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APPLICATION OF ULTRASONIC PHYSICS IN MATERIAL TECHNOLOGY*

IGNACY MALECKI

Institute of Fundamental Technological Research (Warszawa)

Measurements of the velocity and attenuation of ultrasonic wave as the basis of the inspection of technological processes. Ultrasonic defectoscopy as used for the detection of internal defects and measurement errors. Correlation between indications of a defectoscope and the mechanical strength of the construction. Determination of elasticity constants of materials. The effect of material grain size upon the attenuation of ultrasonic waves. Stress action vs. increased attenuation. Continuous inspection of the processes: burning of ceramics, polimerization, metal solidification. The effect of microstructure upon the propagation of ultrasonic waves. Properties of materials at low temperatures.

1. Physical foundations of ultrasonic measurement

The main advantage of ultrasonic methods consists in the fact that by means of relatively simple measurements of a few acoustic quantities we can obtain comprehensive information on the structure and properties of materials.

In principle we are dealing with two measurements:

1. measurement of the time-lag Δt between two signals,
2. measurements of difference Δi in the intensity levels of these signals (in decibels).

When a continuous monochromatic signal is used for making the measurements, the equivalent of measuring the time-lag is the measurement of phase differences; a gradual decrease in the amplitude of the signal (in time or in space) corresponds to a difference in intensity. These measurements are made, as a rule, for a certain frequency band and the characteristics $\Delta t(f)$ and $\Delta i(f)$ are thus obtained.

These two basic measurements permit the determination of a number of the acoustic properties of materials, or of the parameters of the object under investigation.

(a) If the distance travelled by a wave between the point from which it is sent and the point at which it is received is Δl, the measurement Δt determines the velocity of propagation of the wave in a given medium:

$$c = \frac{\Delta l}{\Delta t}. \tag{1.1}$$

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(b) If the velocity of propagation of the wave is known, the measurement of the time-lag determines the distance, e.g. the distance between the surface of a given object and its internal flaw:

$$\Delta l = c\Delta t. \tag{1.2}$$

(c) The difference Δi in the intensity levels between two points divided by the distance Δl determines the attenuation coefficient α of a given material:

$$\alpha = 0.1151\frac{\Delta i}{\Delta l}\left[\frac{\mathrm{dB}}{\mathrm{cm}}\right]. \tag{1.3}$$

(d) The differences in the intensity level of waves reflected from the internal heterogenity of a given material is a function of the magnitude and kind of heterogenity.

(e) The maximum of the characteristic $\alpha(f)$ determines the time constant of the acoustic relaxation of the material:

$$\tau = \frac{1}{2\pi f}. \tag{1.4}$$

(f) The pattern of the functions $c(f)$ or $\alpha(f)$ enables us to draw conclusions concerning the mechanism of molecular and atomic losses and of the dynamic quantities of the elasticity constants.

Quantities Δt and Δi may change over time in the technological process, e.g. in the polymerization of plastics. Observation of these changes provides information on the course of this process.

The most interesting measurements from the point of view of learning about the structure of the material are those of Δl and Δi as functions of temperature, especially at low temperatures close to the superconductivity state.

These dependencies concern the kind of material which can be treated as a continuous medium. At very high frequencies (hypersound), when the length of the acoustic wave becomes commensurable with the free path of the phonon, i.e. with the distance between the nodes of the crystal lattice, a classical approach does not suffice. It is necessary then to resort to the presentation of the acoustic wave as a stream of phonons, i.e. to take into consideration the quantum nature of the phenomena which occur. The methods of measurement change correspondingly; for instance the tunnel effect is used for generation, and opto-acoustic methods are frequently used in reception.

Ultrasonic methods are, in many cases, used in parallel with other methods (e.g. X-rays) for determining the same properties of the materials.

The following considerations are decisive in the choice of the ultrasonic method:

1. the properties of the material – the majority of constructional materials are «transparent» for ultrasound, while electromagnetic waves and streams of elementary particles are reflected almost completely;

2. the ultrasonic equipment is relatively inexpensive and simple in use;
3. the passage of an ultrasonic wave does not, in most cases, produce additional effects similar to those which accompany, for example, the propagation of X-rays.

We shall now review the applications of ultrasonic methods in material engineering, with special emphasis on those problems in which close cooperation between the technologist and the physicist is needed.

2. Detection of internal flaws in materials and geometric measurements

The detection of flaws in materials is the oldest technological application of ultrasound. The first ultrasound flaw detectors (FIRESTONE, SOKOLOV) date back to the 1930's. Even at the first International Conference of Non--Destructive Testing (Brussels 1955) [1] it was shown that ultrasonic methods provide an important tool for technological control in industry. Since that time the importance of these methods has been increasing steadily; it is estimated that at present about 30% of industrial control equipment based on non-destructive methods use ultrasonic techniques.

The most common equipment operates by measuring the delay time and the intensity of an impulse reflected from a flaw. A modern flaw detector has extensive electronics, providing a high degree of automation of measurement. In addition to universal devices a number of specialized devices have been built, e.g. a device serving for detecting internal flaws in rails. There is a whole group of instruments for measuring (usually by resonance methods) the thickness of walls accessible from one side only, as for example in high pressure containers. Actually an ultrasonic control system for testing motor vehicle tyres is progressing; by means of probe-heads switching the high precision and high speed is achieved in testing.

