

**THERMALLY STABLE ORGANOSILOXANE
COMPOSITIONS AND METHOD FOR
PREPARING SAME**

BACKGROUND OF THE INVENTION

1. Field of the Invention

This invention relates to thermally stable organosiloxane compositions. More particularly, this invention relates to unfilled organosiloxane compositions that cure by a platinum catalyzed hydrosilation reaction to yield materials exhibiting a resistance to heat-induced discoloration during long-term exposure to temperatures of up to 200° C.

2. Description of the Prior Art

Polyorganosiloxane compositions which cure by means of a platinum catalyzed hydrosilation reaction to form silicone gels are disclosed, for example, in U.S. Pat. No. 3,020,260, issued to Nelson on Feb. 6, 1962, and in U.S. Pat. No. 4,374,967, issued to Brown, Lee and Maxson on Feb. 22, 1983. The gels described by Nelson are obtained by reacting an organosiloxane containing two silicon-bonded hydrogen atoms per molecule with a vinyl-containing copolymer wherein the repeating units are of the formulae $RViSiO$, R_2SiO , and $(CH_3)R_2SiO_{0.5}$, where R is methyl or phenyl and Vi represents vinyl.

The vinyl-containing copolymers disclosed by Brown et al. consist essentially of $(CH_3)_2SiO$, $(CH_3)_3SiO_{0.5}$ and $(CH_3)_2ViSi_{0.5}$ where Vi again represents vinyl, and are reacted with an organohydrogensiloxane having more than one silicon-bonded hydrogen atom per molecule.

U.S. Pat. No. 4,584,361, which issued to Janik and Lo on Apr. 22, 1986 teaches the use of a platinum catalyst inhibitor of the general formula $R_2^1NR^3NR_2^2$ or R^4NH_2 in an amount sufficient to impart long term storage stability at temperatures of up to 70° C. to a one-part unfilled polyorganosiloxane gel composition that cures by a platinum-catalyzed hydrosilation reaction. In these formulae each R^1 is individually selected from the group consisting of alkyl radicals containing from 1 to 4 carbon atoms, R^2 is R^1 or hydrogen, R^3 represents an alkylene radical containing from 2 to 4 carbon atoms, and R^4 represents an alkyl radical containing from 2 to 4 carbon atoms. Surprisingly the inhibitor does not substantially retard the cure rate of the composition at temperatures above about 80° C.

The use of ammonia, amines, and other nitrogen-containing compounds as inhibitors for the platinum catalyzed reaction of silicon-bonded hydrogen atoms with silicon-bonded hydroxyl groups is taught in U.S. Pat. No. 3,867,343, issued to Garden on Feb. 18, 1975. U.S. Pat. No. 4,281,093, issued to Garden on July 28, 1981, discloses using these inhibitors for the reaction of polyorganohydrogensiloxanes with polyorganosiloxanes containing silicon-bonded vinyl or allyl radicals. The catalysts for the reaction are organic complexes of platinum or rhodium. Neither metallic platinum nor reaction products of platinum compounds with organosilicon compounds are mentioned as catalysts suitable for use with these amine inhibitors.

A shortcoming of the compositions disclosed in the aforementioned Nelson and Brown et al. patents is the tendency of the initially clear and colorless cured materials prepared from these compositions to darken when exposed to temperatures from 100° to about 200° C. for extended periods of time. In addition to being aestheti-

cally unattractive the darkening may adversely affect the utility of the compositions if transparency and the absence of discoloration are requirements.

The present inventors have now discovered that the addition to curable compositions disclosed in the aforementioned Nelson and Brown et al. patents of less than 1 percent by weight of certain aliphatic and cycloaliphatic amines, including the platinum catalyst inhibitors of the aforementioned U.S. Pat. No. 3,867,343 to Garden and the relatively small group of amines disclosed as platinum catalyst inhibitors for the compositions claimed in the aforementioned Janik and Lo patent, imparts a resistance to heat-induced discoloration to these compositions in cured form. This effect is not observed when aliphatic and cycloaliphatic monoamines are added to curable organosiloxane compositions containing a vinyl-substituted polydiorganosiloxane in place of the vinyl-containing polyorganosiloxanes disclosed in the aforementioned Nelson and Brown et al. patents.

SUMMARY OF THE INVENTION

The present invention is based on the discovery that small amounts of aliphatic or cycloaliphatic amines are unique with respect to their ability to impart heat stability to cured materials prepared using the curable organosiloxane compositions disclosed in the aforementioned Brown et al. patent.

**DETAILED DESCRIPTION OF THE
INVENTION**

This invention provides a method for imparting resistance to heat-induced discoloration to a cured unfilled organosiloxane material prepared from a curable composition comprising the product obtained by blending to homogeneity

(A) at least one organosiloxane copolymer consisting essentially of from 80 to 96.5 mol percent of $(CH_3)_2SiO$ units, from 2.0 to 10.0 mol percent of $(CH_3)_3SiO_{1.5}$ units, from 1.25 to 6.0 mol percent of $(CH_3)_3SiO_{0.5}$ units and from 0.25 to 4.0 mol percent of $(CH_3)_2(CH_2=CH)SiO_{0.5}$ units;

(B) an organohydrogensiloxane containing at least two silicon-bonded hydrogen atoms per molecule, no more than one silicon-bonded hydrogen per silicon atom and organic radicals selected from the group consisting of alkyl containing from 1 to 6 carbon atoms per radical, phenyl and 3,3,3-trifluoropropyl, said organohydrogensiloxane providing from 0.7 to 1.2 silicon-bonded hydrogen atoms per silicon-bonded vinyl radical present in said composition; and

(C) a platinum catalyst in an amount sufficient to promote the reaction of (A) and (B), where said platinum catalyst is metallic platinum or a reaction product of a platinum compound and an organosilicon compound; said method comprising adding to said curable composition at least one amine of the formula $R_nNH_{(3-n)}$, $H_2NR''NH_2$, $R'_2NR''NR'_2$ or $H_2NR''N(H)R''NH_2$ in an amount sufficient to inhibit heat-induced discoloration of said cured organosiloxane material at temperatures of up to about 200° C., where each R is individually selected from alkyl radicals and cycloalkyl radicals, R' represents an alkyl radical, R'' represents alkylene and the value of n is 1, 2 or 3.

This invention also relates to curable compositions containing the aforementioned ingredients A, B, C and an amine corresponding to the formula R_mNH_{3-m} ,