

Experimental Determination of Ultrasonic Wave Velocities in Plastics as Functions of Temperature.

I. Common Plastics and Selected Nose-Cone Materials

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Synopsis

An experimental study was conducted to determine previously unavailable data showing the relationship between temperature and the velocity of longitudinal ultrasonic waves through several plastics. The ultrasonic frequencies used were between 1 and 10 Mc./sec., and the temperature range covered was between room temperature and 100°C. Velocity-temperature curves were determined for nylon, a high- and a low-density polyethylene, Plexiglas, Teflon, and for several reentry vehicle nose-cone materials, including chopped nylon phenolic, tape-wound nylon phenolic, Castable 124, Avcoat I, and Avcoat 19. Less complete data were obtained for Avco phenolic fiber glass, General Electric phenolic fiber glass, and oblique tape-wound refracil. These layered materials are difficult to measure, and the velocity of acoustic waves is very dependent on the direction of propagation with respect to the laminations. For most of the materials studied the velocity was found to decrease more or less linearly with increasing temperature. Typically, the velocities at 100°C. were 9-31% less than at room temperature.

Introduction

An investigation was conducted to determine, as functions of temperature, the velocities of longitudinal ultrasonic waves through several plastic materials, including some of the nonmetallic materials used in the nose cones of ballistic reentry vehicles. This information was needed as input data for research in material dynamics, in particular, the propagation of large amplitude disturbances through plastic materials. Without published data showing the effect of temperature upon velocity it would have been necessary to assume the velocity to be constant under temperature changes. This is a broad assumption which the results of this study show to be invalid.

In view of the accuracy of other input data it was determined that a precision of approximately 5% in velocity values was needed. Considering this and the availability of equipment, it was decided to use a simple direct timing method in which transit times through samples of different thicknesses were measured.

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Experimental Method

The technique of determining velocity by measuring the time required for pulses to traverse samples of different thicknesses was chosen to eliminate complicating considerations such as transducer and circuitry delay times. This is best explained by reference to Figure 1.

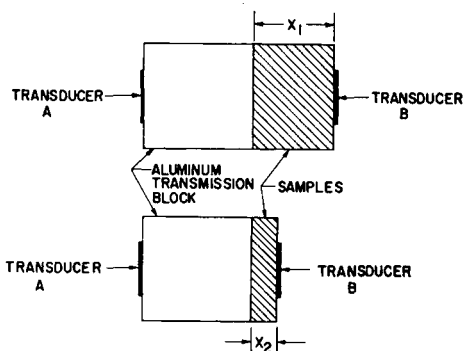


Fig. 1. Two acoustic assemblies, different only in sample thickness.

If the transit time from A to B through the thick sample is t_1 , and the transit time from A to B through the thin sample is t_2 , then the velocity of the pulse through the sample material can be calculated as

$$v = x/t = (x_1 - x_2)/(t_1 - t_2)$$

If the same aluminum block* and the same transducers are used while making the measurements of t_1 and t_2 it is not necessary to determine the transducer delay time and the transit time through the aluminum, since these constant delays disappear on computing the difference in times, $t_1 - t_2$.

Most of the velocity determinations were made by measuring the elapsed times through three samples of different thicknesses, x_1 , x_2 , and x_3 . The corresponding transit times t_1 , t_2 , and t_3 were determined, and then three interrelated velocity calculations were made according to the formulas

$$v_{12} = (x_1 - x_2)/(t_1 - t_2); v_{13} = (x_1 - x_3)/(t_1 - t_3); v_{23} = (x_2 - x_3)/(t_2 - t_3)$$

The arithmetic mean of the three values was computed, and the resultant value was plotted as a point of the velocity-temperature curve. One can also compute the velocity by taking the slope of an x versus t plot. However, considering the limited number of samples used for each velocity determination, no significant advantage is gained.

This method, of course, assumes that the acoustic units can be reproducibly assembled and that the difference in transit times is due only to the difference in sample thicknesses. A bonding clamp, based on a design by Sullivan,¹ was devised to achieve this reproducibility.

