

Abrasion, Fatigue, and Smearing of Rubber

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Synopsis

The abrasion of rubber on tracks without sharp-pointed asperities is dominated by fatigue failure. This is demonstrated by the effect of certain antioxidants on the abrasion of natural rubber compounds in air, which largely disappears in nitrogen. The experimental results obtained in air can be obscured by smearing which seems to originate from the abrasion process itself. In addition, the relative humidity of the surrounding atmosphere has a great, and little-understood influence on the rate of abrasion. The bearing of these findings on the interpretation of road test results is discussed.

Introduction

The rate of abrasion of a typical tread compound on laboratory tracks with different surface characteristics can easily vary by a factor of 50 for the same frictional energy dissipation; even on different stretches of common road surfaces, fourfold differences in abrasion have been found without searching for extremes in roughness.¹ Effects of this magnitude suggest the operation of different abrasion mechanisms on various surfaces, and Russian workers have distinguished between purely mechanical abrasion on sharp tracks, and wear caused by fatigue failure on smoother tracks.²⁻⁸ Fatigue failure during abrasion was diagnosed from the similarity between the power laws connecting abrasion and normal load on the one hand, and fatigue life and maximum stress during cyclic deformation on the other hand.

Brodskii and his co-workers deduced a close connection between fatigue-abrasion and chemical degradation from the difference between the abrasions of various vulcanizates in air and in nitrogen.³ The abrasion of natural rubber (NR) and oil-extended styrene-butadiene (SBR) treads was less in nitrogen than in air, the reduction being considerably greater on a knurled metal track than on abrasive paper; butyl rubber was not affected by the atmosphere. The abrasion resistance of NR compounds depended on the antioxidant used, but the relative differences between the variously protected compounds was maintained in nitrogen.

Work carried out in this laboratory has confirmed the occurrence of two abrasion mechanisms. Abrasion on sharp tracks (silicon carbide paper) is closely related to the energy density at break of the rubber, and originates therefore from tensile failure.⁹ A preliminary account¹⁰ of work on the

effect of the track, surrounding atmosphere, and antioxidant formulation on the abrasion of various elastomers showed that the antioxidant has little influence on the abrasion of NR tread compounds on silicon carbide paper; this lack of response to the antioxidant was to be expected from a tensile failure mechanism. The tensile product—an approximate measure of the energy density at break—of the best protected compound was only about 5% higher than of an unprotected compound.

On a smoother track, where abrasion due to fatigue was anticipated, the presence and nature of an antioxidant had a pronounced effect on the rate of abrasion of NR, but the differences were much reduced in nitrogen, in contrast to the Russian results.³ In certain cases, abrasion was greater in nitrogen than in air. This was attributed to smearing during abrasion in air but not in nitrogen, and subsequent results given in this paper substantiate the earlier conclusions. It is further shown here that the relative atmospheric humidity has an unexpectedly large effect on the abrasion of both natural and synthetic rubbers.

Experimental

All experimental results have been obtained with an Akron abrader constructed to standard specifications (B.S. 903), operated at a slip angle of 15°. The machine was housed in the main chamber of a Perspex enclosure which communicated with a smaller compartment through a port closable from the main chamber. The small chamber, which had a second port in an outside wall, served as lock for conveying batches of samples from and into the main enclosure for weighing between runs without having to admit air into it. During measurements, the inner lock-door and the samples were handled with a long rubber glove whose rim was clamped air-tight around the circumference of a large opening in the front wall of the main chamber.

The motor was mounted outside, the drive-shaft entering the enclosure through a simple gland, and the samples rotated at 62.5 rpm (as against the usual 250 rpm) to avoid undue temperature rises in the box.

