J nat Rubb Res **8(3)**, 190-207

Effects of Carbon Black Distribution on the Physical Properties of a NR/BR Tread Compound

SEE TOH MOOK SANG*

Two different carbon black masterbatch blending procedures were used to achieve a wide range of black distribution The black distribution and blending procedures influenced the physical properties significantly However, the effect of black distribution on blend properties is not always monotonic

The procedures do not always produce significant improvements in the physical properties over the conventional blending method The proper choice of blending technique and black distribution is required to achieve maximum benefits

The effects of carbon black loading on rubbers are not the same for all rubbers Different physical properties have different optimum black loadings. In blends of rubbers, these effects are more complex In conventional mixing of rubber blends, the black has been shown to preferentially locate in a particular rubber phase[1 2 3] Another important feature of blends is the rubber phase configuration Depending on the rubber blend ratio[4], the rubber phases may consist of a discrete and a continuous phase or both the phases are continuous

There were many studies to optimise the performance of blends by preferentially locating the black in a selected rubber phase. The results from different studies were not always in agreement The differences are believed to be due to the differences in materials used or the mixing procedures or both These result in different amounts of carbon black bound rubbers and degree of dispersion[4 8].

This work investigates the effects of carbon black distribution on some of the physical properties of NR/BR tread compounds prepared by two mixing procedures The effects of masterbatch viscosities were also studied.

EXPERIMENTAL

The formulation of the NR/BR compound is shown in *Table 1* This formulation is a typical truck tyre tread compound, obtained by two mixing techniques

TABLE 1 NR/BR TYRE TREAD COMPOUND

Formulation	Tread compound
SMR 20	65
High *cis* BR	35
Carbon black N110	55
Process oil[a]	15
ZnO	4
Stearic acid	2
Antidegradant[b]	2
MBS	0 8
Sulphur	2 2
	181 0

[a]Aromatic oil, *e g* Dutrex 729
[b]Paraphenylene diamine *e g* Santoflex 13

Conventional (CONV) Mixing

The conventional mixing cycle involved mixing of the NR/BR masterbatch in a BR Banbury as follows:

*Rubber Research Institute of Malaysia, P O Box 10150 50908 Kuala Lumpur, Malaysia
Present address Sime Darby Tyre Technology Centre 4 Jalan Tandang, 46500 Petaling Jaya, Selangor, Malaysia

	Time	Operation
1.	0 min	Add NR and BR.
2.	0.5 min	Add ½ black.
3.	1.5 min	Add oil, ½ black, activators, antidegradant.
4.	2.5 min	Sweep.
5.	4.0 min	Dump.

The rotor speed was 116 r.p.m. and the starting temperature was 60°C.

The full compound was then prepared by adding the sulphur and accelerator to the masterbatch on a two-roll mill. The mill mixing cycle was as follows:

	Time	Operation
1.	0 min	Add masterbatch and band on roll.
2.	1 min	Adjust rolling bank.
3.	1.5 min	Add sulphur and accelerator.
4.	5.0 min	Make four ¾ cuts from each side.
5.	6.0 min	Cut and roll up stock. Pass rolled-up stock endwise through the mill six times.
6.	8.0 min	Sheet out.

Blending of NR and BR Black/Oil Masterbatches

The second method of preparation of the NR/BR compound involved initially the preparation of NR and BR masterbatches (M/B) separately. These masterbatches were then blended in correct proportions to achieve the full compound of the formulation. The mixing cycle of the NR and BR masterbatches was as follows:

	Time	Operation
1.	0 min	Add rubber.
2.	0.5 min	Add ½ black.
3.	1.5 min	Add oil, ½ black.
4.	2.5 min	Sweep.
5.	4.0 min	Dump.

The masterbatch viscosities were then determined and adjusted to the desired level by milling on a two-roll mill at 60°C before blending.

Two series of NR and BR masterbatches were prepared. These are given in *Tables 2* and *3*.

Tables 2A and *2B* show the compositions of *Series 1* of NR and BR masterbatches. Note that the black and oil compositions were varied.

Tables 3A and *3B* show the compositions of *Series 2* of NR and BR masterbatches. In this series, the level of oil added was fixed while the level of black was varied. Experiments

TABLE 2A. BR MASTERBATCH COMPOSITIONS: SERIES 1

Ingredient	BR masterbatch					
	1	2	3	4	5	6
BR	100	100	100	100	100	100
CB	110	94.3	78.6	62.9	47.1	31.4
Oil	42.9	42.9	42.9	28.6	14.3	0
Total weight	252.9	237.2	221.5	191.5	161.4	131.4
Weight for 35 BR	88.5	83.0	77.5	67.0	56.5	46.0

COPYRIGHT © MALAYSIAN RUBBER BOARD

TABLE 2B NR MASTERBATCH COMPOSITIONS SERIES 1

Ingredient	NR masterbatch					
	7	8	9	10	11	12
NR	100	100	100	100	100	100
CB	25 4	33 8	42.3	50 8	59 3	67 7
Oil	0	0	0	7 7	15 4	23 1
Total weight	125 4	133 8	142 3	158 5	174 7	190 8
Weight for 65 NR	81 5	87 0	92 5	103 0	113 5	124 0

TABLE 3A BR MASTERBATCH COMPOSITIONS SERIES 2

Ingredient	BR masterbatch					
	13	14	15	16	17	18
BR	100	100	100	100	100	100
CB	85	70	55	40	25	10
Oil	15	15	15	15	15	15
Total weight	200	185	170	155	140	125
Weight for 65 BR	70	64 75	59.50	54 25	49 00	43 75

TABLE 3B NR MASTERBATCH COMPOSITIONS SERIES 2

Ingredient	NR masterbatch					
	19	20	21	22	23	24
NR	100	100	100	100	100	100
CB	38 85	46 92	55 00	63 08	71 15	78 23
Oil	15	15	15	15	15	15
Total weight	153 85	161 92	170 00	178 08	186 15	194.23
Weight for 65 NR	100 00	105 25	110 50	115 75	121 00	126 26

involving compounds prepared as in *Series 2* had been studied by many workers.

The NR/BR masterbatches were prepared by blending the corresponding NR and BR masterbatches, activators and antidegradants in a BR Banbury. The mixing cycle was as follows:

Time	Operation
1. 0 min	Add masterbatches.
2. 0.5 min	Add activators, antidegradants.
3. 2.5 min	Sweep.
4 4.0 min	Dump.

The full compounds were then obtained by adding the sulphur and accelerator on the two-roll mill according to the cycle used in conventional mixing.

