

THE COEFFICIENT OF REFLECTION OF ULTRASONIC WAVES FROM AN ADHESIVE BOND INTERFACE

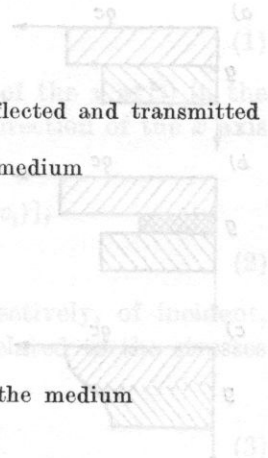
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This paper reports on an attempt to explain the existence of a relation between the reflection coefficient and mechanical strength of an adhesive bond, based on a model of a bond with finite rigidity. On the basis of formulae derived, a correlation relation was determined from experimental results between the modulus of the pressure reflection coefficient of a plane ultrasonic wave incident normal to the interface of an adhesive bond of lucite with epoxy resin, and the tensile strength of the bond.

Notation

- A_p, A_0, B — displacement amplitudes, respectively, of incident, reflected and transmitted waves
- c_i — propagation velocity of ultrasonic waves in the i th medium
- f — frequency of ultrasonic wave
- g — thickness
- K — bond rigidity
- \bar{r}_{21} — complex pressure reflection coefficient
- $(r_{21})_0$ — pressure reflection coefficient for $K = \infty$
- R_0 — tensile strength
- t — time
- u_i — displacement in the direction of the x axis in the i th medium
- x — attenuation over one wavelength
- Z_i — characteristic acoustic impedance in the i th medium
- \bar{Z}_i — complex characteristic acoustic impedance in the i th medium
- a — attenuation coefficient
- σ_i — stress in the i th medium
- σ_g — stress at the interface
- λ_i, μ_i — Lamé constants
- ω — angular frequency
- ρ_i — density of the i th medium



1. Introduction

One of the possible ways of evaluating adhesive strength (degree of adhesion) of layered joints by ultrasonic methods consists in measurement of the pressure coefficient of the reflection of a plane ultrasonic wave incident normal to the bond interface [1]. The results of a number of papers [2-4] indicate the presence of a relation between the reflection coefficient and mechanical strength of a bond developed using different techniques (explosive welding, rolling, gluing etc.). The authors of these papers give correlations for specific bonds obtained from results of strength (destructive) tests and ultrasonic investigations. The present paper reports on an attempt to explain changes in the reflection coefficient accompanying changes in adhesive bond quality, based on a model of a bond with finite rigidity.

2. Complex reflection coefficient

Acoustically, adhesive bonds are characterized by a boundary surface on which a step-like change occurs in characteristic acoustic impedance (Fig. 1a). In practice, this class of bonds also includes, in addition to those with a distinct boundary such as e.g. some bimetals or glue-and-metal bonds, those involving a thin intermediate layer, caused, for example, by diffusion processes,

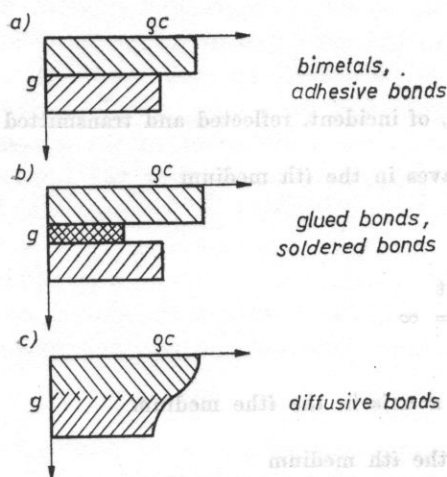


Fig. 1. A division of layered bonds from the acoustical point of view

whose thickness, however, is much less than the length of the ultrasonic wave incident on the boundary. Another group of layered bonds consists of those which involve a layer with thickness comparable to the length of the wave used and with characteristic acoustic impedance distinctly different from that of the materials bound (Fig. 1b). This group includes, for example, glued or

soldered bonds. In investigations of these bonds, a distinction is made between problems related to cohesive strength, i.e. the strength of the binding layer itself, and adhesive strength, which is a measure of the degree of adhesion of elements bound to a binder. The other problem corresponds to the first group of bonds, the so-called adhesive bonds. Finally, a third group consists of diffusive bonds which involve a smooth change in acoustic properties at a thickness comparable to the wavelength.

