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Thermo-mechanical coupling and self-excited oscillation in the neck propagation of PET films

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Abstract

The self-excited oscillation of neck propagation during cold drawing of polymer films has been examined experimentally. On the basis of Barenblatt's model considering a thermo-mechanical coupling at the neck, the temperature rise at the neck has been studied with an infrared camera. The temperature began to rise in a range showing a negative velocity dependence of the applied load. The behavior is consistent with the view of thermo-mechanical coupling. The temperature rise was up to 80°C ($>T_g$) and explains the occurrence of crystallization for faster drawing rates. It has also been confirmed that the temperature rise follows the oscillation of stress due to the coupling. © 2001 Published by Elsevier Science Ltd.

Keywords: Poly(ethylene terephthalate); Necking; Self-excited oscillation

1. Introduction

Self-excited oscillation is a typical example of dissipative structures seen in nonlinear dynamics [1]. In many occasions of polymer processing, we encounter examples of self-excited oscillation, e.g. in neck-propagation during cold drawing of polymer films [2], in the extrusion of polymer melt from a die [3,4], in the process of spinning known as draw resonance [5], during tear of rubber [6], and in the peeling of pressure sensitive adhesives [7]. The mechanisms of the processes are therefore crucial for practical purposes as well as scientific interests.

In our previous papers [8,9], we have reported the experimental and numerical calculation results on the oscillation in the cold drawing of poly(ethylene terephthalate), PET, film, and concluded that the process is successfully explained as a thermo-mechanical coupling in the neck region. In the modeling, the beginning of oscillation is characterized as the Hopf bifurcation of a stable steady state to a limit cycle, which can be expected for a nonlinear dynamical system.

In this paper, we discuss the temperature rise in the neck region, which is a necessary prerequisite of the modeling based on the thermo-mechanical coupling. The temperature rise at the neck region has been studied mainly by two methods; one is a calorimetric method [10,11] to measure the total heat generated during necking and the other method is to measure the temperature rise directly by an infrared camera [12–15]. Both the methods indicate appreciable increase in temperature; in some cases, it has been reported that temperature rises up to 90°C from room temperature [12]. Andriavova et al. [2] estimated the temperature rise by observing the melting of colored organic crystals dusted onto the surface of PET film and observed an oscillatory heat generation by a calorimetric method [11]. In recent years, with availability of infrared camera, it is possible to monitor the change in temperature profile on the film surface as a function of time during self-excited oscillation. We report the direct measurement of the temperature at the neck region by an infrared camera in the course of self-excited oscillation and confirm the thermo-mechanical coupling.

In the following, we first review the characteristics of the oscillation process and the modeling proposed by Barenblatt [16]. Experimental results are discussed subsequently.

1.1. Characteristics of self-excited oscillation in polymeric systems

The oscillation behaviors seen in some of the above

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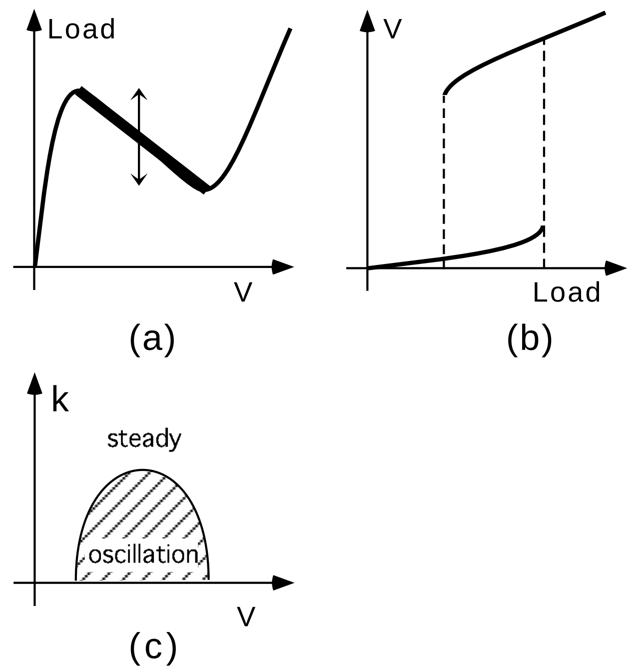

Fig. 1. Schematic plots of deformation rate, V, vs. load at constant speed (a) and at constant load (b). Kinetic phase diagram in (c) shows the regions of steady deformation and self-excited oscillation in the map of the stiffness of the whole system, k, and deformation rate, V.

mentioned systems have the following common features (Fig. 1), which is also common in nonlinear dynamical systems.