* The function of the aluminum block was to make the overall transit time greater than the duration of the ultrasonic pulse so that the transmitted and received signals were well separated in time.

The samples were cut as right circular cylinders approximately 2 in. in diameter, and they ranged in thickness from 3 to 38 mm.

The ceramic transducers used were circular disks designed to operate in the thickness mode, i.e., to produce longitudinal vibrations. They were composed of lead zirconate-titanate mixtures and were operated at frequencies of 1.5, 3.0, 4.33, and 10.0 Mc./sec.

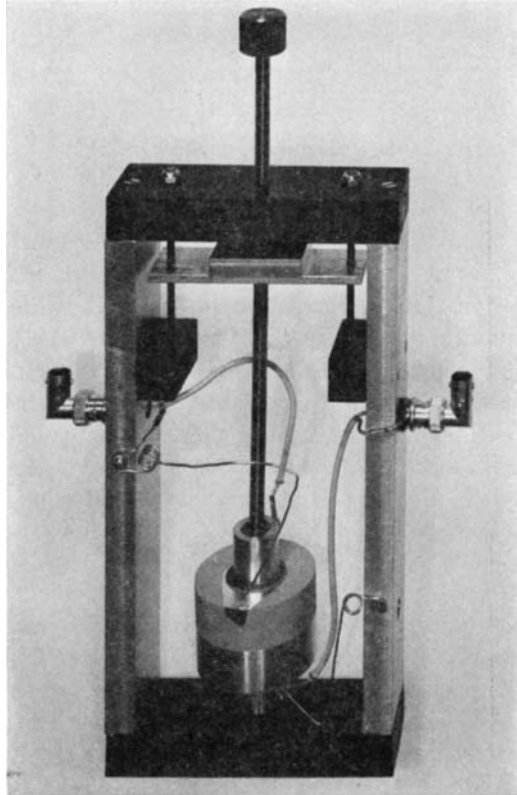


Fig. 2. Bonding clamp with acoustic unit in place. The transducers used here are lead zirconate/titanate.

A light transformer oil (Special Marcol 42-46, Humble Oil and Refining Company) was used as a coupling agent between the various components of the acoustic units. The same oil was used as the bath liquid in the thermostatically controlled bath used to control sample temperatures.

Figure 2 shows the bonding clamp with an acoustic unit in place. The acoustic unit shown in the clamp in this figure is composed of two ceramic transducers (only the upper one being visible), a sample, and an aluminum transmission block. The cylindrical aluminum electrodes of the clamp are mounted on ball joints. Hence they hold the components of the acoustic unit together, accurately aligned and under constant spring pressure (less than 3 psi).

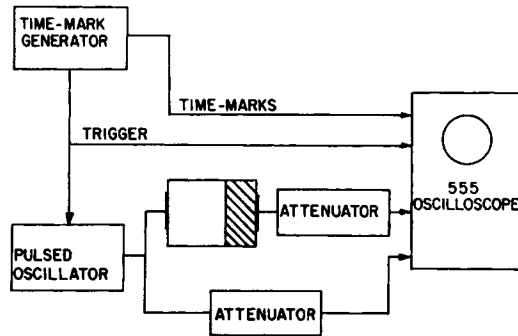


Fig. 3. Block diagram of measuring circuit.

The assembled units were placed in the temperature bath and connected to the measuring circuit shown as a block diagram in Figure 3.

The pulse generating equipment was an Arenberg pulsed oscillator. (Arenberg Ultrasonic Laboratory, Model PG-650C, Boston, Massachusetts.) The attenuators shown in the diagram were also made by Arenberg and were designed to give from 1 to 122 db. of attenuation in 1 db. increments. They were used for convenience in adjusting the amplitudes of the transmitted and received signals so that they could easily be observed on the oscilloscope. The time-mark generator was calibrated and accurate to within 1 ppm. The oscilloscope used for all measurements was a Tektronix Type 555 dual beam scope with two Type CA dual-trace plug-in units.

