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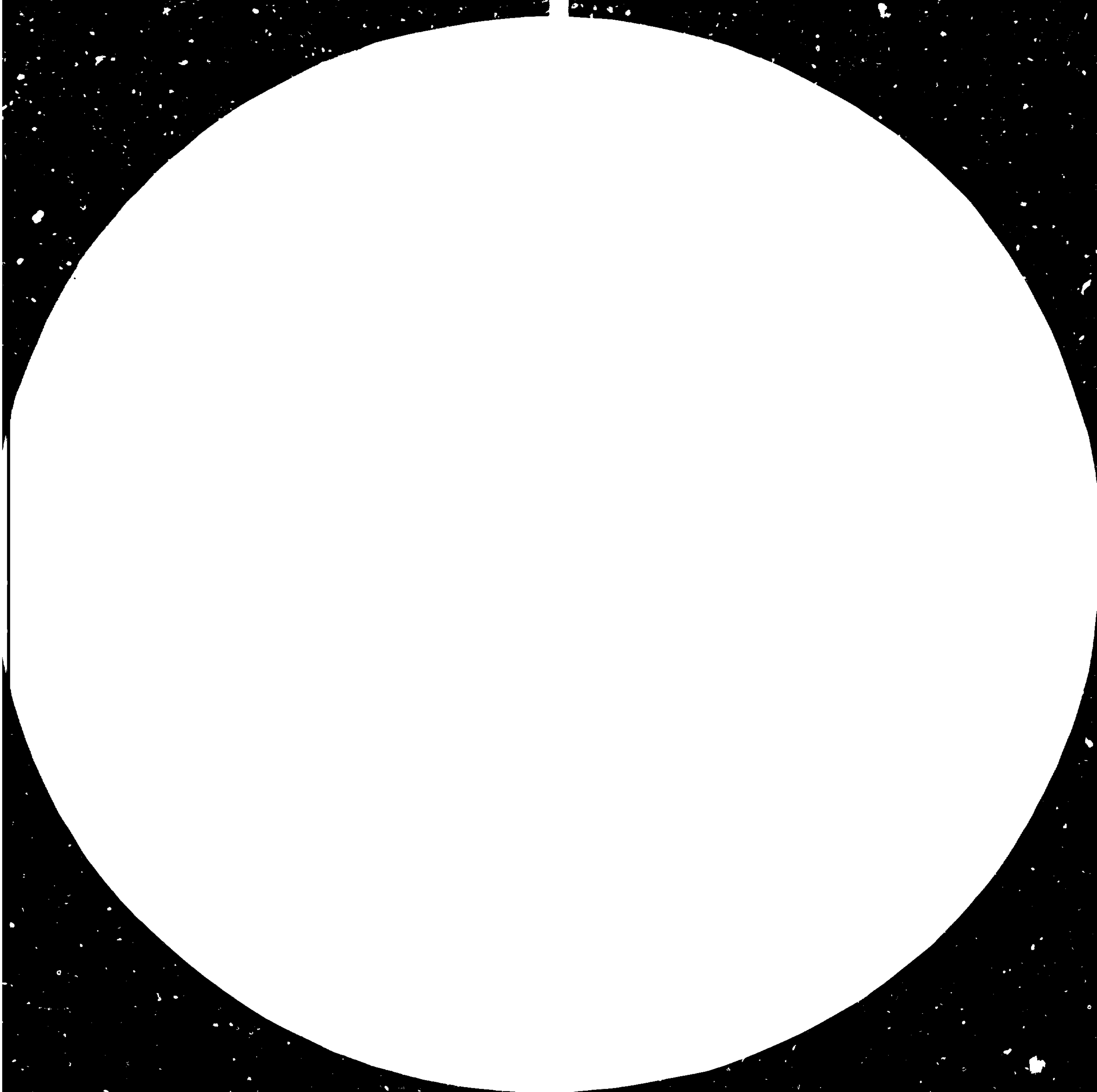

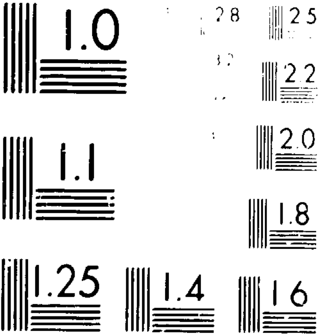
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Distr.
LIMITED

ID/WG.368/2
31 March 1982

ENGLISH

United Nations Industrial Development Organization

Petrochemical and Polymer Consultation Week
Porto Alegre, Brazil, 17 - 21 May 1982

COMPETITION BETWEEN NATURAL AND SYNTHETIC RUBBERS*

by

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TITLE COMPETITION BETWEEN NATURAL AND SYNTHETIC RUBBERS

Part 1	Price and Production of Natural and Synthetic Rubbers.
Part 2	Processing and Service Performance of Natural and Synthetic Rubbers in Tyres and Non-Tyre Applications.
Part 3	New Forms of Natural and Synthetic Rubbers. Their Effect on the Competitive Position of Natural Rubber.

General Introduction

Price, availability, processing cost and service performance are the components of competition. The three papers in this series will look at, in Part 1, the present position and future trends in price and availability of natural rubber and its synthetic competitors; in Part 2 the processing and service performance of NR and its competitors in tyres and non-tyre uses; in Part 3 the development of new forms of rubbers and the effect on the competitive position of NR.

Both natural and synthetic rubbers have their special place; ie some applications need the special properties of one or the other rubber. For many, indeed most applications, however, compositions can vary widely and still give technically satisfactory performance. It is with this competitive area that these papers are concerned.

Part 1 Price and Production of Natural and Synthetic Rubbers

1 Introduction

The major competitors for NR are SBR, (styrene butadiene rubber) and to a lesser extent BR, (polybutadiene rubber) since these three rubbers supply virtually all the rubber for tyres, and since tyres consume 70% of all rubbers. The price and availability of other synthetic rubbers are governed by the same factors which control the price and availability of SBR, ie oil price and competition for raw materials from oil. This paper therefore will deal mainly with NR and SBR, and will assume that other synthetic rubbers will follow the same price trends as does SBR.

Given normal industrial progress it is not possible that either NR or SBR can wholly supplant one another. The demand is huge, and, oil shortage or no, can be expected to grow, albeit slowly, over a considerable number of years. In this paper we shall look at the production capabilities and the likely relative prices of NR and SBR.

2 Synthetic Rubbers

Historically, increased industrialization has been linked with increased transport, not only freight transport but also cars as an integral part of improved living standards. It is to be supposed that this will continue, and that rubber tyred vehicles, propelled by whatever means, will continue to increase in numbers as world industrialization progresses. Since no forecast of future total rubber supplies envisages that sufficient natural rubber will be available to satisfy total demand, the position on the supply of synthetic rubbers can be simply stated. That is that the supply will increase to meet the increased demand. Though currently SR plants may be under-used, if and when demand increases the supply will increase also. It is to be expected too that the price of SR will be determined in good part by the price of oil.

Figs. 1 and 2 show the correlation over a rather short period between the production index for manufacturing industry[1] and consumption of total rubber and synthetic rubber. In developed countries the ratio of rubber consumption to industrial production index is almost constant, indicating that in these countries the industrial scene has changed quantitatively but not qualitatively. In developing countries on the other hand this ratio changes considerably between 1960 and 1977 reflecting a qualitative change to a more industrialized scene.

The changes in rubber consumption are closely paralleled by changes in the number of vehicles in the areas concerned, as shown in Table 1.

SBR is the major synthetic rubber because of its use in tyres together with BR, tyres consuming upward of 60% of total rubber production. If other synthetic rubbers are developed which supplant SBR (or NR) as a major tyre rubber then no doubt their price and supply will be governed by the same factors as govern SBR supply. So far no competitor has emerged, though periodic announcements are made that a new rubber has been developed to take over the tyre market. A more convincing one may have emerged in the recent Dunlop/Shell development of a rubber claimed to have both low rolling resistance and high wet grip.

3 Natural Rubber

Though in Brazil the price of natural rubber may be artificially possibly crosslinked SBR depending on the properties required of the NR substitute. Once lost to NR, this share of the market would be difficult to regain. A painstaking analysis has been made by the Malaysian Rubber Research and Development Board of the realistic maximum market share which NR could obtain on technical grounds modified by certain economic and political factors, and this share is proposed to be 35 - 40%[4]. Obviously to reach the required production will demand a major effort, and this effort is in fact being made, by new planting, by replanting with modern high yielding trees which will mature early, by yield stimulation and improved tapping techniques. The contemplated advance demands major organization and expenditure which must compete with other demands for investment to improve the economy of the countries concerned. All these schemes must necessarily take in existing and new small-holders who produce an increasing proportion of the total NR, and involve not only capital expenditure but also loans, both long and short term, to small-holders. The competition between NR and other crops is illustrated by the loss of area under rubber to particularly palm oil, where there has been a decrease since 1963 of about 30% in area of Malaysian estate rubber, because palm oil has been judged to be a more profitable crop.

In new planting, Malaysia plans to bring in about 40,000 hectares pa.[5] Thailand aims at a similar figure, and estimates that about 500,000 hectares are still available [5]. Indonesia is engaged on large rehabilitation, new planting and replanting schemes to the extent of about a million hectares, and so on [5].

Yield per acre has increased markedly as a result of clonal selection. Table 2 gives the yield per acre of various clones introduced in Malaysia determined, elsewhere it is currently determined entirely by demand, and therefore fluctuates to a considerable extent. This is inherent in the way in which it is marketed, and there seems little possibility of any real change at the present time. This price fluctuation may be ameliorated in two ways, ie by long term contract directly between producer and manufacturer, as is increasingly occurring, and in the future by a price stabilization scheme whose operation still has to be proven. In the long term, the price of NR has been rather above that of SBR (Fig 2), reflecting the fact that NR possesses some valuable processing properties for which manufacturers are prepared to pay, and natural rubber interests no doubt hope that will continue to be so. A stable price a little above that of SBR would be profitable to NR producers without offering too much incentive to manufacturers to convert to SR formulations, or to SR producers to develop new synthetic rubbers. Even at a price below that of SBR, NR can be profitably produced at least by the more efficient estates and larger small-holders.

Production of NR has increased enormously since the 1950s, and all NR producing countries have major plans for further increases. Various forecasts have been made of growth in total rubber demand and of growth in NR demand, and some of these are shown in Fig.4[2]. The most conservative, by T R Riedl,[3] forecasts an NR output of 5.3 million tonnes by 1990, requiring an additional area of one million hectares of mature high-yielding trees by that time. Such an increase is feasible, and was in fact achieved between 1963 and 1973. However, on this basis NR's market share would be only 27%, and this would encourage, indeed necessitate, the production of synthetic rubbers to substitute for the shortfall of NR. These synthetic rubbers could be improved isoprenic types, or SBR, or over the years, the latest yielding over 3000 kg/hectare. Replanting is a continual process as trees pass maturity and better trees become available. Table 3 shows the striking increase in yield of estates and small-holdings since 1950, a consequence of replanting and improved husbandry.

Yield stimulation, which at first sight might seem to be very suitable to small-holders' needs, has been particularly poorly taken up by small holders[6]. This technique calls for the application of ethylene to the tapping cut, liberated from the compound Ethrel (2-chloroethyl phosphoric acid), and can increase rubber yields by 50 - 80% with well nourished trees in tapping for more than fifteen years. In Malaysia in 1977 65 - 70% of estate area over fifteen years in tapping were so stimulated, but only about 2% of small-holder area. There are several reasons. The cost of Ethrel, plus the five times greater cost of fertilizer to feed the tree, cannot readily be met by poor families. This is exacerbated by the system of inheritance, leading to multiple ownership and consequent difficulty in arriving at a decision to spend or borrow the necessary cash. In addition Ethrel demands longer times to elapse between tapping and collection. Since many, or most, small-holders have other jobs, possibly at some distance from the plantation, they cannot afford to wait this longer time. Similar considerations apply to replanting. Since **most rubber comes from smallholders (Table 4) it is particularly important to remedy this situation.**

While Ethrel stimulation is capable of increasing yields by the amount indicated, it is not in fact being so used at the present time[6]. Both estates and small-holders seek to maximize profit, and one component in the equation is the tapping technique and the cost of that technique. Ethrel performs best on trees tapped at less than 100% intensity, and estates have therefore used this technique to reduce tapping, and therefore tapping costs, while maintaining or only slightly increasing output. Small-holders on the other hand, using family labour, will tap at higher intensity, and therefore risk overtapping and dry panels. Obviously, at unduly low NR price there is little incentive to use Ethrel, but at times of increased demand Ethrel will be a potent factor increasing yield and stabilizing price.

The sum of all these endeavours will be a rapid increase in the amount of NR available, and a figure of 10 - 12 million tonnes pa by the year 2000, compared with present production of about 3.5 million tonnes of NR and about 7 million tonnes of synthetic rubbers. Even this high postulated output of NR will not be more than 40% of total forecast demand for all rubbers, a factor which alone will seriously hinder greater use in major applications.

Even if NR output grows, but demand for all rubbers lags below the forecast, NR will still be in a strong position. Reduced costs of production by more efficient growing and tapping, and the realization that NR can be used more widely than at present should ensure that all the NR produced is sold at a reasonable profit. The technical aspect of NR usage will be surveyed in Part 2 of this paper.

Part 2 Technical Merits of NR and SR

Introduction

The choice of natural or synthetic rubber, or a blend is based on an assessment of price and performance in which processability as well as service life is taken into account. The major choice in tyres, and therefore in tonnage, is between NR and SBR, with or without polybutadiene. In non-tyre applications the manufacturer is faced with a wider range of required properties and with a larger number of competing polymers - NR, SBR, polychloroprene, EPDM, polyurethanes etc.

Certain properties are commonly required in various applications eg green strength, tack, adhesion, extrudability, reversion resistance, resistance to wear, tear and fatigue, oil and heat resistance, and this paper deals with NR's competitive position in terms of such properties, and of course require special rubbers. In others the range of choice is wider, and different manufacturers may reach different solutions in apparently similar applications. Table 5 illustrates the sort of choice which is being made in tyre treads in Europe and USA.

The cost can figure differently to the manufacturer or the user. The custom mixer making retread compounds will think very much about the relative costs of NR and SBR. His customer, the retreader, will see the compound cost as one element in a larger equation of processing cost, proportion of returns and customer acceptance. A transport fleet manager, the ultimate user, will be concerned with pence per mile and reliability, in which the initial compound cost is a very small part. The interplay between all these people will ultimately determine polymer usage.

Processing

Cold feed extrusion illustrates very well the behaviour of natural rubber *vs* SBR in processing. As is well known, natural rubber compounds which have been rested after milling become rapidly tough and difficult to process, but after for example a short time on the mill became much more processable. This is a thixtropic effect and the toughening/softening stages can be repeated several times. This is quite a serious disadvantage for natural rubber, and for instance may reduce the proportion of NR in a compound to 50% or 60% purely because of extrusion (or calendering) requirements, though a higher proportion of NR might give better service performance.

The importance will vary according to the type of extruder. In the worst case, of a ram extruder, Table 6 shows the performance of unworked NR and SBR tread compounds, the latter extruding much faster. If however both compounds are passed through the mill two or three times to give light mastication without much breakdown, the position is reversed and NR extrudes much faster. Some retreaders of high quality tyres, earthmover or aircraft, work like this.

Developments in extruders have ameliorated but not removed NR's disadvantage. High intensity screws, and the pin extruder lately developed, give the results typified in Fig 5. Such machines are becoming increasingly used in tyre and retread factories, will enable NR to retain its place in heavy duty treads.

An interesting and potentially important observation is that in oil-extended compounds, the use of oil-extended NR masterbatch (eg 75 NR/25 oil/no black) as made in Malaysia gives excellent extrusion in the ram extruder (Fig 6).

Green Strength and Green Adhesion

In many composite articles it is essential that the rubber phase should have adequate strength in the unvulcanized state to allow manipulation in building processes. The best known example is in radial tyre carcasses where during inflation of the uncured carcass the cords must remain uniformly spaced and must not penetrate the carcass rubber. Natural rubber has excellent green strength and is almost universally used for this application. The reason is believed to be connected with the ready crystallization of NR on extension. SBR and ordinary synthetic polyisoprene have poor green strength (Fig 7). Improvements are claimed for special PI rubbers, for emulsion SBRs containing crosslinks which are labile in processing, and for SBR containing some polystyrene blocks in the polymer chain, but so far none has reached wide commercial acceptance.

