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On the destruction of elastic polymer materials under the action of an electron beam

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An explanation for a feature found in several experiments in the general picture of the destruction of non-brittle polymers under the influence of a shock wave initiated by a powerful electron beam is proposed. The distance of the cracking region from the surface of the material affected by the beam to a finite length in depth is associated with the three-dimensional nature of the propagation of elastic waves. The universality of the effect is demonstrated by the simplest isotropic model, which shows that large tensile stresses are effectively generated inside the target at its sufficiently large transverse and longitudinal size, even without taking into account nonlinear and shear processes.

Keywords: high-current electron beams, shock waves, polymer materials, high pressures.

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Introduction

Explosive and ballistic generators are classical instruments for excitation of shock waves in studies in the field of high dynamic pressure physics. The desire for further progress in the field of previously inaccessible parameters and the possibility of organizing shock-wave studies in the conditions of the ordinary physical laboratory prompted the use of other methods of intensive influence on matter. In particular, high-power pulsed lasers and high-current electron beams are widely used as sources of high dynamic pressures for studying the elastoplastic and strength properties of metals and plastics (see, for example, [1–4]).

Previously, in experimental studies of the effect of a high-power pulsed electron beam on targets made of polymeric materials, in particular, plexiglass (polymethyl methacrylate —PMMA) and epoxy resin (EDP brand, i.e. Epoxy-Diane resin with Plasticizer), carried out on a high-current relativistic electron beam (REB) generator „Kalmar“ jcite1,2, a „non-standard“ pattern of their destruction was recorded. In the experiments under consideration, the maximum beam electron energy varied in the range 200−300 keV, the maximum beam current ranged from 10 to 20 kA at half-height duration of 100 ns. Under conditions where the frontal dimensions of the target are several times greater than the diameter of the spot of interaction of the beam with its surface, and the thickness of the sample is quite comparable with it or greater, between the region of energy release of the REB and the region of internal damage (cracking) of the polymer, there remains transparent zone of material unaffected by erosion along the direction of beam propagation 2−6 mm at beam diameter of 10−20 mm. In the specified range of parameters, the thickness of this region is practically independent on the geometric characteristics of the irradiated sample and is determined only by the parameters of the incident electron beam. In particular, as the beam diameter on the target surface increases, the thickness of the transparent region increases. In this way, the damage pattern of the mentioned polymers differs from that for materials subject „to brittle fracture“ for example, of glass or polystyrene [1,2]. Possible reasons for the significant difference in „brittleness“ for polymers can be related to the features of stress relaxation and are considered in more detail in the works [5,6].

The reason for the described effect has not yet been established, although among which processes one should look for, it was immediately realized. The depth of the energy deposition of the beam is on the order of hundreds of microns, and therefore destruction in the volume and on the back side of the samples occurs under the influence of the elastic perturbation of the material generated by heating from the REB, which propagates along the target and has the character of the shock wave at the leading edge. Particularly, the works [1–4] were devoted to the initiation of such waves.

Typical set up of the discussed experiments is described below (Fig. 1).

Polymer sample in the form of rectangular parallelepiped was placed in the anode assembly of the diode of the „Kalmar“generator, in which, when a high-voltage pulse was applied to the cathode, REB was formed due to explosive emission, irradiating the sample. The radial distribution of the REB energy contribution to the target was determined from X-ray photographs taken with an X-ray pinhole camera located behind the sample. In the standard case, the shape of this distribution was close to Gaussian one. The features of „Kalmar“ accelerator experiments at the and the technique for measuring the beam parameters are described in detail in the works [1,2]. Fig. 2 shows photographs of PMMA and epoxy resin (EDP) samples after electron beam exposure to them; later on, similar effects were observed more than once (see, for example, [7,8]).

It is well known that the destruction of elastic-plastic materials under tension occurs at absolute values of mechanical stress, which are noticeably (by times) lower than under compression. As part of the study of the discovered phenomenon, experiments were carried out on the same „Kalmar“ machine to determine the moment of destruction of PMMA when observing the propagation of a shock wave initiated by REB over the sample with a good time resolution [7]. It turned out that the erosion of the material occurs at its „destressing“ already after the passage of the shock wave front, i.e. just at the stage of tension of a plastic.

