

## EFFECT OF PART REPLACEMENT OF SILICA SAND WITH CARBON BLACK ON COMPOSITE PROPERTIES

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(Received 17 October 2001; accepted 31 October 2002)

Much interest have been shown in the study of filled natural rubber composites (Bristow 1986; Elliot 1986; Barker 1987; Adu and Adeosun 1997; Adeosun *et al* 1999). We have reported the properties of natural rubber filled with locally available materials (Adu *et al* 2000). The effect of local clay, limestone, silica sand and charcoal on the properties of natural rubber has been examined. Results have shown detrimental effects of silica sand on the properties of natural rubber compound. It has been reported that when silica is used as a part for part replacement of carbon black, the heat build up the composite decreased whilst tear resistance improved (Horn 1975; Barbin and Rodgers 1994).

Since the objective of the study is to attempt to find possible industrial utility for the local materials under examination, interest is now focused on possible boosting the reinforcing ability of silica sand by other materials. In the present work, we have replaced silica sand in part with carbon black in natural rubber composites and determined the mechanical and cure properties of the resultant composites. The properties of the composites filled with the mixed silica sand/carbon black were then compared to the properties of composites filled separately with silica sand and carbon black.

Compounding formulation of the composites examined are contained in Table 1. Sample preparation, compounding,

**Table 1**  
Compounding formulations

Composites	Parts per hundred rubber (pphr)						
	100	100	100	100	100	100	100
Natural Rubber	100	100	100	100	100	100	100
Zinc Oxide	5	5	5	5	5	5	5
Stearic acid	3	3	3	3	3	3	3
Mbt <sup>(a)</sup>	1	1	1	1	1	1	1
Suplhur	3	3	3	3	3	3	3
Carbon black	-	10	20	25	30	35	50
Silica sand	50	40	30	25	20	15	-

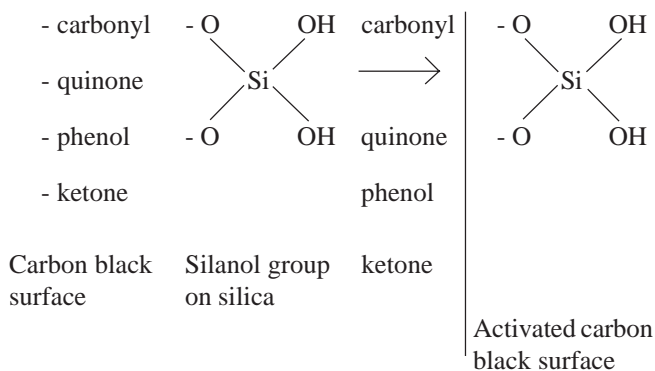
(a) = Mercaptobenzothiazole (accelerator)

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curing and the determination of mechanical and cure properties have been described by Adeosun (2000).

The mechanical properties (tensile strength, modulus and elongation at break, hardness, resilience and hysteresis) and cure properties (scorch time, cure time, torque rise, cure rate index, crosslink density and reversion resistance) have been determined and are presented in Figs. 1-5.

It is observed that the composite loaded (50 pphr) with silica sand showed a strength of 1.14KNm<sup>-2</sup> which improved tremendously on the replacement of 10pphr with carbon black. (Fig 1). The tensile strength increased progressively with increasing carbon black content in the rubber composite matrix. It has been known for some time that the use of silica in rubber compounds is problematic mainly due to the presence of siloxane and silanol groups on the surface of the filler (Wolff *et al* 1994). The silanol or hydroxyl groups have been reported to be acidic in nature (Hair and Hertl 1970). The analysis of dry carbon black surface indicated the presence of a range of hydrocarbon groups, which are capable of reacting with other functional groups (Bras and Papirer 1979; Barbin and Rodgers 1994). The enhancement of tensile strength of silica sand by carbon black can therefore, be suggested to be as a result of reaction of such groups as carbonyl, lac quinone, phenol and ketone on the carbon black surface with the silanol and hydroxyl groups of the silica sand leading to the formation of covalent bond with the carbon black.



In addition to this type of reaction, Le Bras (1979) has demonstrated that carbonyl, phenolic, quinone and other functional groups on the carbon black surface react with the natural rubber polymer. He provided evidence that chemical crosslinks exist between these materials in vulcanizates. Such chemical crosslinks can probably contribute to the enhancement of the energy properties of the composite such as tensile strength and modulus.

