ARCHIVES OF ACOUSTIC
16, 3–4, 451–460 (1991)

SOUND VELOCITY AND LOSS FACTOR OF POLYURETHANE COMPOSITES. PART II

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Elastic, non-foamed polyurethane compositions to be used as vibro-insulating. materials have been developed. The compositions were prepared using poly (propyleno-ethers) of different molecular weights, toluyene diisocyanate and aluminum hydrate as the filling agent. The effect of the composition contents on viscoelastic properties that is, on sound velocity and loss factor has been determined. It has been found that various types of the filler permit to modify properties of composite and especially its stiffness and internal damping.

1. Introduction

The production technology and the methods of investigating the basic viscoelastic properties of polyurethane composites were developed when searching for new acoustic materials to be employed for solving vibration isolation problems. Part one of these investigations has been devoted to polyurethane composites filled with microspheres [1].

In that paper the effect of the chemical compositions of the polyurethane binder and the amount of microspheres, acting as the filling agent, upon the elasticity and damping ability of the composition have been investigated, i.e., the sound velocity and loss factor were determined. The both quantities enable the components of Young's complex modulus to be calculated, thus being of fundamental significance for proper choice of vibroinsulating materials.

The aim of the present paper is to determine the correlation between the type and amount of the filler, and sound velocity and loss factor of the polyurethane PU composite. In order to investigate this correlation we prepared a new kind of PU composite filled with aluminium hydrate $Al_2O_3 \cdot 3H_2O$ instead of microspheres [1, 3].

2. Characteristics of the material

Unfoamed PU composition was the investigation object, where aluminum hydrate in the form of powder was used as the filling agent. The PU binder is composed of poly(oxypropylene)trioles and diisocyanates. The introduction of the filler into the polyurethane composition induces the changes in the PU lattice structure, the mobility of the lattice fragments and the increase in the rigidity of the polymer chain. The investigation was carried out on two series of specimens.

In the first series, one of the PU binder components was Rokopol M–12, i.e., poly(oxypropyleneethylene)triole with the molecular weight of 4800, the other was MDI isocyanate (4.4-diisocyanatodiphenylomethane). The ratio of the number of hydroxyl functional group —OH in macrotriole to that of the —NCO group in isocyanate was 1 : 1.3. The amount of aluminum hydrate serving as the filling agent was varied from 10% to 60% relative to the main component of PU binder, i.e., Rokopol M–12.

In the second series, the properties of the PU binder were changed by replacing Rokopol M–12 with Rokopol 111, characterized by molecular weight of 6000. The ratio of the functional groups —OH and —NCO was 1 : 1.5, and the amount of the filler varied from 20% to 60% relative to Rokopol 111. One more specimen with fixed filler content (30%) and functional group ratio —OH : —NCO = 1 : 1.3 was also prepared in order to compare viscoelastic properties of the composite with those ones obtained earlier for PU composite filled with microspheres [1, 2]. Density of the PU composition with aluminum hydrate as the filling agent is varied from 1200 kg/m^3 to 1300 kg/m^3 depending on the amount of the filler.

3. Measurement method

In order to find the sound velocity and loss factor of the PU composite in frequency function the measurement method of vibration transfer function has been used [4, 5]. Figure 1 presents the idea of the measurement.

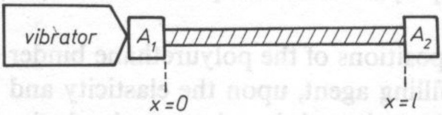

FIG. 1. Rod-like specimen excited to longitudinal vibrations by means of a vibrator

It is assumed that the cross-section of the rod is constant, the relation between stress and strain is linear, lateral motion is not prevented at the rod ends and the lateral dimension is much smaller than the elastic wavelength. The expression for the complex transfer function can be formulated as the ratio of magnitudes of vibration accelerations at both ends of the specimen

$$\mathbf{T} = \frac{A_2}{A_l} = |T| \cdot e^{i\varphi} = \left|\frac{A_2}{A_l}\right| e^{i\varphi} \tag{1}$$

where $|T|$ is the transfer function modulus, A_2 and A_l are vibration accelerations at the end of the specimen, respectively, φ is the transfer function phase and i is the imaginary unit.
The complex transfer function **T** can be expressed by the propagation constant

$$\mathbf{T} = [\operatorname{ch}\gamma l + (M/m)\gamma l \cdot \operatorname{sh}\gamma l]^{-1} \tag{2}$$

where γ is the propagation constant, $\gamma = \alpha + i\beta$, α is the attenuation constant, β is the phase constant, l is the rod length, m is the specimen mass, M is the loaded mass including accelerometer mass.

