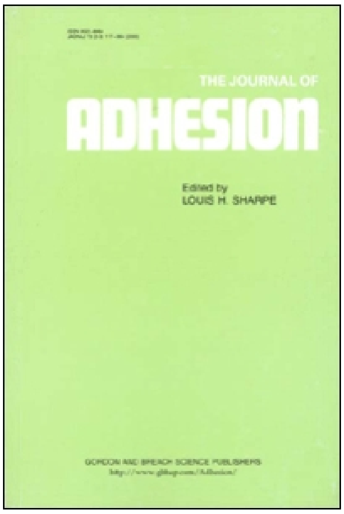

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Hysteresis Loss as a Measure of Metal-Rubber Adhesion

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As in the case of reinforcing-filler-induced increase in hysteresis in rubbers, attachment of aluminium (Al) foil onto the surface of a rubber blend (chlorobutyl rubber-carboxylated nitrile rubber) also causes an increase in hysteresis of the rubber. Changes in the hysteresis loss due to Al foil can be correlated with the Al/rubber adhesion.

KEY WORDS adhesion; chlorobutyl rubber; carboxylated nitrile rubber; rubber blend; self-crosslinkable; hysteresis loss; peel strength.

INTRODUCTION

Hysteresis is a measure of the energy dissipated by a material during a cyclic deformation. Fillers are known to cause an increase in hysteresis loss in rubbers[1–3] and hysteresis measurements can be used to estimate the reinforcing ability of fillers. Hysteresis arises due to wetting (or interaction) between the filler particles and the matrix.[4–12] In the case of metal/rubber adhesion, a similar situation arises in the sense that the higher is the interaction (or wetting) between the metal surface and the rubber matrix, the higher is the adhesion. Accordingly, it is expected that the adhesion of metal with the rubber surface could induce changes in the hysteresis properties of the rubber. With this objective in mind, the present investigation was undertaken.

In the present communication, we report the results of our studies on the effect of attachment of aluminium (Al) foil onto a rubber surface on the hysteresis properties of the rubber. It was reported earlier that the self-crosslinkable rubber blend based on chlorobutyl rubber (CIIR) and carboxylated nitrile rubber (XNBR) acts as an adhesive for Al-Al bonding.[13,14] The peel strength and the type of failure of the Al-Al joints depend, among other factors, on the moulding time.[15,16] In the present studies, the composites based on a 1:1 blend of CIIR and XNBR were moulded for 5, 30 and 60 min with the objective of varying the peel strength of Al-Al joints.

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Attempts were made to correlate the increase in hysteresis loss of the rubber, due to the attachment of the Al foil to one of its surfaces, with the peel strength of an Al-rubber-Al composite.

THEORY

Figure 1 shows the dumbbell-shaped test samples. Their dimensions and physical properties are identical except that in the sample in Figure 1b, Al is bonded to one of its surfaces. If the sample as in Figure 1a is subjected to cyclic deformation along the major axis at a constant test temperature and rate, there will be hysteresis loss due to this cyclic deformation. Let us designate this hysteresis loss as Hy_1.

If the same experiment is repeated for the sample as in Figure 1b, the Al foil attached to the rubber surfaces will cause a hysteresis loss, abbreviated as Hy_2. The increment in hysteresis loss over the neat rubber (that is, $\Delta Hy = Hy_2 - Hy_1$) is likely to depend on the extent of adhesion.

EXPERIMENTAL

Raw Materials

The Al foil (thickness 0.05 mm, hardness 52.70 V.P.N.) was obtained from Indal, Bombay. The chlorobutyl rubber used was chlorobutyl 1168 (chlorine content 1.2 wt%) of Exxon Chemical Company, U.S.A. The carboxylated nitrile rubber used was Krynac 231 from Polysar Ltd., Canada.

