

Observation of Polymer Film Drawing by Use of Thermography. An Introductory Investigation on the Thermodynamics

Tatsuo Yamauchi

Division of Forest and Biomaterials Science, Graduate School of Agriculture, Kyoto University, Sakyo-ku, Kyoto 606-8502, Japan

Received 20 May 2005; accepted 24 October 2005

DOI 10.1002/app.23784

Published online 9 February 2006 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The local heat generation that accompanied tensile drawing of polymer films was successfully observed by infrared thermography as a series of temperature distribution images. These images showed that the high temperature part where rearrangements of molecular orientation and microcrystallines occurred was located mainly at the end of the necked part for the polymers that exhibited necking or appeared uniformly throughout films for the

polymers that did not exhibit necking. The generated heat could be roughly estimated, and the relation between the estimated heat and mechanical work led to the thermodynamic investigation. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 100: 2895–2900, 2006

Key words: polymer film; drawing; necking; thermodynamics; thermography

INTRODUCTION

Polymer films are quite often stretched in their final usage, while the drawn films are also employed for many purposes because of the improvement of the physical properties by their drawing. On stretching, many polymer films, at first, homogeneously extend with an increase in tensile load and then suddenly become thinner at one point by a process known as “necking” just after the yielding point.¹ Necking is an irreversible deformation and is postulated to occur adiabatically with rearrangement of molecular orientation; the energy expended appears as heat, causing a rise in temperature.² However, few reports have been published on these points.³ The heat generation under deformation for other materials has been estimated using various techniques and ideas.⁴ However, the exchange rate from mechanical work to the estimated heat in the literatures showed considerable variation.³ The method based on thermography was first applied to a notched sheet of paper under deformation^{5,6} to show stress concentration through the temperature distribution images. The heat generation was estimated using assumptions of the heat capacity and emissivity. A value of 55% for the exchange rate from mechanical work done to the estimated heat was obtained for paper made from beaten pulp.⁶ In this arti-

cle, some representative polymer films are rapidly stretched and the temperature profiles of the surface and the heat generation under the tensile drawing process, including necking, are examined to show the process of drawing as a series of temperature distribution images and to reveal the possibility of a thermodynamic investigation on polymer drawing by the use of thermography.

EXPERIMENTAL

Materials

Commercially available films of polyethylene terephthalate (PET), polyvinyl chloride (PVC), polypropylene (PP), and polyethylene (PE) were selected as the representatives of films not exhibiting necking (PET and PVC) and those exhibiting necking (PP and PE). The thicknesses of these films were 0.06, 0.09, 0.20, and 0.05 mm, respectively.

Instrumentation

The experimental arrangement for tensile drawing and concurrent thermography observation has been described in the previous reports.^{5,6} The following is a brief description of the method. The specimen of 15 mm in width and 70 mm in length was clamped to an Instron-type tensile testing machine (Shimadzu Autograph AGS-100) with a span distance of 50 mm. The specimen was drawn at a crosshead speed of 500 mm/min under the standard atmosphere (20°C 65%

Correspondence to: T. Yamauchi (yamauchi@kais.kyoto-u.ac.jp).

