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2,751,352

## MAGNETIC FLUIDS

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This invention relates to magnetic fluids and more particularly to magnetic fluids wherein the magnetic particles are inhibited from precipitating or settling out of the fluid system.

Magnetic fluids of the type with which this invention is concerned and their application to various uses such as in hydraulic systems, dashpots and clutches is fully discussed by Jacob Rabinow of the National Bureau of Standards, in a paper presented before the AIEE meeting in Washington, D. C., October 5-7, 1948. In essence the magnetic fluid for use in a clutch or the like operates on the following basic principle:

When the space between two parallel (magnetic) surfaces is filled with finely divided magnetizable or magnetic particles and a magnetic field is established between the two plates, the magnetic particles bind the plates together against movement parallel to their surfaces. The magnetic particles may be finely divided iron which, for most applications, is mixed with a liquid, such as oil, to prevent packing and to afford smoother operation of the clutch. When a portion of this mixture is acted on by a magnetic field, the iron particles are mutually attracted, bind together in the field, and the mixture seemingly solidifies, an effect readily demonstrated by lowering a small permanent magnet into a beaker of iron-oil mixture. As the magnetic field can be produced by an electric current, a very simple means is thus obtained for the control of the binding force over a very wide range.

In fluids of the type under discussion the magnetic particles such as iron or iron oxide and the like tend to settle out of the fluid in which they are dispersed, forming heavy sediment and necessitating the re-mixture of the fluid before it can be used. Various means have been employed in order to prevent settling out of iron particles from the fluid in which they are dispersed. Thus, very viscous fluids have been tried as suspending agents or additives having a tendency for form thixotropic mixtures, as well as other means, all with little success.

It is an object of this invention to produce an improved magnetic fluid. It is another object of this invention to produce a stable magnetic fluid. Still another object of this invention is to prevent settling out of the magnetic particles of the liquid carrier in which they are dispersed. Other objects of this invention will appear as the description proceeds.

It has now been discovered that separation or precipitation of magnetizable or magnetic particles such as iron powder from base magnetic fluids in which they are dispersed, can be prevented by addition to said base fluids of a minor amount sufficient to prevent separation of magnetic particles from such base magnetic fluids, of a material, preferably liquid in character which is insoluble in said base fluid carriers, but which possess ferrophilic properties.

More specifically, it has been discovered that by addition of a minute amount of an oleophobic material to a magnetic fluid, containing magnetizable or magnetic particles such as iron powder, the settling out or precipitation

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of the magnetic particles can be prevented or inhibited. The oleophobic material should also possess ferrophilic properties and can be selected from a wide class of materials. The oleophobic materials suitable as suspending agents for magnetic solids suspended in magnetic fluids of this invention should be low molecular weight oleophobic polar materials of which oxygen and/or nitrogen-containing oleophobic-ferrophilic liquids are particularly preferred. By the terms "magnetophilic" and "oleophobic-ferrophilic liquids" is meant to include such liquids which tend to be readily attracted to magnetic particles such as metal (iron) particles and coat said particles so as to prevent the particles from being wetted by the liquid base such as being oil wetted. Among the oleophobic-ferrophilic liquids which are particularly suitable as dispersing agents for colloidal iron particles and the like, suspended in various liquid mediums, can be included low molecular weight alcoholic materials as well as amide derivatives of low molecular weight acids having not more than five carbon atoms and water-soluble amines. Illustrative examples of dispersing agents for use in composition of this invention are: ethyl alcohol, propyl alcohol, isopropyl alcohol, allyl alcohol, mercaptoethanol, glycerol, ethylene glycol, propylene glycol, pentanediol-2,4; hexanediol-2,5; butanediol-1,3; ethylene diamine, diethylene triamine, N-hydroxy ethyl propylene diamine, morpholine, N-methyl morpholine, triethanolamine; foramide, acetamide and the like.

The base carrier or vehicle for the magnetic particles can be selected from a wide variety of materials preferably oleaginous in character and possessing lubricating properties such as natural and/or synthetic lubricants.

Mineral oils of any viscosity range varying from about 40 to 2,000 SUS at 100° F., having high flash and fire points and mixtures thereof can be used, said mineral oils being obtained from any petroleum crude. Preferred mineral oils are refined ones, e. g., steam cylinder oils, bright stock oils, white oils, light machine oil, etc., 300 to 700 SUS at 100° F., a viscosity index of from about 40 to 70 and an average molecular weight of from about 350 to 550. Mixtures of mineral oil and fixed oils such as castor oil, lard oil and the like can be used as well as organic synthetic lubricants and mixtures thereof, such as:

I. Synthetic lubricants produced by the Fischer-Tropsch, Synthol, Synthine and related processes, e. g.,

A. Polymerization of olefins such as ethylene, butylene, and the like, and their mixtures in presence of a Friedel-Crafts or other type catalysts under elevated temperatures and pressures as well as lubricating bases as described in U. S. Patent 2,526,986.