In spite of tremendous progress in flaw detector design, the process of improving them has not yet been completed. This applies both to electronic and to acoustical parts of equipment. Figure 1a shows a typical transmission system and Fig. 1b – a receiving system for equipment measuring the speed and attenuation of ultrasonic waves. From the research point of view the most interesting problem is that of establishing a correlation between flaw signal in the equipment and the mechanical weakening of the material by a flaw. As we have already mentioned, the method used most frequently compares results of the tested object with those from a sample with a standard flaw. There are, of course, also more general studies of a scientific nature [2, 3]. The problem here is to solve complex diffraction problems. Let us recall that the theory of ultrasonic wave reflection from an obstacle in a solid was solved only 20 years ago and only for the simplest case [4]. In periodicals dealing with acoustics quite a number of articles dealing with this problem may still be encountered. The complication is that both the transducer and the obstacle generate waves of different types, causing complex secondary reflections [5].

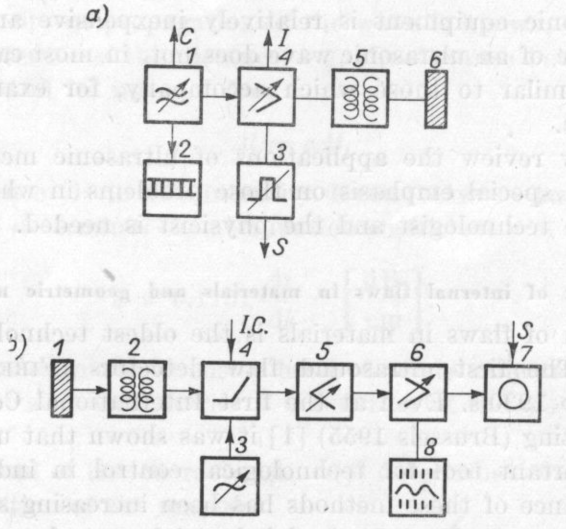

Fig. 1. **The electronic system of the equipment for measuring velocity and attenuation**
a) Transmitting part. *1* – **oscillator,** *2* – **frequency meter,** *3* – **pulse generator,** *4* – **amplifier,** *5* – **matching circuit,** *6* – **transmission head.**
b) Receiving part. *1* – **receiving head,** *2* – **matching circuit,** *3* – **oscillator,** *4* – **frequency converter,** *5* – **attenuator,** *6* – **amplifier,** *7* – **oscilloscope,** *8* – **information processing.**
C – **continuous wave,** *I* – **pulse,** *S* – **synchronizing pulse**

The second part of the problem is an evaluation of the extent to which a single flaw or a set of flaws detected by a flaw detector results in a lowering of the mechanical strength of an object. Extensive experimental work is being carried on in order to determine this relationship for various types of flaws [6].

On this basis standards have been elaborated [7]; in testing welded joints, for example, five classes of flaws have been established and the quality of the joint is determined according to them. This work is conducted by the International Institute of Welding [8]. The best practical results are given by the comparative method, using as a standard an element with artificial flaws [9] (Fig. 2).

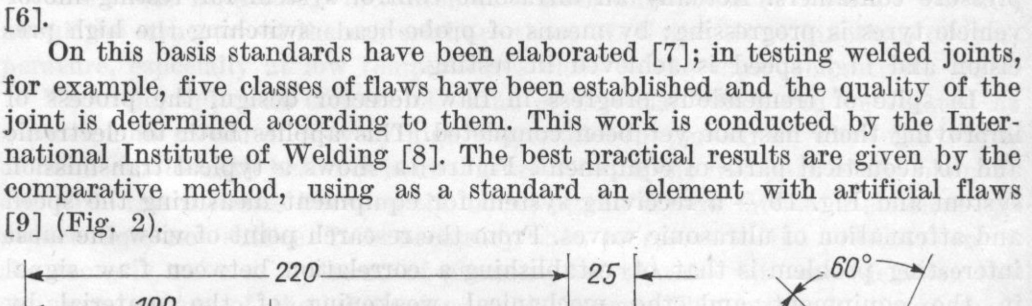

Fig. 2. Standard element for surface waves

3. Determination of the elastic constants

In a homogeneous medium there is a simple relationship between the velocity of acoustic wave propagation and the elastic constants. For the VOIGT model we have

$$\frac{1}{2}\,\varrho c_T^2=\frac{\left[\dfrac{E}{2(1+\nu)}\right]^2+\omega^2\eta^2}{\dfrac{E}{2(1+\nu)}+\sqrt{\left[\dfrac{E}{2(1+\nu)}\right]^2+\omega^2\eta^2}}, \tag{3.1}$$

$$\frac{1}{2}\,\varrho c_L^2=\frac{\left[\dfrac{(1-\nu)E}{(1+\nu)(1-2\nu)}\right]^2+\omega^2\eta^2}{\dfrac{(1-\nu)E}{(1+\nu)(1-2\nu)}+\sqrt{\left[\dfrac{E(1-\nu)}{(1+\nu)(1-2\nu)}\right]^2+\omega^2\eta^2}}, \tag{3.2}$$

where ϱ is the density of the material, c_T, c_L – the velocities of tranverse and longitudinal waves, E, η – elasticity and viscosity elements in the Voigt model, ν – Poisson's ratio.

On the basis of the measurement of the dynamic elastic coefficient, the specific heat at constant volume is determined. It turns out that the materials of the concrete type, i.e. materials [10] with distinct heterogeneity in the macrostructure, display a distinct velocity dispersion (Fig. 3). It should be surmised that the elastic modulus E increases (Fig. 4) and the viscosity decreases (Fig. 5) with frequency.

