ACOUSTIC PROPERTIES OF MULTILAYERED POLYURETHANE/ POLYVINYLCHLORIDE BLENDS

J.P. Szabo (1), J.Y. Guigné (2), and L.A. Utracki (3)

(1) Defence Research Establishment Atlantic, PO Box 1012, Dartmouth, NS, Canada B2Y 3Z7
(2) Guigné International Limited, Paradise, Newfoundland, Canada A1L 1C1
(3) National Research Council Canada, Boucherville, Quebec, Canada J4B 6Y4

1. INTRODUCTION

The purpose of this investigation was to characterize the acoustic properties of ester-based thermoplastic polyurethane (TPU) /polyvinylchloride (PVC) blends as single and multiple layers. We have previously shown [1] these thermoplastic blends to be two phase structures, with an Interpenetrating Polymer Network (IPN) morphology in the concentration range 30-50% PVC. For sound and vibration damping materials, blends based on IPN morphology have been of interest due to their broad frequency response and their improved performance [2,3]. Furthermore, the properties of blends may be easily adjusted, e.g. the impedance of a blend can be modified by combining two polymers with different sound speeds in various ratios. These factors make the system chosen an interesting one for the experimental and theoretical investigation of multilayers.

Due to the difficulty of making large size panels, an experimental technique for acoustic characterization of small panels was chosen, namely the utilization of a truncated parametric array source. This technique, pioneered by Humphrey [4,5], has the advantage of providing an acoustic source with a narrow beamwidth and no side lobes, which reduces the effects of edge diffraction from the small panel. The wide frequency bandwidth of the parametric source is also an advantage for carrying out panel measurements.

In this study, the experimental insertion loss spectra were compared with simulated spectra for individual layers and for a sample containing seven layers with PVC content varying from 0% to 60% through the layers.

2. DYNAMIC MECHANICAL PROPERTIES

The thermoplastic resins used were PVC 87444 (BF Goodrich) and an ester based urethane, TPU PS-49-100 (Morton Thiokol). After overnight drying under vacuum at 95°C, the resins were dry blended at room temperature in the desired proportions, then compounded in a twin-screw, zsk-30 extruder. The melt was then molded under vacuum at 180°C in an autoclave at 41.4 MPa.

For the multilayered sample, the individual layers were demolded and their surfaces were cleaned to remove the mold release agent and to enhance the inter-layer adhesion. The layers were placed on top of one another in the appropriate sequence in the mold, and remolded at 170°C.

The dynamic mechanical properties of the blends are included here for completeness. A more detailed description of the thermal and viscoelastic behaviour of these materials has been published elsewhere [1]. The complex Young's modulus ($E = E' + iE''$) of the samples were determined as a function of temperature using a DuPont DMA 983 dynamic mechanical thermal analysis (DMTA)

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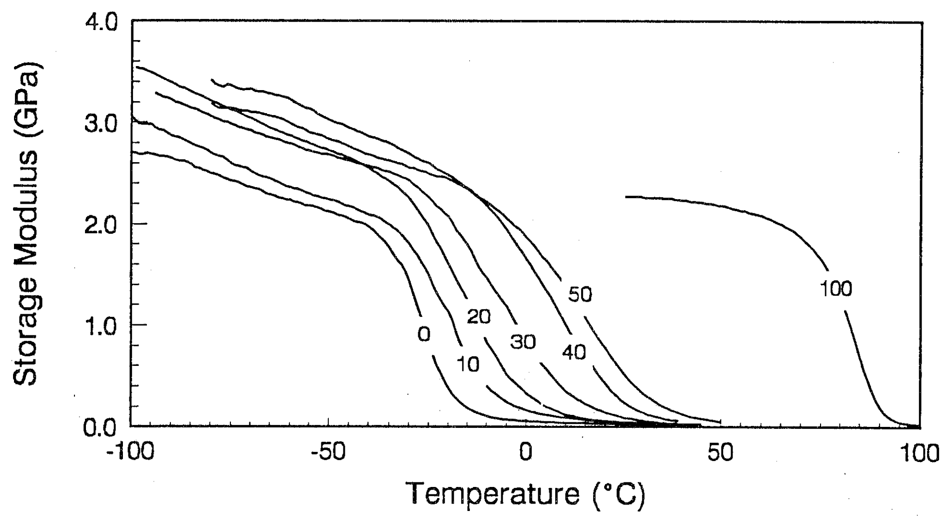

Figure 1. Tensile storage modulus (E') versus temperature for TPU/PVC samples excited in flex mode at 1 Hz. The number on each curve corresponds to the volume concentration of PVC in percent.