For measurements in nitrogen, the gas was passed from a cylinder through the enclosure at a given rate, and the remaining oxygen concentration calculated on the assumption that a volume of nitrogen entering the box expelled the same volume of the existing gas mixture through the outlet. If the rate of nitrogen flow R (in liters per minute) is maintained for t minutes, and the swept volume is V , the oxygen concentration is

$$C = C_0 \exp \{ -Rt/V \}$$

where C_0 is the initial concentration. Once the wanted reduction in oxygen concentration had been attained, the nitrogen flow was reduced to a rate sufficient to maintain a slight pressure during measurements. Data given in this paper for abrasions in nitrogen were obtained at an oxygen concentration of not more than 0.1%, as calculated from the equation.

TABLE I
Composition and Cure of Samples*

	Composition, parts			
	Natural rubber (NR)	Styrene-butadiene rubber (SBR)	Polybutadiene rubber (BR)	Ethylene-propylene rubber (EPR)
RSS1	100	—	—	—
Intol 1500	—	100	—	—
Cis-4	—	—	100	—
Enjay EPR 404	—	—	—	100
Sundex 1585	6	5	—	—
Dutrex R	—	—	5	—
Zinc oxide	5	3.5	3.5	—
Stearic acid	2	1.5	2.5	—
Philblack O	45	45	50	50
Santocure	0.4	1	0.6	—
DiCup R	—	—	—	4
DPG	—	0.4	—	—
Sulfur	2.5	1.8	2.0	0.4
Antioxidant	2	—	—	—
Nonox HFN	—	2	1	—
Cure time, min.	40	25	60	60
Cure temp., °C.	142	153	140	153

* The samples are identified in the text by polymer and antioxidant.

For experiments in dry air, laboratory air was sucked through the chamber after having first passed through two traps immersed in a solid CO₂-alcohol mixture. The humidity obtained by this procedure was less than 20%, as determined with a paper hygrometer which did not give lower readings. To achieve a high humidity, the air was passed over a moistened wick in a long glass tube.

Abrasion experiments were made with a Standard aluminum oxide wheel which, it should be noted, had been in service for many years, and with knurled aluminum and stainless steel wheels whose surfaces were similar to tracks used by Brodskii et al.³ The projections produced by knurling (with an engineering tool) were pyramids with a diamond-shaped base of about 0.5 mm.² area. New wheels of this type have a high initial abrasive power which, however, drops to a much lower stationary level when sharp points have been polished away during use. The height of the projections after running-in was about 0.4 mm.

Dust was fed into the nip between sample and wheel every 25 revolutions of the wheel to obviate or reduce smearing. The dust was either a mixture of 2 parts by volume of Carborundum 180 and 1 part of Fuller's Earth in doses of 16 mg., or 1 mg. of magnesium oxide (Magnesia Levis), the volume of dust being the same in both cases. The samples always looked mat and dry and did not feel tacky with either dust, but the results indicated

that magnesia was the more effective drying agent of the two. The difference between the results obtained with the dust mixture and magnesia is relevant to the argument which is being developed.

All rubbers were compounded as tread mixes according to the formulations given in Table I.

Results

It was pointed out in the previous publication¹⁰ that a survey carried out by K. E. Simpson had shown *p*-phenylenediamine derivatives to be the most efficient antioxidants in increasing the abrasion resistance of NR compounds as assessed by the Akron abrader, and that antioxidants known for producing good heat-aging resistance failed to increase the abrasion resistance; synthetic rubbers were little affected by the antiaging formulation. For this reason, only three NR compounds were used in the present investigation which contained, respectively, 2 parts of Nonox ZA (*N*-isopropyl-*N*-phenyl-*p*-phenylenediamine), 2 parts of Nonox HFN (90% phenyl- β -naphthylamine, 10% diphenyl *p*-phenylenediamine), and no antioxidant. Synthetic rubbers, when protected, contained Nonox HFN.

When the atmosphere is changed during an abrasion experiment, any changes in the rate of abrasion develop only gradually until a constant new level is reached. Figure 1 shows this for an unprotected and a protected NR compound which, after having been abraded in air, were abraded in nitrogen and then in air again. Each point gives the rate of abrasion during the preceding 250 revolutions of the grinding wheel. Attention is drawn to the first reading in air after the nitrogen experiment, which is below the level reached in nitrogen.