TABLE 4. FINAL COMPOUNDS

Final compound number	NR masterbatch			BR masterbatch			M/B viscosity ratio (NR/BR)
	No.	Wt.	VB	No.	Wt.	VB	
1/1	7	606.2	74	1	558.3	98	0.76
1/2	8	568.5	72.5	2	596.0	81	0.90
1/3	9	530.9	74.5	3	633.6	76	0.98
1/4	10	459.0	74.0	4	705.6	68	1.09
1/5	11	387.0	74.0	5	777.5	70	1.09
1/6	12	315.0	71.5	6	849.4	76	0.94
2/1	19	685.0	61.5	13	479.5	133	0.46
2/2	20	721.0	73.5	14	443.5	102	0.72
2/3	21	756.9	66.0	15	407.6	89.5	0.74
2/4	22	792.9	65.0	16	371.6	63.0	1.03
2/5	23	828.9	76.5	17	335.7	44.5	1.71
2/6	24	864.8	80.0	18	299.7	39.0	2.05

Table 4 shows the series of compounds obtained by blending NR and BR masterbatches.

Vulcanisate Preparation

Compounds were cured at 150°C for 20 min or 24 min depending on the *t*90 assessed by the Monsanto Rheometer model TM100. Samples for heat build-up were cured for 25 min.

RESULTS AND DISCUSSION

Parameters for the Carbon Black Distribution

Three parameters have been used to describe the black distribution:

- percentage black in any one rubber phase.
- parts per hundred black in any one rubber phase.
- the ratio of the parts per hundred black in the two phases (black distribution ratio).

The ratio parameter is useful and necessary where there are changes in the total black loadings and the rubber blend ratios. The first two parameters are similar. For the present work both of these can be used since the black loading and rubber ratio are constant. *Table 5* shows the three black distribution parameters for the compounds used in this work. In drawing up this table, it is assumed that the carbon black when incorporated with a rubber phase especially in a highly unsaturated rubber phase, is not transferred to other rubber phases, in the subsequent blending operation[3,4,5,9].

Compound Properties

Compounds in *Series 2* tended to give low masterbatch viscosity (VB) at low black levels, and high masterbatch viscosity at high black levels when the oil loading is constant. The VB in *Series 1* were in a reasonable narrow range as the oil levels were adjusted. The two series of compounds were expected to give different black-rubber structures and hence different blend properties.

The properties of the twelve compounds prepared by blending of NR and BR masterbatches are given in *Tables 6A* and *6B*. The abbreviations used in these tables and elsewhere are explained in *Appendix 1*.

Tables 7A and 7B give some statistics of the data. For *Series 1*, the differences [DIF (%)]

TABLE 5. BLACK DISTRIBUTION PARAMETERS

Vulcanisate		% CB in BR phase	BR (p.h.r.)	NR (p.h.r.)	CB (p.h.r.) BR CB (p.h.r.) NR
1/1	Series 1	70	110	25.4	4.33
1/2		60	94.3	33.8	2.78
1/3		50	78.6	42.3	1.85
1/4		40	62.9	50.8	1.24
1/5		30	47.1	59.2	0.80
1/6		20	31.4	67.7	0.46
2/1	Series 2	54.09	85.0	38.9	2.19
2/2		44.55	70	46.9	1.14
2/3		35.00	55	55.0	1.00
2/4		25.45	40	63.1	0.63
2/5		15.91	25	71.2	0.35
2/6		6.36	10	79.2	0.13

TABLE 6A. EXPERIMENTAL DATA

Vulc.	Series	BBR	BNR	VBR	VNR	BRAT	VRAT	VC	T5	T90	TS	EB
1	1	110.0	25.4	98.0	74.0	4.330	1.324	42.5	48.0	17.2	25.8	657
2	1	94.3	33.8	81.0	72.5	2.789	1.117	51.5	49.5	18.2	23.3	609
3	1	78.6	42.3	76.0	74.5	1.858	1.020	55.5	52.0	19.5	21.2	589
4	1	62.9	50.8	68.0	74.0	1.238	0.918	62.0	54.4	20.7	20.4	628
5	1	47.1	59.2	70.0	74.0	0.795	0.945	69.5	53.8	21.5	19.7	614
6	1	31.4	67.7	76.0	71.5	0.463	1.062	75.0	51.8	23.8	20.9	698
7	2	85.0	38.9	133.0	61.5	2.185	2.162	54.0	46.9	17.3	24.9	601
8	2	70.0	46.9	102.0	73.5	1.492	1.387	57.0	48.4	17.5	24.3	627
9	2	55.0	55.0	89.5	66.0	1.000	1.356	58.5	49.6	18.1	22.5	590
10	2	40.0	63.1	63.0	65.0	0.633	0.969	55.0	50.0	18.3	24.2	654
11	2	25.0	71.2	44.5	76.5	0.351	0.581	50.0	50.0	18.3	22.8	577
12	2	10.0	79.2	39.0	80.0	0.126	0.487	54.0	43.6	16.8	22.1	575

TABLE 6B. EXPERIMENTAL DATA

Vulc.	M100	M300	Hard	TT	DCT	CRT	RFAT	DIN	AKR	DRES	LRES	HBU
1	1.75	8.27	59.5	17.7	81.6	109.0	151	113	163	60.5	55.9	90.0
2	1.78	8.43	62.5	16.1	104.0	114.0	253	114	163	58.0	51.7	95.5
3	1.95	8.45	62.0	16.7	84.7	106.0	289	133	194	55.7	48.7	98.5
4	1.72	6.96	64.0	13.3	83.2	71.3	138	190	254	60.5	54.3	103.0
5	1.76	6.82	63.0	14.4	51.2	89.2	900	190	254	60.5	54.3	103.0
6	1.46	5.00	57.0	13.9	45.1	63.4	1100	259	291	65.5	61.2	113.5
7	1.80	8.32	61.5	19.9	83.3	109.0	125	116	123	60.4	54.7	89.5
8	1.93	8.57	63.0	19.0	88.9	101.0	152	121	119	59.7	53.0	91.5
9	1.94	8.59	64.0	22.1	104.0	105.0	234	120	126	57.8	52.5	92.0
10	1.79	7.80	61.5	24.8	90.2	101.0	287	111	148	57.6	52.2	91.5
11	1.92	8.79	62.5	21.4	88.3	94.8	215	109	130	56.8	49.8	91.5
12	0.90	8.72	63.0	15.2	88.3	107.0	367	104	130	59.9	48.3	96.0