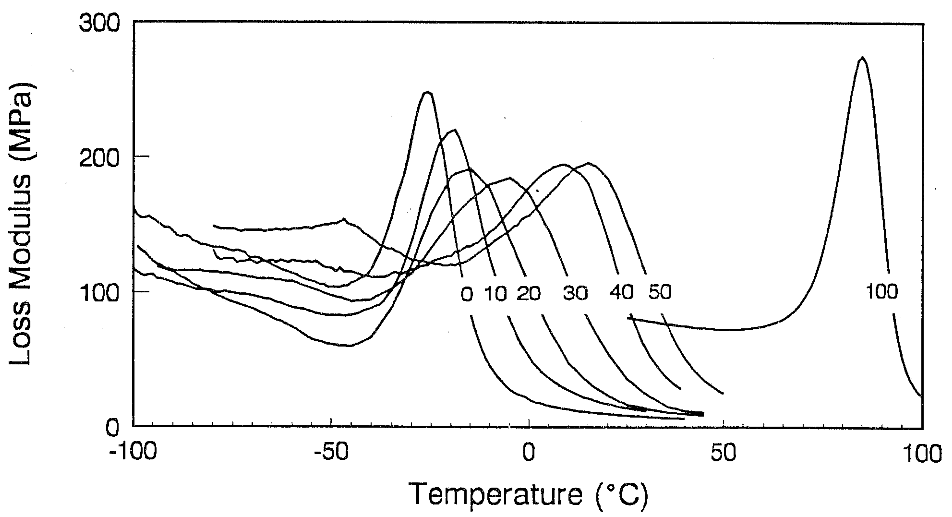

Figure 2. Tensile loss modulus (E") versus temperature for TPU/PVC samples excited in flex mode at 1 Hz. The number on each curve corresponds to the volume concentration of PVC in percent.

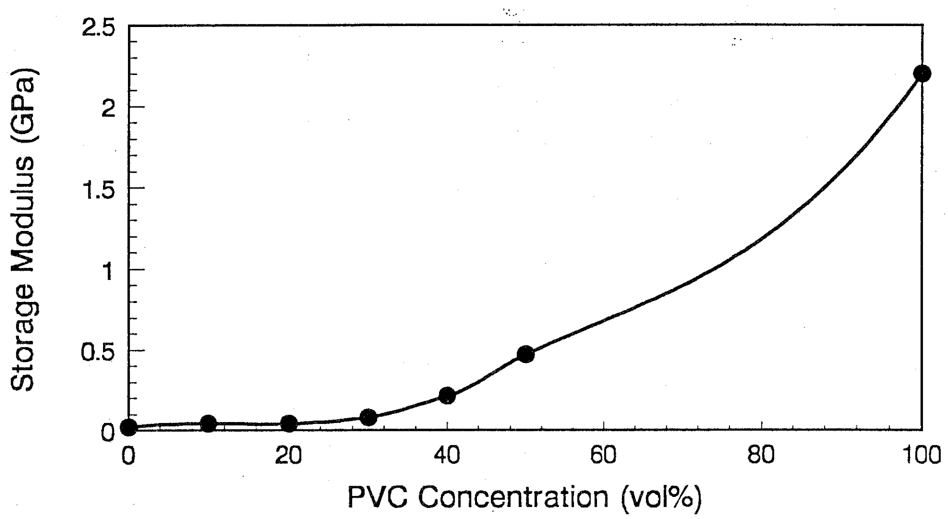

Figure 3. Tensile storage modulus (E') versus PVC concentration for TPU/PVC blends at 23°C and 103 Hz. The solid line is a cubic spline fit (———) to the data points (•).

