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Effect of Silica Filler on Viscosity, Peel Strength, Shear Strength and Tack of Styrene-Butadiene Rubber-Based Adhesive

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ABSTRACT

Viscosity, peel strength, shear strength and tack of styrene-butadiene rubber (SBR)-based pressure-sensitive adhesive were studied using silica as the filler. The silica content was varied from 10-50 parts per hundred parts of rubber (phr). The tackifying resin, solvent and coating substrate used were gum rosin, toluene and poly(ethylene terephthalate) (PET) respectively. Viscosity of the adhesive was determined by a Brookfield Viscometer. Both peel strength, shear strength and tack were measured by a Lloyd Adhesion Tester operating at 10-60 cm/min. Results show that viscosity increases with silica loading due to the concentration effect. Peel strength, shear strength and tack passes through a maximum value at 20 phr silica loading, an observation which is attributed to the culmination of wettability at the optimum silica content. Peel strength decreases with the angle of testing at a fixed silica loading and coating thickness. Both peel strength, shear strength and tack increases with coating thickness and testing rates.

Keywords: Adhesion, adhesive, peel, shear, tack, rate of testing

INTRODUCTION

Several investigations have been carried out to study the effect of fillers on adhesion properties of adhesives. These include the study of the cohesive strength of silicone rubber adhesive containing an organic montmorillonite (Wang et al., 2006), surface modifications and adhesion of SBS rubber containing calcium carbonate filler by treatment with UV radiation (Romero-Sanchez et al., 2007). The effect of nanosilica and precipitated calcium carbonate on the adhesion properties of thermoplastic polyurethane adhesives were reported (Bahattab et al., 2011; Donate-Robles & Martin-Martinez, 2011). The addition of nanosilica increased the surface energy of the polyurethane as indicated by the moderate increase in the single lap shear strength of stainless steel/ polyurethane adhesive joints. It was also observed that 10 wt % of precipitated calcium

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carbonate filler produced the highest final adhesive strength. The adhesion properties of filled epoxy resin adhesives have also been studied by other researchers. Wolf et al. (2012) found that with the addition of less than 1% wt of carbon nanotube/protein moiety filler to epoxy adhesive, peel strength and shear strength were improved by 50% and 24%, respectively. Ghosh et al. (2013) reported that thermal stability and single lap shear joint strength of filled epoxy adhesive showed significant improvement with 10 wt % loading of Al_2O_3, Al and Cu particles compared to neat epoxy adhesive. On the other hand, Ho et al. (2013) observed that silane coupling agent-treated Ag flakes have a significant effect on the electrical resistivity and shear strength in phenolic-based Ag-filled conductive adhesive. Recently, we have investigated the effect of zinc oxide and kaolin on the shear property of natural rubber adhesives (Poh & Ng, 2013; Poh & Sulaiman, 2015). For both fillers used, viscosity increases with filler loading. However, shear strength of adhesive increases up to 20 phr of filler loading after which it decreases with further addition of filler. This observation is attributed to the culmination of cohesive strength at the optimum filler loading for both filler systems. For a fixed filler loading, shear strength increases with coating thickness and testing rate. In view of the scarcity of research carried out on filled rubber adhesives, it is thus the aim of this article to report our findings on the adhesion properties of styrene-butadiene rubber (SBR)-based adhesive using silica as the filler.

MATERIALS AND METHODS

Styrene-butadiene rubber (SBR) with a 33.5% by weight of target-bound styrene and Mooney viscosity of 50 was used as the elastomer. It was supplied by Bayer Company (Penang, Malaysia). Silica with a surface area of 50 m^2/g and specific gravity of 2.0 was selected as the filler. Gum rosin, toluene and poly(ethylene terephthalate) (PET) were used as the tackifier, solvent and coating substrate respectively in this study. All the materials were freshly supplied commercial grades and purification was not carried out prior to use.

