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Starved Extrusion for the Improvement of Mechanical Properties of Rubber Vulcanizates

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ABSTRACT: Filled compounds of natural rubber, isobutylene-isoprene rubber and styrene-butadiene rubber compounds were extruded through a laboratory extruder by varying the feeding rate at different temperatures and revolutions per minute. The extruded compounds were vulcanized up to their optimum cure times and the mechanical properties of the vulcanizates were determined. The properties suggest that there is a particular feeding rate in the starved fed region which results in maximum mechanical properties. The study shows that running the extruder at a slightly starved condition is an attractive means of improving the physical properties.

KEY WORDS: natural rubber, isobutylene-isoprene rubber, styrene-butadiene rubber, laboratory extruder, starved fed.

INTRODUCTION

STARVED FEEDING IS commonly employed for improving the operating versatility of single screw extruders [1]. For example the same screw can handle a wide range of materials and performance requirements often better utilizing horsepower availability [2]. Starved feeding of single screw extruders is regarded as a means of operating at a lower torque [3]

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achieving higher stock temperature and increased variability in output rate [4]. For a properly mixed rubber compound, starved feeding is likely to result in reduced mechanical breakdown and preferential orientation of the molecules which may give rise to improved mechanical properties. In this study we report the effect of starved feeding on the mechanical properties of filled NR, IIR and SBR compounds.

MATERIALS USED

Elastomers

1. Natural Rubber (NR): Mw = 7.70 × 105; Mooney viscosity, ML (1 + 4) at 100°C, 85.3; ISNR-5 supplied by Rubber Research Institute of India, Kottayam.
2. Isobutylene-Isoprene Rubber (IIR): Exxon 065, Mooney viscosity, ML (1 + 8) at 100°C, 50; supplied by Indian Petrochemicals Corporation Ltd.
3. Styrene-Butadiene Rubber (SBR): 23.5% styrene; Mooney viscosity, ML (1 + 4) at 100°C, 49.2; supplied by Synthetics and Chemicals Ltd., India.

Compounding Additives

1. Zinc oxides: supplied by M/s.Meta Zinc Ltd., Bombay
2. Stearic acid: supplied by Godrej Soaps (P) Ltd., Bombay
3. HAF (N-330): supplied by (CACIL, Kerala)
4. Aromatic oil: Commercial grade
5. Paraffinic oil: Commercial grade
6. *N*-cyclohexyl-2-benzothiazyl sulfenamide (CBS): supplied by Indian Explosives Ltd.
7. Dibenzythiazyl disulphide (MBTS): supplied by Bayer Chemicals, Bombay
8. Tetramethyl thiuram disulphide (TMTD) and pilfex-13: supplied by Polyolefins Industries Ltd., Bombay
9. Sulphur: supplied by Standard Chemicals Company (P) Ltd., Madras

EXPERIMENTAL

Studies on starved feeding were done on a laboratory extruder attached to a Brabender Plasticorder Model PL 2000 with an L/D ratio of 10 and a compression ratio of 1 and provided with a feeding roll. Three typi-

cal filled compounds, one based on NR, a second based on IIR and the third based on SBR (Table 1A) were selected for the study.

Compounds were prepared on a laboratory two roll mill as per ASTM D 3184 and 3189 (1973). The compounds were sheeted out by passing through a 1 mm nip of the mixing mill. The sheets were cut into 10 mm strips for feeding into the extruder. The feeding rates were adjusted by placing a different number of layers of the strips on the feeding roll and measured by the rate of output of the extrudate. The compounds were extruded at varying feeding rates mainly in the starved feeding region at 20, 40, 60 and 80 rpms and at different temperatures. The cure curves of the compounds before and after extrusion were taken on a Goettfert Elastograph model 67.85 as per ASTM D 1646 (1981) at 150°C for NR and SBR compounds and at 170°C for IIR compound. The extruded samples were vulcanized up to their optimum cure times (90% of the time required for attaining maximum torque) in an electrically heated laboratory hydraulic press. The moulded samples were then cooled by immersing in water and dumb-bell specimens were cut out of the sheets for tensile testing. The tensile properties of the vulcanizates were measured using a Zwick Universal Testing Machine of Model 1445 at an extension rate 500 mm/min as per ASTM D 412 (1980). The swelling index of the vulcanizates were measured by equilibrium swelling in toluene for NR and SBR and in benzene for IIR [5].

$$\text{Swelling index} = \frac{\text{final weight} - \text{deswollen weight}}{\text{initial weight}} \quad (1)$$

The percent bound rubber content (filler gel) of a few sets of samples of NR, IIR and SBR was determined by immersing the samples in 25 ml of toluene for seven days at room temperature (solvent was renewed after

Table 1A. Formulations of the compounds.