(i) When we plot the load required for the constant speed of deformation, the plot shows an N-shaped dependence on speed, which means that the load becomes smaller for faster deformation rate in a certain velocity range: Fig. 1a.
(ii) When we see the dependence in a different way for the deformation with constant load instead of constant speed, there exists bistable deformation speeds for a single value of load, which consequently introduces a hysteresis: Fig. 1b.
(iii) Self-excited oscillation mainly appears in the velocity range where load becomes smaller for faster deformation speed: Fig. 1a. The stiffness of the whole system must be soft enough to start the oscillation: Fig. 1c.

1.2. Barenblatt's model based on thermo-mechanical coupling

Neck is formed during cold drawing of polymer films and propagates along the draw direction with chain deformation localized at the neck. The propagation becomes unstable in some polymers and shows a self-excited oscillation in load under constant speed of drawing in the velocity range with negative dependence of applied load on the drawing speed (Fig. 2). The drawn film shows a periodic appearance of transparent and opaque parts, which indicates crystallization in the opaque part during oscillation. The period of oscillation sometimes becomes doubled, and hence chaotic behavior after consecutive periodic doubling is expected with a controlled experiment.

Based on a thermo-mechanical coupling at the neck, Barenblatt proposed the modeling of a nonlinear dynamical system. Thermo-mechanical coupling means that the work done on drawing is converted into heat. Since the deformation occurs locally at the neck, the heat increases the temperature at the neck. Barenblatt's model correlates the temperature rise to the oscillation. The modeling is based on the following differential equations.

(i) It is empirically known that the following equation satisfactorily describes the relationship among applied load (stress σ), neck propagation velocity, v, and temperature at the neck, T

$$v = v_0 \exp\left(-\frac{\Delta F}{kT}\right)\exp\left(\frac{\alpha\sigma}{kT}\right) \tag{1}$$

where ΔF represents activation free energy and α is the activation volume [17,18]. It means that the dynamics of neck propagation occurs on a two-dimensional surface in the three-dimensional space of σ, v and T.
(ii) The heat balance at the neck is represented as

$$A\frac{dT}{dt} + Bv(T - T_0) = -C(T - T_0) + D\sigma v \tag{2}$$

where A, B, C and D are physical constants and T_0 is the ambient temperature. In Eq. (2), the second term in LHS represents the heat loss from the neck due to neck propagation, the first term in RHS represents the heat transfer to the surroundings, and the second term represents the heating by the work done at the neck.
(iii) In terms of the relationship between the neck propagation velocity, v, and the drawing velocity, V, neck propagation velocity can be varied even under the condition of constant speed of V because of the elastic

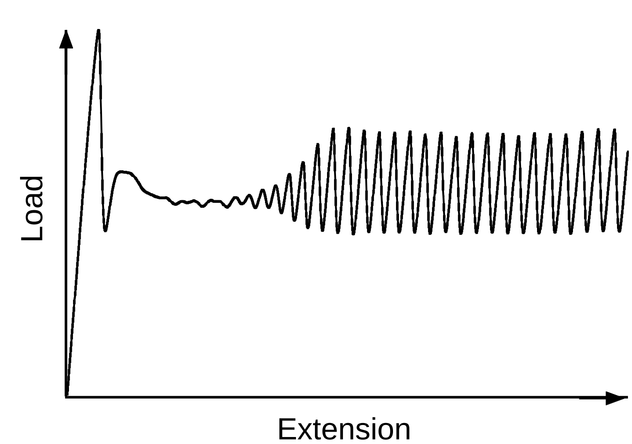

Fig. 2. Typical load–extension curve for oscillatory neck propagation in a polymer film.

deformation of the whole system

$$v + \frac{1}{k}\frac{d\sigma}{dt} = V\ (\text{constant}) \quad (3)$$

where k represents the stiffness of the whole system.

