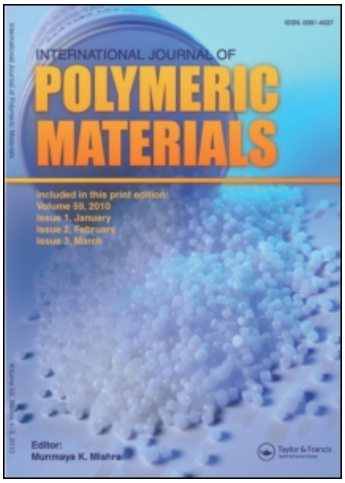

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Mechanical Analysis of "Pullout" Adhesion Test Method

R. A. Turusov[a]; A. S. Freidine[b]; V. N. Kestelman[c]
[a] Institute of Chemical Physics of the Academy of Science, Moscow, Russia [b] Ecolad International, Toronto, Canada [c] JBK International, Radnor, PA, USA

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Mechanical Analysis of "Pullout" Adhesion Test Method

R. A. TURUSOV

Institute of Chemical Physics of the Academy of Science, 4, Kosygin Str., Moscow 117334, Russia

and

A. S. FREIDINE

Ecolad International, Suite 1406, 24 Leith Hill Rd., Toronto, M2J 1Z3, Canada

and

V. N. KESTELMAN

JBK International, Radnor, PA 19087, USA

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The "pullout" adhesion test in the reinforced plastics has some restrictions. The result of these tests depends on geometry of samples and the physical conditions of the testing. This paper represents an attempt to analyze this complex phenomenon. It is shown that the stress depends on the specific properties of boundary layers between the fiber and the polymer matrix. Experimental and theoretical investigations of adhesive strength were carried out in connection with the length of a sample, diameter of a fiber, test temperature and the level of polymer hardening.

KEY WORDS Adhesion, boundary layers, stress, fiber, polymer matrix, adhesive strength.

THEORY AND DISCUSSION

The "pullout" method is used widely for determination of adhesive strength between polymer matrix and fiber in reinforced plastics. According to this method the fiber is pulled out from a small polymer cylinder formed and hardened on this fiber. In these tests the mean (integral) shear strength $\bar{\tau}$ is usually determined as ratio of breaking load to square of polymer-fiber contact.

This strength depends not only on test temperature but also on geometric and physical parameters of specimen and test conditions.[1] Therefore, $\bar{\tau}$ is not true strength of the adhesive-substrate pair. In this paper we made attempt to analyze this problem with the help of the method of boundary layers (MBL).[2,3]

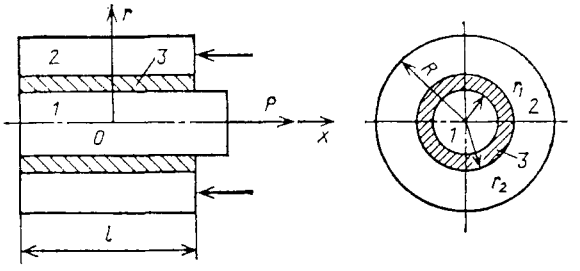

FIGURE 1 The diagram of "pullout" model of reinforced materials. 1. Fiber (rod). 2. Matrix. 3. Boundary layer.

The "pullout" method, diagram of loading of this model, and the boundary layer between adhesive (polymer matrix) and substrate (reinforcing fiber) are shown at Figure 1. Mechanical properties of this boundary layer are anisotropic and different from properties of polymer and fiber.[2,3] The mean shear adhesive strength was measured in test. This strength is integral result of the stresses interaction initiated by outer forces P and residual stresses. Residual stresses are generated by difference between temperature of hardening and testing, density change of polymer matrix due to the hardening crystallization, swelling in liquids, etc.

The coefficient of linear expansion of polymers is usually higher than that of fibers. On cooling after hardening, polymer strives to decrease its axial dimensions. The adhesion opposes this action and boundary tangential stresses are formed on interface. These stresses are relieved by outer force P.

At the same time the deformation of polymer in radial direction produces the friction forces.

