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Thermoplastic Elastomers I. Effect of Processing Variables on Tensile Properties of Natural Rubber/Polypropylene Blends

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The effect of processing conditions and type of mixers on the mechanical performance of different compositions of natural rubber/polypropylene (NR/PP) blends are investigated. The use of 0.3% dicumyl peroxide in 60% natural rubber blends under optimised processing conditions resulted in the best mechanical performance of the blend. High shearing rate mixers (Buss-Ko kneader) afforded blends with higher initial mechanical performance compared with a laboratory-scale internal mixer (Hampden-RAPRA torque rheometer). The evidence presented indicates the lack of interpolymer formation during the mechano-chemical process although a small amount of NR undergoes crosslinking during the processing operation. Both melt stability and mechanical properties of NR/PP blends are adversely affected on remoulding: reinjection showed much greater deterioration compared to recompression, albeit high initial mechanical properties of the injection moulded samples.

A basic requirement of a thermoplastic elastomer is the formation of crosslinks which are stable at room temperature but become unstable at processing temperatures. Thus thermoplastic elastomers exhibit both the characteristics of vulcanised rubber at room temperature and thermoplastics processibility at elevated temperatures. In the case of polypropylene-based thermoplastic elastomers, the micro-crystalline domains in polypropylene (PP) offer 'pseudo-crosslinks' which in turn are responsible for the stiffness and strength of the blends[1]. These blends (e.g. NR/PP) have two distinct advantages over other thermoplastic rubbers; they are made from relatively low cost materials and they have low densities.

The characteristics of polyolefin thermoplastic elastomers depend greatly on the processing method used for their fabrication. Like other polyblends[2], the polyolefin thermoplastic elastomers may be produced by mechanical blending[3], whereby the two different polymers are subjected to high shearing forces in a thermoplastic processing machine at a temperature above their glass transition. Under such processing conditions, chain scission of both polymers takes place and leads to the formation of two different active macro radicals. In the absence of oxygen, recombination of these radicals yields block- or graft-copolymers[4].

To achieve enhanced mechanical performance of polymer blends, the two phases must be compatible to a certain extent. An external solid phase dispersant is used to increase the compatibility between the two phases[5]. Blends based on NR/PP thermoplastic elastomers, on the other hand, show good mechanical performance even in the absence of an external solid phase dispersant[3]. The question arises as to whether an interpolymer formation due to reactions between the two polymeric phases during processing serves as an 'internal' solid phase dispersant.

The purpose of the present work is two-fold First, to examine the question of interpolymer formation under a wide range of processing conditions and to study the effect of these conditions on the mechanical and long-term

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ageing performance of the blends before and after various remoulding cycles (the present investigation) and secondly, to evaluate the use of NR/PP thermoplastic elastomers as solid phase dispersant for other thermoplastic-based polyblends (Part II).

EXPERIMENTAL

Materials

The following polymers were used: stabilised PP (HW 25) supplied by ICI Ltd, natural rubber (SMR 5) supplied by MRPRA and ethylene-propylene-diene monomer supplied by Exxon Chemicals Ltd under the trade name Vistalon 6505. Benzoyl peroxide (ex. BDH) and dicumyl peroxide (ex. Hercules) were used as received without further purification.

Polymer Processing

Processing was carried out either in a closed mixer of a Hampden-RAPRA torque rheometer or in a Banbury mixer or extruded in a Buss-Ko kneader, at different temperatures (160°C–180°C) and times (5–11 min). The polymer was then either compression moulded at 180°C into sheets of thickness 200 μm as described previously[6] or injection moulded on an Edgwick 1214 HY injection moulding machine using barrel temperatures of 170°C, 180°C, and 200°C, mould temperature of 60°C, injection pressure of 47 MPa, screw speed of 50 r.p.m. and cooling time of 16 s. To prepare NR/PP blends in the presence of peroxides, the two polymers were first blended (without the peroxide) for 3 min at the required processing temperature. At the end of this period the required amount of peroxide was added and the processing was continued for various periods of time up to 11 min.

Tensile properties. Tensile tests were carried out on an Instron tensile tester (Model TMTS) using cross head speed of 3 cm per minute at 20°C. Six samples were tested for each measurement.

Melt rheology. Both the melt flow index (MFI) and viscosity of the polymer melt under a range of shear rates at a given temperature were investigated. Measurement of the melt viscosity was carried out on a Weissenberg rheogoniometer which is basically a cone and plate viscometer. The flat platen remains stationary while the conical platen moves with an angular velocity (β) which can be regulated. The temperature was set at 230°C (similar to that used for MFI determination) and a rotational force was applied. The sample (processed in a torque rheometer and compression moulded into film, 20 μm thick) was placed between the two platens and was allowed to reach the test temperature (230°C). Viscosity measurements were carried out at different shear rates *(Equation 1)*. Viscosity of the melt was calculated[7] according to *Equation 2:*

$$\text{Shear rate } (\gamma) = \frac{360}{\alpha t} \ s^{-1} \qquad \ldots 1$$

$$\text{Viscosity } (\eta) = \frac{t \times \alpha \times \Delta T \times K_t}{94.25 \times d^3} \text{ poise} \qquad \ldots 2$$

where the value of

t is dependent on the angular rotation and its value (in seconds per revolution) is calculated from the gear setting of the rheogoniometer

α is angle of cone which is 4° 8′

ΔT is the movement of the torsion head transducer (in thousandths of an inch),

K_t is torsion head constant (dyne cm per thousandths of an inch of movement of torsion head transducer) and is 3.372×10^4

d is diameter of platens which is 2.5 cm.

MFI measurements were done on a Davenport melt flow indexer at 230°C using a load of 50 Newton. Calculated MFI values are the average of five measurements for each sample.

Molecular Weight and Gel Determination

Molecular weight distribution of the soluble fraction (in tetrahydrofuran) of processed

polymer samples was determined by RAPRA using gel permeation chromatography. In these analyses, tetrahydrofuran was used as a solvent and polystyrene for the purpose of calibration. Solvent insoluble gel was measured by dissolving 0.5 ml of processed polymer in 50 ml tetrahydrofuran (90°C, 60 min) followed by filtration under nitrogen. The gel formed was vacuum-dried to a constant weight.

Determination of Insoluble Bound Natural Rubber

The extent of insoluble bound NR on PP was determined as follows: decaline (50 ml) was added to 0.5 g of NR/PP (60/40) blend and the solution was heated to 170°C under nitrogen until all the polymer had dissolved. On cooling, a mixture of PP and any insoluble 'bound' NR precipitated out while the soluble unbound NR remained dissolved in decaline. The precipitate was filtered and redissolved. This was repeated three times and the final precipitate was dried to a constant weight. The bound NR was calculated as shown below,

$$\text{Percentage bound NR} = \frac{A - B}{A} \times 100 - 40\%$$

where A is the weight before extraction

B is the weight after extraction

40% is PP fraction in the blend.

RESULTS AND DISCUSSION

Effect of Processing Variables on Natural Rubber/Polypropylene Blends

The effect of processing conditions (temperature and time using torque rheometer) on the mechanical performance of various compositions of NR and PP (compression moulded samples) was studied. *Figure 1* shows the effect of varying the composition of NR/PP and the effect of processing temperature on the tensile strength of the blends. It is clear that increasing PP content in the composition increases the tensile strength of the blends under the different temperature conditions used; the tensile strength of PP alone is much higher than that of NR. Moreover, all blends processed at 160°C offered slightly higher tensile strength than those produced at higher temperatures. At lower temperatures, the viscosity of the blend is higher (see inset *Figure 1*) leading to higher shear rates. This may contribute to more homogeneous dispersion between the two phases with an overall improvement in mechanical performance.

The effect of processing time (at all temperatures used) on the tensile strength of the blends is less pronounced. Slightly better performance is obtained with samples processed for 7 min and this condition was subsequently used. In order to choose the optimum composition of NR/PP which produces a thermoplastic rubber, the tensile stress of different blends processed under the above conditions (160°C, 7 min) was plotted against their elongation at break *(Figure 2)*. The tensile properties of blends containing low concentrations of NR (e.g. 40%) are typical of thermoplastics, *i.e.* high tensile strength and low elongation at break. This suggests that the dominant phase here is PP. On the other hand, the tensile properties of blends containing high concentrations of NR (e.g. 70%) are typical of uncrosslinked rubbers, *i.e.* low tensile strength and high elongation, suggesting that the dominant phase is NR. The observed tensile properties of blends containing 60% NR support the formation of a thermoplastic rubber. It was shown previously[3] by scanning transmission electron microscopy that both phases of a 60/40 NR/PP blend, processed in an internal (Banbury) mixer, are essentially continuous.