All abrasion data quoted in this paper are averages of the last few readings when a stationary state has been reached, and variations in the

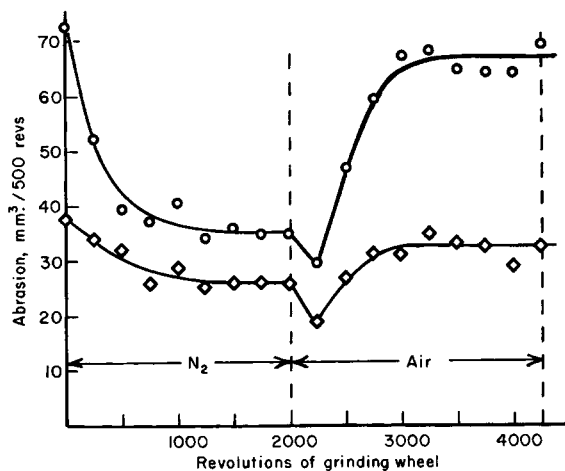


Fig. 1. Change in the rate of abrasion of two NR compounds on changing the atmosphere (grinding wheel): (O) no antioxidant; (◇) 2 phr Nonox ZA.

TABLE II
Abrasion in Air and in Nitrogen under Various Experimental Conditions

Atmosphere	Abrasion, mm. ³ /500 rev.						
	NR			SBR		BR	
	Bayer 4010 (2 phr)	Nonox ZA (2 phr)	Nonox HFN (2 phr)	None	(Nonox HFN, 2 phr)	(Nonox HFN, 1 phr)	EPR (no antioxidant)
Grinding wheel, dust mixture	—	32.9	42.5	66.5	25.2	18.1	43.1
	—	26.1	30.6	36.4	27.0	25.2	29.4
Aluminum wheel, dust mixture	6.2	—	8.0	12.2	9.6	—	—
	15.8	—	14.8	15.8	13.2	—	—
Aluminum wheel, magnesia	—	15.5	25.4	40.2	10.7	0.6	—
	—	4.0	5.2	5.9	2.0	3.7	—
Stainless steel wheel, dust mixture	—	23.4	30.8	48.5	14.8	4.5	—
	—	18.7	23.0	26.8	20.9	19.7	—
Stainless steel wheel, magnesia	—	38.8	52.7	75.0	25.4	12.2	—
	—	10.4	11.9	12.2	7.8	5.0	—

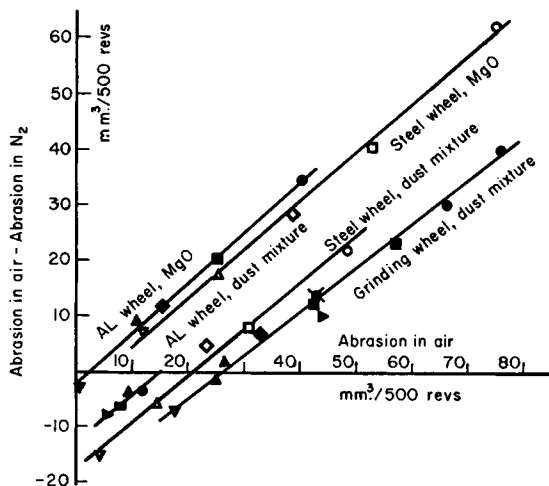


Fig. 2. Difference between the abrasion in air and in nitrogen as function of the abrasion in air: (\diamond) NR, Nonox ZA; (\triangleright) NR, Bayer 4010; (\square) NR, Nonox HFN; (\circ) NR, no antioxidant; (\triangle) SBR, Nonox HFN; (∇) BR, Nonox HFN; (\times) EPR, no antioxidant.

abrasion values are judged to be due to scatter. This can take a long time and limits the number of valid data which can be accumulated with one sample.

Table II lists the abrasion results obtained with NR and synthetic tread compounds in air and in nitrogen on the various wheels using the two drying dusts described in the last section. It was not practical to use magnesia on the grinding wheel because of clogging. The data in the third row are reproduced from a previous report.¹⁰ All values were determined before the importance of the relative humidity for abrasion in air was realized; this aspect of the work will be discussed presently.