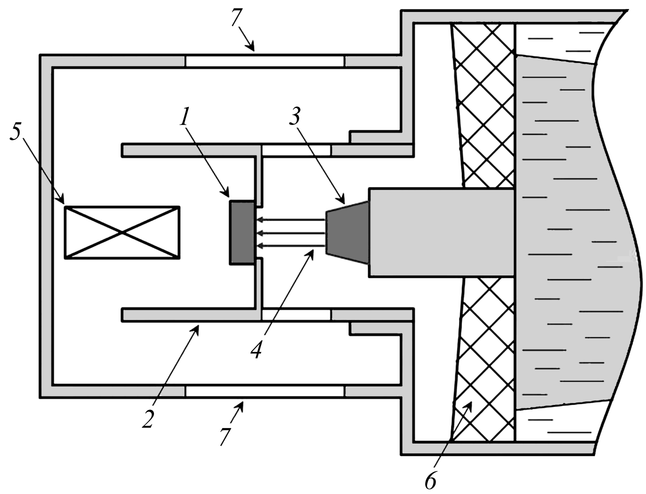

Figure 1. Scheme of experiments to study the effect of REB on polymeric materials: *1* is test sample, *2* is anode assembly, *3* is cathode, *4* is REB, *5* is pinhole camera, *6* is diode insulator, *7* are observation windows.

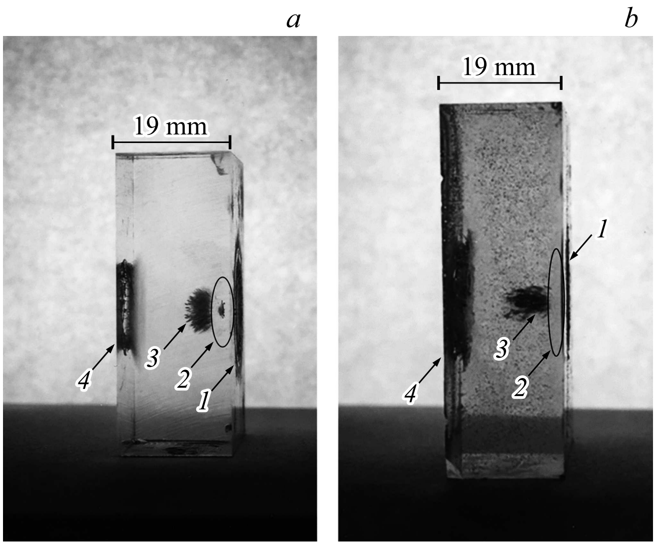

Figure 2. Samples of PMMA 5×5 cm (*a*) and EDP 6.2×6.2 cm (*b*) irradiated with an electron beam; REB was incident on the right surface of the samples. The numbers indicate: *1* is region of evaporation and melting of the target material under the action of an electron beam, *2* is transparent region, *3* is region of internal destruction, *4* is „spall“ region that occurs when a shock wave is reflected from this boundary.

Figure 3. Sample of polystyrene 5.2×5.2 cm, 2.3 cm thick, irradiated with an electron beam; REB was incident on the right surface of the sample.

For comparison, Fig. 3 shows the photograph of the destroyed polystyrene sample irradiated with electron beam under similar conditions. It can be seen that any transparent region between the area of the electron beam exposure on the sample surface and destruction area was not observed.

In this work, based on the above features of the phenomenon, we propose the possible explanation for the „effect of the non-standard“ destruction pattern i.e. the presence of the transparent region between the area of energy release and destruction area, associated with the specifics of the three-dimensional evolution of the elastic wave excited by pressure pulse at the boundary of the medium . It is important that the effect of the appearance of tensile stresses at a distance from the target surface, discussed below, is of an extremely general nature, independent of variations in the mechanical properties of the material. For this reason, the discourse doesn't focus on the specific structure of the polymer, on which, naturally, these properties depend and even anisotropy is possible.