While measuring the transfer function modulus and the phase for each frequency, the components of propagation constant were found from formula (2) and subsequently, sound velocity c and loss factor η were calculated from well-known formulae [5]

$$C = \frac{2\pi f}{\beta}, \qquad \eta = \frac{2d}{1-d^2} \tag{3, 4}$$

where f is the frequency and $d = \alpha/\beta$.

The measurement error of transfer function modulus $|T|$ was not larger than $\pm 5\%$ and the phase was determined with the error not larger than ± 0.4 deg in the whole range of frequency i.e. from 20 Hz to 1000 Hz. It allowed to find the sound velocity with the error of $\pm 5\%$, and the loss factor with the error of $\pm 10\%$.

In investigations a prismatic shaped specimens of 1 cm^2 cross-section and about 12 cm length were used. The amplitude of the vibration acceleration was adjusted carefully not to exceed the dynamic strain limit of linearity of the investigated material. A detailed description of the measurement system and the method of carrying out the measurements of the transfer function modulus and phase have been presented in [1, 6].

4. Results and discussion

The measurements were made at room temperature in frequency range from 20 Hz to 1000 Hz. The measurement of transfer function modulus for frequencies above 1000 Hz was not possible because of the large damping of the signal in the material investigated. For the upper limit of the measurement range, the ratio of the length of the longitudinal wave to the lateral dimensions of the specimen was properly large which allowed to neglect the influence of transverse vibrations on the calculation of sound velocity and loss factor.

The modulus and phase of the transfer function measured for the first series of specimens and the calculated values of the sound velocity and loss factor have been shown in Figures 2, 3 and 4, respectively.

It is seen from the presented results that the change of the number of aluminum hydrate from 10% to 30% practically does not have any effect on the sound velocity in the composition investigated.

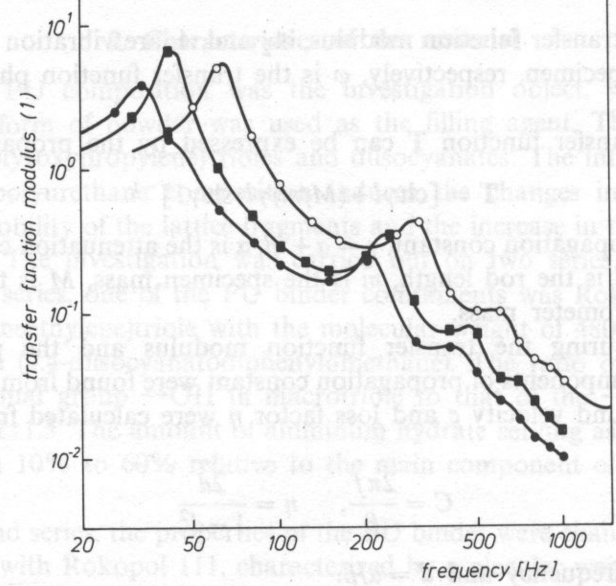

FIG. 2. Measured modulus of the transfer function plotted against frequency for the compositions with Rokopol M–12 and different contents of aluminum hydrate. ■■ 10%, ●● 30%, ○○ 60%. Molar ratio —OH/—NCO = 1 : 1.3

