Measurements of Ultrasonic Velocity and Attenuation in Biaxially Oriented Polypropylene Films Using Fast Fourier Transform

SVETOZÁR MALINARIČ

Department of Physics, University of Education, SK-949 74 Nitra, Slovakia

SYNOPSIS

The first part of this study deals with the design of a new instrument for measuring the velocity and attenuation of ultrasonic waves propagating in thin polymeric films. The experimental results indicate a large anisotropy. In addition, a strong velocity and attenuation dependence on both frequency and temperature has been observed. The results are explained by means of phenomenological theory. © 1994 John Wiley & Sons, Inc.

INTRODUCTION

The basic object in making mechanical wave measurements is to determine the velocity and attenuation. These measures may then be related to structural properties of the material. The wave velocity is primarily a function of the elastic constants, while the attenuation is largely determined by dissipative mechanisms operating in the material.[1]

In general a choice has to be made between the use of continuous wave or pulse. The traveling-wave method has been used for making measurements on thin strips of materials and fibers. The specimen is driven at one end into vibration by means of a suitable transducer. A second transducer, placed in contact with the specimen, is used to record the longitudinal waves generated in the specimen. Disadvantages of this method includes the difficulty of avoiding standing-wave effects.

Pulse methods involve the timing of the passage of a wave packet as it traverses the material and are particularly suited to the study of material properties at high frequencies. It is difficult to extract the phase velocity corresponding to a given frequency unless Fourier analysis is employed. Even the determination of the group velocity is not reliable if appreciable dispersion is present.

Journal of Applied Polymer Science, Vol. 52, 1405–1410 (1994)
 CCC 0021-8995/94/101405-06

The measurements of the propagation velocity have been generally used in the evaluation of molecular anisotropy and orientation in polymeric fibers and films,[2] but very few studies have been reported on biaxially oriented polypropylene (BOPP) films.[3–5] It is often difficult to understand some properties of BOPP films when referring to the general behavior of nonoriented polypropylene films.

The purpose of this study is to provide some insight on the anisotropy of BOPP films in connection with viscoelastic properties.

EXPERIMENTAL

More detailed information on the material behavior may be obtained by making ultrasonic measurements at more than one frequency. Using Fourier transform method one can obtain information on the propagation of ultrasonic waves in a given material system in the form of a response spectrum.

In the first part of this study a special instrument for measuring the ultrasonic wave velocity and attenuation is described. The idea for the mechanical arrangement has been taken from a commercially available instrument (PPM-5, H. M. Morgan Co., Inc.).[6] It consists of two transducers made from bending piezoceramics—one serves as transmitter and the other as receiver. The film strip is 1 mm wide and 200 mm long and is kept taut between the

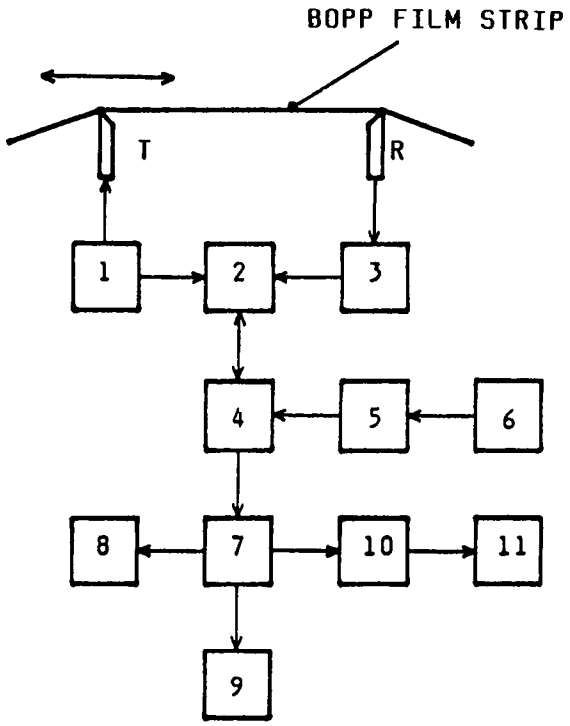

Figure 1 Block diagram of the instrument for ultrasonic velocity and attenuation measurements: T transmitter, R receiver, 1 generator, 2 sampling oscilloscope, 3 preamplifier, 4 PC AT-286, 5 ohmmeter, 6 thermistor, 7 interface, 8 stepping motor, 9 ventilator, 10 digital to analog converter, 11 heating elements.

transducers. The transmitter can be moved along the specimen by means of a screw and a stepping motor.