1. Physical model

The propagation of such elastic waves, as it is known, can be quite effectively represented on the basis of simple acoustic model, according to which the stress evolution in a medium σ is described by linear wave equation with the constant speed of sound c_S:

$$\frac{\partial^2\sigma}{\partial t^2} - c_S^2\Delta\sigma = 0, \quad (1)$$

[9,10]. In this case, such approximation is justified by the fact that the values σ achieved in the experiment, on the one hand, are not too large, and the observed shock wave velocities do not greatly exceed the so-called volume sound velocity (see below), and, on the other hand, are sufficient to overcome the yield strength of the plastic, when the elastic properties of the material become similar to those of a liquid or gas (except that the stress — or pressure can also take negative values). Accordingly, the sound velocity in (1) is of the order of $\sqrt{K/\rho}$, where K is the volumetric modulus of elasticity of the material, and ρ is its density, while shear modulus $G \to 0$ [9,12]. In addition, the simplicity of the model makes the conclusions based on it especially clear and reliable. The effects of nonlinearity, anisotropy and dissipation, of course, are capable of modifying some quantitative characteristics of elastic processes, but cannot change the declared qualitative pattern of the phenomenon. Further, the process within (1) is considered in a cylindrical coordinate system with the axis z directed from the boundary, on which the REB falls, deep into the sample. Since the profile of this beam can be considered axially symmetric, σ in (1) is a function of t, z, and r, but not φ.

No presence as such of tensile stresses is subject to explanation, but the achievement by them of values sufficient for the destruction of the material only with the removal of the perturbation from the boundary to the finite distance. In [7], where it was experimentally proved that plexiglass is cracked exactly under its tension, the appearance of the $\sigma < 0$ region was theoretically simulated according to the standard scheme [9] on the basis of the one-dimensional (according to z, i.e. for $\sigma = \sigma(t, z)$) acoustic equation, when unbounded and uniform in r (recall, in a cylindrical coordinate system) electron beam rapidly heats the near-surface layer of the material, thereby generating the medium's stress perturbation, propagating further according to (1). In this case, since the beam introduces energy into the medium, but not momentum, [1], in the forming elastic wave traveling deep into the sample, almost immediately (at distances of the order of the thickness of the energy input region, which, as indicated above, is only a few hundred microns), sections that compensate each other appear, with $\sigma > 0$ in the head and $\sigma < 0$ (due to the so-called „destressing") on the tail [7,9] and only in this case the medium will have total zero-momentum $\int \rho \mathbf{v} dz$, where $\mathbf{v}$ is mass velocity of the medium. With distancing from the boundary, the perturbation amplitude in reality decreases due to the un-accounting for effects of dissipation and wave divergence along the radius due to the finite thickness of the beam that generated it, so that the maximum tensile stresses in the framework of such a model should be reached near the boundary, which does not correspond to the observed pattern „of the break off" of cracking areas (Fig. 2).

In present paper, we pay attention to the fact that this pattern can be explained by changing the model concept of the mechanism of the beam action on the sample and the idea of the geometry of the problem. The changes by themselves are of not-original type (see links below), but it is interesting that only two modifications lead to the goal. The achievement of large negative values σ only in the depth of the material is due to the combined effect of evaporation and spread of the sample's substance in the REB energy deposition region, together with the limitedness of the excited elastic wave in all three spatial dimensions, i.e. due to its three-dimensional nature. The last circumstance, within the framework of completely different model, was proposed to be taken into account also in [2].