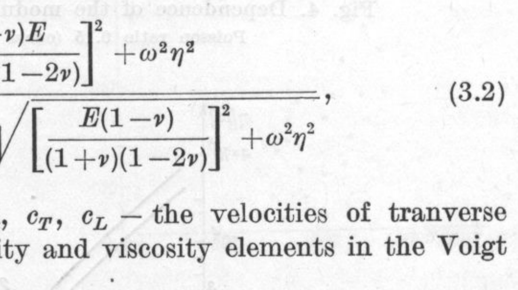

Fig. 3. Wave dispersion in concrete (slab 40× 40× 5 cm) (Drescher)
L – longitudinal wave, T – transversal wave

In recent years composites have been more and more frequently used; they are, as we know, ceramics, metallic materials or polymers strengthened with fibres or whiskers. It has turned out that ultrasonic methods are very useful for testing the elastic properties of these new materials. Generally speaking, the elasticity modulus E_k of the composite is less than the weighted sum of the moduli of the components:

$$E_k < E_a V_a + E_b V_b \tag{3.3}$$

(V_a, V_b are volume proportions).

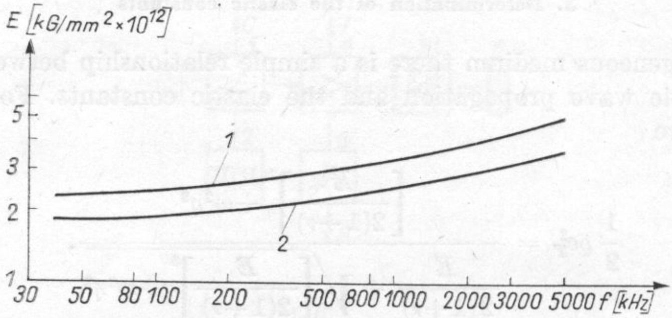

Fig. 4. Dependence of the modulus of elasticity E on frequency

Poisson ratio 0.15 (curve *1*) and 0.30 (curve *2*)

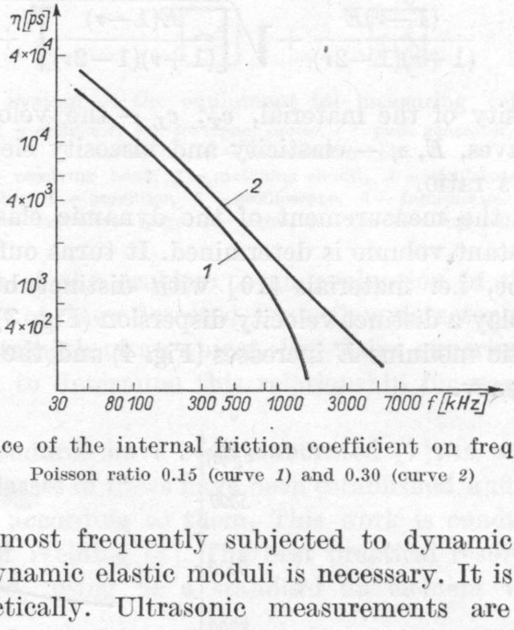

Fig. 5. Dependence of the internal friction coefficient on frequency (Drescher)

Poisson ratio 0.15 (curve *1*) and 0.30 (curve *2*)

Composites are most frequently subjected to dynamic stresses and thus knowledge of the dynamic elastic moduli is necessary. It is difficult, however, to calculate theoretically. Ultrasonic measurements are indispensable for this purpose. The ultrasonic method is especially useful for measuring the elastic constants of brittle materials such as ceramic materials, and semi-brittle ones such as graphite, where other methods produce no results. Resonance methods at frequencies up to 100 kHz, and pulse methods at frequencies between 1 and 100 MHz, are used for these measurements.

4. Testing the mechanical strength

The magnitude of the elastic constants is related directly to the strength of the materials. Research has shown that for some groups of brittle materials (concrete, cast iron) there is a relationship between the velocity of propagation of the ultrasonic waves and the mechanical strength R_c (Fig. 6). This makes possible an extensive application of ultrasonic methods for controlling building constructions, especially bridge constructions. Special flaw detectors are used for measuring the strength of cast iron using scales on which both the velocity of the ultrasonic waves and the strength of the material can be immediately read [12] (Fig. 7). The velocity measurement must be very precise (the error must not exceed 0.2 %) and it is, therefore, necessary to continue improving the electronic systems and to use a quartz frequency stabilizer. Scientific studies are concerned primarily with the mechanism of wave propagation in media with stochastic heterogeneities, such as concrete and polycrystals. This mechanism depends upon many parameters, such as the thermal conductivity at the boundaries of the heterogeneities, their elastic anisotropy, and the ratio of their dimensions to the wavelength. A fairly good approximation is obtained by the formula for the attenuation coefficient [13],

$$\alpha = 4\left(\frac{\overline{\Delta c}}{c}\right)^2 k^4 D^3 \quad \text{for } kD \ll 1, \qquad (4.1)$$

$$\alpha = \frac{\sqrt{\pi}}{2}\left(\frac{\overline{\Delta c}}{c}\right)^2 k^2 D \quad \text{for } kD \gg 1, \qquad (4.2)$$

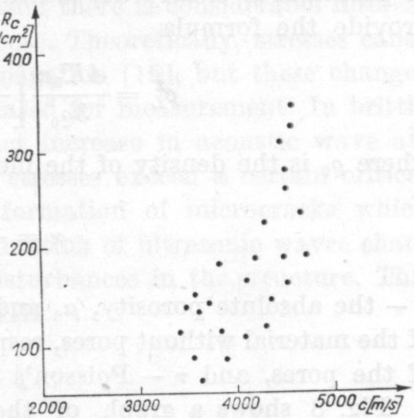

Fig. 6. Dependence of mechanical strength on wave velocity for concrete (experimental data) (Pawłowski)

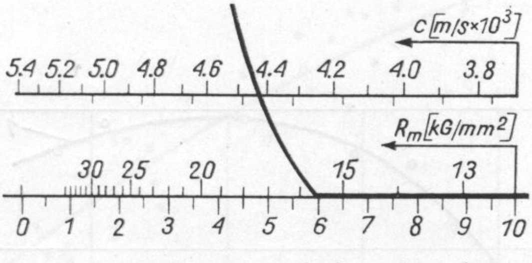

Fig. 7. The meter scale for ultrasonic wave velocity adapted to permit direct reading of the strength of pig iron

where D is average diameter of a heterogenity, k is the wave number and $\Delta c/c$ — relative change of the wave velocity due to the inhomogeneities in the medium.