The steady state solution of these equations gives an N-shaped dependence of the applied stress on drawing velocity [8]. The negative slope is explained as a consequence of softening of material due to temperature rise at fast drawing [19]. The positive slope at still faster drawing is due to the leveling off of temperature rise limited by the heat loss in the second term of Eq. (2). Barenblatt has suggested that the dynamical system of Eqs. (1)–(3) undergoes Hopf bifurcation from the stable steady state to a limit cycle; the limit cycle means a periodic oscillation in applied load and neck propagation velocity. This instability is qualitatively explained as follows. An excess temperature rise due to fluctuation causes more softening and consequently faster propagation. Then, if the whole system is soft enough to have the response of stress with retardation, the heat produced by the second term in RHS of Eq. (2) increases, and consequently temperature rises further. Following this explanation, it will be understandable that the stiffness of the whole system, k, determines the stability, and the stiffness must be smaller than a critical value. The instability occurs only in the velocity range with negative slope in the plot of applied force against deformation speed, where faster propagation results in the increase in temperature and consequently more softening.

In the modeling, temperature rise at the neck plays an essential role. In the present paper, we will experimentally confirm the evidences. First, the relationship between the neck propagation velocity and the temperature is examined by the isothermal experiments of films immersed in a thermostated bath. Secondly, the temperature rise at the neck is directly examined by an infrared video camera monitoring the surface temperature profile in the air. Then, the correlation of the degree of temperature rise with drawing speed in the velocity range of negative slope will be examined. The confirmation of the oscillation in temperature is also the subject of the present paper.

2. Experimental

The samples used were amorphous PET films made by TOYOBO; the film thickness were in the range 0.3–0.6 mm. The film was cut into rectangular strips of 3–10 mm in width and 150 mm gap length. The samples were examined with a Shimazu Auto Graph S-100 or AG-S testing machines. For the isothermal experiments, films were immersed in a silicone oil bath; temperature control of the bath was within ±0.1°C and the development of extension was monitored with constant load by a laser displacement sensor (LB-60, Keyence). For the visualization of temperature profile on the film surface, an infrared video camera (Thermotracer TH1102, NEC) was employed; the detector made of InSb was cooled down by liquid nitrogen and scanned the view to measure the infrared in the wavelength 3–5.3 μm.

3. Results and discussion

Fig. 3 shows the plots of applied load against drawing speed at different temperatures under isothermal conditions. This figure clearly confirms the relationship expected with Eq. (1).

Fig. 4 shows the view of temperature profile on the surface of a film during neck propagation. Temperature rise is clearly seen and the temperature at the neck increases above the glass transition temperature around 67°C. It has been well known that fast drawing induces crystallization of PET film. It will be now obvious that the temperature rise is an important factor in crystallization.

The degree of temperature rise at the neck as well as applied load has been plotted against drawing speed in Fig. 5. As we have expected from thermo-mechanical coupling, steep increase in temperature occurs in the