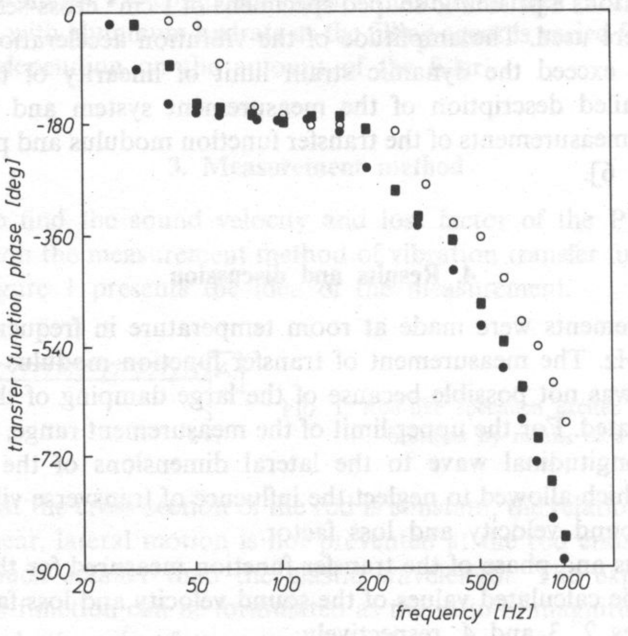

FIG. 3. Measured phase of the transfer function. Further legend as for Fig. 2

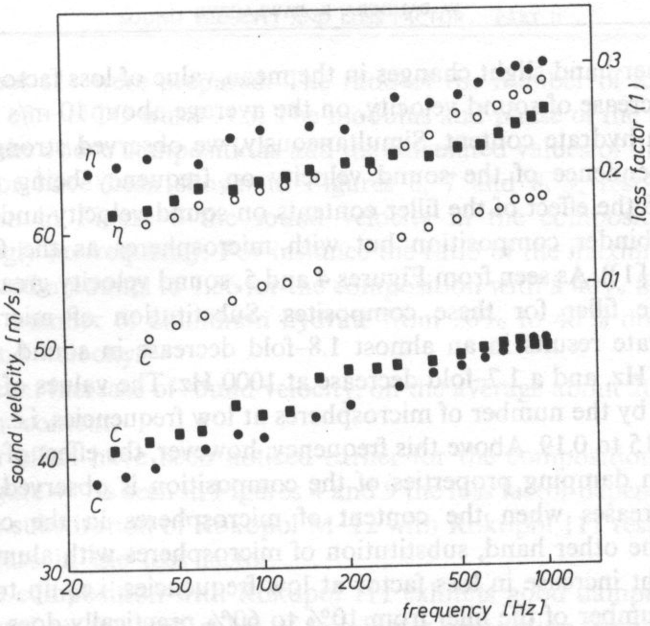

FIG. 4. Sound velocity and loss factor for the compositions with different content of aluminum hydrate. Further legend as for Fig. 2

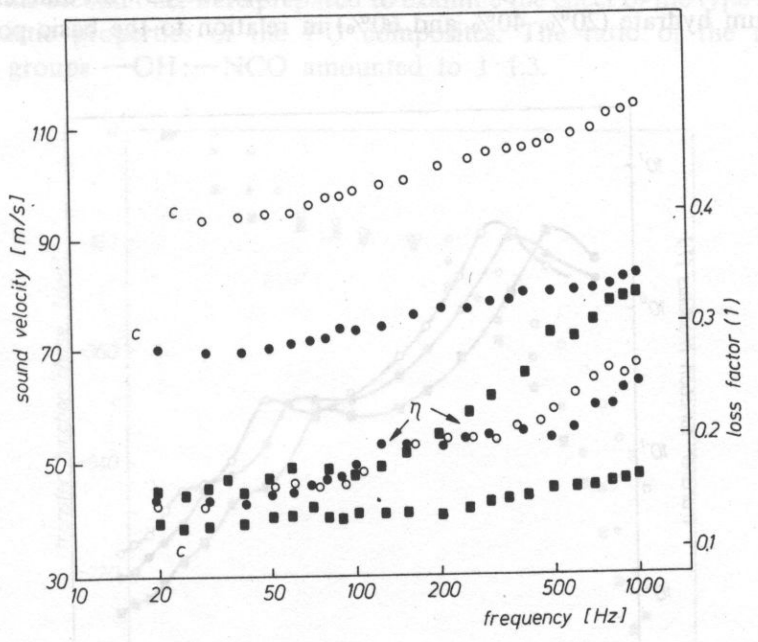

FIG. 5. Effect of microsphere contents on the sound velocity and the loss factor for the compositions with Rokopol M–12. Molar ratio —OH/—NCO = 1 : 1.3 ■■ 10%, ●● 30%, ○○ 60% microsphere content (after [1])