Figure 1 shows the block diagram of the instrument. The whole experiment is controlled and evaluated by the personal computer AT-286. The square-wave pulse is supplied by a generator and converted to mechanical oscillations by the transmitter. After having propagated in the specimen the pulse is converted back to voltage in the receiver and amplified. Then the signal is sampled by PCIP-SCOPE and saved in PC memory. The other instrumentation-like thermistor, ohmmeter, etc., enable automatic temperature control in the range from 30 to 70°C.

The principle of the method resides in sampling signal at two different transmitter locations (a and b). Two files, each containing 1024 samples at 10-MHz sampling rate, are obtained. Using the fast Fourier transform (FFT) algorithm,[7] two complex series in frequency domain are obtained. In accordance with the basic principle of ultrasonic spectrometry, the transfer function of the generator, transducers, and amplifier are eliminated.

Then, the complex transfer function $D_{ab}(f)$ of the strip, between a and b positions of the transmitter, is given by the expression

$$D_{ab}(f) = |D_{ab}(f)|\exp[j\theta_{ab}(f)] = \frac{U_a(f)}{U_b(f)} \quad (1)$$

where $U_a(f)$ and $U_b(f)$ are the voltage responses of the system. $|D_{ab}(f)|$ is absolute value and $\theta_{ab}(f)$ is a phase of the complex number $D_{ab}(f)$. The attenuation $\alpha(f)$, is given by

$$\alpha(f) = -20\frac{\log|D_{ab}(f)|}{x_a - x_b} \quad (2)$$

where x_a, x_b are the distances between transducers at the two transmitter positions. The phase velocity is given by

$$c_f(f) = \frac{x_a - x_b}{\theta'_{ab}(f)/2\pi f} \quad (3)$$

where $\theta'_{ab}(f)$ is the phase of $D_{ab}(f)$ after phase-straightening process.[8]

Two commercially available BOPP films for electrical use (film 1 and BK6) were selected for the present study. Both were made by the "tenter"-type orientation. The machine direction (MD) and the transverse direction (TD) orientations were carried out successively.

RESULTS AND DISCUSSION

Figure 2 shows the results of ultrasonic velocity measurements on film 1. The results obtained by ultrasonic spectrometry are compared with ones obtained by the traveling-wave method. One can see quite good agreement between the two methods. At frequencies below 50 kHz there is larger dispersion in experimental data because of the overlapping of the main pulse with an echo generated by reflection from the strip holders and transducers. The echo must be removed from the signal since it leads to a loss of information in low-frequency range. The reproducibility of the ultrasonic spectrometry was determined as 0.5% for velocity and 10% for attenuation measurements over a frequency range from 50 to 400 kHz.

Figure 3 shows the frequency dependence of the ultrasonic velocity and attenuation measurements on film BK6. In accordance with the other mea-

