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GEL AND LUBRICANT MADE THEREFROM

Samuel S. Kistler, West Boylston, Mass., assignor to Monsanto Chemical Company, St. Louis, Mo., a corporation of Delaware

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This invention pertains to the art of thickening liquids and more particularly it relates to a lubricating oil of increased viscosity and the method of preparing the same.

Oils have heretofore been thickened or rendered more viscous by admixture with other materials, but it has been necessary that the matter introduced be added in relatively large amounts to achieve the desired result.

It is an object of this invention to provide a thickening agent for oils and other liquids which will materially increase the viscosity of the liquid when the amount of added material is small.

A further object of this invention is the provision of a thickened oil which shows an abnormally small change in viscosity with variations in temperature.

This invention also contemplates the provision of an addition agent for oils and similar liquids which under conditions of use is inert to the liquid and the bearings or other parts with which the liquid comes in contact during normal usage.

As an additional object the invention contemplates a thickening agent which is unaffected by operating temperatures, and lubricating oils and greases embodying such thickening agents which possess markedly improved properties when compared to present day lubricating oils and greases.

My invention further contemplates the provision of a lubricating grease having novel and useful properties as to viscosity, appearance, viscosity index, and stability.

This invention includes the discovery that if liquids are thickened with particles of a gel in which the original gel structure is retained substantially intact, the product possesses improved characteristics of great value.

Compositions of the type to which this invention is directed may be prepared by the addition to the liquid to be thickened of the aerogels described and claimed in U. S. Patent Number 2,093,454 to S. S. Kistler. Similar results are obtainable by replacing the swelling liquid of the gel with a portion of the liquid to be thickened.

In my copending application Serial Number 151,969, there is disclosed a process of preparing gels in which the continuous phase is an organic liquid, as an oil of mineral, animal, or vegetable origin.

As illustrative of the processes and products forming part of this invention, the preparation of a lubricating oil having incorporated therein a gel will be discussed. In oils intended primarily for lubrication, it is preferred to use gels of substances having a relatively high melting point, and in that group better results are obtained by use of gels of an inorganic nature. It is to be understood, however, that the principal advantages of my invention can be obtained with aerogels of any material not incompatible with the fluid to be thickened, and the invention contemplates the use of aerogels of silica, magnesia, alumina, gel-forming metal oxides and hydroxides in general, gelatine, albumin, cellulose, and gels of like nature.

Inorganic fillers for lubricating oils have been heretofore used for the purpose of thickening the same, and as representative of this type of addition agent, reference is had to graphite, mica, sulfur, talc, asbestos, and diatomaceous earth. These compounds are reduced to a fine state of subdivision only with some difficulty and the porosity thereof is small. The characteristics of the present compounded oils are attributed in large measure to the high percentage of voids in the thickening agent. It is believed that a comparison of the compositions with those prepared from diatomaceous earth will point out most clearly the advantages of this invention. For the purpose of this comparison tests were made of oils containing silica aerogels.

The gels used in these tests were manufactured from hydro-gels of silica made by precipitating silica by addition of sulfuric acid to sodium silicate solution in the well-known manner and washing the gel relatively free of salts with water. This composition is now used in industry as an intermediate in the making of silica gel. The prior practice has been to dry out the gel in any convenient manner thereby materially reducing the volume of the gel. Such shrinkage is probably due to the force exerted on the solid phase of the gel by the surface tension of the liquid as it recedes in the pores of the material. These materials will be referred to as xerogels. As distinguished from the xerogels, the aerogels are composed of the original solid phase of the gel in substantially the same condition as it exists while filled with the swelling liquid. Aerogels are conveniently made by raising the gel to the critical temperature of the liquid contained therein, while maintaining the pressure on the system sufficiently high to insure that the liquid phase will remain liquid until the critical temperature is reached. At this point, the liquid will be converted into the gaseous state without the formation of menisci at a gas-liquid interface. The degree of porosity may be controlled to a large degree by controlling the concentration of silica in the gel as precipitated.