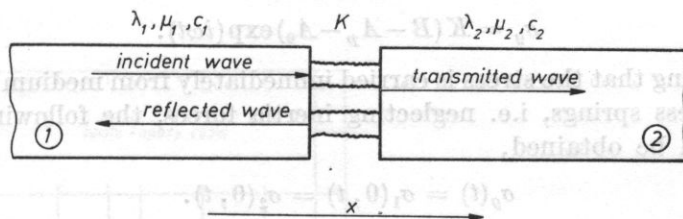


Fig. 2. A model of the interface of an adhesive bond

c_i - density, c_i - ultrasonic wave propagation velocity, λ_i and μ_i - Lamé constants

Fig. 2 shows schematically the model of an adhesive bond interface in which vibrations are carried from medium 1 to medium 2 by weightless springs with equivalent rigidity K [N/m²] per unit area. The assumption of a finite value of the rigidity K permits a modification in boundary conditions, allowing a displacement jump Δu caused by the current stress σ_0 at the interface of the media [9]

$$\sigma_0 = K\Delta u. \quad (1)$$

When the displacements u_1 and u_2 (in the direction of the x axis) in the two media for a plane wave propagating in the positive direction of the x axis are expressed with the formulae

$$\begin{aligned} u_1 &= A_p \exp[i\omega(t-x/c_1)] + A_0 \exp[i\omega(t+x/c_1)], \\ u_2 &= B \exp[i\omega(t-x/c_2)], \end{aligned} \quad (2)$$

where A_p , A_0 and B are displacement amplitudes, respectively, of incident, reflected and transmitted waves; and are subsequently related to the stresses σ_1 and σ_2 by the relations

$$\sigma_i = (\lambda_i + 2\mu_i) \frac{\partial u_i}{\partial x}, \quad i = 1, 2, \quad (3)$$

and considering that

$$Z_i = (\lambda_i + 2\mu_i)/c_i;$$

the displacement jump Δu at the interface ($x = 0$) can be given by

$$\Delta u = u_2(0, t) - u_1(0, t) = (B - A_p - A_0) \exp[i\omega t], \quad (4)$$

while the stresses in media 1 and 2 can be expressed in the following way

$$\begin{aligned}\sigma_1 &= -i\omega Z_1 A_p \exp \left[i\omega \left(t - \frac{x}{c_1} \right) \right] + i\omega Z_1 A_0 \exp \left[i\omega \left(t + \frac{x}{c_1} \right) \right], \\ \sigma_2 &= -i\omega Z_2 B \exp \left[i\omega \left(t - \frac{x}{c_2} \right) \right].\end{aligned}\quad (5)$$

According to formula (1), the stress at the interface is

$$\sigma_g = K(B - A_p - A_0) \exp(i\omega t). \quad (6)$$

Considering that the stress is carried immediately from medium 1 to medium 2 by weightless springs, i.e. neglecting inertia forces, the following boundary condition can be obtained,

$$\sigma_g(t) = \sigma_1(0, t) = \sigma_2(0, t). \quad (7)$$

The satisfaction of this condition leads to a system of equations

$$-i\omega Z_1 A_p + i\omega Z_1 A_0 = -i\omega Z_2 B = K(B - A_p - A_0). \quad (8)$$

The pressure reflection coefficient \bar{r}_{21} can be determined from the system of equations (8). This coefficient is the ratio of the pressure amplitude of the reflected wave to the pressure amplitude of the incident wave,

$$\bar{r}_{21} = \frac{Z_2 - Z_1 - i(\omega Z_1 Z_2 / K)}{Z_2 + Z_1 + i(\omega Z_1 Z_2 / K)}. \quad (9)$$

(1) It is a complex form of the reflection coefficient, neglecting wave attenuation in the two media, which is a result, in contrast to classical solutions, of the assumption of discontinuous displacement at the interface. The introduction of the rigidity K with finite value permits consideration of imperfection of a bond. Similar reasoning led the authors of papers [6-8] to the same relation between the reflection coefficient and the rigidity or compliance of a bond.