Interpolymer Formation in Natural Rubber/Polypropylene Blends

It has been shown that addition of solid phase dispersants to polymer blends enhances their mechanical performance[5]. Mechanochemically formed NR/PP blends may inherently contain an interpolymer which acts as an 'internal' solid phase dispersant and may therefore contribute to improvements in mechanical properties of these blends. This possibility was examined in all the NR/PP

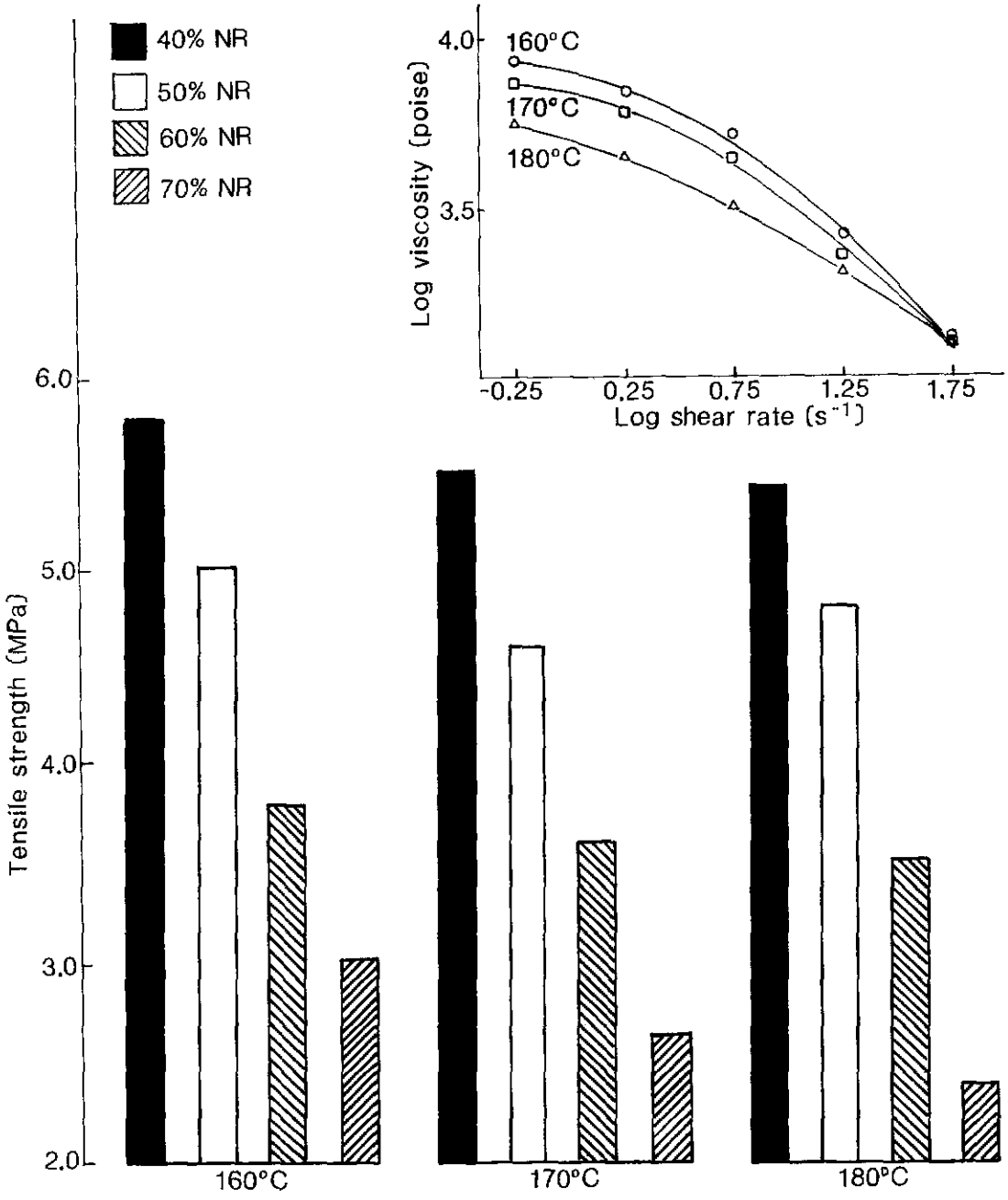


Figure 1. Effect of composition of NR/PP blends on their tensile strength when processed in torque rheometer at different temperatures for 7 min. Inset shows the effect of processing temperatures on the melt rheology of NR/PP blend containing 60% NR (processed for 7 min).

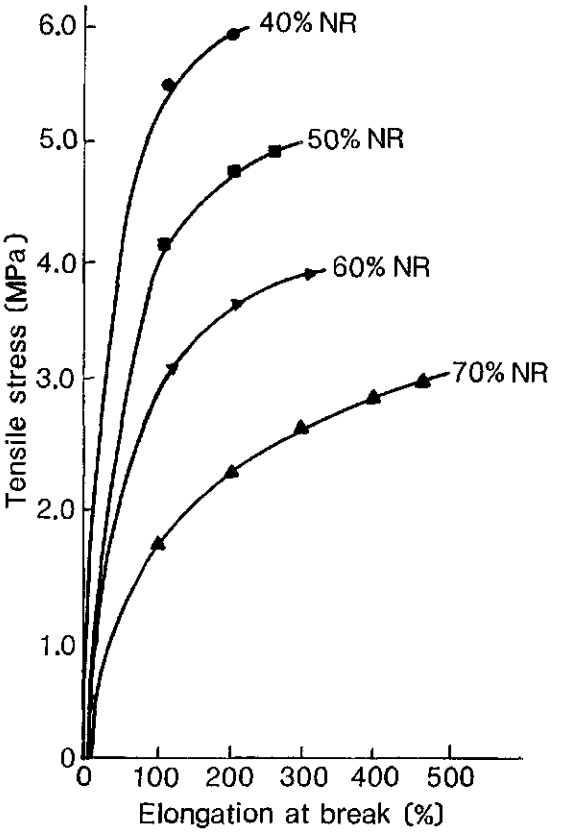


Figure 2. Stress-strain curves of different compositions of NR/PP blends processed at 160°C for 7 min.

compositions used. *Figure 3* shows the effect of processing temperature and time (compression moulded samples processed in torque rheometer) on the molecular weight of extracted NR/PP compositions containing 70% and 40% NR. It is clear that processing at higher temperatures leads to a decrease in molecular weight of the soluble fractions of the blends; the effect is more pronounced with higher NR content. On the other hand, increasing PP content in the blend is accompanied by a decrease in the molecular weight under all processing conditions (see inset).

Measurement of the amount of insoluble 'bound' NR shows that, although the overall percentage of bound NR in all the compositions and under the different processing conditions used is small, its percentage decreases with increasing PP content in the blends *(Figure 4)*. The observed reduction in molcular weight of NR/PP blends when the PP content is increased (see inset *Figure 3*) is accompanied by a concomittant reduction in viscosity which leads to lower shearing forces. However, since the shearing forces exerted during the processing operation play an important role in the formation of macro-radicals, and ultimately to the interchange of the mechano-chemically formed macro-radicals of both polymer components of the blend, lower shear forces in blends containing higher PP contents must contribute to lower amounts of the interpolymer formed [as measured from the insoluble (percentage bound) NR in these blends, see *Figure 4*]. The very small amount of insoluble NR formed under the above processing conditions is not likely to contribute, to any significant extent, to the enhancement of mechanical properties of these blends. The relation between the insoluble NR and the mechanical performance of NR/PP (60/40) blends, when the processing operation is optimised, and hence its role on the performance of the blend is discussed in the next section.

Effect of Free Radical Initiators on Performance of 60/40 Natural Rubber/Polypropylene Blend and Role of Interpolymer Formed

The effect of free radical initiators on mechanical performance of NR/PP blends containing 60% NR was investigated. Benzoyl peroxide and dicumyl peroxide were examined. *Figures 5* and *6* show the effect of the above peroxides on tensile strength of the blends. It is clear that benzoyl peroxide does not offer a significant increase in tensile strength *(Table 1)*. There is, however, a small but steady increase in the amount of insoluble ('bound') NR with increasing concentration of this peroxide *(Figure 5)*. *Figure 6* shows that dicumyl peroxide has a more significant effect on the tensile behaviour *(Table 1)* of the NR/PP blend, at all processing temperatures, with marked improvements obtained at a concentration of 0.3 p.h.r. and a processing temperature of 170°C.