The modulus of the composite loaded with silica sand only showed a magnitude of 0.73KNm<sup>-2</sup> which is more than tripled

on the replacement of 10 pphr of silica sand with carbon black (Fig 1). Modulus increases with increasing carbon black content in the composite mix. The explanation for this enhancement could be found in the suggested reactions between carbon black/silica-sand and carbon black/polymer within the composite matrix. The elongation at break ( $E_B$ ) generally decreased with increasing carbon black content in the mix. This is expected because the brittleness of the composites is known to increase with decreasing strength.

The hardness of the composites loaded with mixed filler (silica sand/carbon black) showed a relatively superior values to that of the composite loaded singly with silica sand (Fig 2). The resilience of these composites (loaded with mixed fillers) however showed inferiority to that of the composite loaded singly with silica sand (Fig 2). The hysteresis also showed higher magnitude for the mixed fillers compared to

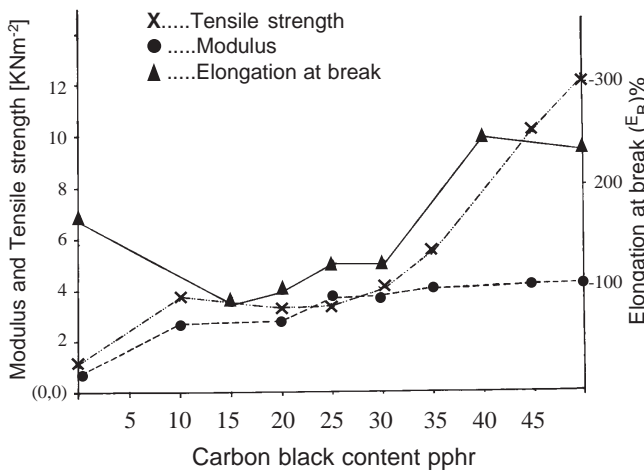


Fig 1. Plots of Tensile strength, modulus and elongation at break versus filler content in the mix.

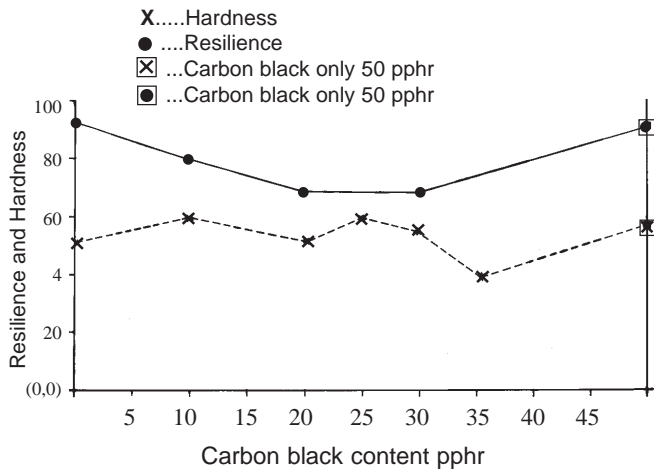


Fig 2. Plot of resilience and hardness versus carbon black content in the mix.

silica sand within the concentration range of fillers used in the present work (Fig 3).

The scorch time which is the time between the beginning of the heat cycle to the time curing begins, measures the processing safety period of the composite. The scorch time (Fig 4) of the composite loaded with silica sand only showed a magnitude of 28.8sec. Which decreased on the replacement of 10pphr of silica sand with carbon black. The scorch times of composites loaded with the mixed fillers showed comparable values similar to that composite loaded with carbon black alone. The present results indicate that the composite loaded with carbon black alone and the mixed fillers have inferior processing safety period compared to the composite loaded with silica sand alone. The cure time (Fig 4) followed the same trend as the scorch time but with different implication. The composites loaded with mixed fillers cures faster than the composites loaded singly with silica sand or carbon black. The composite loaded with carbon black has a faster curing than the composite loaded with silica sand. Long cure times and slow cure tares have been reported for silica

