Research Article

Effect of the addition of selected silicon fillers on Si-PSA shrinkage

Abstract

The concept of shrinkage phenomenon is widely described in the available literature. With respect to pressure-sensitive adhesives (PSA) in general, the definition of shrinkage is understood to be "less than its original size" and is closely related to the crosslinking process and the effect of the crosslinker on the test adhesive. Shrinkage alongside adhesive properties (adhesion, tackiness) and mechanical (cohesion) is one of the most important characteristics of a self-adhesive adhesive. It is very important in terms of production when receiving, for example, decorative banners or self-adhesive films where crosslinked adhesive and thus shrinkage can affect the surface of the adhesive material and create deformations. In the case of PSA, the acceptable adhesive pressure shrinkage must not exceed 0.5 %. Contraction is an important criterion for assessing the aging resistance of PSA materials. There are no studies on the shrinkage of silicone pressure-sensitive adhesives in literature, but many references to carbon-based adhesives have been reported.

Keywords: silicone pressure-sensitive adhesives, shrinkage, adhesion, cohesion

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Introduction

Pressure-sensitive adhesives (PSA) are materials defined as a special group of adhesives that exhibit significant adhesive forces and stickiness as a result of contact with the substrate at room temperature without the need for a chemical reaction. PSA, applied as a polymer layer on a flexible carrier (fabric, foil, paper), show a very long gluing life. They are applied in liquid form by rollers onto the substrate and subjected to evaporation of the solvent in the drying channel. This leads to the hardening of the polymer layer within a few seconds. Pressure-sensitive adhesives are used to produce a variety of materials, such as mounting tapes, labels, protective films, masking tapes, advertising banners, as well as for the production of a wide range of self-adhesive medical products in the form of patches, bandages, surgical tapes and biomedical electrodes. Adhesive tapes are used to bond various materials together, such as metal, paper, plastics, glass, wood and leather. They are characterized by a constant level of peel and stick strength, as well as excellent aging resistance, both at room temperature and elevated temperature. High-quality self-adhesive adhesives are highly resistant to light, oxygen and moisture.[1-3]

Silicone pressure-sensitive adhesives were initially developed on the basis of solvent adhesives. Dexter's 1956 patent on adhesives describes them as a mixture of silicone resin and silicone polymer obtained by removal of a solvent with a metal oxide or titanium catalyst. These adhesives are used in the production of insulating tapes used in extremely high temperatures. Silicone pressure-sensitive adhesives (Si-PSA) are characterized by high elasticity, flexibility and hydrophobicity (impermeable to water). They show a low glass transition temperature and at room temperature they keep a stable elastic consistency. The low surface tension of self-adhesive silicone adhesives allows them to be easily spread over a variety of materials. They have been used in the production of patches and other medical devices (they show no negative effect on human skin). Si-PSA exhibits excellent ion barrier properties. Silicone pressure sensitive adhesives are produced by controlled polycondensation. The silicone monomers polymerize to form silicone polymers that exhibit adhesive and cohesive properties. Silicone pressure sensitive adhesives typically consist of heavy molecules of silane-functional silicone polymers and silane-functional MQ silicone resins, and may also contain vinyl-functional polymers (although vinyl-functional polymers are used as PSA cured additive). The dimethyl groups around the Si-O-Si bonding of polysiloxanes are responsible for the surface tension properties and the ability of silicone PSAs to moisturize and bond to low surface energy substrates such as Teflon® and Kapton®. MQ silicone resin provides traction and better stability at high temperature.[1-6]

Shrinkage is next to adhesive (tack, adhesion) and mechanical properties (cohesion) important parameter useful for characterizing the nature of pressure-sensitive adhesives. Shrinkage is the most important property, especially when using adhesive for production of films used in the manufacture of decorative banners and other adhesives product where dimensions are necessary. It depends among others on the type of monomers used, the polymer molecular weight and methods or compounds of crosslinking used in the technology of self-adhesive materials. The explanation of shrinkage phenomenon is not widely described in the available literature (including pressure-sensitive adhesives). In the case of articles on PSA's are comes to generally understood definition shrinkage like "becomes smaller than its original size" (not described in the article) and effect of tested agent on adhesive. In general, it is presented as a percentage or millimeter change of PVC or PET film coated with pressure-sensitive adhesives dimensions after crosslinking and fixed to a steel sheet. Typically, the contraction test is performed by the cross-method. The PVC/PET film is applied to the degreased steel plate and two cuts are made at right angles. The cut width is then measured immediately after cutting and after a given sample residence time at 70°C (usually 4/8 weeks shrinkage higher than 0.5 % or than 0.5 mm exceeds the shrinkage that is acceptable in self-adhesive technology.[7-11]

In this paper influence of selected silicon filler (kaolin, montmorilonite and silca) on shrinkage properties of silicone pressure-sensitive adhesives were presented. The adhesive tapes with varying of filler were obtained for the tests. The materials thus obtained can be used, e.g. in heat engineering and fireplace installations.