Transformation of formula (9) gives a relation between the rigidity K and the modulus of the reflection coefficient, $|\bar{r}_{21}|$,

$$K = \omega \left[A + \frac{B}{|\bar{r}_{21}|^2 - C} \right]^{1/2}, \quad (10)$$

where

$$A = -[Z_1 Z_2 / (Z_1 + Z_2)]^2, \quad B = -A [1 - (r_{21})_0^2],$$

$$C = (r_{21})_0^2 = [(Z_2 - Z_1) / (Z_2 + Z_1)]^2, \quad \omega = 2\pi f, \quad f - \text{frequency}.$$

It can be noted that without adhesion ($K = 0$) the modulus of the reflection coefficient is 1, while for $K = \infty$ (an ideal bond)

$$|\bar{r}_{21}| = (r_{21})_0 = (Z_2 - Z_1) / (Z_2 + Z_1).$$

Fig. 3 gives a frequency relation between the bond rigidity K and the modulus of the reflection coefficient $|\bar{r}_{21}|$ calculated from formula (10) for an adhesive bond of epoxy resin ($\rho = 1.189 \cdot 10^3 \text{ kg/m}^3$, $c_L = 2650 \text{ m/s}$) and lucite ($\rho = 1.182 \cdot 10^3 \text{ kg/m}^3$, $c_L = 2740 \text{ m/s}$). It can be seen that at higher frequency a weakening of the bond with respect to an ideal one causes a greater change in the modulus of the pressure reflection coefficient. Moreover, changes in the reflection coefficient, which correspond to frequency changes, are greater for mean values of bond stiffness than for bonds which are either extremely "good" or "bad".

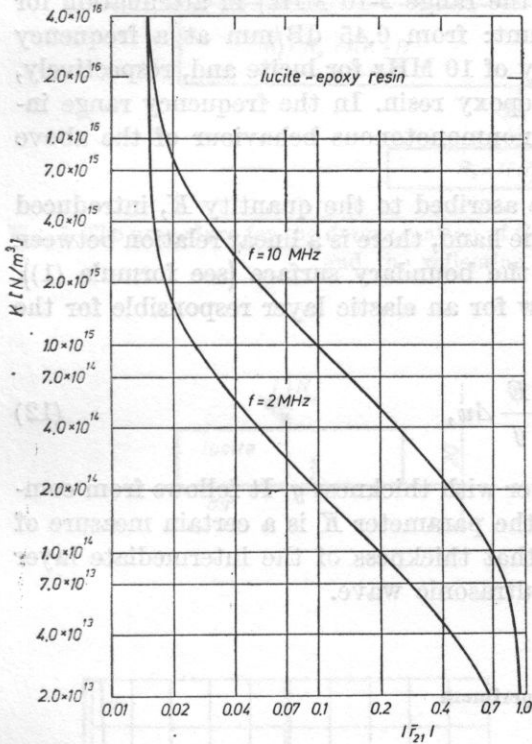


Fig. 3. The frequency relation between bond rigidity and the modulus of the reflection coefficient

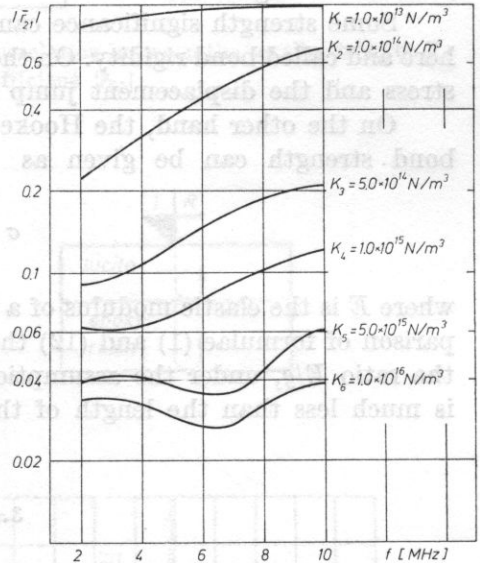


Fig. 4. The frequency dependence of the modulus of the reflection coefficient for different rigidity of a lucite — epoxy resin bond, with consideration of attenuation in materials bound

In terms of the measurement resolution of the coefficient $|\bar{r}_{21}|$ the combination of materials bound is also significant. From this point of view, pairs of materials with close values of characteristic acoustic impedance $(r_{21})_0 \rightarrow 0$ are most convenient. Yet, particularly in such cases, additional changes in the value of the reflection coefficient, caused by attenuation changes in the two materials bound as frequency changes, must be expected. These can be noticed

particularly with materials which differ greatly in the attenuation coefficient (by a few orders). These changes can be evaluated by introducing into formula (9) the complex characteristic wave impedances

$$\bar{Z}_i = Z_i/(1+x_i)^2 - ix_i Z_i/(1+x_i)^2, \quad i = 1, 2; \quad (11)$$

where $x_i = c_i \alpha_i / \omega$ is the attenuation over one wavelength, c_i the propagation velocity of the ultrasonic wave, and α_i is the attenuation coefficient.