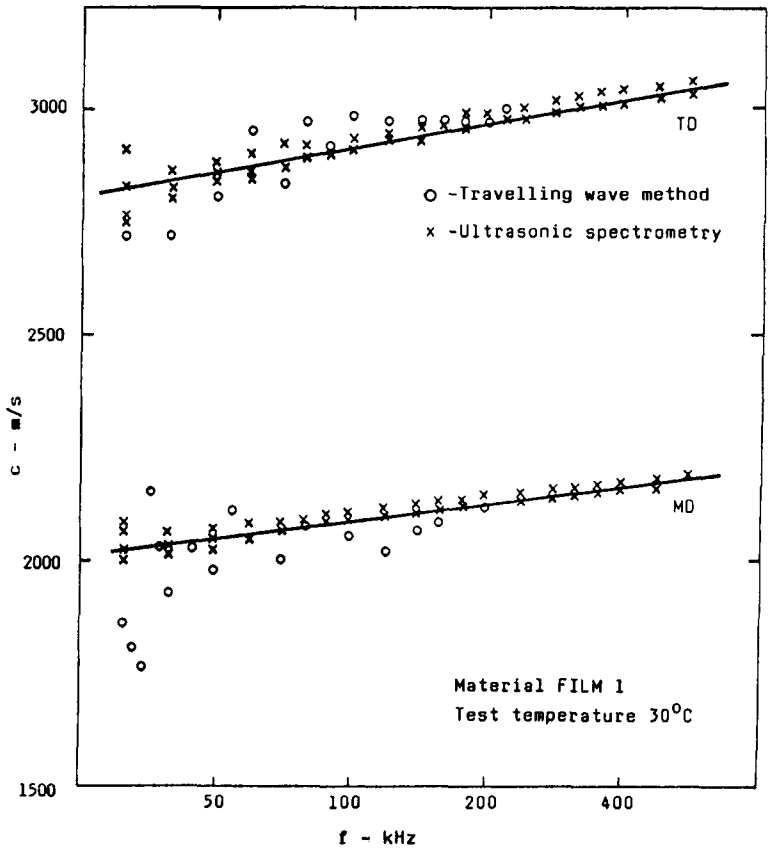

Figure 2 *Results of the ultrasonic velocity measurements of the film 1.*

surements on polymers,[9] the linear growth of attenuation with frequency is referred to as hysteresis absorption. If we extrapolate the curve to intersect the α axis then at $f = 0$, we will not get precisely $\alpha = 0$, which seems to have no physical sense. But we must realize that the attenuation measuring accuracy is not very good and it can cause this phenomenon.

The velocity shows a linear growth with the logarithm of excitation frequency and considerable drop in magnitude with temperature. This observed phenomenon is referred to as relaxation behavior of polymers.[10] By applying the time–temperature superposition principle the curves measured at different temperatures can be shifted to generate a master curve.

Figure 4 illustrates the algorithm used to generate the master curve for the square of the ultrasonic wave velocity versus the logarithm of frequency. The straight lines corresponding to different temperature can be described by equations:

$$c^2_{1(2)}(f) = a_{1(2)} + b_{1(2)} \ln f \qquad (4)$$

The y coordinate of a center y_c can be defined as

$$y_c = [c_1^2(f_1) + c_2^2(f_2)]/2 \qquad (5)$$

Then, the shift factor can be determined

$$\ln a_T(1, 2) = (y_c - a_1)/b_1 - (y_c - a_2)/b_2 \qquad (6)$$

so that point P_2 is moved into point P_1 and the abscissa T_2 is shifted to the left to a new position in Figure 4.

Figure 5 shows the plot of c^2 versus $\ln f$ from the measurements made on film BK6 before and after time–temperature superposition. From the four curves one master curve is obtained in the interval from 20 Hz to 400 kHz. Now the frequency range is broad enough to compare measurements with theory. There are a number of existing models[1] describing the mechanical relaxation process. Viscoelastic properties of polymers are usually described by the Zener solid model, giving the relation