In most of the pulse methods of measuring ultrasonic velocities which have been reported in the literature, the receivers have contained detectors so that pulse envelopes were observed. Eros and Reitz,² however, have described a single-transducer method based on the observation of the unrectified pulses. The latter was the practice used in this work.

In making velocity determinations by a timing method it is necessary to have, on both the transmitted and received pulses, reference points between which the elapsed time can be measured. Therefore, the unrectified pulses were observed in their entirety on the oscilloscope so that the time could be measured from one particular oscillation of the transmitted pulse to the corresponding oscillation of the received pulse. With this method the velocity that is measured is the phase, rather than the group, velocity. However for the generally nondispersive media investigated here, these velocities are identical.

During the course of this investigation several different time-measuring techniques were used. In the preferred method which was finally developed, ultrasonic pulses of approximately 3 μ sec. duration were used. The position and sweep delay controls of the oscilloscope were used to superimpose the received and transmitted signals on the oscilloscope in order to identify as reference points two corresponding peaks near the middle of the respective pulses. The transit time was then determined by

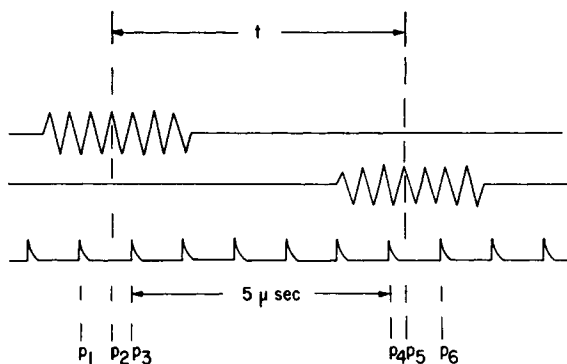


Fig. 4. Schematic representation illustrating interpolation time-measuring technique.

measuring the delays needed to start the sweeps of expanded oscilloscope presentations at the selected reference points. Figure 4 illustrates the method.

The illustration shows three hypothetical oscilloscope traces with a fixed time relationship to each other. They represent, from top to bottom, the transmitted pulse, the received pulse, and a series of $1 \mu\text{sec.}$ time marks. The desired time interval t is that between the fourth positive peaks of the transmitted and received pulses. Readings of the oscilloscope delay potentiometer corresponding to the six events marked P_1 through P_6 were recorded, and the elapsed time was computed by assuming that the oscilloscope delay circuit was sufficiently linear over any $1 \mu\text{sec.}$ interval to permit linear interpolation. With this time-measuring technique, successive measurements with the same acoustic unit regularly agreed to within $\pm 0.004 \mu\text{sec.}$, and most measurements agreed even closer than that. Most of the velocities reported here were determined by this technique.

Experimental Results

Velocity determinations were made on five common plastics and several of the special plastics used in the nose cones of reentry vehicles. The data were then plotted on graphs, longitudinal velocity being used as the ordinate and temperature as the abscissa.

The first material studied over a range of temperatures was a nylon (Poly-Penco nylon, density 1.15 g./cc.). Temperatures ranged from a low of 114°C. to a high of 250°C. The values obtained are plotted in Figure 5. The frequency utilized for most of the measurements was 3.0 Mc./sec. Determinations were also made at frequencies of 1.5 , 4.33 , and 10.0 Mc./sec. with the use of the interpolation timing technique. The later values were consistent with the earlier ones, and all of them are plotted on the same graph. The frequency-dependent deviation appears to be no greater than the deviation due to experimental error, indicating that the material is not dispersive in this frequency range.