Materials and methods

Materials

In this study, a commercial silicone adhesive was used (resin content 50 % wt.), a product of Dow Corning (USA). Dichlorobenzoyl peroxide (DClBPO) supplied by Peroxid-Chemie (Germany) was used as the crosslinking agent. Bentonite from Zębiec (Poland) was used as a natural material to obtain montmorillonite. Kaolin and silica was used as fillers, which was a product of BASF (Germany).

Preparation of Si-PSA

To study the effect of silicon filler addition on shrinkage properties of the pilot silicone composition, a base resin and the crosslinking agent (1.5 wt. % 2.4-dichlorobenzoyl (DClBPO) peroxide according to polymer content) were mixed to obtain a homogenous organic composition containing 50 wt. % of polymer. The composition thus obtained was mixed with 1 % wt. silicon filler (calculated on the dry weight of the resin). The mixture prepared in this way was left for 24 hours for degassing, after which it was coated on a polyester film with a thickness of 36 μm using an automatic coating machine developed in the Laboratory of Adhesives and Self-Adhesive Materials of the West Pomeranian University of Technology in Szczecin in order to obtain the same thickness of the adhesive layer over the entire surface. The coated film with the adhesive composition was crosslinked for 10 min in a drying channel at 110°C, and then the obtained adhesive layer was protected with polyester film.

Methods

Measurements of adhesion, cohesion, tack and shrinkage were carried out according to international standards (Association des Fabricants Europeens de Rybans Auto-Adhesifs and Fédération Internationale des Fabricants et Transformateurs d'adhesifs et thermocollants sur papiers et autres support) respectively AFERA 4001, FTM 8, AFERA 4015 and cross method developed at the German company BASF.

Results and discussion

The obtained compositions were characterized by relatively high adhesion and tackiness (above 10 N/2.5 cm and 8 N). The addition of silicone fillers caused a decrease in their value as compared to the sample without fillers (Table 1).

Table 1 Impact of the additive selected silicon filler on adhesion and tack of Si- PSA

Selected filler in composition	Tack [N]	Adhesion [N/25mm]
-	12.5	13
montmorilonite	8.1	8.6
kaolin	11	12.2
silica	6.7	8.2

It is a common phenomenon observed with this type of modification, known from the literature. Moreover, the adhesive films had very high cohesion both at room temperature and elevated temperature (Table 2); both compositions without and with silicon fillers. Particularly noteworthy is the high increase in thermal resistance expressed as time (h) and the change in temperature (ºC) between the initial load on the sample until cohesion/adhesion failure. The lowest increase in resistance was recorded for kaolin, where from 133ºC the resistance increased to 147ºC. The same measurement for silica and montmorillonite showed 210ºC and 242ºC, respectively. These values are very high for self-adhesive materials, even for Si-PSA because the test was performed under load (conditions sample preparation as for cohesion tests).

Table 2 Impact of the additive selected silicon filler on cohesion of Si-PSA at 20°C and 70°C, expressed as time (h) and the change in temperature (°C) between the initial load on the sample until cohesion/adhesion failure

Selected filler in composition	Cohesion in 20°C [h]	in 70°C [h]	[°C]
-	> 72	> 72	133
montmorilonite	> 72	> 72	242
kaolin	> 72	> 72	147
silica	> 72	> 72	210

In all tested cases, the contraction increased up to about 4 days after the start of the study and then stabilized (Table 3). The compositions containing silicone fillers showed shrinkage of less than 0.5%, which is below the acceptable limit for pressure-sensitive materials for industrial application. The shrinkage values for these tapes oscillated around 0.25%.

Table 3 Impact of the additive selected silicon filler on shrinkage of Si-PSA in time

Selected filler in composition	Shrinkage [%]									
	10 min	30 min	3 h	8 h	24 h	2 days	3 days	4 days	5 days	6 days	7 days
-	0.1	0.25	0.3	0.4	0.4	0.45	0.5	0.6	0.7	0.7	0.7
montmorilonite	0	0.05	0.05	0.05	0.1	0.15	0.15	0.2	0.2	0.2	0.2
kaolin	0	0	0	0	0.05	0.1	0.2	0.25	0.3	0.3	0.3
silica	0	0	0.05	0.1	0.15	0.2	0.2	0.2	0.2	0.2	0.2

Conclusion

The tested composition based on a commercial silicone pressure-sensitive adhesive was characterized by very good adhesion, cohesion and tack, as well as high cohesion at room temperature and elevated temperature; and high shrinkage. The addition of selected fillers, such as montmorillonite, kaolin or silinonite, decreased the tack and adhesion values while increasing the cohesion and increasing the thermal resistance of the tested samples. There has been a significant decrease in the shrinkage value. The obtained compositions were characterized by relatively high thermal resistance, so they can be used in heat engineering and fireplace installations. The conducted research confirmed the high quality of commercial silicone pressure-sensitive adhesives.

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Conflicts of interest

The author declares there is no conflict of interest.

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