A survey of the results obtained in air shows that the effect of the antioxidant on the abrasion of the NR compounds is practically independent of the experimental conditions although the absolute values vary from case to case. The superiority of the *p*-phenylenediamine derivatives Nonox ZA and Bayer 4010 over Nonox HFN (which is mostly phenyl- β -naphthylamine) is clearly shown; the unprotected rubber wears at about twice the rate of the rubber containing Nonox ZA as antioxidant. Similarly, the ratio between the abrasion of NR and SBR is approximately constant, with the exception of the data found on the aluminum wheel with the dust mixture. The abrasion of *cis*-polybutadiene rubber (BR) tends to be very low on the metal wheels. Abrasion on the metal wheels depends in all cases greatly on the dust and is without exception higher with magnesia than with the mixture.

The effect of nitrogen is, at first sight, confusing. The natural expectation of lower abrasion in nitrogen than in air is not universally borne out, the most flagrant example being the results on the aluminium wheel with

TABLE III
Abrasion in Different Atmospheres

	Abrasion, mm. ³ /500 rev.			
	NR		SBR	BR
	Nonox ZA (2 phr)	Nonox HFN (2 phr)	(Nonox HFN, (2 phr)	(Nonox HFN, 1 phr)
		None		
		Grinding wheel, dust mixture		
Laboratory air (open box)	36.1	47.8	26.0	15.3
Stream of laboratory air (<20% R.H.)	25.5	30.6	24.7	23.9
Cylinder air	31.5	36.6	30.5	24.1
Nitrogen, dry	29.0	33.3	29.6	23.0
Nitrogen, moist (70% R.H.)	27.0	30.6	21.9	15.4
Laboratory air	40.2	55.2	29.4	18.9
Oxygen	27.6	39.0	31.2	24.2
		Stainless steel wheel, magnesia		
Stream of laboratory air (53% R.H.)	37.0	49.7	19.8	6.7
Stream of laboratory air (<20% R.H.)	20.4	32.1	11.4	8.7
Cylinder air	22.9	33.7	11.9	7.1
Nitrogen	18.9	23.8	11.8	5.9

the dust mixture. Other cases of a higher abrasion in nitrogen than in air are SBR and BR on the grinding and stainless steel wheels when using the dust mixture. The application of magnesia, however, results with one exception in a drastic reduction of the abrasion in nitrogen.

A more coherent picture of the nitrogen effect emerges when the difference between the abrasions in air and in nitrogen are plotted against the abrasion in air (Fig. 2). This graph contains all the data in Table II and those given in reference 10, Table I. It is seen that for any one experimental condition, all points fall very nearly on a straight line, irrespective of the polymer or, in the case of NR, of the antioxidant. The simple linear relation exhibited by Figure 2 may be fortuitous because no account has been taken of the effect of the atmospheric humidity on the abrasion in air. The graph suggests, nevertheless, that only those compounds which have a low rate of abrasion in air tend to increase it in nitrogen. For a given condition of testing there appears to be a critical value of the abrasion in air which remains unchanged when the atmosphere is changed to nitrogen. With magnesia as dusting agent, this critical abrasion approaches zero, that is to say, the abrasion is expected to be always higher in air than in nitrogen with this dust.

p-Phenylenediamine derivatives are known to be antiozonants, and this could conceivably be the reason for their increasing the abrasion resistance of NR. The *prima facie* case for ozone attack is weak because the ozone concentration in the laboratory is extremely low; also, the abrasion in air and nitrogen of ethylene-propylene rubber (EPR), which has an outstanding ozone resistance, conforms to the behavior of the other polymers (Fig. 2). Fragmentary experiments were carried out in air which had been passed through a saturated potassium iodide solution without revealing any changes in the relative abrasion resistance of the the NR compounds.

