

Effect of latex coated coconut fibre on cure characteristics and physico-mechanical properties of natural rubber-coir composites

D G Edirisinghe*, W D M Sampath* and P C Wettasinghe*

**Rubber Research Institute of Sri Lanka, Telewala Road, Ratmalana, Sri Lanka*

Abstract

A wide variety of natural fibres such as coconut fibre (coir), bagasse, banana fibre, sisal, etc. can be used to reinforce polymers. Natural fibre reinforced, biodegradable composites are eco-friendly. However, the interfacial adhesion between natural fibre and most of the biodegradable polymers is not adequate. The objective of this study was to modify readily available coir by coating with natural rubber (NR) as well as synthetic rubber latices with the aim of improving adhesion between coir and rubber in order to develop a coir filled NR based composite suitable in manufacture of tyre treads.

Coir was coated with different compounded latices namely, neoprene, nitrile and NR and with uncompounded NR latex. Thereafter, composites were produced by mixing 15 phr of the latex coated coir fibres with virgin NR according to a tyre tread formulation and properties of the composites were evaluated and compared. Results revealed that processing safety of the compounded NR latex coated coir filled rubber composite is higher than that of the other three composites, whereas uncompounded NR latex coated coir filled rubber composite showed a similar processing safety to that of the two synthetic latex coated coir filled rubber composites. However, the two NR latex coated coir filled rubber composites were faster curing than the synthetic latex coated coir filled rubber composites. Minimum torque results indicated that compounded NR latex coated coir filled rubber composite has a higher processability when compared to the other composites. However, the latter composites indicated a higher state of cure and cross-link density in comparison to the former composite. Tensile strength, elongation at break, resilience and abrasion volume loss of the compounded NR latex coated coir filled rubber vulcanisate are superior to those of the other three vulcanisates.

In overall cure characteristics and mechanical properties, especially abrasion resistance of the composite based on compounded NR latex coated coir indicates that it could be used in manufacture of tyre treads.

Keywords: coir, natural rubber composites, natural rubber latex, neoprene latex, nitrile latex

Introduction

Natural fibres are generally lignocellulosic in nature, consisting of helically wound cellulose microfibrils in a matrix of lignin and hemicellulose. Natural fibres have been used to reinforce materials for over 3,000 years. More recently they have been employed in combination with rubber and plastics (Sakdapipanich *et al.*, 2007). Many types of natural fibers have been investigated for use in rubber and plastic including coconut fibre (coir), bamboo fibre, oil palm fibre, isora fibre, banana fibre, sisal, *etc.* Natural fibres have the advantage that they are renewable resources and have marketing appeal. They are increasingly used in automotive and packaging materials.

Natural fibres degrade faster than synthetic fibres in natural environments, minimizing environmental pollution. However, biodegradation of natural fibre can decrease the life time of fibre reinforced polymer composites. To prolong their performance in natural environments, natural fibres are treated to protect from any circumferential agents. Hydroxyl groups from cellulose and lignin are natural substrates for modification. In addition, coating of fibres often reduces water absorption, protection from bacteria and fungi attack. The treatment methods which have been used for fibre reinforced polymer composites are not suitable for protection of the fibres in contact with soils (Farshid *et al.*, 2011).

Coir is a versatile lignocellulose fibre obtained from the coconut tree (*Cocos nucifera*), which grows extensively in tropical countries. Because of its hard-wearing quality, durability and other advantages, it is used for making a wide variety of floor furnishing materials, yarn and rope. However, these traditional coir products consume only a small percentage of the potential total world production of coconut husk. In the past, composites of coconut fibre/natural rubber (NR) latex were extensively used by the automotive industry (Geethamma *et al.*, 1998) and presently mattresses, pots, *etc.* are widely produced using this combination. Apart from the conventional uses of coir as mentioned above, research and development work has been conducted to find new applications for coir, including utilization of coir as a reinforcing material for plastics (Owolabi *et al.*, 1985).