The next material studied was the low-density polyethylene (0.916

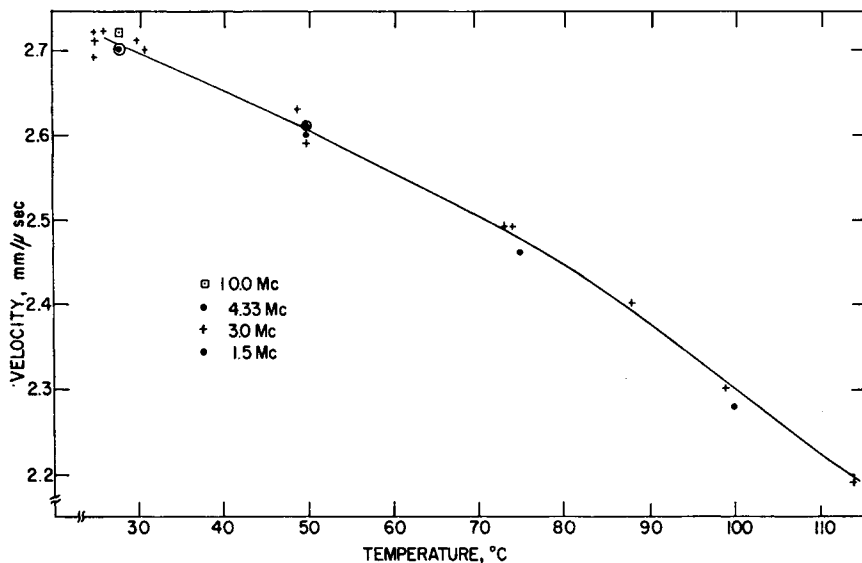


Fig. 5. Longitudinal wave velocity in nylon, density 1.15 g./cc.

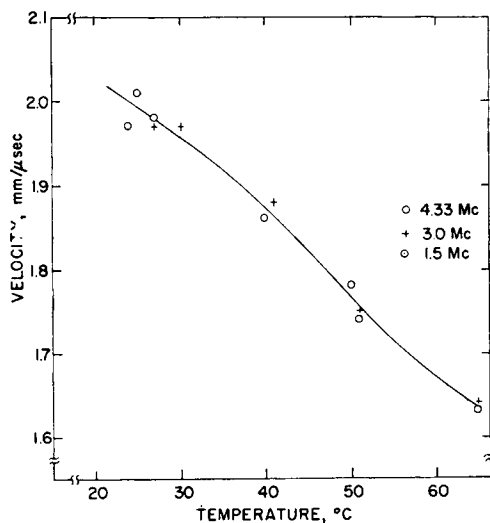


Fig. 6. Longitudinal wave velocity in low-density polyethylene, density 0.916 g./cc.

g./cc.). The velocity data are plotted in Figure 6. The temperature range used here was necessarily limited to avoid melting or distorting the samples.

The longitudinal wave velocity in Plexiglas (density 1.19 g./cc.) is plotted in Figure 7. All measurements for this material were made by using the interpolation timing method. There is close agreement between the values obtained at the different frequencies.

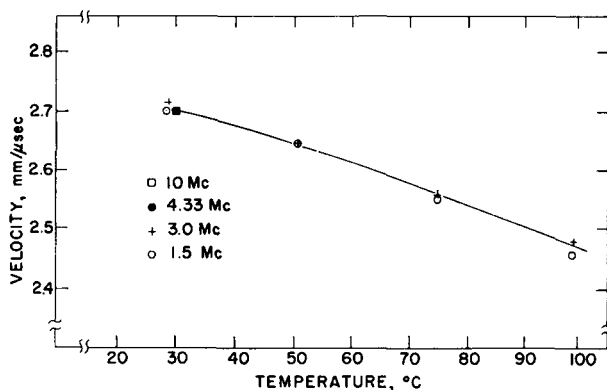


Fig. 7. Longitudinal wave velocity in Plexiglas, density 1.19 g./cc.

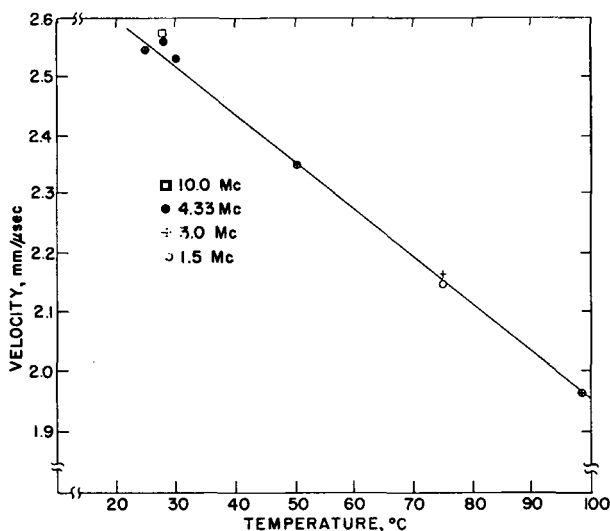


Fig. 8. Longitudinal wave velocity in high-density polyethylene, density 0.964 g./cc.