On the other hand slight changes in the mean value of loss factor are observed. A significant increase of sound velocity, on the average about 10 m/s is observed for 60% aluminum hydrate content. Simultaneously, we observed strong dispersion of sound, the dependence of the sound velocity on frequency being almost linear.

In Figure 5 the effect of the filler contents on sound velocity and loss factor for the same PU binder composition but with microspheres as the filling agent is presented (after [1]). As seen from Figures 4 and 5, sound velocity greatly depends on the type of the filler for these composites. Substitution of microspheres with aluminum hydrate results in an almost 1.8–fold decrease in sound velocity at the frequency of 25 Hz, and a 1.7–fold decrease at 1000 Hz. The values of the loss factor are not affected by the number of microspheres at low frequencies, i.e., up to 200 Hz, varying from 0.15 to 0.19. Above this frequency, however, the effect of the number of microspheres on damping properties of the composition is observed. The value of loss factor decreases when the content of microspheres in the composition is increased. On the other hand, substitution of microspheres with aluminum hydrate results in a slight increase in loss factor at low frequencies, i.e., up to 300 Hz. The change of the number of the filler from 10% to 60% practically does not cause any influences of loss factor, however, a slight increase of the loss factor for the composition with 30% content of the filler is observed.

In the next step of investigation, three PU compositions with different contents of aluminum hydrate (20%, 40% and 60%) in relation to the basic polyurethane

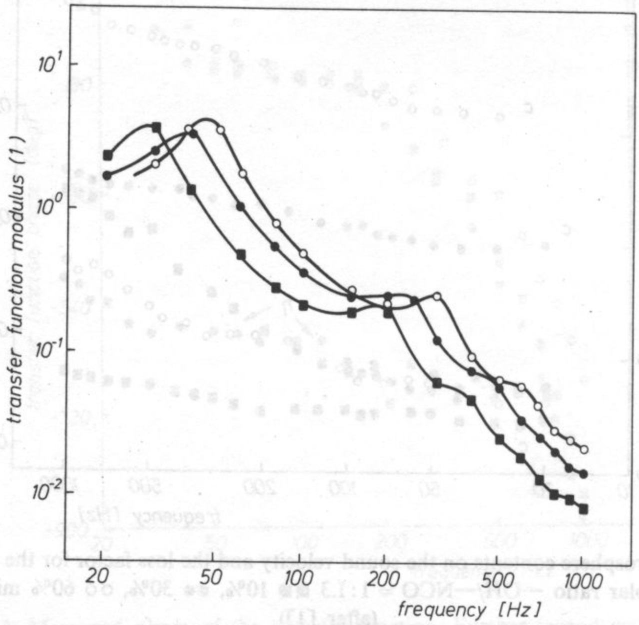

FIG. 6. Measured modulus of the transfer function plotted against frequency for the compositions with Rokopol 111. Molar ratio—OH/—NCO = 1:1.5 ■■ 20%, ●● 40%, ○○ 60% aluminum hydrate content

binder Rokopol 111 were prepared. The ratio of the number of functional groups —OH to that of —NCO was 1:1.5. The modulus and phase of the transfer function measured for the above compositions and the calculated values of the sound velocity and loss factor have been shown in Figures 6, 7 and 8, 9, respectively.

As is seen in Figure 8 the sound velocity in the composition investigated depends strongly on frequency. For instance the ratio of the maximum to minimum velocity of sound amounts to 1.56 for the composition with a 40% filler content. The change of the number of aluminum hydrate from 20% to 40% does not have any effect on sound velocity.

A significant increase of sound velocity, on the average about 20 m/s is observed for 60% filler content.