TABLE 7A. SOME STATISTICS OF RESULTS – SERIES 1

Property	Mean	SD	Low	High	Dif (%)	LSD (%)	CONV
VC	59.25	12.028	42.50	75.00	54.8	3.0	60.0
t5	51.58	2.460	48.00	54.40	12.4	3.0	39.0
t90	20.15	2.3882	17.20	23.80	32.8	3.0	18.0
TS	21.88	2.269	19.70	25.80	27.9	5.0	24.4
EB	632.5	39.24	589.0	698.0	17.2	5.0	640.0
M100	1.737	0.15807	1.460	1.950	28.2	8.0	1.93
M300	7.337	1.3699	5.000	8.540	48.2	10.0	8.73
IRHD	61.33	2.6013	57.00	64.00	11.4	3.0	64.5
TT	15.35	1.739	13.30	17.70	28.7	20.0	23.3
DCT	74.97	22.389	45.10	104.0	78.6	10.0	92.1
CRT	92.1	21.086	63.40	114.0	54.9	8.0	105.0
DIN	158.0	58.872	113.0	259.0	92.4	2.5	139.0
AKR	213.2	51.24	163.0	291.0	60.0	5.0	152.0
DRES	59.62	3.424	55.70	65.50	16.4	4.0	60.6
LRES	53.80	4.420	48.70	62.20	23.2	4.0	53.1
RFAT	471.8	418.00	138.0	1100.0	204.0	13.0	194.0
HBU	100.08	7.921	90.00	113.50	23.5	8.0	97.0

TABLE 7B. SOME STATISTICS OF RESULTS – SERIES 2

Property	Mean	SD	Low	High	Dif (%)	LSD (%)	CONV
VC	54.75	2.928	50.00	58.50	15.5	3.0	60.0
t5	48.08	2.500	43.60	50.00	13.3	3.0	39.0
t90	17.72	0.6145	16.80	18.30	8.47	3.0	18.0
TS	23.47	1.143	22.10	24.90	11.9	5.0	24.4
EB	604 .0	31.00	575.0	654.0	13.1	5.0	640.0
M100	1.880	0.06723	1.790	1.940	7.98	8.0	1.93
M300	8.465	0.3635	7.800	8.790	11.7	10.0	8.73
IRHD	62.58	0.9704	61.50	64.00	3.99	3.0	64.5
TT	20.40	3.240	15.20	24.80	47.1	20.0	23.3
DCT	90.50	7.020	83.30	104.0	22.9	10.0	92.1
CRT	103.0	5.123	94.80	109.0	13.8	8.0	105.0
DIN	115.2	4.792	109.0	121.0	10.4	2.5	139.0
AKR	129.3	10.07	119.0	148.0	22.4	5.0	152.0
DRES	58.20	1.501	56.80	60.40	6.19	4.0	60.6
LRES	51.75	2.312	48.30	54.70	12.4	4.0	53.1
RFAT	230.0	88.73	125.0	367.0	105.0	13.0	194.0
HBU	92.00	1.145	89.50	96.00	7.07	8.0	97.0

Dif(%) = (High – Low) / Mean * 100
LSD(%) = Least significant difference (% of mean) of test method
CONV = Results from conventional blend mixing

between the highest and lowest values are significantly greater than the testing errors [LSD (%)]. For *Series 2*, the differences for M100, M300, hardness and HBU are not significant. The standard deviations (SD) in *Series 1* are generally larger than in *Series 2* indicating a greater spread of data in *Series 1*.

For most of the properties measured, the value from the conventional blending technique (CONV), is within the range of values obtained by blending of M/B for *Series 1* and *2* except in the following properties

- Mooney scorch (CONV lower in both cases)
- Trouser tear strength (CONV higher than *Series 1*)
- DIN abrasion index (CONV higher than *Series 2*)
- AKRON abrasion index (CONV lower than *Series 1*)

To compare the effect of M/B blending to the conventional method, the differences between the data were calculated (*Table 8*) The interpretations are summarised in *Table 9* showing the general trend of results that can be achieved by M/B blending Significant improvements are seen in scorch, abrasion and fatigue Other properties are not significantly affected.

Graphical Analysis

The physical properties are plotted against:

- parts per hundred black in BR phase (BBR)
- black distribution ratio (BRAT)
- viscosity ratio of the M/B (VRAT),

The plots against parts per hundred black and black distribution ratio are similar, and generally the parts per hundred black plots show better correlation, suggesting that the parts per hundred black may be a better black distribution parameter to use The plots show

TABLE 8 DIFFERENCES BETWEEN RESULTS FROM CONVENTIONAL BLENDING AND MASTERBATCH BLENDING

Property	Series 1		Series 2	
	High (%)	Low (%)	High (%)	Low (%)
VC	25 0	–29	–2 5*	–16 7
t5	39 5	23	28 2	11 8
t90	32 2	–4 4	1 7*	–6 7
TS	5 7	–19 3	2 0*	–9 4
EB	9 1	–8 0	2 2*	–10 2
M100	1 0*	–24 4	0 5*	–7 3*
M300	–2 2*	–42 7	0 7*	–10 7*
IRHD	–0 8*	–11 6	–0 8*	–4 7
TT	–24 0	–43 0	6 4*	–34 8
DCT	12 9	–51 9	12 9	–9 6*
CRT	8 6*	–39 6	–3 8*	–9 7
DIN	86 3	–18 7	–12 9	–21 6
AKR	91 4	7 2	–2 6*	–21 7
DRES	8 1	–8 1	–0 3*	–6 3
LRES	15 3	–8 3	3 0*	–9 0
RFAT	467 0	–28 9	89 2	–35 6
HBU	17 0	–7 2*	–1 0*	–7 7*

$$\text{High (\%)} = \frac{\text{High} - \text{Conv}}{\text{Conv}} \times 100\% \qquad \text{Low (\%)} = \frac{\text{Low} - \text{Conv}}{\text{Conv}} \times 100\%$$

High = Highest value from M/B blending
Low = Lowest value from M/B blending
Conv = Value from CONV blending
* = Not significant

TABLE 9. EFFECTS OF M/B BLENDING ON PROPERTIES COMPARED TO CONV BLENDING

Property	Series 1	Series 2
VC	Increase/Decrease	Decrease
t5	Increase	Increase
t90	Increase	Decrease
TS	Decrease	Decrease
EB	Increase/Decrease	Decrease
M100	Decrease	Unchange
M300	Decrease	Unchange
IRHD	Decrease	Decrease
TT	Decrease	Decrease
DCT	Decrease	Decrease
CRT	Decrease	Decrease
DIN	Increase/Decrease	Decrease
Akron	Increase	Decrease
DRES	Increase/Decrease	Decrease
LRES	Increase/Decrease	Decrease
RFAT	Increase	Increase
HBU	Increase	Unchange

that the data from *Series 1* and *Series 2* are different and should be analysed separately.

