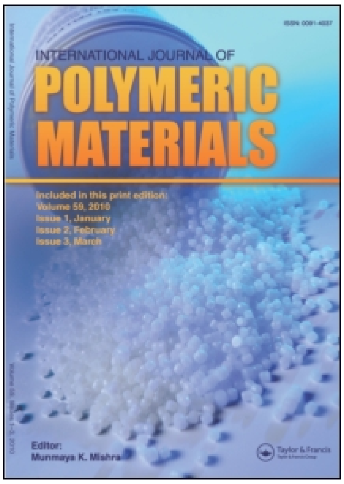

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Tensile Strength and Ultimate Elongation of Rubber-Fibrous Compositions†

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Data are presented which show that the mechanism of fracture in rubber-fiber composites is governed by the adhesion of the matrix material to the fiber as well as the strength of the matrix. Degree of orientation of the fiber is also important.

INTRODUCTION

The reinforcement of plastics (thermoplastic and thermosetting) with short fibers of various nature leads, as a rule, to an increase in their tensile strength.[1,2] Yet, it is known that incorporation of fibrous fillers into elastomeric matrices (filled and unfilled rubbers) results in a substantial breaking strength reduction.[3,4] In this case the dependence of the composition strength $|\sigma_c|$ on the fiber concentration $|V_f|$ has a well defined minimum. An equivalent strength of the matrix and the composition is achieved, at best, when the fiber concentration is from 10 to 15 volume per cent. However the nature of the minimum in the dependence $\sigma_c|V_f|$ and the versatility of its shape for the elastomer-based compositions of various types have not yet been elucidated. As was shown by Derringer,[3] the nature of the fiber, degree of orientation and adhesion to the elastomeric matrix influence the strength and ultimate elongation of the rubber-fibrous compositions. Yet any quantitative estimates of the required adhesion level and of the orientation influence have not been known.

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This article deals with the results of experimental investigation of: (a) the dependence of strength $|\sigma_c|$ and ultimate elongation $|\varepsilon|$ of compositions on the fiber concentrations $|V_f|$ in various types of elastomeric matrices; (b) the influence of the adhesion level on the composition failure character and the type of dependences $\sigma_c|V_f|$ and $\varepsilon|V_f|$; (c) the influence of the fiber concentration on the degree of its orientation in the elastomeric matrix.

EXPERIMENTAL

Materials

According to the character of the stress-strain curves two types of elastomeric matrices have been chosen (Figure 1): (1) rubbers (vulcanizates) of high strength within 15–30 MPa based on caoutchouks filled with carbon black (for instance, PIR, SBR, etc.) in which the most intensive growth of stress occurs at deformations $\varepsilon > 2.5$–3.0; (2) rubbers (vulcanizates) based on unfilled noncrystallizable caoutchouks (SBR) which possess low strength of the order 2–4 MPa, in which the stress monotonously grows with increase of ε. Vulcanizers, antioxidants, modifiers are also added into the rubbers. Various adhesion to fiber was obtained by varying the concentration of a special modifier in the composition.

High-strength fibers were used as reinforcing elements—nylon 6 ($\sigma_f \cong 700$ MPa), length $L \cong 3$ mm, diameter 2.7×10^{-2} mm.