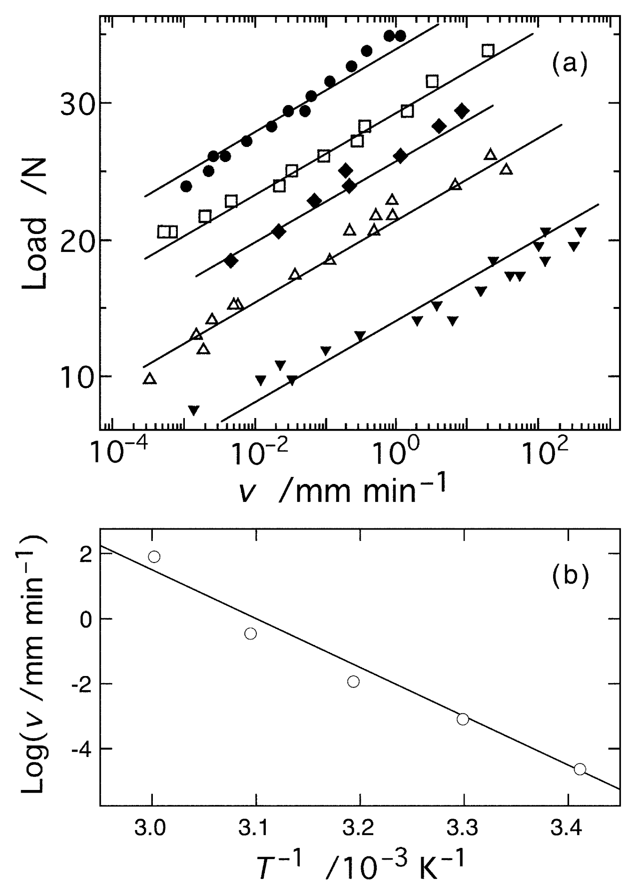

Fig. 3. (a) Plots of applied load against neck propagation velocity at different temperatures under isothermal conditions: ● 20, □ 30, ◆ 40, △ 50, ▼ 60°C. The films were 0.3 mm thick and 3.0 mm in width. (b) Temperature dependence of the neck propagation velocity shown in (a) at load = 20 N.

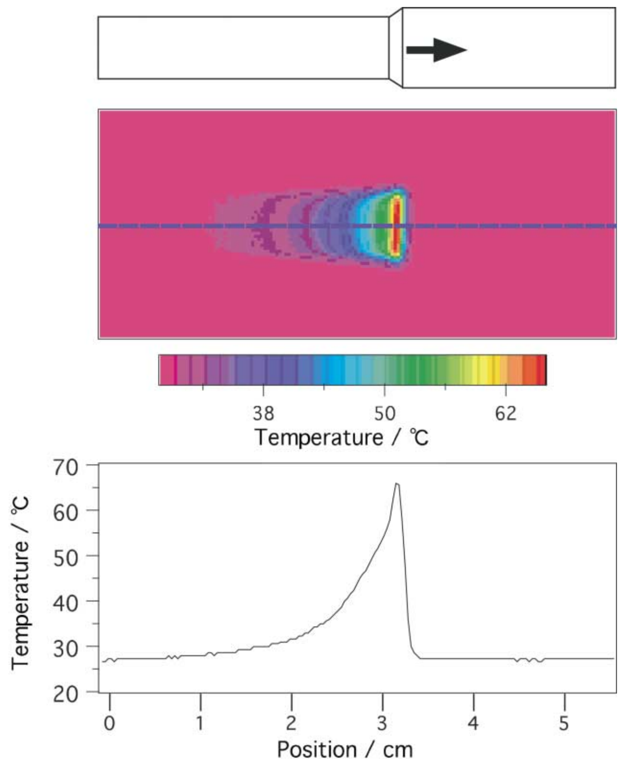

Fig. 4. The view of temperature profile on the surface of a film (0.6 mm in thickness and 10 mm in width) during neck propagation at $V = 100$ mm min^{-1}. The arrow indicates the direction of neck propagation.

velocity range with negative slope of applied load. We will be able to conclude from this result that the negative slope is due to the softening of film on temperature rise.

Fig. 6 shows the oscillation behavior of temperature rise during the stress oscillation. The time development of temperature profile clearly shows the oscillation in temperature. A simultaneous measurement of temperature and stress oscillation is shown in Fig. 7. Here, based on Eq. (3), the time derivative in Fig. 7b represents the propagation velocity of neck. The temperature change is in phase with the propagation velocity, and hence the changes in temperature and propagation velocity are mutually correlated with each other.

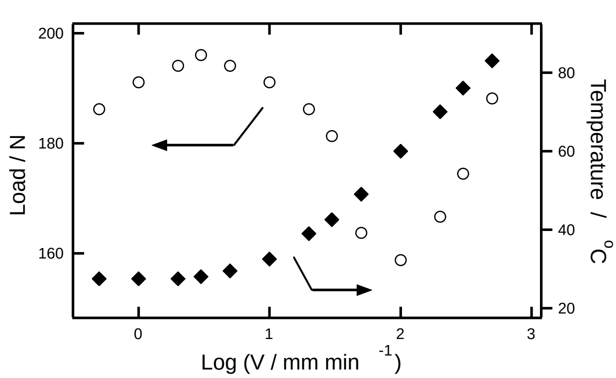

Fig. 5. Plots of applied load and temperature at the neck against drawing velocity. The films were 0.6 mm thick and 10.0 mm in width.