Fig. 4 shows results of calculations of the modulus of the reflection coefficient as a function of frequency for some values of the rigidity K of a lucite-epoxy resin bond. Linear changes (in the range 2-10 MHz) in attenuation for both materials were taken into account: from 0.45 dB/mm at a frequency of 2 MHz to 1.0 dB/mm at a frequency of 10 MHz for lucite and, respectively, from 0.88 dB/mm to 2.0 dB/mm for epoxy resin. In the frequency range investigated it is interesting to note a nonmonotonous behaviour of the above relation for higher K .

Some strength significance can be ascribed to the quantity K , introduced here and called bond rigidity. On the one hand, there is a linear relation between stress and the displacement jump at the boundary surface (see formula (1)).

On the other hand, the Hooke law for an elastic layer responsible for the bond strength can be given as

$$\sigma = \frac{E}{g} \Delta u, \quad (12)$$

where E is the elastic modulus of a layer with thickness g . It follows from comparison of formulae (1) and (12) that the parameter K is a certain measure of the ratio E/g , under the assumption that thickness of the intermediate layer is much less than the length of the ultrasonic wave.

3. Experiment

In order to evaluate the usefulness of the model of an adhesive bond with finite rigidity for explanation of the relation between the modulus of the pressure reflection coefficient of an ultrasonic wave incident normal to the interface of two bodies and the strength of their bond, experiments were performed according to the following procedure (Fig. 5):

— Different (mechanical and chemical) preparation of the surface of lucite yielded 29 samples of the adhesive lucite — epoxy resin bond with different degree of adhesion. The shape of the samples is shown in Fig. 7.

— Ultrasonic investigations were performed to determine the modulus of the pressure reflection coefficient of a wave at a frequency of 10 MHz, according to the procedure shown in Fig. 6.

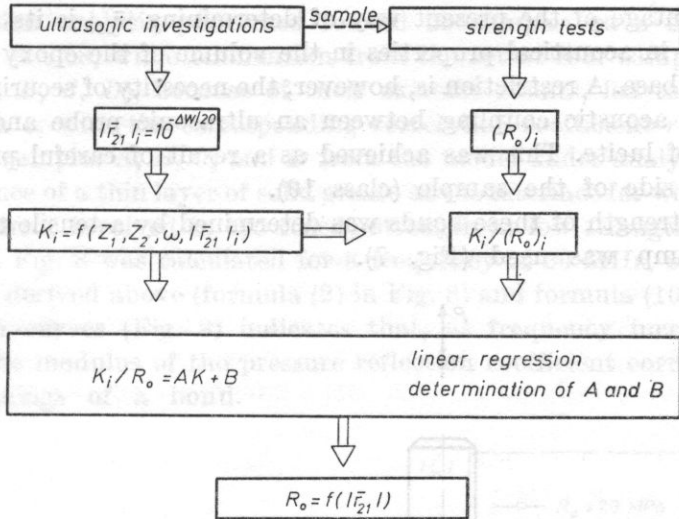


Fig. 5. The procedure for the determination of the correlation between the tensile strength R_0 and the reflection coefficient $|\bar{r}_{21}|$.