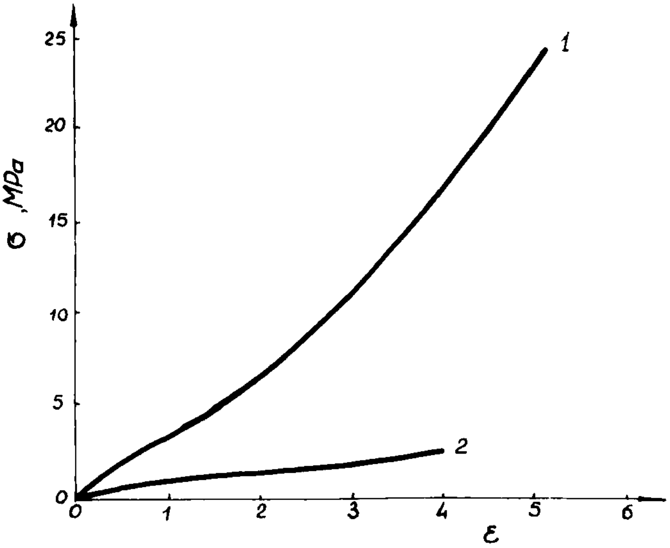

FIGURE 1 Stress-strain relation for elastomers: 1—PIR-based vulcanizate + technical carbon; 2—SBR-based vulcanizate.

The fibers were fed into the rubber mixtures in rolls. In the process of blending treatment the fibers remained practically undamaged. Vulcanization of the rubber-fibrous compositions was effected: PIR + black carbon ПM-100 – 45 min at 143°C, SBR – 120 min at 143°C.

Investigation methods

The strength of standard specimens ($50 \times 6.5 \times 2$ mm) was determined on a universal tensile tester of the Instron Company at a stretching rate of 100 mm/min.

The adhesion of rubber to fiber, i.e. the minimum shear stress on the boundary τ (MPa) was measured using H-method, by pulling the thicker nylon 6 fibers (0.8 mm) out of a rubber block. The PIR-based compositions had three values of the bond strength (MPa): 0.8; 4.1; 5.30.

RESULTS AND DISCUSSION

Figure 2 shows the relation of σ_c and ε of the PIR-based compositions (type 1 matrix) reinforced with short nylon 6 fibers, to V_f. The specimens were cut along the calendering line where the effect of fiber orientation is at its maximum. Nevertheless, the growth of composition strength and the shape of the minimum in the relation $\sigma_c(V_f)$ depend to a large extent on the degree of adhesion. Figure 3 shows the character of specimens failure ($V_f = 10$ volume per cent) when $\tau = 0.8$ MPa and $\tau = 5.30$ MPa. In the first case pulling of fibers out of rubber has been observed while in the second case the character of sample failure indicates that fibers were broken, i.e. adhesion governs the mechanism of rubber-fibrous compositions failure.

When $\tau \rightarrow 0$, the fibers of the test specimen under tension slide relative to the matrix. At high τ values the fibers are loaded via the matrix thus limiting the deforming ability of the latter (Figure 2b). The composition will not break until the fibers oriented in the direction of stretching break. It is proved by Figure 3, as well as by the following experimental fact: the compositions with $\tau = 5.3$ MPa did not lose their strength after thermal ageing (72 hours at 100°C). The strength of rubber is known to be reduced under such conditions by nearly one half while that of a thermally stabilized fiber remains substantially unchanged. The adhesion modifier used in the compositions which were studied permits maintenance of τ under the conditions specified. The retention of the composition strength proves that breakage of fibers is the initial act of failure.

One of the prerequisites for efficient reinforcement of materials with short fibers lies in complete utilization of their strength. This is achieved when the