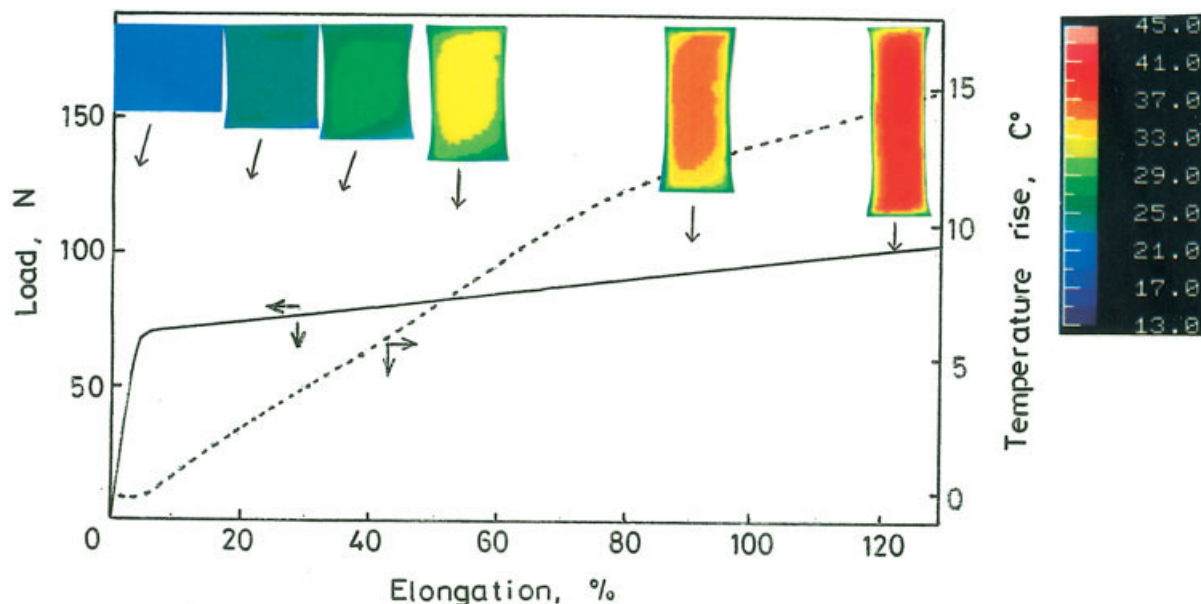


Figure 1 Relationships for load versus elongation and temperature rise versus elongation and some selected temperature distribution images from the drawing of PET film. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

RH). The extreme high speed of the crosshead was expected to provide a nearly adiabatic drawing.³

The infrared thermal imaging system used here was an Infrared Thermo-Tracer 6T62 (NEC San-ei Instruments, Ltd., Kodaira, Tokyo, Japan). The temperature determination was made, assuming that the specimen was a perfect black body, its emissivity being 1.0. In practice, the emissivity of the polymer was considered to be about 0.9.⁷ The errors arising from the assumption are small and are generally less than the errors in the measurement system.⁶ They were therefore neglected in this work. The surface temperature distribution image of the specimen was obtained at a rate of 4 images/s with a temperature sensitivity of 4, 0.3, 4, and 0.4°C, respectively, for PET, PVC, PP, and PE. Because of the large stretching up to 130% elongation, all temperature distribution images of the specimen were recorded as contracted to one fourth in length. Heat emission from the background can affect the temperature images, since all polymer films are to some extent infrared transparent. However, tensile drawing was carried out about 50 cm apart from the wall whose temperature was the same as that of the sample before drawing. Therefore, the effect of background was negligible, unless the sensitivity was very high. Actually, a shadow of the background marginally appeared on the temperature distribution image as the sensitivity increased to 0.2°C and disappeared at the sensitivity of 0.3°C. By means of an image processing system for the thermography, a rectangular area was defined within the drawn specimens and an average temperature within the area was provided for PET, PVC, and PE before necking.

RESULTS AND DISCUSSION

Tensile drawing not accompanied by necking

An example of the relationships for load versus elongation and average temperature rise versus elongation for PET film, along with selected temperature distribution images, is shown in Figure 1 as an example of film drawing not accompanied by necking. At the beginning of the drawing, the elastic deformation was accompanied by a steep increase in load and a very small decrease in temperature of $\sim 0.25^\circ\text{C}$, which is consistent with Kelvin's thermoelastic theory.^{5,6} The load gradually increased with an increase in elongation during the plastic deformation period after the elastic deformation. A nearly even temperature distribution throughout the specimen except for the clamping zone was observed throughout the entire drawing period. This corresponded to homogeneous deformation. The average temperature rise gradually increased during the plastic deformation period, probably because of the accumulation of the generated heat. Since heat was generated uniformly throughout the specimen, heat generation within a selected period was estimated from the rise of the average temperature and the mass of the specimen, using an assumed heat capacity of 1.26 J/g K.⁸ For example, the temperature rise within 0.25 s at $\sim 85\%$ elongation was 0.5°C and the estimated heat generation was 36 mJ. On the other hand, the mechanical work done during the period was estimated to be 180 mJ from the average force of elongation (90 N) and the amount of elongation during 0.25 s (2 mm). The heat generation tended to decrease with an increase of elongation while the