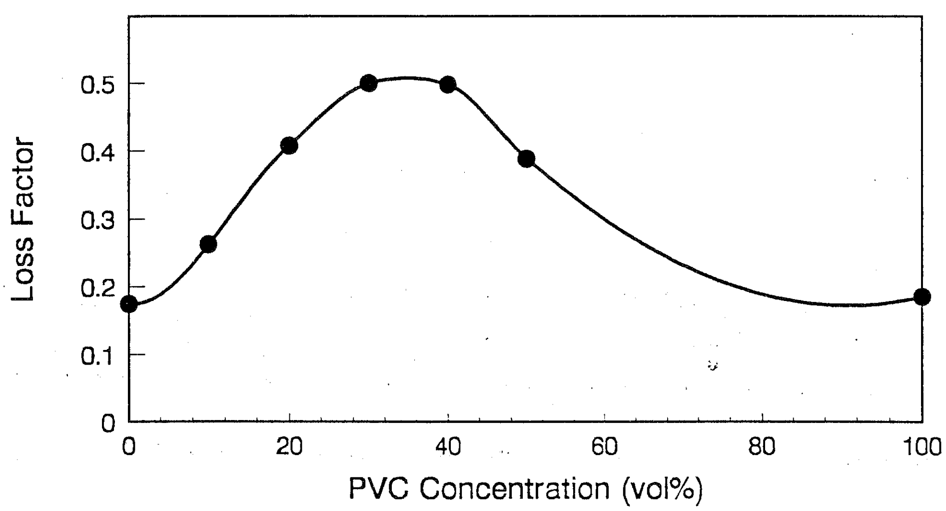

Figure 4. Material loss factor (tan δ) versus PVC concentration for TPU/PVC blends excited in tension at 23°C and 103 Hz. Data points (•), cubic spline fit (———).

instrument in fixed frequency mode. Figure 1 shows the storage modulus (E') versus temperature for samples excited in flex mode at 1 Hz. The glass transition temperatures of the blends, as determined by the inflection points on the storage modulus curves or the peaks in the loss modulus (E") curves, Figure 2, increase linearly with PVC content. A single transition in the DMTA thermogram is usually associated with single component materials or miscible blends, yet transmission electron microscopy studies clearly show these blends to be two phase systems [1].

The presence of two phases is of interest here, since the acoustic properties of a two phase system may be different than for a one phase system. The properties of miscible blends often follow a linear additivity rule, while two phase systems often display an abrupt change in properties in a composition range where phase inversion occurs [6], i.e. where a component changes from a continuous network to a dispersed state or vice versa. Figures 3 and 4 show the storage modulus and material loss factor ($\tan \delta = E''/E'$) measured using a direct stiffness method [7] plotted versus concentration of PVC. Note that rather than being linear with concentration, the loss factor curve peaks and the E' curve has an inflection point around 40% PVC. Based on the dynamic mechanical data, one might expect the sound speeds or attenuations to have a non-linear dependence on concentration due to the presence of two phases.

3. INSERTION LOSS EXPERIMENTS

The theoretical and practical details of using parametric array sources for materials characterization have been described in detail elsewhere [4,5]. For these experiments, a parametric array transducer with a 1 MHz primary frequency was used to carry out insertion loss measurements, as shown in Figure 5. A short pulse of the carrier was modulated with a 40 kHz raised cosine bell envelope in order to obtain a secondary signal suitable for the 10-100 kHz frequency range. A 0.75 mm thick stainless steel plate was inserted in the beam 375 mm from the source in order to filter out most of the primary signal. Both reference and sample signals were captured and digitized using 12 bit resolution and 50 signal averages. Samples were approximately 350 mm x 350 mm x 25 mm in size and were oriented normal to the axis of the projector.