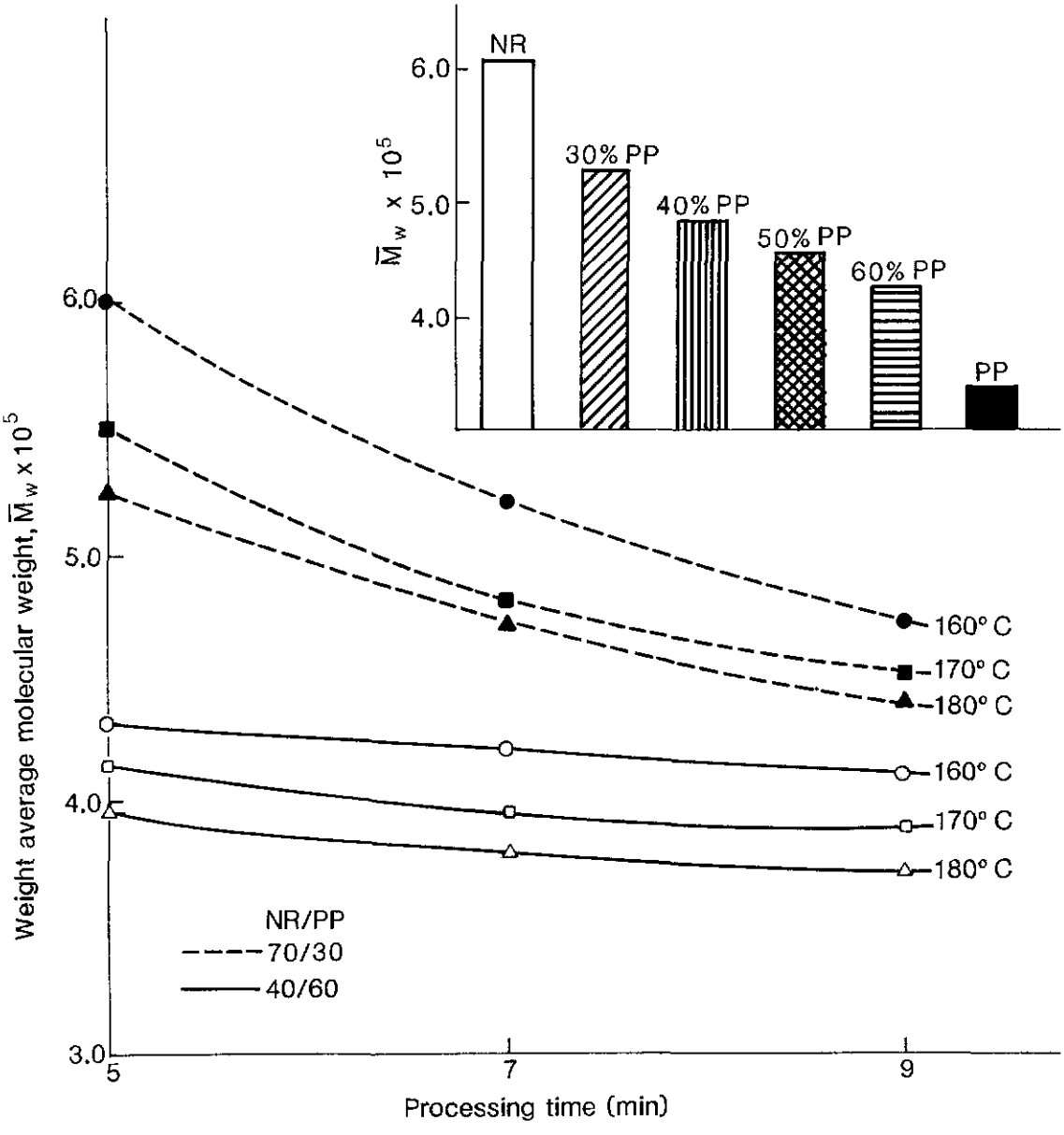


Figure 3. Effect of processing time and temperature on weight average molecular weight ($\bar{M}_w$) of tetrahydrofuran-soluble fraction of two different NR/PP blend compositions. Inset compares $\bar{M}_w$ of different NR/PP compositions with those of the NR and PP controls; the blends were processed at 160°C for 7 min.

Similarly, the amount of 'bound' NR present in samples prepared with dicumyl peroxide is higher than those processed with benzoyl peroxide *(Figure 7)*. Further improvement in the tensile properties of the NR/PP blend, which contains the optimum dicumyl peroxide concentration (0.3 p.h.r.), was achieved by increasing processing time *(Figure 8)*. For

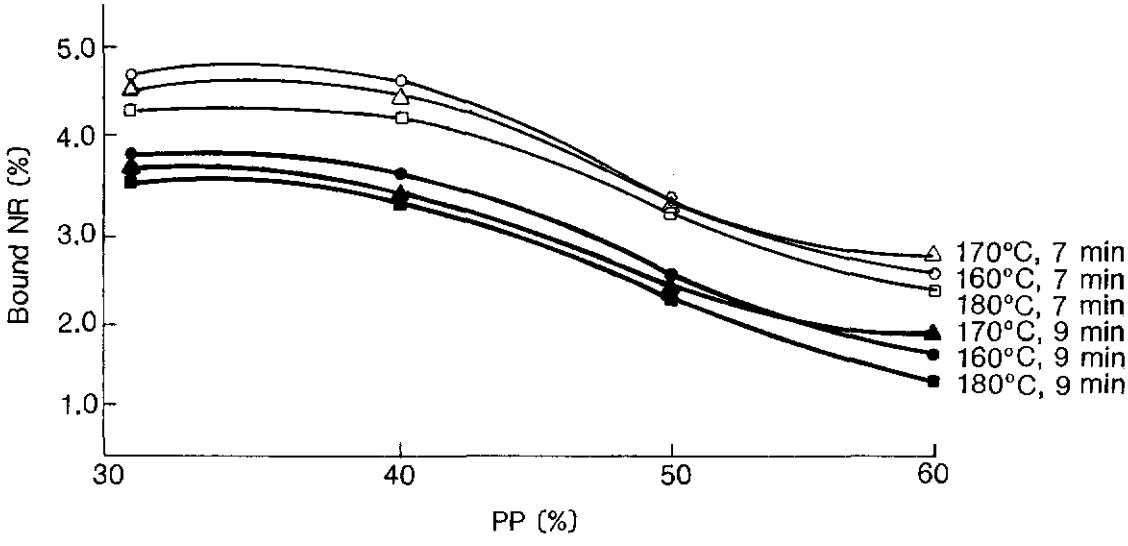


Figure 4. Effect of composition of NR/PP blends on extent of 'bound' NR formed during processing at different temperatures and times.

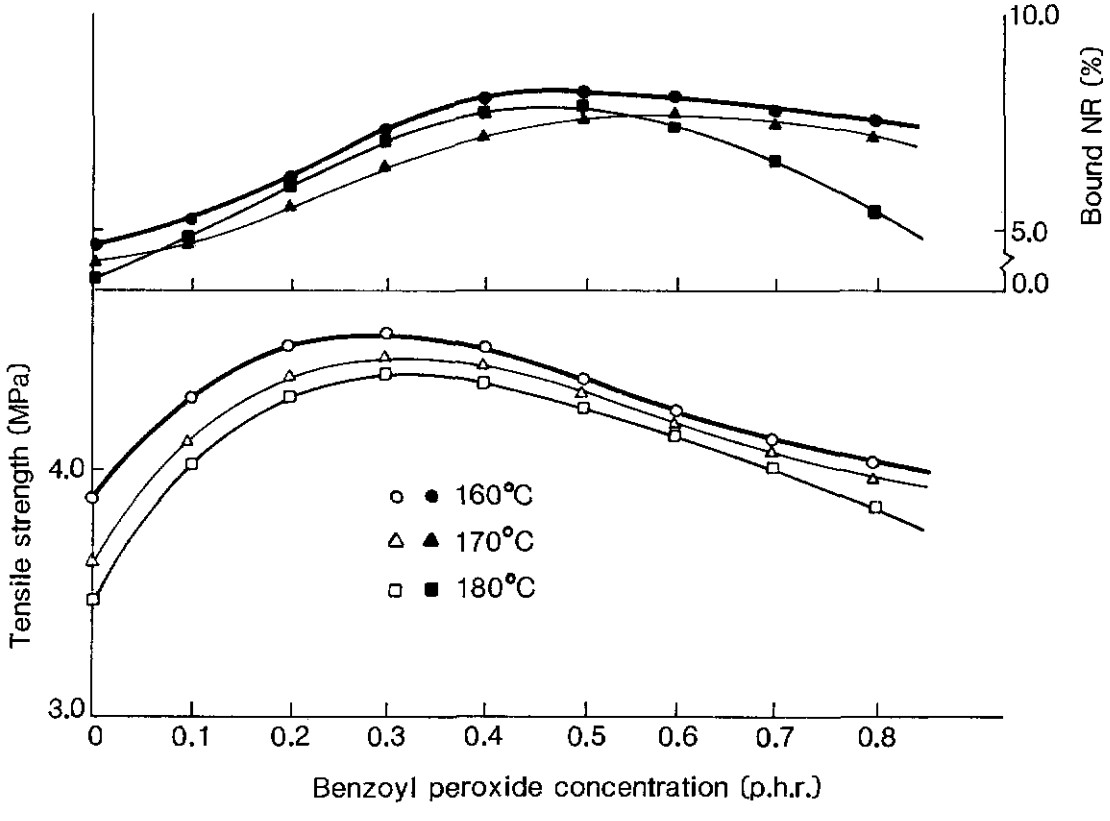


Figure 5. Effect of benzoyl peroxide concentration on extent of 'bound' NR and on the tensile strength of NR/PP (60/40) blends processed at different temperatures for 7 min.