The plots against viscosity ratio show that this variable is generally less important, possibly because its range is narrow in *Series 1* and that *Series 2* itself generally causes less variation in the results.

Some of the plots are curves with maximum or minimum turning points. This means that in some properties the change is not monotonic with respect to the variables.

An example of the plots is shown in *Figure 1* for tensile strength. The plots for the other properties are summarised in *Figures 2a - 2d.*

Computer Analysis

The data were analysed by the statistical package GLIM 3.77 (1985). GLIM (Generalised Linear Interactive Modelling) is a special-purpose package primarily intended to facilitate the fitting and investigation of generalised linear models.

Preliminary studies have shown that the M/B blending procedure is a very important factor and the data should be analysed in two separate groups. The linear models consist of the black distribution parameter (BBR), viscosity ratio of the masterbatch (VRAT) and the square terms of these parameters where appropriate.

Significant models are selected and investigated further by plotting:

- the observed and fitted values against the variables in the models.
- the normal plot of ordered residuals.
- the residuals against the fitted values.

These are to ensure the assumptions in the models are not invalidated.

Tables 10A and *10B* summarise the significant models (in the form of response equations) for the various properties. In *Series 1*, fourteen properties were affected and in *Series 2*, eight properties. Black distribution is a significant factor in all these affected properties. Viscosity ratio is a significant factor in three properties in *Series 1* and *Series 2*.

The properties most affected by the M/B blending techniques were fatigue, Akron and DIN abrasion in *Series 1* and fatigue in *Series 2*.