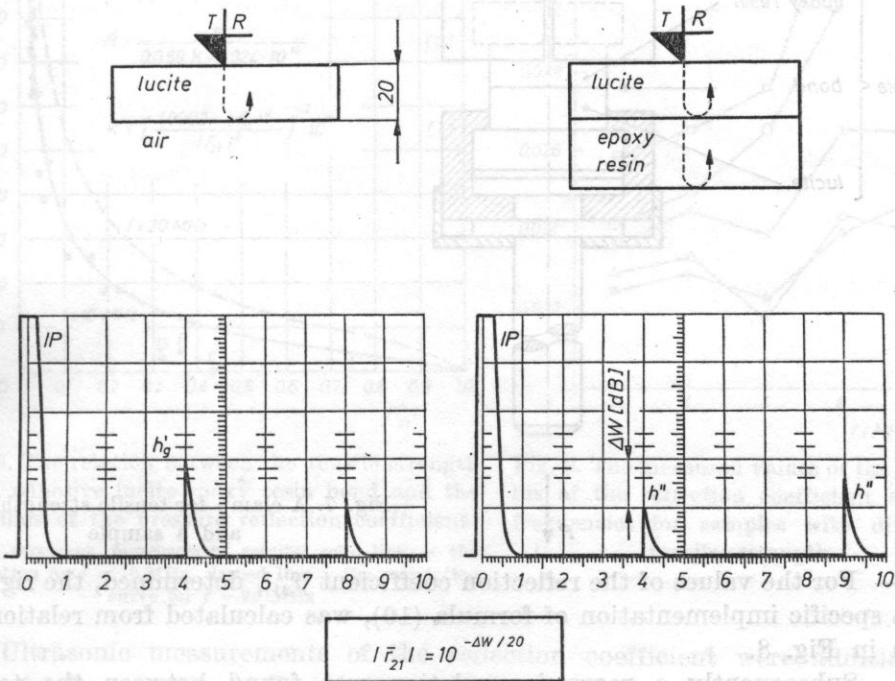


Fig. 6. The procedure for the determination of the modulus of the pressure reflection coefficient of ultrasonic waves $|\bar{r}_{21}|$.

An advantage of the present way of determining $|\bar{r}_{21}|$ is its independence from changes in acoustical properties in the volume of the epoxy resin poured on to a lucite base. A restriction is, however, the necessity of securing repeatable conditions of acoustic coupling between an ultrasonic probe and the sample on the side of lucite. This was achieved as a result of careful preparation of the external side of the sample (class 10).

— The strength of these bonds was determined by a tensile test, in which a special clamp was used (Fig. 7).

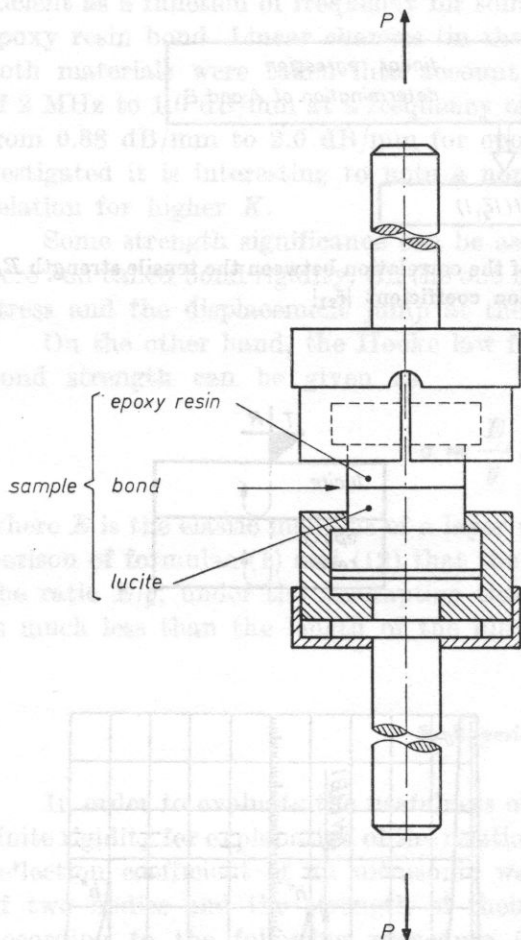


Fig. 7. A clamp for tensile strength tests and a sample

— For the values of the reflection coefficient $|\bar{r}_{21}|_i$ determined, the rigidity K_i , a specific implementation of formula (10), was calculated from relation (1) given in Fig. 8.

— Subsequently a regression relation was found between the tensile strength R_0 and the bond rigidity K , under the assumption of a homographic functional relation between R_0 and K .

Correlation analysis performed for all 29 samples gave the correlation coefficient $r = 0.68$, while elimination from the set of four samples with least strength (*A, B, C, D*), because of their extreme results, led to the equation given in Fig. 8, with the corresponding correlation coefficient $r = 0.998$. The deviation of samples *A, B, C*, and *D* from the model under analysis is justified by the presence of a thin layer of solid grease at the interface for weakest samples and by the error involved in the tensile strength at low strength. The dashed line curve in Fig. 8 was calculated for a frequency of 20 MHz, on the basis of a correlation derived above (formula (2) in Fig. 8) and formula (10). Comparison of these two curves (Fig. 8) indicates that, as frequency increases, greater changes of the modulus of the pressure reflection coefficient correspond to the same weakenings of a bond.

