HEAT-CURABLE ORGANOPOLYSILOXANE COMPOSITIONS

FIELD OF THE INVENTION

This invention relates to heat-curable organopolysiloxane compositions. More particularly, the invention relates to heat-curable organopolysiloxane compositions improved in shelf life or storage stability.

DESCRIPTION OF THE PRIOR ART

Heat-curable organopolysiloxane compositions of the addition reaction type comprising a vinyl-group-containing organopolysiloxane, an organohydrogenpolysiloxane, a filler, and a platinum catalyst are well known and, since they usually tend to gradually cure during long storage at room temperature, commercially available as formulations of the so-called two-package system wherein the component in one package is mixed with the other just prior to use. It is also known that the room-temperature curing on storage of the conventional organopolysiloxane compositions can be prevented by use of an additive, such as tin compounds, phosphorus compounds, or acetylenically unsaturated hydrocarbon group-containing organic compounds, whereby the troublesome mixing of the two packages prior to use can be dispensed with. However, the use of these additives works disadvantageously to reduce the flame retardancy of the resultant elastomers.

OBJECT OF THE INVENTION

It is an object of this invention to introduce novel heat-curable organopolysiloxane compositions having improved storage stability and allowing for imparting an excellent flame retardancy to silicone elastomers made therefrom.

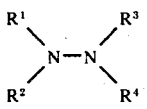
It is a further object of the invention to provide a process of preparing such compositions.

DETAILED DESCRIPTION OF THE INVENTION

The heat-curable organopolysiloxane composition introduced by the invention comprises (1) 100 parts by weight of a diorganopolysiloxane having the average formula

$$R_aSiO_{4-a/2} \qquad (I)$$

where R is a substituted or unsubstituted monovalent hydrocarbon group and a has an average value of from 1.98 to 2.01, which contains at least two vinyl groups per molecule and the viscosity of which exceeds 100 centistokes at 25° C, (2) from 10 to 200 parts by weight of an inorganic filler, (3) an organohydrogenpolysiloxane containing at least two Si—H linkages per molecule in an amount sufficient for giving the Si—H linkages from 0.5 to 15 times in number compared to the vinyl groups contained in component (1) above, (4) platinum or a platinum-containing compound in a catalytic amount, (5) a hydrazine compound having the general formula



(II)

where R^1, R^2, R^3 and R^4 each represent a hydrogen atom, phenyl group or alkyl group, provided always at least one of them is a phenyl or alkyl group, in an amount larger than 10% by weight of the amount of component (4) as platinum, and optionally (6) an organic peroxide in an amount of from about 0.01 to 5 parts by weight.

This invention is based, in part, on the discovery that a mixture of a diorganopolysiloxane having two or more vinyl groups per molecule as the main component with an organohydrogenpolysiloxane, an inorganic filler, a platinum catalyst, and a hydrazine compound is remarkedly excellent in long storage stability at room temperature and further that elastomer products produced by curing the mixture possess excellent flame retardant properties.

The diorganopolysiloxane useful as component (1) in the compositions of this invention is represented by formula I in which the monovalent hydrocarbon groups denoted by R are, illustratively, alkyl groups (for example, methyl, ethyl and propyl), halogen-substituted alkyl groups, alkenyl groups (for example, vinyl and allyl), aralkyl groups (for example benzyl), aryl groups (for example, phenyl), and halogen-substituted aryl groups; and the value of a is from 1.98 to 2.01. Further, this component (1) is required to have a viscosity exceeding 100 centistokes at 25° C, preferably exceeding 1,000 centistokes at 25° C, and contain at least two vinyl groups per molecule.

Illustrative of the diorganopolysiloxanes of the above nature are those which are terminated with the dimethylvinylsilyl, trimethylsilyl or dimethylhydroxysilyl groups and which contain methyl and other hydrocarbon groups including vinyl groups as organic groups on their main chains.

The inorganic fillers as component (2) in the composition of the invention serve to impart mechanical strengths to the finished elastomers.

The inorganic fillers are usually powdery silicious oxides, such as fume silica, precipitated silica, quartz flour and diatomaceous earth. In practice, 10 to 200 parts by weight calculated on 100 parts by weight of component (1) are used.

The organohydrogenpolysiloxanes useful as component (3) incorporated in the compositions of the invention are, necessarily, those containing at least two Si-H linkages per molecule. They can be prepared by known processes, and exemplified by methylhydrogenpolysiloxanes terminated with trimethylsilyl groups and varied in polymerization degrees, tetramethyltetrahydrogencyclotetrasiloxane, siloxane copolymers consisting of units SiO_2 and $(CH_3)_2Si(H)O_{0.5}$, and copolymers of methylhydrogensiloxane units and diorganosiloxane units.

The organohydrogenpolysiloxanes are necessarily incorporated in amounts such that the Si-H linkages can be given in a number from 0.5 to 15 times the whole number of the vinyl groups contained in component (1).

The platinum or platinum-containing compounds of component (4) are useful as the curing catalyst and for imparting flame retardancy to the resultant silicone elastomers. Illustrative of this component (4) are platinum black, chloroplatinic acid and complexes of chloroplatinic acid with ethylene or propylene. A further example is a complex formed from chloroplatinic acid with alcohols and the like as described in U.S. Pat. No. 3,220,972.