Coir is an inexpensive fibre among the various natural fibres available in the world. Furthermore, it possesses the advantages of a lignocellulose fibre. It is not toxic and possesses no waste disposal problem. Unfortunately, the performance of coir as a reinforcement in polymer composites is unsatisfactory and not comparable even with other natural fibres. This inferior performance of coir is due to various factors such as its low cellulose content, high lignin and hemicellulose content, high microfibrillar angle and large and

variable diameter (Geethamma *et al.*, 1998).

The hydrophobic NR matrix and hydrophilic cellulose fibre can be made compatible through modification of polymer or fibre surface. The extent of adhesion is usually increased by the use of bonding agents and chemical modification of fibres. The effects of a silane coupling agent (Si69) on curing characteristics and mechanical properties of bamboo fibre filled NR composites were studied (Ismail *et al.*, 2002). The mechanical properties of composites such as hardness, modulus, tensile and tear strengths improved with the addition of Si69. Chemical treatment of cellulose fibres usually changes the physical and chemical structure of the surface. The use of maleic anhydride grafted copolymer and alkaline treatment of the natural fibre are the most used techniques with the aim to improve the fibre-matrix interfacial bonding (Ku *et al.*, 2011). Arumugam *et al.* (1989) evaluated mechanical properties of NR composites prepared with pristine and sodium hydroxide treated coconut fibers. They added the same coupling agents as Ismail *et al.* (2001) in order to improve rubber-filler interactions. Results of the compounds prepared with pristine and treated fibres were compared with those of untreated fibres and an enhancement in the mechanical properties was observed. It was reported that alkali treatment on coir fibre enhances the thermal stability and maximum moisture retention (Mahato *et*

al., 1993). Prasad *et al.* (1983) reported that the use of alkali treatment on coir fibres improves the mechanical properties of coir-polyester composites. Jacob *et al.* investigated the effect of chemical modification of banana fibre in NR (Communicated). Modification of banana fibre resulted in superior mechanical properties. Chemical modification of both sisal and oil palm fibres was imperative for increased interfacial adhesion and resulted in enhanced properties (Jacob *et al.*, 2004). The influence of alkali treatment on oil palm fiber reinforced NR composites was investigated by Joseph *et al.* (2006). They observed superior mechanical properties for the composites after chemical modification due to better adhesion. Effects of alkali, silane coupling agent and acetylation on oil palm fibres have been studied by Mahato *et al.* (1993). De *et al.* (2006) were able to improve adhesion between grass fiber and NR matrix by alkali treatment.

Chemical modification of pineapple leaf fiber in NR was investigated by Lopattananon *et al.* (2006). Sodium hydroxide and benzoyl peroxide were used to treat the surfaces of fiber. It was found that all surface modifications enhanced adhesion and tensile properties. The effects of different chemical treatments, including mercerisation, acetylation, benzoylation and treatment with toluene diisocyanate (TDI) and silane coupling agents, on isora fibre properties and mechanical properties were analyzed. Isora fibre

was seen to have immense potential as reinforcement in NR. Acetylation and TDI treatments gave higher tensile strength values compared to other treatments (Mathew *et al.*, 2004). Maize stalk fibres were also chemically treated with acetic anhydride (acetylation) to enhance their compatibility with the hydrophobic rubber matrix (Chigondo *et al.*, 2013). The NR-maize stalk fibre filled composites showed good processing safety and when compared with untreated NR-maize stalk fibre composites, the acetylated composites exhibited higher mechanical properties, reduced moisture absorption and higher resistance to hydrothermal aging.

The effect of plasma treatment on cellulose fibers in NR composites was investigated by Ahlblad *et al.* (1994). Chemiluminescence analysis was used to indicate the grafting on the surface of the cellulose fibres and also to estimate the effect of the plasma on the cellulose fibres. The results indicated the possibility of obtaining a surface layer on the fibres, which would lead to improvement of mechanical properties of rubber composites.

The use of chemical treated natural fibres such as coir in composites has increased due to their benefits such as high strength with very low weight and recyclable nature. However, no work on latex coating of natural fibres has been reported in the past. Hence, this research was conducted to study the effect of latex coating of coir on properties of NR-coir composites.