Let's suppose that rupture of such a simple adhesion model of reinforced plastic is a result of shear stresses on the matrix-polymer boundary. We shall not consider the destruction by normal radial stresses σ_r along circle $x = 1/2$, $r = r_1$. These stresses may appear because of axi-symmetrical bending of polymer cylinder by distributed force P. The force P is applied to polymer cylinder by support ring (Figure 1). With increasing radial forces σ_r will increase and influence the breaking force value. However, it was shown that the change of support ring dimensions had no influence on the rupture force.

Tangential stresses consist of internal stresses and stresses from outer forces. Internal stresses press the polymer to the fiber and cause the friction forces which prevent pullout fiber from polymer.

When hardening is occurred at high temperature, thermal internal stresses appear. These stresses are created during the time of cooling from the T_0 to temperature of testing T_t because of difference of the coefficient of the thermal extension of the polymer and fiber. The tangential stresses on the fiber-polymer border are:

$$\tau_{rx}(x, r) = \tau_{rx,p} = \tau_{zx,T} + \tau_{rx,S} \tag{1}$$

where $\tau_{rx,p}$ = shear stresses on the border from outer force P; $\tau_{rx,T}$ and $\tau_{rx,S}$ the temperature and shrinkage stresses. Temperature stresses in the first approximation are:

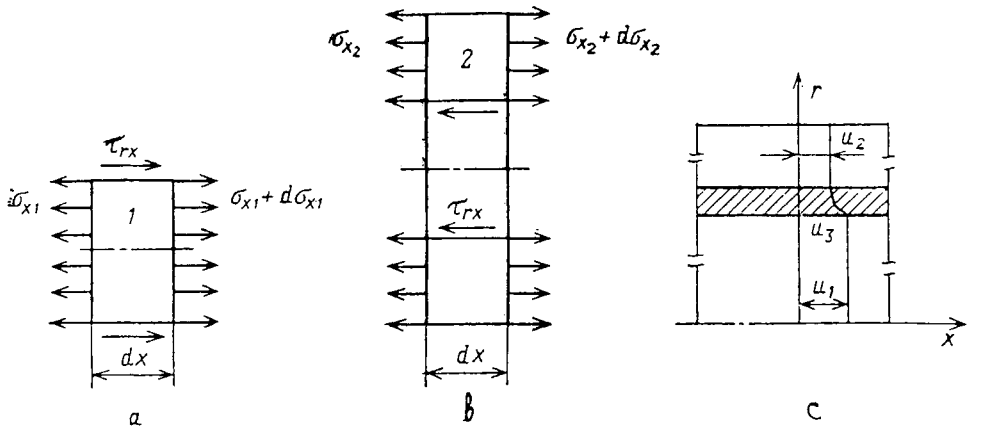

FIGURE 2 Stresses in the central (a) and outer (b) cylindrical rods; (c) deformations "u" along the axis in the rods and boundary layer.

$$\tau_{rx,T}(x, r) = \tau_{rx,T}(\sigma_x, T) + \tau_{rx,T}(\sigma_r, T)$$
$$\tau_{rx,S}(x, r) = \tau_{rx,S}(\sigma_x, S) + \tau_{rx,S}(\sigma_r, S) \quad (2)$$

The first parts in these equations correspond to the tangential stresses which prevent the relative displacement of fiber and polymer along X-axis. The second parts are the analogue of friction forces caused by normal stresses in radial direction. Accordingly it is possible to write:

$$\tau_{rx,T}(\sigma_r, T) = K_1\sigma_{r,T}; \quad \tau_{rx,S}(\sigma_r, S) = K_2\sigma_{r,S} \quad (3)$$

where K_1 and K_2 = the coefficients of friction.