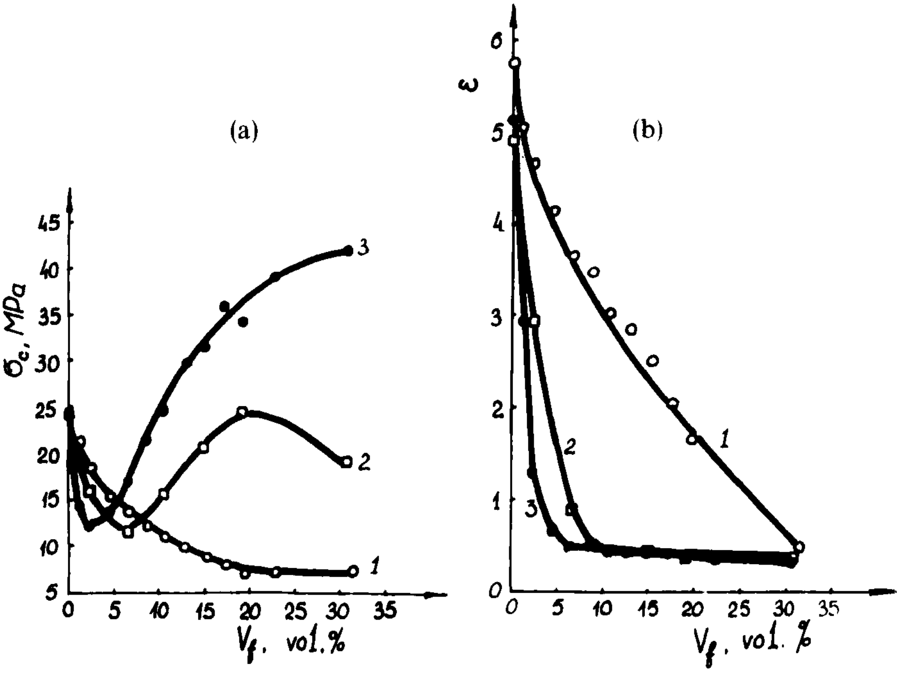

FIGURE 2 Breaking stress (a) and ultimate elongation (b) relations of a rubber-fibrous composition (type 1 matrix) to nylon 6 concentration (parallel to the calendering line): 1—τ = 0.8 MPa; 2—τ = 4.1 MPa; 3—τ = 5.3 MPa.

length of the fibers L exceeds a certain ineffective length l_c which can be calculated on the condition that the force required for breaking the fiber is equal to the maximum shear force on the fiber-rubber boundary[5]:

$$l_c = \frac{\sigma_f \cdot d}{2\tau} \tag{1}$$

where σ_f and d is the strength and diameter of the fiber respectively.

Hence, the efficiency of reinforcement with the fiber of a finite length is a function of τ value. For curves 1, 2 and 3 (Figure 2) l_c equals, mm: 11.7; 2.3; 1.8; i.e. when $L = 3$ mm one can obtain a composition with a higher strength as compared to the matrix rubber if $\tau \geqslant 3.15$ MPa.

To explain the shape of dependences $\sigma_c(V_f)$ let us use the theoretical diagram proposed by A. Kelly and W. Tyson for computing the efficiency of filamentary reinforcement of metals:[6]

$$\sigma_c = \sigma_f V_f\left(1 - \frac{l_c}{2L}\right) + \sigma_m V_m \tag{2}$$

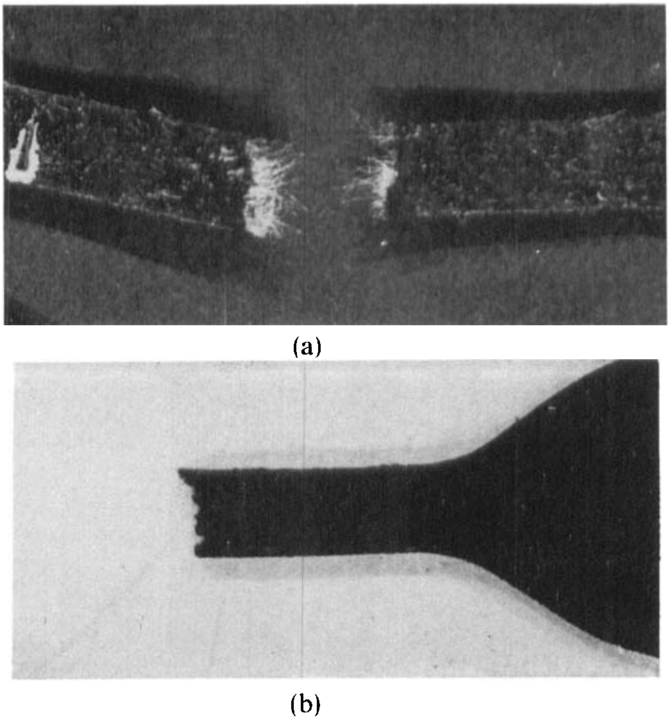

FIGURE 3 Fractured by stretching rubber-fibrous specimens with various bond strength values (τ) between fiber and matrix: (a) $\tau = 0.8$ MPa; (b) $\tau = 5.3$ MPa.

where σ_m and V_m is the strength and the volumetric portion of the matrix respectively.

