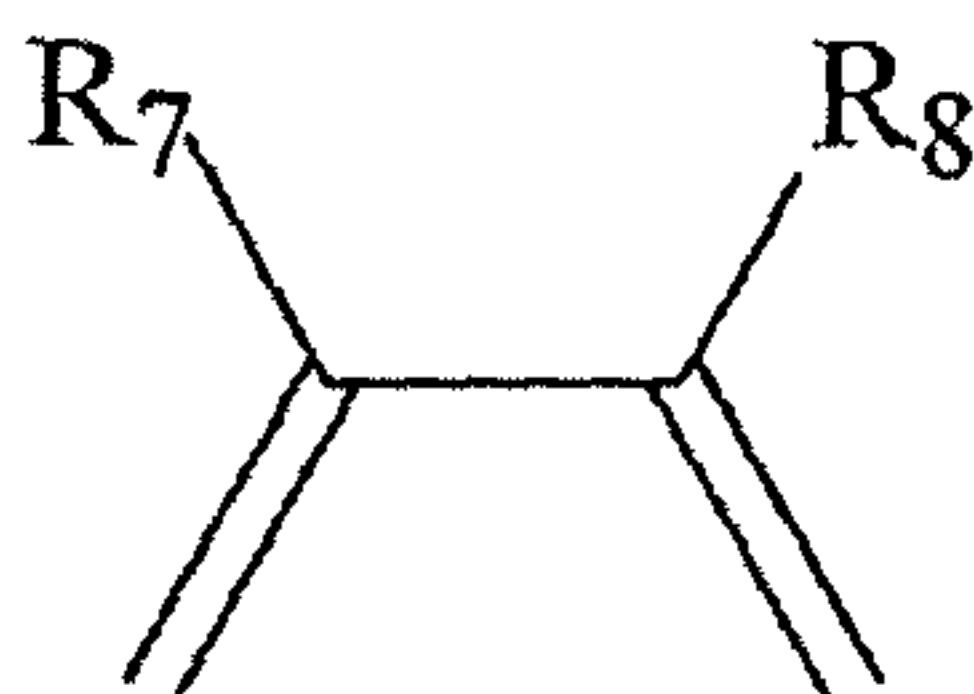
wherein R_2 is $-C(O)OR_3$, phenyl or aryl, heterocyclic, $-OR_4$, or $-O(O)CR_5$; wherein R_3 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; wherein R_4 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; wherein R_5 is a C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, or aryl-substituted alkyl; and/or

b)



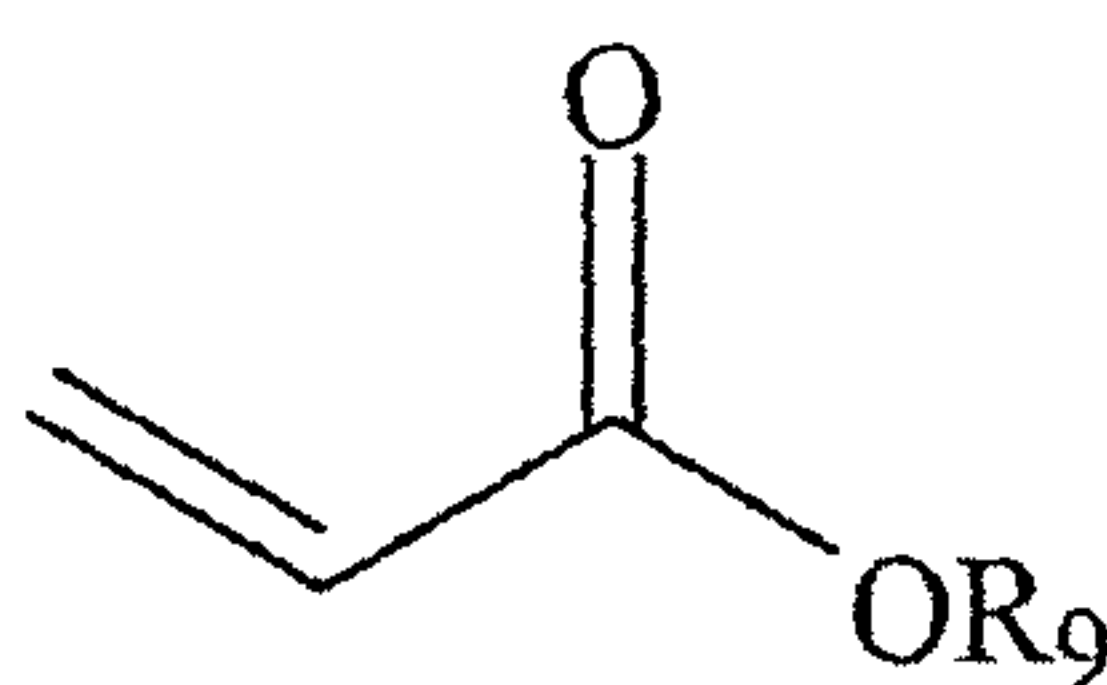
wherein X is O , or $N-R_6$, wherein R_6 is H , C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, aryl-substituted alkyl, or ring-opened alkyl diesters; and/or