In the method of preparing the aerogel now in use, the liquid is removed by heating in an autoclave. It is the practice to remove the product by a suction line. The gel is drawn through the centrifugal fan employed to create the suction and during its passage through the fan is reduced to small particles. About 70% of the mass will pass a 100 mesh screen; 10% remains on a 60 mesh screen. It will be understood that when unground aerogel is hereafter referred to, the product referred to is that collected after passage through the fan without further subdivision.

A thickened oil is prepared by mixing the thin oil with a small proportion of aerogel with vigorous mechanical agitation. In order to inhibit settling of the thickening agent, it has been found advisable to grind the mixture in a ball mill for a period depending upon the properties desired, the properties of the oil, and the fineness of the aerogel. The following data is illustrative of results obtained by thickening oils with silica aerogel:

Material tested	Viscosity at 27° C.
SAE No. 10 motor oil	11
SAE No. 20 motor oil	24
SAE No. 40 motor oil	54
SAE No. 10 oil plus 2% unground aerogel	123
SAE No. 10 oil plus 2% aerogel ground 1 hr	54
SAE No. 10 oil plus 4% aerogel ground 18 hrs	53

The viscosity measurements were taken on a MacMichael viscosimeter with a No. 26 wire. The ground gels were ground for the periods indicated in a stone ball mill and the percentages are based on weight.

The desired result may also be achieved by forming a grease in the manner of any of the processes described below and adding more oil to the composition by grinding in a ball mill.

Study of the above table will definitely show that an oil having markedly increased viscosity can be prepared from light oils. A further advantage of oils prepared in accordance with the present invention is their relative stability to temperature change. The following table demonstrates this fact by comparison of aerogel containing oils and untreated oils of comparable viscosity. In this table oil A is a pure SAE No. 20 lubricating oil. Oil B is SAE No. 10 oil containing 3% by weight of silica aerogel ground in a ball mill for 24 hours. Oil C is a pure SAE No. 40 oil, while oil D is a No. 10 oil containing 7% by weight of silica aerogel ground for 24 hours.

Oil	Temperature, degrees C.	Viscosity in centipoises	Percentage decrease in viscosity per 10° rise in temp.
A	17	270	
	27	130	48.1
B	17	190	
	27	130	31.6
C	17	630	
	27	300	47.6
	37	165	45.0
D	17	450	
	27	340	24.4
	37	315	7.3

These results indicate quite clearly that the aerogel oils demonstrate a stability toward temperature change which is very desirable in lubricants, especially those intended for use on bearings which are idle for periods long enough for them to become cold. Moving parts which are subjected to a range of temperatures or those normally used at high temperature are especially benefited by lubrication with the present compounded oils.

It has been found desirable to use finely ground aerogel in preparing thickened lubricating oils, since a smoother, more uniform suspension is obtained. It should be noted in this connection that a larger amount of the ground gel is required to give the same results than are gained by a small proportion of the unground aerogel.

Incorporation of a larger amount of aerogel than is required to merely thicken the oil results in a grease of excellent qualities. As is the case with thickened oils, it is found that the amount of aerogel required to make a grease of desired qualities varies with the fineness of the aerogel. It has been determined that a good grease may be prepared from motor oil with a viscosity of ten seconds, Saybolt, by incorporation therein of 10% of unground aerogel on the weight of the mixture. If the aerogel has been ground one to two hours, about 20% by weight will be required to form a grease of satisfactory consistency, while 30% or more of aerogel ground 20 to 24 hours may be used.

A grease of good properties was formed by adding 39 grams of unground aerogel to 350 grams of No. 10 motor oil with vigorous mechanical stirring and subsequently running the resultant slurry through a roller mill several times. The mill used was of the type commonly used to grind paints. It consists of three steel rolls revolving at different speeds. The first and third rolls revolve in the same direction and opposite to the direction of rotation of the second roll; the rolls revolve progressively faster in the direction of the flow of the material. The shearing action of the rollers serves to grind the oil into the pores of the aerogel, which becomes visibly stiffer after the first passage through the mill and at the end of the sixth pass is found to be a stiff, transparent, even bodied grease. Such grease is stiff and dry and does not exude oil upon standing for prolonged periods.