Preparation of Adhesive

SBR was masticated on a two roll-mill for 10 minutes. An amount of 5 g of the masticated rubber was cut into small pieces to facilitate easy dissolution in 30 mL of toluene. The rubber solution was tightly closed and kept overnight at 30° C until complete dissolution of SBR occurred. This was followed by the addition of 2 g, corresponding to 40 phr of tackifier, of pulverised gum rosin into the rubber solution with constant stirring. The rubber adhesive thus produced was kept for 2 hours before the addition of silica. In this study, five different loadings of silica i.e. 0.5, 1.0, 1.5, 2.0 and 2.5 g corresponding to 10, 20, 30, 40 and 50 phr of filler were used. One control sample without silica loading was also prepared for comparison purposes.

Measurements

Viscosity. A Brookfield viscometer (model DV-II + Pro) (Middleboro, MA, USA) fitted with spindle (CPE-51) and metal cup (CPE-44Y) was used to determine the viscosity of the silica-filled SBR adhesive. The testing speed was set at 1 rpm. Both the platform and spindle head were cleaned with isopropyl alcohol to eliminate any trace of contamination. A drop of adhesive

was then put in the middle of the platform and measurement was carried out for one minute. The average viscosity was computed from at least five readings displayed.

Peel strength. Three peel modes i.e. T-, 90º and 180º peel were used in the determination of peel strength using PET film as the coating substrate. Table 1 shows the respective dimensions of the peel testing samples. The effect of angle on the peel strength was also investigated using PET substrate having the same dimension as that of a 90º sample. For all the modes of peel testing, the end of a base stock was coated using a SHEEN hand coater (Teddington, Middlesex, UK) at a coating area of 10 cm × 4 cm at 60 µm and 120 µm coating thicknesses. The face stock was then gently laid on the coated base stock to form the test specimen, which was then conditioned at 30° C for 24 hours. A Lloyd Adhesion Tester (Hampshire, UK) operating at 10-60 cm min^{-1} was used to determine the peel strength of the samples. The average peel force was calculated from the three highest peaks recorded from the load-propagation plot. Peel strength is expressed as the average load per width of the bond line required to separate progressively a flexible member from a rigid member or another flexible member (ASTM D 907).

Table 1
Dimensions of peel test sample

Mode of Peel Test	Base Stock	Face Stock
T- Peel	20 cm x 4 cm	20 cm x 4 cm
90° Peel	20 cm x 4 cm	15 cm x 7 cm
180° Peel	25 cm x 4 cm	10 cm x 10 cm

Shear strength. PET film with dimension of 20 cm x 4 cm was used as the substrate. A SHEEN hand coater was used to coat the base stock from the end of the film at a coated area of 10 cm x 4 cm at 60 µm and 120 µm coating thicknesses. One end of the uncoated PET film (face stock) was then carefully placed on the coated area of the base stock so that the testing distance was 10 cm corresponding to the coated length. The test sample was conditioned at 30° C for 24 hours before testing on a Lloyd Adhesion Tester operating at 10-60 cm min^{-1}. Shear strength was defined as the shear force per unit area of testing (N m^{-2}).

Loop tack. A PET film with dimensions of 4 cm × 25 cm was used as the substrate. It was coated at the centre with a coated area of 4 cm × 4 cm using a SHEEN hand coater at 60 µm and 120 µm coating thicknesses. The coated sample was then conditioned at 30° C for 24 hours. This was followed by the formation of a loop and the outer coated surface was gently brought into contact with a glass plate. A Lloyd Adhesion Tester operating at 10-60 cm min^{-1} was used to determine the debonding force to detach the loop from the glass plate. The loop tack was expressed as the average debonding force per area of contact (N m^{-2}).

RESULTS AND DISCUSSION

The effects of silica loading, testing rate and angle of testing on the adhesion properties are systematically discussed below.

Viscosity

The dependence of viscosity of SBR-based adhesive on silica loading is shown in Figure 1. The graph shows that viscosity of adhesive increased steadily with silica loading; the increase was greater as the filler concentration was increased. This observation is attributed to the concentration effect of the silica whereby interaction between silica and SBR occurred, resulting in the increase in viscosity of adhesive. The nonlinear dependence of viscosity on silica concentration indicates that the molecular interaction between SBR and silica did not increase linearly with silica loading, suggesting that the adhesive system was a non-ideal mixture. This observation is consistent with our previous results reported on zinc oxide and kaolin-filled natural rubber adhesives (Poh & Ng, 2013; Poh & Sulaiman, 2015).