Experimental

Materials

Neoprene and nitrile latex compounds were supplied by Dipped products Plc., Sri Lanka. Compounded NR latex, uncompounded NR latex and virgin NR (RSS 2) were purchased from a local supplier. Zinc oxide (ZnO), stearic acid, carbon black (N 330-high abrasion furnace black), rubber processing oil (Dutrex R), IPPD (N-isopropyl, N'-phenyl paraphenylene diamine), TMQ (2,2,4 trimethyl 1,2-dihydroquinoline), TBBS (N-t-butyl-2 benzothiazole sulphenamide) and sulphur were purchased from the local market. Coconut fibre (coir) was supplied by a local supplier and was used as received.

Method

Coconut fibre was coated with compounded neoprene latex, compounded nitrile latex, compounded NR latex and uncompounded NR latex. Neoprene and nitrile latices were chosen as coating materials for coir as these are readily available and existence of some similarities between each of the structures of the two synthetic rubbers and the structure of NR. Further, better adhesion between each of the two polar synthetic rubber latices and coir would be expected due to the presence of hydroxyl groups in cellulose and lignin of coir. Four composites were produced by mixing the latex coated fibres with virgin NR according to the formulations given in Table 1.

Table 1. *Formulations of latex coated coir filled rubber composites*

Ingredient	Formulation			
	S1	S2	S3	S4
Natural rubber (RSS 2)	100	100	100	100
Zinc oxide	5.0	5.0	5.0	5.0
Stearic acid	1.5	1.5	1.5	1.5
Processing oil	3.0	3.0	3.0	3.0
Sulphur	2.5	2.5	2.5	2.5
Flectol H	1.0	1.0	1.0	1.0
MBT	1.5	1.5	1.5	1.5
Uncompounded NR latex coated coir	15	00	00	00
Compounded neoprene latex coated coir	00	15	00	00
Compounded nitrile latex coated coir	00	00	15	00
Compounded NR latex coated coir	00	00	00	15

Properties of the composites containing compounded latex coated fibres were evaluated and compared with those of the composite containing uncompounded NR latex coated fibres. The mixing cycle used in the preparation of latex coated coir filled rubber composites is given in Table 2.

The first stage of mixing was carried out using a Baker Perkins Engineers, Petersborough and London, laboratory Banbury and the second stage was carried out using a David Bridge & Co. Ltd. Castleton Rochdale, England, laboratory two-roll mill (6" x 13").

Table 2. *Mixing cycle used in the preparation of latex coated coir filled rubber composites*

Stage 1: Internal mixer	Total time (min.)
Added NR (RSS 2)	0
Added ZnO + Stearic acid + Flectol H	2
Added ½ (latex coated coir + oil)	4
Added remaining ½ (latex coated coir + oil)	6
Dumped	7
Stage 2: Two-roll mill	
Added MBT	8
Added sulphur	9
Sheeted out	10

Determination of cure characteristics of the latex coated coir filled rubber composites

Cure characteristics such as the minimum torque (M_L), maximum torque (M_H), scorch time (TS_2), optimum cure time (T_{90}), cure rate index (CRI) and delta cure ($M_H - M_L$) of the four rubber compounds were obtained by a MDR 2000 Moving Die Rheometer (M/S Alpha Technologies, USA) at 150°C.

Preparation of latex coated coir filled rubber vulcanisates

The four latex coated coir filled rubber composites were placed in test piece moulds and pressed between the platens of a hydraulic press (Yeji Corporation, Taiwan). The samples were cured at 150°C temperature and at an applied pressure of 20 MPa according to respective optimum cure times obtained from the rheographs. After curing, the test pieces were removed from the moulds and immediately cooled under tap water to prevent further curing.