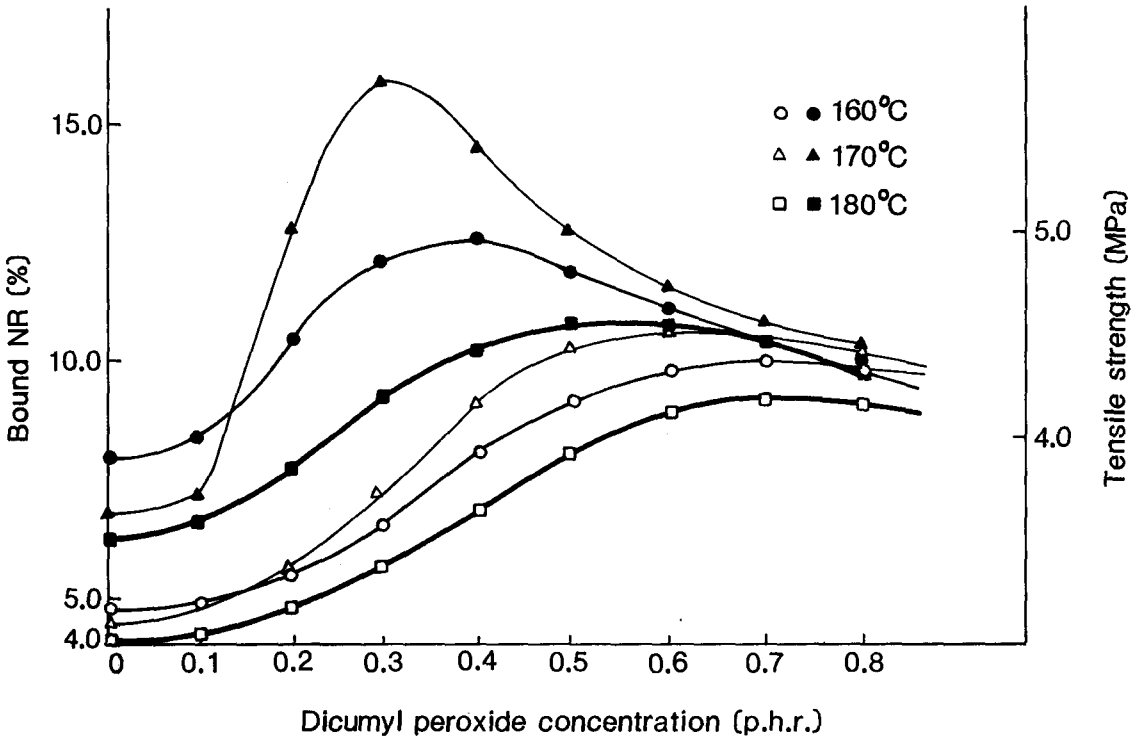


Figure 6. Effect of dicumyl peroxide concentration on extent of 'bound' NR and tensile strength of NR/PP (60/40) blends processed at different temperatures for 7 min.

TABLE 1. EFFECT OF PEROXIDES ON TENSILE STRENGTH OF NR/PP (60/40) BLENDS PROCESSED AT DIFFERENT TEMPERATURES FOR 7 MIN

Temperature (°C)	Peroxide (p.h.r.)	Improvement in tensile strength (%)
170	0.3 (dicumyl peroxide)	45
160	0.4 (dicumyl peroxide)	27
180	0.5 (dicumyl peroxide)	22
160	0.3 (benzoyl peroxide)	16

example, under the optimum processing conditions (160°C, 10 min) addition of 0.3 p.h.r. dicumyl peroxide gives rise to 88% improvement in tensile strength when compared to a similarly processed control in the absence of peroxide (or 34% improvement when compared to the 7 min processed analogue). The improvement in tensile properties under all processing conditions, however, does not appear to be related to the insoluble 'bound' NR content formed during processing. Inset of *Figure 7* for example shows this clearly at 170°C whereby the sharp drop in tensile strength, observed after the addition of 0.3 p.h.r. dicumyl peroxide, corresponds to a gradual increase in the amount of 'bound' NR. This suggests that

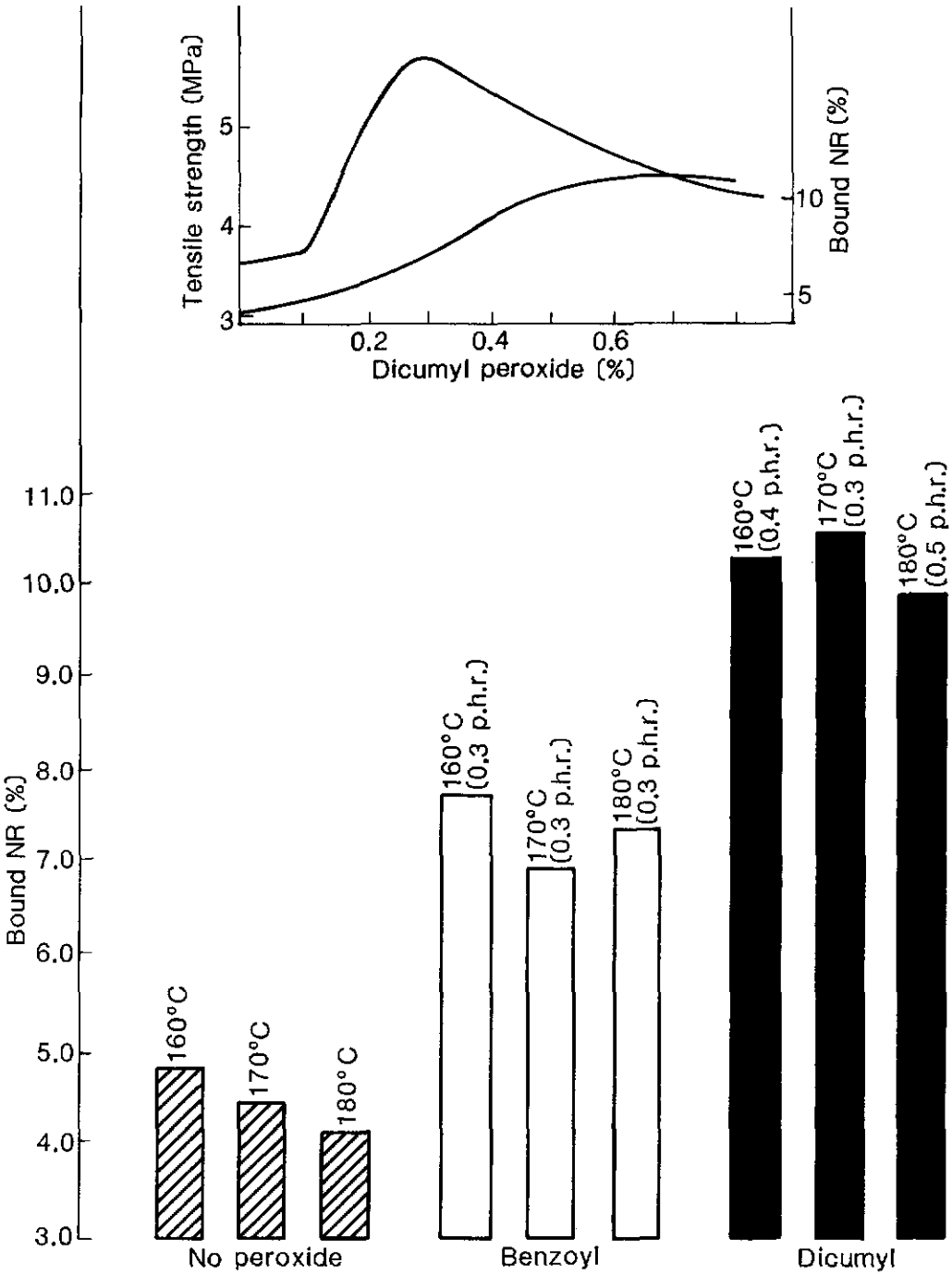


Figure 7. Maximum concentration of insoluble ('bound') NR in blends of NR/PP containing 60% NR in the presence and absence of peroxides. Blends were processed at different temperatures for 7 min. Inset shows the effect of dicumyl peroxide on extent of 'bound' NR and tensile strength of the above blend when processed at 170°C for 7 min.