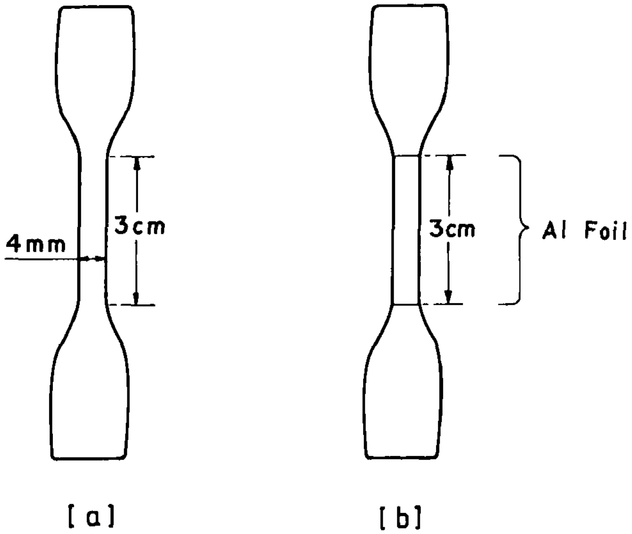

FIGURE 1 Sample geometry: a) Rubber sample for hysteresis test. b) Aluminium-rubber composite sample for hysteresis test.

Preparation of the Rubber Blend

CIIR was first masticated for one minute in a 35 × 15 cm^2 two-roll mill. Next XNBR was blended in and further masticated for 2 minutes.

(a) Preparation of samples of the neat rubber blends (without Al foil): The masticated rubbers were moulded in a Teflon® coated mould (4 × 8 × 1.5 cm) in a hydraulic press at a temperature of 180°C and a moulding pressure of 0.35 MPa.

(b) Preparation of samples of rubber blend with Al foil on one surface: Rubber samples bonded to Al foil were prepared by moulding the masticated rubber with Al foil (3 × 4 cm) on one side. Figure 1 shows the sample geometry. Al foil was bonded to the mid portion of the sample. The two ends were kept free for gripping with the Zwick Universal testing machine (UTM), model No. 1445.

Hysteresis Measurement

A dumbbell-shaped specimen, according to ASTM D412-80, was used for hysteresis measurement.[17] The hysteresis experiments were carried out on the Zwick UTM, at 20% elongation. Experimental results were reproducible to within ±1%.

The tension set was determined after each cycle by measuring the distance between two gage marks of the dumbbell-shaped specimen. The measurements were made after 24 hr of rest time.

Preparation of the Composite for Peel Testing

Aluminium foils were cut into 15 cm × 20 cm sizes and cleaned with acetone. Three gm of the rubber blend was passed four times through the two-roll mill at the closest nip gap and a thin rubber layer was obtained. This rubber layer was placed between the two aluminium foils and the assembly was bonded under a pressure of 0.35 MPa at 180°C, for various times, in a rubber moulding press. A part of the metal foil assembly was not filled with rubber. It was kept outside the mould during moulding and the two parts of it were fixed in the grips of an Instron tensile testing machine for measurement of the peel strength. The dimensions of the test specimens were 10 cm × 2.5 cm, while the bonded region was 5 cm × 2.5 cm. Figure 2 is the schematic diagram of the test specimen for peel testing.

Determination of 180° Peel Strength of the Composite

The 180° peel strength was determined in accordance with ASTM D413-82 using an Instron UTM, model 1195. The test specimen was fitted horizontally to the grip of the Instron machine through the non-bonded part of the metal foils. A steady load was applied at the rate of 50 mm/min until separation was complete. The average peel strength in terms of force/width was calculated from the plot of force *versus* displacement obtained in the Instron machine according to the following equation,

$$\text{Peel strength} = \frac{2\,F}{w} \qquad (1)$$

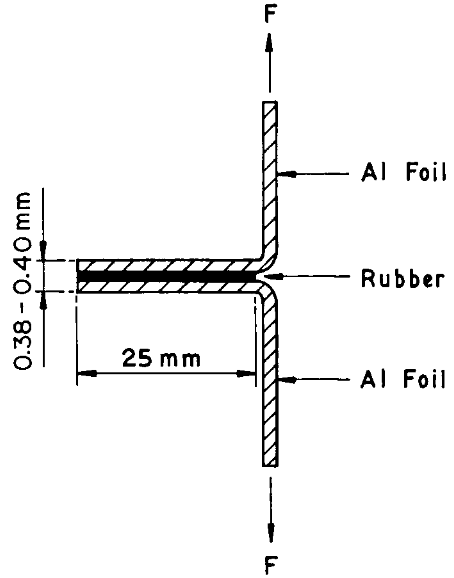

FIGURE 2 Schematic diagram of the Aluminium-rubber-Aluminium sample for peel test.

where F is the average force in Newton and w is the width of the specimen in meter. The results were found to be reproducible within ±5%.

RESULTS AND DISCUSSION

Table I gives the composite designations and the results from the hysteresis experiments.