More complete measurements were made on the grinding wheel in air taken from a cylinder, which should also be free from ozone. The upper part of Table III gives the results; the absolute values of the abrasion in air and nitrogen differ slightly from those in Table II which were obtained with a different batch of samples. The abrasion of the NR compounds is lower in cylinder air than in laboratory air, and the relative difference between them is somewhat smaller; the abrasion of the synthetic rubbers, however, is increased in cylinder air. Oxygen, surprisingly, gives results which are broadly similar to those in cylinder air, but the difference between the variously protected NR compounds is significantly widened in this gas. The preliminary conclusion drawn from these findings is that oxygen, and not ozone, produces the difference between the abrasions in air and in nitrogen. This leaves the question open why abrasions in laboratory air and cylinder air should differ at all.

Cylinder air is dry, and experiments were therefore carried out in laboratory air of controlled humidity. The results are shown in Figure 3. Although there are only three points to each curve, and the humidity of

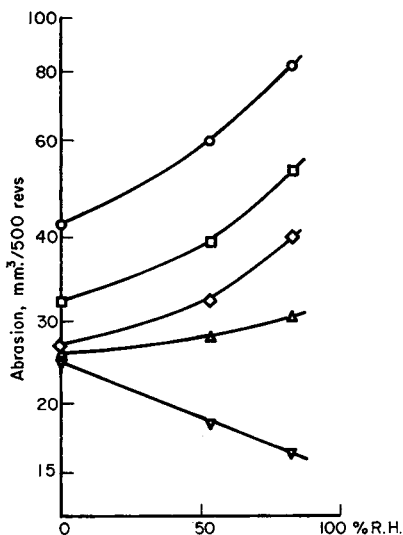


Fig. 3. Effect of relative humidity on the abrasion in air with the grinding wheel. Key as in Fig. 2.

the air which had been sucked through the cold traps (see the section on experimental procedure) has arbitrarily been put down as zero, the graph serves to show the great effect of the relative humidity on the rate of abrasion of NR and BR. It also demonstrates the narrowing of the difference between the rubbers in a dry atmosphere, in agreement with the data for cylinder air in Table III. Abrasion in cylinder air is, for unknown reasons, about 20% higher than in dry air, but the relative values are approximately equal in both atmospheres. Table III gives furthermore the results of a single experiment in moist nitrogen in which both synthetic rubbers abraded less than in the dry gas.

The humidity effect, once established, has led to an explanation of the puzzling fact that the level of abrasion in laboratory air depended on whether the rubber glove was removed, and the machine operated with bare hands, or a stream of air was sucked through the enclosure and the glove used. In the first case, perspiration most probably increased the relative humidity around the sample. The data given in Figure 3 had therefore all been determined in an air stream of the indicated humidity.

The influence of the relative humidity on the abrasion with the stainless steel wheel and magnesia is shown in the lower part of Table III. The abrasion of the BR compounds was so low that the scatter obscured finer differences. (The low abrasion of this polymer in the repeat experiments was probably due to its low coefficient of friction. The angle at which the tips of the small pyramids on the metal wheel meet the oncoming rubber may be very near the critical angle, and a small decrease in its slope during prolonged use can lead to a large decrease in the local stress.) The other rubbers follow the pattern of the results with the grinding wheel

but it is important to notice that the antioxidant effect on the abrasion of NR is here larger in the dry than in a humid atmosphere. The abrasion in nitrogen, though low, was not reduced as much as in the first experiments with the stainless steel wheel; we cannot account for this lack of reproducibility.

Discussion and Conclusions

The main argument for oxidative attack during abrasion in air on the type of track employed in our experiments is the behavior of variously protected NR samples in different atmospheres, in particular, the narrowing of the differences between them in nitrogen. In this latter respect, there is a serious discrepancy between our and the Russian results.³ Also, the ratios between the abrasion in air of an unprotected and similarly protected compound found by us is greater than those reported by Brodskii and his co-workers. It is significant in this context that the difference between the NR samples is reduced in nitrogen even when the absolute values are higher than in air (aluminum wheel with the dust mixture), and that furthermore the difference between NR and the synthetic rubbers is less in nitrogen than in air (Table II).