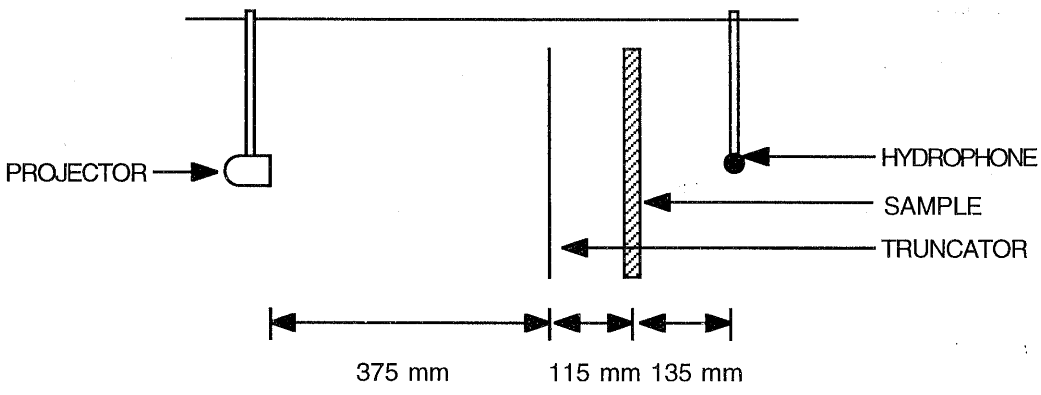

Figure 5. Schematic diagram of insertion loss experiment illustrating relative positions of parametric array projector, truncating plate (acoustic filter), sample, and hydrophone.

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Figure 6 shows the time domain data for a 10% PVC blend sample pulse and a reference pulse taken with no sample present. Note the shift of the sample signal relative to the reference signal along the time axis. The time shift, Δt, may be used to calculate an approximate sound speed [8], roughly equal to the group velocity in the sample (c):

$$c = \left[\frac{1}{c_w} - \frac{\Delta t}{d} \right]^{-1} \tag{1}$$

where c_w is the sound speed in water and d is the sample thickness. Figure 7 shows the sound speeds calculated using equation (1) for various compositions of the blend. The sound speed appears to be less sensitive than the storage modulus (Figure 4) to the phase changes occuring in the 30-50% PVC concentration range, although more data points in the 60-100% PVC region would be useful to test whether the curve in Figure 7 is approximately linear or has a minimum as suggested by a cubic spline fit to the data points.

Fast Fourier Transforms (FFT) of the time signals were carried out to obtain the frequency content of the pulses and the insertion loss spectra. The insertion loss is defined here by

$$IL = -20 \log |T| \tag{2}$$

where T is the ratio of the sample and reference signal pressures. Figure 8 shows the experimental insertion loss spectrum for 10% PVC, and a theoretical curve calculated as described below.

A multilayered panel with seven 3.18 mm layers was fabricated. The layers were composed of TPU/PVC blends with blend compositions of 0, 10, 20, 30, 40, 50, and 60% PVC. The experimental insertion loss spectra are shown in Figure 9 for two orientations: one with the 60% PVC side facing the transducer and another for the sample rotated 180°. In theory, these two curves should be identical for infinite sized panels. Differences in the transmission coefficient due to the orientation of a multilayer sample have been explained in terms of finite sample size effects [9], a possibility which cannot be ruled out here.

4. ACOUSTIC SIMULATIONS

In this study, the theoretical framework developed by Brekhovskikh [10] was used to model the acoustic response of single and multiple layers. It follows from equations presented by Mikesa and Behrens [11] that the insertion loss of a single homogeneous layer may be expressed

$$IL = -20 \log \left| \frac{4Ak}{(A+k)^2 \exp(ikd) - (A-k)^2 \exp(-ikd)} \right| \tag{3}$$

$$k = \omega/c - i\alpha \tag{4}$$

$$A = \rho\omega/Z_o \tag{5}$$

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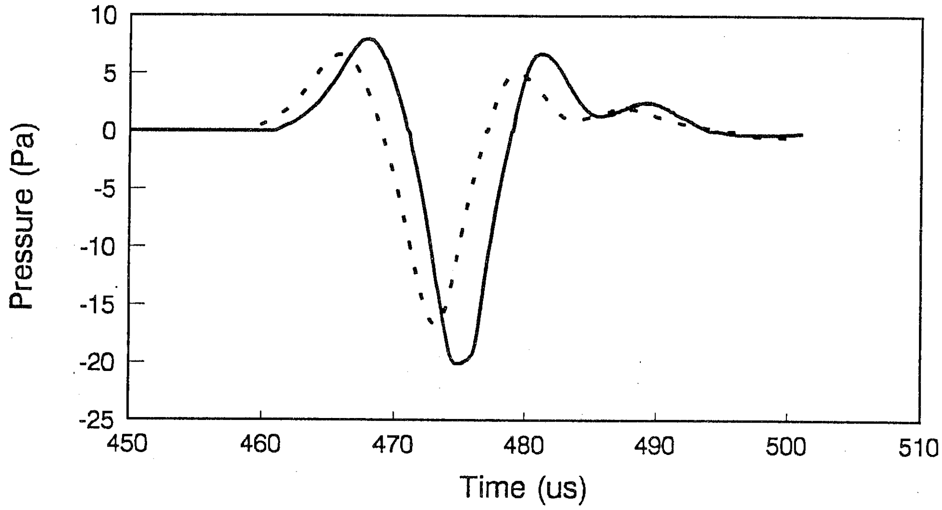