The propagation of ultrasonic waves may then provide some idea of the nature of heterogeneity.

Experience shows that the velocity of wave propagation in such materials as porcelain is clearly dependent upon porosity. Theoretical calculations [14] provide the formula

$$c_L^2 = \frac{4K_0\mu}{3\varrho_0}\left[\frac{3}{4\mu+3\varepsilon(1+\varkappa)\sigma^2}+\frac{1-\varkappa\varepsilon}{1-\varepsilon}\right], \tag{4.3}$$

where ϱ_0 is the density of the medium without pores,

$$\varkappa = \frac{\mu_0}{K_0} - \frac{3}{2}\,\frac{(1-\nu)}{(1+\nu)}, \tag{4.4}$$

ε — the absolute porosity, μ_0 and K_0 — the shear modulus and bulk modulus of the material without pores, respectively, σ^2 — the standard volume deviation of the pores, and ν — Poisson's ratio.

Fig. 8 shows a graph of the relationship $c_L(\varepsilon)$ versus temperature of burning. Measurements confirm theoretical expectations.

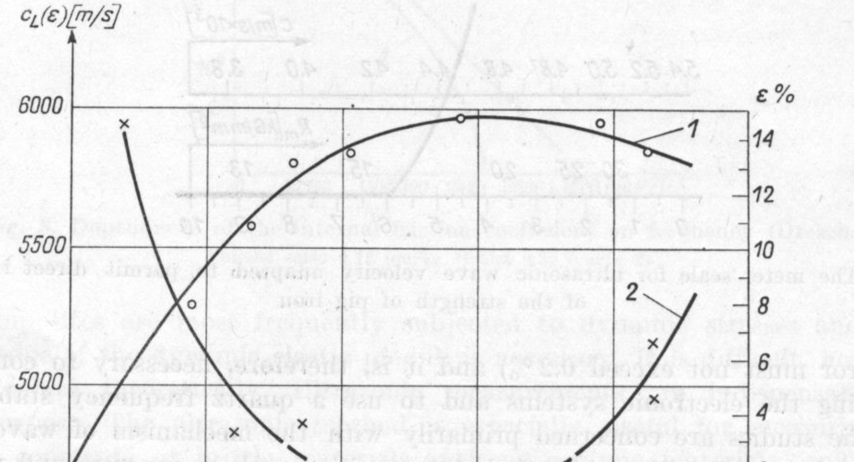

Fig. 8. Relation between wave velocity c (curve *1*) and porosity ε (curve *2*) for technical porcelain (Ranachowski)

The mechanical strength of porcelain depends, of course, upon its porosity. The ultrasonic method is the only one which can be used for testing the mechanical strength of large insulators in which pores are sometimes not uniformly distributed and which thus may be subject to the danger of local cracks.

5. Testing the stress of materials

The knowledge of the spatial distribution of stresses in mechanical elements is of great importance for the construction and operation of machines. Tensometric methods may only be used for the testing of surface stresses, while elastooptic methods require the construction of models not fully commensurable with the original machine. For this reason there is considerable interest in the use of ultrasonic methods for this purpose. Theoretically, stresses cause changes in the velocity of acoustic wave propagation [15], but these changes are so small that in practice they cannot be used for measurement. In brittle and semi-brittle materials, however, a distinct increase in acoustic wave attenuation occurs in areas where the internal stresses exceed a certain critical value. This phenomenon is caused by the formation of microcracks which disperse the wave. Fig. 9 shows how the attenuation of ultrasonic waves changes as a function of the percentage of the disturbances in the structure. This applies to testing frequencies from 0 to 30 MHz.

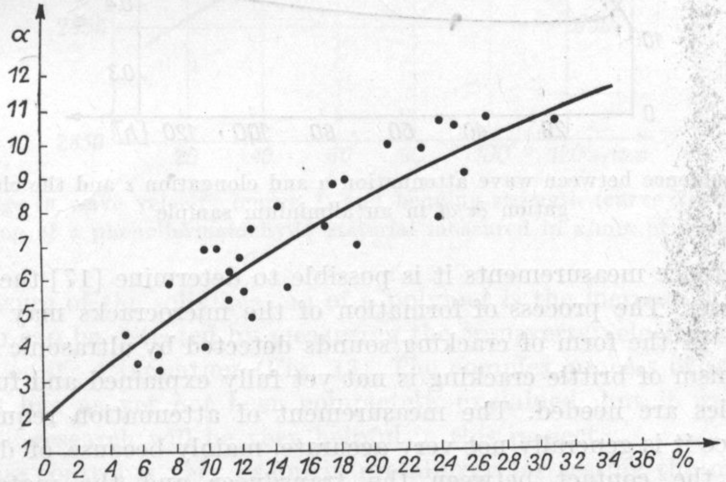

Fig. 9. Wave attenuation as a function of the percentage of area of the disturbed structure of a ceramic insulator

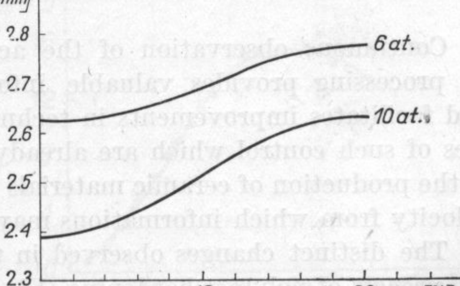

Fig. 10. Attenuation α of the wave in polypropylene in the elongation function ε, with different injection pressures

Changes in the attenuation of a 2.5 MHz ultrasonic wave can be clearly observed (Fig. 10) during the stretching of a polymer (polypropylene) sample. This is caused by reorientation of the macromolecules due to stretching. The creep effect in metals also produces changes in attenuation (Fig 11) and thus opens the possibility of determining the limits of elasticity and plasticity [16].