As a result the tangential stresses on the border along the X-axis are:

$$\tau_{rx}(x_1r_1) = \tau_p + \tau_T + K_1\sigma_{r,T} + \tau_S + K_2\sigma_{r,S} \quad (4)$$

To find the function $\tau_{rx}(x, r)$ let's consider the fiber 1 as the continuous rod and the matrix 2 (Figure 1) as the hollow rod. We assumed the existence of some anisotropic boundary layer with small thickness δ between polymer matrix and fiber. This layer can transfer the normal σ_r and tangential $\tau_{r,x}$ stresses. The equilibrium equations of rods elements 1 and 2 with length dx (Figure 2) for element of central rod (fiber) 1 are:

$$(d\sigma_{x1}/dx)dx\pi r_1^2 + \tau_{rx}(r_1)dx2\pi r_1 = 0 \quad (5)$$

For element 2 (matrix):

$$(d\sigma_{x2}/dx)dx\pi(R^2 - r_2^2) - \tau_{rx}(r_2)dx2\pi r_2 = 0 \quad (6)$$

After reduction:

$$\tau_{2x}(x_1r_1) = -(r_1/2)(d\sigma_{x1}/dx); \quad \tau_{2x}(x, r_2) = \frac{R_2^2 - r_2^2}{2_{r_2}} \cdot \frac{d\sigma_{X2}}{dx} \tag{7}$$

If the normal forces in the rods are $N_1 = \pi r_1^2 \sigma_{x1}$, we can rewrite expression 7 in more convenient form for further analysis:

$$dN_1/dx = -2\pi r_1 \tau_{rx}(x_1r_1); \quad dN_2/dx = 2\pi r_2 \tau_{2x}(x, r_2) \tag{8}$$

One of these equations can be changed to equilibrium equation of any part of the model in section x:

$$N_1 + N_2 = 0 \tag{9}$$

The total deformations in layers 1 and 2 are composed of elastic e_{xi}, temperature ε_{Ti} and shrinkage ε_s deformations. Suppose that there are no shrinkage deformations in reinforcing rod:

$$\varepsilon_{x1} = e_{x1} + \alpha(T - T_0); \quad \varepsilon_{x2} = e_{x2} + \alpha_2(T - T_0) + \varepsilon_S \tag{10}$$

where T and T_0 = the current and start temperature.

The elastic deformations correlate with stresses by means of Hook's law:

$$e_{xi} = \sigma_{xi}/E_i = N_i/S_iE_i, \quad i = 1, 2 \tag{11}$$

In layer 3 only shear stresses connected with tangential stresses are originated:

$$\varepsilon_{rx} = e_{rx} = \tau(x, r)/G \tag{12}$$

To get the value of stresses from the frictional forces we need at first to find normal stresses which press polymer to fiber σ_r. For this purpose we solved the flat axi-symmetric problem of residual (from temperature and shrinkage) stresses in composed cylinder. So we found that the tangential stresses from frictional forces on the border between polymer and fiber are:

$$\tau_{rx,T} = \frac{K_1(\varepsilon_{q2} - \varepsilon_{q1})E_1E_2(R^2 - r_1^2)}{E_1[R^2(1 + \mu_2) + r_1^2(1 - \mu_2)] + E_2(1 - \mu_1)(R^2 - r_1^2)} \tag{13}$$

where K_1 = coefficient of friction.

In this case we found the tangential stresses nondependable on the coordinate x. The rupture of our model will start with force P_p^0 and critical maximal stresses near edge of the model (without frictional forces). The breaking is finished by shear the fiber from the matrix by increase force $P_p = P_P^0 + P_f$, where friction force P_f.

$$P_f = -K_1\sigma_{r1}2\pi r_1 l = \tau_{2x,l}2\pi r_1 l \tag{14}$$

The τ_{max} is found from:

$$\tau_{rx}(x, r) = -\frac{f(x)}{r} = -\frac{1}{r}\left(\frac{1}{2\pi}\cdot\frac{dN_1}{dx}\right) = -\frac{\omega}{2\pi}\left[P\,\frac{ch\omega(l/2 + x)}{sh2\nu} - \beta(\varepsilon_{q2} - \varepsilon_{q1})\,\frac{sh\omega x}{ch\nu}\right], \quad \nu = \omega l/2 \tag{15}$$