Indeed, damage to experimental samples and observations of plasma spread from the irradiated surface [13] illustrate that heating of their REB not only creates an elastic stress in the near-surface layer, but evaporates (and ionizes) it, after which this layer begins to spread indefinitely, being under the influence of the return elastic force present in the heated, but remaining solid material in the model [7,9]. Such spread exerts a long-term pressure compared to the time of the beam exposure on the material layers unaffected by evaporation due to the reactive force, introducing into the condensed medium an already sizeable impulse, which is detected experimentally [14]. In other words, the generation of an elastic perturbation in the target in the framework of (1) in the region $z > 0$ is adequately described by setting the positive pressure pulse at the boundary $z = 0$. In a one-dimensional situation, the resulting acoustic wave would have exclusively non-negative values σ. However, since in reality, as emphasized in the Introduction, the region of the REB energy deposition across the diameter is noticeably smaller than the size of the sample, this wave is completely three-dimensional, and it is well known (see, for example, [15,16]) that in higher dimensionalities, unlike one-dimensionality, within (1) the monopolar wave with $\sigma \geq 0$ cannot exist. This fact is usually explained by the following consideration. The presence of stress in a linear isotropic medium puts it in motion according to Newton's second law $\rho\partial\mathbf{v}/\partial t = -\nabla\sigma$). The velocity of the medium in the acoustic approximation is potential velocity $\mathbf{v} = -\nabla\theta$, so that the linkage $\int \sigma dt \propto \theta$ is obtained. If at the observation point before and after the sound wave passes through it, the medium is at rest, characterized by zero potential, then, therefore, in any localized wave there are necessarily sections with σ of both signs : otherwise the time integral will not be zeroised. One-dimensional wave divides the space into in regions in front of and behind it ones, and the values θ on $\pm\infty$ may differ, but when the wave is localized in all directions, the region of rest outside it, is connected and therefore is characterized by

[1] Of course, the absorbed electrons also have momentum too, but its influence is negligible due to their small mass.

one value of the potential. Thus, if the reactive pressure initially creates the quasi-one-dimensional (because its size in r significantly exceeds the thickness in z, see below) compression wave in the elastic sample, then with its removal from the boundary by distances comparable to its own transverse dimension, three-dimensionality comes into play, resulting in „generating" tension regions.

Note that the effect of reaching the maximum tension of the material in the depth of the target was observed in calculations [4] related to the excitation of shock waves in steel samples. However, the mechanism of this phenomenon was different, the region with $\sigma < 0$ initially putting in appearance at the beam edge and at the target boundary, and the subsequent increase in the perturbation amplitude occurred when the wave converged from the edge to the axis. Most likely, this was anyway due to the destressing during incomplete evaporation of the steel by beam and the fact that in the tension formation region $G \neq 0$ (the description of the calculations is not too detailed). Our mechanism is believed by us to be more universal.

2. Analytical and numerical calculations

Let's pass on to strict calculation. It is required to find solution (1) $\sigma(t, \mathbf{r}, z)$, where $\mathbf{r}$ is a two-dimensional vector in the xy plane, i.e. $\sigma(t, r, \varphi, z) = \sigma(t, r, z)$ due to the above symmetry of the problem, in the region $z > 0$ for given initial conditions $\sigma = 0$, $\partial\sigma/\partial t = 0$ at $t = 0$ and the boundary condition, which, to simplify the formulas, is written as a function with separable variables, $\sigma = g(t)P(r)$ for $z = 0$ conditions. Specific profiles $g(t)$ and $P(r)$ will be given below Due to the linearity of the problem, this solution is expressed by convolution of the boundary condition with some characterizing (1) function $L(t, r, z)$, which is to be determined. Indeed, by applying the Laplace transformations in t and the two-dimensional Fourier transformations for $\mathbf{r}$ and using the fact that $\partial^2/\partial t^2\ top^2$, $\Delta \to -k^2 + \partial^2/\partial z^2$, one can write that

$$\frac{d^2\sigma_{p\mathbf{k}}}{dz^2} = \left(\frac{p^2}{c_S^2} + k^2\right)\sigma_{p\mathbf{k}} \to \sigma_{p\mathbf{k}} = g_p P_{\mathbf{k}} L_{p\mathbf{k}},$$

where

$$L_{p\mathbf{k}} = \exp\left[-z\sqrt{(p/c_S)^2 + k^2}\right].$$

Here the standard addition of subscripts p for the Laplace and $\mathbf{k}$ for the Fourier transforms of the corresponding functions is used (see, for example, [17]). Inverting the Fourier transformation, we obtain

$$L_p = \frac{1}{2\pi}\int_0^\infty \exp\left[-z\sqrt{(p/c_S)^2 + k^2}\right] J_0(kr)kdk$$

$$= \frac{z}{2\pi(r^2+z^2)^{3/2}}\left(1+\frac{p\sqrt{r^2+z^2}}{c_S}\right)\exp\left(-\frac{p\sqrt{r^2+z^2}}{c_S}\right),$$