Adhesion of unvulcanized to unvulcanized rubber is of importance in many articles besides tyres. Natural rubber is acknowledged the best, and even in some circumstances may be too good. A special requirement for tyre retreading, which uses perhaps a million tons of rubber hydrocarbon a year, is adhesion of unvulcanized rubber to a vulcanized buffed surface. Fig 8 shows this green adhesion for blends of NR and SBR, where clearly there is a certain minimum proportion of NR which will secure good green adhesion. The importance of this is that it allows the tyres to be handled before curing with less risk of separation in the green state which might give rise to separation in service.

Cut growth and fatigue

A property of fundamental importance in many applications is the growth of small cuts under repeated stress, and the manner in which a rubber responds to different levels of stress. At a sufficiently high stress the rubber will tear, and in this sense fatigue and tear are opposite extremes of the same process, cut growth. It is however illuminating to think of them as separate effects which are of different importance in different applications, and will therefore affect the choice of rubber. This is particularly useful in considering wear, as will be seen below.

The cut growth behaviour of NR and black-filled SBR[7] is shown in Fig 9 (the addition of filler does not substantially alter the behaviour of NR). At low tearing energies (low stresses), cut growth is mechano-chemical in nature, and SBR is superior to NR. At higher tearing energies a region of catastrophic tear is approached, and under these conditions NR is superior. An important point is that under conditions where the stress is <u>not</u> completely relaxed during each stress cycle the fatigue life of NR is greatly improved, and this is of considerable importance in many engineering applications.

Typical uses illustrating the relative fatigue performances are:

a) sidewalls of bias ply tyres, where SBR is used under conditions of low strain

b) sidewalls of radial ply sidewalls, which operate at higher strains and demand more NR

c) rubber springs, where NR stands up to high tearing energies without catastrophic failure.

Vulcanization

Considerable strides have been made in improving both reversion resistance of natural rubber on extended cure, and its efficiency of vulcanization at high curing temperatures, thereby improving the performance of NR in such processes as continuous curing, injection moulding, and the curing of thick articles, including tyres. By decreasing sulphur level from the conventional 2.5 parts to about 0.3 parts or about 1.2 parts (the so called efficient vulcanization, EV, and semi-efficient vulcanization, S-EV, systems) with accompanying increase in accelerator level, reversion resistance is greatly improved as shown in Fig 10[8]. Recently a number of other improvements have been reported, and some of these, in addition to improving reversion, also increase efficiency of cure at high temperatures. These are as follows:

1 The use of a high stearic acid level in a semi-EV compound improves not only modulus reversion but also laboratory abrasion resistance (Table 7) on overcured samples[9]. The cause of the improvement has not been investigated. It is likely to involve some tendency of the system towards an EV system. The high stearic acid level does not adversely affect bonding. Truck tyres have been built and rig tested up to 25% overload without failure.

2 The use of a co-vulcanizing agent (Si 69, a tetrasulphide) as an addition to a black-filled semi-EV compound (1.5 phr of sulphur, 1.5 MBS accelerator)[10]. This is believed to act by slow co-vulcanization, replacing crosslinks lost by reversion. In addition to improving reversion it also significantly improves efficiency of cure at high temperatures (Figs 11 and 12). Other tetrasulphides do not act similarly.

3 Kempermann and Holzer have developed pentachlorophenyl sulphenamide compounds which are added to a semi-EV compound, and are thought to act as slow accelerators rather than co-vulcanizers[11]. These are even more effective in improving efficiency of cure at high temperatures but at the time of writing are development products only.

4 All the above are improvements to an established sulphur system. A new departure has been the development of urethane crosslinking agents (trade name Novor)[12]. These introduce a totally new type of crosslink which is thermally very stable and is almost immune to reversion or to loss of efficiency or raising cure temperature. A valuable aspect is that they can be combined with sulphur systems, when pronounced synergism is found. This allows the proportion of both urethane and sulphur crosslinking agents to be reduced, while retaining valuable reversion resistance. A particular advantage is the retention of fatigue resistance after ageing (Fig. 13). They have been successfully evaluated in a large number of evaluations and have reached commercial acceptance in a number of products, for instance engine mounts for hot environments. An interesting application is in heating pads for repair of earthmover tyres, where NR/Novor is giving several times the life of the silicone rubber formerly used.

Wear and other properties of tyres

Most rubber articles fail through wear, ie tyres, conveyor belts, shoe soles. It is well known, particularly for tyre treads, that if two rubbers are compared under different conditions reversals of ranking may occur. Grosch and Schallamach in investigations of the mechanic dependence of wear have identified two abrasion mechanisms, fatigue failure and tensile, or tear, failure, which depend on type of compound and test conditions, ie pressure on the abrading surface, and temperature[13].

The effect of temperature on tread wear has been investigated by Grosch[14] who found that the effect of speed, manner of driving and ambient conditions could be compounded into the effect of one parameter, the temperature of the surface of the tyre, as shown in Fig 14 . At high tyre surface temperatures, where fatigue is more important, SBR and SBR/BR blends are superior to NR and NR/BR blends. At low tyre surface temperatures NR is superior.

The effect of pressure as found by Grosch and Schallamach[13] is shown in Fig 15 where reversals of ranking are observed when NR and SBR are abraded on surfaces of different sharpness, ie different pressure on the rubber. On smooth surfaces SBR is superior, but with increasing pressure NR becomes superior.

These two effects are reflected in the use of NR or SBR in different treads. Car tyre treads are normally synthetic rubber, because they operate under light loading ie fatigue conditions. The larger the tyre the greater the load, and therefore the greater the use of NR in large truck tyres and even more in aircraft and earthmover tyres, to withstand tearing. Other factors such as heat build-up are obviously important.

An important illustration of the effect of conditions on wear is given in Table 8 which gives the results of wear trials of winter tyres [15].

These tyres were tested on hire fleet cars in Montreal, Minnesota, New England and Germany. Under the coldest conditions, Montreal, the NR treads were considerably superior, but in the more temperate conditions of New England and Germany they were equivalent. Results such as these have led to the use of NR not only in winter tyres (up to 75% NR), but also in the more important of the new all-weather tyres, up to 30% NR.

The tyre tests mentioned above were run not just in the depths of winter, but on to May or June, and suggest the possibility that general purpose car tyres could contain substantial proportions of NR without loss in current wear standards in normal service.

Conveyor belts with NR and SBR covers have shown a marked superiority for NR in severe conditions including arc conveying in a steel works or coarse basalt in a quarry, Table 9.

Other important properties of tyre treads are wet grip and groove cracking. In both respect oil-extended NR/BR treads are as good as the usual OESBR/BR.

A topic of current importance is energy consumption, and the effect of various factors, ie radial or bias-ply, inflation pressure and

and polymer type, E.J. Buckler (ref. 5). has published data on the effect of construction and tyre pressure, showing the considerably greater importance of the gasoline consumed in rolling the wheels than the petroleum equivalent of the rubber consumed in the life of the tyre. Data from his paper are contained in Table 10. If the SBR in the tread were replaced by NR, at similar compound composition, ie oil-extended to the same level, a saving of about 20% of the gasoline used in rolling the wheels would be expected. Since NR takes less energy to produce (Tables 11 and 12) there would be a dual saving in energy consumption which would amount to about 3% of the gasoline used to move the <u>car</u>. This would be a worthwhile saving on a large scale, though the private motorist would not notice it. Though wet grip would be reduced slightly, this would not be significant on the radial-ply tyre. In fact this replacement is unlikely to occur on a large scale simply because of supply, but also because of the introduction by Dunlop and Shell of a synthetic rubber (undisclosed) claimed to have good wet grip <u>and</u> low rolling resistance.

Fluid resistance and ozone resistance of blends

As is well known, natural rubber does not take up water to any significant extent, but is not normally considered as an oil-resistant rubber. Two points can be made, first that in a thick article, the rate of diffusion of viscous oils is so slow as to be negligible[16].

and natural rubber is of course widely used in engineering applications where it is contaminated by oil, such as engine mounts, vibrating screen mounts, railway wagon springs and the like. Second, natural rubber can be blended with polychloroprene or acrylonitrile-butadiene rubbers to give useful compounds [17,18]. For example up to 20% of NR may be substituted for polychloroprene with little if any adverse effect on properties which would reduce service performance (Table 13). At higher proportions of NR oil resistance is reduced (Fig 16) but water resistance is improved (Fig 17). Ozone resistance varies with hardness and with NR/CR ratio (Fig 18).

Blends of NR with acrylonitrile-butadiene rubbers may similarly be made. Volume swelling and ageing is shown in Figs 19 and 20 . A 20 NR/80 ABR blend may be used for most CR applications except where high resilience is required (Fig 21). In many ABR applications such a blend may be used except where maximum oil resistance is required, and indeed the slight swelling shown by a 20 NR/80 ABR blend, against the slight shrinkage of 100% ABR, may be advantageous in sealing applications.

Engineering Applications

Rubber springs, bushes etc require adequate strength and fatigue resistance, low creep and compression set, and usually low hysteresis. As far as possible these properties should have minimal dependence on strain amplitude, frequency of deformation, and temperature. Increasingly, reproducibility of properties within close limits is being required. Natural rubber can fulfill these total requirements possibly better than any other rubber, though naturally there will be special circumstances which will require particular synthetic rubbers.

To obtain reproducible modulus within say 5% of the mean mastication and mixing must be carefully controlled. By using viscosity controlled natural rubber such as SMR 5CV premastication can be eliminated, and uniform compound viscosity and vulcanizate modulus more easily achieved. NR can be compounded to low hardness and still retain excellent tear and fatigue resistance (Table 14)[19]. Because of its low glass transition temperature (Table 15) its modulus is constant over a temperature range of -20°C to +100°C, and/or over a wide range of frequencies[19].

Low creep rate is important in many applications. Very low creep can be obtained by the use of low sulphur (EV) systems (see section on Vulcanization) in which all the added components and their reaction products are fully soluble in the natural rubber at room temperature, ie the operating temperature for most of such applications. In addition, such systems have reproducible modulus, good dynamic properties and better fatigue resistance than most other EV systems.

In bridge bearings and similar applications NR's only current competitor is polychloroprene. As shown in Figs 22 and 23, the principal disadvantage of CR is in low temperature resistance and in water absorption, giving increased creep.

Part 3

Introduction

There are a number of developments in the rubber industry which pose a challenge for natural rubber. Injection moulding has become prominent in the last 15-20 years but has still to realise its full acceptance. Thermoplastic rubbers are becoming slowly more popular. Powdered rubbers and their possible processing advantages have still to make any major progress. All these change NR's competitive position in some way, and NR must develop and be modified to retain its places in any new scene which becomes important.

Injection moulding[20,21]

When the modern interest in injection moulding began, in the 1960s, it was assumed by some manufacturers that natural rubber would be unlikely to be usable in this technique because of the high curing temperatures involved. This is very far from being the case, and natural rubber is widely used, generally without requiring special compounding to meet injection moulding conditions. Most natural rubber compounds designed for compression moulding and having a scorch time of ten minutes or more at 150°C can be satisfactorily injection moulded provided machine controls are properly adjusted. Indeed, one advantage of natural rubber is the substantial heat build-up at the injection stage, so that the compound arrives in the mould at near mould temperature while being adequately safe in the barrel.

It is good general advice that in adapting a compound for injection moulding scorch safety should be ensured by adding cure inhibitor, then machine variables should be set before further varying the compound. For example Fig. 24shows the effects of barrel temperature in a Rep machine in which the compound is plasticized by a screw, fed into a separate injection chamber then injected by ram through a nozzle into the mould. Fig.25 shows the effect of injection pressure or injection time and injection temperature. By proper selection of these and other machine variables it is possible to arrive at the shortest mould time consistent with adequate resistance to scorch. Fig.26 gives an example.

Choice of vulcanizing system is governed by required scorch resistance, and by the maximum rate of cure once the mould is filled. In many cases, especially for thin articles of uniform thickness, conventional sulphur systems are satisfactory up to a mould temperature of 180°C.

For thick articles, and those of varying thickness, EV or semi-EV systems are generally satisfactory. The cure inhibitor, Santogard PVI (N Cyclohexyl thio-phthalimide, Monsanto) is not very effective in EV or semi-EV systems, but scorch time can be adjusted by varying ratio of primary to secondary accelerator. All sulphur systems lose efficiency at high temperatures (Fig. 27). This may be met by

a. Increasing the level of all vulcanizing components (Fig.28);

b. In conventional systems by increasing the level of primary accelerator (Fig. 29); and

c. Using a mixed Novor/sulphur system (Fig.30).

Thermoplastic rubbers

There are many versions of thermoplastic rubbers, whose principal attraction of course is the elimination of the vulcanization step and the ability to be handled in plastics processing machinery. In principle a range of hardness is available either through variation of the polymer composition or through normal compounding, addition of fillers, oils, resins, etc. Such compounding requires high shear equipment, eg internal mixers or double screw compounding extruders.

Of the four types of thermoplastic rubbers, two are cheap enough to be potential competitors with general purpose rubbers. These are the styrene block polymers, developed in the early 1960s, and the thermoplastic olefinics, in the early 1970s. The latter consist of physical blends of EPDM rubber and polypropylene in which the rubber phase is partially crosslinked.

The various thermoplastic rubbers have competed with vulcanizable rubber and PVC in applications such as shoe soling, wire and cable insulation, automotive components, and some mechanical goods. Total tonnages are not large, about 200,000 tons in 1981, and their principal limitation is that at moderately high temperatures they soften. For example, sports shoe soles in strenuous sports such as squash may develop sufficient frictional heat to soften soles of styrenic block copolymers though satisfactory for less demanding sports.

Natural rubber can be blended with crystalline polyolefins such as high density polyethylene and/or isotactic polypropylene[22]. The blending is easily performed in internal mixers in which the temperature of the mix can be raised above the melting point of the polyolefin (165°C-175°)

early in the mixing cycle. After blending the batch while still hot must be passed once through a mill to give a sheet 4-8 mm thick suitable for granulating.

In the blend the hard microcrystalline regions of the polyolefins play the part of crosslinks. A degree of light crosslinking of the rubber phase improves flow, shrinkage and surface finish, and this is accomplished by adding an organic peroxide in the mixing cycle sufficient to gel the rubber without impairing thermoplastic properties. This addition is preferably made after the polyolefin has melted (indicated by recorder temperature or ammeter). A mix cycle in a steam heated internal mixer (eg K2A Intermix) is as follows for a wide range of NR/PP/black combinations:

0 minutes - add black, polypropylene, NR
3 - 4 minutes - add dicumyl peroxide
6 - 7 minutes - add antioxidant
6.5 - 7.5 minutes - dump

Machine conditions for injection moulding the blend are shown in Table 16. The minimum extruder, injection chamber and nozzle block temperature is 190°C. Injection pressure should be high enough to give the shortest possible injection time; and should be held long enough to prevent sink marks due to thermal contraction of material in the mould. Mould temperature should be high enough to avoid flow patterns, usually 90-100°C. Table 17 gives typical physical properties of some blends covering a good range of modulus and strength. The major factor governing properties is the ratio of NR to polyolefin. General purpose blacks add little to the properties of a peroxide modified mix, but antistatic materials suitable for painting by electrostatic spraying can be made by adding conductive blacks. Weather and ozone resistance are excellent. Adhesion of paints requires simple surface chlorination, and is better the higher the proportion of NR in the blend.

These materials can be prepared by the rubber manufacturer, given that adequate mixing temperature can be attained. In large mixers, even unheated, this has been no problem. The main application is seen to be in vehicle body components and in footwear, a stiff flexible component, suitable for a working range of -30 to +70°C. They have a small temperature coefficient in this range which makes these competitive with or superior to polyurethanes or EPDM.

Powdered rubber

General purpose powdered rubbers have been of interest for some years, for the possibility that they might offer cheaper or better processing by current or novel processing methods. Published results are conflicting, but in general they give little or no advantage in conventional processing, and only limited promise in the new continuous mixer extruders developed by Farrel Bridge (the MVX) and Werner-Pfleiderer (the EVK). In these novel machines, and also in injection moulding there is the possibility of using a comparatively light weight dry blender to mix powdered rubber with other ingredients, then passing the result to an injection moulding machine or to a continuous mixer extruder followed by a continuous curing bath. A particular advantage might be the ability to set up a relatively small operation, too small to use economically the conventional mixer and take-off arrangement.

