

SILICONE SELF-ADHESIVE COMPOSITION

This invention relates to a silicone self-adhesive composition having improved self-adhesive properties, curing to heat resistant layers and thus finding use in the manufacture of heat resistant adhesive tapes and labels.

BACKGROUND OF THE INVENTION

Among prior art well-known silicone self-adhesive compositions is a composition comprising a condensate between a polysiloxane having $(\text{CH}_3)_2\text{SiO}_2$ and SiO_2 units and a dimethyl-silicone raw rubber. This self-adhesive composition is crosslinked with peroxides for the purpose of enhancing adhesive force and cohesive force whereas the curing temperature is as high as 180° to 200° C. There is a need for low-temperature curable silicone self-adhesive compositions from the standpoints of energy cost, productivity and application to substrates having a low heat distortion temperature.

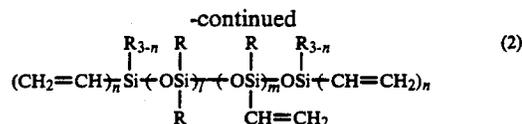
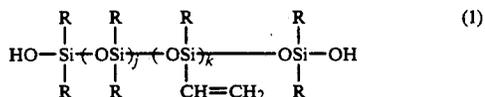
Japanese Patent Publication No. 37907/1979 proposes a low-temperature curable silicone self-adhesive composition utilizing addition reaction between an alkenyl group-containing organopolysiloxane having a viscosity of 200 to 1,000 poise and a SiH-containing organopolysiloxane. This self-adhesive composition, however, is difficult to form a layer having good physical properties and consistent adhesive force on substrates. To obviate these drawbacks, Japanese Patent Application Kokai No. 22886/1088 proposes a silicone self-adhesive composition utilizing addition reaction between an alkenyl group-containing organopolysiloxane having a viscosity of at least 500,000 centipoise (raw rubber) and an SiH-containing organopolysiloxane. The composition is alleged to show satisfactory, consistent adhesive force. However, when exposed to elevated temperatures beyond 200° C. for an extended time, this self-adhesive composition gives rise to the problems of foaming from the raw rubber triggered by cracking, a limited adhesive area, and a noticeable loss of adhesive force, with the additional drawback of a product value lowering due to deteriorated appearance. The composition is not suitable for heat resistant adhesive tapes, labels and other applications where it should withstand elevated temperatures for a long time.

Therefore, an object of the present invention is to provide a silicone self-adhesive composition which not only meets the general demand for brief curing at low temperatures, but also forms a heat resistant silicone adhesive layer which has excellent adhesive properties and undergoes no foaming even after long-term exposure to elevated temperatures.

SUMMARY OF THE INVENTION

According to the present invention, there is provided a silicone self-adhesive composition comprising

(A) at least one organopolysiloxane selected from organopolysiloxanes of the following general formulae (1) and (2):



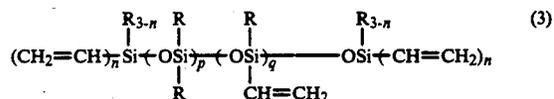
wherein R is a monovalent hydrocarbon group excluding alkenyl, letter n is an integer of from 0 to 3, k and m each are 0 or a positive integer, with the proviso that m is an integer of at least 2 when n is 0, and j and l each are an integer of at least 2,000,

(B) an organopolysiloxane comprising R_3^1SiO_2 and SiO_2 units wherein R^1 is a monovalent hydrocarbon group or a hydroxyl group, the molar ratio of R_3^1SiO_2 unit to SiO_2 unit being in the range of from 0.5 to 1.2,

(C) an organohydrogenopolysiloxane containing at least two hydrogen atoms attached to silicon atoms in a molecule, and

(D) a platinum catalyst.

The composition further includes (E) an organopolysiloxane of the following general formula (3):



wherein R and n are as defined above, p is an integer of from 0 to 1,000, q is an integer of from 10 to 1,000, and $0 \leq p/q \leq 1$.

In general, the use of a high viscosity raw rubber like component (A) in a silicone self-adhesive composition is, on one hand, effective for stabilizing and improving the adhesive force thereof. On the other hand, exposure to 200° C. of higher temperatures for a long time causes the raw rubber to be thermally cracked into dimethyl cyclics which escape in gas form from the adhesive layer which is, in turn, foamed or expanded, resulting in a noticeable drop of adhesive force. Addition of component (E), high vinyl content organopolysiloxane is effective for preventing cracking of the silicone raw rubber upon heating, so that the composition may cure and crosslink to provide an appropriate close polymer network without foaming.

Since component (E), high vinyl content organopolysiloxane participates in crosslinking, it is possible to use a dimethylsilicone raw rubber terminated with silanol at either end as component (A) rather than a vinyl-containing raw rubber. Then if a vinyl-containing raw rubber base system is relatively short of adhesive force, the system can be improved in adhesive force by adding thereto a dimethylsilicone raw rubber terminated with silanol at either end. The silicone self-adhesive composition of the invention can be cured at low temperatures because it is of the addition curing type.

The silicone self-adhesive composition of the invention cures at low temperatures of about 100° to 150° C. into a silicone adhesive layer which has improved adhesive properties including adhesive force, tack and cohesive force and undergoes little or no foaming and little or no loss of adhesive force even when exposed to 250° C. or higher temperatures. Therefore, the composition is well suited for such applications as heat resistant adhesive tapes and labels.