Rubber reveals its strength properties under high elongations ($\varepsilon > 4$), though, as it is shown in Figure 2b, breakage of the rubber-fibrous composition takes place at far lower elongations. Therefore the influence of the matrix on the value of σ_c will depend not on σ_m but on its stretching resistance at the maximum composition deformation $\sigma_{m(\varepsilon)}$. In addition, it is necessary to take into account the actual orientation of the fiber. Then the strength of a rubber-fibrous composition will be expressed by the following equation:

$$\sigma_c = \sigma_f V_f \left(1 - \frac{l_c}{2L}\right) K + \sigma_{m(\varepsilon)} V_m \qquad (3)$$

where K is the coefficient of fiber orientation.

The deformability of the fibers is much lower than that of the matrix ($\varepsilon_f = 0.2$; $\varepsilon_m = 5$). When the adhesion is sufficiently high ($L \geqslant l_c$) the fibers located parallel to the direction of stretching break thus causing fracturing of the entire material. Therefore $\varepsilon \ll \varepsilon_m$. Even at the fiber concentration of 2 volume per cent $\varepsilon = 1.65$ and the ratio $\sigma_m/\sigma_{m(\varepsilon)}$ continues to grow with the increase of V_f. In such case the weakening influence of the matrix on the strength is not compensated by the effect of the fiber. This is the cause of the minimum on the dependence $\sigma_c(V_f)$ at low values of V_f. With the increase of V_f the effect of the fibers on σ_c increases and we observe an increase in the strength of the composite.

The shape of the curve $\sigma_c(V_f)$ is determined to a large extent by the fiber orientation. Figure 4 shows the strength and ultimate elongation of test specimens ($\tau = 5.3$ MPa) cut out transversely to the calendering line. The strength of the matrix has not been achieved within the investigated range of fiber concentrations due to the low value of the orientation factor $K_\perp < K_{||}$.