c)



wherein R_7 and R_8 can independently be H , C_1 to C_{20} alkyl, isoalkyl, cycloalkyl, aryl, aryl-substituted alkyl, or heterocyclic.

53. The drag reducing composition of any one of claims 46 to 51, wherein the comonomer is an acrylate having the following structure:



wherein R₉ is a straight or branched aliphatic alkyl group having 8 or fewer carbon atoms.

54. The drag reducing composition of claim 53, wherein R₉ is a straight or branched aliphatic alkyl group having 6 or fewer carbon atoms.

55. The drag reducing composition of any one of claims 46 to 51, wherein the comonomer is selected from the group consisting of n-butyl acrylate; 2-ethylhexyl acrylate; 3,5,5-trimethylhexyl acrylate; tert-butyl acrylate; 2-phenylethyl acrylate; isobutyl acrylate; isobornyl acrylate; n-hexyl acrylate; sec-butyl acrylate; ethyl acrylate; and lauryl acrylate.

56. The drag reducing composition of any one of claims 46 to 51, wherein the comonomer comprises n-butyl acrylate.

57. The drag reducing composition of any one of claims 46 to 56, wherein the copolymers comprise the comonomer in an amount in the range of from about 5 to about 60 wt% based on the total weight of the copolymer.

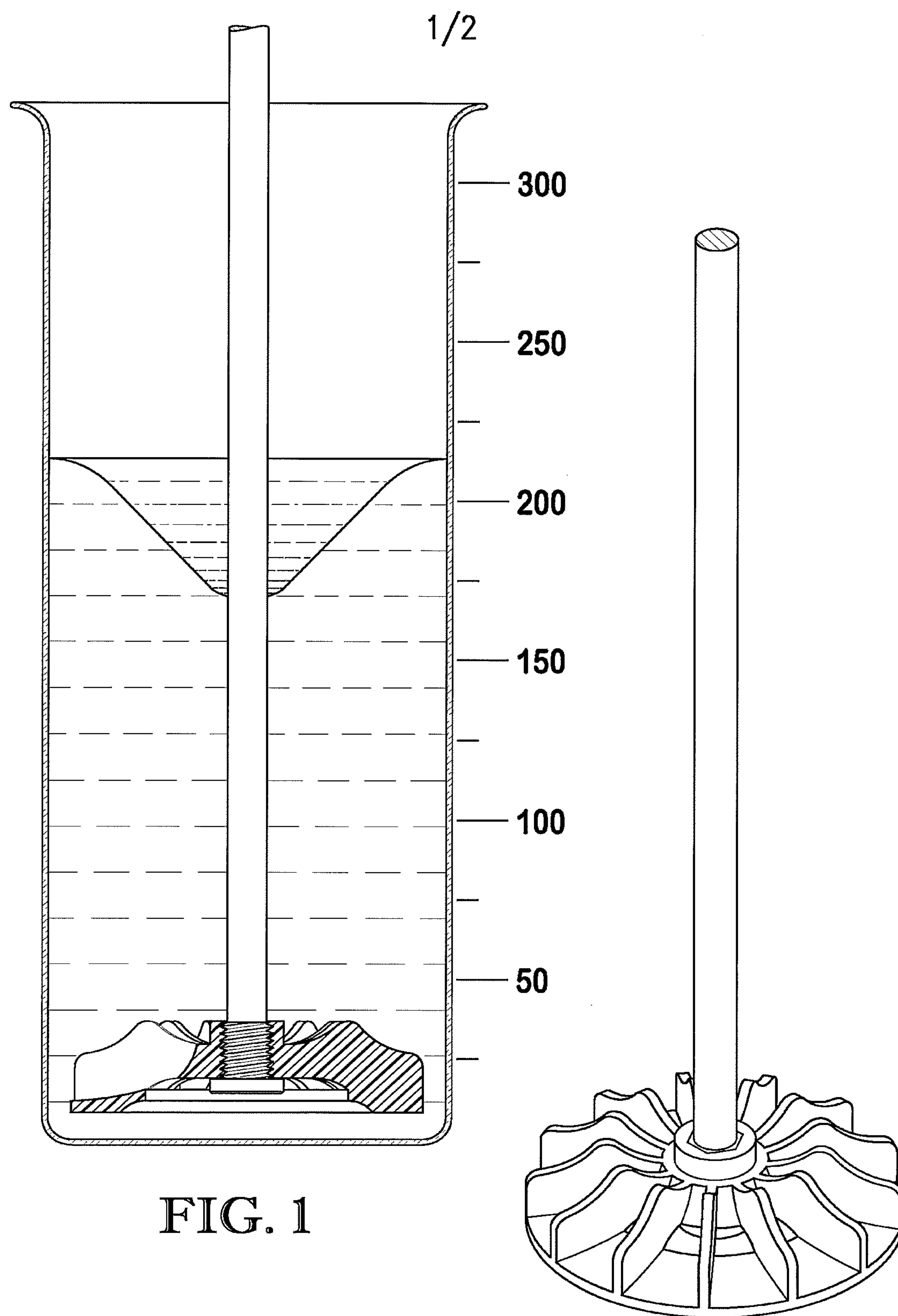
58. The drag reducing composition of any one of claims 46 to 57, wherein said copolymers comprise the methacrylate monomer in an amount in the range of from about 40 to about 95 wt% based on the total weight of the copolymer.

59. The drag reducing composition of any one of claims 46 to 58, wherein the polymer particles have a hydrocarbon dissolution rate constant of at least about 0.01 min⁻¹ at 20°C in a mixture of kerosene and isopropyl alcohol ("IPA") in a volume ratio of 95:5 kerosene:IPA.

60. The drag reducing composition of any one of claims 46 to 59, wherein the continuous phase comprises water, one or more alcohols, one or more polyols, and mixtures thereof.

61. The drag reducing composition of any one of claims 46 to 60, wherein the drag reducing composition comprises the polymer particles in an amount in the range of from about 10 to about 60 wt% based on the total weight of the drag reducing composition.

62. The drag reducing composition of any one of claims 46 to 61, wherein the polymer particles have a weight average molecular weight of at least 1×10^6 g/mol.
63. The drag reducing composition of any one of claims 46 to 62, wherein the drag reducing composition comprises an emulsion drag reducing polymer wherein the polymer particles have an average particle size in the range of from about 10 to about 500 nm.
64. The drag reducing composition of any one of claims 46 to 62, wherein the drag reducing composition comprises a suspension drag reducing polymer wherein the polymer particles have an average particle size in the range of from about 1 to about 500 micrometers.