$$c^2 = A - B/[1 + (2\pi f\tau)^q] \qquad (7)$$

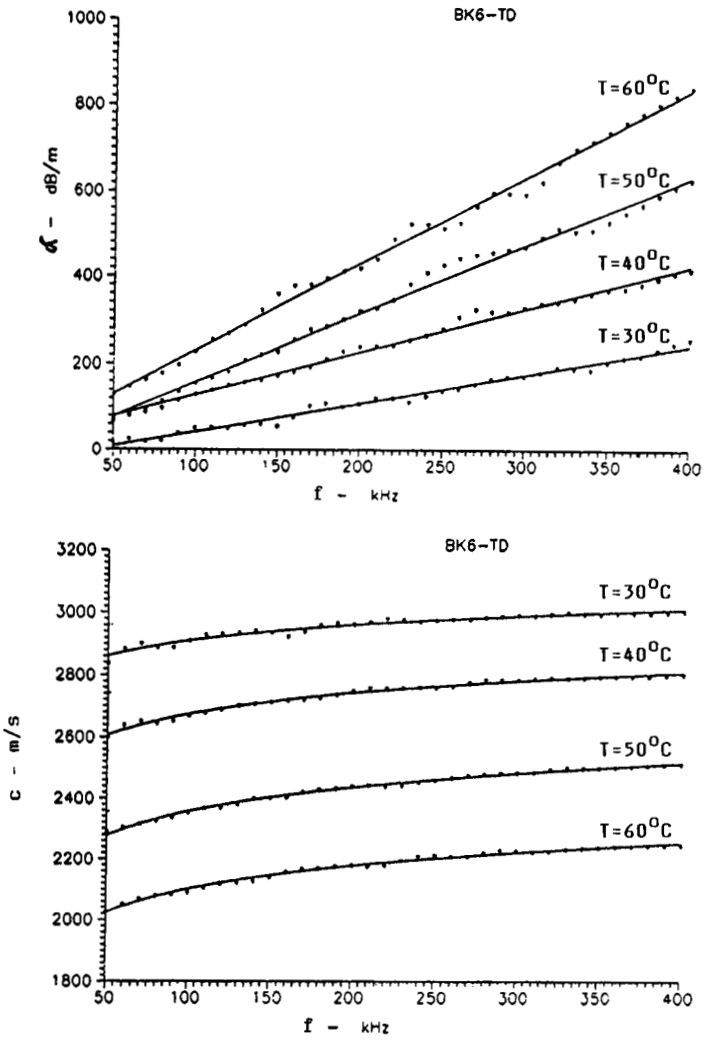

Figure 3 Results of the ultrasonic velocity and attenuation measurements of film BK6.

where c is the phase velocity, A and B are material constants, τ is the relaxation time, and q is a parameter of relaxation time distribution.

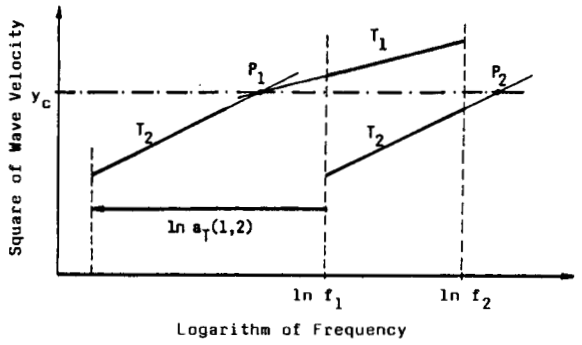

Figure 4 Illustration of the time–temperature superposition algorithm.

Figure 6 shows three curves computed by using Eq. (7) for parameter $q = 0$, 0.5, and 2, where $A = 9.5 \times 10^6$ m^2 s^{-2}, $B = 6 \times 10^6$ m^2 s^{-2}, and $\tau = 53$ μs. The curve $q = 2$ refers to the Zener solid with a single relaxation time. Because the transition region is very narrow, this curve cannot describe the measurements on Figure 5. The curve $q = 0.5$ refers to broadened relaxation time distribution and fits the measurements on Figure 5. For $q = 0$ the relaxation times are extremely spread and so the relaxation behavior is lost.

Only one term in Eq. (7) depends on temperature, that is, the relaxation time τ. The relation is given by

$$\tau = \tau_0 \exp(U/kT) \tag{8}$$

where U is an activation energy, k is the Boltzmann constant, and T is the absolute temperature. The

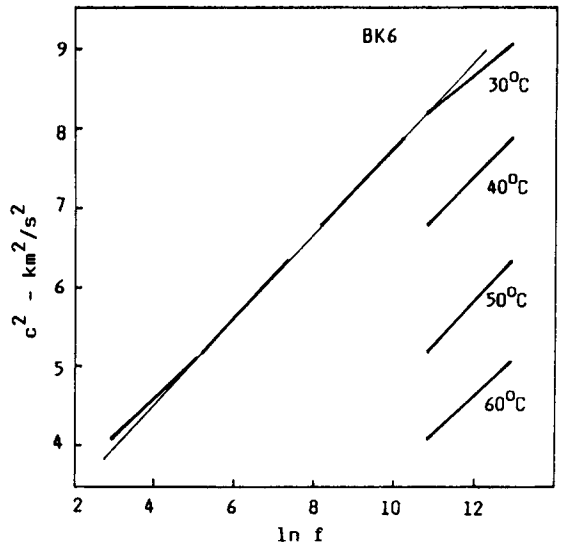

Figure 5 Application of the time–temperature superposition for measurements of film BK6.

shift factor, using the well-known Williams–Landel–Ferry (WLF) equation may be written as

$$\ln a_T = \ln\left[\frac{\tau(T)}{\tau(T_0)}\right] = \frac{U}{kT} - \frac{U}{kT_0} \qquad (9)$$

Figure 7 shows the dependence of $\ln a_T$ versus $1/T$ for the time–temperature superposition shown in Figure 5. According to the relation,[9] the dependence should be a straight line from which the activation energy $U = 2.3$ eV is calculated. Despite the large anisotropy, the activation energy gave nearly the same value for both films and both directions (MD, TD).

