Simultaneous Measurements of Stress and Birefringence Development during Extensional Deformation of Cyclic Olefin Copolymer

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Introduction

Market for the flat panel displays is expanding in these years, and therefore research activities for the development of optical parts such as polarizing filters and phase-retarder films for better quality of display is extremely high. As polymeric materials for the optical use, cyclic olefin polymers (COPs) and cyclic olefin copolymers (COCs) attract much attention because of their low intrinsic birefringence, high transparency, low moisture absorption and high glass transition temperature. For the development of optical devices from these polymers, effect of processing conditions on the development of molecular orientation and anisotropic optical properties are important. Therefore, to clarify the mechanism of structure development of cyclic olefin copolymers (COCs) in stretching process, simultaneous on-line measurements of optical retardation and tensile force during elongation and relaxation processes of films were performed in this study.

Experimental

The Cyclic olefin copolymer (COC) with the glass transition temperature (Tg) of 150°C (Polyplastics, Topas 6015) was used in this study. The polymer was compression molded to form films. The film of 25 x 7 mm was stretched at three different elongational strain rates and subsequently relaxed after the application of a pre-determined level of stretching. Variations of tensile force and optical retardation were measured simultaneously during the elongation and relaxation processes. Schematic diagram of the optical system for the measurement of retardation is shown in Fig. 1. The optical system was consisted of a laser light source, polarizing filters, quarter wave plates, a rotating polarizing filter etc. Changes in the cross-sectional area and thickness of the film were estimated from the measured length and width assuming the constant volume. Subsequently, tensile stress and birefringence were evaluated from tensile force and optical retardation. The COC fibers were also prepared by the melt spinning and drawing processes, and the changes of tensile force during the heating process was investigated.

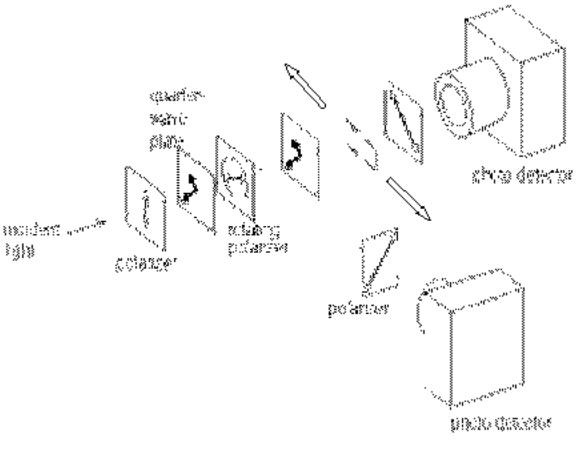

Figure 1. Schematic diagram of optical system for on-line retardation measurement used for the analysis of birefringence development during stretching and relaxation processes of COC films.

Results and discussion

Typical example of the result of measurement for the variations of stress and birefringence with time is shown in Fig.2. At the drawing temperature of 160 °C, stress showed yielding when the tensile strain reached about 5 % whereas birefringence showed continuous increase. It should be noted that the films were stretched uniformly even though strong yielding was observed. At the drawing temperature of 170 °C, both stress and birefringence increased with an increase in the tensile strain. At 165 °C, tensile behavior was similar to that of 160 °C at a high strain rate, whereas the behavior resembled the one observed at 170 °C when the tensile strain rate was low. In the relaxation stage after the cessation of stretching, both stress and birefringence decreased monotonously at all the temperatures.

Stress versus birefringence relations during the stretching and relaxation processes were plotted in Fig.3. The slope at the initial stage of stretching observed at a temperature of 160 °C was small (= 0.1 GPa^{-1}). This result indicated that the material behaved like a solid. In this case, birefringence increased steeply during the yielding of the stress. In the process of stress relaxation, strong hysteresis with respect to the stretching process was observed. The slope for the relation between stress and birefringence was much larger than that in the stretching stage. In other words, the material exhibited liquid-like behavior. The stress-optical coefficient estimated from the end part of the stress relaxation process was 1.2 GP^{-1}. When the stretching temperature was increased to 170 °C, in both stretching and relaxation processes, linear relation was observed for the relation between stress and birefringence, and the slope was about 1.2 GPa^{-1}.

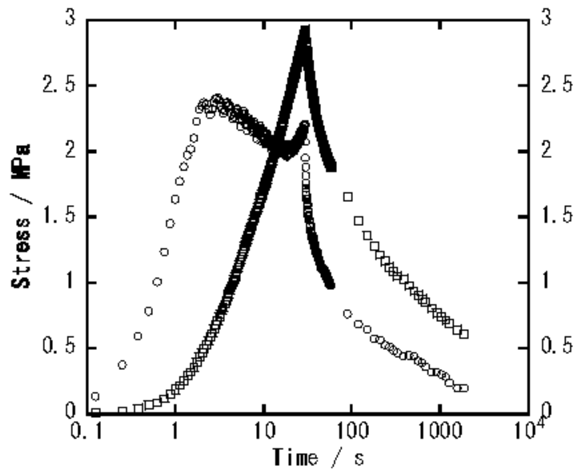

Figure 2 Typical example of stress versus time and birefringence versus time relations measured at 160 °C. Film sample was stretched at strain rate of 1 s^{-1} up to the strain of 0.5, and then relaxed by stopping the stretching.

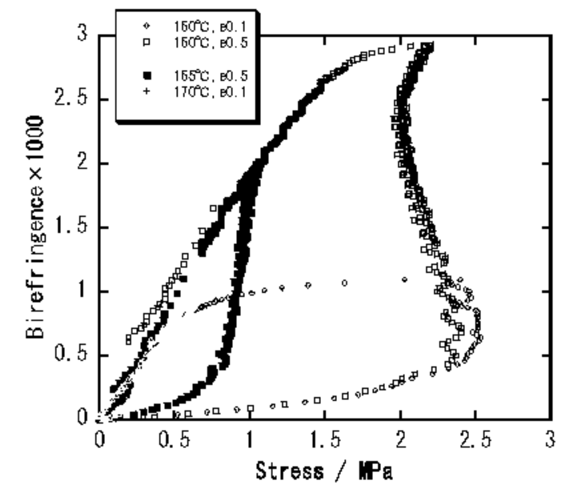

Figure 3 Birefringence versus stress relation during stretching and relaxation processes. Stretching temperature and maximum strain are indicated in the figure.