B. Polymerization of unsaturated hydrocarbons in presence of a catalyst and then condensing said polymerized product with an aromatic hydrocarbon such as xylene, benzene and naphthalene.

C. Oxidation of polymerized olefins of lubricating range as noted under A and B.

D. Conversion of natural gas to carbon monoxide and hydrogen, followed by catalytic reaction under elevated temperature and pressure to produce hydrocarbons of lubricating range (Synthol Process).

II. Bergius process for producing synthetic lubricants, e. g.,

A. Hydrogenation of coal, peat, and related carbonaceous materials under pressure and elevated temperature in presence of a catalyst.

B. Hydrogenation of asphalts, petroleum residues and the like.

### III. Voltolization process for producing synthetic lubricants, e. g.,

- A. Voltolization of fatty materials such as fatty oils.  
 B. Voltolization of mixtures of fatty oils and petroleum hydrocarbons.  
 C. Voltolization of unsaturated hydrocarbons, their mixtures, and the like.

### IV. Organic synthetic lubricants

A. Complete alkyl esters of organic acids, e. g., alkyl lactates, alkyl oxalates, alkyl sebacates [di(2-ethylhexyl) sebacate], alkyl adipates [di(2-ethylhexyl) adipate], alkyl phthalates (dioctyl phthalates), alkyl ricinoleates (ethyl ricinoleate), alkyl benzoates.

B. Alkyl or alkylaryl esters of inorganic acids, e. g., tricresyl phosphate, trioctyl phosphate, dibutyl trichloromethanephosphonate, trixylenyl phosphate, tributyl phosphate, triethyl phosphate.

V. Synthetic lubricants made from polymerization of alkylene oxides at elevated temperatures in the presence of catalysts such as iodine, hydrogen iodide, etc.

A. Polymers of: ethylene glycol, trimethylene glycol, propylene glycol, tetramethylene glycol, hexamethylene glycol, pentamethylene glycol.

B. Copolymers of: trimethylene glycol and triethylene glycol, trimethylene glycol and hexamethylene glycol, trimethylene glycol and B-methyltrimethylene glycol, trimethylene glycol and diethylene glycol.

C. Copolymers prepared from certain peroxides at elevated temperatures and in the presence of alkali metal base (KOH) or BF<sub>3</sub>-ether catalyst, e. g., ethylene oxide and propylene oxide, isobutylene oxide and propylene oxide.

D. Sulfur-containing reaction products obtained by treating allyl alcohol, divinyl ether, diallyl ether, dimethylallyl ether and glycols with H<sub>2</sub>S in the presence of a catalyst such as toluene sulfonic acid, e. g., dihydroxy dipropyl sulfide, trimethylene glycol and dihydroxy dipropyl sulfide, trimethylene glycol and hydroxy diethyl sulfide.

VI. Polymers obtained from oxygen-containing heterocyclic compounds, e. g., polymerization of tetrahydrofuran in the presence of a catalyst.

VII. Silicone compounds, e. g., silicate esters (alkyl and/or aryl silicates, e. g., tetraphenyl silicate, tetra ethyl hexyl silicate), polyalkyl silicone polymers (dimethyl silicone polymers), Dow-Corning fluids, alkylaryl silicone polymers, e. g., (methyl phenyl silicone polymers).

VIII. Fluoro and/or chloro carbon oils, e. g., fluorinated petroleum fractions, e. g., kerosene, trifluorochloroethylenes, hexachlorobutadiene, chlorinated biphenyls.

The metal magnetic particles or powders used to form magnetic fluids of this invention can be any materials possessing magnetic properties having dimensions of from 2 to 100 microns and preferably from 5 to 30 microns. Powder iron produced by decomposition of carbonyl iron is particularly suitable for use in magnetic clutch fluids of this invention, although powder iron and/or iron oxide produced by any other suitable means can be used.

Highly desirable magnetic fluids such as magnetic clutch fluids can be prepared by using formulations within the following range:

	Amount percent wt.	
	Broad Range	Narrow Range
Base carrier, e. g., natural and/or synthetic lubricant.....	3-79	10-40
Magnetic material, e. g., powder iron.....	97-10	60-90
Dispersing agents, e. g., oleophobic-ferrophilic material.....	0.001-10	0.01-5
Optional addition anti-oxidant, corrosion inhibitor, etc.....	0-1	0-0.1

Specific compositions of this invention are:

Composition A:	Percent weight
Kerosene .....	14
Iron powder $d_0=7-10 \mu$ .....	84.9
Ethylene glycol .....	0.1
Composition B:	
Dimethyl siloxane polymer .....	15
Iron powder (prepared from iron carbonyl) $d_0=7-10 \mu$ .....	84.9
Ethylene glycol .....	0.1
Composition C:	
Mineral oil .....	15
Iron powder .....	84.95
Propylene glycol .....	0.05
Composition D:	
Mineral oil .....	15
Iron powder .....	84.95
Glycerol .....	0.05
Composition E:	
Di(2-ethyl hexyl sebacate) .....	15
Iron powder .....	84.9
Ethylene glycol .....	0.1
Composition F:	
Mineral oil .....	15
Iron powder .....	84.9
Formamide .....	0.1
Composition G:	
Tetraphenyl silicate .....	20
Iron powder .....	79.9
Ethylene glycol .....	0.1
Composition I:	
Trifluorochloroethylene polymer .....	25
Iron powder .....	74.9
Ethylene glycol .....	0.1
Composition J:	
Fluorinated petroleum oil .....	28
Iron powder .....	72.95
Ethylene glycol .....	0.05
Composition K:	
Chlorinated biphenyl .....	22
Iron powder .....	77.9
Ethylene glycol .....	0.1
Composition L:	
Mineral oil .....	23
Iron oxide .....	76.5
Ethylene glycol .....	0.5
Composition M:	
Mineral oil .....	39.9
Iron powder .....	60
Acetamide .....	0.1
Composition N:	
Mineral oil .....	30
Iron powder .....	69
Ethylene diamine .....	0.1
Ethylene glycol .....	0.9
Composition O:	
Mineral oil .....	20
Iron powder .....	75
Propylene glycol .....	0.5
2,6 ditert-butyl-4-methyl phenol .....	4.5
Composition P:	
Mineral oil .....	25
Iron powder .....	74
Ethylene glycol .....	0.1
Phenyl-2-naphthylamine .....	0.9
Composition Q:	
Mineral oil .....	20
Iron powder .....	79.8
Ethylene glycol .....	0.1
Sodium nitrite .....	0.1

To illustrate the [pronounced dispersing or suspending] properties which additives of this invention impart when added to magnetic fluids the following tests were made and the results are tabulated below.

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The compositions identified hereinbelow were placed in 2 oz. bottles and allowed to stand for 5 days at room temperature undisturbed and the amount of oil separation observed.

Composition	Percent oil separation
A -----	0
B -----	0
C -----	0
E -----	0.1
G -----	0
X <sup>1</sup> -----	40
Y <sup>2</sup> -----	50
Z <sup>3</sup> -----	70

<sup>1</sup> Composition X=1 part mineral oil+6 parts iron powder.

<sup>2</sup> Composition Y=Composition X+0.1 part of oleophilic surface active agent (oil-soluble ester).

<sup>3</sup> Composition Z=Composition X+0.1 part of oleophilic surface active agent (oil-soluble fatty acid).

Compositions of this invention can be modified by addition thereto of minor amounts (0.01-1%) of anti-oxidants, corrosion inhibitors, extreme pressure agents, anti-wear agents and the like such as alkyl phenols (2,6 di-tertbutyl-4-methyl phenol), amines, e. g., phenyl-naphthylamine; organic phosphorus compounds, e. g., dilauryl phosphate, dialauryl phosphite, tributyl phosphate, tricresyl phosphate; sodium nitrite, lithium nitrite, etc.

I claim as my invention:

1. A stable magnetic fluid consisting essentially of a mixture of from about 60% to 90% of iron particles having dimensions of from 5 to 30 microns and from about 10% to about 40% of a liquid hydrocarbon, said mixture containing from about 0.01% to about 5% of an alkylene glycol selected from the group consisting of ethylene glycol and propylene glycol.

2. A stable magnetic fluid consisting essentially of a

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mixture of from about 60% to 90% of iron particles having dimensions of from 5 to 30 microns and from about 10% to about 40% of mineral oil, said mixture containing from about 0.01% to about 5% of an alkylene glycol selected from the group consisting of ethylene glycol and propylene glycol.

3. A stable magnetic fluid consisting essentially of a mixture of from about 60% to 90% of iron particles having dimensions of from 7 to 10 microns and from about 10% to about 40% of mineral oil, said mixture containing from about 0.01% to about 5% of ethylene glycol.

4. A stable magnetic clutch fluid consisting essentially of a mixture of iron particles having dimensions of 7 to 10 microns and kerosene in the ratio of 6 to 1 respectively, and about 0.1% of ethylene glycol based on the total composition.

5. A stable magnetic fluid consisting essentially of a mixture of from about 60% to 90% of iron particles having dimensions of from 7 to 10 microns and from about 10% to about 40% of mineral oil, said mixture containing from about 0.01% to about 5% of propylene glycol.

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