The rupture starts when $\tau_{max} = \tau_{ad}$ and $P = P_p^0$. For real reinforced plastics in processs of hardening and cooling we usually have $-(\varepsilon_{q2} - \varepsilon_{q1})$. In this case we found the maximal stresses from (15) in the point ($x = l/2$, $r = r_1$):

$$|\tau|_{max} = |\tau_{2x}(l/2, r_1)| = \omega[p(cth2\nu - \beta(\varepsilon_{q2} - \varepsilon_{q1}) + h\nu]/2\pi r_1 \tag{16}$$

If the $|\tau|_{max} = \tau_{ad}$, from (16) we found the force P_p^0:

$$P_P^0 = \frac{\tau_{ad}2\pi r_1}{\omega}\,th2\nu + \beta(\varepsilon_{q2} - \varepsilon_{q1})\,\frac{th\nu}{cth2\nu} \tag{17}$$

The full rupture force is $P_p = P_p + P_f$. The mean rupture stress $\bar{\tau}$ measured in tests is:

$$\bar{\tau} = \frac{P_p}{2\pi r_1 l} = \frac{\tau_{ad}}{2\nu}\,th2\nu + \frac{\beta(\varepsilon_{q2} - \varepsilon_{q1})}{2\pi r_1 l}\,th\nu th2\nu - \frac{K_1(\varepsilon_{q2} - \varepsilon_{q1})E_1E_2(R^2 - r_1^2)}{E_1[R^2(1 + \mu_2) + r_1^2(1 - \mu_2)] + E_2(1 - \mu_1)(R^2 - r_1^2)} \tag{18}$$

We use (18) for the analysis of the dependence of the mean shear strength on some geometric and physical parameters.

Dependence of $\bar{\tau}$ on Glue Line Length

The length in Equation (18) as $\nu = \omega l/2$ is in the first $\tau^{(1)}$ and the second $\tau^{(2)}$ parts of (18). We determine the extreme of l. The extremal significance of $\tau^{(1)}$ can be only if $l = 0$ and $l = \infty$. Dependence $\tau^{(1)}$ on l is shown in Figure 3a. For $\tau^{(2)}$ there are several extreme points. On Figure 3b we can see function $\tau^{(2)}(l)$, if $(\varepsilon_{q2} - \varepsilon_{q1}) < 0$ (hardening or cooling). We obtain $\bar{\tau}(l)$ by adding to $\tau^{(1)}(l)$, $\tau^{(2)}(l)$, the l independent item τ_f. The diagram of this function may be one of the three (Figures 3c; d; e). The dashed line equals to the stress from friction τ_f. With increasing l, all the curves asymptotically approach to τ_f. The curve (Figure 3c) is the same as experimental curve of dependence $\bar{\tau}$ on length of adhesive joint (Figure 4, curve 3).[1] In Figure 3e zone l_1^* l_2^* with negative $\bar{\tau}$ is shown. This zone is an interval of the length of adhesive joints which are ruptured without the external load. Therefore, for some adhesive-substrate pairs and testing temperature $T_{exp} \neq T_0$ we cannot obtain good (non-ruptured) joints with fiber's length $l_1^* \leq 1 \leq l_2^*$.

So when $l \to 0$ significance of $\bar{\tau}$ approaches $(\tau_{ad} + \tau_f)$ where τ_f = the stress from