Higher abrasions in nitrogen than in air have previously been ascribed by us to the incidence of smearing in air but not in nitrogen.¹⁰ This explanation is, in our opinion, strikingly confirmed by a comparison between the results obtained on the metal wheels with the Fuller's Earth-Carborundum mixture and with magnesia as dusting agents. It has been shown in an earlier investigation that magnesia is a better adsorber of the contaminants occurring during abrasion than Fuller's Earth (which we take to be the active component in the dust mixture).⁹ Hence it always produces, with one badly documented exception, a higher abrasion in air than in nitrogen. On the other hand, Fuller's Earth, though a poor adsorber, had a particle size some 70 times larger than magnesia and appears to be intrinsically a better abrasive than magnesia which, according to Grosch,¹¹ acts very much like a solid lubricant; the rates of abrasion in nitrogen are therefore greater with the dust mixture than with magnesia.

The effect of visible smearing on the rate of abrasion and, vice versa, the intensity of smearing at different rates of abrasion have been investigated by the author some time ago when he found that smearing increased pronouncedly with decreasing rate of abrasion (Fig. 11 in reference 12). These findings are in keeping with our results as represented by the graphs in Figure 2 which have demonstrated that, under given conditions, a rubber with a low rate of abrasion in air will tend to abrade at a higher rate in nitrogen. If this is accepted, magnesia must be considered as virtually preventing the effects of smearing.

It appears from Figure 1 that oxidative degradation affects a thin surface layer on the sample which, on changing the atmosphere to nitrogen, must first be worn off before a stationary state is reestablished. Similarly, when continuing in air a degraded layer is slowly re-formed. The first low

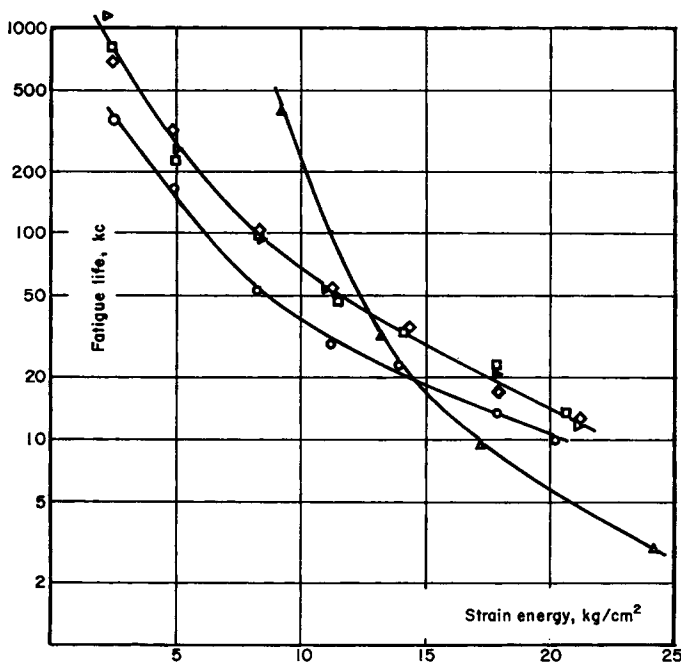


Fig. 4. Fatigue life of the NR and SBR samples as function of the maximum strain energy during a cycle. Key as in Fig. 2.

reading in air after abrasion in nitrogen is then explained by still undegraded rubber being abraded but smearing setting in instantaneously. The source of smearing must lie in the abrasion process itself and originate from local degradation in the presence of oxygen. The effectiveness of magnesia as a drying agent can be gauged from the observation that when this dust is used, the increase in the abrasion of an NR sample in air after abrasion in nitrogen is continuous, and not preceded by the initial drop found with the dust mixture (Fig. 1).