Measurement of physico-mechanical properties of the latex coated coir filled rubber vulcanisates

A Gotech Testing Machines Inc. tensile testing machine was used to measure the tensile properties of the rubber vulcanisates in accordance with ISO 37: 2011 at room temperature ($27 \pm 2^\circ\text{C}$) at a grip separation rate of 500 mm/min. Tear strength of the vulcanisates was measured using angle (Die B) test pieces with the aid of the same machine as per ISO 34-1:2010. Hardness of the vulcanisates was measured by a “Digi Test” hardness tester for hardness in the

IRHD N-scale as per ISO 48:2010. Resilience of the vulcanisates was measured by a Wallace Lupke pendulum in accordance with ISO 4662: 2009. Abrasion volume loss of the vulcanisates was determined using a DIN abrasion tester in accordance with DIN 53516.

Determination of aging properties of the vulcanisates

Tensile and tear strengths were evaluated after aging. Aging was carried out in an air circulating oven at 100°C (Sanyo Gallenkamp, UK) for 22 h according to ISO 188: 2011.

Results and Discussion

Cure characteristics of the latex coated coir filled rubber composites

Cure characteristics of the four rubber composites are given in Table 3. Minimum torque (M_L) is an indication of the processability and is related to the stock viscosity of the compound. The minimum torque of compounded NR latex coated coir filled rubber composite (S4) is lower and hence the processability is higher than that of the other composites (Table 3) and it may be due to better distribution of the compounded NR latex coated coir in the NR matrix. Maximum torque (M_H) indicates the state of cure and delta cure ($M_H - M_L$) is an indication of the crosslink density. M_H and delta cure of compounded NR latex coated coir filled composite is lower than those of the others. Neoprene latex coated coir filled composite (S2) has the highest crosslink density compared to S1 and S4 composites (Table 3).

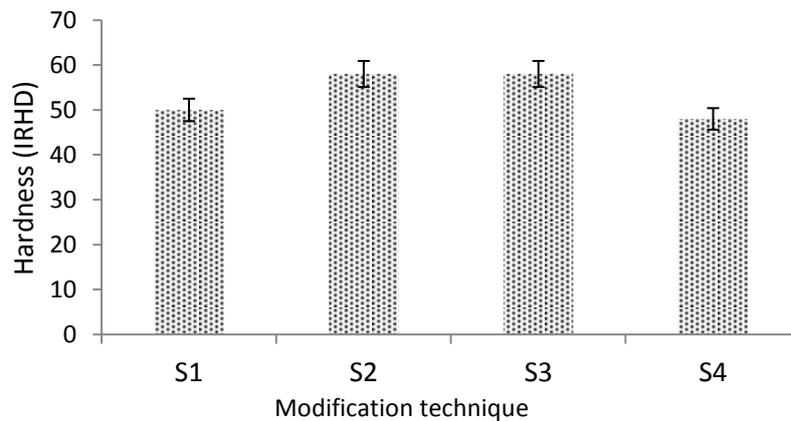
Table 3. Cure characteristics of the latex coated coir filled rubber composites

Sample No	Minimum torque (M_L) dNm	Maximum torque (M_H) dNm	Delta cure ($M_H - M_L$) (dNm)	Scorch time (ts_2) min.	Cure time (t_{90}) min.	Cure rate index min^{-1}
S1	2.25	9.34	7.09	0.31	2.27	51
S2	2.39	12.90	10.51	0.29	3.32	33
S3	2.42	11.48	9.06	0.31	3.11	36
S4	1.62	7.66	6.04	1.13	2.34	83

S4 composite containing compounded NR latex coated coir has the highest scorch time (ts_2) or in other words the highest processing safety. However, there is no significant difference between the scorch time of the composites S1, S2 and S3. Table 3 indicates that composites S1 and S4 containing NR latex coated coir are faster curing than the other two composites. In S1 and S4 composites, there are more crosslinking positions due to the presence of a greater number of double bonds and this is probably the reason for faster cure of the composites.

Physico-mechanical properties of latex coated coir filled rubber vulcanisates

Hardness variation of the four rubber composites is shown in Figure 1. This figure shows that there is no significant difference between the hardness of S2 and S3 vulcanisates and they are markedly higher than that of the S1 and S4 vulcanisates. When synthetic latices are coated on coir it becomes stiff due to reduced elasticity and this is probably the cause for higher hardness shown by the S2 and S3 vulcanisates compared to S1 and S4 vulcanisates.