A similar effect have been noticed earlier for the compositions with Rokopol M–12 (see Figure 4). As seen in Figures 4 and 9 the loss factor depends on the type of Rokopol. The substitution of Rokopol M–12 with Rokopol 111 results in an almost 1.5–fold increase in the loss factor.

Thus, the composition with Rokopol 111 exhibits good damping properties. It has been proved by the high value of loss factor amounting from 0.3 to 0.4 in the investigated frequency range. In the last step of our investigations two compositions with Rokopol 111 and microspheres as the filling agent in the first and aluminum hydrate in the second one, were prepared to examine the effect of the type of the filler on viscoelastic properties of the PU composites. The ratio of the number of functional groups —OH:—NCO amounted to 1:1.3.

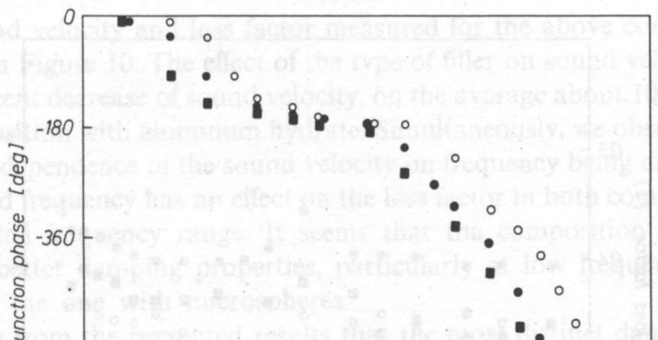

FIG. 7. Measured phase of the transfer function. Further legend as for Fig. 6

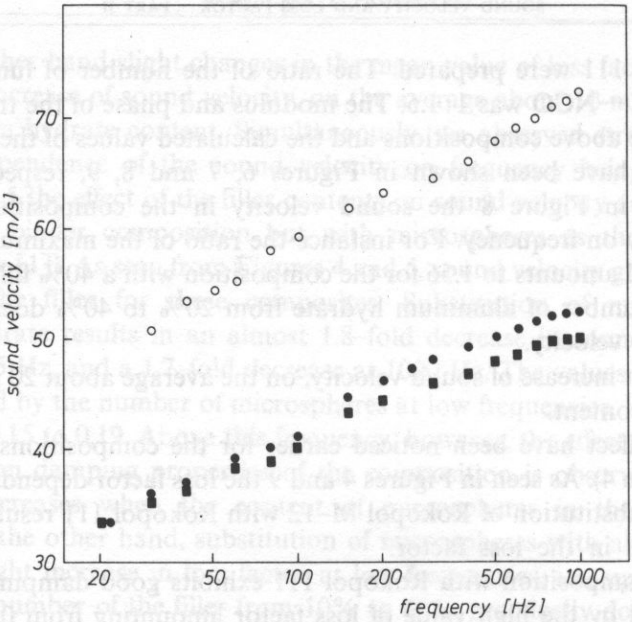

FIG. 8. Sound velocity for the compositions with different content of aluminum hydrate. Further legend as for Fig. 6

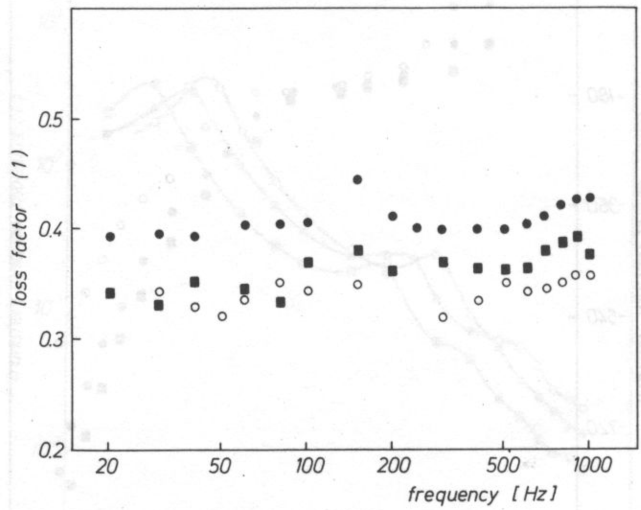

FIG. 9. Loss factor for the compositions with different content of aluminum hydrate. Further legend as for Fig. 6