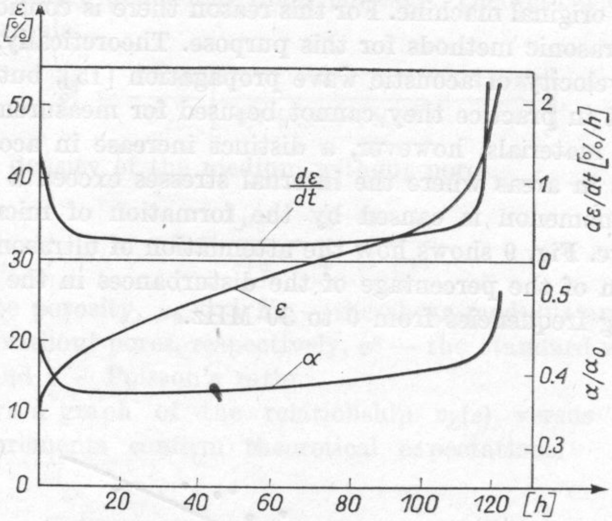

Fig. 11. Dependence between wave attenuation α and elongation ε and the change in elongation $\partial\varepsilon/\partial t$ in an aliminium sample

By difference measurements it is possible to determine [17] the anisotropy of the stresses. The process of formation of the microcracks may be monitored directly in the form of cracking sounds detected by ultrasonic sensor [18]. The mechanism of brittle cracking is not yet fully explained and further theoretical studies are needed. The measurement of attenuation requires improvement, since it is generally not very accurate, mainly because of disturbances caused by the contact between the transducer and the material. Pulse methods or resonance ones are used depending upon the parameters.

6. Control of technological processes

Continuous observation of the acoustic parameters of a material during its processing provides valuable information on the course of this process and facilitates improvements in technical control. We shall give several examples of such control which are already used in practice. The reheating process in the production of ceramic materials is controlled by measurement of the wave velocity from which informations may be obtained concerning pores formation.

The distinct changes observed in the velocity of wave propagation during the process of polymer hardening (Fig. 12) may be used as a test of the pattern of the process. The changes in wave velocity are caused by the reorientation θ of polymer molecules in accordance with the relationship

$$\theta = \frac{1-(c_n/c)^2}{1-(c_n/c_0)^2}, \tag{6.1}$$

where c_n is the wave velocity in a non-orientated amorphous polymer, c_0 — the wave velocity in an ideally orientated polymer, and c — the actual wave velocity.

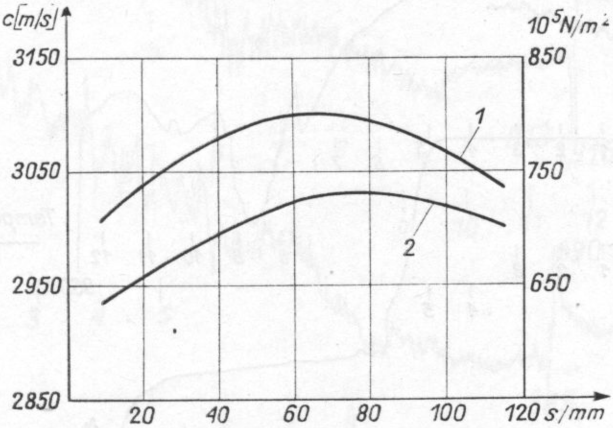

Fig. 12. Change in wave velocity (curve *1*) and bending strength (curve *2*) in the process of hardening of a phenolformaldehyde material measured in s/mm of pressing time

The measure of the solidification of a polymer is the increase in shear elasticity, which can be detected by measuring the transverse velocity of the ultrasonic wave or its attenuation (Fig. 13). The complex process of solidification of polymers has as yet not been completely explained, but it would appear that acoustic research will be very helpful in this respect.

Ultrasonic control is very useful in metallurgy for testing the melting process and phase changes. Both occur at virtually constant temperature, but

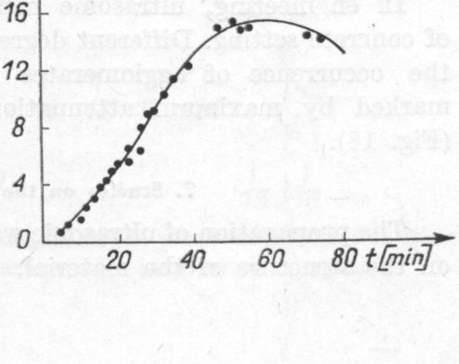

Fig. 13. Attenuation as a function of polymerization time for a polyester ($f = 3$ MHz) (Kunnert)

the ultrasonic attenuation is considerably increased during melting (Fig. 14). In phase transformations the temperature hysteresis takes place and sudden changes in attenuation can be observed, as in Fig. 15, which pertains to pure iron. In Fig. 16 we have an analogous graph for low carbon (0.1 %) Armco steel. Probably, because of the influence of the impurities, the transformation in this material takes place gradually with declining temperature. Another example is the influence of temperature on wave attenuation (Fig. 17).