NR Compound		IIR Compound		SBR Compound	
NR	100	IIR	100	SBR	100
ZnO	5.0	ZnO	4.0	ZnO	4.0
Stearic acid	2.0	Stearic acid	2.0	Stearic acid	1.5
Antioxidant (Pilflex-13)	1.0	Antioxidant (Pilflex-13)	1.0	Antioxidant (Pilflex-13)	1.0
HAF black (N 330)	50	HAF black (N-330)	45	HAF black (N-330)	45
Aromatic oil	7.0	Paraffinic oil	7.5	Aromatic oil	6.0
MBTS	0.6	MBTS	0.5	MBTS	1.0
Sulfur	2.5	TMTD	1.0	TMTD	0.25
		Sulfur	1.5	CBS	0.8
				Sulfur	2.0

three days). Then the sample was dried for one day in air at room temperature and then for 24 hours in an oven at 105°C [6]. The percent bound rubber of the polymer, R_B was then calculated according to the following equation

$$R_B = \frac{w_{fg} - w[m_f/m_f + m_p]}{w[m_p/(m_f + m_p)]} \times 100 \quad (2)$$

where w_{fg} is the weight of carbon black and gel, m_f the weight of the filler in the compound, m_p the weight of the polymer in the compound and w the weight of the specimen. The carbon black dispersions in a few typical compounds were examined using a polaroid microscope.

RESULTS AND DISCUSSION

Figure 1 shows the variation of tensile strength with feeding rate for NR, IIR and SBR vulcanizates at different rpms at 80°C. The highest feeding rate for each rpm represents the feeding rate recommended by the manufacturer. It is found that irrespective of rpm, the tensile strength initially increased with feeding rate, reaches a maximum and thereafter decreases. This shows that for a given shear rate and temperature there is a particular feeding rate in the starved region, which results in maximum tensile strength. This is possibly due to the improved uniformity in temperature of the compounds since thick sections are not properly heated to uniform temperatures in the case of thermal insulators like rubber. Further, the lower shear breakdown and preferential orientation of the molecules may be the other reasons for the higher tensile strength at this feeding rate. The preferential orientation effect is clearly seen from the higher difference between the tensile strength measured in the longitudinal (extrusion) and transverse directions in this case (Figures 1 and 2).

Figures 3 and 4 show the variation in elongation at break of NR, IIR and SBR vulcanizates with feeding rate in the longitudinal (extrusion) and transverse directions at different rpms. As in the case of tensile strength, unimodal curves are obtained for each rpm. This further shows the efficiency of the starved extrusion, in getting maximum physical properties.

Figures 5 and 6 show the variation in tensile strength of NR, IIR and SBR vulcanizates with feeding rate at different temperatures at a fixed rpm. The tensile strength improves with temperature when the temperature is raised from 30°C to 90°C in the case of NR, from 60°C to 120°C in the case of IIR, and from 80°C to 100°C in the case of SBR showing that the deterioration due to thermal degradation is not serious in this range. This result is