Figure 3a shows the hysteresis plots of the CX_1 and CX_1/Al systems. It is evident that:

(a) Attachment of Al foil onto the rubber surface increased the hysteresis loss of the rubber due to (i) the absence of deformation within the rigid Al foil and (ii) the immobilization of the rubber on the surface of the Al foil due to wetting.

(b) As the number of cycles increased, the hysteresis loss of the neat rubber (CX_1) became reasonably constant after the third cycle. However, in the case of the composite (CX_1/Al), a higher number of cycles were required for the constancy to occur than in the neat rubber.

(c) In all cycles, CX_1/Al showed the higher hysteresis loss compared with the CX_1 systems. The results are summarised in Table I.

Figure 3b shows similar plots for CX_2 and CX_2/Al. The results are similar to those of the CX_1 and CX_1/Al systems. However, it is to be noted that although the hysteresis loss values of CX_1 and CX_2 are close to each other, the CX_2/Al system

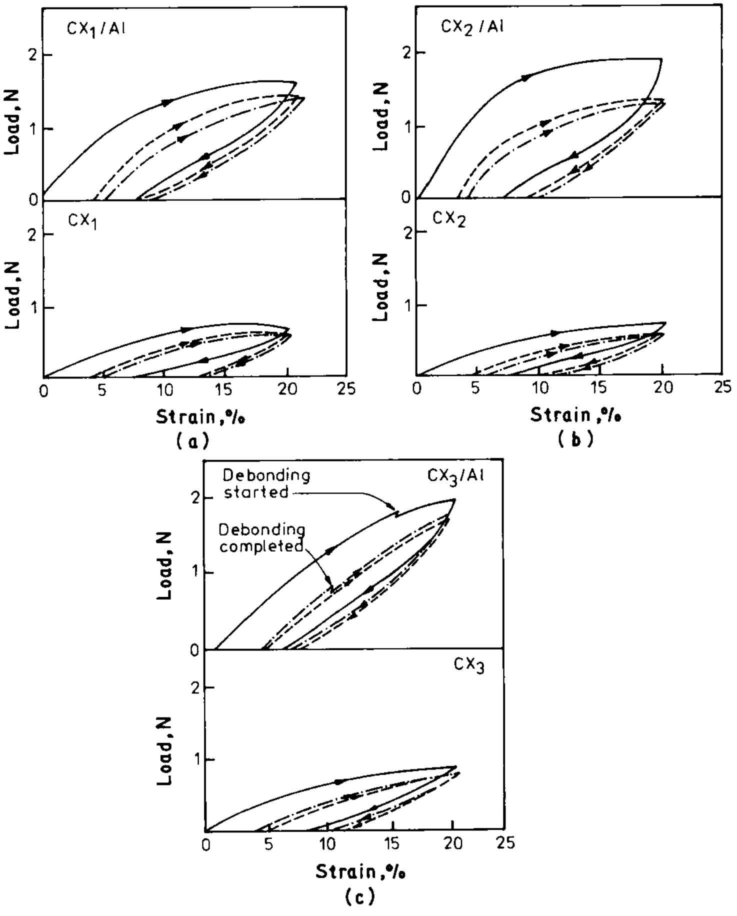

FIGURE 3 Hysteresis loss plots. a) CX_1 and CX_1/Al, b) CX_2 and CX_2/Al, c) CX_3 and CX_3/Al. ——— cycle 1, – – – – cycle 2, — · — cycle 3.

showed higher hysteresis loss than the CX_1/Al system. It was observed earlier that the CX_2 system showed a higher extent of adhesion with Al than the CX_1 (Table I).[14]

Figure 3c shows the hysteresis plots for the CX_3 and CX_3/Al systems. Here again, CX_3 showed hysteresis behaviour similar to CX_1 and CX_2, but the CX_3/Al composite exhibited a behavior different from the CX_1/Al and CX_2/Al systems. Furthermore, from the third cycle onwards the hysteresis loss of the CX_3/Al system was similar to that of the CX_3 system, in that there was complete debonding between Al and the matrix in CX_3/Al after the third cycle. It was reported earlier[14] that CX_3 showed much less adhesion (180° peel strength) with Al foil as compared with CX_1 and CX_2 and the nature of the failure of Al-adhesive-Al joints during peeling was interfacial in the case of CX_3, in contrast to its cohesive nature in the case of CX_1 and CX_2 (Table I).