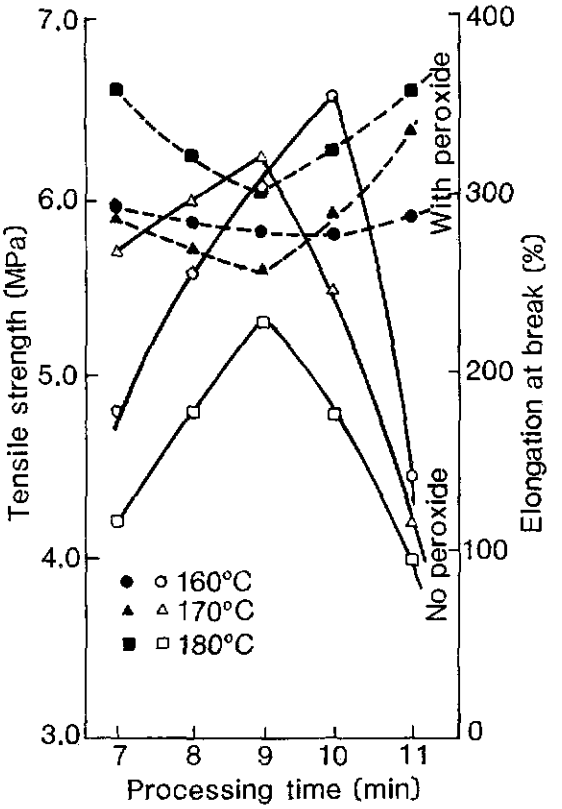


Figure 8. Effect of processing time and temperature on tensile properties of NR/PP (60/40) blends containing 0.3 p.h.r. dicumyl peroxide. Tensile strength of above samples when processed without peroxides is also shown for comparison.

the 'bound' NR as determined here is not in the form of interpolymer, but may have resulted from a chemical reaction (crosslinking) in the rubber phase itself without any significant participation from the PP phase. Peroxides are known to promote crosslinking of rubbers[8]. In the case of polyolefins, on the other hand, the effect of peroxides during high temperature processing is an increase in the level of thermal degradation[9]. If peroxides affect the components of NR/PP blends in this way then this may account for the decrease in tensile strength which is observed at higher peroxide concentrations (≥ 0.3 p.h.r.).

To examine the above possibilities, the behaviour of each component of the blend *i.e.* NR and PP, was compared with that of the blend itself under identical processing conditions and similar concentrations of dicumyl peroxide. If degradation of the counter polymer, *i.e.* PP, does take place with increasing peroxide concentration then this must be due to two factors; an increase in viscosity of the blend and the effect of peroxide on the PP phase. *Figure 9* shows changes in viscosity of NR/PP blends (with 60% NR) containing different concentra-

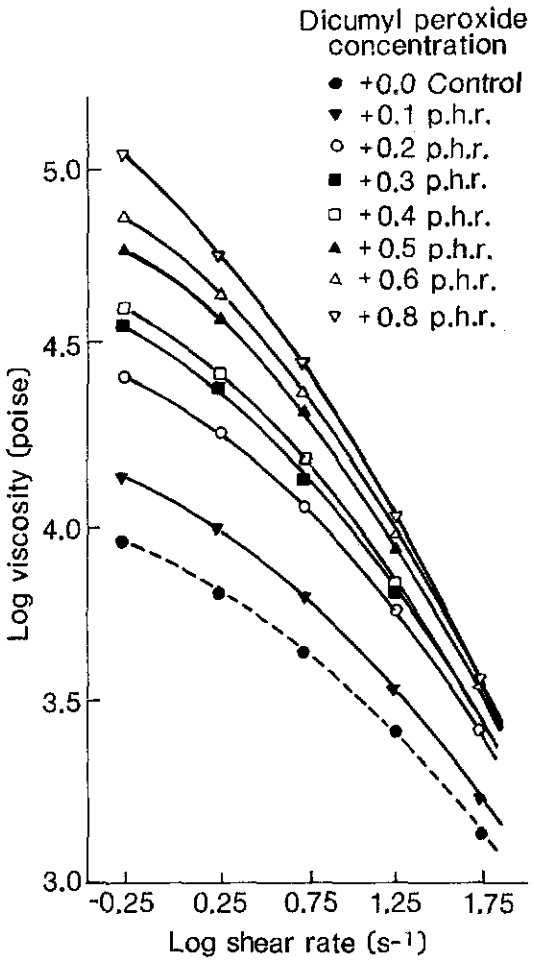


Figure 9. Effect of dicumyl peroxide on the melt rheology of NR/PP (60/40) blends processed at 160°C for 7 min.

tions of dicumyl peroxide at different shear rates. It is clear that increasing the concentration of dicumyl peroxide causes a remarkable increase in viscosity of the blends at low shear rate but this difference decreases dramatically with increasing shear rates. This is even more pronounced for blends which have higher viscosities. Increasing viscosity of the blends causes higher shearing forces which may lead to greater degradation of the PP phase. *Figure 10* compares changes in MFI of PP and NR samples processed separately in the presence of varying concentrations of dicumyl peroxide. It is clear that the MFI of NR decreases dramatically with increasing concentration of dicumyl peroxide under all processing temperatures suggesting a crosslinking reaction. In contrast, MFI of PP processed under the same conditions increased very rapidly with increasing concentration of the peroxide indicating a chain scission process. Inset of *Figure 10* shows clearly that the gel content in NR increases dramatically with increasing concentration of dicumyl peroxide while the molecular weight of the gel-free remaining solution (the soluble fraction) decreases. Increasing concentration of dicumyl peroxide, therefore, leads to the formation of an insoluble high molecular weight fraction in NR (see also *Figure 6,* note the continuous increase in the unextractable NR content with increasing dicumyl peroxide concentration in the blend). This must give rise to the overall observed decrease in MFI of these NR samples. In the case of PP, on the other hand, no gel was formed under all concentrations of dicumyl peroxide used and the weight average molecular weight of PP (all of which is soluble) decreases with increasing concentration of dicumyl peroxide. This finding is in accord with the earlier observation where MFI of these samples show an increase with increasing peroxide concentration *(Figure 10).* These observations are attributed to the degradation of PP by a chain scission process. High temperature thermal processing of polyolefins may lead to oxidative degradation *(Scheme 1a),* crosslinking *(Scheme 1b)* or chain scission *(Scheme 1c)*[10,11]. Crosslinking reactions take place only under restricted oxygen access and the structure

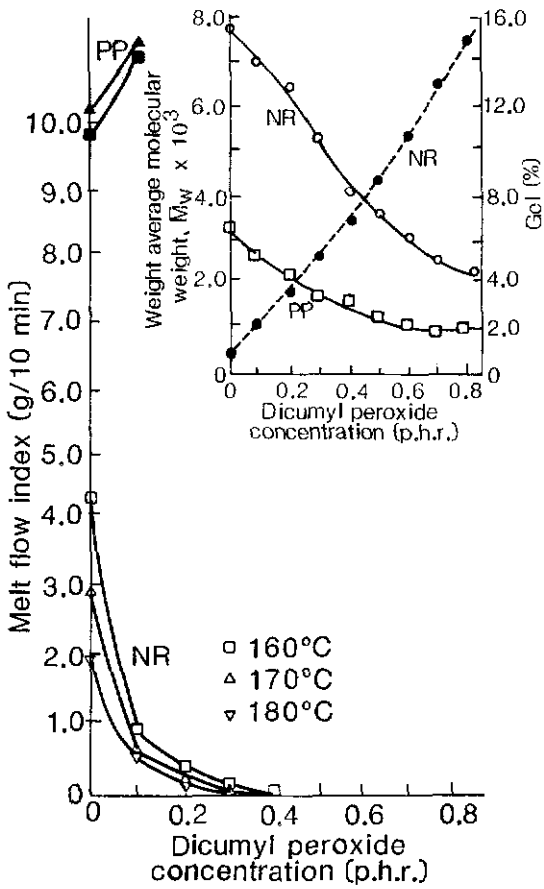
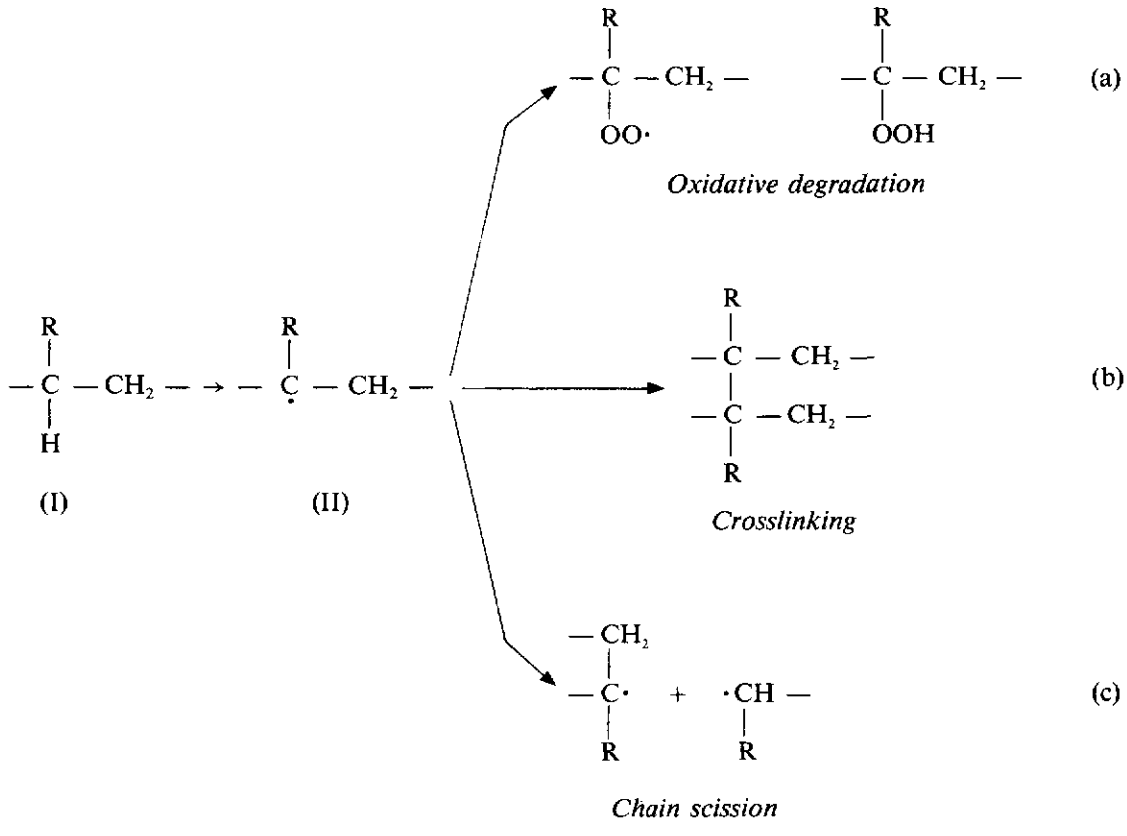