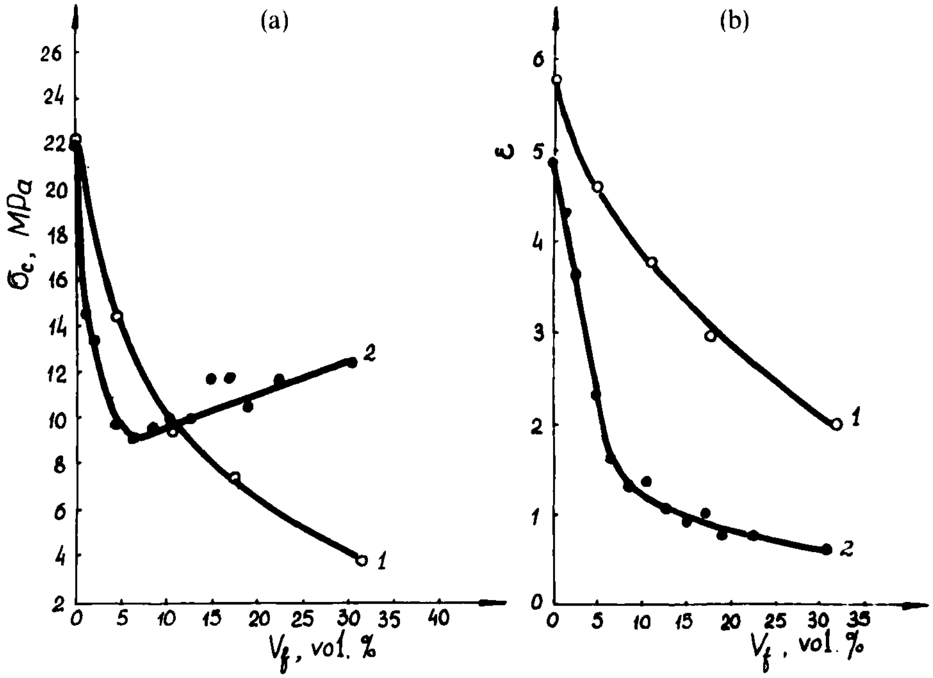

FIGURE 4 Breaking stress (a) and ultimate elongation (b) relations of a rubber-fibrous composition (type 1 matrix) to nylon 6 concentration (perpendicular to the calendering line): 1—$\tau = 0.8$ MPa; 2—$\tau = 5.3$ MPa.

The experimental results illustrated in Figure 1 (curve 1), 2 and 4 were used for calculating values $K_\perp$ and $K_{||}$.

The values obtained show that the orientation factor depends not only on the calendering direction but also on the fiber concentration. The fibers are oriented mainly parallel to the direction of calendering only at a low concentration of 2.5 volume per cent. Then the chaoticity of their distribution increases and $K_{\parallel}$ is ~3.9 times higher than $K_{\perp}$ within the whole range of concentrations, i.e. the relative quantity of fibers parallel and perpendicular

V_f	$K_{\parallel}$	$K_{\perp}$	$K_{\parallel}/K_{\perp}$
2.5	0.71	0	—
5	0.59	0.10	5.9
7.5	0.55	0.14	3.9
10	0.52	0.13	4.0
15	0.45	0.12	3.9
20	0.40	0.10	4.0
25	0.35	0.09	3.9
30	0.31	0.08	3.9

to the calendering line remains unchanged. With the increase of V_f by 20 volume per cent σ_c growth rate (Figure 2a) is not reduced, thus explaining the reduction of $K_{\parallel}$. An increase in the quantity of fibers in the composition above 5 volume per cent hampers their orientation. An increase in the length of fibers has a similar influence on σ_c. According to Eq. (3) an increase in fiber length must lead to an increase of σ_c. However, as it is shown in Figure 5, $L = 10$ mm fibers result in obtaining a composition with a lower strength as compared to $L = 3$ mm fibers. Thus, an increase of L does not

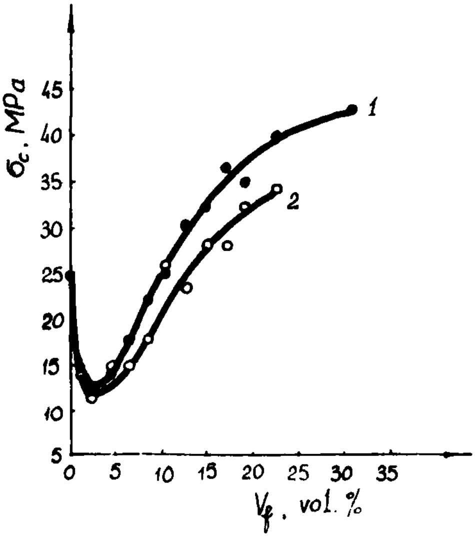

FIGURE 5 Breaking stress relation of the rubber-fibrous composition (τ = 5.3 MPa) to the concentration of 3 mm long (curve 1) and 10 mm long (curve 2) pieces of nylon 6.

lead to an increase of σ_c. An increase of τ has a more effective influence on the strength of a rubber-fibrous composition. It should be noted that the processability of a composition is also material improved when $L \geqslant 3$ mm. An optimum value of L/d for reinforcing fibers is often given in literature. The experimental results prove that this value is governed by the value of τ: an increase of the latter enables to use shorter fibers, thus materially improving the processability of rubber-fibrous compositions. Figure 6 shows the strength and ultimate elongation of compositions based on unfilled SBR