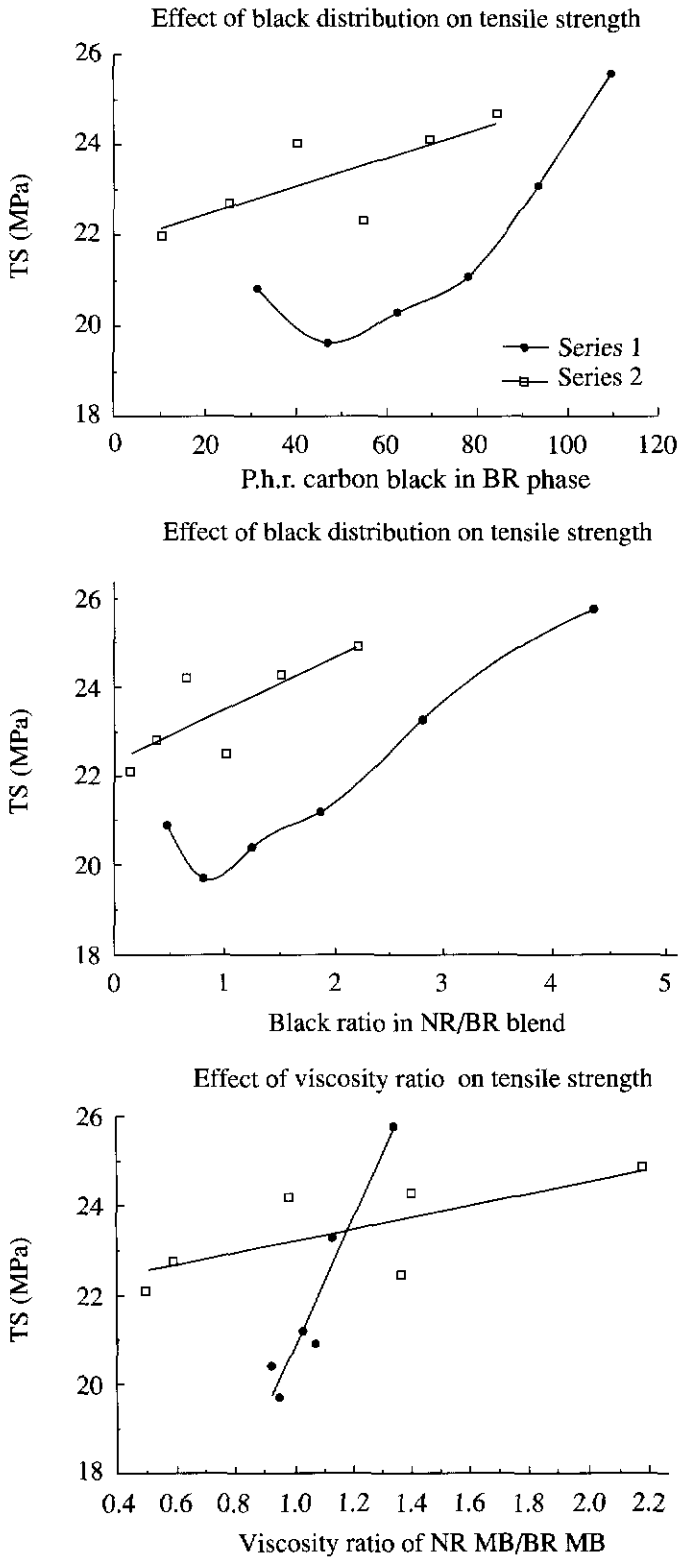

Figure 1. Tensile strength vs *blending variables.*

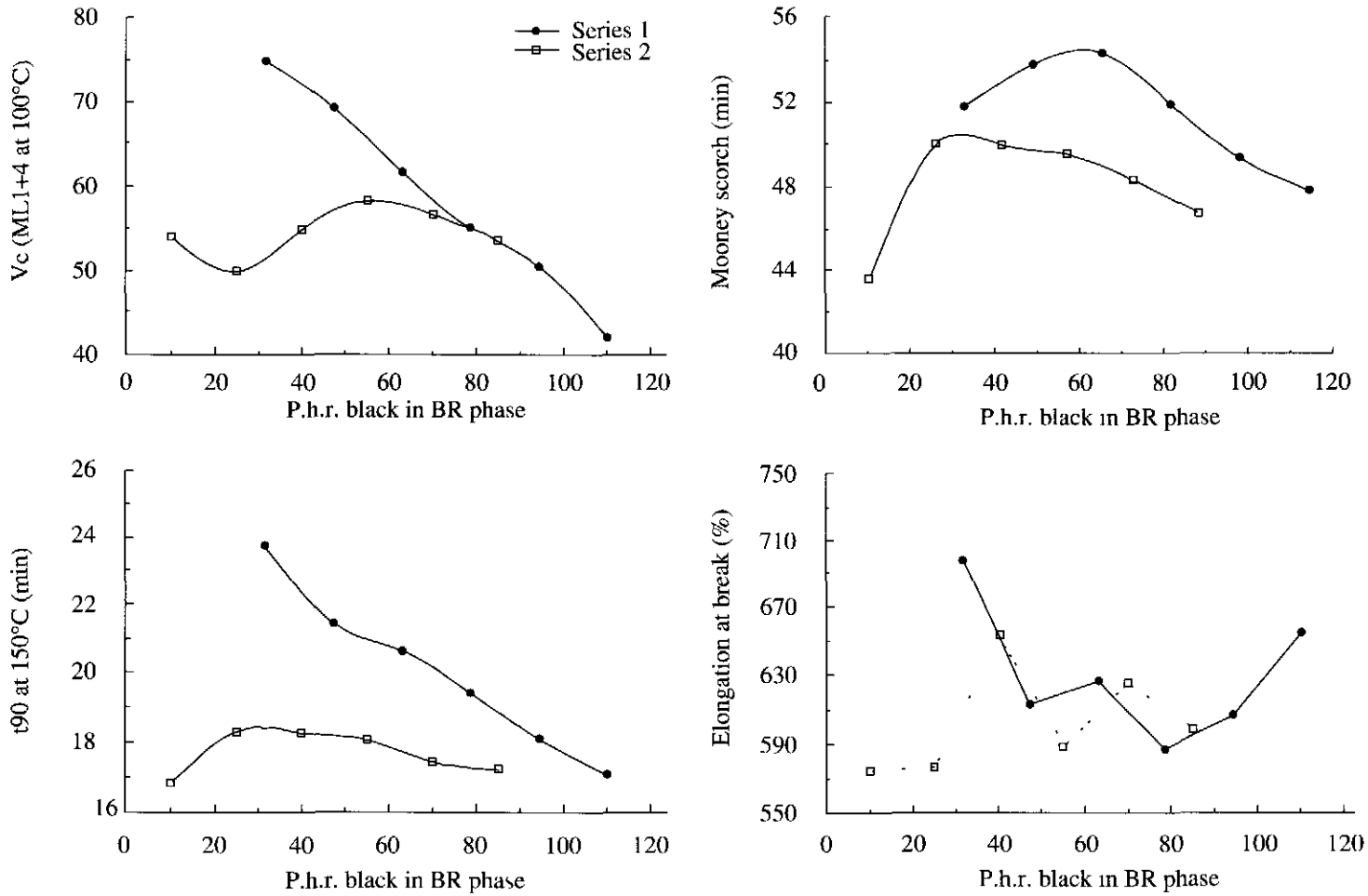

Figure 2a. Physical properties vs *black distribution.*

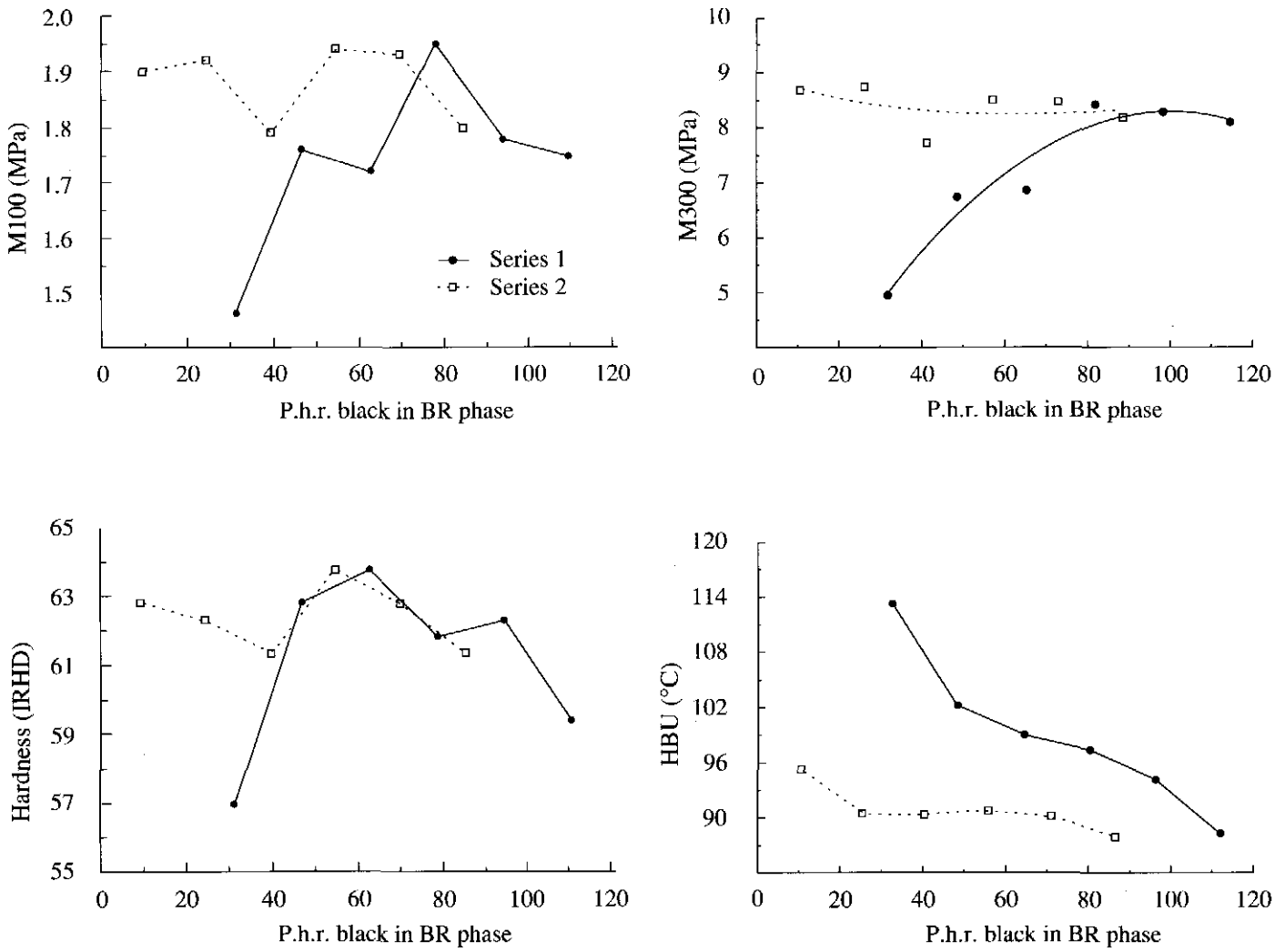

Figure 2b. Physical properties vs *black distribution.*

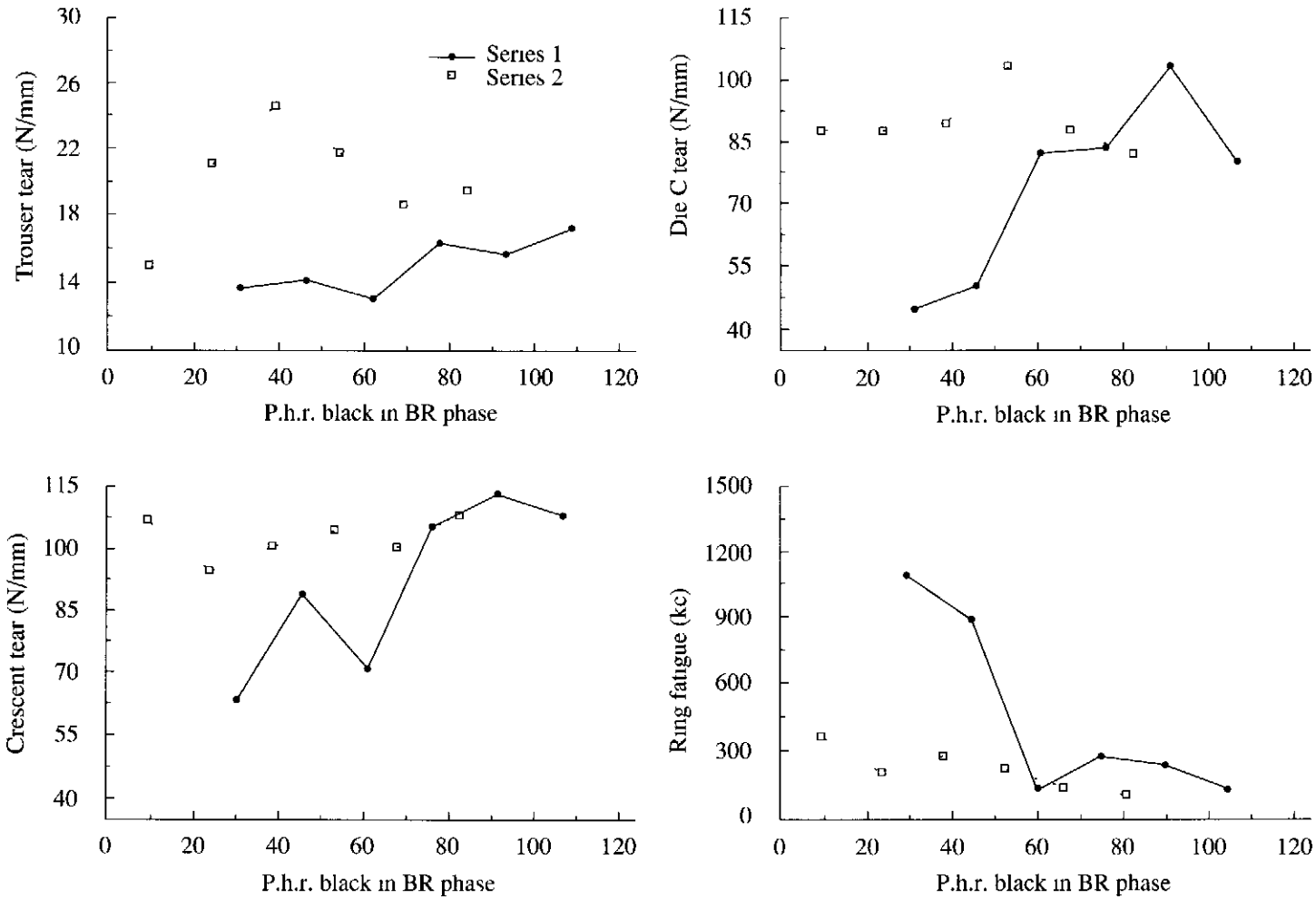

Figure 2c Physical properties vs black distribution

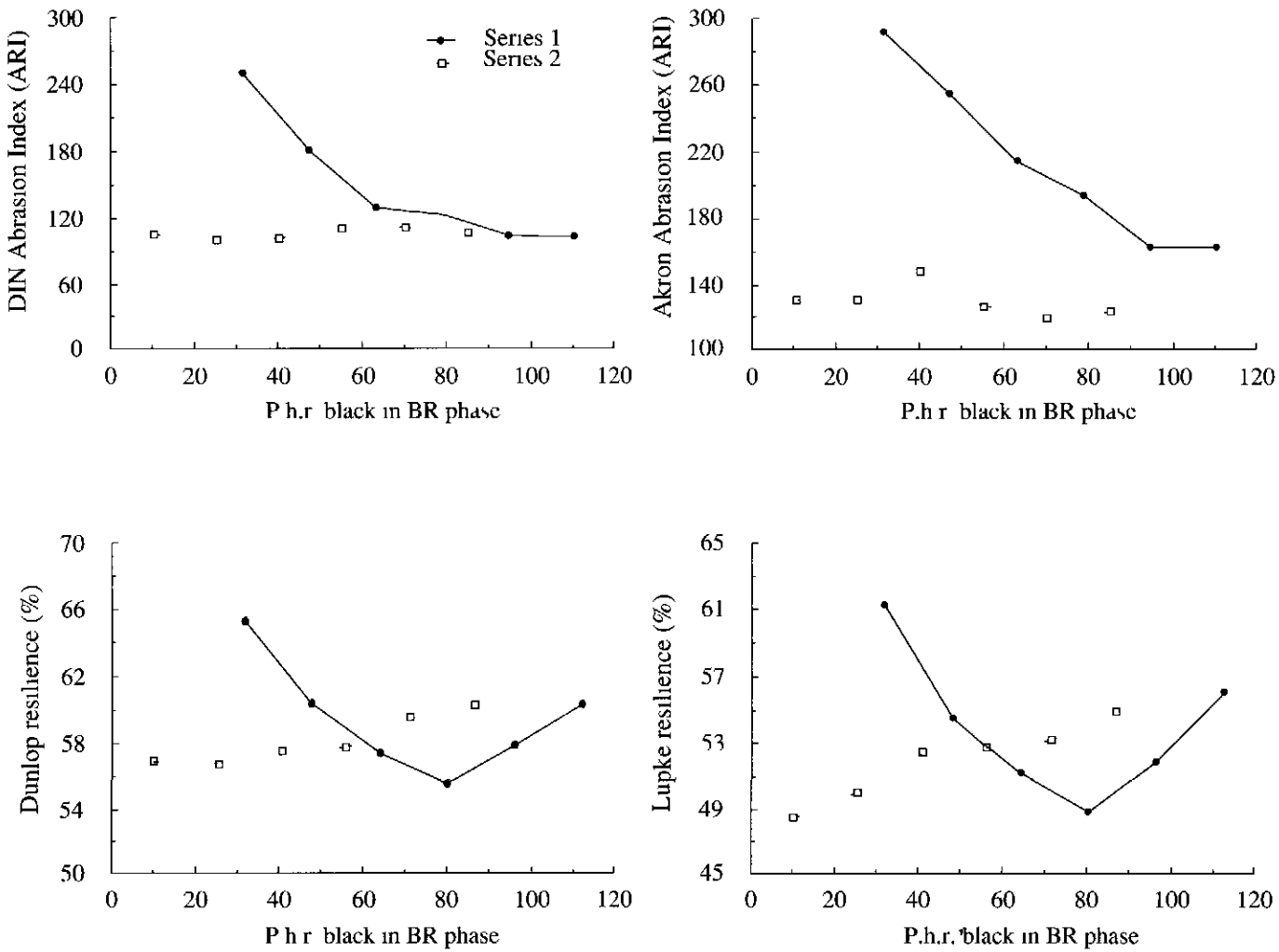

Figure 2d Physical properties vs black distribution

TABLE 10A RESPONSE EQUATIONS FOR EXPERIMENTAL PARAMETERS – SERIES 1

Response	Const	BBR	BBR2	VRAT	VRA2
V_C (ML 1+4)	88 10	–0 4080	–	–	–
Scorch (min)	107 60	–	–0 0002	–88 18	33 68
Op cure (min)	25 82	–0 0801	–	–	–
TS (MPa)	24 70	–0 1809	–0 0017	–	–
M300 (MPa)	1 050	0 1525	–0 0008	–	–
TT (N/mm)	13 30	–	0 0004	–	–
DCT (N/mm)	30 94	0 6225	–	–	–
CRT (N/mm)	48 84	0 6124	–	–	–
DIN (ARI)	97 25	–2 540	–	225 7	–
AKR (ARI)	248 10	–2 540	–	103 9	–
DRES (%)	82 16	–0 662	0 0042	–	–
LRES (%)	83 44	–0 8908	0 0058	–	–
RFAT (kc)	1312 12	–11 88	–	–	–
HBU (°C)	118 3	–0 2571	–	–	–

TABLE 10B RESPONSE EQUATIONS FOR EXPERIMENTAL PARAMETERS – SERIES 2

Response	Const	BBR	BBR2	VRAT	VRA2
Scorch (min)	46 82	0 7588	–0 0066	–24 63	7 861
Op cure (min)	17 53	0 1873	–0 0018	–6 01	2 137
TS (MPa)	21 95	0 0320	–	–	–
CRT (N/mm)	86 66	–1 512	0 0094	80 28	–19 44
DRES (%)	56 64	–	0 0005	–	–
LRES (%)	47 96	0 0798	–	–	–
RFAT (kc)	361 4	–2 766	–	–	–
HBU (°C)	94 90	–0 0610	–	–	–

BBR = p h r black in BR phase
VRAT = viscosity ratio (NR M/B)/(BR M/B)
BBR2 = BBR * BBR
VRA2 = VRAT * VRAT