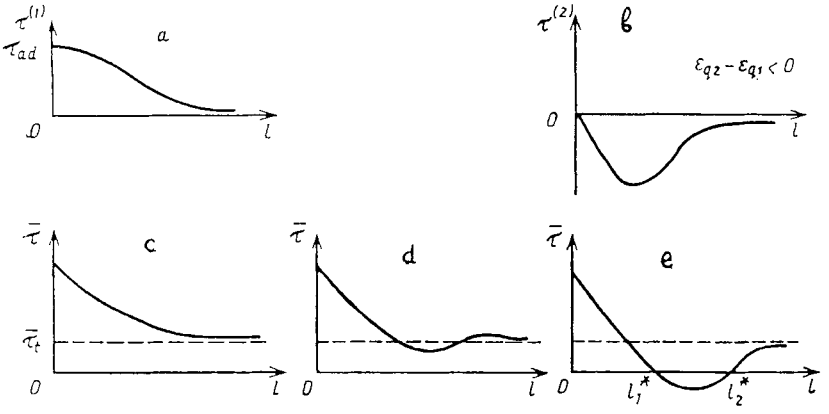

FIGURE 3 Theoretical dependence of the mean strength τ of the polymer-fiber joint on the glue line length l: a, b—dependence $\tau^{(1)}$ and $\tau^{(2)}$ on l; c, d, e—the possible dependence $\bar{\tau}$ on l.

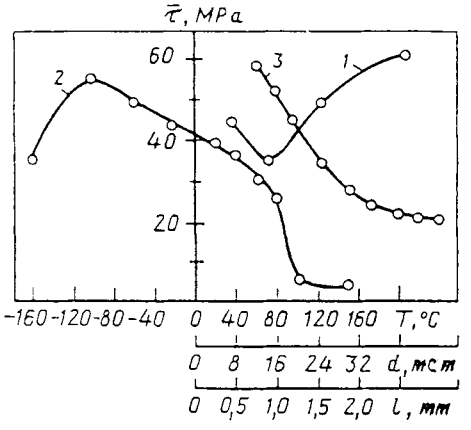

FIGURE 4 Experimental dependence $\bar{\tau}$ for pullout fiber from polymeric matrix on the diameter of glass fiber (1); the temperature (glass fiber) (2) and the length of joint (steel wire) (3).

friction and when $l \to \infty$ this value goes to τ_f. If l is big enough, then the major part of $\bar{\tau}$ comes from friction forces. These facts give the possibility for measuring the "true" shear adhesive strength in "pullout" tests:

$$\tau_{ad} = \lim_{l \to 0} [P_p/(2\pi r_1 l)] - \tau_f; \quad \tau_f = \lim_{l \to 0} [P_p/(2\pi r_1 l)] \tag{19}$$

The method of determination τ_{ad} and τ_f from experimental $\bar{\tau}(l)$ is more realistic provided the theoretical dependence (18) holds for the experimental curve.

Dependence of $\bar{\tau}$ on Diameter of Fiber

Let's consider the first, second and third terms in (18) as $\tau^{(1)}$, $\tau^{(2)}$, and $\tau^{(3)}$. Suppose that thickness of boundary layer δ and layer of matrix $\Delta = R - r$ are constant.

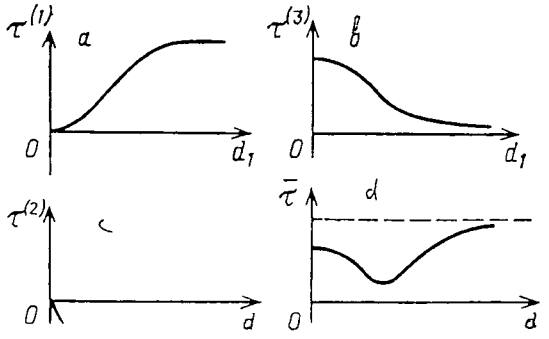

FIGURE 5 Theoretical dependence of the strength of joints τ on the diameter of the fiber d: a, b, c—dependence of functions $\tau^{(1)}$, $\tau^{(2)}$ and $\tau^{(3)}$ on d_1, d—one of the dependence τ (d_1).