($J_0(\alpha) = \int_0^\pi \exp(i\alpha\cos)d\varphi/\pi$ — Bessel function) [18]. It follows from this that

$$\sigma(t, r, z) = \int \frac{zP(\mathbf{r} - \mathbf{r}_1)}{2\pi c_S(r_1^2 + z^2)}$$

$$\times \left(\frac{c_S}{\sqrt{r_1^2 + z^2}} + \frac{d}{dt}\right) g\left(t - \sqrt{r_1^2 + z^2}/c_S\right) d^2\mathbf{r}_1.$$

The terms responsible for the monopolar ($\propto g \geq 0$) and sign-alternating ($\propto \dot{g}$) terms of the acoustic wave are clearly visible. Unfortunately, this integral can be taken analytically only in two extreme situations: one-dimensional one with $P = \text{const}$, where $\sigma \propto g(t - z/c_S)$, as it should be (sign-alternating term „is amortized" by interference), and limitingly three-dimensional one, when the reactive pressure is delta-functional one $P \propto \delta(\mathbf{r})$,

$$\sigma \propto \frac{(\mathbf{e}_z \cdot \mathbf{R})}{R^2}\left[\frac{g(t - R/c_S)}{R} + \frac{\dot{g}(t - R/c_S)}{c_S}\right], \qquad (2)$$

where $\mathbf{R} = \{x, y, z\}$ is 3-d radius vector and $\mathbf{e}_z$ is unit vector (ort) of applicate axis. Here, the monopolar term is the so-called „near field" of a point acoustic source at the boundary, and the alternating term is the standard dipole mode wave [15,16].

Nevertheless, the found integral expression is a convenient tool for wave simulation, since the numerical calculation of integrals is much more reliable and steady than solving differential equations. For these purposes, it is convenient to rewrite the convolution in the form

$$\sigma = -\int_0^\infty \left[\int_0^{2\pi} P\left(\sqrt{r^2 + r_1^2 - 2rr_1\cos\varphi_1}\right) d\varphi_1\right]$$

$$\times \frac{z}{2\pi}\frac{\partial}{\partial r_1}\frac{g(t - \sqrt{r_1^2 + z^2}/c_S)}{\sqrt{r_1^2 + z^2}} dr_1. \qquad (3)$$

The calculation requires setting the boundary condition in the form of quite specific function $\sigma(t, r, 0)$ Based on the experimental facts indicated in the Introduction, we chose for it the Gaussian spatial dependence $P = P_0\exp(-r^2/2\vartheta^2)$ with $\vartheta \sim 3.5$ mm. The shape of the temporal pressure pulse $g(t)$ is shown in Fig. 4.

This is the spline of total duration τ, smoothly „joined together" from two cubic polynomials, the rise time of which, according to the physics of the case, is comparable to the duration of the beam exposure on the sample, was taken three times less than the decay time associated with a decrease in pressure in the jet stream due to its irreversible expansion. Change in the ratio of these times had practically no effect on the nature of the process. Due to the linearity of the problem, the pattern observed in the calculation depends only on one dimensionless parameter $c_S\tau/\vartheta$, equal to the ratio of the longitudinal and transverse scales of the excited

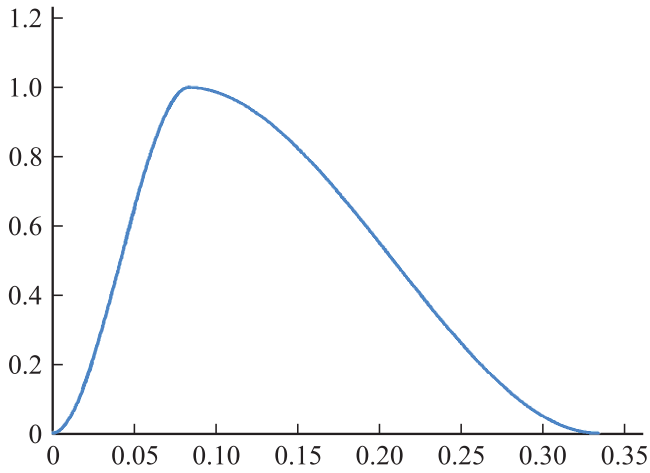

Figure 4. Dependence of the pressure impulse at the boundary on time in ϑ/c_S units for the main calculation variant.