**Fig. 1.** Variation of hardness of latex coated coir filled rubber vulcanisates

Similar to hardness, modulus at 300% elongation of synthetic latex coated coir filled vulcanisates is markedly higher than that of the NR latex coated coir filled vulcanisates (Fig. 2).

As expected, tensile strength of the NR latex coated coir filled vulcanisates is greater than that of the synthetic latices coated coir filled vulcanisates (Fig. 3). This indicates good adhesion between virgin NR and NR latex coated coir. In other words, NR latex coating has improved adhesion between NR and coir. Jacob & Anandjiwala (2009) reported that when fibres are aligned parallel to the stress direction, tensile strength develops a characteristic drop with increasing fibre volume content until a critical fibre level is reached. Further, it is important to ensure

homogeneous fibre dispersion in order to achieve maximum strength and performance of the composite materials (Visakh *et al.*, 2012).

There is no significant difference between the elongation at break of S1 and S4 vulcanisates (Fig. 4). The lower hardness shown by the S1 and S4 vulcanisates reflects higher elongation at break for the same compared to S2 and S3 vulcanisates. However, elongation at break of all the four vulcanisates is at a level acceptable for tyre treads. Generally, higher homogeneity or compatibility indicates higher elongation at break. Since S1 and S4 vulcanisates are more homogeneous, higher elongation at break has resulted for the same.

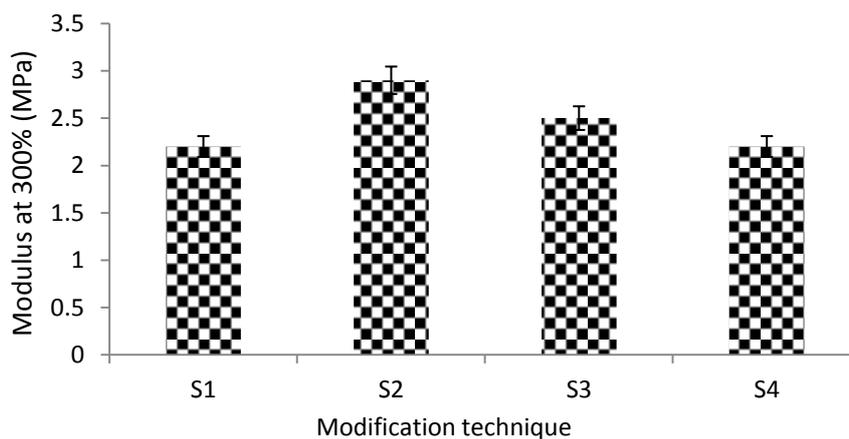


Fig. 2. Variation of modulus at 300% elongation of latex coated coir filled rubber vulcanisates

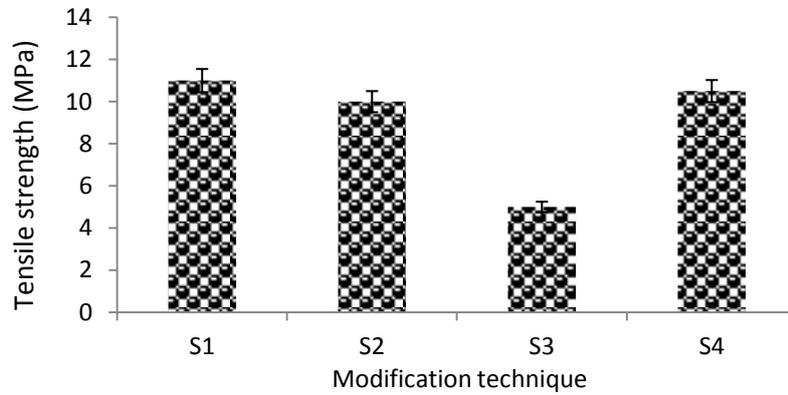


Fig. 3. Variation of tensile strength of latex coated coir filled rubber vulcanisates