Consider the limit of terms in (18) when diameter of fiber $r \to 0$ and $r \to \infty$. In the first case:

$$\lim_{r_1 \to 0} \tau^{(1)} = \lim_{r_1 \to 0} \tau_{ad}/2\nu = 0; \quad \lim_{r_1 \to 0} \tau^{(2)} = \lim_{r_1 \to 0} E_1 r_1^2/(2r_1 l) = 0;$$

$$\lim_{r_1 \to 0} \tau^{(3)} = \frac{K(\varepsilon_{q2} - \varepsilon_{q1})E_1E_2}{E_1(1 + \mu_2) + E_2(1 - \mu_1)} \tag{20}$$

In the second case:

$$\lim_{r_1 \to \infty} \tau^{(1)} = (\tau_{ad}/2\nu_l)th2\nu_l; \quad \lim_{r_1 \to \infty} \tau^{(2)} = \frac{(R - r_2)E(\varepsilon_{q2} - \varepsilon_{q1})}{l} thv_l th2\nu_1;$$

$$\lim_{r_1 \to \infty} \tau^{(3)} = \lim_{r_1 \to \infty} \frac{K_1(\varepsilon_{q2} - \varepsilon_{q1})E_1E_2\Delta(2r_1 + \Delta)}{E_1[(r_1 + \Delta)^2(1 + \mu_2) + r_1^2(1 - \mu_2)] + E_2(1 - \mu_1)\Delta(2r_1 + \Delta)} = 0 \tag{21}$$

Dependence of the $\tau^{(1)}$, $\tau^{(2)}$ and $\tau^{(3)}$ on diameter of fiber is shown in Figure 5 (curves a, b, c). Curves of dependence of τ on the diameter of fiber are obtained by addition of three items in (18). It is known that diversity of these curves may be high. One of the curves is presented in Figure 5d. This theoretical curve is similar to experimental curve (Figure 4, curve 1) for polyepoxide-glass fiber (1).

Dependence $\bar{\tau}$ on the Outer Radius of the Model

It was shown earlier that the outer radius R of the model increases as the radius of fiber increases. Let's consider the case when r = constant and $R \to \infty$:

$$\lim_{R \to \infty} \beta = \pi E_1 r_1^2 = \beta_R; \quad \lim_{R \to \infty} \nu = l/2 \sqrt{\frac{2G}{E_1 r_1^2 \ln(1 + \delta/r_1)}} = \nu_R \tag{22}$$

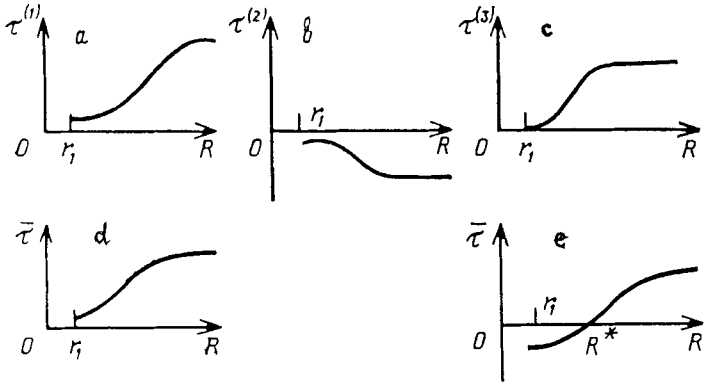

FIGURE 6 Theoretical dependence of the strength of joints τ on the outer radius R of model: a, b, c,—dependence of functions $\tau^{(1)}$, $\tau^{(2)}$ and $\tau^{(3)}$ on R; d, e—variants of dependence $\bar{\tau}$ on R.

Curves $\tau^{(1)}(R)$, $\tau^{(2)}(R)$ and $\tau^{(3)}(R) = \tau_f(R)$ are shown in Figure 6 (curves a, b, c). Curves d, e of Figure 6 present different possibilities of the dependence $\bar{\tau}$ on R. When $R \leq R^*$, the spontaneous rupture of the model without outer loading is possible (Figure 6e).

Dependence $\bar{\tau}$ on the Test Temperature and the Matrix Modulus

Let's consider only residual stress from the test temperature and disregard the residual stress from adhesives hardening. In (18) for $\bar{\tau}$ instead of $(\varepsilon_{q2} - \varepsilon_{q1})$ we use $(\alpha_2 - \alpha_1)(T_{exp.} - T_0)$ and suppose that at starting temperature T_0 the residual stresses are extremely low. The temperature interval $\Delta T = (T_{exp.} - T_0)$ has influence on polymer properties. It is known that Poisson coefficient slightly changes with temperature. But the polymer elasticity modulus E_2 increases with decreasing temperature. Simultaneously β also rises and ν may increase or decrease. Therefore, ν includes elasticity modulus G of boundary layer. Probably this modulus slightly increases with increasing of temperature. For this reason the first item $\tau^{(1)}$ in (18) depends on temperature.