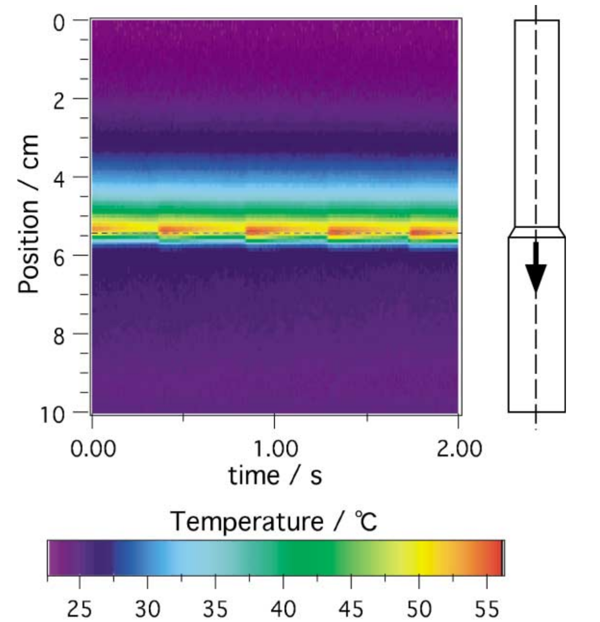

Fig. 6. The view of oscillation behavior of temperature rise during the stress oscillation in a film (0.4 mm in thickness and 10 mm in width) with drawing at $V = 100$ mm min^{-1}. The change in temperature profile along the broken line is shown. The arrow indicates the direction of neck propagation. The horizontal line is for indicating the moving down of the neck region.

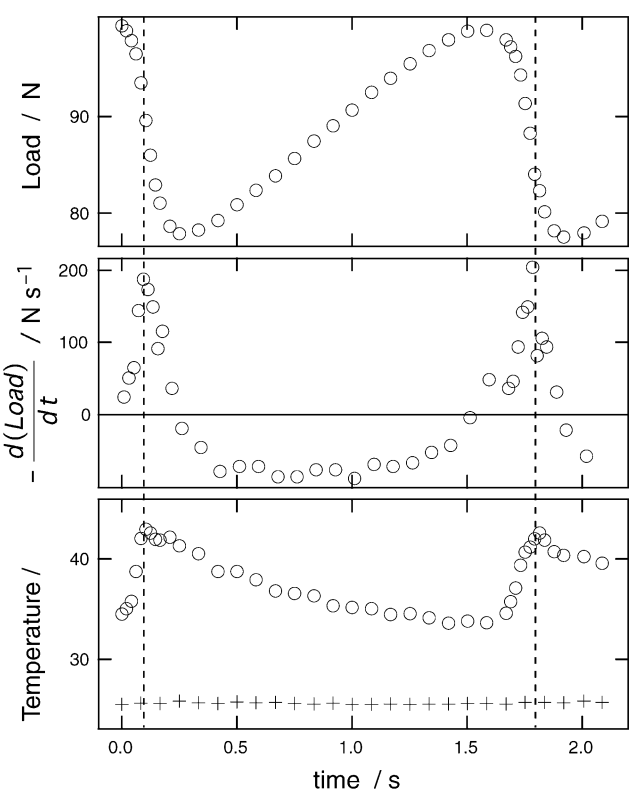

Fig. 7. Simultaneous measurement of oscillation in load (a) and temperature (c) in a film (0.3 mm in thickness and 10 mm in width) with drawing at $V = 50$ mm min^{-1}. In (b), the time derivative of load represents the propagation velocity of neck, based on Eq. (3). The symbol + in (c) represents ambient temperature.

From those experimental evidences, it is confirmed that the self-excited oscillation in the neck propagation of PET film is caused by the instability of steady propagation because of the temperature rise at the neck. Those evidences strongly support the mechanism of thermo-mechanical coupling, on the basis of which Barenblatt's model has been constructed. Here, it should be mentioned that the model has a limit in its applicability to the actual oscillation process: i.e. we know that oscillation can occur in the velocity range of non-negative slope in the plot such as the one shown in Fig. 1a, the amplitude of stress oscillation depends on drawing velocity, oscillation period can be doubled under certain conditions. All these features cannot be expected by the model having freedom only in two-dimensions, and should be accounted for with the introduction of other parameters such as temperature profile in the film, as has been proposed in our previous paper [9]. In order to examine the possibilities, numerical calculations based on a finite element method with the consideration of temperature profile is in progress.

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