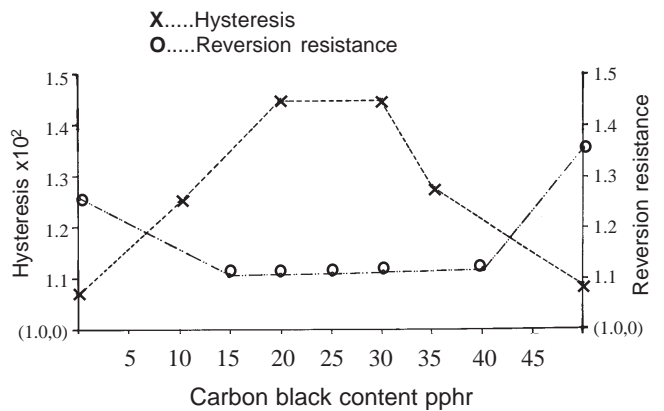


Fig 3. Plot of hysteresis and reversion resistance versus carbon black content in the mix.

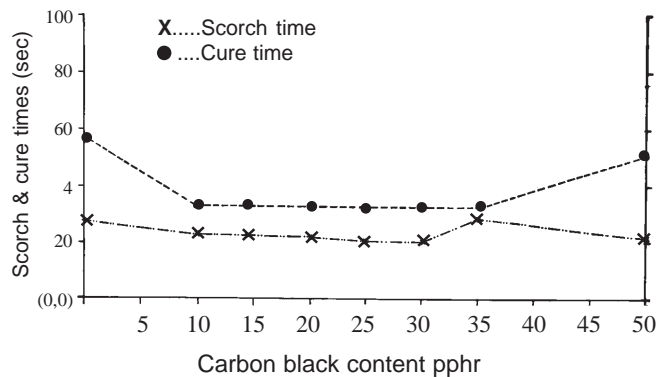
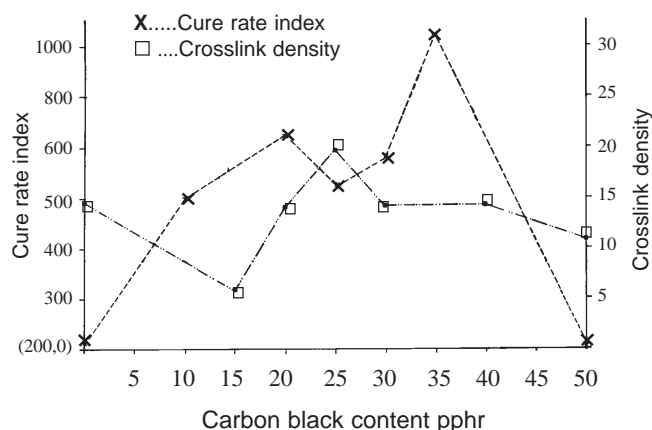


Fig 4. Plots of cure and scorch times versus carbon black content in the mix.



**Fig 5.** Plots of cure rate index and crosslink density versus carbon black content in the mix.

loaded rubber compound (Wolff *et al* 1994). This observed behaviour has been explained to be due to the strong tendency of the silica filler, (which is polar and hydrophilic) to absorb moisture (Hockley and Pethica 1961; Basset *et al* 1968). The amount of water adsorbed on the surface of the filler controls the ionization of the hydroxyl groups (Wagner 1976), resulting in detrimental effects on the curing attributes of compounds.

The cure rate index of the composites loaded with silica sand or carbon black showed slow cure rates compared to the composites loaded with mixed silica sand/carbon black (Fig 5). The reason adduced for the long cure time of silica sand explains why the cure rate of composite loaded with silica sand is slow. The crosslink density (Fig 5) of the composite loaded with carbon black or with mixed silica sand/carbon black showed superiority (except the composite filler of ratio 3:7) to that loaded with silica sand alone. This might be due to the presence of carbon atoms in carbon black filler, which would be made readily available for crosslink purposes. The reversion resistance decreased on the replacement of 10pphr of silica sand with carbon black as depicted in Fig 3, remained fairly unaffected by further replacement of silica sand with carbon black.

Results revealed that within the filler content range used in the present work, the hardness, modulus, and tensile strength of composites loaded with silica sand/carbon black showed enhanced magnitude over the composite loaded singly with silica sand. These parameters generally increased with increasing carbon black content in the composite. New area of use requiring moderate level of tensile strength, hardness and

modulus (as in soles of shoes and engine mounts) is therefore opened up for silica sand.

**Key words:** Silica sand, Carbon black, Cure properties, Mechanical properties, Natural rubber composite, Filler loading.

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