

## OPTICALLY CLEAR REINFORCED ORGANOSILOXANE COMPOSITIONS

### BACKGROUND OF THE INVENTION

#### 1. Field of the Invention

This invention relates to organosiloxane compositions that cure by a platinum-catalyzed hydrosilation reaction to form optically clear elastomers. More particularly, this invention relates to curable organosiloxane compositions that are reinforced using a specified class of organosiloxane copolymers and exhibit an unexpected decrease in viscosity with increasing concentration of these copolymers.

#### 2. Description of the Prior Art

It is known to improve the tensile properties of organosiloxane elastomers, particularly tensile strength and tear strength, by including in the curable compositions used to prepare these elastomers a finely divided reinforcing filler such as fume or precipitated silica.

Another method for improving the tensile properties of cured organosiloxane elastomers prepared from compositions that cure by a platinum-catalyzed hydrosilation reaction is to include in the curable composition a resinous organosiloxane copolymer containing SiO<sub>2</sub> units and vinyl-containing siloxane units.

U.S. Pat. No. 3,436,366, which issued to Modic on Apr. 1, 1969 discloses organosiloxane compositions yielding cured elastomers exhibiting tear strengths greater than 60 pounds per linear inch (10.5 kilonewtons per meter). The compositions contain a polydiorganosiloxane having vinyl radicals at the terminal positions, an organosiloxane copolymer with SiO<sub>2</sub>, triorganosiloxane and, optionally, diorganosiloxane units, an organohydrogensiloxane in an amount sufficient to cure the composition, a platinum-containing hydrosilation catalyst in an amount sufficient to promote curing of the composition and an optional filler. Either or both of the diorganosiloxane and triorganosiloxane units comprising the organosiloxane copolymer contain a vinyl radical.

One of the two exemplified organosiloxane copolymers in the aforementioned patent to Modic contains SiO<sub>2</sub>, trimethylsiloxy and methylvinylsiloxy units. Elastomers prepared by curing compositions containing this copolymer, a dimethylvinylsiloxy-terminated polydimethylsiloxane having a viscosity of 3.3, 80 or 550 Pa.s at 25° C. and no reinforcing filler exhibited tear strengths of 22, 64 and 80 ppi, respectively. Example 15 of this patent discloses a composition containing a diphenylvinylsiloxy-terminated dimethylsiloxane/diphenylsiloxane copolymer having 3 diphenylsiloxane units per 100 dimethylsiloxane units and exhibiting a viscosity of 150 Pa.s at 25° C. In this instance the aforementioned organosiloxane copolymer contained SiO<sub>2</sub>, dimethylvinylsiloxane, methylvinylsiloxane and trimethylsiloxy units, and the reported tear strength of a cured elastomer prepared by curing the composition was "in excess of 60 p.p.i.", however no absolute value is reported.

U.S. Pat. No. 4,500,584, which issued to Modic on Feb. 19, 1985 discloses dirt-resistant silicone coating compositions curable by a platinum-catalyzed hydrosilation reaction. The compositions comprise a vinyl-terminated polydiorganosiloxane having a viscosity of up to 2,000 Pa.s wherein at least 50 percent of the silicon-bonded hydrocarbon radicals are methyl and a vinyl-containing organosiloxane copolymer consisting essentially of triorganosiloxane and optionally diorganosilox-

ane units in combination with SiO<sub>2</sub> units. All of the exemplified copolymers contain only trimethylsiloxy, methylvinylsiloxy and SiO<sub>2</sub> units.

U.S. Pat. No. 3,284,406, which issued to Nelson on Nov. 8, 1966 discloses compositions consisting essentially of (1) a vinyl terminated polydiorganosiloxane having a viscosity of from 500 to 500,000 centistokes wherein at least 80 percent of the silicon-bonded hydrocarbon radicals other than vinyl are methyl and any remainder are phenyl, (2) an organosiloxane copolymer containing specified proportions of trimethylsiloxy, dimethylsiloxy and SiO<sub>2</sub> units, (3) an organohydrogensiloxane curing agent and (3) a platinum-containing hydrosilation catalyst. The two exemplified polydiorganosiloxanes have viscosities of 2,000 centistokes and 9,000 centistokes.

The organosiloxane copolymers disclosed in the aforementioned patents to Modic and Nelson are typically solid, resinous materials under ambient conditions, and their presence in curable organosiloxane compositions would be expected to increase the viscosity of these compositions relative to the viscosity of the unmodified polydiorganosiloxane. This effect is observed with both of the polydiorganosiloxanes exemplified in the aforementioned Nelson patent.

An objective of this invention is to provide curable organosiloxane compositions yielding optically clear elastomers that exhibit substantially superior tensile properties relative to elastomers prepared from prior art curable compositions of comparable viscosity.

### SUMMARY OF THE INVENTION

The present inventor discovered that the presence of up to about 40 percent by weight of a resinous organosiloxane copolymer consisting essentially of trimethylsiloxy, dimethylvinylsiloxy and SiO<sub>2</sub> units in a diorganovinylsiloxy-terminated polydiorganosiloxane containing at least 95 mol percent of dimethylsiloxane units and having a viscosity greater than about 12 Pa.s at 25° C. unexpectedly decreases the viscosity of the mixture relative to the viscosity of the polydiorganosiloxane. Cured elastomer prepared from curable compositions containing these mixtures exhibit better tensile properties relative to elastomers prepared from prior art curable compositions of comparable viscosity. Some of these prior art compositions comprise a polydiorganosiloxane in combination with vinyl-containing organosiloxane copolymers other than those of the present invention as the reinforcing agent.

### DETAILED DESCRIPTION OF THE INVENTION

This invention provides an improved curable composition comprising

1. a diorganovinylsiloxy-terminated polydiorganosiloxane,
2. an amount of a resinous organosiloxane copolymer sufficient to improve the physical properties of the elastomer obtained by curing said composition,
3. an amount of an organohydrogensiloxane sufficient to cure said composition, and
4. an amount of a platinum-containing hydrosilation catalyst sufficient to promote curing of said composition,

where the improvement comprises (a) selecting said polydiorganosiloxane from those exhibiting a viscosity of at least 12 Pa.s at 25° C. and wherein at least 95% of