The second item in (18) increases with decreasing temperature. Therefore, β and ΔT also increase. The third item $\tau^{(3)}$ can be presented as:

$$\tau^{(3)} = \frac{K_1(\alpha_2 - \alpha_1)(T_{exp.} - T_0)E_1(R^2 - r_1^2)}{(E_1/E_2)[R^2(1 + \mu^2) + r_1^2(1 - \mu_2)] + (1 - \mu_2)(R^2 - r_1^2)} \tag{23}$$

Since we know the dependence of $E_2(T)$, we can discuss the influence of temperature on this function. When $T_{exp.} > T_0$, the first item in (18) changes slightly. Change of $\tau^{(2)}$ depends on competition between increasing of $\Delta T = T_{exp.} - T_0$ and diminishing of β. At first $\tau^{(2)}$ increases with temperature, but later decreases when $E_2 \to 0$ (Figure 7a). When $T_{exp.} > T_0$, the pressure on the fiber from polymer by the radial stress changes to the pressure from normal stress. In practice the rupture load is reduced because the load includes normal tension stress. Curve for $\tau^{(3)}(T)$ is shown in Figure

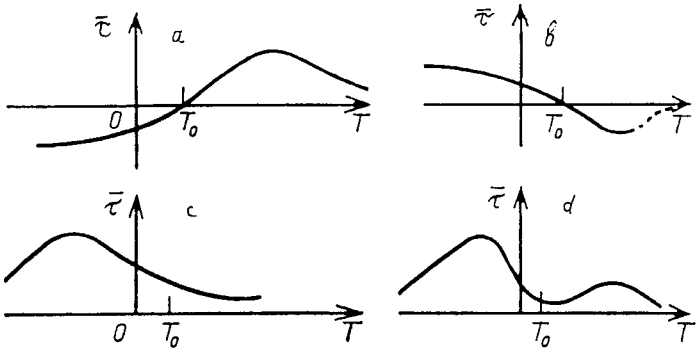

FIGURE 7 Theoretical dependence of the mean strength of joints on temperature: a, b—dependence of the functions $\tau^{(2)}$ and $\tau^{(3)}$ on T; c, d—summary dependence $\bar{\tau}$ on T.

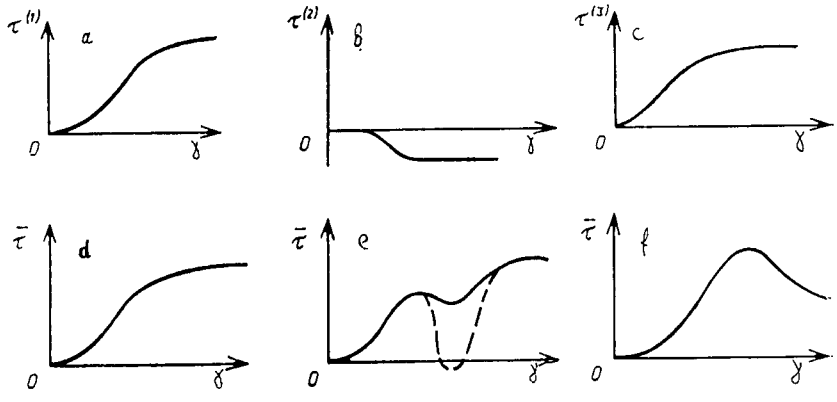

FIGURE 8 Theoretical dependence of the mean strength of the adhesive joints on extent of hardening γ: a, b, c—dependence $\tau^{(1)}$, $\tau^{(2)}$ and $\tau^{(3)}$ on γ if shrinkage is negative; d, e, f—variants of curves for dependence $\bar{\tau}$ on extent hardening.