For the purposes of comparison, it was attempted to make a grease by adding 30.7% of fuller's earth to a No. 10 oil. The slurry was as thick as could be agitated practically with the mixing device used for the aerogel thickened oils and greases, but could not be formed into a grease on the roller mill. After several passes through the mill, the mixture remained sloppy, and a film of oil separated on top when it was allowed to stand.

A test was made to determine the relative stability of aerogel greases and a good grade of commercial cup grease to heat. The commercial cup grease used for this purpose was of the type in which body is imparted by the addition of a water insoluble soap, while the aerogel grease was composed of 90% No. 10 motor oil and 10% of unground silica aerogel. At room temperature the aerogel grease possesses a cleaner, more attractive appearance. The color is opalescent yellow, and a one-inch layer is translucent. No difference between the two could be detected in viscosity, texture, or lubricating qualities.

Samples of the greases were then placed on watch glasses and heated to 100° C. in an oven. No change in properties could be detected in the grease containing silica aerogel. The commercial grease, on the other hand, was materially affected by the higher temperature. The viscosity was decreased substantially, and the

grease became much shorter than it had been in the cold state.

It has been found that a grease of good quality can be made by successive replacements of the continuous phase of a gel to obtain a substance which may be denominated as "petro-gel." This latter composition is composed of the original gel structure, with the continuous phase being a petroleum fraction. It is to be understood that gels containing lubricating fluids other than those derived from mineral oil may also be formed, and the following discussion of petro-gels is merely illustrative of this feature of the invention.

The liquid continuous phase in a gel may be replaced by another liquid, provided the two liquids are mutually miscible. By employment of that principle, a gel may be prepared in which the continuous phase is a lubricating liquid such as a mineral, vegetable, or animal oil. Exemplary of such gels is the result of replacing the water in a silica gel with acetone, replacing the acetone with textile spirits, and then substituting a petroleum lubricating oil for the textile spirits. Such replacements are accomplished by extraction of the gel with the replacing liquid. If the petroleum oil is a No. 10 lubricant, a grease having desired properties can then be obtained by the incorporation of additional oil on the roller mill.

By similar manipulative steps, a lubricant containing the structure of an organic gel, such as a swelled resin, can be prepared. A sheet of cellulose (Cellophane) was swelled in sodium hydroxide solution, after which the sodium hydroxide was removed by washing with water and the water in turn was displaced by alcohol. The alcohol was then displaced by ether in order to obtain a liquid phase that is miscible with an oil of mineral or vegetable origin. The ether gel was thereupon washed with a light petroleum lubricating oil whereby the ether was displaced and there resulted a material in sheet form that is suitable for some types of bearings, and packing for bearings, shafts, etc. since it is essentially a relatively firm sheet of soft material consisting predominately of lubricant.

Alternatively, a petro-gel may be formed by immersing in a desired oil, a textile spirits gel, such as silica gel in which the continuous phase is textile spirits, and distilling off the textile spirits by any suitable method. The preferred method contemplated by this invention is the preparation of a slurry or suspension of textile spirits gel in lubricating oil from which the lighter oil is removed by heating the mixture above the boiling point thereof with agitation. It is to be understood that the invention is not restricted to the use of textile spirits, but rather that any liquid boiling at a temperature substantially below the boiling point of the lubricant may be suitable if miscible with the liquid of the previous step and also with the lubricant.

A petro-gel may be made in various ways other than by an intimate mixing or grinding of an aerogel in a ball mill or a colloid mill with the petroleum oil, or fraction. One method which has been already indicated consists in displacing the water in a hydro-gel such as a silica hydrogel with alcohol and thereafter displacing the alcohol with a liquid which is miscible with the petroleum fraction as, for example, ether, acetone and the like. The general procedure used is described in my co-pending application, Serial No. 151,969, filed July 3, 1937, and my Patent No. 2,093,454. The resulting acetone gel or ether gel is mixed with a petroleum cut to give the petro-gel. The low-boiling liquid may thereafter, if desired, be removed as by distillation as described.