Figure 10. Effect of dicumyl peroxide on melt flow index of NR and that of PP processed separately at different temperatures. Inset shows the effect of dicumyl peroxide on weight average molecular weight of tetrahydrofuran-soluble fraction of PP and of NR processed separately at 160°C for 7 min. Amount of gel formed in NR is also shown.

of the R group *(Scheme 1, I)* determines the preferred reaction pathway. In the case of PP (R = CH_3) the positive induction effect of the methyl group facilitates a homolytic scission of the C-H bond and the tertiary macroradical *(II)* formed will readily undergo chain scission *(Scheme 1c)* even under low partial pressure of oxygen; in the presence of oxygen, *Reaction 1a* predominates.



Scheme 1. Primary degradative processes occurring as a result of thermal processing of polyolefins.

In the light of the above results, the effect of dicumyl peroxide on the behaviour of NR/PP blends was examined. *Figure 11* shows that in NR/PP blends increasing concentration of dicumyl peroxide leads to a dramatic overall decrease in the MFI. The effect of the peroxide on MFI of the blends must then be two-fold: a reduction in MFI due to the crosslinking process in the rubber phase, and an increase in MFI (or decrease in molecular weight of the soluble fraction of the blend, *Figure 11*) due to degradation of PP contents. However, since the amount of NR in this blend is higher than that of PP, the process of crosslinking predominates as shown in *Figure 11*. The effect of degradation of PP is likely to be more important in determining the tensile properties of the blend since in such a thermoplastic elastomer, the strength relies on the PP phase and hence a consequent drop in tensile strength of the blend after addition of 0.3 p.h.r. dicumyl peroxide is observed *(Figure 6)* even though the amount of crosslinked rubber continues to increase.

Effect of Processing and Remoulding Operations on Natural Rubber/Polypropylene

Mechanical properties and melt stability of injection moulded NR/PP samples when processed in the three different mixers at different temperatures are compared in *Table 2*. It is clear that blends processed in the Buss-Ko kneader and Banbury mixer have better initial mechanical properties than those processed in the torque rheometer. The first two processing

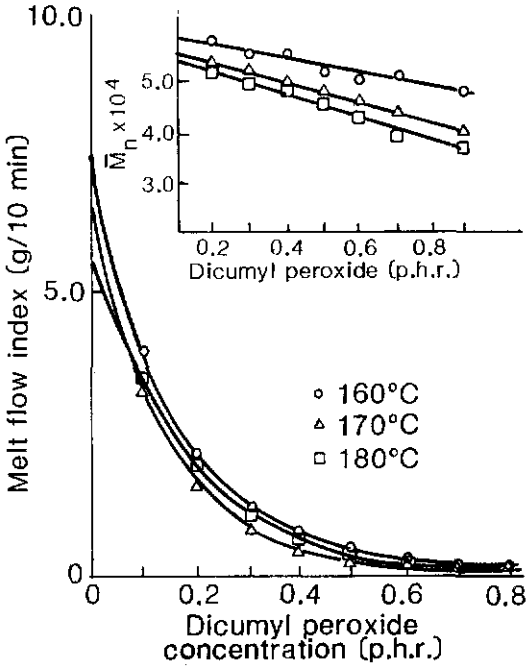


Figure 11. Effect of dicumyl peroxide on melt flow index of NR/PP (60/40) blends processed at different temperatures for 7 min. Inset shows changes in number average molecular weight of the soluble fraction of the blends under the same conditions.

machines offer higher shearing forces than the laboratory-scale internal mixer; the Buss-Ko kneader offers the highest shearing forces due to the double (reciprocating and rotating) action of the screw. The higher shearing forces exerted by the former processing machines must help in rupturing the molecular chains of NR to a larger extent than is the case in the torque rheometer leading to a decrease in viscosity of the rubber phase hence offering better dispersion of the two phases which in turn contribute to the observed higher mechanical properties in the blend. It is clear that a combination of high temperature and shearing forces reduces the thermal stability of the blends as reflected in the higher MFI values *(Table 2)*.

Recycling of polymer blends based on combinations of polypropylene with other thermoplastic polymers such as polyethylene or polystyrene has been studied and antioxidants have been used to protect the blends under conditions encountered during recycling operations with the production of products with good initial and long-term performance[12]. The idea of being able to remould rejects and scraps of NR/PP blends is an attractive one and is examined here. Both recompression and reinjection mouldings were investigated. *Figure 12* shows the effect of ten successive recompression moulding cycles (at 180°C) on the tensile properties and melt stability of the NR/PP blends processed in the three mixers (at 160°C). It is clear that in all cases the mechanical properties are retained up to three or four recompression cycles before a sharp decline in the properties takes place. This coincides with only little changes in the MFI during the first three cycles (inset *Figure 12*). In this respect the behaviour of thermoplastic rubber tested here is similar to that of a typical thermoplastic in being able to be remoulded. Although during the first few recompression cycles, blends processed in the Buss-Ko kneader (where the shearing forces are high) show higher mechanical performance than when processed in a laboratory-scale torque rheometer (see, for example, tensile strength curves in *Figure 12*), at a larger number of recompression cycles (more than five) the curves cross over, such that performance of blends processed under conditions of high shear (Buss-Ko kneader) becomes lowest. This emphasises the importance of the initial thermal history on subsequent operations. Initially, the more homogeneous mixing offered by processing mixers with high shearing forces is the main contributory factor to the better initial properties. However, at the same time, under these high shear rate conditions the thermal stability of the blends is lower than if processed under lower shearing conditions as reflected by the higher MFI (inset *Figure 12*). This thermoxidative instability becomes the dominating factor responsible for the much lower strength of the blends observed after larger numbers of recompression moulding cycles.

Figure 13 shows the effect of reinjection moulding on the behaviour of NR/PP blends.