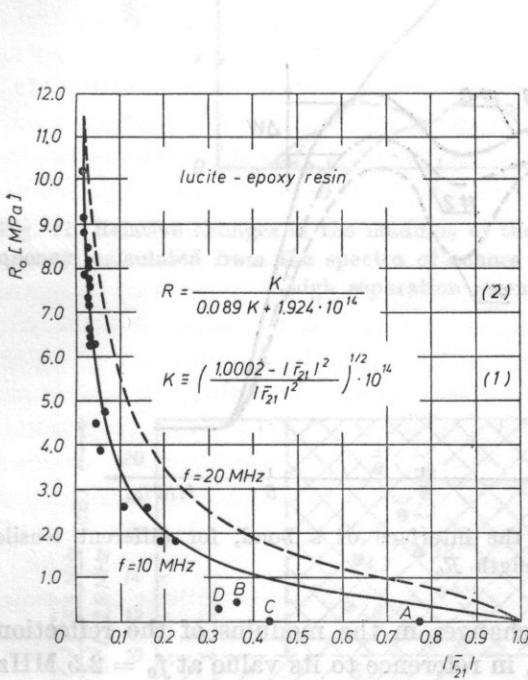


Fig. 8. The relation between the tensile strength of an adhesive lucite-epoxy resin bond and the modulus of the pressure reflection coefficient points represent measurement results; solid line — the correlation for $f = 10$ MHz, dashed line — the calculation curve for $f = 20$ MHz

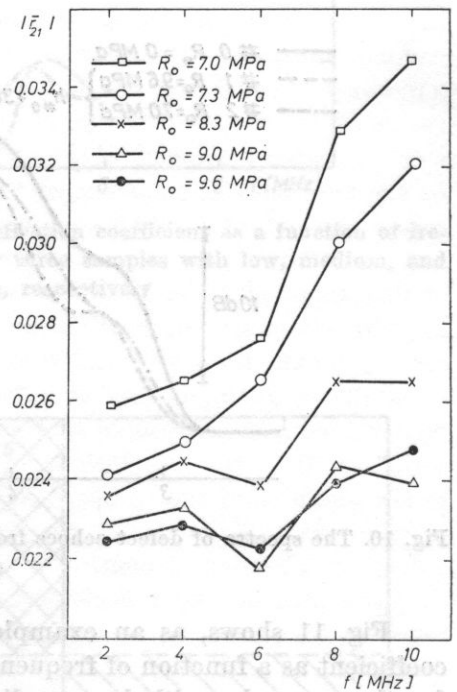


Fig. 9. The measured values of the modulus of the reflection coefficient at five frequencies for samples with different tensile strength

Ultrasonic measurements of the reflection coefficient were additionally taken for five samples at five frequencies, i.e. 2, 4, 6, 8 and 10 MHz. The results of the measurements are given in Fig. 9. Its similarity with Fig. 4 is noteworthy. These results also confirm the thesis that measurement sensitivity increases

as frequency increases. It is also possible to see the nonmonotonous character of changes in the reflection coefficient, depending on frequency for "good" bonds, with corresponding larger K .

In order to investigate the frequency dependence more precisely, on ultrasonic spectral analysis was used. At the Nondestructive Testing Laboratory of the Institute of Machinery Construction (SVUSS) in Prague (Czechoslovakia), measurements were taken on 20 samples of a lucite - epoxy resin bond using an analogue spectral analyzer manufactures by Hewlett Packard. Fig. 10 shows some examples of echoes from interface for two samples with different tensile strength against the background of the spectrum for a "zero" sample, i.e. without connection. On the basis of the spectra obtained, the moduli of the reflection coefficient were calculated at 0.25 MHz intervals over the frequency range 2.5-6 MHz.