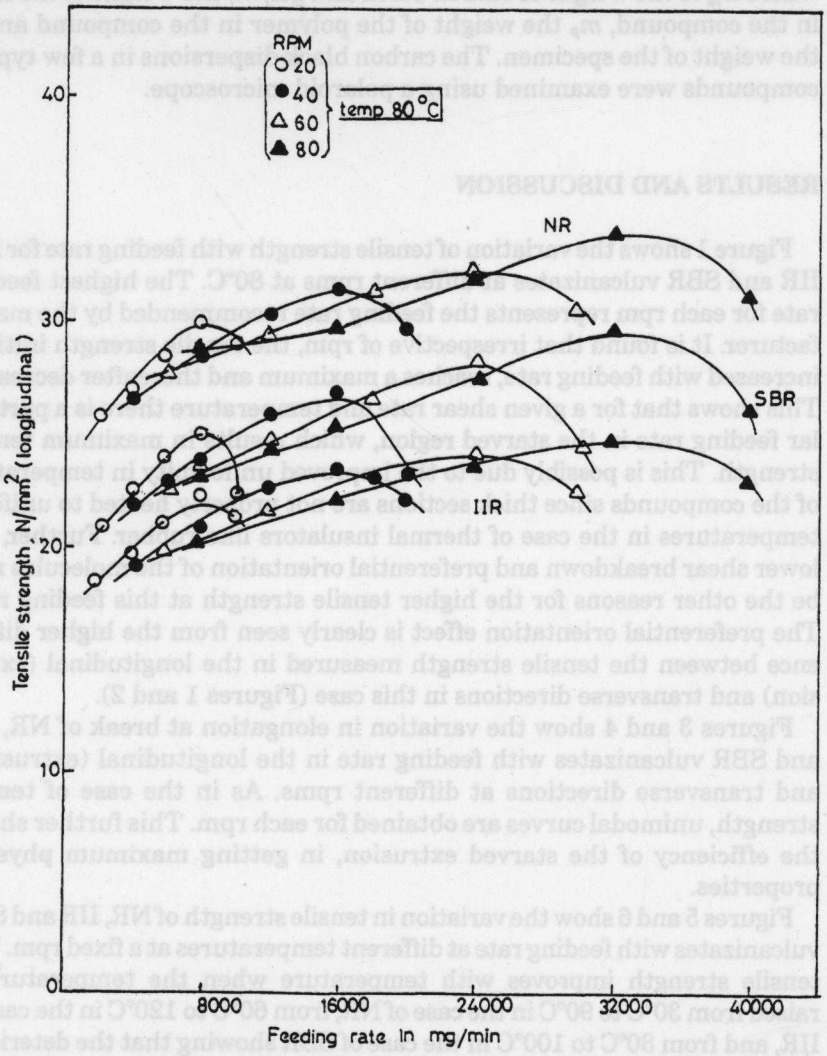

FIGURE 1. Effect of feeding rate on the tensile strength of NR, IIR and SBR vulcanizates at different rpms in longitudinal (extrusion) direction.

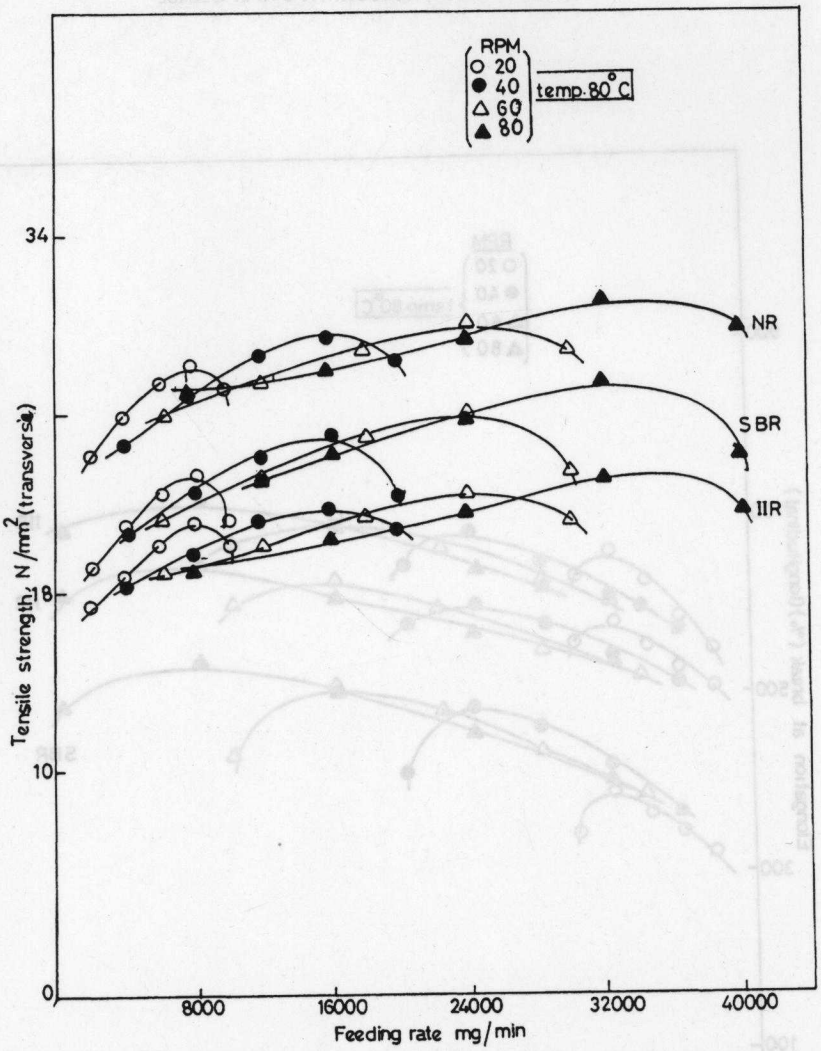

FIGURE 2. Effect of feeding rate on the tensile strength of NR, IIR and SBR vulcanizates at different rpms in transverse direction.