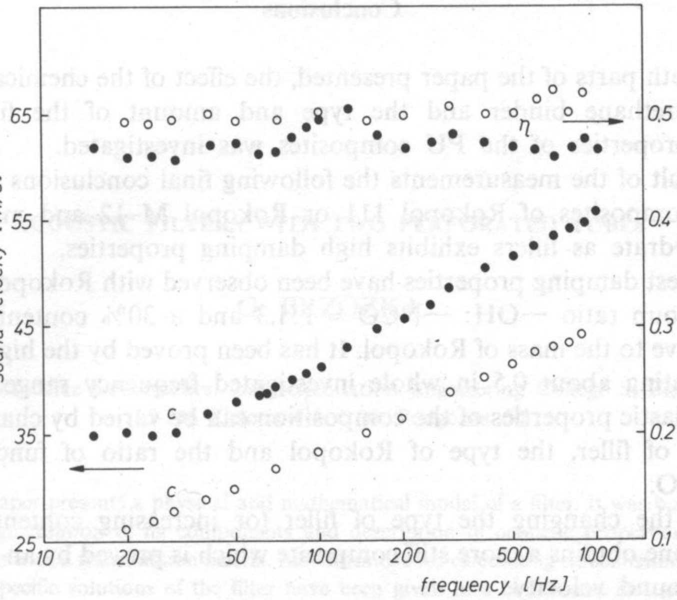

FIG. 10. Effect of the type of filler on the sound velocity and loss factor for the compositions with Rokopol 111 and a 30% filler content. ●● microspheres, ○○ aluminum hydrate. Molar ratio —OH/—NCO = 1:1.3

The sound velocity and loss factor measured for the above compositions have been shown in Figure 10. The effect of the type of filler on sound velocity is evident.

A signifıcent decrease of sound velocity, on the average about 10 m/s is observed for the composition with aluminum hydrate. Simultaneously, we observed dispersion of sound, the dependence of the sound velocity on frequency being almost linear. On the other hand frequency has no effect on the loss factor in both compositions within the investigated frequency range. It seems that the composition with aluminum hydrate has better damping properties, particularly at low frequencies, i.e., up to 100 Hz then the one with microspheres.

It is seen from the presented results that the most distinct damping properties have been observed in case of Rokopol 111, with the functional group ratio —OH/—NCO = 1:1.3 and a 30% content of the filler in relation to the mass of Rokopol. As was stated in a previous paper [1] the effect of the type of Rokopol upon the sound velocity and the loss factor can be explained in terms of the structure of the both composition types. The flexible segments of the composite containing Rokopol 111 are longer than those in the composite with Rokopol M–12 due to the difference in the molecular weights of those polyetherols. In addition, the composite containing Rokopol M–12 is solidified faster and deaerated more readily compared with the compositions with Rokopol 111 which is related with lower viscosity of Rokopol M–12, affecting in turn, the damping properties of the composition.

Conclusions

In the both parts of the paper presented, the effect of the chemical composition of the polyurethane binder and the type and amount of the filler upon the viscoelastic properties of the PU composites was investigated.

As a result of the measurements the following final conclusions can be drawn.

1. PU composites of Rokopol 111 or Rokopol M–12 and microspheres or aluminum hydrate as fillers exhibits high damping properties.

2. The best damping properties have been observed with Rokopol 111, with the functional group ratio —OH: —NCO = 1:1.3 and a 30% content of aluminum hydrate relative to the mass of Rokopol. It has been proved by the high value of loss factor amounting about 0.5 in whole investigated frequency range.

3. The elastic properties of the composition can be varied by changing the type and number of filler, the type of Rokopol and the ratio of functional groups —OH:—NCO.

4. With the changing the type of filler (or increasing contents of filler) in a composite one obtains a more stiff composite which is proved by an almost double increase of sound velocity.

5. It seems that the investigated PU composites could be used as vibroisolating material in constructions of various types.

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