Natural rubber has particular disadvantages in this scheme. First, its natural tack is so good that agglomeration in transit or in store is not easy to prevent. Second, if powdered NR is made in the producing country, the major consumers are so far away that freight costs on a bulky material are very high. If however powdered or fine crumb rubber were to be made and <u>used</u> in the producing country then disadvantages would be minimized. It is possible to visualize a hypothetical set up where fine crumb is produced by one of the existing methods in a rubber processing plant and would receive a minimal coating of talc, zinc stearate or other powder to keep the rubber particles separate while drying. The powder could then be passed immediately to a dry blender then to an internal mixer or mixer extruder to produce masterbatch for sale or for further processing on site.

In practice at present there are two forms of particulate NR available, ie the spray-dried latex called 'Crusoe'[23] and material made from bale rubber by granulation.

Crusoe rubber is a fine powder of particle size 100% less than 4 mm, coated with fine silica as a partitioning agent. The premium is high, but it has gained limited commercial acceptance in the field of solvent based adhesives and similar operations, where solution times are considerably reduced compared to the usual dissolution of chopped crepe.

Wheelans[24] has investigated the granulation of bale rubber and its subsequent treatment by a number of techniques, and has costed each process. He estimates the cost of granulation as about £100/t, and this of course has to be recovered in subsequent economy of processing some of his processing cost comparisons are given in the following Table 18. The figures given are those judged most likely to be found in practice, and clearly show no advantage for powdered natural rubber over bale rubber. Even in the most favourable case no advantage could be found.

At present the only outlet for powdered or granulated general purpose rubber appears to be in special compounds which are for some reason difficult to mix, e.g. very hard compounds where heat history may be reduced by using powdered rubber in a continuous mixer-extruder.

Table 19 compares the cost of producing unvulcanized compound in strip form using a Banbury plus mill on a powder blender plus MVX. Wheelans works on a number of possibilities in terms of labour, mixing cycle etc, but Table 19 gives the most likely cost estimates showing that the Banbury is more economical. Note that the granulation cost is not included, and that even omitting this cost the Banbury using bale rubber is cheaper.

Secondly, Wheelans calculated costs of producing a vulcanized extrudate either by a Banbury/mill/cold feed extruder/LCM bath or by a dry blender/MVX/LCM bath where the extrudate from the MVX is fed straight through the LCM bath. In the best possible case, taking into account all the savings in building and machinery costs, he estimates that the saving in production cost using the MVX only just compensates for the cost of granulation.

Similar conditions will apply to SBR, though in this case the overall costs might be even less in favour of the powder route.

Conclusions

The most important conclusion is that there is no possibility that either NR or SR will exclude each other from the major markets, given any reasonable estimate of the role of industrial expansion. The special properties of NR, and its ability to be produced at a lower cost than SBR, ensures its place in its present established uses, though it may be subject to some competition from eg special high green strength elastomers. The special properties of SBR will assure its place in car treads, but if prices rise too high, NR could be used in substantial proportion with little loss in wear and grip, particularly in temperate climates, and with an advantage in rolling resistance which would outweigh any extra rubber or processing cost. Potential newcomers to the scene are the recently announced high grip/low rolling resistance tread rubber of Dunlop and Shell, and the possibility of injection moulded polyurethane tyres. The former will compete at least initially with SBR, and in truck tyres is a completely undisclosed quantity. Polyurethane tyres are likely to be introduced as tractor tyres in the next few years; published data is sparse, and their cost competitiveness with a modern radial tractor tyre is still to be established. Their greatest demerit appears to be in wet grip.

In the complex scene of non-tyre uses natural rubber has lost some important markets with little hope of regaining them, and is all the time under pressure as a general purpose rubber from a variety of synthetic rubbers which each have their own special attributes. For the many applications which demand wear and tear resistance, green strength, resilience or low temperature resistance NR is still the best choice. NR's heat resistance has been substantially improved and blends with oil resistant polymers have been readily acceptable in an increasingly cost conscious market.

Powdered rubbers do not seem to be of widespread advantage, but continuous mixing and injection moulding may make use of them in a relatively small factory where conventional large scale mixing cannot be economically installed. Thermoplastic natural rubber/polypropylene blends are well suited to current and future applications for a general purpose material of this type. Both thermoplastic and powdered rubbers should be considered in a rubber producing country which is expanding its manufacturing industry.

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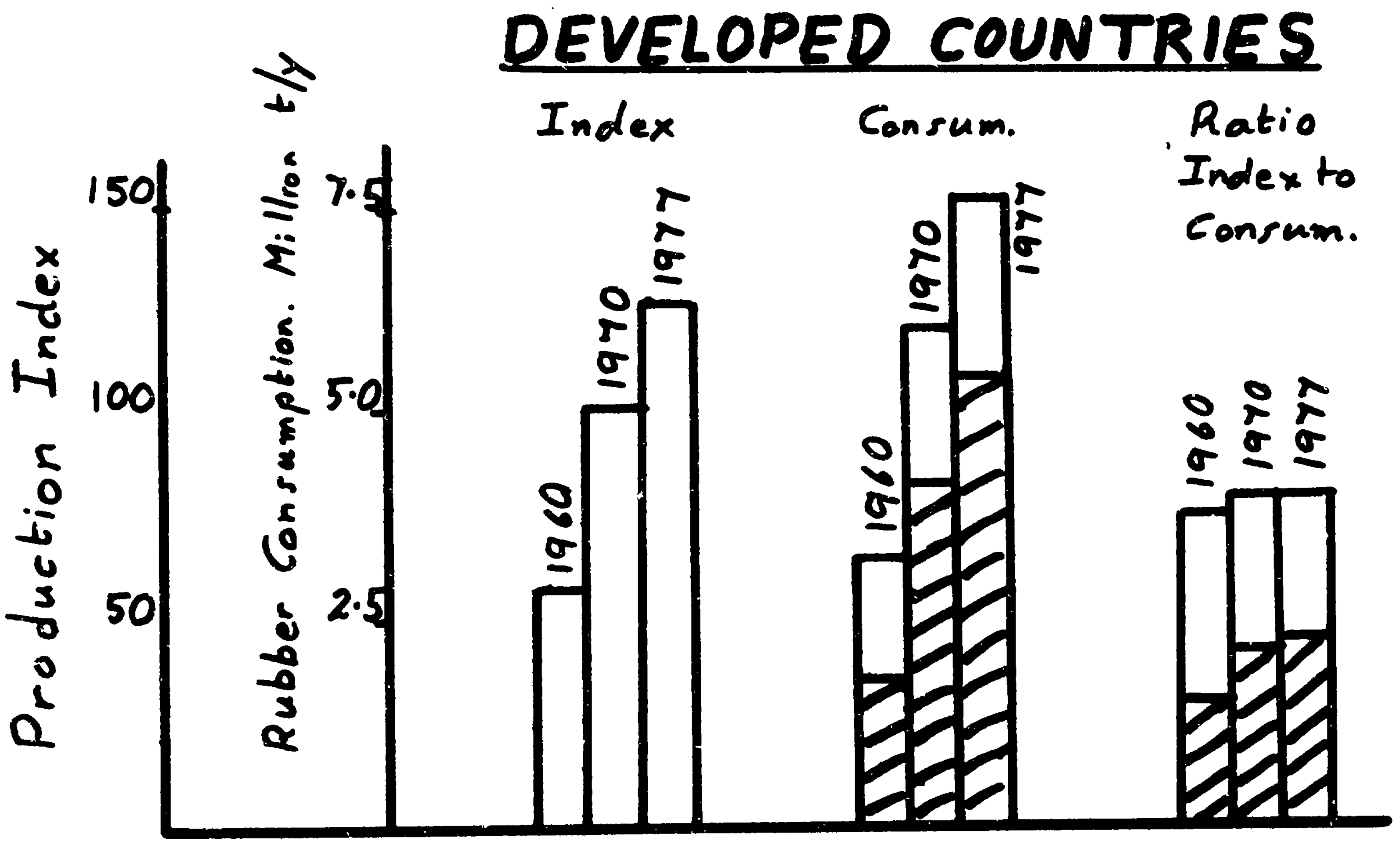


FIG. 1. Production index, SR consumption ▨, and total rubber consumption ☐ in developed countries.

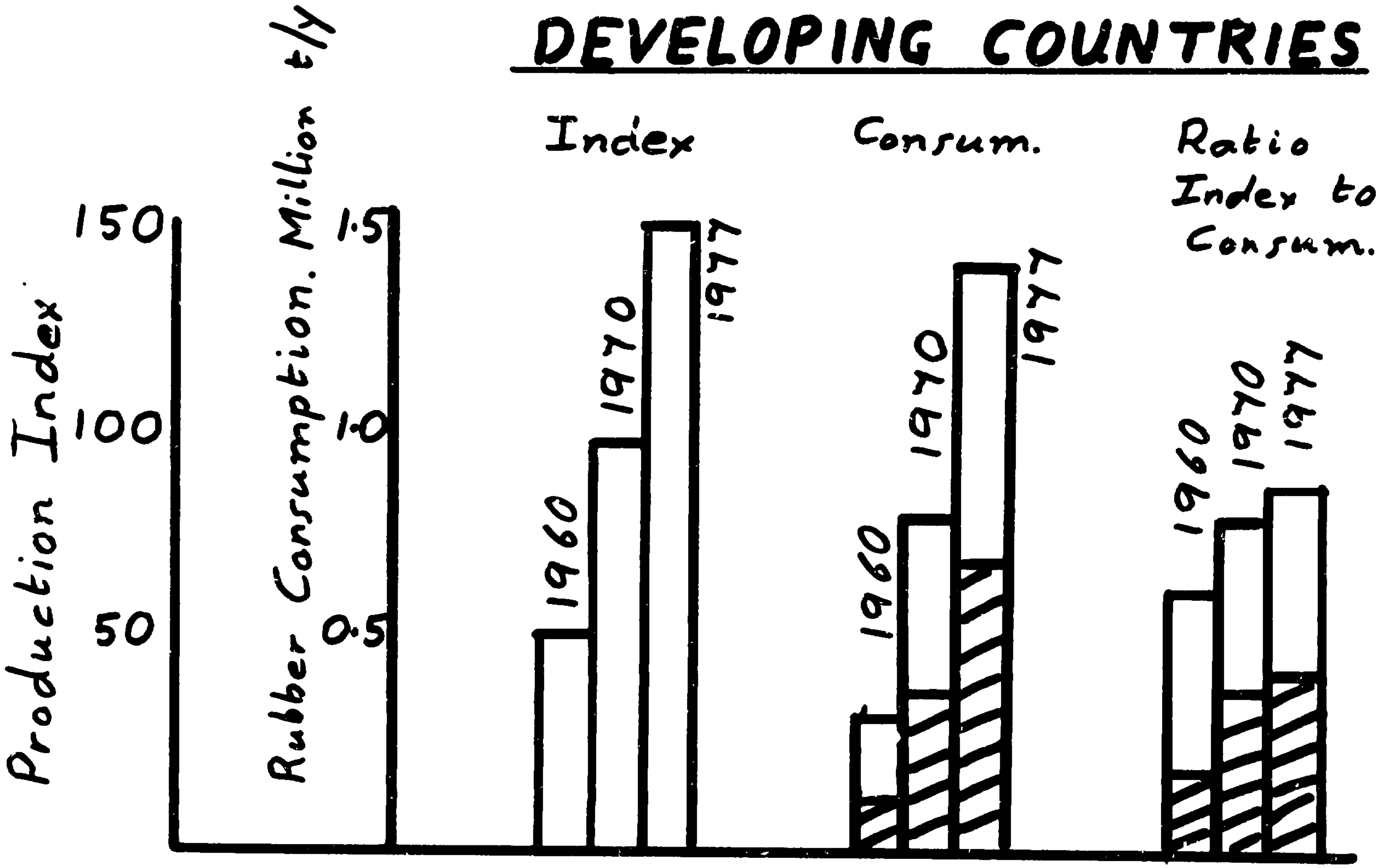


FIG.2. Production index, SR consumption ▨, and total rubber conrumption □ in developing countries.

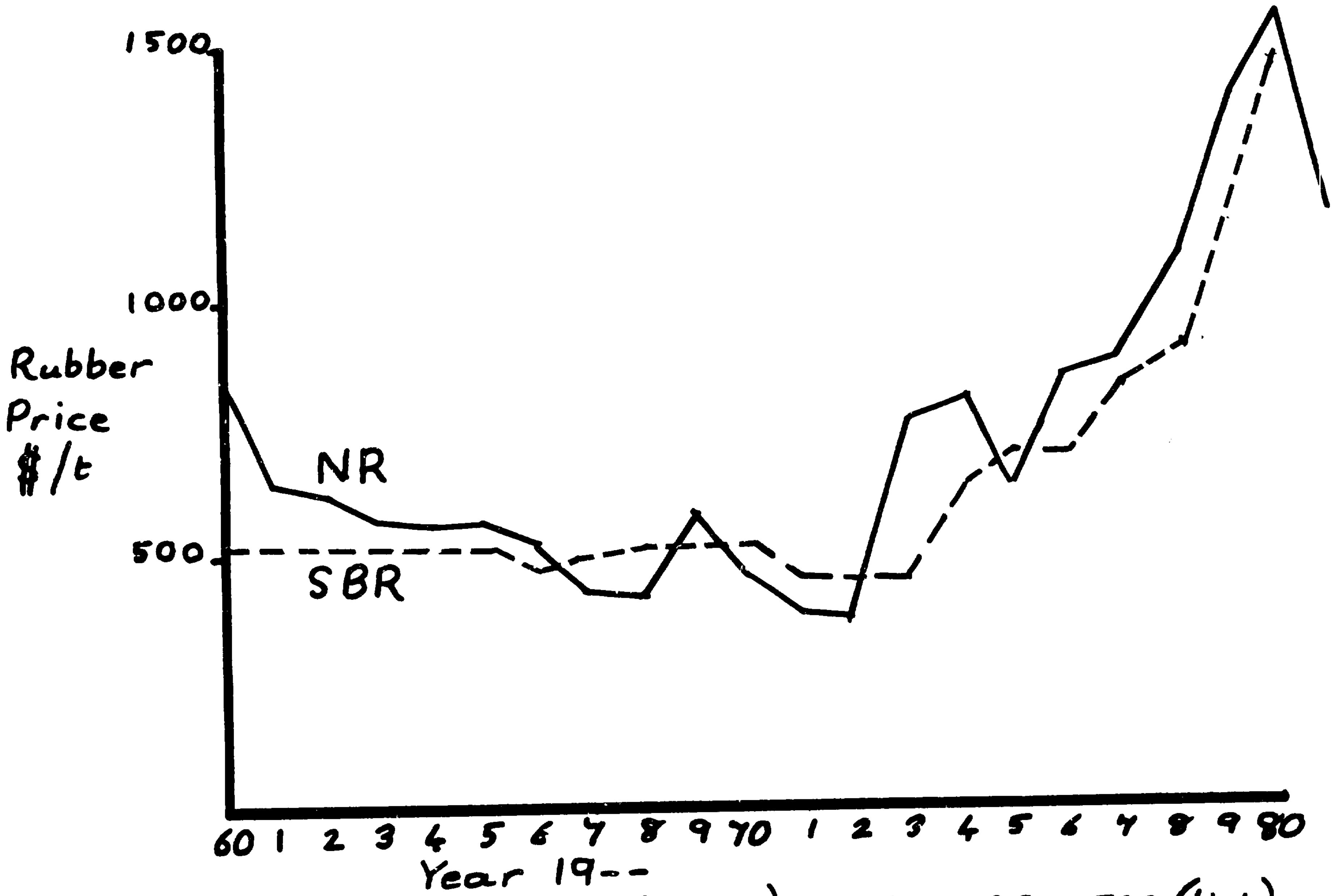
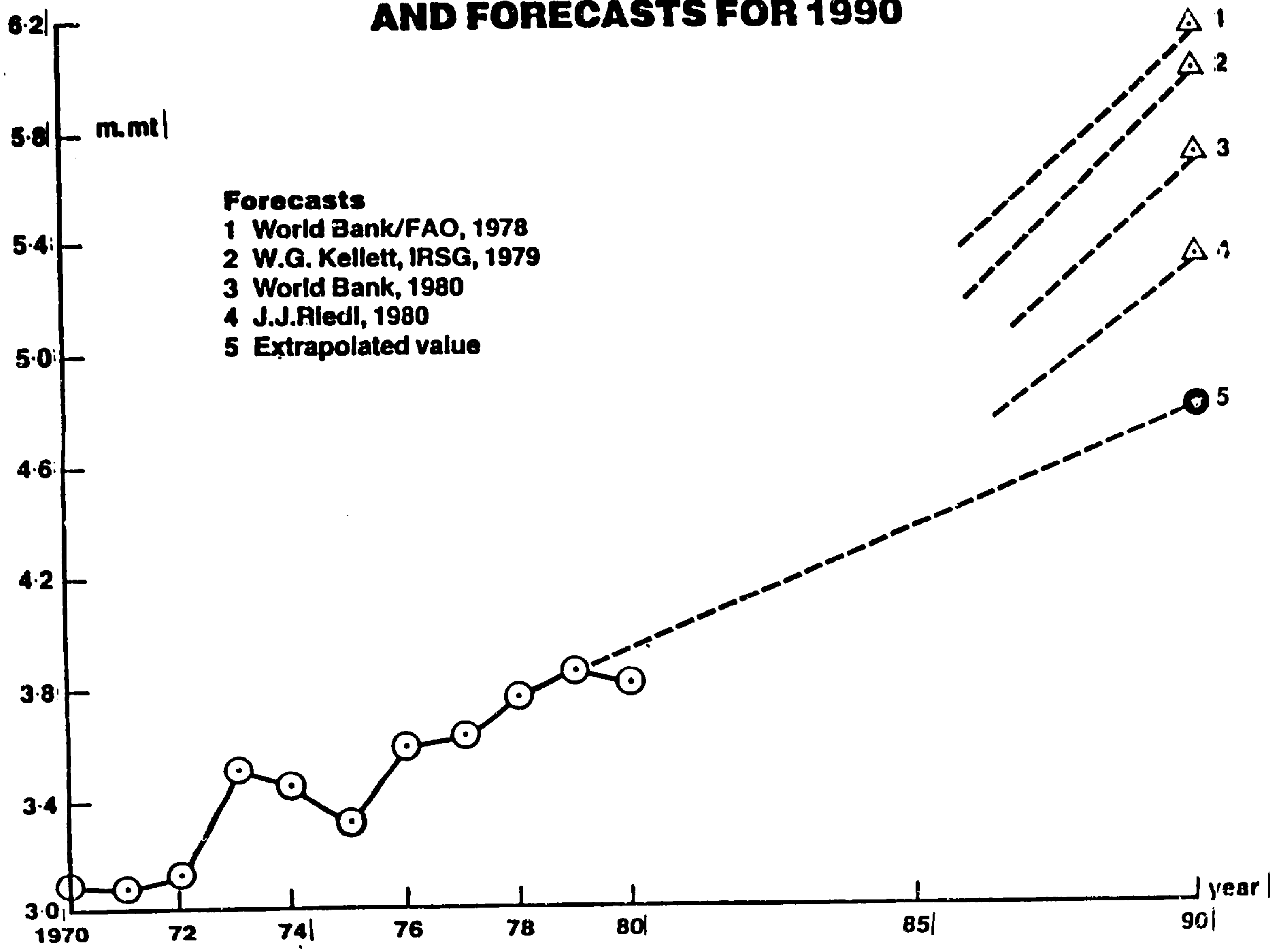