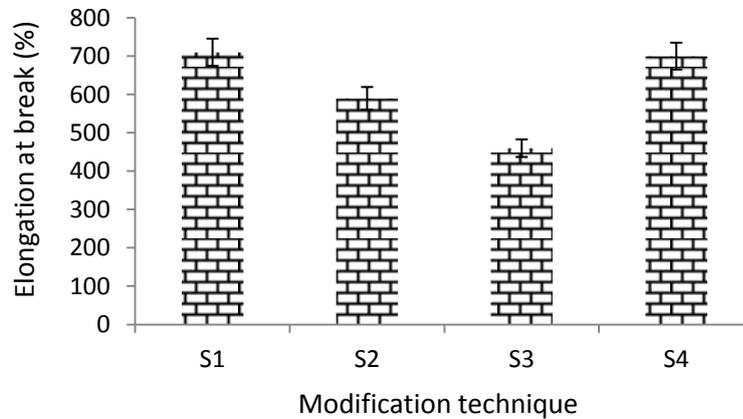


Fig. 4. Variation of elongation at break of latex coated coir filled rubber vulcanisates

Tear strength of the vulcanisate S1 is higher than that of the other vulcanisates (Fig. 5). This indicates that S1 is the most homogeneous vulcanisate. The initiated crack would tend to propagate through the interface and the extent of propagation depends on interfacial adhesion between the latex coated coir and NR matrix. NR latex coated coir improves its interfacial

shear strength in polymer matrix (Yousif & Tayeb, 2007). With NR latex coated coir, fiber is exposed more to the polymer matrix, leading to proper interaction between the surfaces (Khalid *et al.*, 2008). Additionally, since S1 contains coir coated with uncompounded NR latex, crosslinks would be evenly distributed between the latex coating and NR matrix unlike in

the case of composite S4. Hence an interface would not exist between coir and the matrix. This results in higher tear strength for the S1 composite.

The ratio of energy returned to the energy applied is termed as the resilience (Fig. 6). When the deformation is an indentation due to single impact, this ratio is termed “rebound resilience”. If the elasticity is higher, then less deformation energy is dissipated as heat. Heat build-up is a

measure of the amount of energy that is absorbed. The highest resilience is shown by the S4 vulcanisate and is in agreement with the results of elongation at break. The resilience of nitrile latex coated coir filled vulcanisate (S3) is very much lower than that of the other three vulcanisates. The lower elasticity of the S3 vulcanisate could be attributed to lower elongation at break of the same.

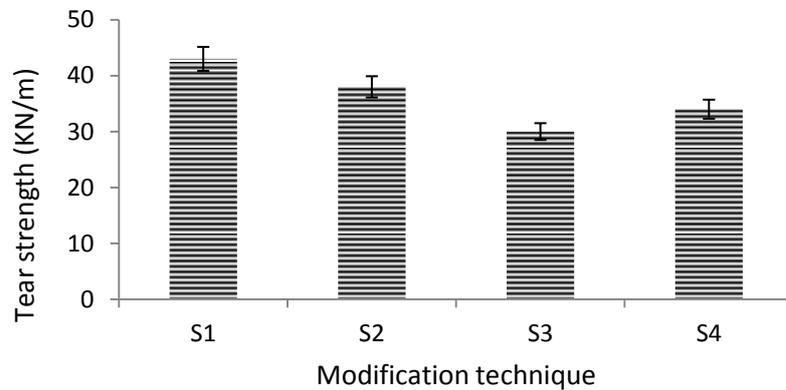


Fig. 5. Variation of tear strength of latex coated coir filled rubber vulcanisates