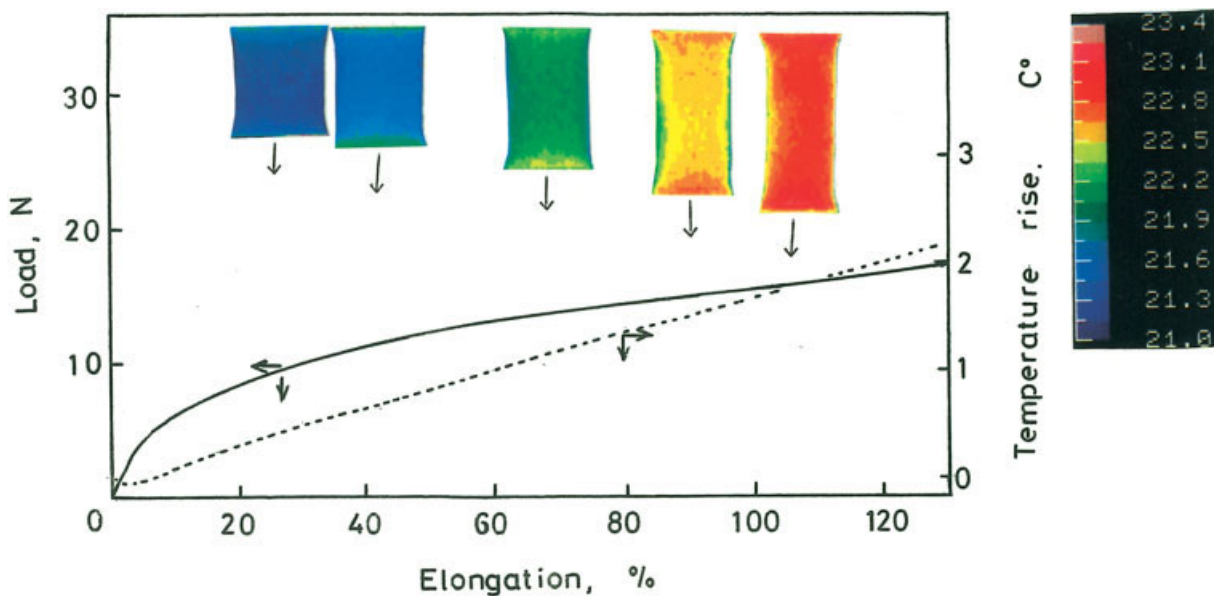


Figure 2 Relationships for load *versus* elongation and temperature rise *versus* elongation and some selected temperature distribution images from the drawing of PVC film. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

mechanical work tended to increase. Eventually, the exchange rate from mechanical work to the heat decreased with an increase of elongation from 36% at 30% elongation to 14% at 120% elongation.

The result for PVC film is shown in Figure 2 as another example. Although a mild degree of stress concentration was observed at the top and bottom ends of the specimen, as indicated by the higher temperature part in yellow in the middle stage of elongation, a nearly even temperature distribution throughout the specimen was observed throughout most of the drawing period. Thus, the heat changes occurred uniformly throughout the PVC film. After a tiny decrease in the temperature of 0.1°C, consistent with Kelvin's theory, during the elastic deformation period, the average temperature throughout the specimen increased during the entire elongation period. Compared with the temperature increase for PET, the extent of temperature increase was very small. Heat generation and further the exchange rate from mechanical work to heat were also estimated with an assumed heat capacity of 1.26 J/g K.⁸ As a result, the generated heat was 8.9 mJ within 0.25 s at 50% elongation and the exchange rate increased from 27% at 30% elongation to 36% at 50% elongation and then gradually decreased to 13% at 120% elongation. Since entropic heat generation accompanying molecular orientation is very small,⁹ most of the generated heat must arise from a rearrangement of molecular or crystal structure. PET is actually a crystalline polymer and its drawing causes crystal orientation and crystallization, as well. The latter generates heat. These are irreversible processes, and thus a large portion of the

mechanical work was converted to heat. On the other hand, PVC film generally includes a considerable amount of plasticizer and the degree of crystallinity is very low. Thus, the majority of the deformation process is reversible. Therefore, the generated heat from irreversible rearrangement of molecular structure in PVC is relatively low.

Tensile drawing accompanied by necking

An example of the relationships for load versus elongation and average temperature rise versus elongation for PP film, along with selected temperature distribution images, is shown in Figure 3(a) as an example of film drawing accompanied by necking. At the beginning of the stretching, PP film showed an elastic deformation, which was accompanied by a tiny decrease in the temperature up to 0.25°C, in accordance with Kelvin's theory. After passing the yielding point with maximum load and as the load was decreasing to a constant level, an extremely high temperature region about 15°C higher than other regions—as indicated by the red-colored zone—suddenly appeared at a point near the side edge. This represents the real start of the necking phenomenon. The extremely high temperature, however, was far lower than the melting temperature of 176°C.⁸ Figure 3(b) shows the temperature distribution image at this point in more detail with 16 times higher temperature sensitivity. On an extended line from the higher temperature zone shown in pink color at the side edge, a moderately high temperature region in yellow color developed. Within this yellow region, several red spots indicate regions of somewhat