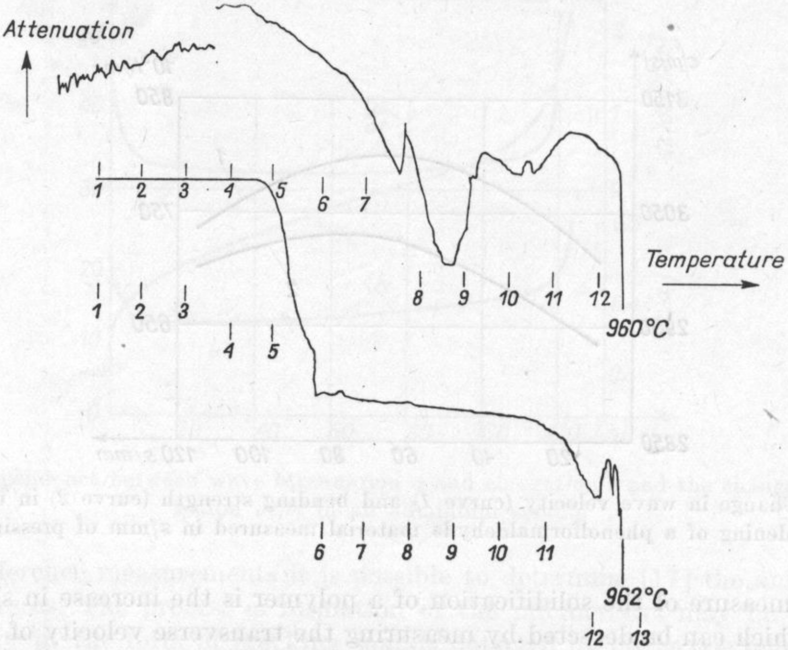

Fig. 14. Changes in wave attenuation as a function of temperature in melting and solidifying pure silver (Kiedrzyński)

In view of the small number of research papers on the influence of the melting or of the solidification of metals on acoustic wave propagation, generalizations concerning these relationships are still lacking. Nevertheless, at the present time the research in this area may be very helpful in the optimization of the parameters of the heat treatment process.

In engineering, ultrasonic methods are used for studying early stages of concrete setting. Different degrees of hydration may be distinguished, while the occurrence of agglomerates C$-$S$-$H ($CaSiO_2$) and C$-$H($CaOH_2$) is marked by maximum attenuation. Audible frequencies are also used here (Fig. 18).

7. Studies on the microstructure of materials

The propagation of ultrasonic waves produces direct or indirect information on the structure of the material.

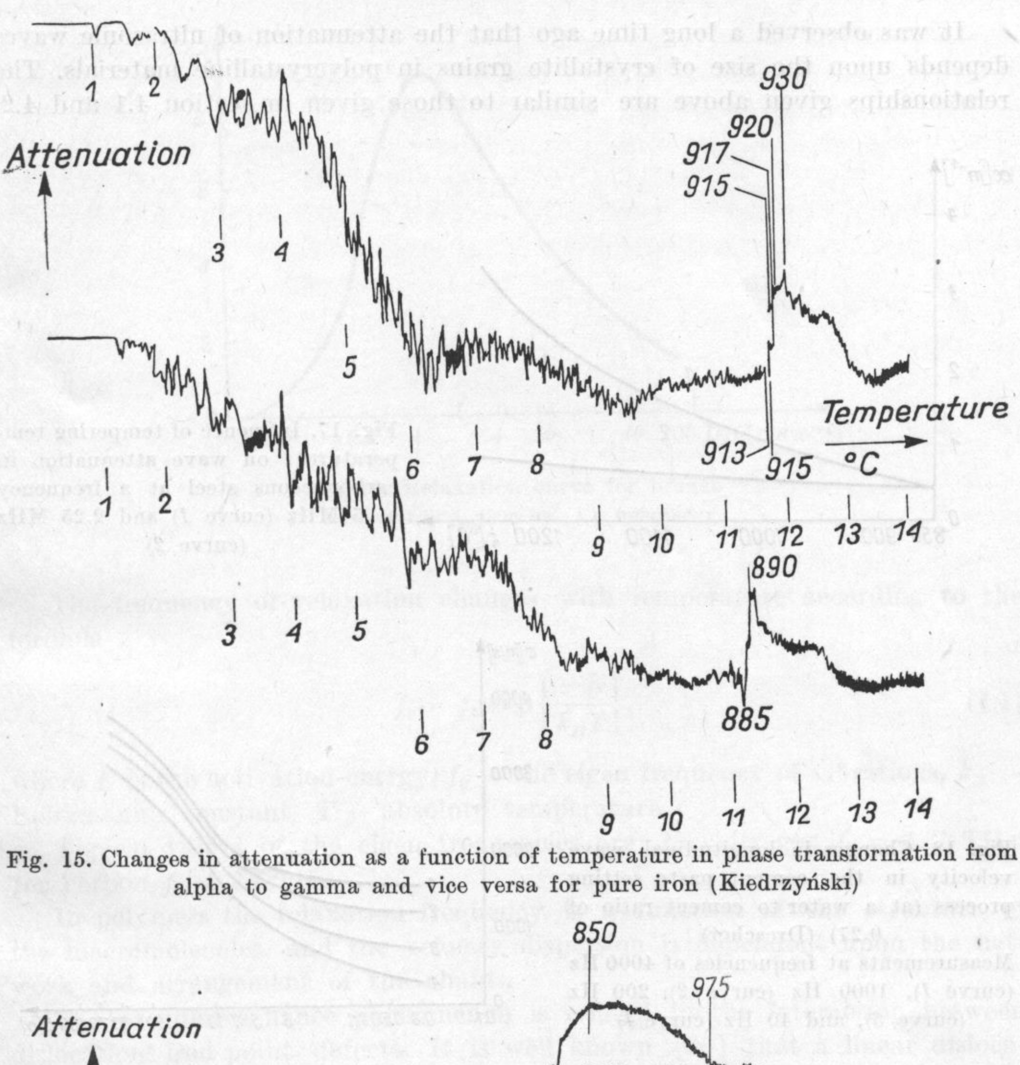

Fig. 15. Changes in attenuation as a function of temperature in phase transformation from alpha to gamma and vice versa for pure iron (Kiedrzyński)