In some models, square terms are included to account for curvatures and turning points (maximum or minimum). Published reports have not mentioned or highlighted these turning points Their general conclusions were that a particular physical property is better/worse with more black in a particular rubber phase These maximum/minimum turning points should not be a surprise, bearing in mind that the individual rubber shows this type of behaviour with increasing black loading Published works did not have the wide range of black distribution used in this work

Comparison with Some Published Data

In the literature, the reports on the effects of black distribution were based on many different formulations They differred in the individual rubbers, the rubber blend ratio, black type and loading, oil type and loading and cure systems The mixing techniques were different In some studies the black distribution was just one of the many variables Comparing results from such a wide range of conditions is difficult and the conclusions should be treated with caution.

Krakowski and Tinker[8] used a central composite design with four variables rubber blend ratio (BR/NR), black level (13-47 p h r N330), black distribution ratio and DPG levels The compounds contained 3 p h r Dutrex 729 They reported that the black distribution ratio was a significant factor for DIN (increase), M300 (increase) and trouser tear (decrease)

Hess and Chirico[6] used a 50/50 NR/BR blend and the black level was 60 p.h.r. N220 and 20 p.h.r. oil. The compounds contained 25% and 75% black in the BR phase. More black in the BR phase affected tear strength (reduced), fatigue (reduced), tensile strength (slight increase) and tread wear (slight increase).

Hess *et al.*[4] studied the effect of rubber blend ratio (NR/BR) and the black distribution. The black used was N229 (60 p.h.r.) and the black level in BR phase were 30, 60 and 90 p.h.r. with 25 p.h.r. aromatic oil. Increasing the level of black in the BR phase affected fatigue (reduced), resilience (reduced), HBU (increased) and tear strength (reduced).