Figure 6. Insertion loss time domain data for reference (———) and 10% PVC blend sample (--------) signals. The polymer sample was 24.4 mm thick.

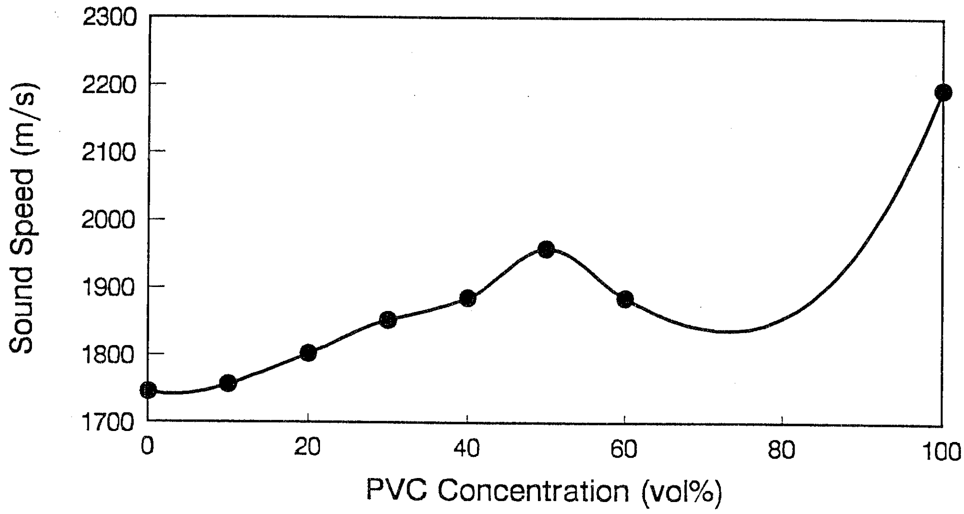

Figure 7. Sound speed calculated from equation (1) versus PVC concentration for TPU/PVC blends. Data points (•), cubic spline fit (———).

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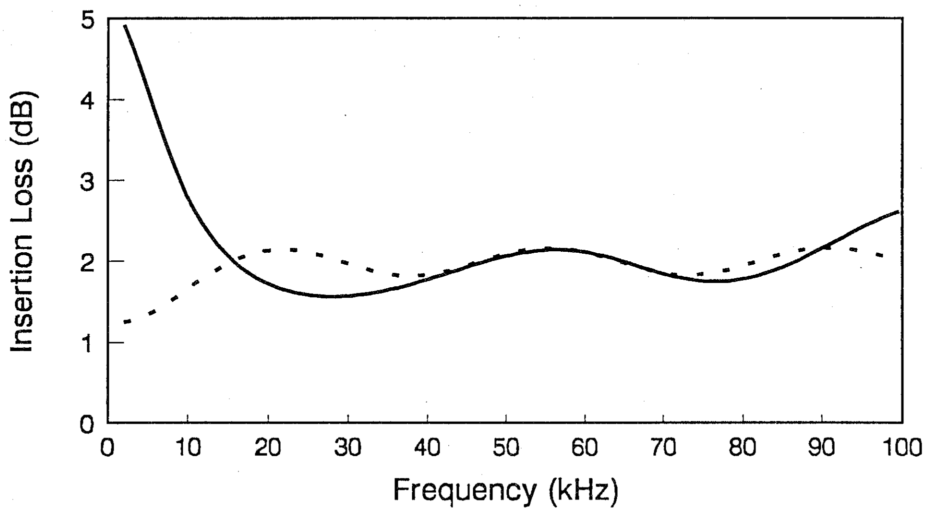

Figure 8. Insertion loss spectra for 10% PVC blend determined by experiment (———) and simulation (--------) using c = 1756 m/s and α = 8.2 Np/m.