7b. The curve of τ may have one (Figure 7c) or two (Figure 7d) maxima. The curve in Figure 7c is the same as the experimental curve (Figure 4, curve 2).

Dependence $\bar{\tau}$ on the Hardening Extent

One of the most important factor question is the moment of forming of the adhesive joint. Two cases are considered. The first-adhesive joint is formed when the adhesive is liquid. the second-adhesive interaction is formed during the time of hardening.

It is known that the more the hardening process is completed the larger is the elastic modulus of the polymer. The hardening is usually accompanied by a change of the polymer volume.

In considerations of these factors we discuss our problem.

According to the first case hypothesis the real adhesive strength τ_{ad} is not changed in the process of the polymer matrix hardening. Correspondingly, it is possible to imagine that shear modulus of the boundary layer also is not changed. Therefore, the parameter ν in (18) may be changed only when E_2 ($0 \leq E_2 \leq E_{2,\max}$) = *varia in*

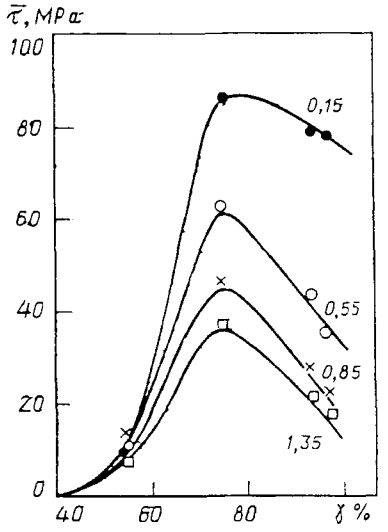

FIGURE 9 Experimental dependence $\bar{\tau}$ for polyepoxide EDT-10—steel wire (d = 15 mcm) on hardening and contact area.

β. The value of β in this case changes from zero to some definite value. The interval of ν change is from infinity (when $E_2 = 0$) to some definite value ν_K (when $E_2 = E_{2,max}$): $\infty \geq \nu \geq \nu_K$ ($o \leq \alpha \leq 1$). Therefore, the first item in (18) during the time of hardening will increase from 0 to some definite value $\tau_K^{(1)}$.

Let's assume that modulus G of the boundary layer increases from some small but definite value to G_{max}. Then the dependence $\tau^{(1)}$ on extent of hardening γ is not changed. Then in the items $\tau^{(2)}$ and $\tau^{(3)}$ we will substitute deformation from polymer's shrinkage $-\varepsilon_S(\gamma)$ for $(\varepsilon_{q2} - \varepsilon_{q1})$. This deformation can be positive or negative. During the time of hardening the parameter β is changing within the limits $0 \leq \beta \leq \beta_K$ and ν—from zero to ν_K. Therefore, $\bar{\tau}$ depends on completeness of hardening, the value and the sign of shrinkage. The first item is always positive, the second one depends on the sign of shrinkage and the third one is the same, but with the opposite sign of shrinkage. In Figures 8a, b, c these three items (the shrinkage is negative) are presented. In Figures 8d, e, f the possible variants of dependence of $\bar{\tau}$ on extent of hardening are shown. If the shrinkage is positive, (i.e. expansion takes place on hardening) the signs of $\tau^{(2)}$ and $\tau^{(3)}$ will change to opposite, but dependence $\tau(\gamma)$ remains the same as in the first case.

According to the second case hypothesis we consider that the value τ_{ad} raises with the extent of hardening, this will lead to only quantitative, but not qualitative change of curves $\tau^{(1)}(\gamma)$.

In Figure 9 the experimental data for polyepoxide matrix on pullout steel wire are shown (1). These curves are similar to theoretical curves (Figure 8e, f).

The similar curves are also typical for the same dependence of hardening of different adhesive joints (shear of lap joints, tension, cleavage, etc.) of metal, wood, and other construction materials (4). Only pure shear (torsion of hole tubular butt joints) gives this dependence without maximum (4). It is explained by very small concentration of stresses in the last case.

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