FIG 3 Price of NR (RSS 3) and SBR 1500 (List) 1960 – 1980

FIG. 4. PRODUCTION OF NR, 1970-80 AND FORECASTS FOR 1990



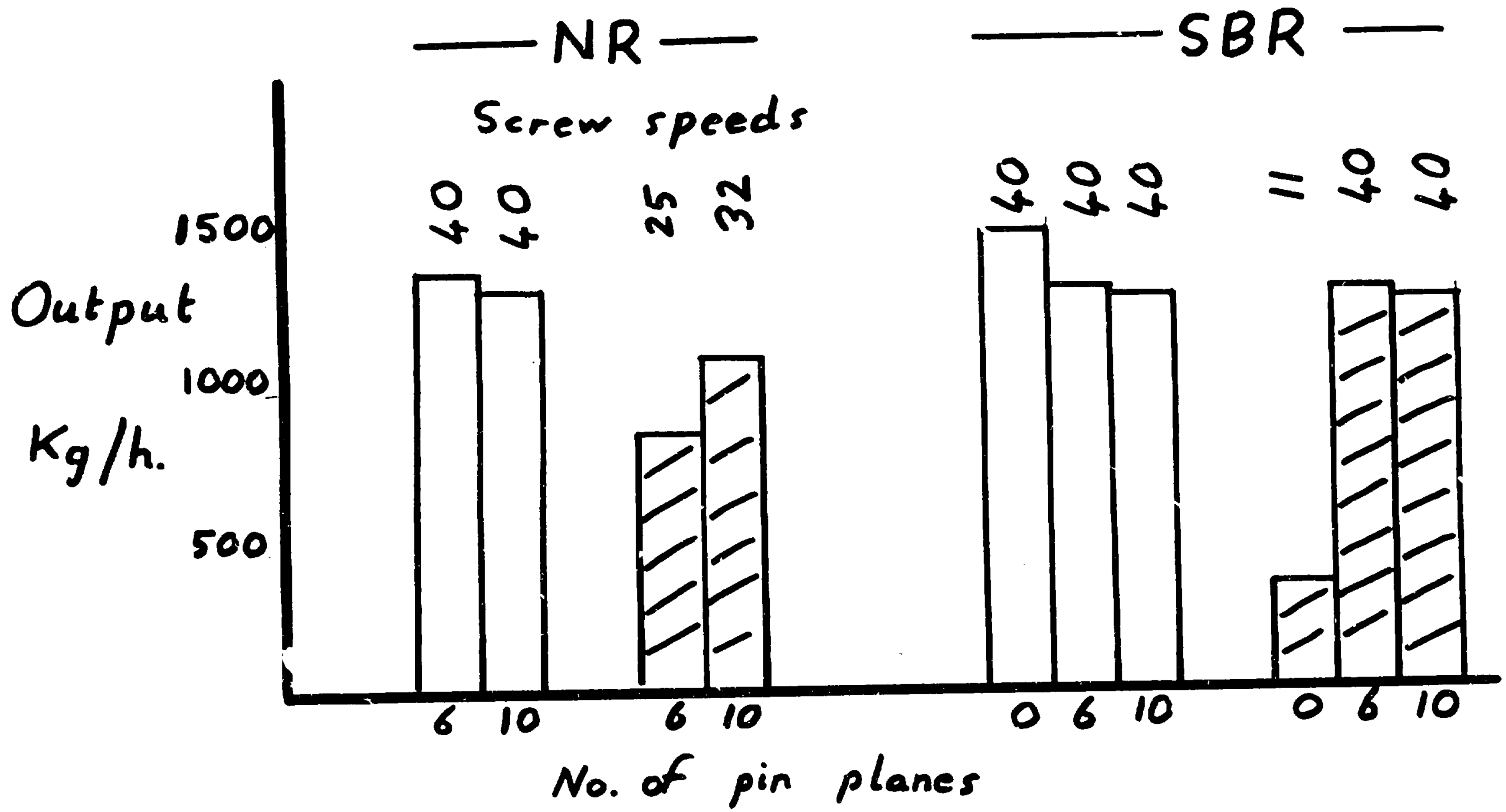


FIG 5. Extrusion of NR and SBR black mixes in Troester pin extruder at maximum screw speed □ and at quality limit ▨

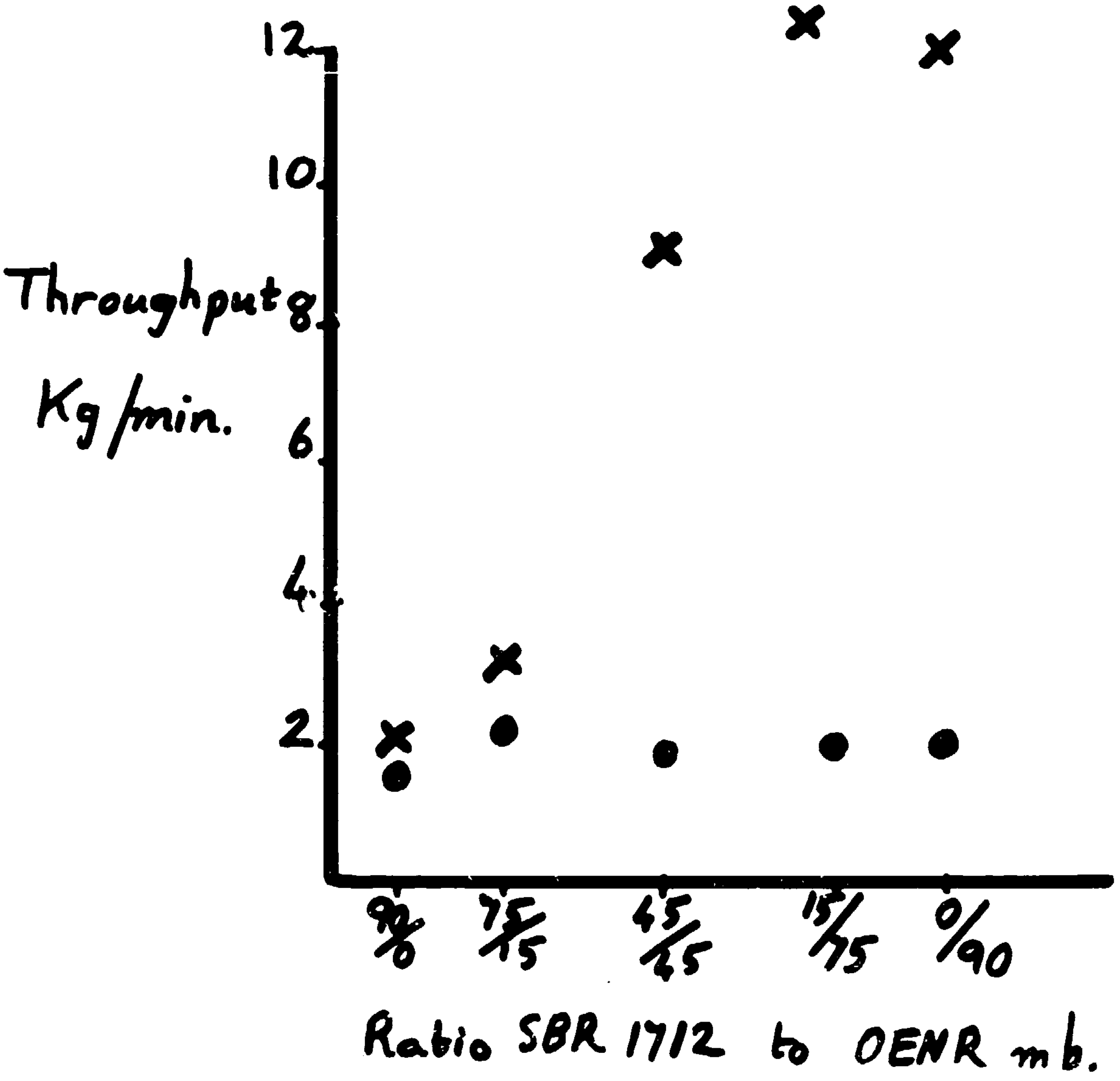


FIG 6. Effect of replacing SBR 1712 by OENR masterbatch on extrusion rate in Barwell ram extruder (x) and in 2½" cold feed screw extruder (●)