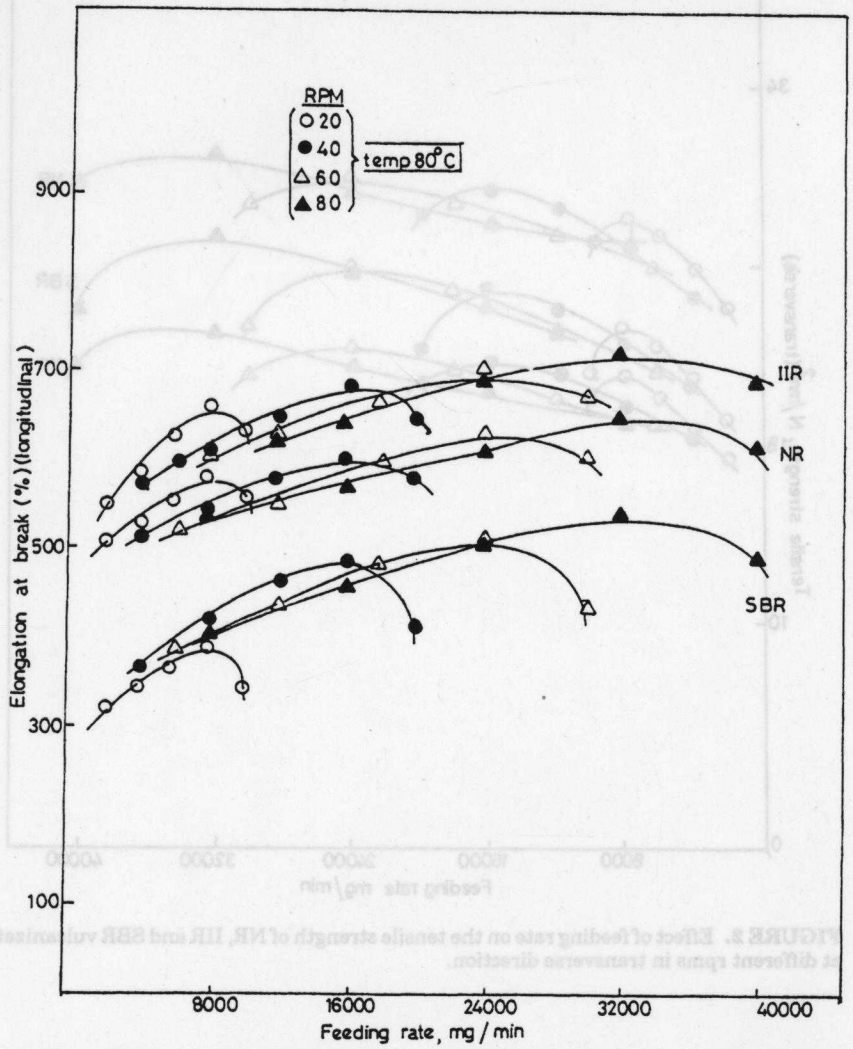

FIGURE 3. Effect of feeding rate on the elongation at break of NR, IIR and SBR vulcanizates at different rpms in longitudinal (extrusion) direction.

