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LOW TEMPERATURE VULCANIZING COMPOSITION AND ARTICLE MADE THEREFROM

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No Drawing. Filed Dec. 15, 1965, Ser. No. 514,140
Int. Cl. C08g 47/04, 47/10

U.S. Cl. 260—37

6 Claims

ABSTRACT OF THE DISCLOSURE

Compositions which cure to transparent silica reinforced potting and molding materials comprise (1) a silanol-terminated polydiorganosiloxane in which from 11% to 13.3% of the organo groups are phenyl, with substantially all of the remaining groups being methyl, (2) a resinous copolymer of trimethylsiloxane units and SiO₂ units, and (3) a reinforcing silica filler.

This invention relates to a new low temperature curing composition and to an article made from that composition. More particularly, this invention relates to a two-component low temperature vulcanizing organopolysiloxane composition which contains reinforcing silica fillers for forming a high strength article, such as a clear, "see-through" mold, and to that clear, transparent article. The composition of matter consists essentially of (1) a resinous copolymer of R₃SiO_{0.5} units and SiO₂ units, (2) a silanol-terminated diorganopolysiloxane fluid, and (3) a reinforcing silica filler. The composition is catalyzed by a curing agent for (1) and (2).

So-called "room temperature vulcanizing" compositions are well known in the art and generally comprise, as a base material, a silanol-terminated diorganopolysiloxane material. When it is desired to form an organopolysiloxane material with a thick cross section that can be cured at low temperatures, for example at or near room temperature, it is desirable that a two-component system be utilized, rather than the one-component system where thick section cures are achieved only with greater difficulty. On the other hand, most two-component low temperature vulcanizing organopolysiloxane systems utilize ethyl silicate as the cross-linking agent. When a reinforcing silica filler is blended into a composition containing a silanol-terminated diorganopolysiloxane and ethyl silicate, a gel is formed and the mixture may solidify, while standing at room temperature for a very short period of time, even prior to the addition of a curing agent. Such a system is obviously unsuitable for commercial application. Due to the increased strength of polymeric systems containing reinforcing silica fillers and to the reduced cost which is inherent in replacing a portion of the organosiloxane polymer with such a silica filler to achieve the same volume, a means of combining a reinforcing silica filler with a two-component low temperature vulcanizable organopolysiloxane system has been sought.

In accordance with the present invention, I have discovered that when a resinous copolymer consisting of R₃SiO_{0.5} units and SiO₂ units is used as a cross-linking agent for the silanol-terminated diorganopolysiloxane fluid, a quantity of reinforcing silica filler can be blended into the mixture without any significant effect upon the fluidity of the mixture prior to the addition of catalyst and application of the material for the desired use. Thus, by using this resinous copolymer of R₃SiO_{0.5} units and SiO₂ units as the cross-linking agent in a two-component low temperature vulcanizing organopolysiloxane system, in place of the formerly used ethyl silicate cross-linking agent, the previously described benefit of including a reinforcing silica filler can be realized.

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Additionally, the silanol-terminated diorganopolysiloxane fluid can be formulated with a refractive index such that, when the reinforcing silica filler and the sinuous copolymer of R₃SiO_{0.5} units and SiO₂ units are blended in, and the mixture cured, a potting or molding resin which is clear, transparent, and strong results. Thus, this invention also relates to the clear, transparent, strong material formed from curing the composition described above when the refractive index of the silanol-terminated diorganopolysiloxane fluid is in a particular range.

Resinous copolymers of R₃SiO_{0.5} units and SiO₂ units are well known in the art, being described for example in U.S. Patent 2,857,356 of John T. Goodwin, Jr. The resinous copolymer is the cohydrolysis product of a trialkylhydrolyzable silane and an alkyl silicate, either in the monomeric or polymeric state, the cohydrolysis product containing a plurality of silicon-bonded hydroxy groups. The trialkylhydrolyzable silane used in the preparation of the resinous copolymer is one which corresponds to the general formula:



where R is a lower alkyl radical, e.g., methyl, ethyl, propyl, isopropyl, butyl, isobutyl, etc., and X is a hydrolyzable group, e.g., halogen, such as chlorine, bromine, fluorine, etc., an alkoxy radical, such as, methoxy, ethoxy, etc., or an acyloxy radical, etc. It is essential that R be a lower alkyl radical since higher alkyl radicals undesirably slow down the hydrolysis of the R₃SiX compound and cause a different type of intercondensation with the alkyl silicate, thus leaving two products which are not as desirable for the present application. Obviously, R may be the same or different lower alkyl radicals.

The alkyl silicate employed for cohydrolysis with the trialkylhydrolyzable silane is one which corresponds to the general formula:



or a polyalkyl liquid silicate obtained by hydrolyzing the monomeric silicate to a stage where it is still liquid and preferably has a viscosity, for ease of handling, below about 0.1 × 10⁶ centipoises. R in Formula 2 is the same as R described for the trialkylhydrolyzable silane and again can be the same or different lower alkyl radicals.

The method of formation of the resinous copolymer of R₃SiO_{0.5} units and SiO₂ units is completely described in the aforementioned Goodwin patent and that portion of the Goodwin patent is herein incorporated by reference. The ratio of the trialkylhydrolyzable silane and the alkyl silicate employed in forming the resinous copolymer is the same as the finally desired ratio of R₃SiO_{0.5} units to SiO₂ units in the resinous copolymer. For the present invention, the ratio of R₃SiO_{0.5} units to SiO₂ units should be in the range of from about 0.5:1 to 1:1, and preferably, the ratio is in the range of from about 0.525:1 to 0.775:1.

The silanol-terminated diorganopolysiloxane which is employed in the composition of the present invention has the formula:



where R' is a monovalent hydrocarbon radical and n is an integral number of from about 250 to 1500. Preferably, n is an integral number of from about 250 to 1000. Among the monovalent hydrocarbon radicals which R' represents are alkyl radicals, such as methyl, ethyl, propyl, butyl, octyl, etc.; cycloalkyl radicals, such as, cyclopentyl, cyclohexyl, cycloheptyl, etc.; aryl radicals, such as, phenyl, tolyl, xylyl, ethylphenyl, naphthyl, biphenyl, etc. These silanol-terminated diorganopolysiloxanes are, as previ-