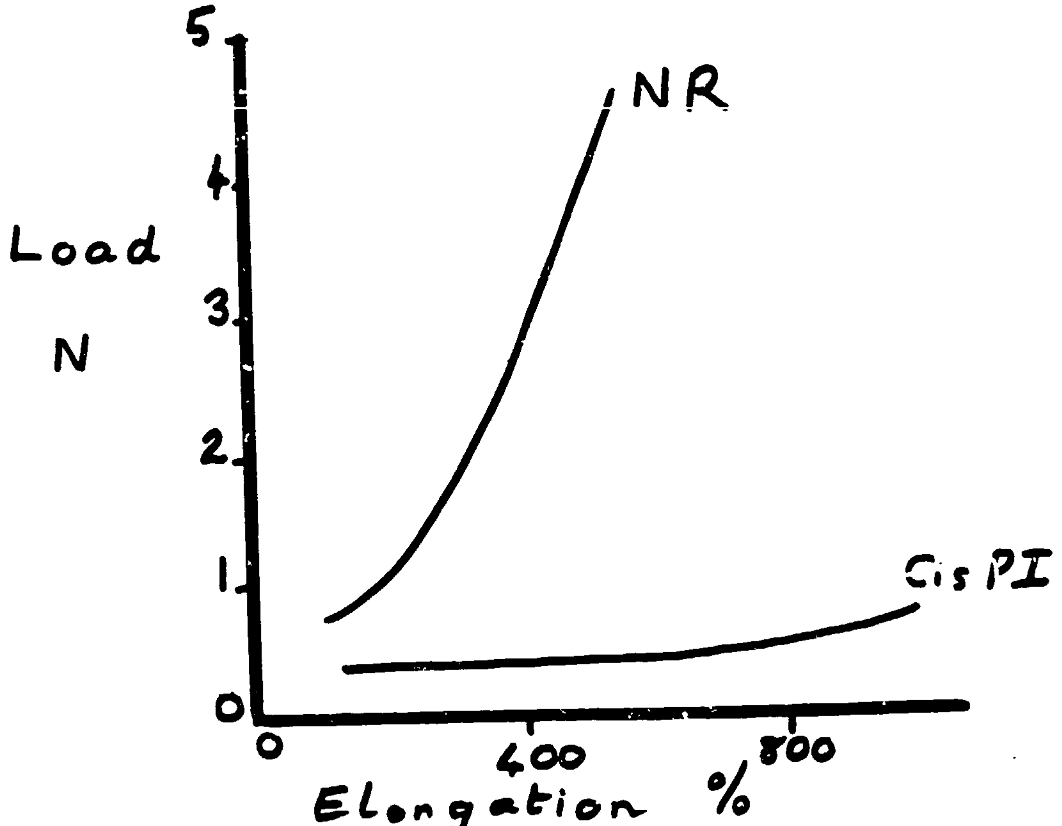


FIG 7A. Green strength of NR and high Cis PI tread compounds

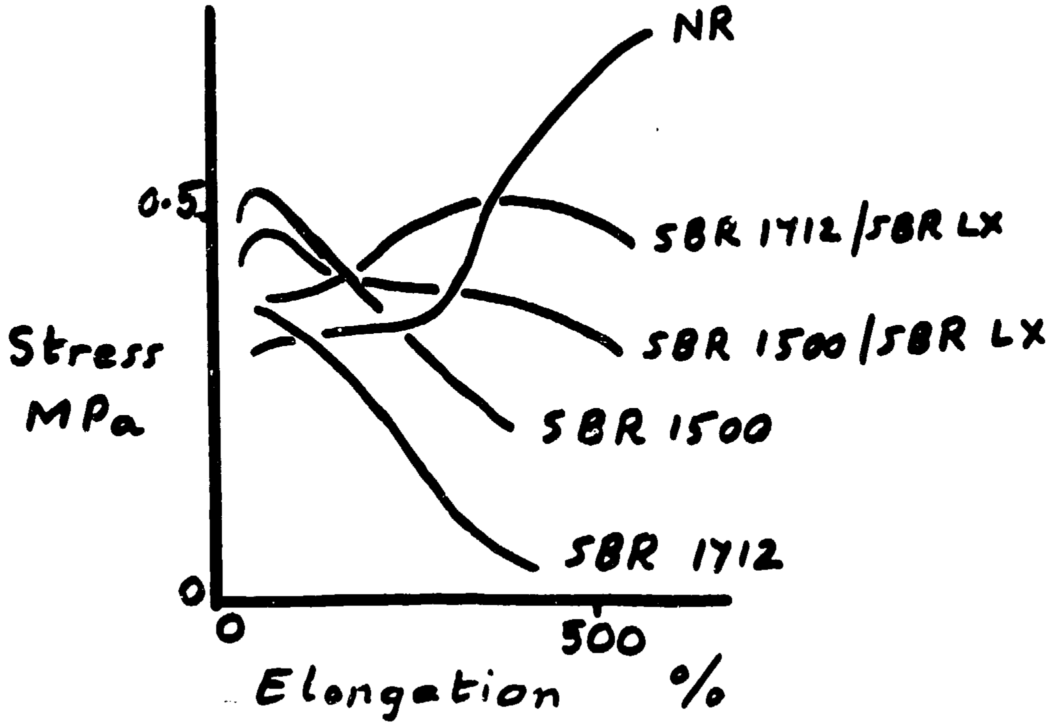


FIG 7B. Effect of SBR LX on green strength

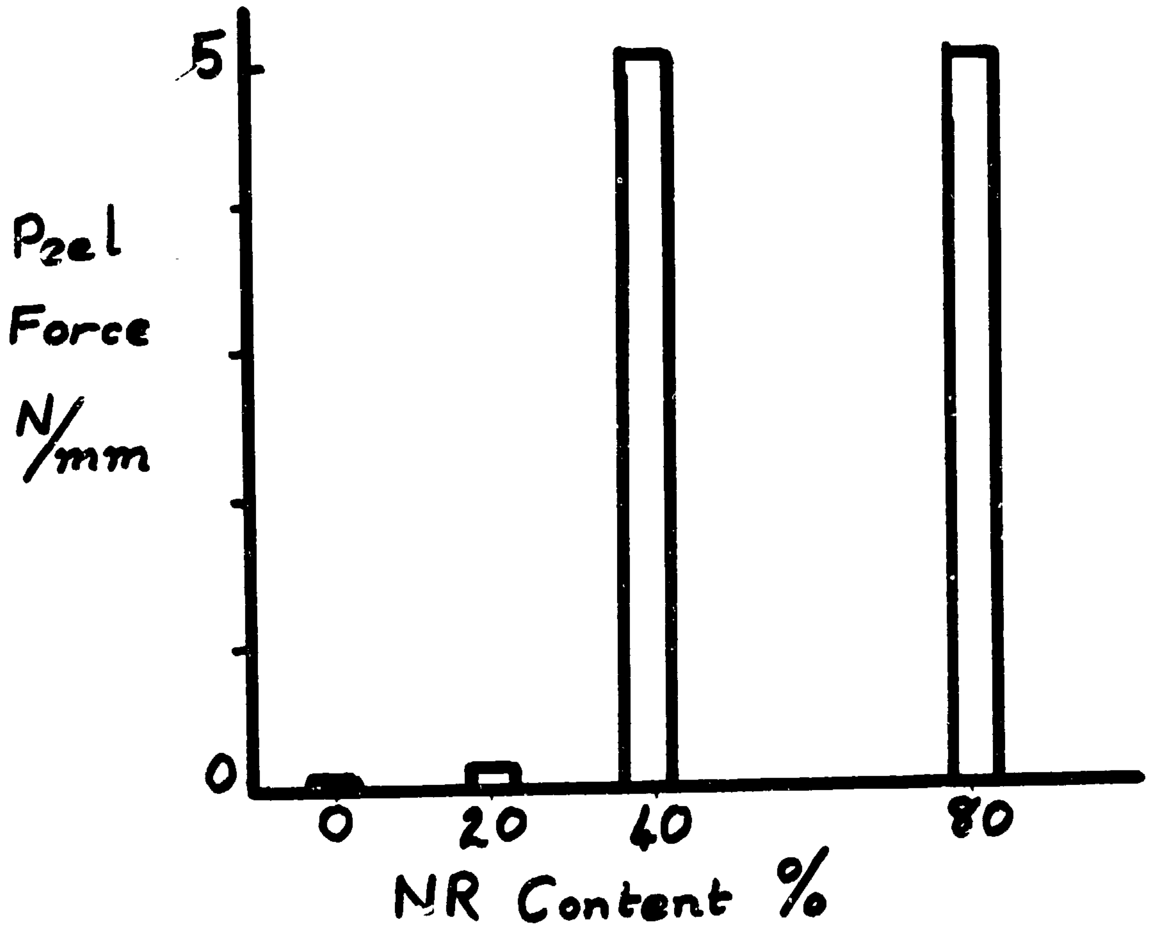


FIG-8. Adhesion of an uncured retread compound to a vulcanized substrate as function of NR content of uncured mix.

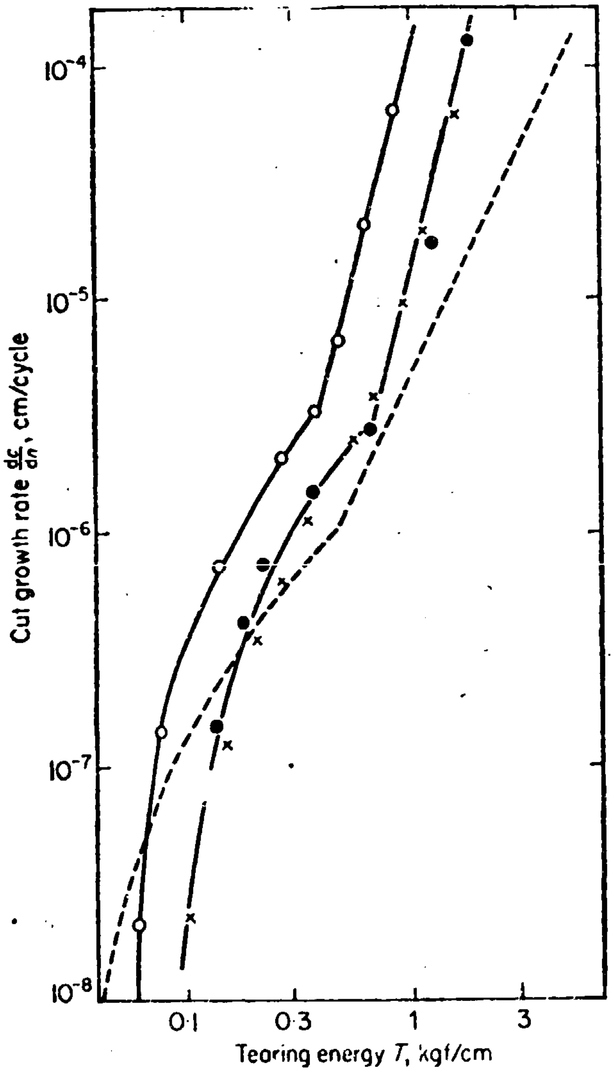
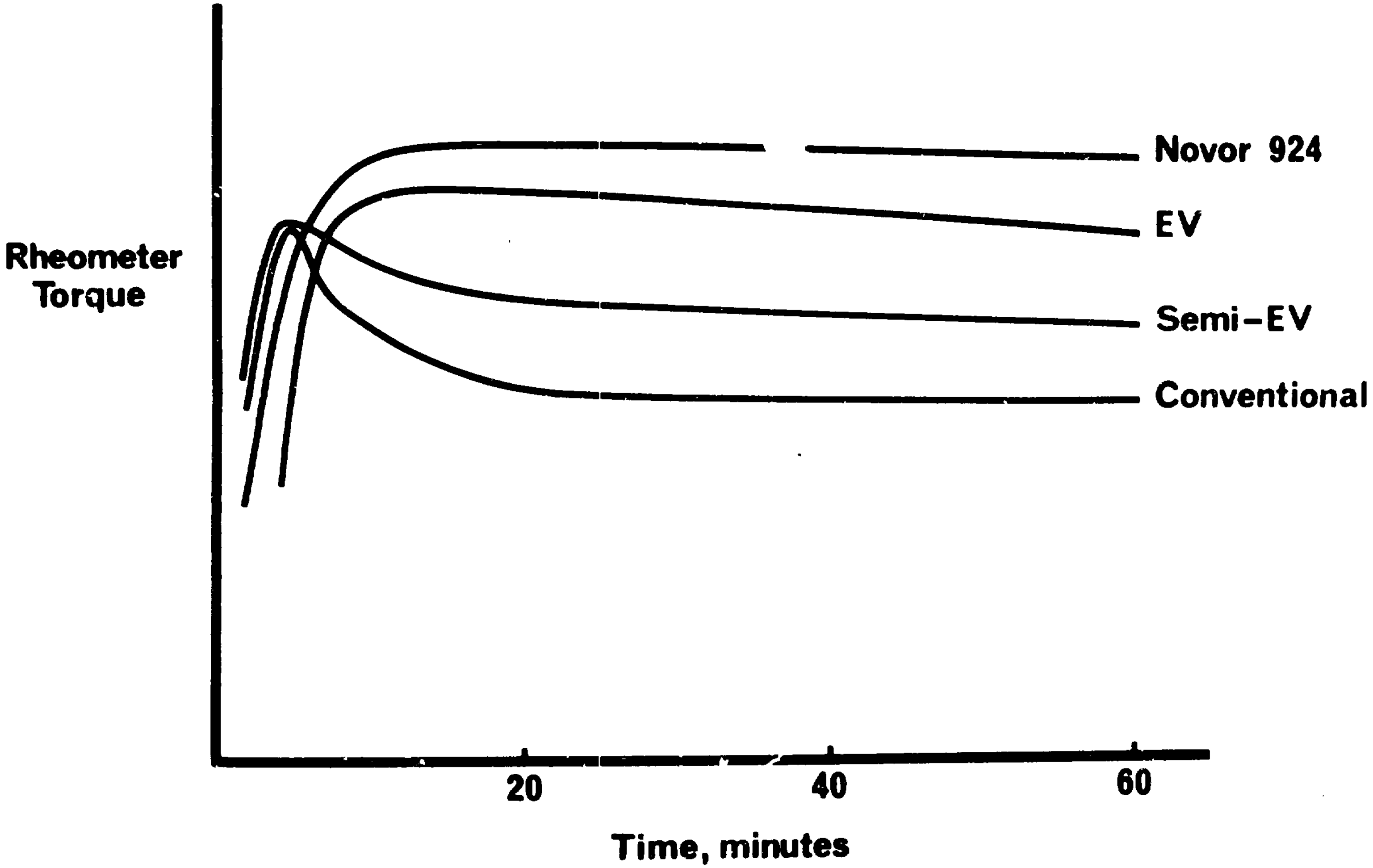


Fig. 9 Cut growth characteristics for natural rubber gum vulcanizate (broken line) and for SBR vulcanizates: gum vulcanizate (O), with 50 pphr MT black (X) and with 50 pphr HAF black (●)

FIG10 REVERSION RESISTANCE AT 180°C



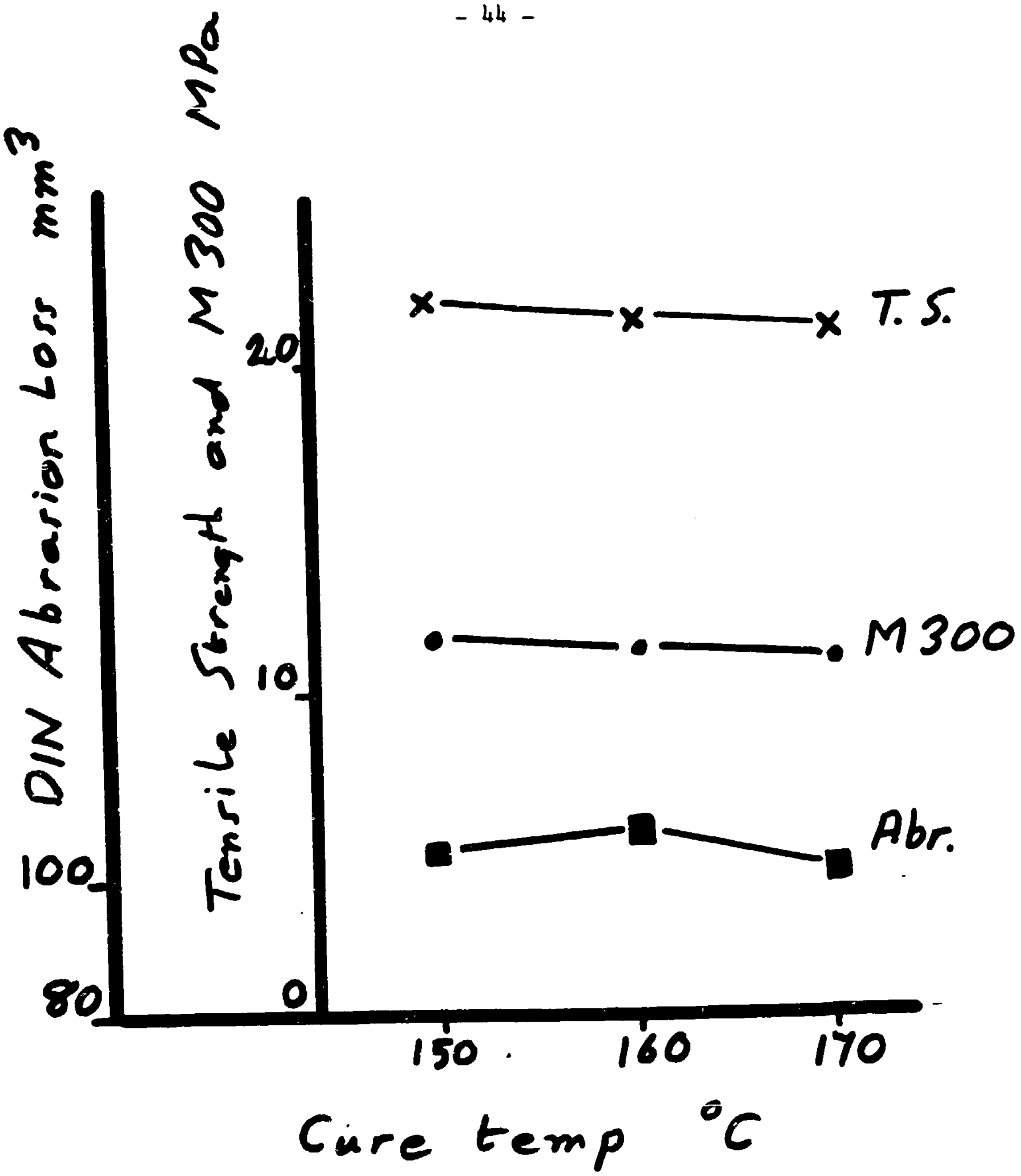


FIG 11 Retention of curing efficiency at increasing cure temperature of an Si69/MBS/S mix.