Figure 8 shows the almost perfectly linear velocity-temperature relationship determined for the high-density polyethylene (0.964 g./cc.). All measurements were made again by using the interpolation timing method. It is interesting to note the variation of velocity in polyethylene for the two densities reported here. Davidse³ et al. has investigated the dependence of velocity on density for a wide range of densities in polyethylene and concludes that the sound velocity is determined only by the density in this material. The data in the present case should not be compared quantitatively with his, since he measured the extensional or thin rod velocity, and the longitudinal bulk velocity was measured here. However, the present results compare qualitatively with his in regard to the dependence of velocity with density.

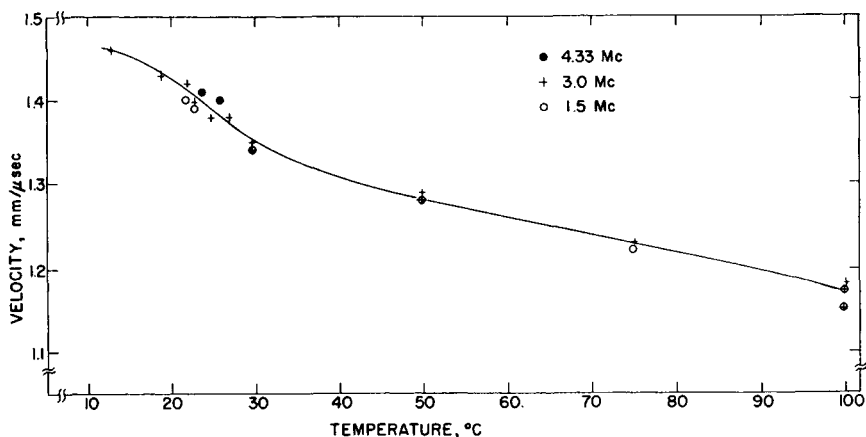


Fig. 9. Longitudinal wave velocity in Teflon, density 2.19 g./cc.

The last common plastic investigated was Teflon (density 2.19 g./cc.). For this material the temperature range was extended to below room temperature in order to obtain a more accurate picture of the shape of the velocity curve in the range between 20 and 30°C. As can be seen in Figure 9, there is a fairly sharp inflection point in the velocity curve which corresponds to a second-order transition in the molecular structure occurring in this region. The velocities measured in Teflon appear to be somewhat more frequency-sensitive than they were in the other materials. Nevertheless, the difference was not sufficient to justify separate plots, and all measurements at the three frequencies used are plotted on the same graph.

The other group of materials studied consisted of the nose-cone materials. In certain of these materials the measurement of ultrasonic velocities was very difficult, and the results obtained should be viewed with caution. This is particularly true in the case of the laminated materials. In the graphs which follow it will be noted that fairly complete and self-consistent sets of measurements have been obtained for some of the materials. In other cases the results have been poor, undoubtedly due to nonuniformity of the samples.

The materials represented in Figures 10 and 11 are chopped nylon phenolic (CNP, density 1.68 g./cc.) and Castable 124 (C124, density, 1.22 g./cc.). In the CNP the axis of propagation was approximately perpendicular to the layers of reinforcing material.

