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3,036,985

COMPOSITION COMPRISING A SILOXANE COPOLYMER

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This invention relates to transparent organopolysiloxane elastomers and to the fillers employed therein.

Various types of silica fillers and modified silica fillers have been employed in organopolysiloxane rubbers. The use of silica fillers has given rubbers of varying degrees of strength. In general naturally occurring silica such as diatomaceous earth or powdered quartz give rubbers of relatively low stress-strain properties, i.e., rubbers having efficiencies of less than 100. This is likewise true of unmodified silicas which are prepared by evaporating the solvent from hydrogels or organogels at a temperature below the critical temperature of the solvent. Such silicas are known in the art as xerogels.

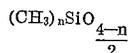
Superior stress-strain properties are generally obtained by employing extremely finely divided silicas. These are generally of three classes: silicas prepared by removing the solvent from an organogel at a temperature above the critical pressure of the solvent which are known as aerogels; silicas prepared by burning volatile silanes such as trichlorosilane, ethyl silicate, or silicon tetrachloride which are generally known as fume silicas and modified xerogels such as those disclosed in the copending application of Leslie J. Tyler, Serial No. 460,773, filed October 6, 1954. Of these latter 3 types the modified xerogels give superior elastomers with respect to stress-strain properties.

It is known that fume silicas and silica aerogels can be modified with organosilicon compounds in a variety of ways. These include treating of a fume silica with hydrolyzable silanes as is disclosed in U.S. Patent No. 2,610,167 and the modification of silica aerogels by incorporating limited amounts of monoorganosiloxane salts in a sodium silicate solution and thereafter preparing a cogel by the aerogel technique as is set forth in U.S. Patent 2,441,422. None of the above silicas whether modified or unmodified, with the exception of certain modified xerogels hereinafter defined, give transparent elastomers when incorporated in organosiloxane polymers and thereafter vulcanized.

Applicant has found most unexpectedly that the specific fillers hereinafter defined do give optically clear materials.

It is the object of this invention to produce novel compositions of matter which when incorporated in methyl polysiloxanes give elastomers having a high degree of optical clarity. It is a further object of this invention to provide elastomeric materials suitable for making optically clear safety glass. Another object is to provide elastomeric materials which possess a high degree of optical clarity without undue sacrifice of desirable stress-strain properties. Other objects and advantages will be apparent from the following description.

This invention relates to a composition of matter in a finely divided state having a pore volume of at least 3 cc. per gram, said composition being a copolymer of (A) from 1 to 50 mol percent siloxane units of the formula $RSiO_{3/2}$ in which R is an aliphatic hydrocarbon radical of less than 5 carbon atoms, (B) from 4 to 30 mol percent siloxane units of the formula



in which n is an integer from 2 to 3 inclusive and (C) from 46 to 95 mol percent SiO_2 units.

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For the purpose of this invention the beneficial properties of the compositions claimed herein are not realized when any of the above critical limitations are exceeded. Thus the combination of optical clarity and good stress-strain properties is imparted to silicone rubber by these materials only when the proportions of the various polymer units are within the ranges specified. Furthermore, this combination of properties is imparted only when the material is in a finely divided state, i.e., having pore volume of at least 3 cc. per gram. The pore volume of these materials is determined in accordance with the method described in U.S. Patent 2,541,137 except that octamethylcyclotetrasiloxane is employed in the place of water. When the compositions of this invention are solvent free they are fluffy, light powders having bulk densities of the order of .05 to 1.1 g. per cc.

In the monoorganosiloxane units (A) of the fillers of this invention R can be any aliphatic hydrocarbon radical of less than 5 carbon atoms such as methyl, ethyl, propyl, butyl, vinyl and allyl radicals. The siloxane units (B) are all methylsiloxane units in which there are 2 or 3 methyl groups attached to each silicon atom. For the purpose of this invention it is possible to use a combination of dimethyl and trimethylsiloxane units provided the total mols of the dimethyl and trimethylsiloxane units are not less than 4 mol percent nor more than 30 mol percent of the total composition.

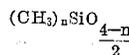
The compositions of this invention may be prepared in any suitable manner. The best method is that described in the copending application of Leslie J. Tyler aforementioned. Briefly this method comprises preparing a silica cohydrogel composed of $RSiO_{3/2}$ and SiO_2 units where R is as above defined. Such cogels may be prepared by acidifying an aqueous solution of a mixture of sodium silicate and the desired monoorganosiloxane salt, for example, $MeSiO(ONa)$. The cohydrogel may then be reacted with dimethylsiloxane or hexamethyldisiloxane or copolymers and mixtures thereof in the presence of an acid catalyst whereupon the dimethyl and/or trimethylsiloxane units copolymerize with the cogel.

A modification of this method involves replacing the water in the cohydrogel with an organic solvent such as toluene, methylene chloride and the like and then reacting the resulting organogel with dimethyldichlorosilane, trimethylmonochlorosilane or the corresponding alkoxy silanes or combinations thereof. The solvent is then evaporated to give the compositions of this invention.

This invention further relates to transparent elastomeric compositions which consist essentially of (1) an organopolysiloxane in which at least 50 mol percent of the polymer units are dimethylsiloxane units, any remaining siloxane units being of the formula



in which R' is a monovalent hydrocarbon radical of less than 10 carbon atoms and n has an average value from 1.9 to 2.1 inclusive and (2) a filler having a pore volume of at least 3 cc. per gram which is composed of a copolymer of (A) from 1 to 50 mol percent $RSiO_{3/2}$ units in which R is an aliphatic hydrocarbon radical of less than 5 carbon atoms, (B) from 4 to 30 mol percent



units in which n is an integer from 2 to 3 inclusive and (C) from 46 to 95 mol percent SiO_2 units.

The proportion of filler in the elastomers of this invention is not critical although preferably the amount of filler should range from 5 to 50 parts by weight based on 100 parts by weight of the polymer. These ranges give elastomers of superior stress-strain properties.