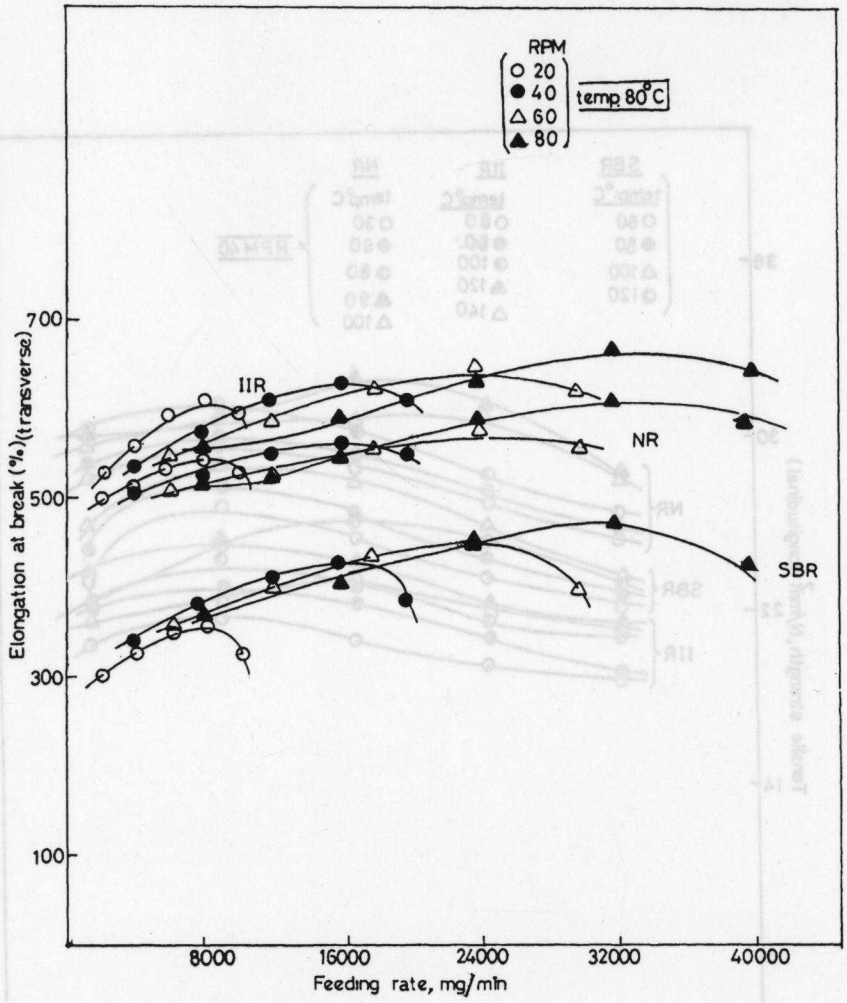

FIGURE 4. Effect of feeding rate on the elongation at break of NR, IIR and SBR vulcanizates at different rpms in transverse direction.

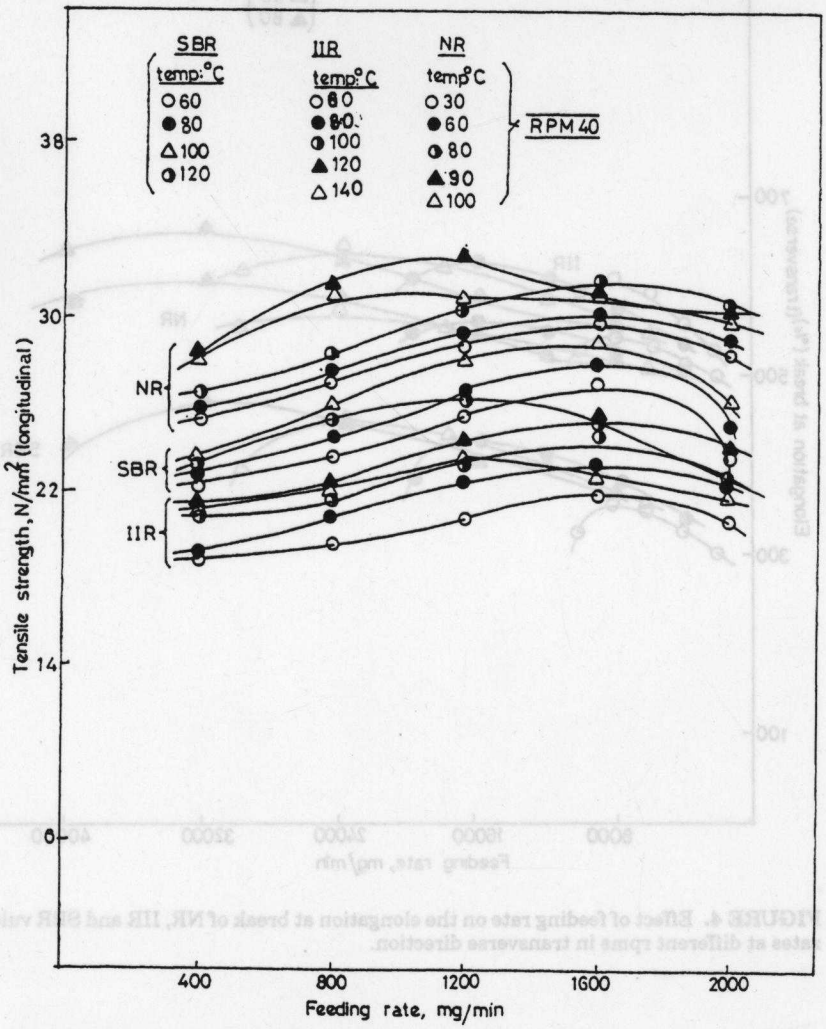

FIGURE 5. Effect of feeding rate on the tensile strength of NR, IIR and SBR vulcanizates at different temperatures in longitudinal (extrusion) direction.

