Effects of tensile stress on the hypersonic wave velocities in polymer films

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1. Introduction

Brillouin scattering has been known as a powerful technique to investigate the hypersonic wave properties of thin films. This method also enables the real-time and nondestructive measurement in the GHz range. Krüger has first applied the Brillouin scattering method to observe the anisotropy of highly oriented polymer films [1]. It, however, has not been attempted to measure the successive changes in wave velocities of these films as a function of applied stress.

In this study, we have focused on the real-time measurements of wave velocities in polymer films during a tensile test using the Brillouin scattering method. This effect can be observed as a change of the longitudinal wave velocities in the film. The profile of velocity changes during the test will be discussed together with the mechanical behavior of the film.

2. Experiment

The polymer film samples used are polypropylene (PP) and polyethylene (PE) manufactured by Hanazono Co. Samples of 0.03 mm or 0.1 mm thickness were prepared. These samples were fabricated by uniaxial stretching; however, the anisotropy of the longitudinal wave velocity in the sample was small (less than 2%). In addition, common polymethylmethacrylate (PMMA) sheet (thickness 0.5 mm) was used as an amorphous polymer sample. As shown in Fig. 1, we used the samples of a dumbbell shape for the tensile test, based on the Japanese Industrial Standard (K 7113-1995).

Brillouin scattering measurement is carried out, using a high performance six pass tandem Fabry-Perot spectrometer, with an argon-ion laser at the vacuum wavelength \( \lambda_0 = 514.5 \text{ nm} \). The laser power at the sample is approximately 10 mW, which does not degrade the sample. The experimental system is also shown in Fig. 1. The scanning time of the spectrometer is less than 1 s and the scattered light passed through the spectrometer was detected by the photon counting system made by Hamamatsu Co. The detailed characteristic of the tandem spectrometer is discussed elsewhere [2].

For the Brillouin scattering measurement, 90° A-scattering geometry is used. In this geometry, hypersonic wave velocities are obtained from the equation, \( v = f_\lambda = f\lambda_0/\sqrt{2} \), which shows that the velocity depends only on the Brillouin shift frequency \( f \). We measure the longitudinal wave velocities in the directions of the stress or perpendicular to the stress during the tensile test.

The tensile test was carried out using the tensile test apparatus, EZ-test (supplied by Shimadzu Co.). The cross head speed was 0.5 mm/min.

3. Results and discussion

Figure 2 shows the changes in the stress and longitudinal wave velocities in the directions of the stress (\( q_\theta \)) and perpendicular to the stress (\( q_\phi \)) during the tensile test. The sample used were PP films (thickness: 0.03 mm or 0.1 mm). Velocities are the averaged values obtained from five different samples. The velocity dispersion due to the sample was less than 2%.

As shown in Fig. 2, longitudinal wave velocity in the stress direction shows a small peak in the initial state and almost constant before the slow increase. The longitudinal wave velocity in the direction perpendicular to the stress decreases in the initial state and becomes constant until the sample is ruptured. As for the velocities in the stress direction, the profile in the initial state seemed to be independent of the thickness. The values of strain at the velocity peaks were almost identical. In the region of the slow increase (strain was more than 100%), however, there exists the effect of thickness. Considering that the samples are being strongly drawn in this region, this difference possibly seems to come the crystallinity and structure of the samples. This problem, however, should be discussed carefully with other experimental studies. Velocity profiles in PE films are shown in Fig. 3. These results also indicate the similar velocity peaks in the initial state.

The stress-strain behavior of these samples is known as a typical process of crystalline materials under the tensile stress [3]. The small initial increase in the Hooke’s region finishes at the yield point. Due to the propagation of necking, the stress keeps constant and finally shows gradual increase in the region of ultra-drawing. This is considered as oriented crystallization in the sample. The velocity profile in the stress direction (\( q_\theta \)) was similar with the stress behavior, however, the initial peak and slow increase were found in the earlier state. Because the scattering measurement always treats the small area (less than 1 mm²) of the sample, one can easily find that the longitudinal wave velocity reflects the small-
scale elastic behavior and deformation. The similarity between the velocity and stress behavior does not seem contradictory, considering the basic relation between the elastic constants and wave velocities.

In order to investigate the dependence of the velocity and stress behaviors precisely, we have next investigated amorphous polymers. Figure 4 shows the initial longitudinal wave velocities in a thin PMMA sheet. The velocities in the directions of the stress and perpendicular to the stress were observed during the tensile test. Here, one can easily find that the velocities decreased monotonously as the stress increased, which was very different from the behavior of the above crystalline samples. A similar decrease of the hypersonic wave velocity was also observed in the amorphous epoxy resin layers under the tensile stress [4]. The monotonous decrease of initial velocity is very interesting because it can be found well before the stress peak near the yield point. From the ultrasonic wave measurements in the bulk PMMA sample, Matsushige has reported a similar decrease of longitudinal
wave velocities in the MHz range [5]. He has then pointed out the possible effect of a softening phenomenon caused by the micro cracks in the sample under high stress. In our measurements, we could find white crazing due to necking from the initial state. Following the Matsushige’s softening idea, it implies that very small cracks have already generated in the sample from the initial state of the tensile test.

4. Conclusion

Brillouin scattering measurements have enabled the real-time measurement of longitudinal wave velocity. In the crystalline PE and PP films, the initial velocity profile during the tensile test was similar with the stress behavior, reflecting changes in the elastic properties of the small measured area. From the measurement of a PMMA sheet, the possibility of the evaluation of micro cracks was suggested. These results should be discussed carefully with other experimental studies of the sample. They, however, show the future possibility of Brillouin scattering as a strong tool for the measurements of stress and deformation.

References