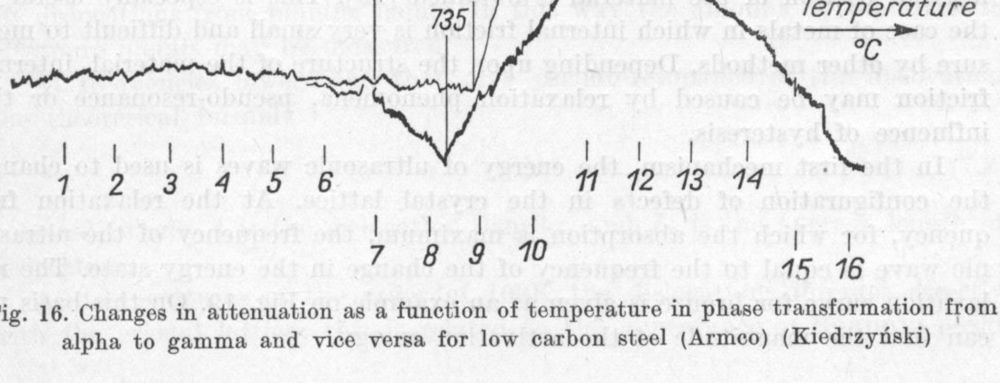

Fig. 16. Changes in attenuation as a function of temperature in phase transformation from alpha to gamma and vice versa for low carbon steel (Armco) (Kiedrzyński)

It was observed a long time ago that the attenuation of ultrasonic waves depends upon the size of crystallite grains in polycrystalline materials. The relationships given above are similar to those given in section 4.1 and 4.2.

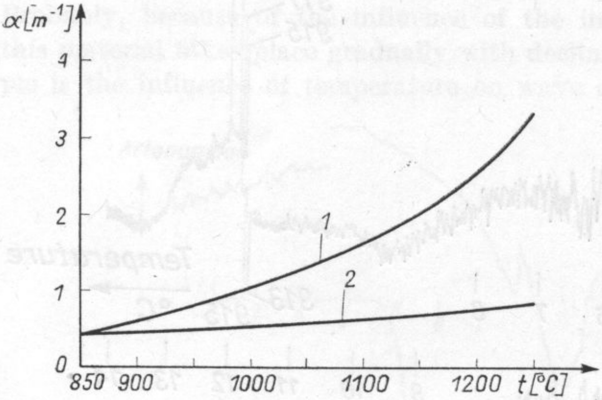

Fig. 17. Influence of tempering temperature t on wave attenuation in carbonaceous steel at a frequency of 5 MHz (curve *1*) and 2.25 MHz (curve *2*)

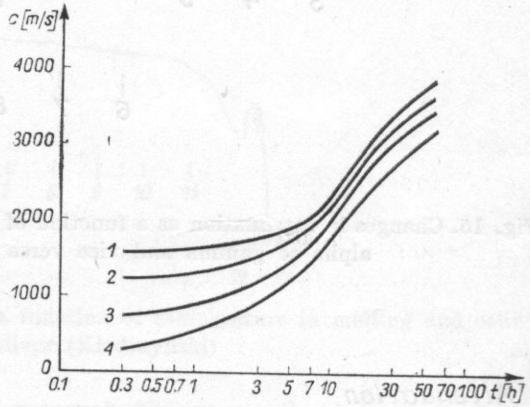

Fig. 18. Changes in longitudinal wave velocity in the cement paste setting process (at a water to cement ratio of 0.27) (Drescher)
Measurements at frequencies of 4000 Hz (curve *1*), 1000 Hz (curve *2*), 200 Hz (curve *3*), and 40 Hz (curve *4*)

By measuring the attenuation of the ultrasonic waves, the magnitude of internal friction of the material is obtained [19]. This is especially useful in the case of metals in which internal friction is very small and difficult to measure by other methods. Depending upon the structure of the material, internal friction may be caused by relaxation phenomena, pseudo-resonance or the influence of hysteresis.

In the first mechanism, the energy of ultrasonic waves is used to change the configuration of defects in the crystal lattice. At the relaxation frequency, for which the absorption is maximum, the frequency of the ultrasonic wave is equal to the frequency of the change in the energy state. The relaxation curve for bronze is given as an example on Fig. 19. On this basis we can find the magnitude of the activation energy.

