OPTICALLY CLEAR SILICONE COMPOSITIONS CURABLE TO ELASTOMERS

This application is a continuation-in-part of application Ser. No. 156,003, filed June 3, 1980, now abandoned.

BACKGROUND OF THE INVENTION

1. Field of the Invention

This invention relates to optically clear silicone compositions which are curable to elastomers comprising at least one polydiorganosiloxane and a hydrophobic reinforcing silica filler wherein the silica filler particles are sufficiently small in their largest dimensions that optically clear compositions result even when the refractive index of the polydiorganosiloxane is not matched with that of the filler. This invention also relates to a method of preparing such compositions. This invention also relates to the optically clear elastomers obtained when such compositions are cured.

2. Description of the Prior Art

Silicone elastomers, more commonly known as silicone rubbers, fine use in many diverse areas such as sealants, tubing, prosthetic devices, wire insulation, molding compounds, contact lenses, solvent resistant hoses and many other uses. In applications where optical clarity is a necessity, compositions consisting of polydiorganosiloxanes such as polydimethylsiloxane gums can be vulcanized or cured to form optically clear elastomeric products. However, when the cured elastomer consists solely of a cured polydiorganosiloxane, the physical properties of the elastomer are poor. To improve these properties, reinforcing fillers such as finely divided silica are added to the gum prior to curing. A surface-treated (hydrophobic) reinforcing silica filler is often employed because such a filler not only improves the physical properties of the cured elastomer as compared with curing the polydiorganosiloxane alone, but also avoids the well-known problem of crepe-aging or structuring of the uncured composition. The addition of hydrophobic reinforcing silica fillers has the disadvantage of increasing the optical haze of the compositions and cured elastomers depending upon the types and amounts of polydiorganosiloxanes, filler and other components present in the formulation.

Thus the compositions and cured elastomers are no longer as optically clear as the cured polydiorganosiloxanes themselves although they may appear transparent in thin sheets where the haze is less noticeable. Contact lenses consisting of transparent silicone rubber containing silica fillers are described in U.S. Pat. No. 3,228,741 to Becker and British Pat. No. 922,871. Transparent room-temperature vulcanizable silicone rubber stocks are described in U.S. Pat. No. 3,408,325 to Hittmair et al. and in U.S. Pat. No. 3,474,064 also to Hittmair et al. Hittmair et al. teach compositions which are composed of organopolysiloxanes which can also contain fillers. The latter '0.64 patent teaches that a cured elastomer derived from 100 parts of a hydroxyl-endblocked dimethylpolysiloxane, 10 parts of fume silica, 5 parts of a hydroxylated methylpolysiloxane fluid and a small amount of a curing agent was sufficiently transparent that print from a typewriter could readily be seen and read through a 4 millimeter (mm) sheet of the cured elastomer although nothing was mentioned regarding the haziness of the sample.

A commonly accepted explanation for the amount of haze which results when a silica filler is added to a polydiorganosiloxane which would otherwise cure to form an optically clear elastomer is that the haze appears to be related to the difference in refractive index at 25° C., Sodium D wavelength (R.I.) between the silica filler (R.I. approximately 1.42 to 1.46) and the polydiorganosiloxane. The haze is apparent to the naked eye in 2.54 mm (100 mil) thicknesses when a hydrophobic reinforcing silica filler is mixed with a polydimethylsiloxane gum (R.I. approximately 1.40) and especially when mixed with a fluorine-containing polydiorganosiloxane gum such as poly-3,3,3-trifluoropropylmethylsiloxane gum (R.I. approximately 1.38). One prior art solution to the problem of haziness involves the use of polydiorganosiloxanes containing silicon-bonded organic radicals such as phenyl radicals in sufficient amounts such that the refractive index of the polydiorganosiloxanes in the composition matches the refractive index of the silica filler, thereby obtaining optically clear compositions and cured elastomers. Examples of such an approach are found in U.S. Pat. Nos. 3,996,189 and 3,996,187, both to Travnicek, which teach optically clear silicone elastomers and contact lenses reinforced with fume silica fillers. These patents teach that 80 to 95 parts of one or two polydiorganosiloxanes containing from 6 to 16 mole percent phenyl groups can be mixed with from 5 to 20 parts of a fume silica to obtain optically clear reinforced silicone elastomers while a polydiorganosiloxane which is free of phenyl groups such as a dimethylpolysiloxane containing a small percentage of vinyl groups results in a hazy material which had inadequate optical clarity. In U.S. Pat. No. 3,624,023, Hartlage teaches compositions containing a surface-treated fume silica filler and a hydroxyl-enblocked polydiorganosiloxane containing phenylmethylpolysiloxane units which are vulcanizable to transparent silicone rubbers under ambient conditions.

In U.S. Pat. No. 4,008,198, Krohberger et al. teach that highly transparent or optically clear elastomers can be obtained by mixing (1) a nitrogen compound containing at least one triorganosilyl group of a particular type, (2) a hexaorganodisilazane, (3) silicon dioxide having a surface area of at least 50 m^2/g and (4) a highly viscous polydiorganosiloxane and then kneading the mixture at 150° C. under a vacuum until there is no evidence that nitrogen compounds are being evolved. The examples contained therein report that a composition prepared using a polydimethylsiloxane containing a small percentage of vinyl radicals possessed a light transparency value of 91% and a composition prepared using a polydimethylsiloxane containing 5.5 mole percent of diphenylsiloxane units and a larger percentage of methylvinylsiloxane units than in the previous example possessed a light transparency value of 96% which is consistent with the prior art technique of obtaining compositions possessing improved optical clarity by matching the refractive index of the filler with that of the polydiorganosiloxane. The haze value of each composition is not reported.

Iler, in U.S. Pat. No. 2,786,042, teaches the preparation of sols of surface-treated colloidal silica particles which are said to have an average diameter of from 10 to 150 millimicrons (100 to 1500 Angstroms). Iler teaches that such particles can be added to plastics in the form of an organosol to improve the physical properties of the plastics and teaches that even transparent