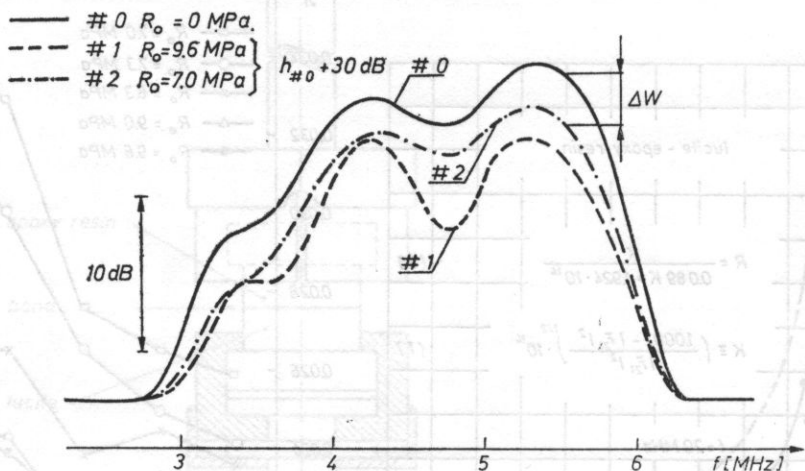


Fig. 10. The spectra of defect echoes from the interface of a bond, for different tensile strength R_0

Fig. 11 shows, as an example, changes in the modulus of the reflection coefficient as a function of frequency, in reference to its value at $f_0 = 2.5$ MHz for three samples with low, medium and high tensile strength, respectively.

Fig. 12, in turn, shows collectively results obtained for all samples in the system: the horizontal axis - the values of the moduli of the reflection coefficient at a frequency of 5.5 MHz, the vertical axis - the difference, in dB, between the values of the reflection coefficient at frequencies of 5.5 MHz, and 2.5 MHz. Respective strength determined in a tensile test was assigned to each point on the curve. Accordingly, three groups of results can be distinguished. The first group consists of high-strength bond samples, with corresponding low values of the moduli of the reflection coefficient and small changes as a function of frequency. The second group is composed of medium-strength

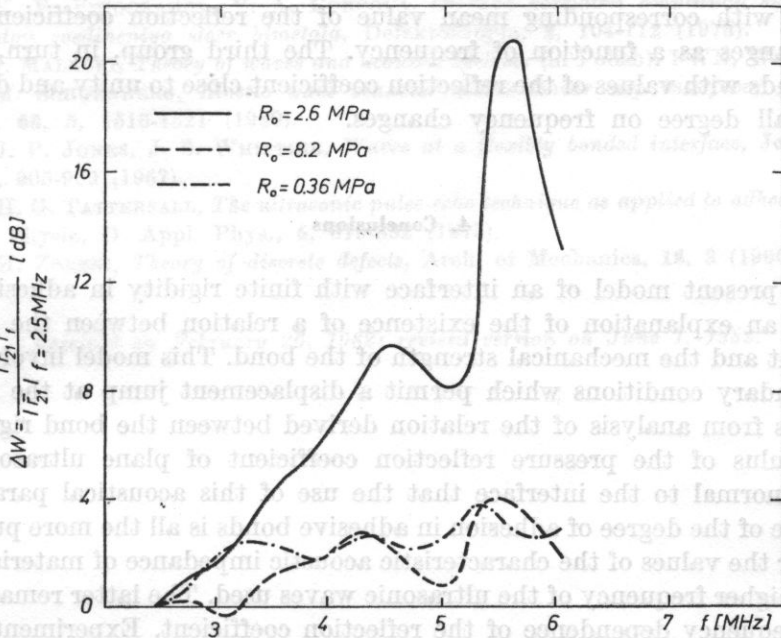


Fig. 11. Relative changes in the modulus of the reflection coefficient as a function of frequency, calculated from the spectra of echoes for three samples with low, medium, and high separation strength, respectively