TABLE I
Effect of Al foil on the hysteresis loss of 1:1 CIIR-XNBR blend moulded for different times.[a] Values in parenthesis are the tension set results, (—) indicates nil tension set

		Hysteresis loss × 10^5 (j/m^2)					
Moulding time (min)	Composite designation	1st cycle (w_1)	2nd cycle (w_2)	3rd cycle (w_3)	4th cycle (w_4)	5th cycle (w_5)	Peel strength of Al-rubber-Al joints and type of failure[14] (N/m)
5	CX_1	52 (1.7)	31 (3.0)	26 (6.8)	25 (6.8)	25 (6.0)	—
5	CX_1/Al	123 (—)	67 (—)	54 (1.3)	50 (1.8)	49 (1.7)	4800, cohesive
30	CX_2	52 (—)	32 (—)	27 (1.7)	27 (3.4)	27 (5.1)	—
30	CX_2/Al	156 (—)	84 (—)	61 (1.0)	60 (2.3)	58 (2.7)	7200, cohesive
60	CX_3	50 (—)	33 (—)	28 (1.4)	28 (1.7)	28 (1.7)	—
60	CX_3/Al	99 (—)	58 (—)	32[a] (1.0)	32[a] (1.3)	30[a] (1.7)	960, interfacial

[a] debonding between Al foil and the rubber surface occurred in third cycle.

Results of tension set measurements after each hysteresis cycle is shown in Table I. The set values decreased with increase in moulding time due to the decrease of plastic flow. The set developed in the neat rubber was higher than that of the corresponding composite. The set which developed in the neat rubber and the composites became almost constant from the third cycles onwards, which showed that the deformation achieved by the rubber became reasonably constant within the third cycle. Presumably, this is due to reorientation and regrouping of long-chain rubber molecules in the direction of elongation which, in turn, decreases the hysteresis values at the higher number of cycles.

Since the CIIR-XNBR rubber blend is self-crosslinkable, the extent of crosslinking increases with the moulding time. Although the hysteresis loss values do not differ much, the corresponding composites such as CX_1/Al, CX_2/Al and CX_3/Al showed a wide variation in hysteresis loss with the moulding time. This variation in hysteresis loss with the moulding time for the composites can be explained on the basis of differences in extent of adhesion.[14]

From the above discussions, it can be argued that the hysteresis behaviour and the peel strength originate from similar mechanisms. It is apparent, therefore, that the increment in hysteresis loss (ΔHy) can be correlated with the extent of metal-rubber adhesion. In Figure 4 we have plotted ΔHy against the peel strength of the Al-rubber-Al system. From Figure 4, it is clear that ΔHy increases with the increase in extent of adhesion. The stronger is the adhesion, the stronger is the immobilization in the metal-rubber interface and the higher will be the viscoelastic energy loss. This is true for all cycles of hysteresis measurement, but the increment of ΔHy with respect to peel strength is prominent in the first two cycles. At higher numbers of cycles of hysteresis measurements, the rate of increase of ΔHy with respect to the

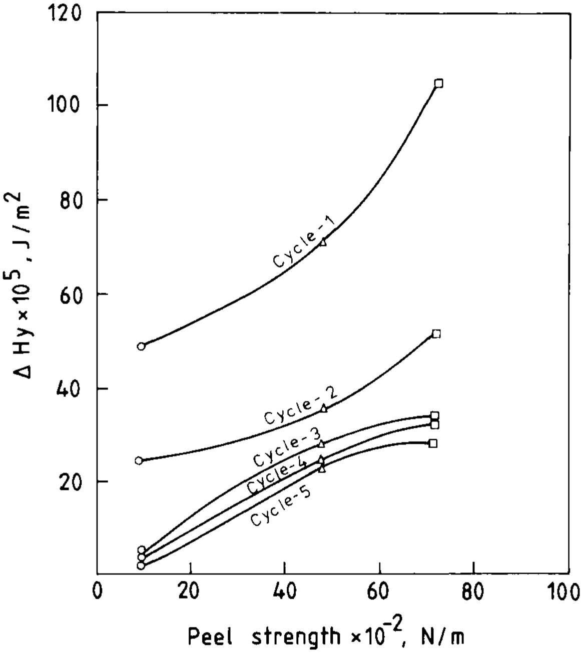

FIGURE 4 Variation of ΔHy with peel strength.

peel strength of the Al-rubber-Al system is less marked. This is ascribed to the disentanglement and reorientation of long-chain rubber molecules during stretching along the direction of the major axis.[7]

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