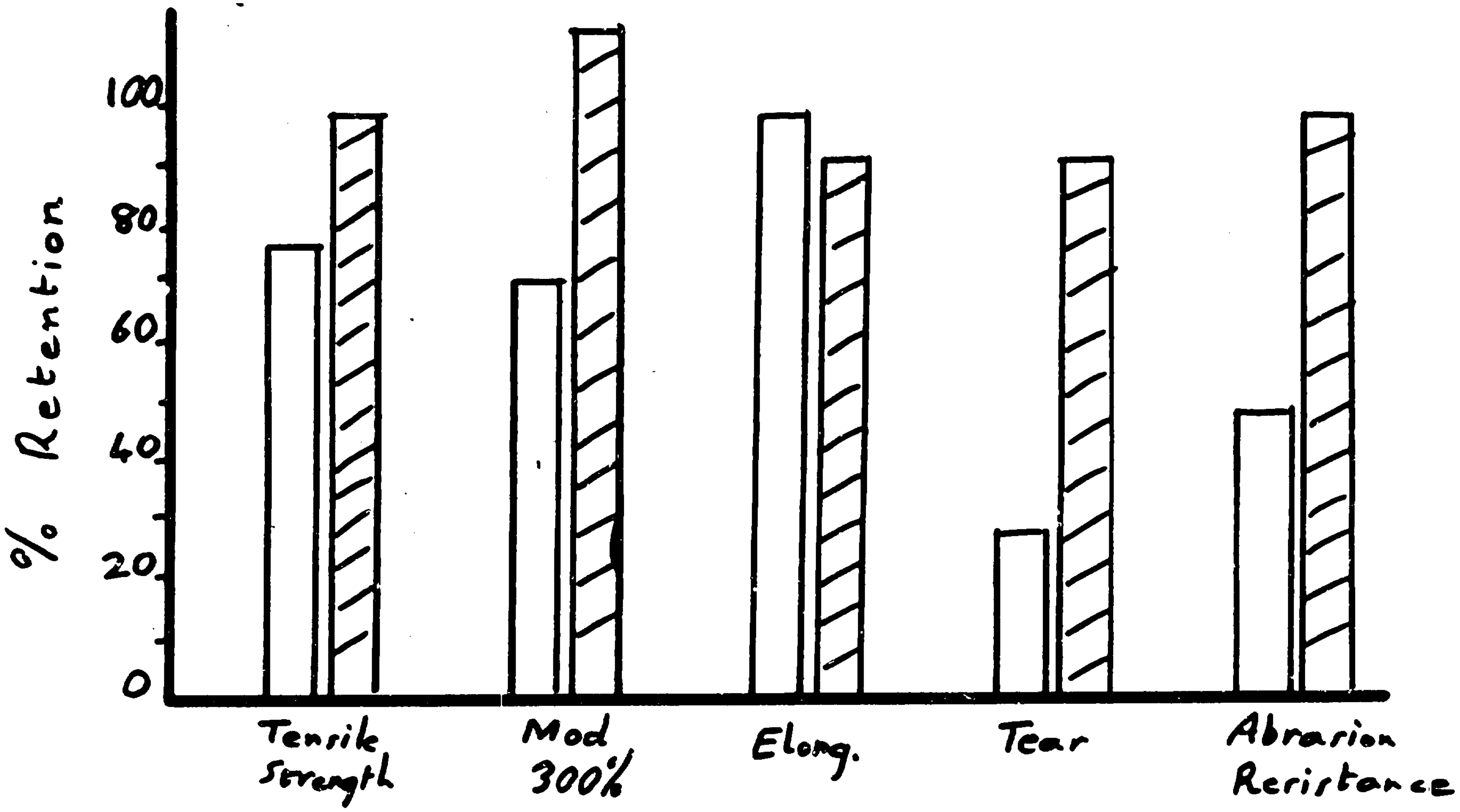
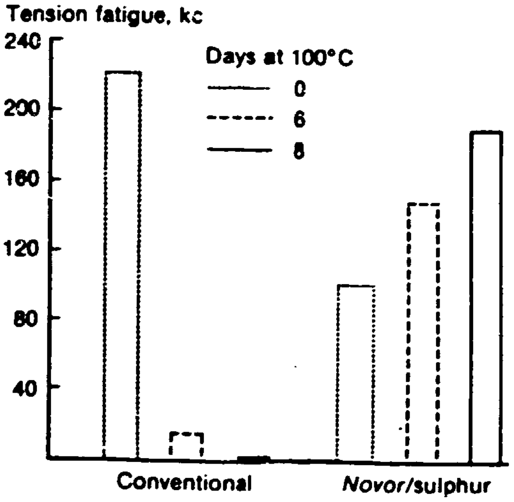


FIG 12. Effect of Si 69 on reversion resistance after 100 min. overcure at 170°C of MBS/S comp □ and Si 69/MBS/S comp. ▨

Figure 13

Comparison of initial and aged tension fatigue performance given by a conventional sulphur and an 80/20 Novor/sulphur system. (The masterbatch used for both systems contained SRF black, 5; process oil, 12.5 parts phr)



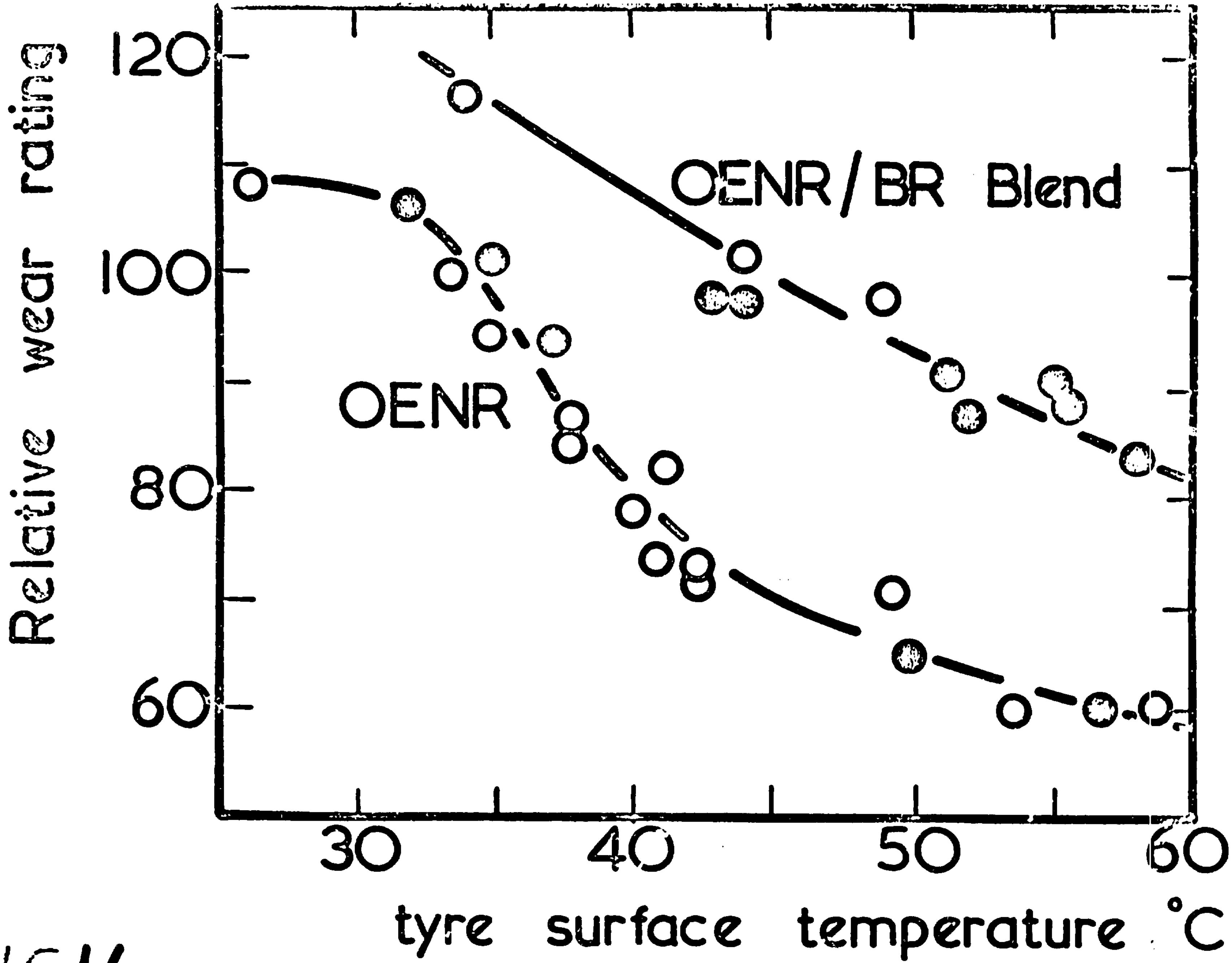

Relative wear rating
120
100
80
60
OENR/BR Blend
OENR
30
40
50
60
tyre surface temperature °C

FIG 14

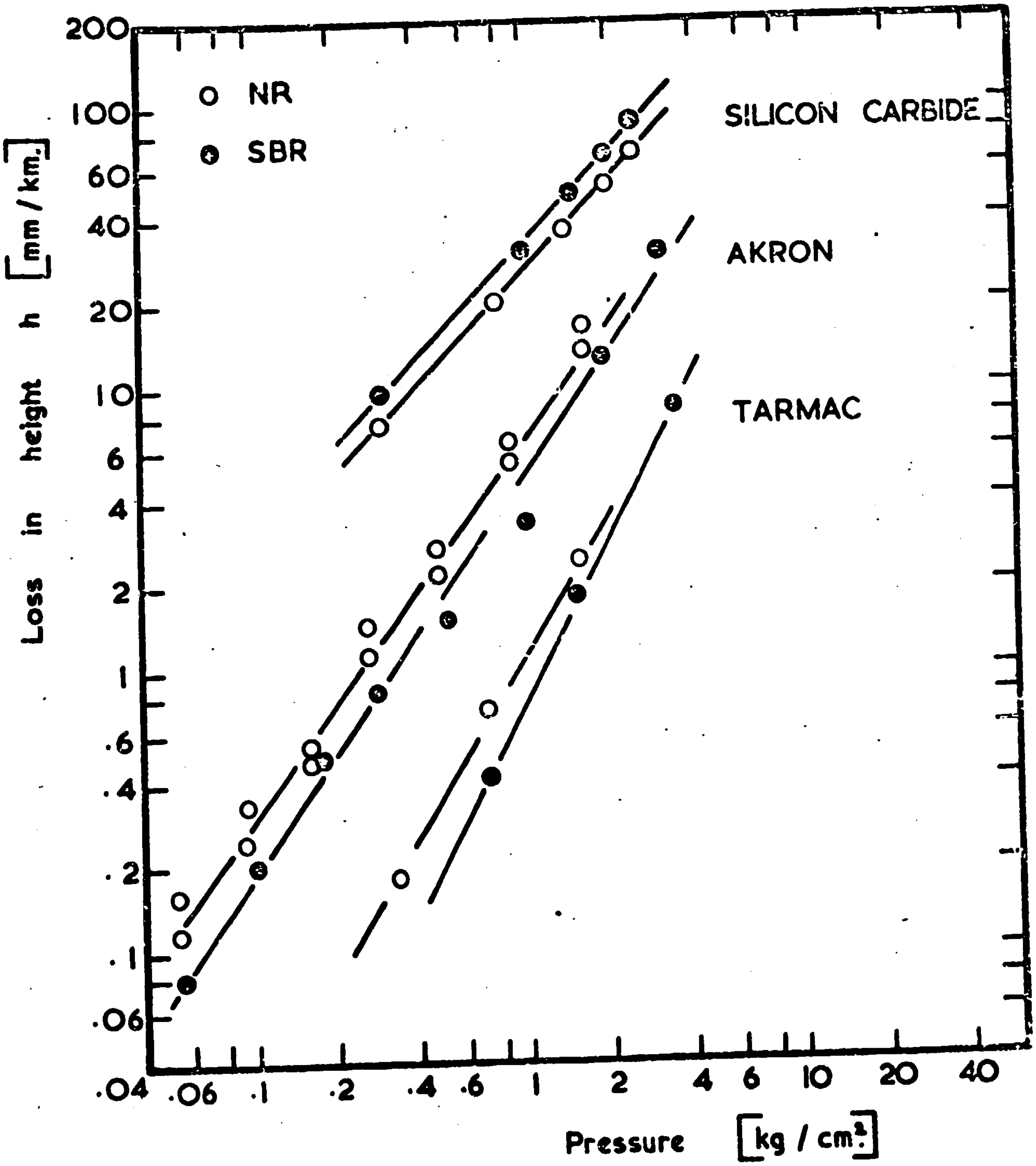


FIG 15 Abrasion of NR and SBR as a function of pressure on tracks of different sharpness

Figure 16 a
Volume swelling in ASTM No 1 at room temperature for NR/CR blends

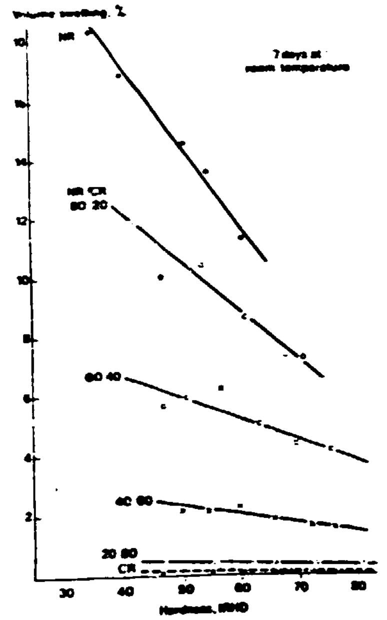


Figure 16 b
Volume swelling in ASTM No 1 oil at 70° for NR/CR blends

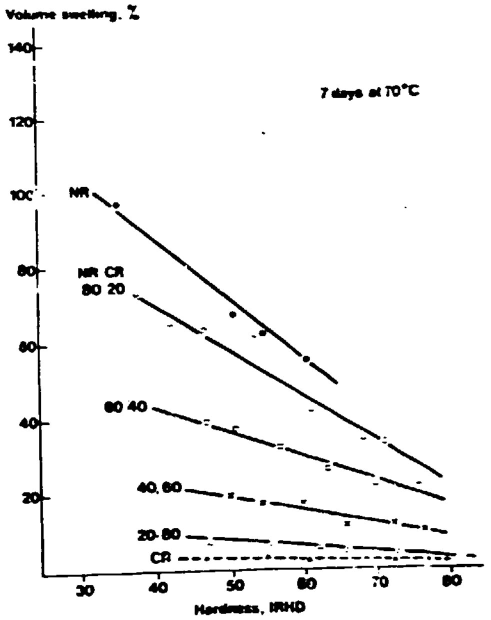


Figure 17 a
Volume swelling in water at room temperature for NR/CR blends

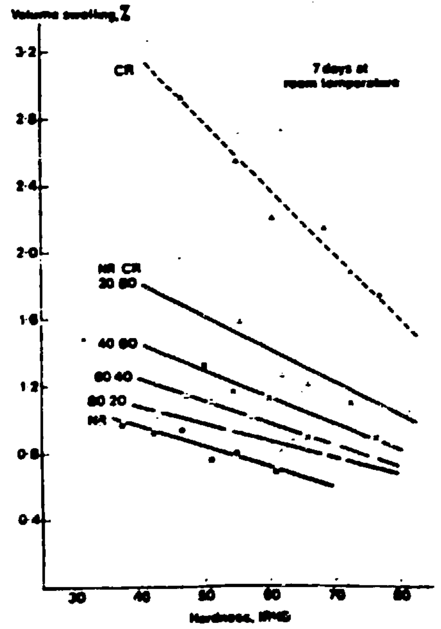


Figure 17 b
Volume swelling in water at 70°C for NR/CR blends

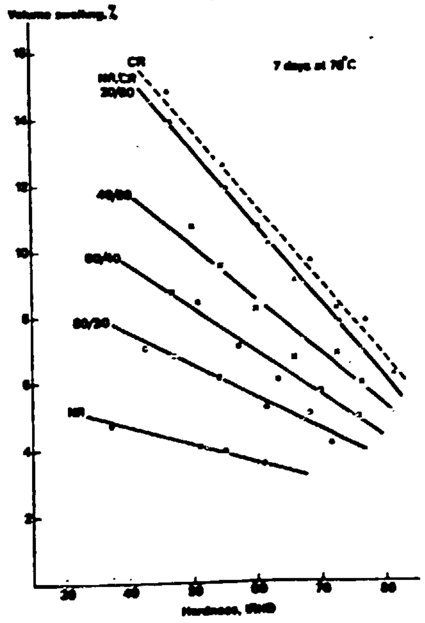
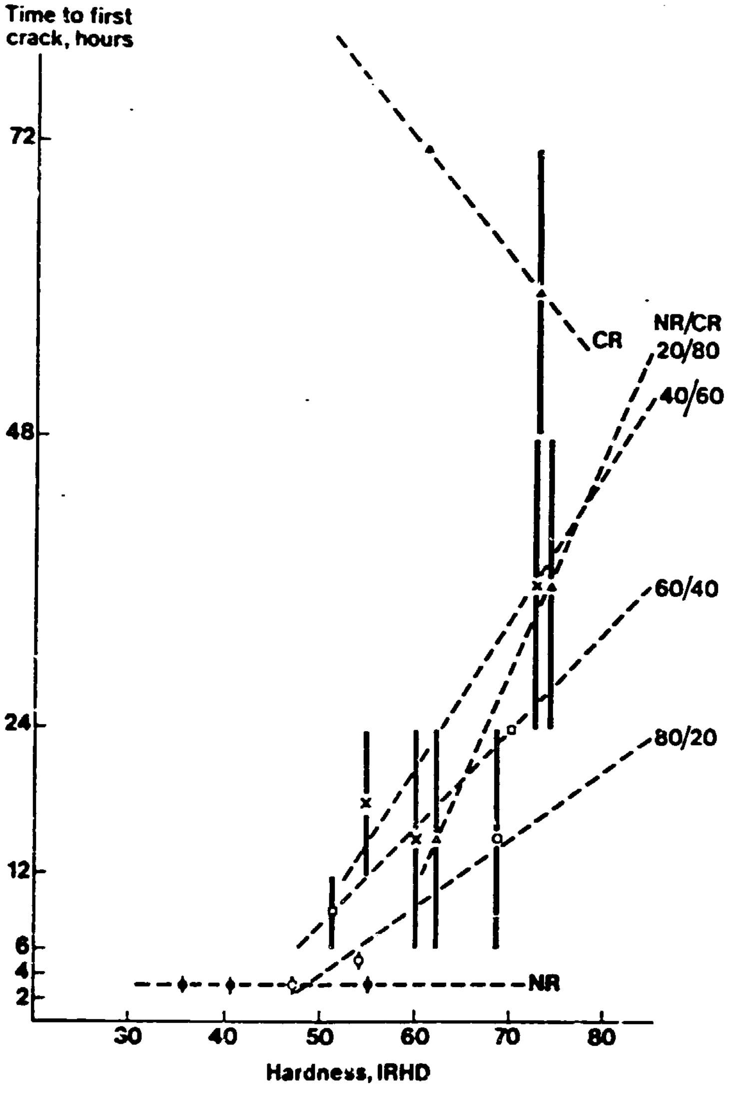