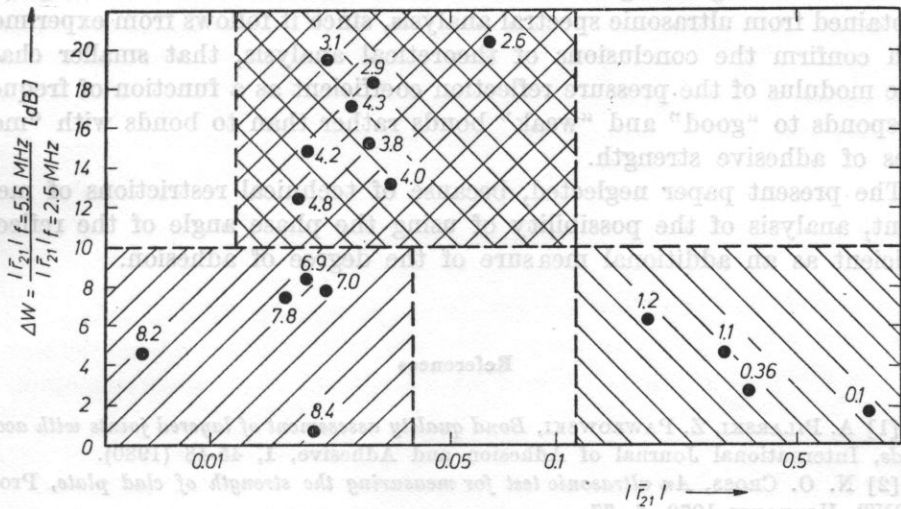


Fig. 12. Moduli of the reflection coefficient $|\bar{r}_{21}|$ at the frequency $f = 5.5 \text{ MHz}$ and the ratio of the moduli $|\bar{r}_{21}|$ at $f = 5.5 \text{ MHz}$ and 2.5 MHz . The figure also shows numerical values of bond tensile strength

samples, with corresponding mean value of the reflection coefficient but its large changes as a function of frequency. The third group, in turn, contains weak bonds with values of the reflection coefficient close to unity and dependent to a small degree on frequency changes.

4. Conclusions

The present model of an interface with finite rigidity in adhesive bonds provides an explanation of the existence of a relation between the reflection coefficient and the mechanical strength of the bond. This model involves modified boundary conditions which permit a displacement jump at the interface. It follows from analysis of the relation derived between the bond rigidity and the modulus of the pressure reflection coefficient of plane ultrasonic wave incident normal to the interface that the use of this acoustical parameter as a measure of the degree of adhesion in adhesive bonds is all the more purposeful, the closer the values of the characteristic acoustic impedance of materials bound and the higher frequency of the ultrasonic waves used. The latter remark relates to the frequency dependence of the reflection coefficient. Experimental verification showed the usefulness of relation (10) derived here for the search for a relationship between the rigidity of a bond and its strength. The advantages of this approach include the possibility of diminishing the number of standard samples necessary for correlation analysis, by the performance of ultrasonic measurements for some different frequencies. Additional information, which facilitates a distinguishing of adhesive bonds in terms of their strength, can be obtained from ultrasonic spectral analysis, since it follows from experiments, which confirm the conclusions of theoretical analysis, that smaller changes in the modulus of the pressure reflection coefficient as a function of frequency corresponds to "good" and "weak" bonds rather than to bonds with "mean" values of adhesive strength.

The present paper neglected, because of technical restrictions of measurement, analysis of the possibility of using the phase angle of the reflection coefficient as an additional measure of the degree of adhesion.

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Measurements of the absorption coefficient of ultrasonic waves in the frequency range 10-100 MHz and of the propagation velocity of ultrasonic and hypersonic waves in aqueous solutions of hexamethylphosphortriamide (HMPT) were carried out. In addition the density of the solutions and the coefficient of shear viscosity was measured. On the basis of the quantities measured, the coefficient of bulk viscosity, relaxation parameters of the process observed and, on the basis of the theory of compressibility relaxation, change in free energy and volume between two structural states were measured.

The analysis of the results of measurements of the absorption coefficient of ultrasonic waves, depending on the frequency, temperature, and composition of solutions, shows the presence in aqueous solutions of HMPT of a relaxation process related to the formation and disintegration of stable structures with the composition HMPT-17 H₂O. On the basis of compressibility relaxation theory, it was shown that the process of structural relaxation is related to a change in volume ΔV and in free energy ΔF between two structural states.

1. Introduction

The character of changes in the absorption coefficient of ultrasonic waves depending on the concentration of a solution varies greatly for a great number of liquid mixtures [1, 6, 11, 10, 3]. An interesting group is made of aqueous solutions of non-electrolytes for which the absorption coefficient of ultrasonic waves reaches a maximum for a strictly defined composition [2, 5, 11-14]. The occurring maximum of the absorption coefficient of ultrasound indicates the presence in the solution of a relaxation process related to the formation and disintegration of strictly defined structures (molecular complexes) between water molecules and the molecules of the medium dissolved.