The effect of the relative humidity on the rate of abrasion is hard to explain. Part of the effect could be due to a trivial cause. The amount of dust picked up by the dosing device and its adherence to sample and abrasion wheel may be different in a dry and a humid atmosphere, but this would not account for the dependence of the ratings in dry air on the nature of the dust nor for the results obtained with the BR compound. It must be concluded, though, that the observed abrasion levels in nitrogen and oxygen are partly due to dryness.

The interpretation of these data would be facilitated by a knowledge of the effect of the relative humidity on the fatigue life which we assume to be the relevant strength property governing the abrasion process on our tracks.

Our evidence for a fatigue mechanism is based on the observation that the influence of antioxidants and atmospheric oxygen on the abrasion of

NR is similar to their effect on the fatigue life of these compounds.¹³⁻¹⁵ Figure 4 shows the fatigue life during cyclic extension of the NR and the SBR compounds (median of six determinations), plotted against the maximum strain energy during a cycle. The difference between the lives of the protected and the unprotected compounds is clear but we have not been able to differentiate between the effects of the various antioxidants which all approximately double the life of an unprotected compound. The two curves converge at high strain energies where the failure becomes more nearly akin to a tensile break, and antioxidants become less effective.

The difference between the strain-energy dependence of the fatigue life of NR and SBR is a well-established effect;¹⁴ the strain energy at which the curves for the protected NR and the SBR compounds cross is an estimate of the strain energy arising in abrasion because the abrasion resistances of the two polymers are of the same order of magnitude; its value is about 12 kg.-cm./cm.³ and corresponds to a strain of roughly 120%.

The effect of atmospheric oxygen on the fatigue life of NR has been investigated by Gent¹³ and by Lake and Thomas.¹⁵ Gent found a tenfold difference between the life of an unprotected gum NR *in vacuo* and in air, and a twofold difference for a rubber protected by 1 part of Nonox ZA or Bayer 4010 (at 200% extension). Lake and Thomas, working with an unfilled vulcanizate containing phenyl- β -naphthylamine, ascertained that the ratio between the fatigue lives *in vacuo* and in air increased with decreasing strain energy; at a strain energy roughly equivalent to that thought to obtain in abrasion, the ratio is about 6. For comparison, the largest ratio between abrasion in air and in nitrogen is, from Table II, 6 for the unprotected samples, and 4 for the rubber containing Nonox ZA (metal wheels dusted with magnesia).

It would be idle to attempt a more quantitative comparison between abrasion and fatigue because of the complications arising from smearing and humidity. The circumstantial evidence, taken together with the Russian results, is nevertheless strongly in favor of a fatigue mechanism.

The effect of dust and humidity on abrasion must have a bearing on tire wear, and could be responsible for the large scatter of road test results sometimes observed under ostensibly similar test conditions.¹⁶ The nature of the dust on the road can depend on the strength and direction of the wind preceding and during the test. The only published data on the effect of the relative humidity on the rate of tire wear have been given by Geesink and Prat,¹⁷ who report a decrease in the wear of, presumably, NR tires with increasing humidity but do not state whether any accompanying changes in the ambient temperature had been allowed for; their evidence does not appear to be conclusive.

The relevance of fatigue abrasion for tire wear may be assessed from the results of road tests carried out with variously protected NR passenger tires.¹⁰ *p*-Phenylenediamine derivatives did indeed significantly increase the wear resistance as compared with Nonox HFN but the improvement was not as great as that in Akron abrasion. It was deduced from these

findings that the tensile failure mechanism plays an important role in tire wear in this country; this conclusion agreed with the results of abrasion measurements on a road surface in "crab walk" which revealed only the relatively small differences in the abrasion resistance of the NR compounds which had been found on silicon carbide paper as abrasive.¹⁰ Fatigue is therefore not the only, and possibly not the dominating, mechanism in tire wear. It must also be borne in mind that wear due to tensile failure, because of its intensity, can determine the life of the tire, even if present only for a fraction of the total mileage.

The abrasion experiments referred to in this paper were made by Mr. N. Sathiamoorthy, and the fatigue measurements in the Physical Testing department of these laboratories. The author is obliged to Dr. K. A. Grosch for carrying out the road tests.

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