TABLE 2. INITIAL MECHANICAL PROPERTIES OF INJECTION MOULDED NR/PP (60/40) BLENDS PROCESSED IN BUSS-KO KNEADER, BANBURY MIXER AND A RAPRA-HAMPDEN TORQUE RHEOMETER AT DIFFERENT TEMPERATURES

Mechanical properties	Processing temperatures (°C)		
	150	160	170
Torque rheometer			
Tensile strength (MPa)	—	7.60	7.30
Elongation at break (%)	—	390	350
Tensile modulus (MPa)	—	63	60
Flexural modulus (MPa)	—	145	136
Flexural strength (MPa)	—	13	13
MFI (g/10 min)	—	1.1	1.2
Buss-Ko kneader			
Tensile strength (MPa)	10.20	9.50	8.2
Elongation at break (%)	340	360	370
Tensile modulus (MPa)	75	70	67
Flexural modulus (MPa)	162	155	148
Flexural strength (MPa)	18	16	15
MFI (g/10 min)	1.6	1.8	2.0
Banbury mixer			
Tensile strength (MPa)	9.10	8.30	7.50
Elongation at break (%)	360	360	380
Tensile modulus (MPa)	68	65	65
Flexural modulus (MPa)	150	145	140
Flexural strength (MPa)	15	13	13
MFI (g/10 min)	1.4	1.6	1.8

It is clear that the effect of reinjection moulding on properties of the blends is much more severe than recompression moulding (shear rate exerted during injection moulding is about one hundred times higher than in the case of compression moulding). Although the initial properties of injection moulded samples are higher than the compression moulded ones (see for example inset *Figure 13*), in the case of the former there is sharp drop in properties (e.g. tensile strength) after the first three reinjection cycles, while changes in the recompression moulded samples are minimal even after seven cycles. Moreover, the high shearing forces involved in the injection moulding process contribute to a lower melt stability (higher MFI values). Further, the extent of changes in mechanical properties with the increasing number of recycling operations of blends processed under both high (e.g. in Buss-Ko kneader) and low shear (e.g. in torque rheometer) forces is more pronounced in injection moulded samples compared to compression moulded analogues. For example, tensile strength of blends processed in a Buss-Ko kneader decrease by a factor of six after seven reinjection moulded cycles, while those processed in a torque rheometer decrease by a factor of two only at the end of the seventh cycle. This does suggest that although NR/PP blends can be reinjection moulded like thermoplastics, incorporation of antioxidants is essen-

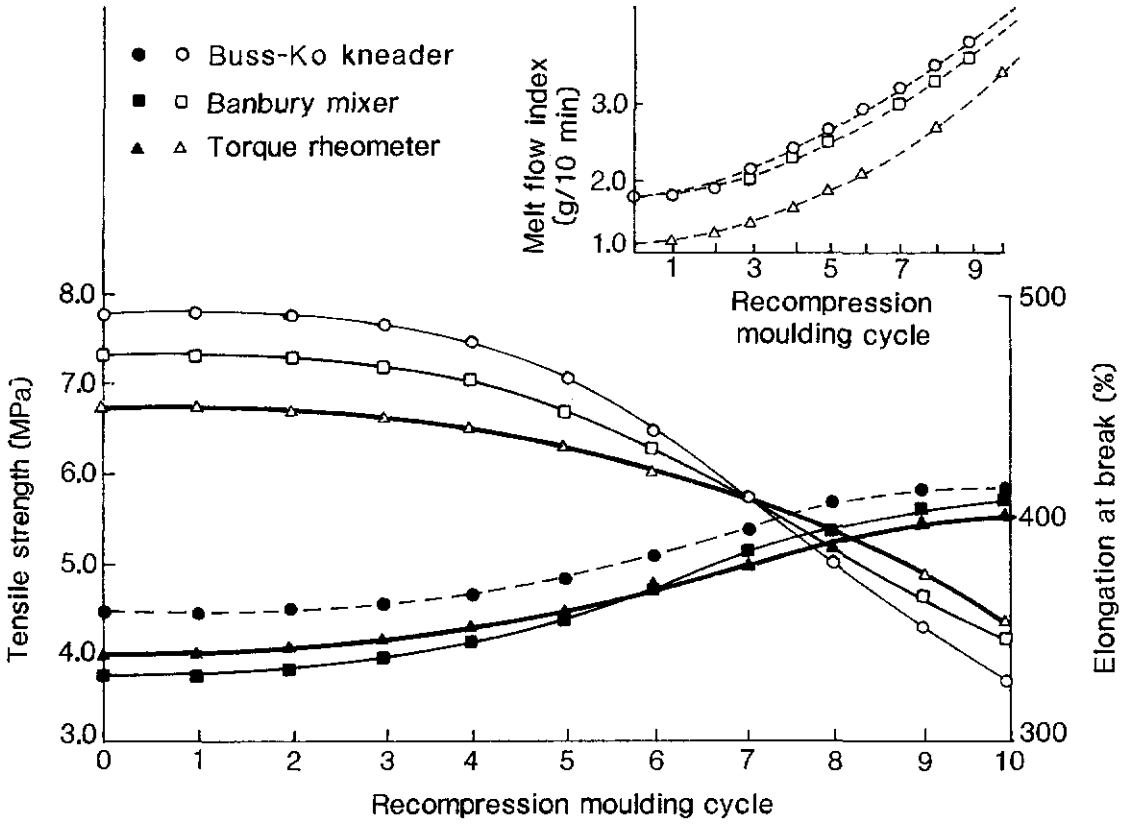


Figure 12. Effect of recompression moulding on tensile properties of films of NR/PP (60/40) blends containing 0.3 p.h.r. dicumyl peroxide processed in torque rheometer, Buss-Ko kneader and Banbury mixer. Inset shows changes in melt flow index of the same samples.

tial if the melt stability and the level of their initial mechanical properties are to be maintained throughout the reprocessing cycles.

The behaviour of the NR/PP blends under environmental conditions which may be encountered in service such as exposure to ultraviolet light and high temperatures was also examined. *Figure 14* shows changes in tensile properties and the build-up of carbonyl functional groups in NR/PP compression moulded films processed in a Buss-Ko kneader and in the torque rheometer. The high photoxidative instability manifested in the rapid growth of carbonyl index may be expected due to the high levels of NR (60%) in the blend. Higher shearing forces (in Buss-Ko) which contribute to high initial mechanical properties are also responsible for higher build-up of polymer hydroperoxides whose detrimental effect becomes very clear after the first few hours of irradiation; blends processed in a Buss-Ko kneader lose about 60% of their original tensile strength value after 11 h of irradiation while the torque rheometer-processed blends lose less than half (about 40% only) their original tensile strength under the same conditions.

Thermoxidative stability of injection moulded NR/PP samples processed in the three mixers is shown in *Figure 15*. In general, samples processed in Buss-Ko kneader offer higher mechanical properties than those processed in the torque rheometer. Blends in both cases

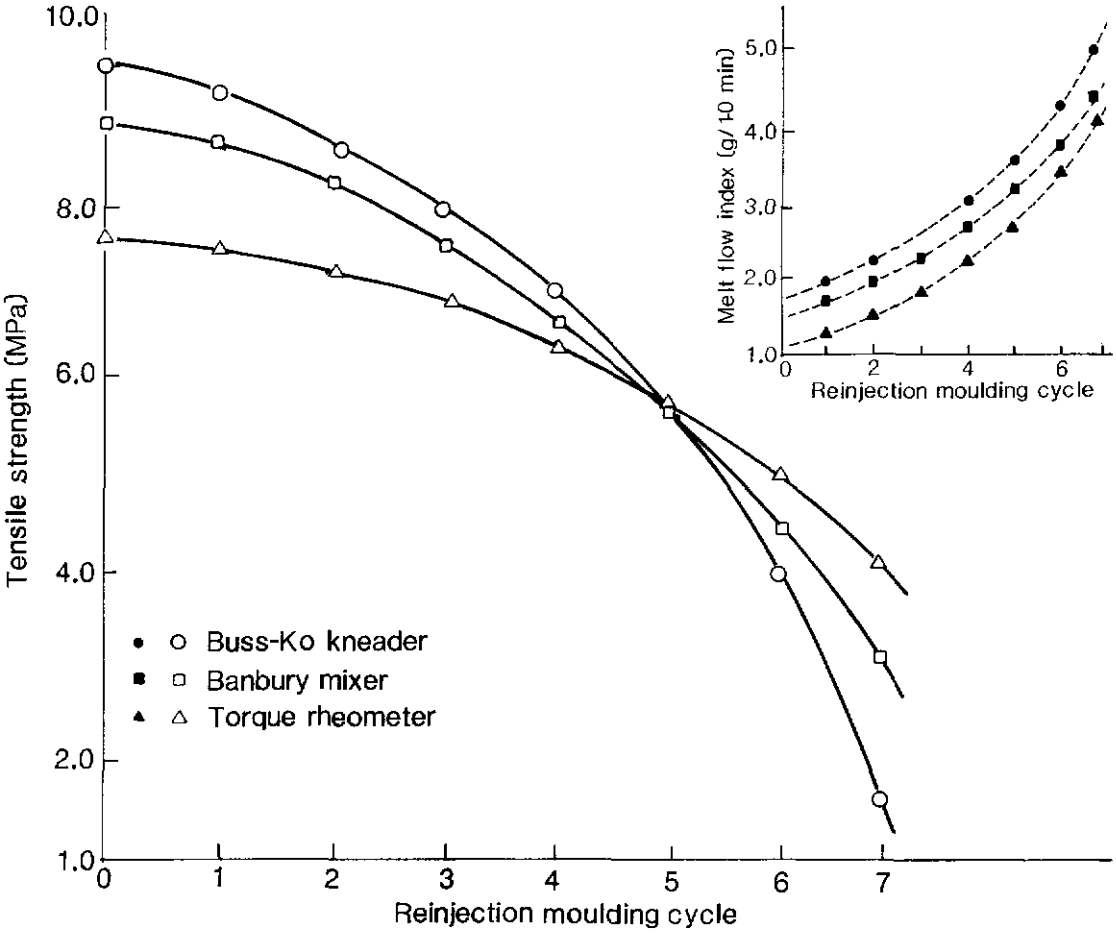


Figure 13. Effect of reinjection moulding on tensile strength and melt flow index (inset) of NR/PP (60/40) blends containing 0.3 p.h.r. dicumyl peroxide processed in torque rheometer, Buss-Ko kneader and Banbury mixer.