2/2

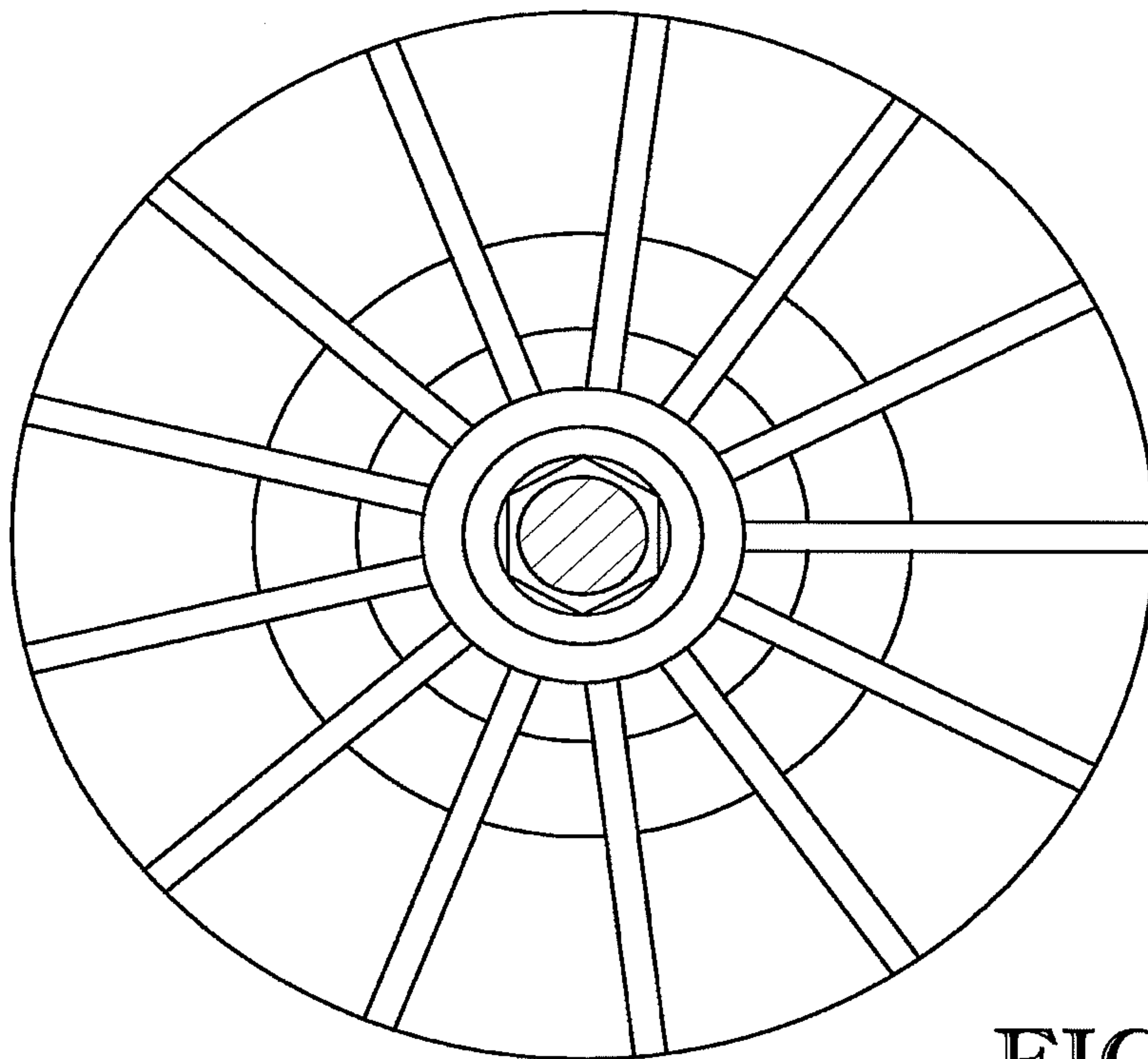


FIG. 3

