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Improvements in the degree of filler dispersion and tensile properties of N550 and N220 carbon blacks-filled natural rubber composites using alkanolamide

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Abstract. Effects of incorporation of ALK – alkanolamide - on the degree of carbon black (CB) dispersion and tensile properties of two kinds of CBs-filled natural rubber (NR) composites were examined. The N550 and N220 kinds of CBs were utilized, each of CBs was separately incorporated to composites with a certain incorporation, thirty parts per-hundred rubber (phr). The ALK was laboratory prepared and added into the composites at 3, 5, 7 and 9 phr. From the overall results, ALK was an internal plasticiser for the composites. It enhanced degree of filler dispersion by decrease the composites viscosity. The higher ALK loading caused in a better degree of filler dispersion. At a similar ALK incorporation, degree of N220 dispersion was poorer compared to N550. ALK also increased torque difference, tensile strength and also tensile moduli up to optimum incorporations. A five phr of ALK was the optimum incorporations for N550 and three phr was for N220. The ALK enhanced elongation at break of the composites. The higher ALK incorporation, the higher were elongation at break. At a similar ALK incorporation, elongation at break of N220 was longer compared to N550.

1. Introduction

The reinforcing fillers are used in achieving rubber composites with a sufficient level of mechanical properties [1]. The silica and CB-carbon black are the very well-known fillers in rubber reinforcement. CB has used for manufacturing black-coloured composites of rubber, whilst silica is used for manufacturing coloured composites. Both of them also can be used in a consolidation form for the aim of gaining their symbiotic contribution in order to manufacture higher complete mechanical properties [2].

In the activity of production of many rubber composites, both silica and CB are utilized to reinforce the rubber composites [3-4]. Nevertheless, at a relatively higher filler addition, filler particles of them are likely to form bigger agglomeration and poorer the filler dispersion that will degrade properties of the composites. Basically, to overcome the degree of filler dispersion as the root of the problem, some certain additives such as dispersant and processing aids are added during the making of composites.

The ALK - alkanolamide [5] - was suggested to overcome the problem. Due to its oily nature, it was used as an internal plasticiser in the compounding processes of some rubbers composites [5-7]. In this study, the ALK was compounded with natural rubber (NR), CB and others rubber chemicals to achieve CB-filled NR composites with ALK. Effects of ALK on CBs dispersion and also properties of tensile of CBs-filled NR composites were checked into thoroughly. Two kinds of CBs, N550 and



N220, were utilized as reinforcing fillers. They are dissimilar in the size of particle. N550 has a bigger particle size compared to N550 [8].

2. Experimental

2.1. Materials for the research

The raw NR was prepared by Guthrie (M) Sdn. Bhd., Malaysia. N220 and N550 kinds of CB were bought from Cabot Corporation. The others chemicals such as sulphur, IPPD antioxidant, stearic acid, ZnO and MBTS accelerator were prepared by Bayer Company, Malaysia. The ALK - $\text{CH}_3(\text{CH}_2)_{14}\text{CON}(\text{CH}_2\text{CH}_2\text{OH})_2$ - was made in the laboratory using RBDPS and diethanolamine [5].

2.2. Compounding for the making of composites

The semi efficient vulcanization was utilized in making of composites. Recipe for making the composites is presented in Table 1. The procedures of composites making were done according to ASTM D3184-80 and the composites were made on a lab scale mill.

Table 1. The recipe for the making of CBs-filled NR composites.

Materials	Composition, phr
NR	100
IPPD	2
Stearic acid	2
ZnO	5
S	1.5
MBTS	1.5
CB (N550/N220)	30
ALK	0; 3; 5; 7; 9

2.3. Rheometric behaviour

Scorch time, cure time, maximum and minimum torques and also torque differences of CB-filled NR composites with and with no ALK were determined utilising an MDR2000 (Monsanto Moving Die Rheometer) ASTM, D2084-11. The temperature for the testing was 150 °C.

2.4. Tensile properties

Certain numbers of dumbbell formed composites were cut from composites sheets (ISO 37). The tensile properties measurements were done at 500 mm.min⁻¹ of speed and they were performed on an Instron 3366 tensile meter to determine the tensile moduli, M300, M100; tensile strength, TS; elongations at break, EB.

3. Results and Discussion

3.1. The properties of torques

From Table 2, the additions of three phr of ALK reduced min torques of both control composites. The min torque is viscosity of a composite [9]. A lower M_L value is a lower viscosity. The incorporation of ALK lowered viscosity of the composites. It was because of an additional usage of ALK as a plasticizing agent that plasticized, softened and reduced viscosity, respectively. The increases the ALK loading causing in a lower viscosity of the composites.