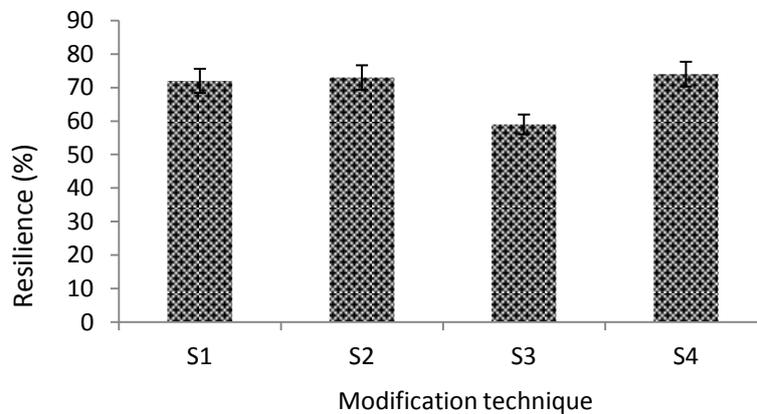


Fig. 6. Variation of resilience of latex coated coir filled rubber vulcanisates

The structure of neoprene is somewhat similar to NR and hence there is not much of a difference between the resilience of S1, S2 and S4 vulcanisates. Figure 7 shows that there is a marked difference between the abrasion volume loss of the three compounded latex coated coir filled rubber vulcanisates S2, S3 and S4. S4 is the highest abrasion resistance vulcanisate.

Abrasion volume loss results also indicate good adhesion between NR latex coated fibre and the NR matrix. The vulcanisate S3 containing nitrile latex coated coir shows the highest abrasion volume loss and can be attributed to poor adhesion or incompatibility between nitrile latex coated coir and the NR matrix and is in agreement with the results of tensile

strength, elongation at break and tear strength.

Aging properties of the latex coated coir filled rubber vulcanisates

Table 4 indicates that there is no marked difference between tensile strength and elongation at break, before and after aging at elevated temperatures. Generally, results of properties of vulcanisates after ageing are low in comparison to results of properties before aging due to conversion of polysulphidic linkages to short mono and disulphidic linkages at elevated temperatures. Formation of a large number of these mono and disulphidic crosslinks would increase the crosslink density and hence modulus increase as indicated from Table 4.

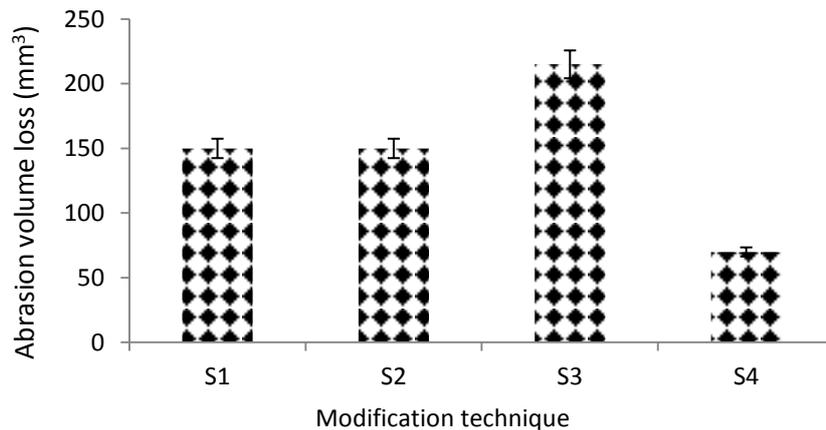


Fig. 7. Variation of abrasion volume loss of latex coated coir filled rubber vulcanisates

Table 4. Percentage retention of properties of the vulcanisates after aging at 100°C for 22 hrs

Sample	Modulus at 300% (MPa)	Tensile strength (MPa)	Elongation at break (%)	Tear strength (KN/m)
S1	111	92	92	86
S2	110	95	95	110
S3	102	84	100	100
S4	131	90	87	109

Conclusions

Processing safety of the S4 composite is higher and it is faster curing compared to the other composites. Further, most of the physico-mechanical properties of this composite are superior to those of the other composites and are at a level acceptable for tyre treads. Hence, the compounded NR latex coated coir filled rubber composite could be used in the manufacture of tyre treads.

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- Address for correspondence:* Dr (Mrs) D G Edirisinghe, Head, Rubber Technology Dept., Rubber Research Institute of Sri Lanka, Telewala Road, Ratmalana, Sri Lanka.
e-mail: dilharae@yahoo.com