The next graph, Figure 12, shows the velocities measured in two similar materials, Avcoat I (a yellow ceramic, density 1.10 g./cc.) and Avcoat 19 (a green elastomer, density 1.07 g./cc.). The temperature range reported here was reduced to extend between room temperature and 75°C. Measurements were attempted at higher temperatures, but the results were certainly in error because the materials became very soft and visibly

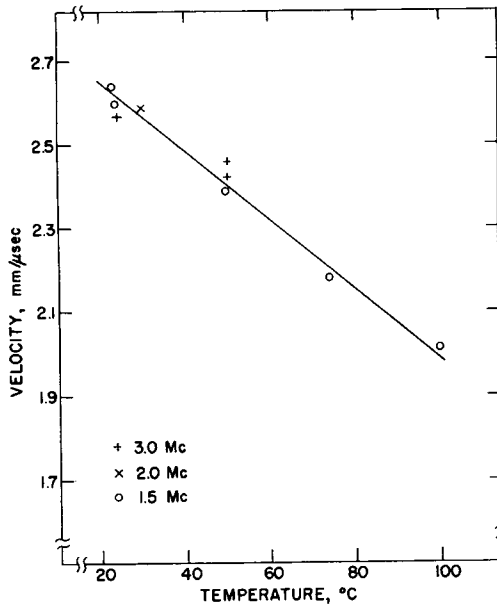


Fig. 10. Longitudinal wave velocity in chopped nylon phenolic, density 1.68 g./cc.

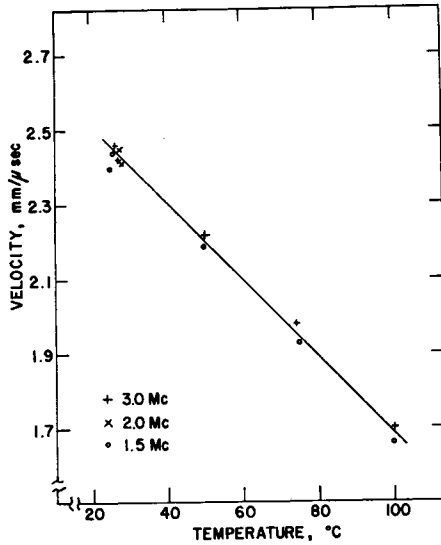


Fig. 11. Longitudinal wave velocity in Castable 124, density 1.22 g./cc.

distorted under even moderate pressure from the spring of the bonding clamp.

The points in Figure 12 which are coded "echo technique" represent velocity determinations made by a technique which was a modification of the method reported by Nolle and Sieck.⁴ This method permitted the use

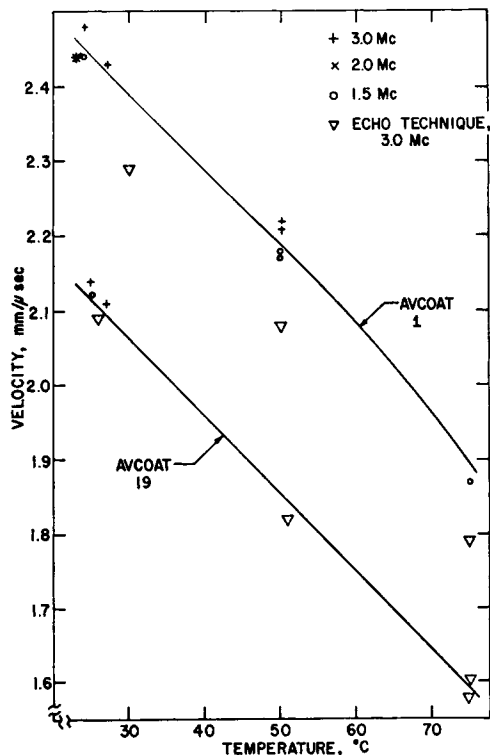


Fig. 12. Longitudinal wave velocities in Avcoat I, density, 1.10 g./cc. and Avcoat 19, density, 1.07 g./cc.

of only one thin sample and was used because of the very high attenuation of ultrasonic waves caused by the thicker samples of these materials at elevated temperatures. In the case of Avcoat 19, the echo technique gave values agreeing with those obtained by the multiple sample technique. For the Avcoat I, on the other hand, the results obtained by use of the echo technique were from 4-5% lower than those obtained by the use of more than one sample.*