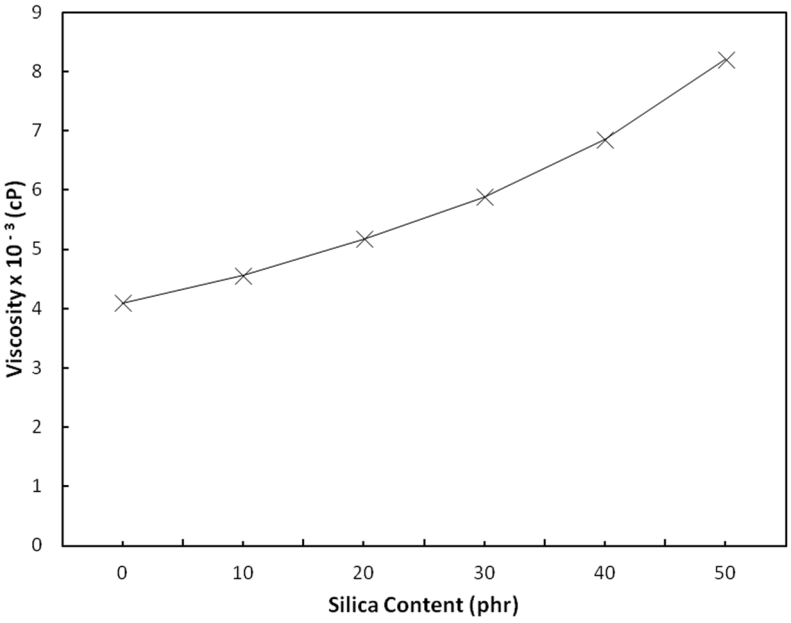

Figure 1. Variation of viscosity of SBR-based adhesive with silica content.

Peel Strength

Figures 2-4 show the dependence of peel strength on silica loading for the T-, 90º and 180º peel tests at 60 μm and 120 μm coating thicknesses. For the three modes of peel tests, peel strength increased with filler loading up to 20 phr silica content before it decreased with further addition of silica for both coating thicknesses. The initial increase in peel strength is attributed to the increase in wettability, which enhanced mechanical interlocking and anchorage of the adhesive in pores and irregularities in the adherent (Lee, 1991; Gierenz & Karmann, 2001). At the optimum silica content of 20 phr, culmination of wettability occurred whereby the lowering of surface tension provided proper flow and wetting characteristics (Poh & Chow, 2007). However, at higher filler content, the wettability of adhesive decreased as viscosity increased

with further addition of silica filler. The viscous component of adhesive was decreased by the increase in viscosity, which hindered the rubber chain mobility, hence decreasing the wettability of adhesive as shown by the drop in peel strength at the higher loading of silica. Figures 2-4 also show that for a fixed silica loading, the 120 μm coated sample consistently indicated higher peel strength compared to that of 60 μm coating thickness. This phenomenon is associated with the higher amount of adhesive in 120 μm coated substrate, which enhances the viscoelastic response of the adhesive (Leong et al., 2003) and is consistent with the general belief that peel force increases with increasing adhesive thickness up to a certain limit (Satas, 1982). The effect of testing rate on the peel strength (90º peel) at the optimum silica loading of 20 phr for 60 μm and 120 μm coating thicknesses is shown in Figure 5. Peel strength increased steadily with testing rate for both coating thicknesses, an observation that is attributed to the different viscoelastic responses as the testing rate was increased. At low testing rate, the viscoelastic response was predominantly viscous in nature and cohesive failure occurred. However, at high testing rate, the viscoelastic response was predominantly elastic, resulting in adhesive failure mode (Satas, 1982). The dependence of peel strength on peel angle is shown in Figure 6 at the optimum silica loading of 20 phr for 120 μm coating thicknesses. The plot indicates that the peel strength decreased steadily as the angle of peel test was increased. This observation is attributed to the increase of dissipation of energy in bending the tape away from the substrate at the line of detachment as the angle of detachment increased (Gent, 1987).