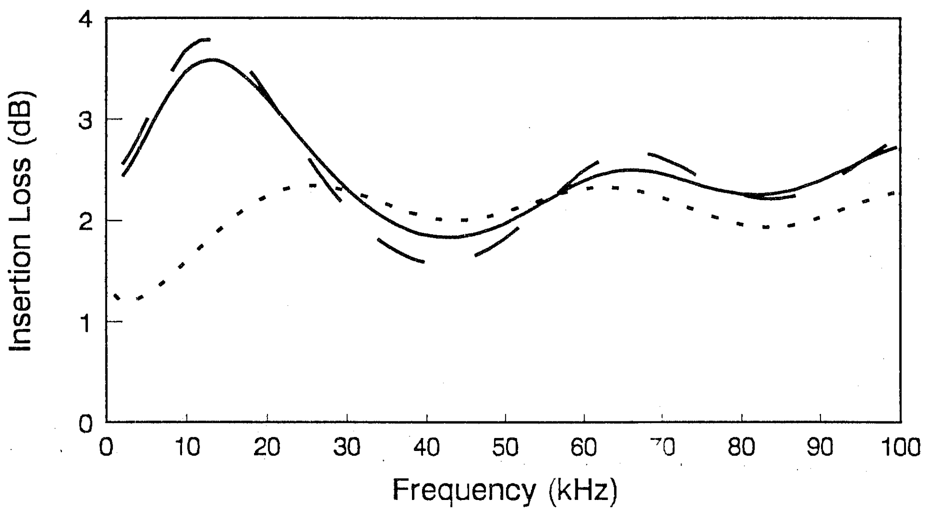

Figure 9. Insertion loss spectra for multilayered sample. The experiment was carried out with the 0% PVC side facing the projector (—— ——) and with the 60% PVC side facing the projector (———). The simulation (--------) was carried out assuming values of c and α shown in Table I.

where k is the complex wavenumber, d is the sample thickness, α and c are the attenuation and sound speed, ω is the angular frequency, and Z_0 is the impedance of water. It is possible to numerically solve for the complex wavenumber in equation (3) at each frequency provided the magnitude and phase of the complex transmission coefficient are accurately known. However, we have found that even a small error in the transmission coefficient will lead to non-convergence or an unacceptable solution (e.g. negative attenuation coefficient). We have therefore found it more useful with experimental data to apply an iterative least-squares method for arriving at values of α and/or c that result in closely matched calculated and experimental insertion loss spectra. For simplicity, we have chosen frequency independent values of α and c, although one could assume a linear or quadratic frequency dependence as well [10].

For each blend, the experimental insertion loss spectrum and sound speed were used to estimate the attenuation coefficient by curve fitting the theoretical to the experimental data. Table I summarizes the densities, sound speeds, and attenuation coefficients for various blends. Figure 8 shows the best fit insertion loss spectrum calculated for a 10% PVC blend using c = 1756 m/s and α = 8.2 Np/m. The agreement between the simulation and experiment was affected by a number of factors, including accuracy of the experimental data (especially at low frequencies where signal strength is low) and the use of frequency independent acoustic properties in the simulations. In addition, the theoretical calculations were based on plane wave theory, which may be inadequate to describe the experimental data for cases in which the transmission coefficient varies rapidly with angle of incidence [5].

Table I. Properties of TPU/PVC Blends.