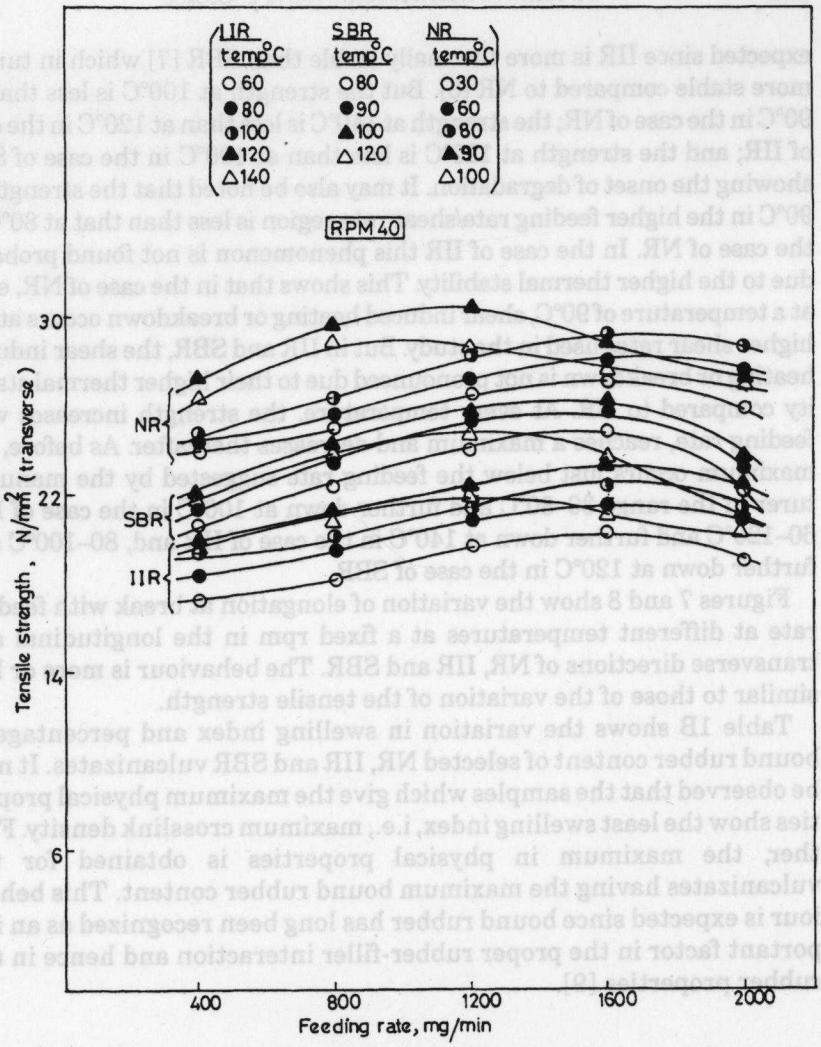

FIGURE 6. Effect of feeding rate on the tensile strength of NR, IIR and SBR vulcanizates at different temperatures in transverse direction.

expected since IIR is more thermally stable than SBR [7] which in turn is more stable compared to NR [8]. But the strength at 100°C is less than at 90°C in the case of NR; the strength at 140°C is less than at 120°C in the case of IIR; and the strength at 120°C is less than at 100°C in the case of SBR showing the onset of degradation. It may also be noted that the strength at 90°C in the higher feeding rate/shear rate region is less than that at 80°C in the case of NR. In the case of IIR this phenomenon is not found probably due to the higher thermal stability. This shows that in the case of NR, even at a temperature of 90°C, shear induced heating or breakdown occurs at the higher shear rates used in the study. But in IIR and SBR, the shear induced heating or breakdown is not pronounced due to their higher thermal stability compared to NR. At every temperature, the strength increases with feeding rate, reaches a maximum and decreases thereafter. As before, the maximum occurs just below the feeding rate suggested by the manufacturer in the range 30–80°C and further down at 100°C in the case of NR, 60–120°C and further down at 140°C in the case of IIR and, 80–100°C and further down at 120°C in the case of SBR.