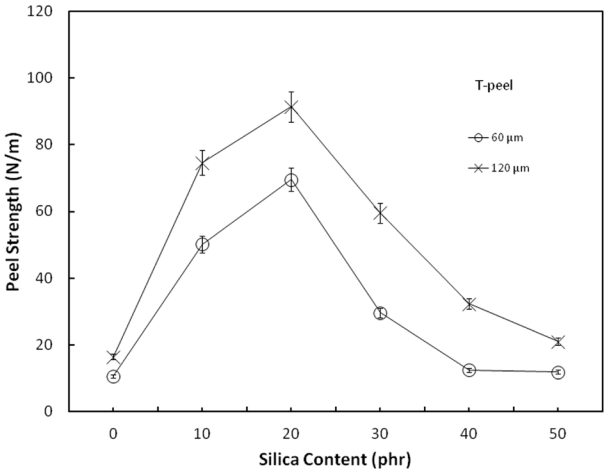

Figure 2. Variation of peel strength (T-peel) with silica content at 30 cm/min for 60 micron and 120 micron coating thicknesses.

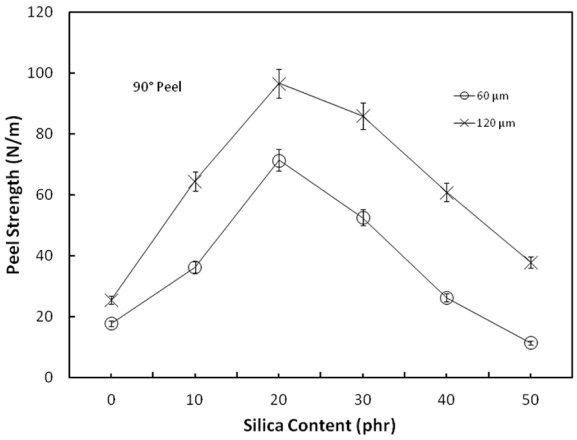

Figure 3. Variation of peel strength (90° peel) with silica content at 30 cm/min for 60 micron and 120 micron coating thicknesses.

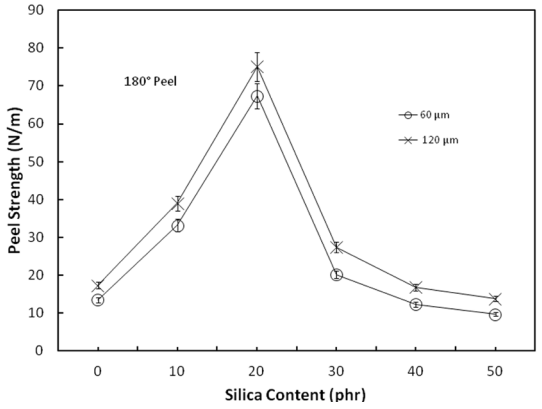

Figure 4. Variation of peel strength (180° peel) with silica content at 30 cm/min for 60 micron and 120 micron coating thicknesses.

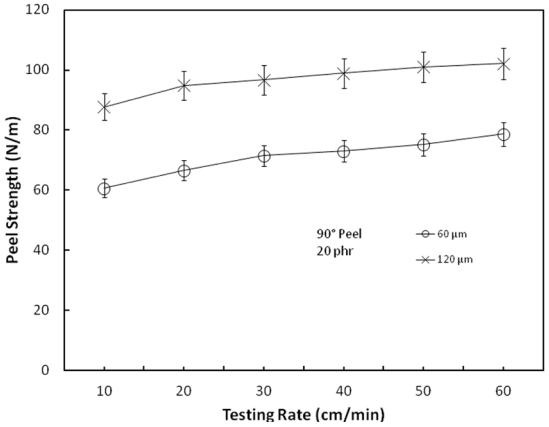

Figure 5. Dependence of peel strength (90° peel) on testing rate at 20 phr silica content for 60 micron and 120 micron coating thicknesses.