Blend Composition % PVC	Density, ρ kg/m^3	Sound speed[1], c m/s	Attenuation coefficient[2], α Np/m
0	1165	1745	2.0
10	1185	1756	8.2
20	1205	1802	11.9
30	1215	1852	11.1
40	1220	1885	11.5
50	1235	1959	10.9
60	1257	1885	7.5
100	1320	2195	4.4

[1]Determined from equation (1)
[2]Calculated by curve fitting insertion loss spectra to equation (3)

Figure 10 shows the calculated attenuation coefficients as a function of PVC concentration. The curve has an inverted U shape very similar to the that of the loss factor - PVC concentration curve in Figure 4. The similarity between the two curves may be understood in terms of a simplified relation [12] in which α is directly proportional to the loss factor,

$$\alpha = \frac{\pi}{\lambda} \tan \delta \qquad (6)$$

where λ is the wavelength and tan δ is the material loss factor for longitudinal deformation. Equation (6) would apply only when viscoelastic absorption is the primary mechanism responsible for acoustic attenuation, a case that would seem to apply here.

The non-linear dependence of the attenuation on PVC concentration reflects the changes in the morphology of the two phase system from a TPU continuous phase to a PVC continuous phase, with a region of co-continuity in the 30-50% PVC range [1]. As noted above, these phase changes are also reflected in the storage modulus and loss factor, but not in the sound speed.

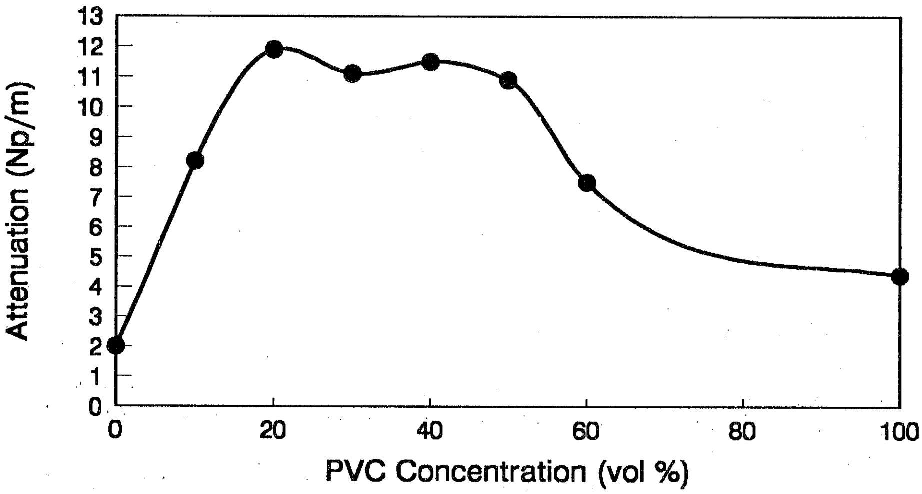

Figure 10. Attenuation coefficients obtained by curve fitting versus PVC concentration for TPU/PVC blends. Data points (•), cubic spline fit (———).

A simulation for the multilayered sample described above was carried out using values for α and c in Table I. The algorithm for simulating multilayers was based on plane wave theory described by Brekhovskikh [10] and Thomson [13,14]. Brekhovskikh uses a matrix approach in his multilayer model. The model can be used for any number of of layers by setting up a matrix for each layer based on its properties, and then multiplying these matrices together to get a final matrix for all thelayers. Each layer is assumed to be elastic, parallel, and isotropic. For the case of normal incidence studied here, the inputed shear properties did not affect the final result. The matrix based algorithm used in these simulations has been described in detail elsewhere [15].

As shown in Figure 9, the agreement between the theoretical and experimental data for the multilayered sample is quite good above 20 kHz, both in terms of the positions of the minima and the absolute values of the insertion loss. Differences between the simulated and experimental curves could be due to any of the factors mentioned above for single layers, as well as uncertainty over the exact thickness and uniformity of the individual layers after molding has taken place.

5. CONCLUSION

The sound speed and attenuation of TPU/PVC blends have been determined by insertion loss experiments using a parametric array source. Curve fitting of theoretical to experimental data was found to be an effective method of determining the attenuation coefficients. The sound speed increased with PVC concentration, while the attenuation went through a maximum at 40% PVC. The concentration dependence of the attenuation paralleled that of the material loss factor-PVC concentration curve.

The insertion loss spectrum of a seven layer TPU/PVC system was determined experimentally and by simulation using plane wave theory. The experimental insertion loss was slightly different when the sample was rotated by 180°, which may indicate an influence of finite sample size. The calculated insertion loss was in satisfactory agreement with the experimental insertion loss from 20 - 100 kHz.

6. REFERENCES

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