Figures 7 and 8 show the variation of elongation at break with feeding rate at different temperatures at a fixed rpm in the longitudinal and transverse directions of NR, IIR and SBR. The behaviour is more or less similar to those of the variation of the tensile strength.

Table 1B shows the variation in swelling index and percentage of bound rubber content of selected NR, IIR and SBR vulcanizates. It may be observed that the samples which give the maximum physical properties show the least swelling index, i.e., maximum crosslink density. Further, the maximum in physical properties is obtained for the vulcanizates having the maximum bound rubber content. This behaviour is expected since bound rubber has long been recognized as an important factor in the proper rubber-filler interaction and hence in the rubber properties [9].

CONCLUSION

Starved feeding of partial filling of the screw channels during extrusion results in better heat transfer characteristics, proper preferential orientation, less mechanical breakdown and hence in a more uniform crosslink density and rubber-filler interaction. For a given screw there is an optimum feeding rate in the starved fed region which results in maximum physical properties. Therefore, running an extruder at a slightly starved condition is an attractive means of improving the physical properties in addition to running the extruder at a lower torque.

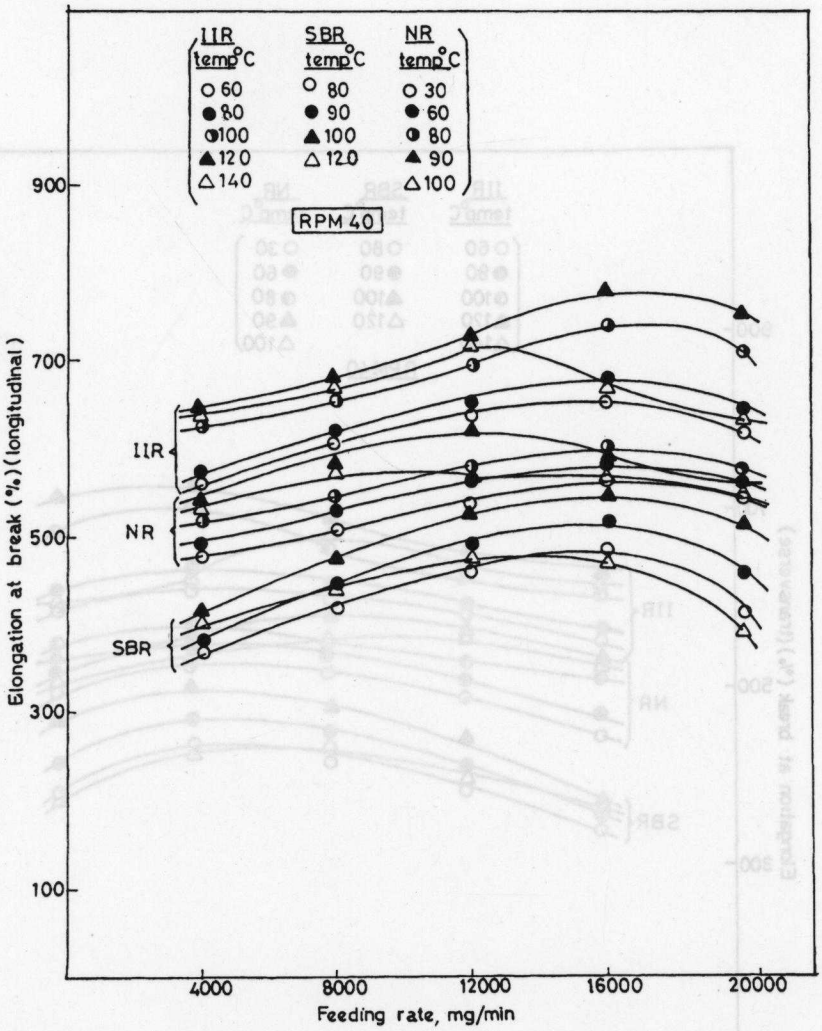

FIGURE 7. Effect of feeding rate on the elongation at break of NR, IIR and SBR vulcanizates at different temperatures in longitudinal (extrusion) direction.