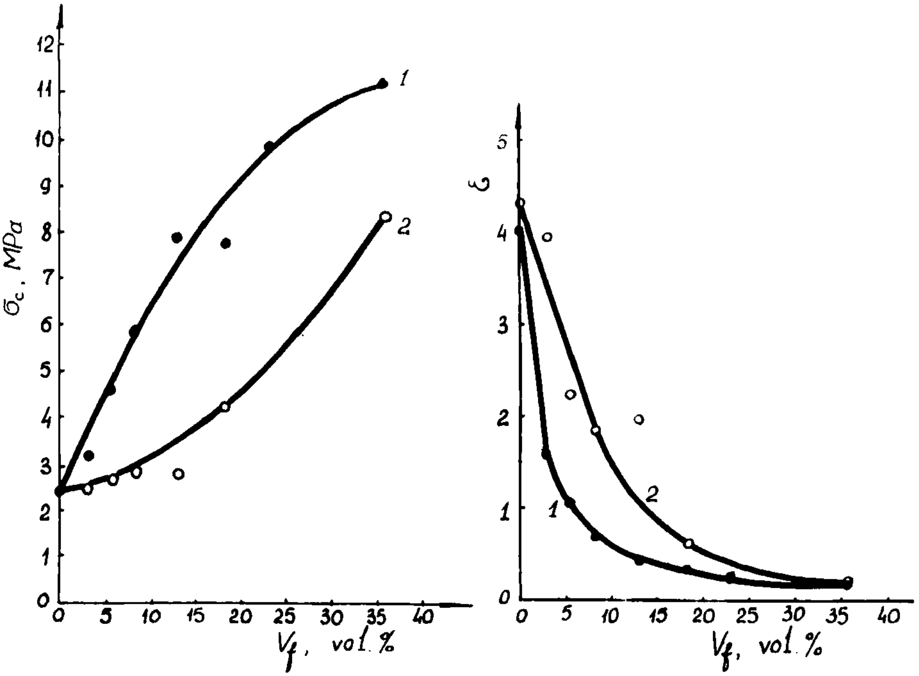

FIGURE 6 Breaking stress (a) and ultimate elongation (b) relation of a rubber-fibrous composition (type 2 matrix) to nylon 6 concentration: 1—parallel to the calendering line; 2—perpendicular to the calendering line.

(type 2 matrix) as a function of fiber concentration. In this case $\sigma_{m(\varepsilon)}$ differs little from σ_m, hence the fiber effectively influences σ_c even at low concentration. According to Eq. (3) the growth of σ_c is observed within the whole range of fiber concentrations, the rate being a little lower when $V_f > 20$ volume per cent (curve 1). In the direction perpendicular to the calendering line σ_c is also higher than σ_m (curve 2), though the anizotropy is sufficiently high.

As the type 2 matrix has low strength, the stretching of the test specimen does not lead to breakage of the fibers, i.e. their strength properties are not utilized completely. Yet, there hasn't been observed any reduction in strength similar to the composition on the type 1 matrix (Figure 2). Hence, the shape of the curve $\sigma_c(V_f)$ for the rubber-fibrous compositions depends on the value of the ratio $\sigma_m/\sigma_{m(\varepsilon)}$. If this ratio approaches unity, then there is no minimum on the curve $\sigma_c(V_f)$. The lower the value of $\sigma_m/\sigma_{m(\varepsilon)}$ ratio, the higher is the relative strength increase σ_c as compared to σ_m.

Thus, for the rubber-fibrous compositions the shape of the curves referring to relationships $\sigma_c(V_f)$ and $\varepsilon(V_f)$ is determined by the type of the matrix, fiber orientation and adhesion on the rubber-fiber boundary.

CONCLUSION

The strength of a rubber-fibrous composition may be described by the additivy rule provided that adhesion and orientation coefficients are introduced and the true influence of the matrix is considered. The orientation coefficient depends not only on the method of processing but also on the fiber concentration.

The mechanism of fracture of rubber-fibrous compositions is governed by the adhesion of the matrix material to the fiber, as well as by the strength of the matrix.

The type of the elastomeric matrix determines the shape of the $\sigma_c(V_f)$ relation curve of the rubber-fibrous composition.

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