It is to be understood that this invention is not limited to any specific gel or petro-gel and that aerogels of alumina, tin oxide and the like may be substituted for silica as has previously been indicated. It is likewise to be understood that the lubricants and greases of the present invention may include the present day compounding agents used in this art such as the water-soluble soaps, graphite, fats, and material for improving the oiliness characteristics such as chlorinated hydrocarbons of high boiling point; materials which improve the viscosity index characteristics such as polymerized olefins of high molecular weight, such as Paratone; corrosion inhibitors; chemical stabilizing agents; pigments and the like may be added to the lubricant within the contemplation of the present invention.

In lieu of the lubricants of petroleum origin one may substitute wholly or in part vegetable or animal oils as well as synthetic oils such as those obtained by alkylating aromatic hydrocarbons as is well understood by those skilled in the art.

It has been found preferable for some purposes to prepare a thicknened oil from a previously formed grease of the gel to be used in thickening the oil. Vigorous mechanical agitation of the grease with untreated oil results in the formation of an oil having a viscosity intermediate between the viscosities of the grease and the untreated oil.

This invention provides a means of thickening oils and forming greases by incorporation therein of a previously formed gel in substantially the form of the gel as originally prepared. By this means it is possible to use relatively small proportions of thickening agent. A further important advantage lies in the provision of a means to incorporate into the liquid a thickening agent of high melting point, in order that the effect of the thickening agent will be retained over a much wider range of temperatures than is possible with thickening agents now used in the preparation of of greases.

This application is a continuation in part of my application, Serial Number 151,969, filed July 3, 1937, now Patent 2,188,007 of January 23, 1940.

I claim:

1. A lubricant comprising an oily liquid substance and a solid substance consisting principally of an inorganic gel formed apart from the liquid substance but retaining in the thickened liquid the gel structure substantially as formed, said gel being substantially free from liquids other than said oily liquid substance.

2. A lubricating composition comprising an oily liquid substance and a proportion less than 50% by weight of a substantially water free gel originally formed apart from the said composition in an aqueous medium and characterized by the fact that the structure of the water free gel is substantially that of the gel as originally formed.

3. As a lubricant, a gel having as its characterizing constituents, an inorganic solid phase and a liquid phase consisting of a water immiscible organic oil selected from the group consisting of petroleum hydrocarbons, animal and vegetable oils.

4. As a lubricant, a gel having as its characterizing constituents an inorganic solid phase composed essentially of silica and a liquid phase

consisting of a water immiscible organic oil selected from the group consisting of petroleum hydrocarbons, animal and vegetable oils.

5. A lubricant containing an oily liquid substance and, in an intimate mixture therewith in an amount sufficient to thicken or increase the viscosity of said oily liquid substance, an inorganic aerogel, said lubricant being substantially free of water.

6. A thickened lubricant comprising an oily liquid substance and a solid substance consisting principally of a substantially water free gel originally formed apart from the liquid substance in an aqueous medium, said gel retaining in the thickened liquid substance the gel structure substantially as originally formed.

7. A lubricant containing an oily liquid substance and an aerogel in an intimate mixture therewith in an amount sufficient to thicken or increase the viscosity of said oily liquid substance, said lubricant being substantially free of water.

8. A thickened lubricant comprising an oily liquid substance and a solid substance consisting principally of a substantially water free organic gel originally formed apart from the liquid substance in an aqueous medium, said gel retaining in the thickened liquid substance the gel structure substantially as originally formed.

9. A lubricant containing an oily liquid substance and an organic aerogel in an intimate mixture therewith in an amount sufficient to thicken or increase the viscosity of said oily liquid substance, said lubricant being substantially free of water.

10. A lubricant containing an oily liquid substance and from 10 to 49% by weight of an aerogel based on the weight of the mixture, said aerogel being present in an amount sufficient to thicken or increase the viscosity of said liquid substance and form a grease therefrom, and said lubricant being substantially free of water.

SAMUEL S. KISTLER.