acoustic wave (see above). The calculation was carried out in the dimensionless form with $c_S = P_0 = \vartheta = 1$, so that the only variable parameter was τ. The smaller it is, the closer the generated wave is to one-dimensionality (in physical variables, the decrease τ is equivalent to increase ϑ), and the region of negative values arises in it and grows in size (with simultaneous increase here also absolute values σ) on the rear side of the profile with respect to the direction of its propagation, at the greater distance from the boundary. To demonstrate the declared effect, we chose ($\tau = 1/3$: taking into account the fact that the volumetric velocity of sound in plexiglass is ~ 2.2 km/s [14], this corresponds to $\tau \sim 500$ ns in physical variables, which is in good agreement with the experimentally observed width of the perturbation traveling along the sample [7,12]

When performing calculations, the nondimensionalized formula (3) by replacing the independent $u^2 = r_1^2 + z^2$ and the dependent $P(s) = F(s^2)$ of variables was written in the form

$$\sigma = -\int_0^{\infty}\left[\int_0^{2\pi} F\left(r^2+u^2-z^2-2r\sqrt{u^2-z^2}\cos\varphi_1\right)d\varphi_1\right] \times \frac{z}{2\pi}\frac{\partial}{\partial r_1}\frac{g(t-u(r_1))}{u(r_1)}dr_1$$

$$= \int_z^{\infty}\left[\int_0^{2\pi} F\left(r^2+u^2-z^2-2r\sqrt{u^2-z^2}\cos\varphi_1\right)d\varphi_1\right] \times \frac{z}{2\pi}\frac{g(t-u)+ug'(t-u)}{u^2}du.$$

Since the function g is identically equal to zero outside the segment $[0, \tau]$, the outer integral is calculated in fact within $[\max(z, t-\tau), t]$. In addition, due to the symmetry of the argument of the function F in the inner integral, the calculation was carried out not for the segment $[0, 2\pi]$, but for $[0, \pi]$. Thus, the formula was obtained

$$\sigma = \frac{z}{\pi}\int_{\max(z,t-\tau)}^{t}\left[\int_0^{\pi} F\left(r^2+u^2-z^2 - 2r\sqrt{u^2-z^2}\cos\varphi_1\right)d\varphi_1\right]\frac{g(t-u)+ug'(t-u)}{u^2}du. \quad (4)$$