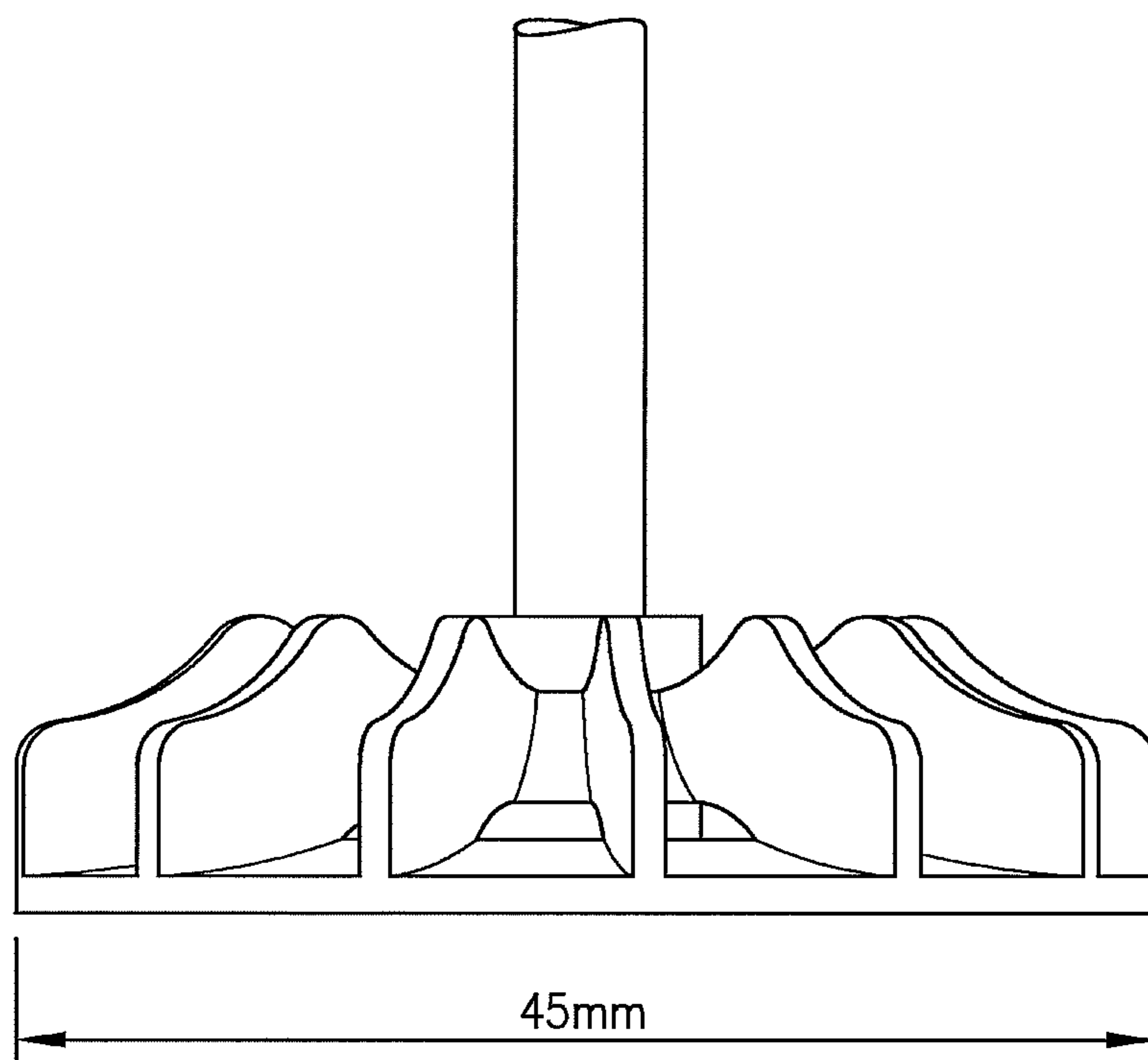


FIG. 4