Figure 18
Ozone resistance of NR/CR blends: Variation of time to first crack with hardness (50pphm ozone, 40°C, 20% strain)



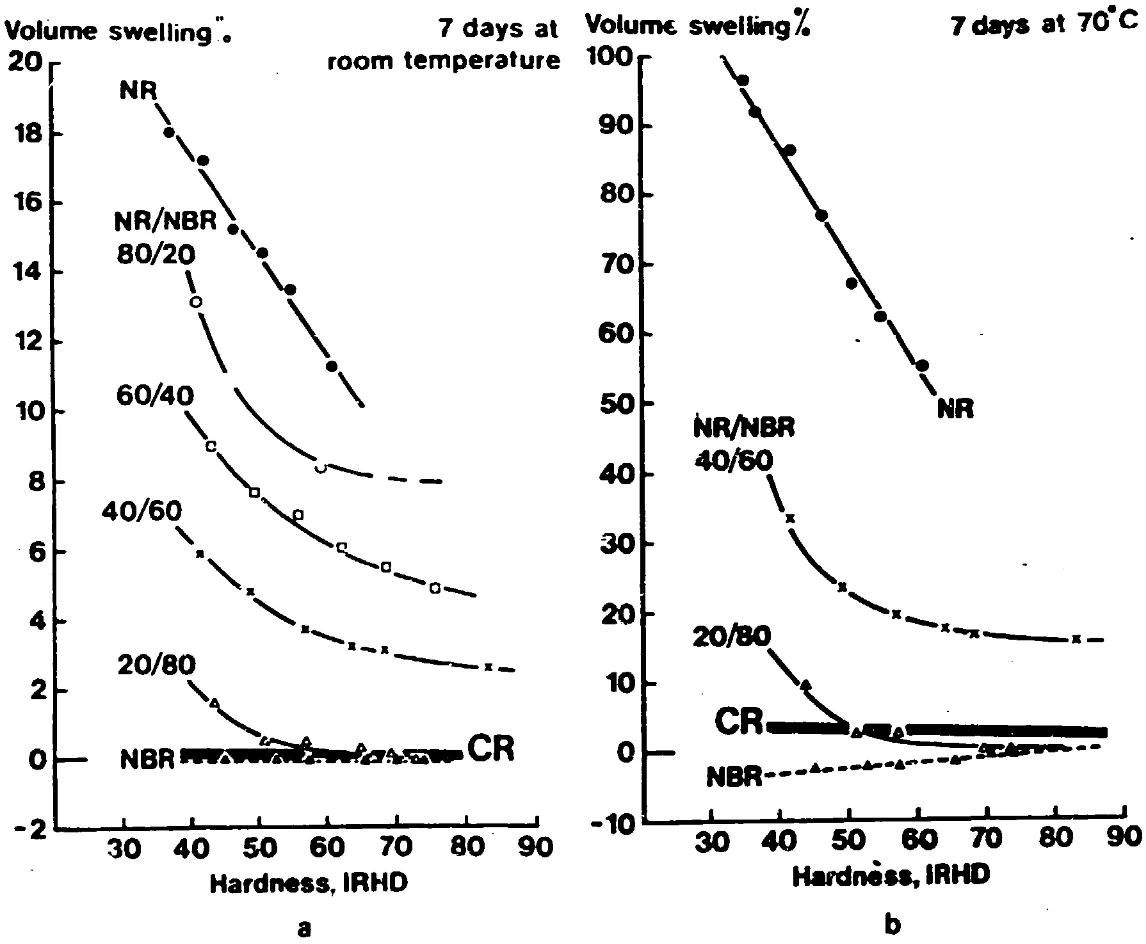


Fig. 19 Volume swelling in ASTM No.1 oil for NR/ABR blends

a. at room temperature

b. at 70°C

Figure 20
Ageing of NR/NBR blends: retention of tensile strength after 4 days ageing in air at 100°C

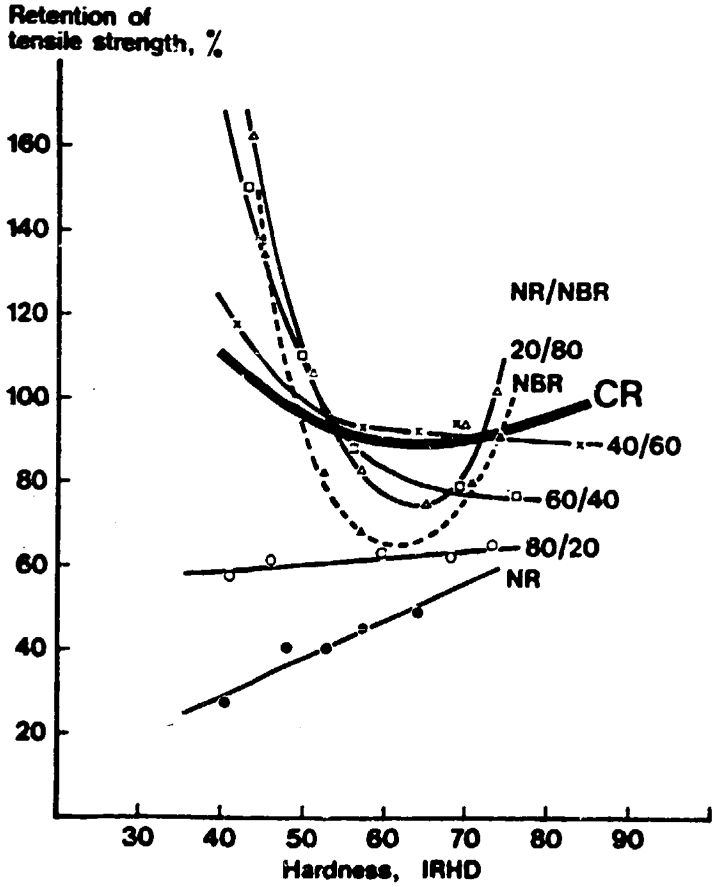
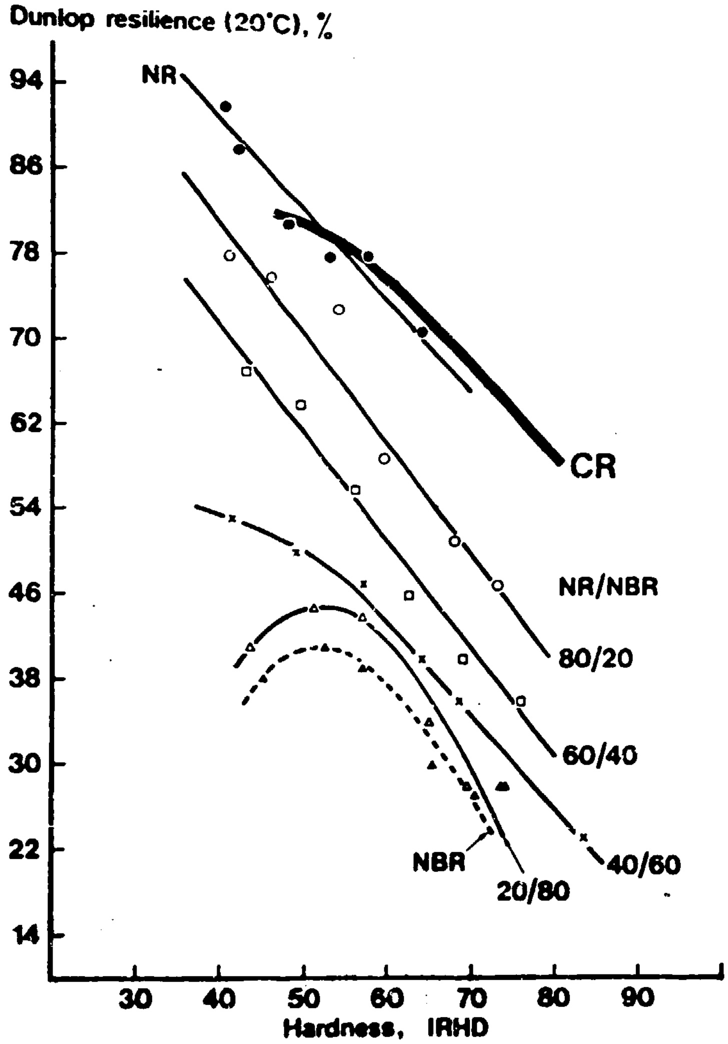


Figure 21
Variation of resilience with hardness for NR/NBR blends



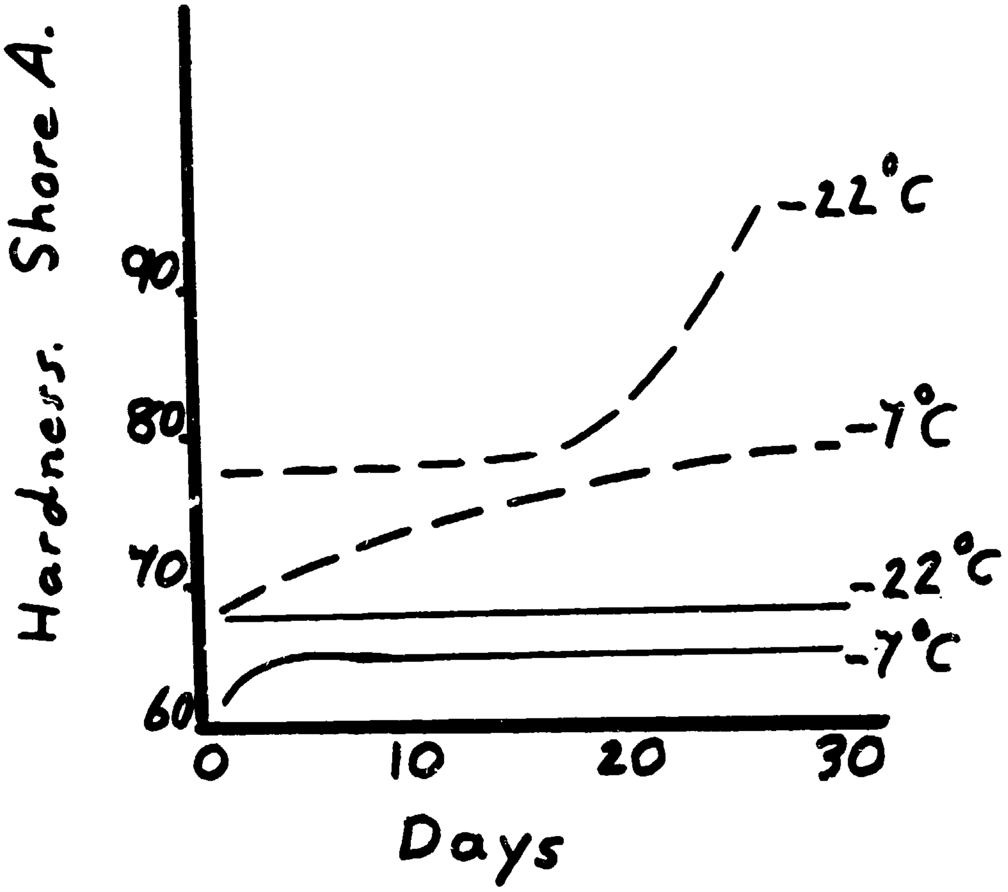


Fig. 22 Variation of herdness with time of NR and CR at -7°C and -22°C. NR ——————, CR ------------.

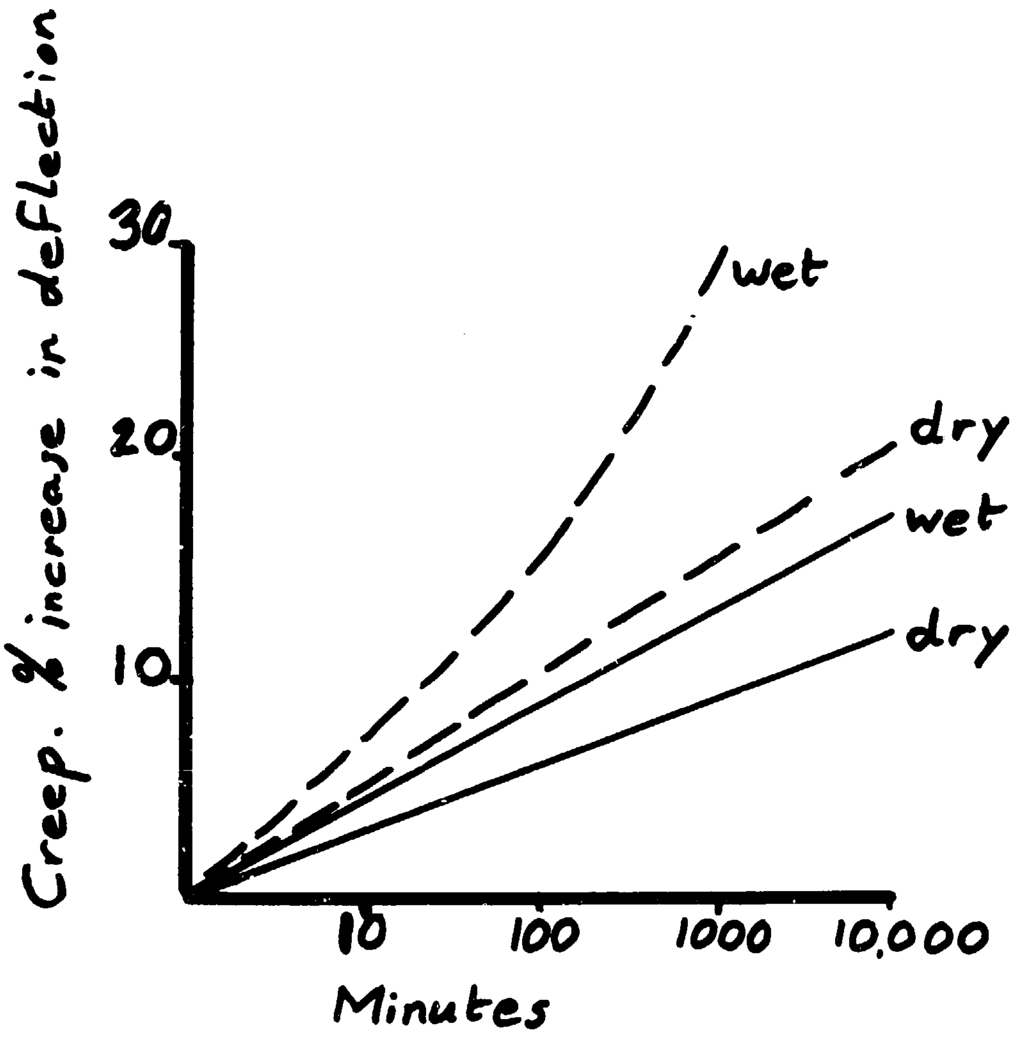


FIG 23. Creep of thin strips of rubber in tension. NR——. CR----
Dry is 36% relative humidity.
Wet is 88% " "

Figure 24
Dependence of injection chamber mix temperature (○) and injection temperature using a 3·5mm nozzle (□) on extruder and injection chamber jacket temperature

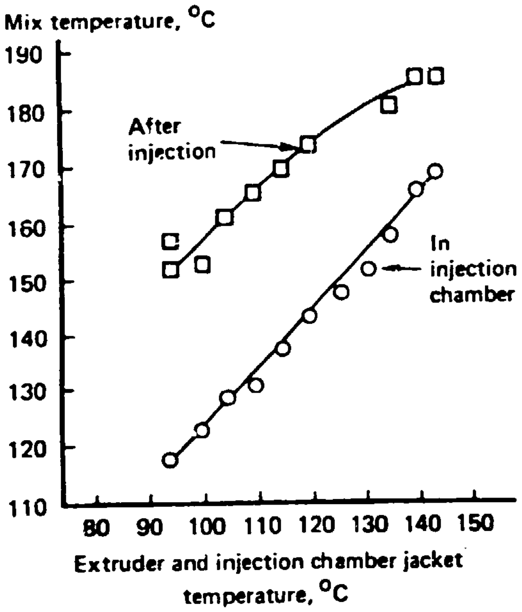


Figure 25
Effect of injection pressure on injection temperature (a) and injection time (b). The work done in forcing the mix through the nozzle is clearly shown by the difference in temperature with (□) and without (○) a 3·5mm nozzle