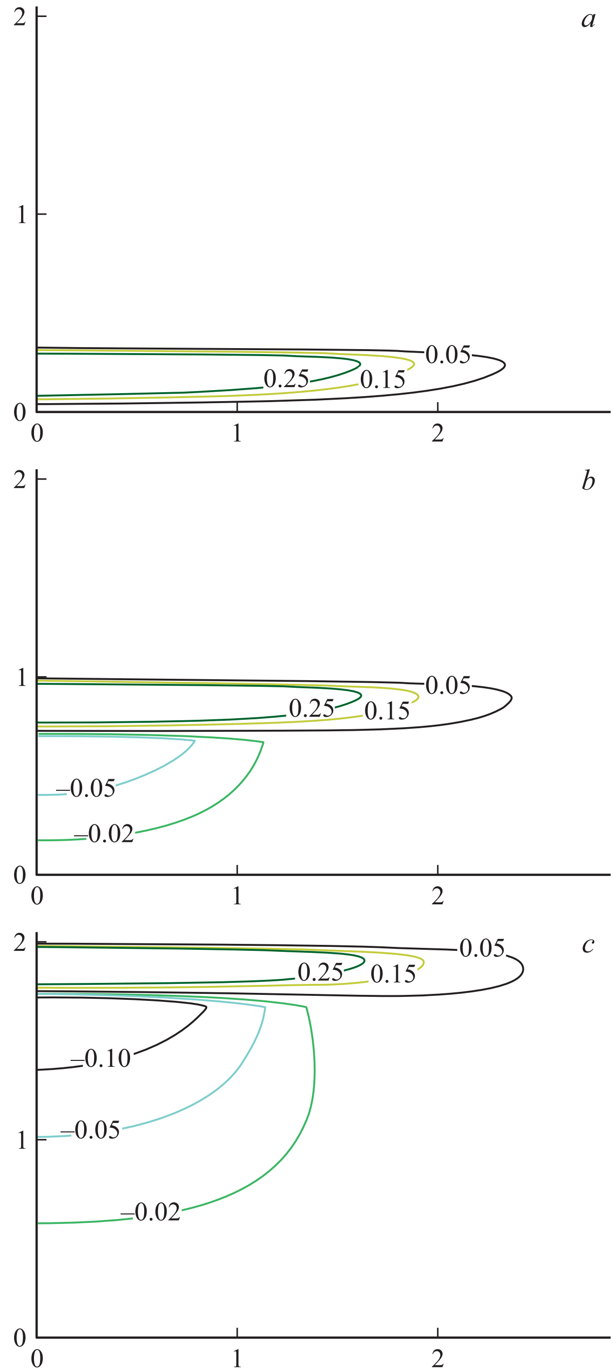

Figure 5. Stress isolines in the elastic wave for different values t: a is 0.34 (i.e. immediately after the end of the pressure pulse), b is 1.0 and c is 2.0. The horizontal axis corresponds to the radial coordinate, the vertical one to the z axis.

By it in the rectangle $\{0 \le r \le 2.8,\ 0 \le z \le 2.1\}$ the matrix of values $\sigma(r_i, z_j)$ was calculated for $r_i = 0.004i$, $0 \le i \le 700$ and $z_j = 0.004j$, $0 \le j \le 525$.

The results of these calculations for three successive moments of time: immediately after the end of the pressure pulse and during subsequent propagation into the depth of the medium are shown in Fig. 5.

The fact that there are points in the pictures for which the wave has not yet reached does not affect the calculation time, since in this case the limits of the outer integral in (4) immediately show that the point should not be calculated. It can be seen that the tension region in the elastic perturbation from the beam genuinely appears at the finite distance from the boundary (at the level of -0.1 from $\sigma_{\max}$ no closer then ϑ). This region, in comparison with the compression region, is characterized by smaller absolute values of σ (which is not surprising for the given perturbation generation mechanism) and smaller transverse dimensions. The latter is also in good agreement with the real pattern of destruction. The attenuation of the wave, which leads to the boundedness of the observed destruction region along z from two sides, is apparently associated with dissipation not taken into account in (1) (after all, this is precisely the shock wave at the leading front), but, possibly, also with wave divergence according to (2).

Conclusion

Thus, we have shown that the mysterious nature of the destruction of PMMA and other non-brittle polymers under the action of an elastic perturbation excited by REB can be explained in the rather simple and clear way. Wherein, it should be noted that PMMA and epoxy resins exhibit similar relaxation features (see, for example, [19]). In more brittle materials, stress relaxation mechanisms can differ significantly [5,6], and cracking is not so dramatically associated with high values of $\sigma < 0$. Due to this, the pictures in Figs 2 and 3 also differ. In principle, the effect „of generation" of the tension region should have been observed in the calculations [20], where the main attention was given „to the full-scale" description of the elastic medium with non-zero values of both K, and G (excluding the taking into account the excess of the yield strength), but the text does not say anything about it. Although the evolution of the wave in this case is more complicated, and the medium velocity in it is no longer potential, but the phenomenon itself, as stated in the Introduction, is at the qualitative level resistant to such perturbations. Perhaps the point is that the authors focused on simulation the destruction of brittle glass that cracks even at $\sigma > 0$, and therefore did not pay attention to the seemingly small negative stress values.

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Conflict of interest

The authors declare that they have no conflict of interest.

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