Table 2 also presents that each three phr the incorporation of ALK causing increases in torque difference values for both control composites. The N550 and N220 have dissimilar behaviours in torque difference by further increases ALK incorporation. Difference in torque of N550 was further enhanced by the ALK incorporation up to five phr and reduced after the incorporation. The difference in torque of N220 was enhanced by the ALK incorporation at three phr and started to reduce after the

incorporation. The difference in torque is degree of crosslinking density of a composite [5-6, 10]. The greater in value is the higher in crosslinking density. The increasing in torque difference or crosslinking density up to optimum ALK incorporations (five phr for N550; three phr for N220) was because of physical and chemical ALK natures. The nitrogen of ALK activated chemically rubber and elemental sulfur when vulcanization. The combination of others curatives made the amine created complexes that attached sulfur to NR with more sufficient [8], caused higher cure degree [8].

Table 2. The properties of torque of N550/N220-filled NR composites

NR Composites	ALK incorporation (phr)				
	0 (Control)	3	5	7	9
CB N550					
Maxim. torque in dN.m	6.66	8.40	8.66	8.56	8.18
Minim. torque in dN.m	0.33	0.30	0.16	0.14	0.12
(Maxim. – Minim.) in dN.m	6.33	8.10	8.50	8.42	8.06
CB N220					
Maxim. torque in dN.m	8.42	11.95	9.80	9.52	9.42
Minim. torque in dN.m	0.39	0.37	0.33	0.32	0.29
(Maxim. – Minim.) in dN.m	8.03	11.58	9.47	9.20	9.13

Basically, the oily ALK has the usage as a plasticizing material. Discussed previously, ALK lowered the composites viscosity. It enhanced dispersion degree and CB to NR interaction, respectively. The CB to NR interaction is additional physical crosslinking [11], together with sulphide crosslinking are total crosslinking density [7, 12].

The decreases in torque differences, after the optimum incorporations, were because of the dilution/more amount effects of ALK.

3.2. The degree of CB dispersion

Using Equation (1), torques properties in Table 2 can be elaborated in calculating degree of filler dispersion in NR composites [13-14].

$$L = [M_{Lf} / M_{Lg}] - [M_{Hf} / M_{Hg}] \quad (1)$$

In which, M_{Lf} and M_{Hf} are min and max torques of filled composites. The M_{Lg} and M_{Hg} are the min and the max torques of unfilled one. A lower L is a better filler dispersion. The max and min torques of unfilled NR compound were 4.85 and 0.05.

Figure 1 presents L values for N220/N500 dispersion in NR at different ALK incorporation. The values of L of CB-filled composites with ALK were lower compared to with no ALK. The higher ALK tend to decrease L value. It was because of the ALK plasticization effect that lowered min torque/viscosity of composites and consequently, enhanced degree of CBs dispersion.

At a similar ALK incorporation, The value of L of N550 was lower compared to N220. It was because of the sizes of CBs particles. Decreasing in particle-size can cause decreasing in filler dispersibility [15]. The N220 has a smaller size compared to N550 and hence, its smaller particles caused the decreases in degree of dispersibility.