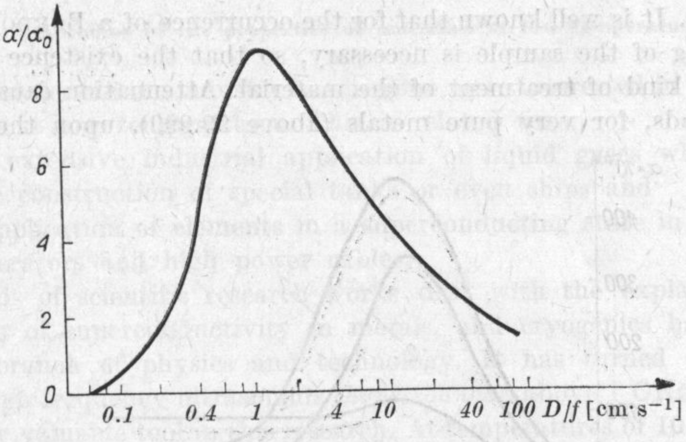

Fig. 19. Relaxation curve for bronze
D – crystallite diameter, f – frequency

The frequency of relaxation changes with temperature according to the formula

$$f_r = f_0 \exp\left\{\frac{-E}{k_B T}\right\}, \tag{7.1}$$

where E is the activation energy, f_0 – the eigen frequency of vibrations, k_B – Boltzmann's constant, T – absolute temperature.

Typical values of the eigen frequencies are: for nitrogen $f_0 = 4.35$ THz, for carbon $f_0 = 2.5$ THz.

In polymers the relaxation frequency is a function of the structure of the macromolecules, and the velocity dispersion is dependent upon the network and arrangement of the chains.

The pseudo-resonance phenomenon is related to the interaction between dislocations and point defects. It is well known [20] that a linear dislocation may be treated as a vibrating string which, at the pseudo-resonance frequency, absorbs maximum energy. Acoustic energy is also used in tearing off dislocations from pinning points. In this way the amount of impurities in ultrapure metals may be measured.

At frequencies lower than that of a pseudo-resonance of the dislocation the theoretical formula

$$a = \Lambda L_0^4 \omega^2 \tag{7.2}$$

is tested, where Λ is the dislocation density, and L_0 is the average length of dislocation.

At temperatures of the order of 100K the dislocations interact directly with the crystal lattice; this is related to the appearance of **Bordoni** peaks

[21] (Fig. 20). It is well known that for the occurrence of a **Bordoni** peak previous crushing of the sample is necessary, so that the existence of the peak indicates the kind of treatment of the material. Attenuation caused by dislocations depends, for very pure metals (above 99.999), upon the percentage of impurities (Fig. 21). As the amount of impurities increases, the dislocation loops become shorter, the frequency at which the maximum occurs increases and the maximum itself becomes flattened.

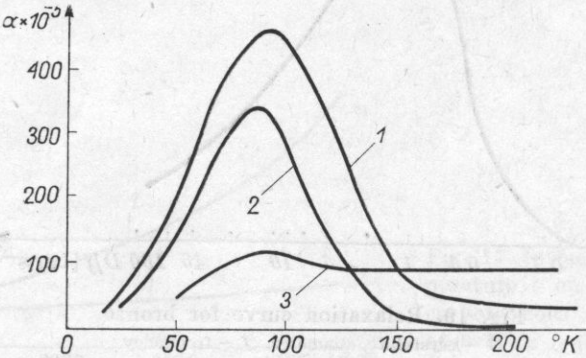

Fig. 20. Wave attenuation for copper indicating the presence of Bordoni peaks
Curve *1* – unannealed copper, curve, *2* – annealed copper, curve *3* – monocrystal of copper

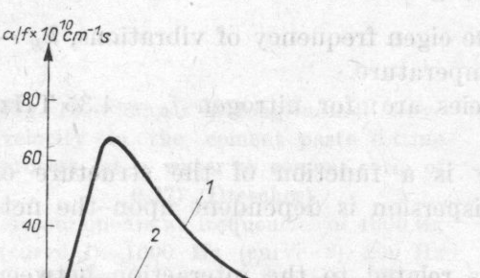

Fig. 21. The dependence of attenuation in very pure aluminum on the impurity density

Curve *1* – 4×10^{-5} % impurities, curve *2* – 19×10^{-5} % impurities, curve *3* – 65×10^{-5} % impurities

Attenuation arising from hysteresis in the material results from the non-linearity of the stress-strain function, which is related to the lowering of strength which, in turn, is related to fatigue processes. The problem is of great practical importance because of the danger of the excessive fatigue of material.

It is worth noting that ultrasound can very well be used not only for measurement, but also for bringing about fatigue in the material. The time of operation for ultrasound is many times shorter than for mechanical fatigue. Especially encouraging results were obtained in the ultrasonic fatigue of hoisting ropes [22]. The frequency of 22 kHz and an electrical power of 600 W applied about 2 minutes are sufficient to obtain visible fatigue in the sample.

8. Studies of the properties of materials at low temperatures

In the last decade the industry has become more interested in the behaviour of materials at low temperatures. This is related to:

1. more extensive industrial application of liquid gases which, in turn, requires the construction of special tanks or even ships and
2. the application of elements in a superconducting state in the construction of generators and high power cables.

Thousands of scientific research works deal with the explanation of the phenomenon of superconductivity in metals, and cryogenics has become an important branch of physics and technology. It has turned out, however, that very high frequency ultrasounds (*hypersounds*), above 1 GHz, may become an especially valuable tool in this research. At temperatures of 10 K the average free path of the phonon is of the order of $10^{-2} \div 10^{-4}$ cm, and so it is of the same order of magnitude as a hypersonic wave length at a frequency of the order of 10 MHz $\div$ 1 GHz.

Under these conditions quantum phenomena are clearly observed and the hypersonic wave must be treated as a flux of phonons. The theory of acoustic phonon collisions [23] with electrons and thermophonons provides an explanation of many phenomena occurring in the state of superconductivity. The observation of the propagation of hypersonic waves gives indirect measurements of the mechanical and electromagnetic parameters of superconductive materials. Studies in this field are at present only in the initial stage, but it should be expected that they will produce valuable results as a result of the common efforts of physicists and technologists.

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