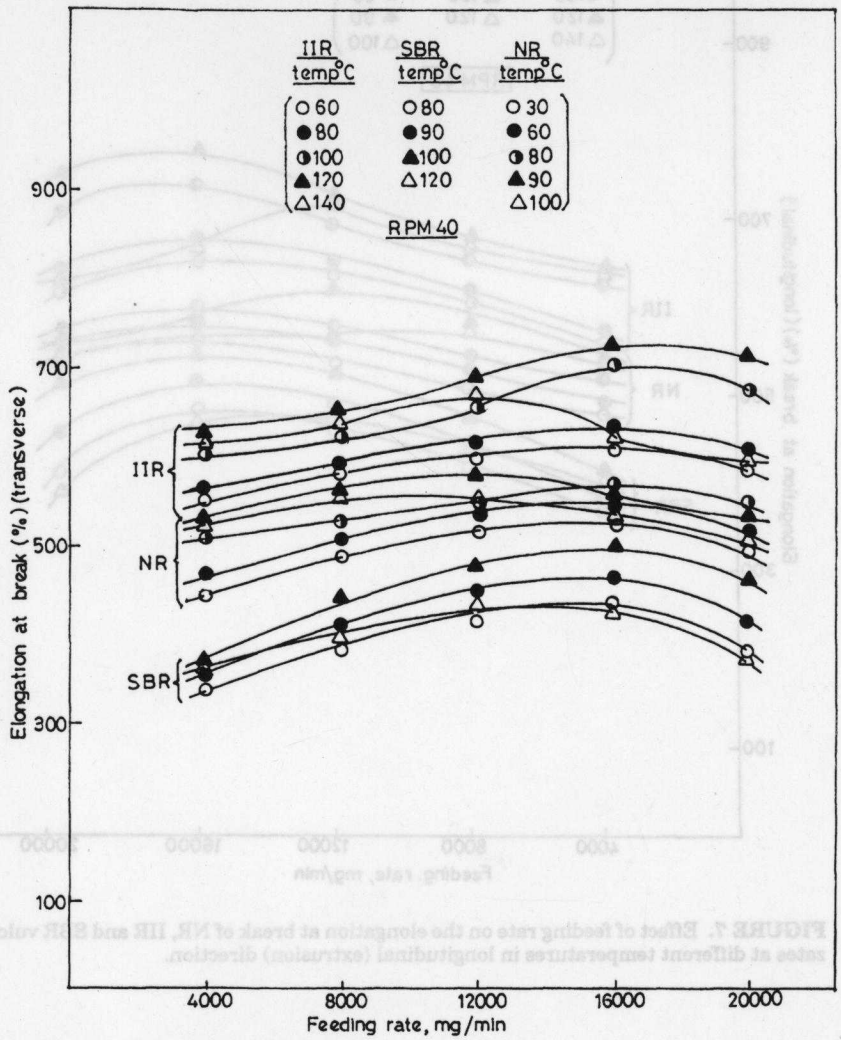

FIGURE 8. Effect of feeding rate on the elongation at break of NR, IIR and SBR vulcanizates at different temperatures in transverse direction.

Table 1B. The variation of swelling index and percentage of bound rubber content of selected NR, IIR and SBR vulcanizates.

	Feeding Rate in mg/min	Swelling Index	Bound Rubber Content in %
Unextruded NR	—	2.70	22
NR extruded at rpm = 80	8000	2.52	28
Temperature = 80°C	16,000	2.38	33
	24,000	2.16	39
	32,000	2.09	48
	40,000	2.33	30
NR extruded at rpm = 40	4000	2.91	24
Temperature = 90°C	8000	2.40	28
	12,000	2.27	36
	16,000	2.36	32
	20,000	2.47	27
Unextruded IIR	—	2.00	27
IIR extruded at rpm = 80	8000	1.83	32
Temperature = 80°C	16,000	1.52	37
	24,000	1.44	43
	32,000	1.25	51
	40,000	1.48	34
IIR extruded at rpm = 40	4000	1.62	36
Temperature = 120°C	8000	1.50	39
	12,000	1.36	46
	16,000	1.20	57
	20,000	1.43	41
Unextruded SBR	—	2.99	25
SBR extruded at rpm = 80	8000	2.56	31
Temperature = 80°C	16,000	2.03	36
	24,000	1.74	45
	32,000	1.24	54
	40,000	1.91	33
SBR extruded at rpm = 40	4000	2.82	27
Temperature = 100°C	8000	2.50	32
	12,000	2.01	37
	16,000	1.51	48
	20,000	2.32	31

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