Sircar *et al.*[5] studied the effects of black distribution in 50/50 NR/BR using 50 p.h.r. N347 black and 7.5 p.h.r. naphthenic oil. The amount of black in the BR phase were 0%, 50% and 100%. Increasing the level of black in the BR phase affected the following,properties: resilience (increase), TS (increase), EB (increase), M300 (lowest at 50%), HBU (highest at 50%) and Mooney viscosity (highest at 50%).

Panciroli[7] prepared blends of 50/50 NR/BR on the two-roll mill by adding the black (N220, 48 p.h.r.; Dutrex 50, 3 p.h.r.) separately on one polymer and then adding the other polymer to the batch. A further type of black distribution was realised by adding black to the pre-blended polymers. The black distribution for the first two mixes were 0% and 100% black in the BR, and for the pre-blend it was undefined.

The effects of increased level of black in BR on properties are as follows (the blend with the highest value for the property was ranked first):

VC (decrease, pre-blend > 0% > 100%)

Scorch (decrease, 0% > pre-blend > 100%)

M100 (increase, 100% > 0% > pre-blend)

M300 (increase, 100% > 0% > pre-blend)

TS (decrease, pre-blend > 0% > 100%)

EB (decrease, pre-blend) 0% > 100%)

Tear (decrease, pre-blend > 0% > 100%

DIN (decrease, pre-blend > 0% > 100%)

Resilience (increase, 100% > pre-blend > 0%)

HBU (decrease, pre-blend > 0% > 100%).

Table 11 shows a comparison of results from different sources. There was poor agreement in most properties except in tear strength and fatigue which decrease with increasing level of black in the BR phase. Results from *Series 1* was in agreement for fatigue but not for tear. But for *Series 2*, good agreement in both tear and fatigue was observed, taking into account the range of black distribution.