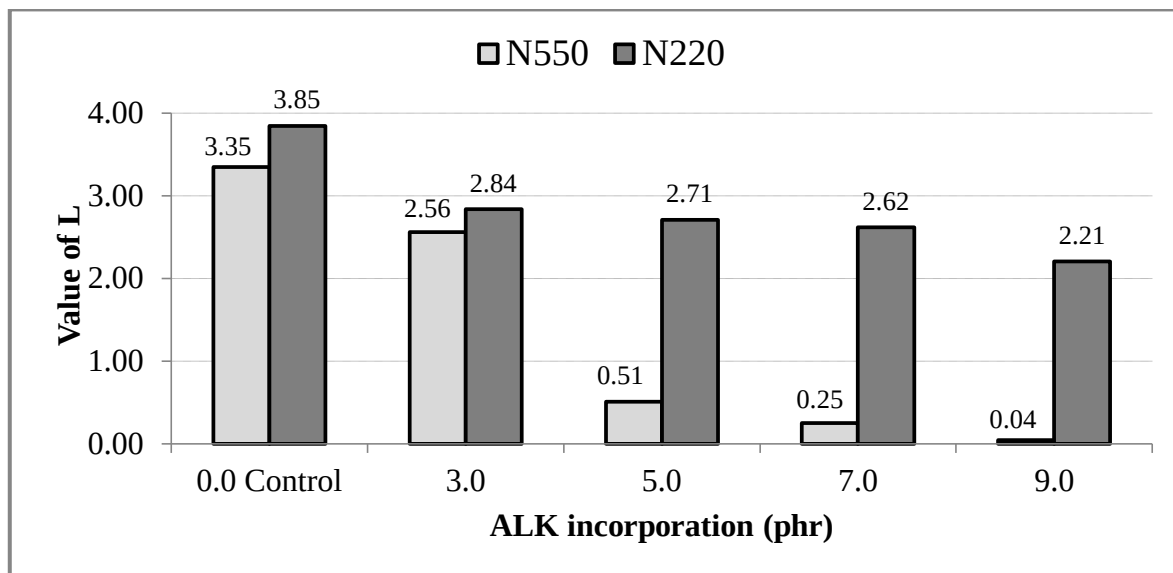


Figure 1. Effect of ALK incorporation on value of L of the carbon blacks-filled NR composites.

3.3. Tensile properties

The effects of ALK loading on tensile properties of N550 and N220-filled NR composites are presented in Table 3. The ALK incorporation up to five phr for N550 composites; whilst at one phr for N220 composite improved M100, M300 and TS into maximum levels and after those loadings displayed decreasing trends. The tensile moduli of a composite are only dependent on degree of crosslinking [16-19]. Enhancements in tensile moduli up to optimum incorporations were simply due to a higher crosslinking density; deteriorations in those properties after optimum incorporations were because of a lower crosslinking density. It was line with difference in torque data (Table 2).

At a similar ALK incorporation, tensile moduli (M100 and M300) of N220 were higher compared to N550. It was because of the crosslink density of N220 composites were higher compared to N550 composites.

From Table 3, ALK incorporations up to five phr, for N550 composites; whilst one phr for N220 composite increased the TS, after those incorporations displayed a reduction of TS.

The TS enhancement up to optimum incorporations was due to a better degree of CBs dispersion. It is in line with the result in Fig. 1. The deterioration in TS after optimum incorporations was due to the more amount of ALK.

At a similar ALK incorporation, TS of N220 was higher compared to N550. It was because of some reasons including smaller particle size and higher crosslink density.

Table 3. The properties of tensile of N550/N220-filled NR composites

NR Composites	ALK incorporation (phr)				
	0 (Control)	3	5	7	9
CB N550					
M100, MPa	1.193	1.386	1.442	1.248	1.209
M300, MPa	4.805	5.163	5.397	4.284	4.087
TS, MPa	19.8	22.2	26.3	24.6	23.8
EB, %	625.0	701.7	751.6	766.7	810.0
CB N220					
M100, MPa	1.276	1.579	1.486	1.318	1.237
M300, MPa	5.220	5.744	5.409	4.652	4.315
TS, MPa	20.2	28.8	27.2	26.4	24.7
EB, %	716.7	758.3	828.3	841.7	878.3

From Table 3, the ALK extent the EB to both CBs-filled NR composites. The EB was increased by increasing the ALK incorporations. It was because of the usage of ALK a plasticizing agent that modified flexibility of NR composites. ALK offered some free volumes that allowing more flexibility for the NR molecules to move. The higher ALK incorporation causing broader in the free volumes and hence, the more flexible the NR molecules.

At a similar ALK incorporation, the EB of N220 was higher compared to N550. Since a greater rubber to filler interaction offers higher reinforcement efficiency [20]; the higher EB of N220 was because of a higher reinforcement efficiency of N220 compared to N550. A higher reinforcing efficiency tends to keep stable the strain-induced crystallization of NR which delays the strain to break of the NR composite until a relative higher strain is got.

4. Conclusions

Alkanolamide enhanced torque difference, tensile modulus and tensile strength of carbon blacks-filled natural rubber composites up to optimum incorporations. A five phr was the optimum incorporation of alkanolamide for N550, and three phr was for N220.

Alkanolamide also enhanced degrees of filler dispersion of N550 and N220 in natural rubber composites. Increases alkanolamide incorporation caused further increasing in the degree of carbon blacks dispersion. At similar alkanolamide incorporation, the degree of N220 dispersion was poorer compared to N550.

Alkanolamide enhanced elongation at break of carbon blacks-filled natural rubber composites. The higher alkanolamide incorporation, the higher were elongations at break. At similar alkanolamide incorporation, elongation at break of N550 was lower compared to N220.

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