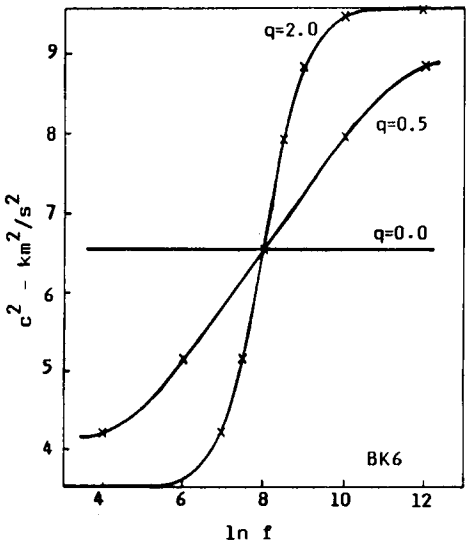

Figure 6 Theoretical curves that should fit the measurements in Figure 5.

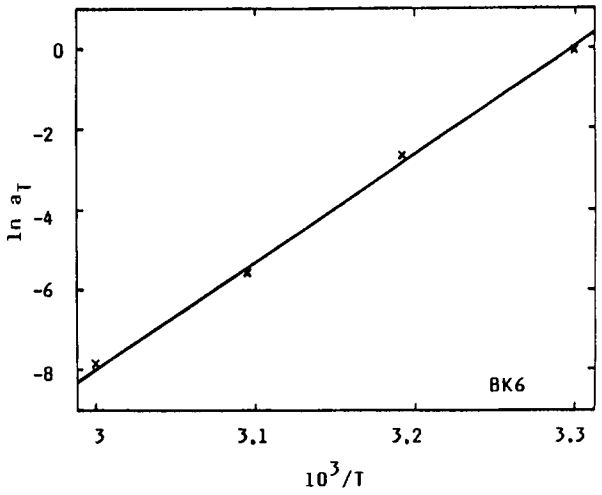

Figure 7 Dependence of the shift factor on temperature for measurements of film BK6.

CONCLUSIONS

The first part of this study dealt with the design of a new instrument for measuring ultrasonic wave velocity and attenuation.

The second part dealt with the application of it for BOPP films. A large anisotropy in ultrasonic wave velocity has been observed. Besides this, a strong dependence of wave velocity and attenuation on both frequency and temperature has been observed. The results are interpreted by phenomenological theory using the Zener solid model. A simple algorithm for time–temperature superposition has been designed, and the model for the relaxation behavior has been extended on relaxation time distribution. The activation energy has been determined and gave nearly the same value for both films and both directions (MD, TD).

The method and instrument are excellent for the study of orientation, anisotropy, and homogeneity.[11-13] But it would be necessary to extend the temperature range of the experiment in order to obtain a deeper understanding of the relaxation behavior in the material.

REFERENCES

1. H. F. Pollard, *Sound Waves in Solids,* Pion Ltd., London, 1977.
2. V. Kalousková, *Europhysics Conference Abstracts, Morphology of Polymers,* Vol. 9E, Praha, 1985.

3. T. Umemura and K. Akiyama, Conference Record of 1984 IEEE International Symposium on Electrical Insulation, Montreal, 1984.
4. R. J. Samuels, *J. Appl. Polym. Sci.*, **26,** 1383 (1981).
5. G. Metzger and A. Vicaud, Conference Record of 1984 IEEE International Symposium on Electrical Insulation, Montreal, 1984.
6. R. J. Samuels, *Structured Polymer Properties,* Wiley, New York, 1974.
7. J. Obraz, *Ultrasonic Testing of Materials* (in Czech), SNTL, Praha, 1989.
8. R. K. Otnes and L. Enochson, *Applied Time Series Analysis,* Wiley, New York, 1977.
9. B. Hartmann and J. Jarzynski, *J. Appl. Phys.*, **43,** 4304 (1972).
10. J. J. Aklonis, W. J. MacKnight, and M. Shen, *Introduction to Polymer Viscoelasticity,* Wiley, New York, 1972.
11. S. Malinarič, *Plasty a kaučuk* (in Slovak), **28,** 198, (1991).
12. S. Malinarič, *Elektroizolačná a káblová technika* (in English), **45,** 68, (1992).
13. S. Malinarič, *Plasty a Kaučuk* (in Slovak), **30,** (1993).

Received January 26, 1993
Accepted December 8, 1993