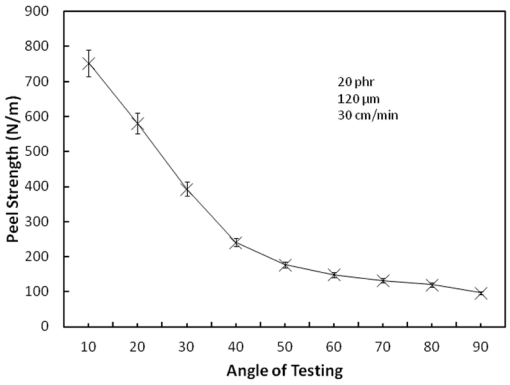

Figure 6. Dependence of peel strength on angle of testing at 30 cm/min and 20 phr silica content for 120 micron coating thicknesses.

Shear Strength

Figure 7 shows the effect of silica loading on the shear strength of SBR-based adhesive at 60 µm and 120 µm coating thicknesses. As in the case of peel strength, the shear strength also increased with silica content up to an optimum loading of 20 phr filler. This observation is attributed to the steady increase in the cohesive and adhesive strength up to the optimum filler loading where maximum cohesive strength occurred as reflected by the peak value of shear strength in Figure 7. However, after the optimum filler loading, cohesive strength decreased due to the increasing dilution effect of silica with further filler loading. For a fixed silica loading, the shear strength increased with coating thickness. The observation is attributed to the increase in the amount of adhesive in thicker coated samples, which enhanced the shearing resistance; hence, this increased the cohesive strength of the adhesive. Figure 8 shows the effect of testing rate on shear strength of silica-filled SBR adhesive at 20 phr filler for 60 µm and 120 µm coating thicknesses. As in the case of peel strength, shear strength also increased steadily with testing rate for both coating thicknesses. This finding is attributed to the difference in viscoelastic response of the rubber adhesive as testing rate was increased. At low testing rate, the viscoelastic response was predominantly viscous in nature, resulting in lower shear strength. On the other hand, at higher testing rate, the viscoelastic response was predominantly elastic, contributing to higher shear strength where adhesive failure mode occurred.

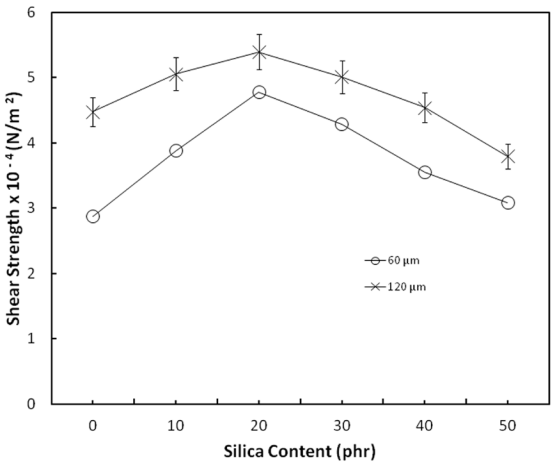

Figure 7. Variation of shear strength with silica content at 30 cm/min for 60 micron and 120 micron coating thicknesses.

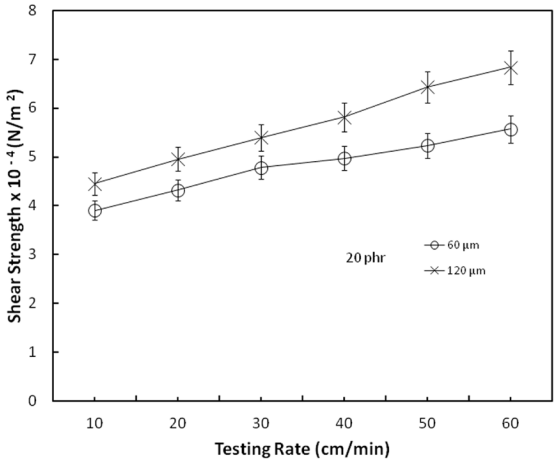

Figure 8. Dependence of shear strength on testing rate at 20 phr silica content for 60 micron and 120 micron coating thicknesses.