Two different configurations of Avco phenolic fiber glass (Avco FG, density 1.70 g./cc.) samples were used. When the axis of propagation of the ultrasonic waves was perpendicular to the reinforcing layers of glass it was found that, for frequencies higher than 1 Mc./sec., the material apparently acted as a low-pass filter. When frequencies higher than this were transmitted into a sample, the received signal was observed to have a lower frequency than that of the transmitted signal. At a frequency of 1 Mc./sec. or below, however, the transmitted and received signals were

* For Avcoat I the disagreement between the two techniques is very probably related to misassignment of reference peaks. The velocity values for both materials were later checked by a different technique which yielded values agreeing with the least-squares fit (solid curves) of the values obtained by the interpolation method.⁵

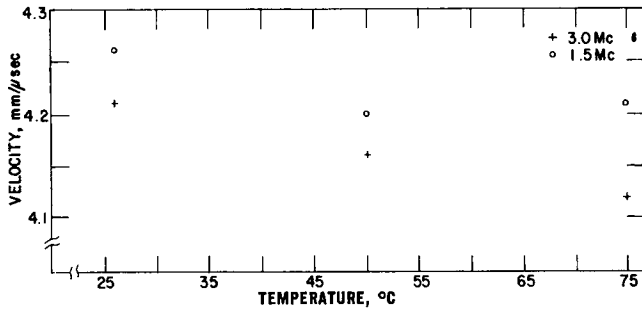


Fig. 13. Longitudinal wave velocity in Avco fiberglass density, 1.70 g./cc., parallel to layers.

identical in frequency. In addition, the attenuation of ultrasonic waves passing through this material perpendicular to the layers increased greatly with increasing temperature. For these reasons, the only measurements made of the "perpendicular" velocity were at room temperature and at a frequency of 1 Mc./sec. The longitudinal velocity under these conditions was found to be 2.3 mm./μsec. This filtering effect has been observed in several, but not all layered, materials and appears to be a complicated combination of layer spacing, relative impedances of each layer, construction geometry of the layers, wavelength and specimen thickness. Further work is being continued to examine this effect and arrive at a more analytical explanation.

The results obtained by passing longitudinal waves through the material in a direction parallel to the layers are shown in Figure 13. This figure shows that the velocity of waves parallel to the layers is much higher than that of waves traveling perpendicular to the layers. The dependence of ultrasonic velocity on the direction of transmission through materials of this type has been reported by Hand⁶ and Dorr and Guenther.⁷

The velocity of waves moving parallel with the laminations of GEFG was measured at 26°C. using frequencies of 1.5 and 3.0 Mc./sec. The values calculated were 4.3 and 4.2 mm./μsec., respectively. These values agree very closely with similar measurements in Avco FG. Further velocity determinations in GEFG were not made because of time limitations. It is, however, expected that the velocity of waves traveling parallel with the layers would be similar to those observed in Avco FG since the velocity of propagation in this direction appears to be primarily determined by the velocity of acoustic waves in the laminating material itself.

Numerous efforts to take meaningful velocity measurements in oblique tape-wound refracil (OTWR, density 1.54 g./cc.) were unsuccessful, in that very erratic results were obtained. The best that can be said is that, between 25 and 75°C., the velocity of waves traveling approximately perpendicular to the layers was found to lie in the range between 2.5 and 2.9 mm./μsec.

Discussion

As stated previously, the intended application of the data presented here did not require a high degree of accuracy. For this reason the points plotted in the data curves show a dispersion considerably greater than that obtainable by this method under optimum conditions. Excluding the rather erratic results obtained with some of the laminated materials, the greatest deviation between the maximum and minimum velocity values obtained at the same temperature was somewhat less than 3%. It should be recalled, however, that this includes determinations made at different frequencies and by different time-measuring techniques. In addition, there are included in the plotted points velocity measurements which were later discovered to have been made with samples which had become irregular in thickness because of dimensional instability.

The velocities calculated for the five common plastics were corrected for thermal expansion of the samples by using handbook⁸ values for the coefficients of linear expansion. There was no information available on the thermal expansion of the nose-cone materials, and the velocities computed for these materials have not been corrected for expansion. Because of this, the velocities calculated at the higher temperatures are too low, probably by not more than 1%.