retain their properties for up to seven days at 70°C. However, at higher ageing temperatures considerable loss in tensile and flexural properties occur (see inset *Figure 15*); at 100°C for example, the tensile strength decreases by almost 50% of its initial value after three days only. The decay of properties at higher temperatures (e.g. 100°C) must be due to degradative processes in both NR and PP phases, while at lower ageing temperatures (e.g. 50°C) it is due mainly to NR. The effect of antioxidants and stabilisers on the subsequent in-service performance will be the subject of another publication.

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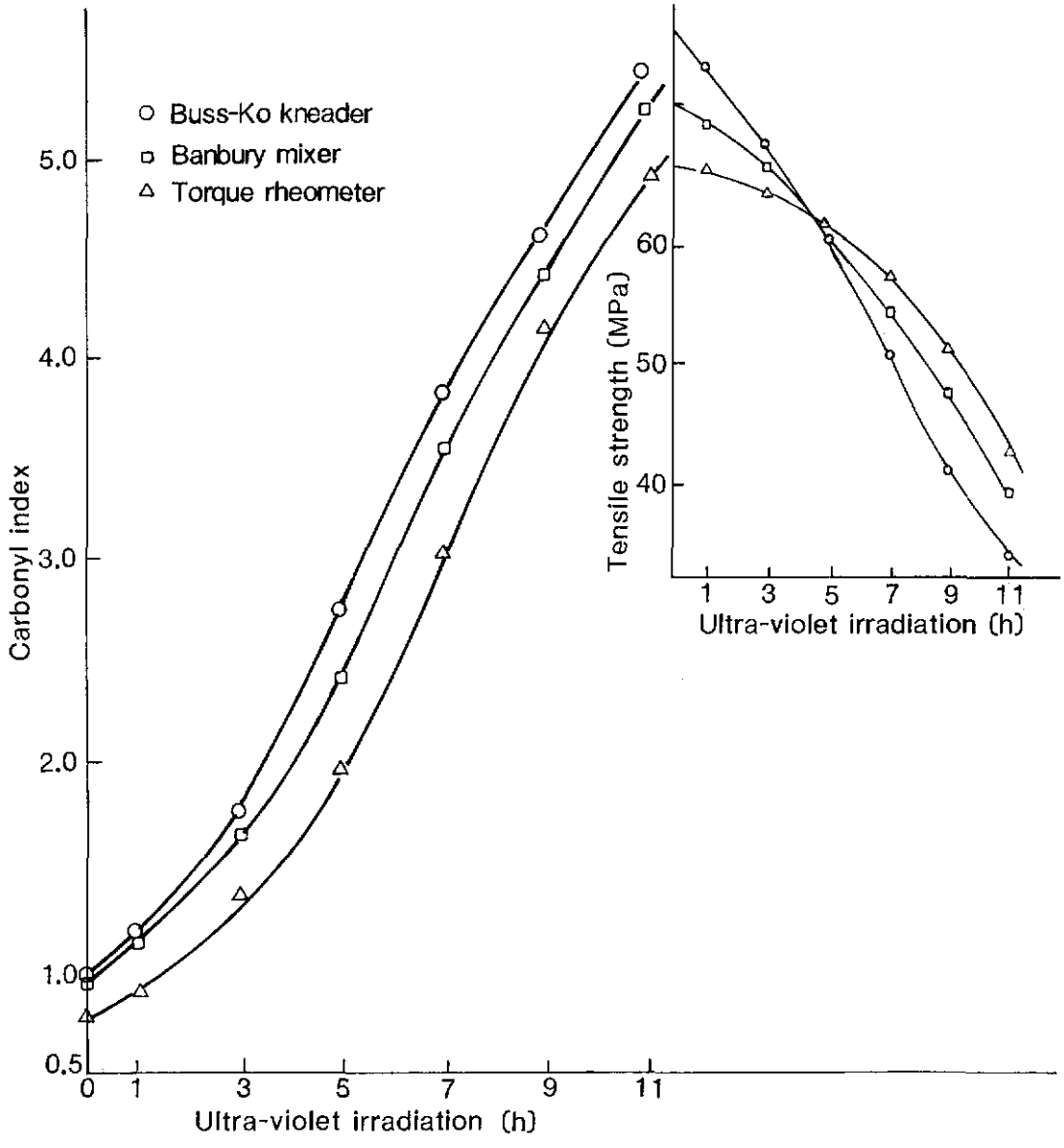

Figure 14. Effect of irradiation time on formation of carbonyl groups and changes in tensile strength (inset) of films of NR/PP (60/40) blends containing 0.3 p.h.r. dicumyl peroxide processed in torque rheometer, Buss-Ko kneader and Banbury mixer, (ultra-violet cabinet containing eight sunlamps and twenty-four actinic blue lamps arranged in symmetrical sequence was used as a source of irradiation).

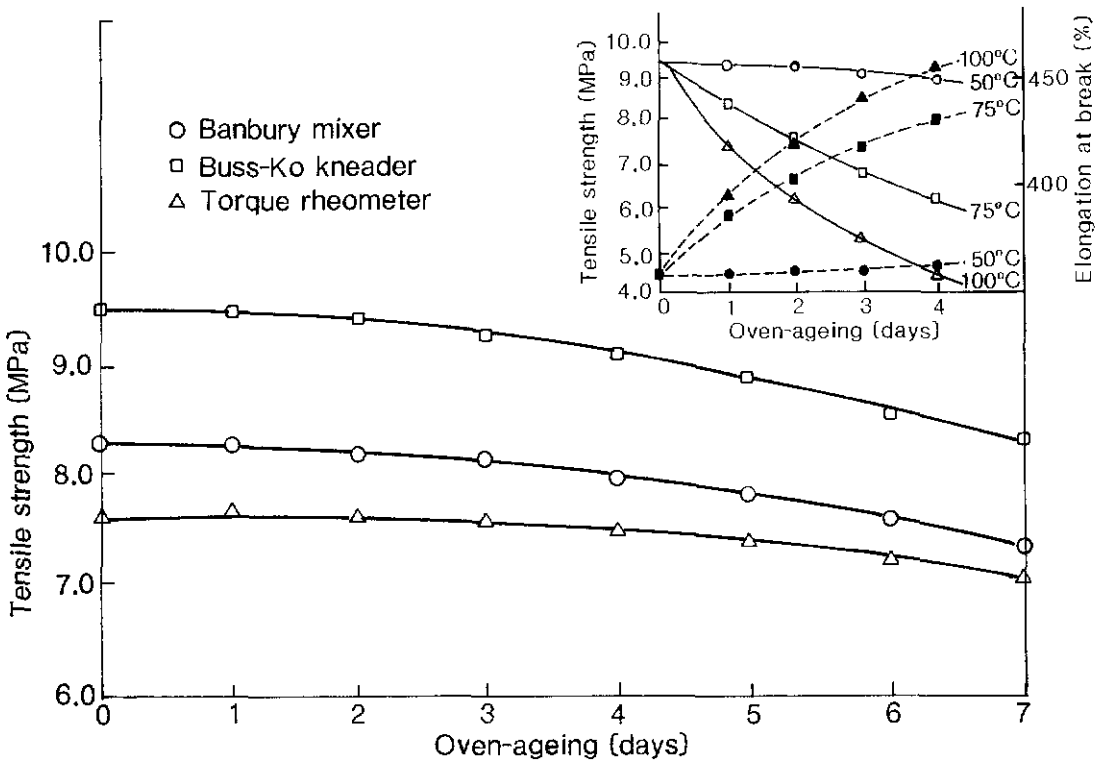

Figure 15. Effect of oven ageing (50°C, using single cell Wallace oven and air flow of 0.07 m^3 per hour on tensile strength of films of NR/PP (60/40) blends containing 0.3 p.h.r. dicumyl peroxide processed in torque rheometer, Buss-Ko kneader and Banbury mixer. Inset shows changes in mechanical properties at higher temperatures.

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