SUMMARY

The effects of carbon black distribution on the properties of a 65/35 NR/BR tyre tread compound were investigated using two masterbatch blending procedures. In *Series 1*, the black and oil levels in the M/B were varied and the M/B viscosities were in a narrow range. In *Series 2*, the black levels were varied with constant oil level in the M/B. The M/B viscosities were in a wide range. These two series are expected to give different black-rubber structures and hence different blend properties, depending on the black distribution and the viscosity ratio of the M/B.

M/B blending *Series 2* did not show any improvement over the conventional mixing method except for scorch and fatigue. *Series 1* can be used, for some properties, to get either higher or lower values than the conventional mixing method depending on the choice of black distribution. *Series 1* is better for scorch and Akron abrasion and worse for trouser tear. At low level of black in the BR, *Series 1* can produce large improvements in abrasion (DIN and Akron) and fatigue.

TABLE 11. EFFECTS OF BLACK DISTRIBUTION TAKEN FROM DIFFERENT SOURCES

Property	Increase p h r. black in BR phase						
	Series 1	Series 2	Krakowski and Tinker	Hess and Chirico[6]	Hess *et al*[4]	Sircar *et al.*[5]	Pancirolli[7]
VC	Dec	NS	NS	NA	NA	Max at 50%	Dec
Scorch	Max at 63 p.h.r	Max at 40 p.h.r	NS	NA	NA	NA	Dec
TS	Inc	Inc	NS	Inc	NS	Inc	Dec
EB	Min. at 78 p.h.r.	NS	NS	NA	NA	Inc	Dec
M100	Min. at 78 p.h.r.	NS	NS	NA	NA	NA	Inc
M300	Inc	NS	Inc	NS	NS	Min. at 50%	Inc
Trouser tear	Inc	Max at 40 p.h.r.	Dec	Dec	Dec	NA	Dec
Din/Wear	Dec	NS	Inc	Inc	NA	NA	Dec
Resilience	Min. at 78 p.h.r.	Inc	NS	NS	Dec	Inc	Inc
Fatigue	Dec	Dec	NA	Dec	Dec	NA	NA
HBU	Dec	NS	NA	NS	Inc	Max at 50%	Dec

Inc = Increase Dec = Decrease
NS = Not significant NA = Not reported
Units in Sircar[5] and Pancirolli[7] data were percentage carbon black in BR phase

Plotting the data shows clearly that the results from the two series are significantly different. The plots based on black distribution show patterns of correlation for fourteen physical properties in *Series 1* and eight properties in *Series 2*. The viscosity ratio has less influence on properties.

Response equations relating the physical properties to black distribution and viscosity ratio were obtained. Some equations contain square terms indicating curvatures. These curves with turning points mean that the effect of black distribution on properties is not monotonic. This has not been well highlighted in the literature.

Data from the literature have been examined and compared to those from this work. It is noted that data from different sources do not always agree. This has been attributed to differences in materials and/or mixing procedures. This work shows clearly the important effect of mixing procedures.

CONCLUSION

This work shows that using two different masterbatch blending procedures, to achieve a wide range of black distribution, produces significantly different results.

These procedures, however, do not always produce significant improvements in physical properties over the conventional mixing method. The proper choice of blending technique and black distribution is required to achieve maximum benefits.

Black distribution is shown to affect many blend properties but the effect is not always monotonic. Viscosity ratio of the masterbatches has less influence on properties.

ACKNOWLEDGEMENTS

This work was done in the Malaysian Rubber Producers' Research Association (MRPRA) where the author was on a one-year secondment

from the Rubber Research Institute of Malaysia (RRIM)/Malaysian Rubber Research Development Board (MRRDB) The writer thanks the MRPRA and RRIM/MRRDB for the opportunity to carry out this study, and Dr C Metherell for assistance on the GLIM programmes

Date of receipt February 1993
Date of acceptance August 1993

REFERENCES

1 WALTERS, M H AND KEYTE, D W (1962) Heterogeneous Structure in Blends of Rubber Polymers *Trans Inst Rubb Ind* **38,** T40

2 HESS, W M , SCOTT, C E AND CALLAN, J E (1967) Carbon Black Distribution in Elastomer Blends *Rubb Chem Technol* **40,** 371

3 CALLAN, J E , HESS, W M AND SCOTT, C E (1971) Elastomer Blends Compatibility and Relative Response to Fillers *Rubb Chem Technol* **44,** 814

4 HESS, W M , VEGVARI, P C AND SWOR, R A (1985) Carbon Black in NR/BR Blends for Truck Tires *Rubb Chem Technol* , **58,** 350

5 SIRCAR, A K , LAMOND, T G AND PINTER, P E (1974) Effect of Heterogeneous Carbon Black Distribution on the Properties of Polymer Blends *Rubb Chem Technol* , **47,** 48

6 HESS, W M AND CHIRICO, V E (1977) Elastomer Blend Properties Influence of Carbon Black Type and Location *Rubb Chem Technol* , **50(2),** 301

7 PANCIROLI, G Effect of Carbon Black Distribution in Binary Polymer Blends Technical Report C I N-02, Cabot Italiana S p A

8 KRAKOWSKI, F AND TINKER, A J (1990) Effect of Polymer Ratio, Carbon Black and Accelerator Level on NR/BR Blends *Elastomerics* June 1990, 31 Effect of Compounding Ingredient Levels on NR/BR Blends *Elastomerics* July 1990, 24

9 HESS W M SCOTT, C E AND CALLAN, J E (1971) Factors Affecting Filler Transfer in Rubber Blends *Rev Gen Caoutch Plast* , **48,** 155

APPENDIX 1

List of Abbreviations

AKR	Akron abrasion index (ARI)
BBR	P.h.r. black in BR
BNR	P.h.r. black in NR
BRAT	Black distribution ratio
CRT	Crescent tear strength (N/mm)
DCT	Die C tear strength (N/mm)
DIN	DIN abrasion index (ARI)
DRES	Dunlop tripsometer resilience (%)
EB	Elongation at break (%)
HARD	Hardness (IRHD)
HBU	Heat build-up (ΔT, °C)
LRES	Lupke pendulum resilience (%)
M100	Stress at 100% strain (MPa)
M300	Stress at 300% strain (MPa)
RFAT	Ring fatigue resistance (kc)
SERIES	Series 1 or 2
T5	Mooney scorch at 120°C (min)
T90	Rheometer optimum cure time at 150°C (min)
TS	Tensile strength (MPa)
TT	Trouser tear strength (N/mm)
VBR	Viccosity of basestock of BR masterbatch
VC	Compound Mooney viscosity (ML 1 + 4 at 100°C)
VNR	Viscosity of basestock of NR masterbatch
VRAT	Viscosity ratio of NR masterbatch to BR masterbatch