Tack

The dependence of tack on silica loading at 60 µm and 120 µm coating thicknesses is shown in Figure 9. As in the case of peel and shear strength, tack also increased with silica loading up to the optimum value of 20 phr content for both coating thicknesses. This observation is again attributed to the increase in wettability, which culminated at 20 phr silica loading as shown by the maximum tack value. At the optimum silica loading, the adhesive was able to conform to the irregularities of the adherend i.e. low surface energy condition was observed (Satas, 1982). However, as the filler loading was further increased beyond 20 phr, wettability decreased due to the increase in viscosity, which reduced the ability of the adhesive to flow and wet the substrate. For a fixed silica content, the 120 µm coated substrate consistently showed higher tack value compared to the 60 µm coated sample. This observation is ascribed to the higher

amount of adhesive, which increased the mechanical interlocking of adhesive molecules in the porous or rough substrates (Rezaeian et al., 2012) as coating thickness was increased and consequently, increased the wettability as shown by the higher tack value in thicker-coated samples as shown in Figure 9. The effect of testing rate on the tack property is illustrated in Figure 10. The graph indicates that tack increased steadily with testing rate for both coating thicknesses. This phenomenon is associated to the change in viscoelastic responses as testing rate was increased. At low testing rate, the response was predominantly viscous and cohesive failure occurred whereas at high testing rate, the response was predominantly elastic, resulting in the adhesion failure (Satas, 1982). The increase in tack value at high testing rate is thus attributed to the increase in elastic component of the viscoelastic response of the silica-filled adhesive.

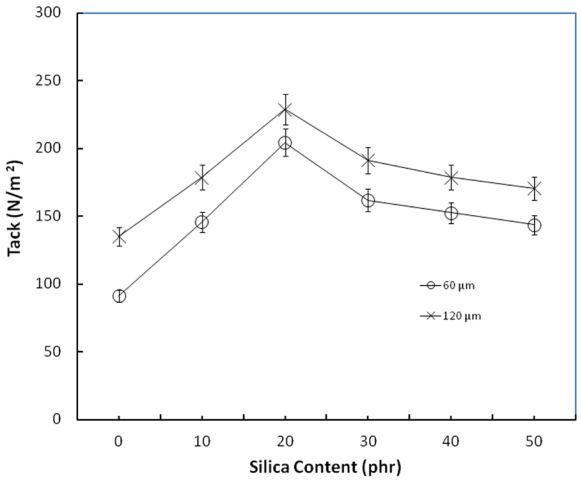

Figure 9. Variation of tack with silica content at 30 cm/min for 60 µm and 120 µm coating thicknesses.

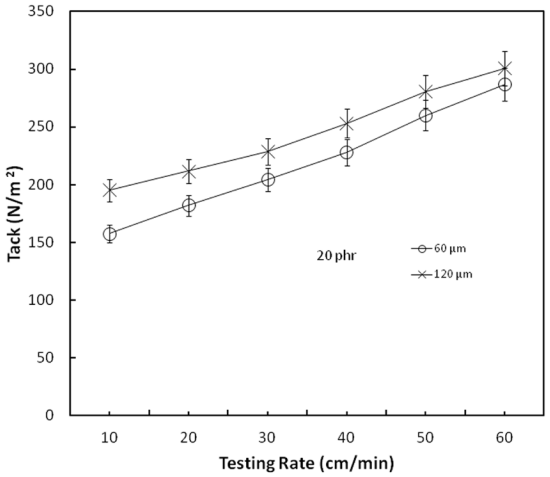

Figure 10. Dependence of tack on testing rate at 20 phr silica content for 60 µm and 120 µm coating thicknesses.

CONCLUSION

The viscosity of SBR-based adhesive increased with increases in silica loading, an observation which is attributed to the molecular interaction between silica and SBR. This concentration effect increased as silica content was increased. Peel strength, shear strength and tack increase with silica loading up to 20 phr of the filler before decreasing with further addition of silica filler. This observation is attributed to the increase in wettability and cohesive strength, which culminates at 20 phr of silica loading. Peel strength decreased with angle of peel test. In all cases, peel strength, shear strength and tack increased with coating thicknesses, an observation that is attributed to the higher amount of adhesive in thicker-coated samples. All the adhesion properties increased with increase in testing rate due to the difference in viscoelastic response as testing rate was increased, whereby viscous and elastic response occurred at low and high testing rates, respectively.

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