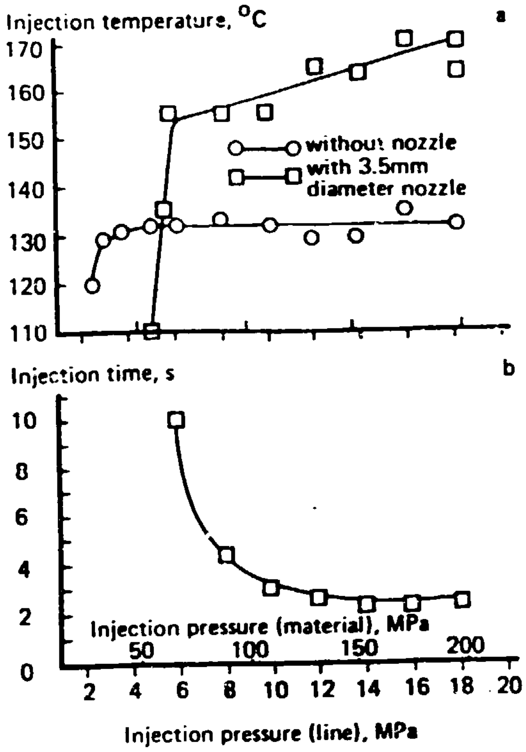


Figure 26
Dependence of mould filling time and scorch on injection pressure for a sheet mould, 260 × 260 × 4mm (shot volume 270cm³). Mix as Table 1 plus: stearic acid, 2; CBS, 0·5; sulphur, 2·5; Santogard PVI *(Monsanto), 0·4 parts phr. Mooney viscosity, ML1 + 3, 120°C, 36; 135°C, 33. Mooney scorch t₅, 120°C, 75 min; 135°C, 21 min*

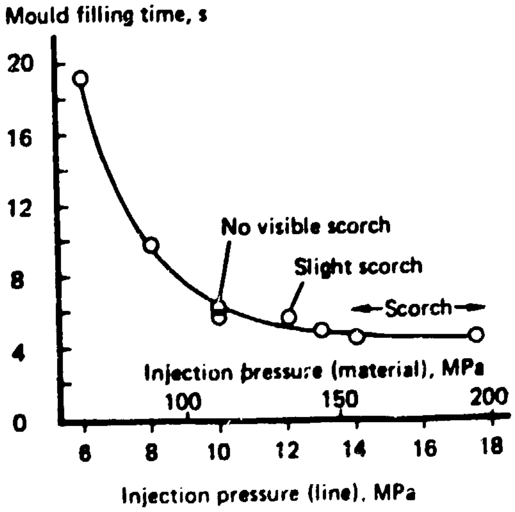


Figure 27

Dependence of optimum properties of a conventional high sulphur vulcanizate on cure temperature. The results are rated against those of mixes vulcanized at 160°C. Vulcanizate properties measured on test samples prepared in a REP B43K *injection moulding machine (REP K International Rue du Dauphiné, 69800 Lyon Corbas, France). Curemeter torque increase,* $M_{HR}-M_L$ *measured on a Monsanto* Rheometer

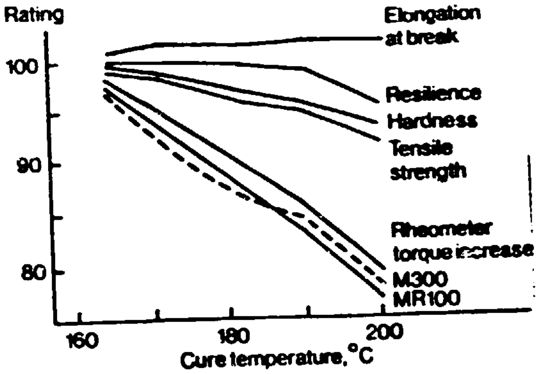


Figure 28

Increasing accelerator and sulphur levels to compensate for the effect of increasing cure temperature. The results for an injection moulded conventional high sulphur vulcanizate show that the crosslink density obtained at 160°C can be matched at 180°C by adding approx 20% more accelerator and sulphur. MR100 *is used as a sensitive measure of cure rate, but similar results are obtained with hardness and modulus at 300% elongation*

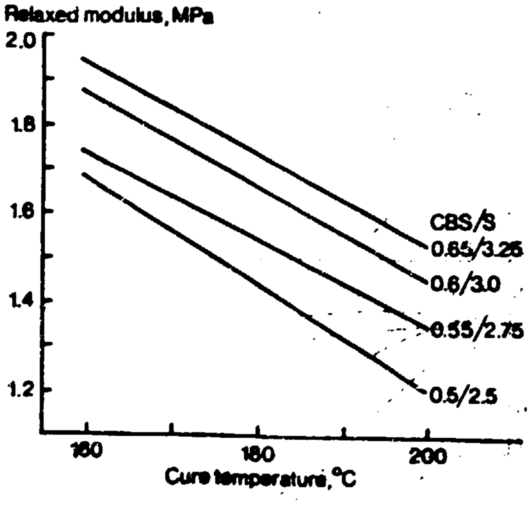


Figure 29

Increasing the level of accelerator to compensate for the effect of increasing cure temperature. The results show how a simple increase in accelerator level (from 0·5 to 1 part phr) can be more beneficial than a 20% overall increase in accelerator and sulphur. A similar improvement is shown with vulcanizate properties

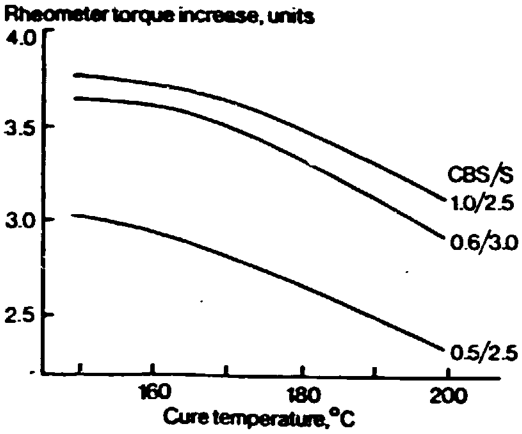


Figure 30

Dependence of tensile strength and modulus at 300 per cent elongation on cure time and cure temperature. The dotted lines are for a mix containing a conventional sulphenamide-accelerated system moulded at 190°C

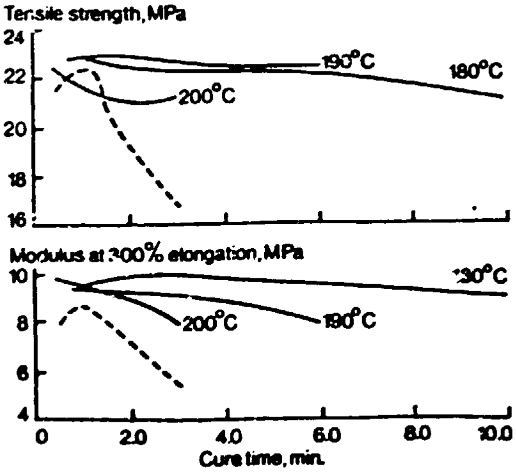


Table 1 Correspondence between rubber consumption and vehicles in use

		1965	1970	1977
World	Rubber consumption in '000t	4825	6850	9125
	No. of vehicles in millions	178	233	362
Brazil	Rubber consumption in '000t	64	122	276
	No. of vehicles in millions	1.8	3.0	7.5
Canada	Rubber consumption in '000t	141	186	278
	No. of vehicles in millions	6.6	8.1	12
Japan	Rubber consumption in '000t	378	779	1010
	No. of vehicles in millions	6.5	17.6	31.4

Table 2 Evolution of planting materials through breeding

Period	Type of planting material	Yield (kg/ha/yr)
1920s	Unselected	560
1930s	Pil B 84	975
1940s - 50s	PB 86	1120
1950s	RRIM 501	1460
1960s	RRIM 600	2350
1970s	RRIM 703	3360

Table 3 Proportion of high yielding material and yield per hectare per year on Peninsular Malaysian estates and smallholdings, 1950-77

Year	Proportion of high yielding material replanted and new-planted (%)		Yield (Kg/ha/yr)	
	Estates	Small-holdings	Estates	Small-holdings
1950	10	1	570	465
1955	24	8	550	432
1960	48	29	758	437
1965	68	54	953	590
1970	89	63	1189	752
1974	95	80	1389	929
1975	95	82	1300*	940
1976	95	85	1500	1040
1977	95	87	1550	1100

*This figure is affected by the National Crash Programme introduced in late 1974 through 1975 to raise the price of rubber. (For details, see Lim S C, 'Towards an equitable international trade in natural rubber', Malaysian Rubber Review. 1976, 1, (1), 13.

Table 4 Proportion of smallholder in rubber growing countries

Country	Total area under rubber ('000 ha)	Smallholding % of total area
Malaysia	2002	67
Indonesia	2369	78
Thailand	1374	95

Table 5 Polymer Ratios in Parts of Radial Tyres

Tyre	Polymer Range	Tread	Sidewall	Belt
Car, general purpose	NR	0	45 - 80	100*
	SBR	55 - 100	0	0
	BR	0 - 45	20 - 55	0
Car, all weather	NR	0 - 30	45 - 80	100*
	SBR	45 - 80	0	0
	BR	20 - 25	20 - 55	0
Car, winter (USA)	NR	0 - 40	40 - 70	100
	SBR	30 - 70	0 - 30	0
	BR	30 - 40	30 - 50	0
Car, winter (Europe)	NR	0 - 75	40 - 70	100
	SBR	0 - 100	0 - 30	0
	BR	0 - 25	30 - 50	0
Truck 24.5"	NR	55 - 100	45 - 60	100
	SBR	0 - 30	0 - 35	0
	BR	0 - 20	20 - 45	0

Table 6 Effect of ratio of NR to SBR on rate of extrusion of oven heated and freshly milled tread mix (V_c 60) in a Barwell extruder

NR/SBR Ratio	Throughput kg/min	
	Oven heated	Freshly milled
100/ 0	11	25
80/20	16	22
60/40	16	17
40/60	13	13
20/80	10	12

Table 7 Effect of stearic acid in maintaining abrasion resistance of an overcured semi-EV NR tread mix

Stearic acid	0	1	2	4	6
Optimum cure at 150°C	10	11	11	13	14
Laboratory abrasion rating. Control (= 100) is 60m cure with 2 parts stearic acid					
(a) optimum cure	106	106	120	128	125
(b) 60 min cure	72	86	100	115	120

Table 8

Wear rating of NR vs SBR in winter tyre treads

NR compound is an oil-extended blend of NR and polybutadiene. SBR compound is a commercial product.

Location	Rating $\frac{\text{Wear loss of SBR}}{\text{Wear loss of NR}} \times 100$
Montreal in winter	127
Europe in winter	100
Texas in summer	60-70

Table 9 Service performance of cover compounds on sectional conveyor belt carrying iron ore

Rubber	Wear rating of cover	Ranking of resistance to cover/carcass separation
NR	100	1
OENR (20% oil)	90	2
OENR (33% oil)	67	2
SBR 1500	82	1
OESBR 1712	62	3

Table 10

Tyre	5.60 x 13 Bias	165SR13 Radial
Tyre inflation pressure KPa	160	240
Tyre life assured	45000	80000
Petroleum equivalent of rubber worn away kg	7.6	4.9
Gasoline to move car kg	1071	750
Gasoline to roll wheels kg	214	97

Table 11 Energy Content of Some Synthetic Rubbers

	GJ/tonne
Polychloroprene	144
SBR	156
EPDM	170
Polyurethane	209
Butyl rubber	209

Table 12 Natural Rubber: Components of Energy Content

	GJ/tonne
Fertilizers and other chemicals	5
Processing	3
Transport	5- 8
	13-16

Table 13 Vulcanizate properties of 100% CR and a 20 NR/80 CR blend

	100 CR	20 NR/80 CR
Black loading phr	40	40
Tensile strength MN/m^2	22.0	19.5
Tear strength N/mm	37	22
Resilience, Dunlop, %	71	71
Volume swelling, 7d/20°C, %		
in water	2.2	1.3
in ASTM No 1 oil	0.1	0.4
in ASTM No 3 oil	1.6	1.9
Ozone resistance, 50 pphm at 40°C. Time to first crack, hours	48-72	24-48

Table 14 Hardness, tear and fatigue of some NR compounds

Hardness IRHD	39	50	60	78
Tear resistance, KN/m (split strip test)	11	11	13	7
Fatigue resistance Kc to failure. Ring fatigue test	271	146	98	105

Table 15 Glass transition temperatures of NR and synthetic rubbers

Rubber	T_g °C
cis BR	-105
NR	- 70
Butyl	- 65
EPDM	- 58
SBR	- 60
CR	- 49

Table 16

Injection moulding conditions with the REP B43K machine

Extruder barrel temp °C	200-220 (390-430°F)
Injection chamber temp °C	200-220
Nozzle block temp °C	190-210 (375-410°F)
Back pressure	none
Screw speed, rev/min	80-180
Screw drive pressure,	
line, MPa (psi)	4-6 (580-870)
Injection pressure 1,	
line, MPa (psi)	10-14 (1450-2030)
material, MPa (psi)	111-156 (16 100-22 600)
Injection pressure 2,	
line, MPa (psi)	9-10 (1300-1450)
material, MPa (psi)	89-100 (12 900-14 500)
Injection speed	max
Nozzle diameter, mm	3.5
Mould dimensions, mm	260 x 260 x 2
Mould volume, cm^3	135
Mould temperature, °C	90-110 (195-230°F)
Cycle times:	Seconds
injection	3
injection pressure 1, hold time	2
injection pressure 2, hold time	5
cooling	3-30
total*	13-40

*Excluding mould opening and closing and stripping times

Table 17

Properties of NR/PP and NR/PP/PE blends

	Parts by weight							
Natural rubber, SMR 5L	65	60	50	40	65	60	50	40
Polypropylene, MFI 20	35	40	50	60	17.5	20	25	30
High density polyethylene, MFI 5.5	-	-	-	-	17.5	20	25	30
Dicumyl peroxide[1]	0.39	0.36	0.30	0.24	0.39	0.36	0.30	0.24
Antioxidant[2]	1	1	1	1	1	1	1	1
Moulding properties								
Tensile strength, MPa a[3]	11.0	13.6	16.1	19.8	14.4	15.0	20.0	21.0
b[4]	10.0	10.6	14.8	16.7	12.6	13.0	14.8	14.0
Elongation at break, % a	65	55	60	115	55	55	40	60
b	405	330	475	540	400	465	495	495
Flexural modulus, MPa, at								
23°C a	400	500	620	880	300	440	620	840
b	200	240	370	580	140	220	380	580
70°C a	150	160	240	300	90	150	200	290
b	70	80	130	210	40	60	120	190
-30°C a	1200	1400	1900	2100	750	1100	1550	2100
b	600	750	1100	1650	400	600	850	1550
$\frac{\text{Flex mod at -30°C}}{\text{Flex mod at +70°C}}$	8.2	8.9	8.1	7.4	8.8	8.1	7.5	7.5

[1] eg Di-Cup R (Hercules)

[2] 2,2-Methylene-bis-(4-methyl-6-t-butylphenol) eg Antioxidant 2246 (Anchor)

[3] Radial direction

[4] Tangential direction

Table 18 Mixing costs with powdered NR

Machine	Output	Production Cost £/t
F80 Banbury 3 min mix cycle with second stage. Using bale rubber	1000 litre/h	750
3 MVX 134/120 machines	Each 300 litre/h	807
2 MVX 240/150 machines	Each 500 litre/h	788

Table 19 Processing cost (excluding cost of granulation) for product of unvulcanized strip by conventional Banbury/mill using bale rubber and by MVX continuous mixer/extruder using crumb NR

Process	Cost £/t
Banbury F80 using 3 min cycle with second stage	158
3 MVX 134/120 300 L/h One man each	173
2 MVX 240/150 500 L/h One man each	154