An examination of the results obtained for the homogeneous and isotropic materials by use of the interpolation timing method gives an indication of the precision attainable by this method. In these cases, there was no instance in which there was more than 1% deviation between the extreme velocity values determined under the same conditions of temperature and frequency.

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References

1. Sullivan, P. F., *J. Acoust. Soc. Am.*, **34**, 1879 (1965).
2. Eros, S., and J. R. Reitz, *J. Appl. Phys.*, **29**, 683 (1958).
3. Davidse, P. D., Waterman, H. I., and Westerdijk, J. B., *J. Polymer Sci.*, **59**, 389 (1962).
4. Nolle, A. W., and P. W. Sieck, *J. Appl. Phys.*, **23**, 888 (1952).
5. Asay, J. R., Air Force Weapons Laboratory, Kirtland AFB, New Mexico, private communication.
6. Hand, W., *Plastics Technol.*, **8**, 35 (February 1962).
7. Dorr, A. J., and A. H. Guenther, *J. Appl. Polymer Sci.*, **10**, 745 (1966).
8. *Handbook of Chemistry and Physics*, 44th Ed., The Chemical Rubber Publishing Company, Cleveland, Ohio, 1962.

Résumé

On a effectué une étude expérimentale en vue de déterminer des données jusqu'ici non disponibles montrant le rapport existant entre la température et la vitesse de propagation d'ondes ultrasoniques longitudinales à travers différents plastiques. Les fréquences

ultrasoniques utilisées étaient entre 1 et 10 mégacycles et le domaine de température couvert s'étendait depuis la température ordinaire jusqu'à 100°C. Les courbes vitesse-température ont été déterminées pour le nylon, un polyéthylène à haute et basse densité, le plexiglas, le téflon, et pour de nombreux matériaux servant à la réentrée des véhicules, y compris le nylon phénolique, le nylon phénolique sous forme de rouleau, le Castable 124, Avcoat I et Avcoat 19. Des données moins complètes ont été obtenues pour la fibre de verre Avco phénolique, la fibre de verre phénolique de la General Electric, et le rouleau refrasil. Ces matériaux en couche sont difficiles à mesurer et la vitesse des ondes acoustiques est fortement fonction de la direction de propagation par rapport aux couches laminées. Pour la plupart des matériaux étudiés, la vitesse décroissait plus ou moins linéairement avec une température croissante. Typiquement, les vitesses à 100°C étaient de 9 à 31% inférieures à celles de la température de chambre.

Zusammenfassung

Eine experimentelle Untersuchung zur Bestimmung bisher nicht zugänglicher Daten über die Beziehung zwischen Temperatur und der Geschwindigkeit longitudinaler Ultraschallwellen in mehreren plastischen Massen wurde ausgeführt. Die verwendeten Ultraschallfrequenzen lagen zwischen 1 und 10 Mhz und der überstrichene Temperaturbereich lag zwischen Raumtemperatur und 100°C. Geschwindigkeits- und Temperaturkurven wurden für Nylon, Polyäthylen hoher und niedriger Dichte, Plexiglas, Teflon und für einige Stoffe für Raumfahrzeugspitzen für den Wiedereintritt in die Atmosphäre, einschliesslich der Nylonphenolharze, Castable 124 Avcoat I und Avcoat 19. Weniger vollständige Daten wurden für Avco-Phenolharzfaserglas, General Electric Penolharzfaserglas und Schrägband-umwickeltes Refrasil erhalten. Diese Schichtmaterialien sind schwer zu messen und die Geschwindigkeit von Schallwellen hängt stark von der Fortpflanzungsrichtung gegen die Schichtrichtung ab. Bei den meisten untersuchten Stoffen nahm die Geschwindigkeit mehr oder weniger linear mit steigender Temperatur ab. Die Geschwindigkeit bei 100°C war in typischen Fällen 9 bis 31% geringer als bei Raumtemperatur.

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