

at 60° C. for 1 hour, during which time it was quickly converted to an optically clear, hard, glass-like polymer containing a first class of organocyclotetrasiloxane units and a second class of organocyclotetrasiloxane units, with there being substantially twice the number of said second class of organocyclotetrasiloxane units as the number of said first class of organocyclotetrasiloxane units, with substantially every unit of said first class being attached to four units of said second class through a silicon-bonded ethylene radical, with substantially every unit of said second class being attached through its 1- and 5-silicon atoms, respectively, to two units of said first class through said silicon-bonded alkylene radicals, with the valences of silicon other than the valences satisfied by oxygen in the cyclotetrasiloxane rings and the valences satisfied by silicon-bonded ethylene radicals being satisfied by methyl radicals. This polymer had a structure similar to the structure shown in Formula 7. The polymeric material exhibited no change in weight or physical appearance when maintained at a temperature of 200° C. for 48 hours, and exhibited no changes in physical appearance or weight when boiled for 48 hours in acetone, toluene or trichloroethylene. Soaking the polymer in concentrated alcoholic potassium hydroxide solution for 2 weeks had no visible effect on the material.

#### Example 2

When the procedure of Example 1 was repeated up to the point where the reaction mixture reached the viscosity of molasses, a glass tape was dipped into the mixture and excess mixture allowed to drain off. A coated glass tape was formed by heating the tape at a temperature of 160° C. for 1½ hours. The resulting coated tape had a hard, clear, smooth coating and was useful as insulation for a dynamo electric machine.

#### Example 3

When the procedure of Example 1 was repeated except that 1,5-dimethyl-1,5-dihydro-3,3,7,7-tetraphenylcyclotetrasiloxane was substituted for the hydrogen-containing siloxane of Example 1, a product comparable to that of Example 1 was formed. This material differed, however, from the product of Example 1 in that the cyclotetrasiloxane units derived from the difunctional cyclotetrasiloxane contained a silicon-bonded methyl radical on the 1- and 5-silicon atoms and contained two silicon-bonded phenyl groups on the 3- and the 7-silicon atoms.

#### Example 4

A solution was formed from a mixture of ingredients in the ratio of two moles of 1,5-diallyl-1,3,3,5,7,7-hexamethylcyclotetrasiloxane and 1,3,5,7-tetrahydro-1,3,5,7-tetramethylcyclotetrasiloxane. To this solution was added a sufficient amount of the catalyst of Example 1 as a 5% solution of the catalyst in amyl alcohol to provide  $1 \times 10^{-5}$  gram atoms platinum per mole of the allyl radical-containing cyclotetrasiloxane. This mixture was heated at a temperature of 75° for 1 hour, during which time the solution was converted into an optically clear, hard, tough organopolysiloxane resin within the scope of the present invention. This material was substantially identical to the material described in Formula 7 except that the cyclotetrasiloxane radicals were joined by propylene (trimethylene) radicals. This organopolysiloxane resin exhibited properties substantially identical to the resin of Example 2.

While the foregoing examples have illustrated the preparation of a number of organopolysiloxane resins within the scope of the present invention, it should be understood that these resinous materials can be prepared from tetrafunctional cyclotetrasiloxanes within the scope of Formula 1 other than those specifically illustrated in

the examples. Likewise, difunctional cyclotetrasiloxanes within the scope of Formula 2 other than those specifically illustrated, can also be employed to prepare organopolysiloxane resins of the scope previously defined. In the examples, the tetrafunctional cyclotetrasiloxane resin has been illustrated by materials in which the four silicon-bonded monovalent hydrocarbon radicals were the same. It should be understood, however, that these four silicon-bonded monovalent hydrocarbon radicals free of aliphatic unsaturation can be different materials. Thus, for example, two of the monovalent hydrocarbon radicals free of olefinic unsaturation can be methyl while the other two could be phenyl. Such a material would be prepared, for example, by the cohydrolysis and cocondensation of ethylvinylchlorosilane and phenylvinylchlorosilane by methods previously described.

It should also be understood that the products of the present invention can contain various fillers such as carbon black, clay, zinc oxide, whiting slate flour, finely divided silica such as fume silica, precipitated silica, silica aerogel, etc. These fillers can be incorporated in amounts of from 10 to 200 or more parts per part of the organopolysiloxane resins of the present invention. These fillers can be incorporated into the organocyclotetrasiloxane starting materials or can be added after the reaction begins but before the reaction passes the viscous fluid stage. While the presence of fillers interferes with the optical clarity of the products of the present invention, the fillers increase the toughness of the resins.

What I claim as new and desire to secure by Letters Patent of the United States is:

1. An organopolysiloxane resin comprising a first class of organocyclotetrasiloxane units and a second class of organocyclotetrasiloxane units, there being present in said resin substantially twice the number of said second class as of said first class, with substantially every unit of said first class being attached to four units of said second class through a silicon-bonded alkylene radical of at least two carbon atoms, with substantially every unit of said second class being attached through its 1- and 5-silicon atoms, respectively, to two and only two units of said first class through said silicon-bonded alkylene radical, substantially all of the valences of silicon other than the valences satisfied by oxygen in the cyclotetrasiloxane rings and the valences satisfied by silicon-bonded alkylene radicals being satisfied by monovalent hydrocarbon radicals free of aliphatic unsaturation.

2. The organopolysiloxane resin of claim 1 in which the monovalent hydrocarbon radicals are selected from the class consisting of phenyl radicals and methyl radicals.

3. The composition of claim 1 in which the monovalent hydrocarbon radicals are methyl radicals.

4. The composition of claim 1 in which the alkylene radicals are ethylene radicals.

5. The organopolysiloxane resin of claim 1 in which the alkylene radicals are propylene radicals.

6. A clear, hard organopolysiloxane resin comprising a first class of organocyclotetrasiloxane units and a second class of organocyclotetrasiloxane units, with there being present substantially twice as many of said second class as said first class, with substantially every unit of said first class being attached to four units of said second class through an ethylene radical, with substantially every unit of said second class being attached through its 1- and 5-silicon atoms, respectively, to two and only two units of said first class through said ethylene radical, substantially all of the valences of silicon other than the valences satisfied by oxygen in the cyclotetrasiloxane rings and the valences satisfied by silicon-bonded ethylene radicals being satisfied by methyl radicals.

7. An organopolysiloxane resin comprising a first class of organocyclotetrasiloxane units and a second class of organocyclotetrasiloxane units, there being present in said resin substantially twice the number of said second class of organocyclotetrasiloxane units as of said first