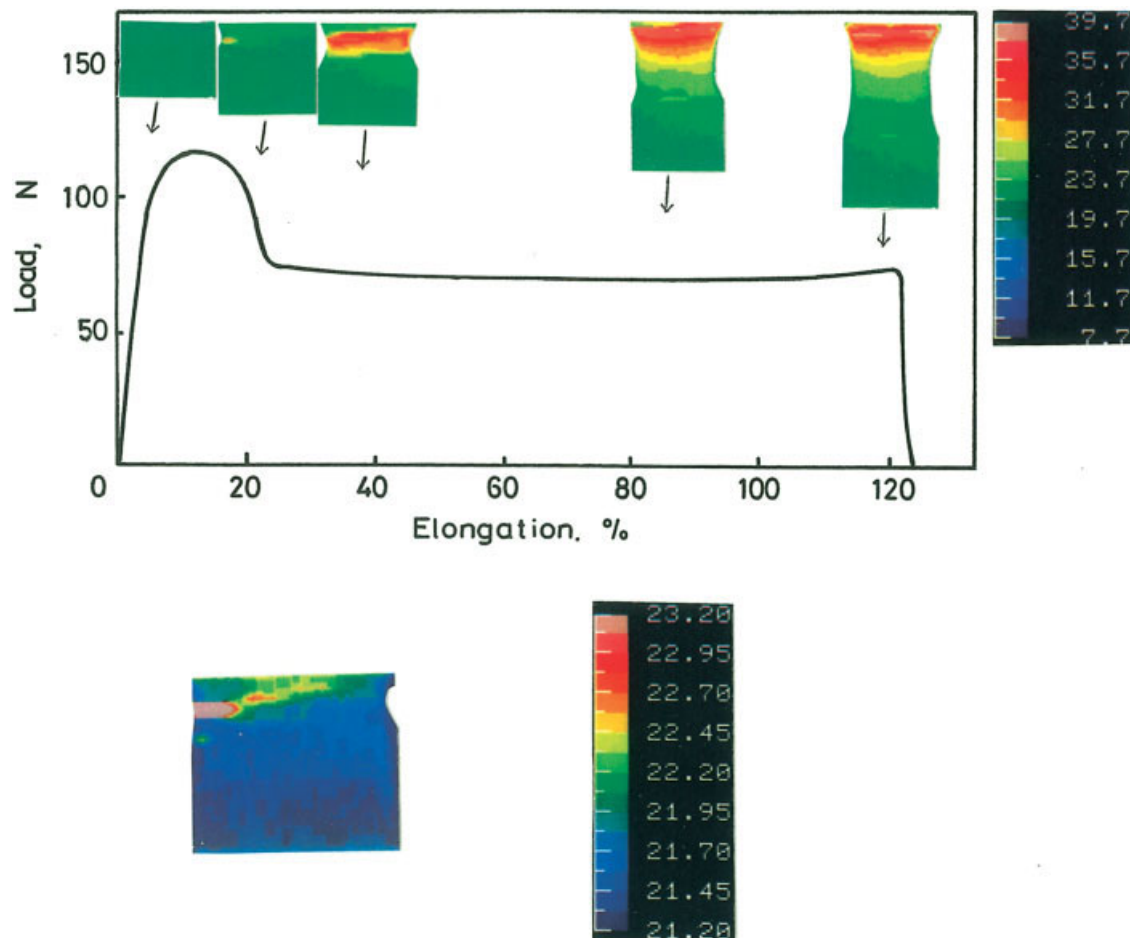


Figure 3 (a) Load/elongation relationship and some selected temperature distribution images from the drawing of PP film. (b) Temperature distribution image shown with 16 times higher temperature sensitivity of (a) at 35% elongation. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

higher temperature. The higher temperature part near the edge extended horizontally almost instantaneously and reached the other edge while the load decreased to the level-off value. The thermographic observation precisely described the start of necking process as follows. When the origin of the necking first appeared at the side edge, other prenecking points successively and instantaneously developed in a short period along the stressing zone. The necking from the side edges developed and connected with the prenecking points, and then the necking was thus completed throughout the width. Deformation and fracture of spherulites and friction between them in the necking zone were assumed to generate heat. Furthermore, rearrangements of molecular orientation occurred in the necking zone from the folded chain crystal to the extended chain crystal. These rearrangements may be accompanied by an adiabatic heat generation, which would cause a temperature rise.¹⁰ When these structural changes began to occur throughout the width, as shown by the red color in the figures, the load kept nearly constant and the necking

apparently seemed to start. While the necking was propagating with nearly constant load, the high temperature part as shown in red color appeared constantly at the upper neck end. A slightly high temperature portion, shown in light green, also appeared in the center just below the neck end during the necking period, which showed that prenecking or light necking with molecular and crystal rearrangement may occur at the other stress concentrating zone at the other end of the neck. After the necking front at the upper end propagated, the temperature of the necked part gradually decreased because of thermal conduction and radiation to the surroundings without heat generation. Thus, the molecular and crystal rearrangement exclusively occurred at the necking front at the upper end and additionally at the prenecking part of the lower end of the neck. As the drawing continued, the drawn portion increased in length at the expense of the undrawn portion of the specimen. The widths of the drawn and undrawn portions remained nearly constant throughout the necking process. The breakage of film occurred when the necking reached the

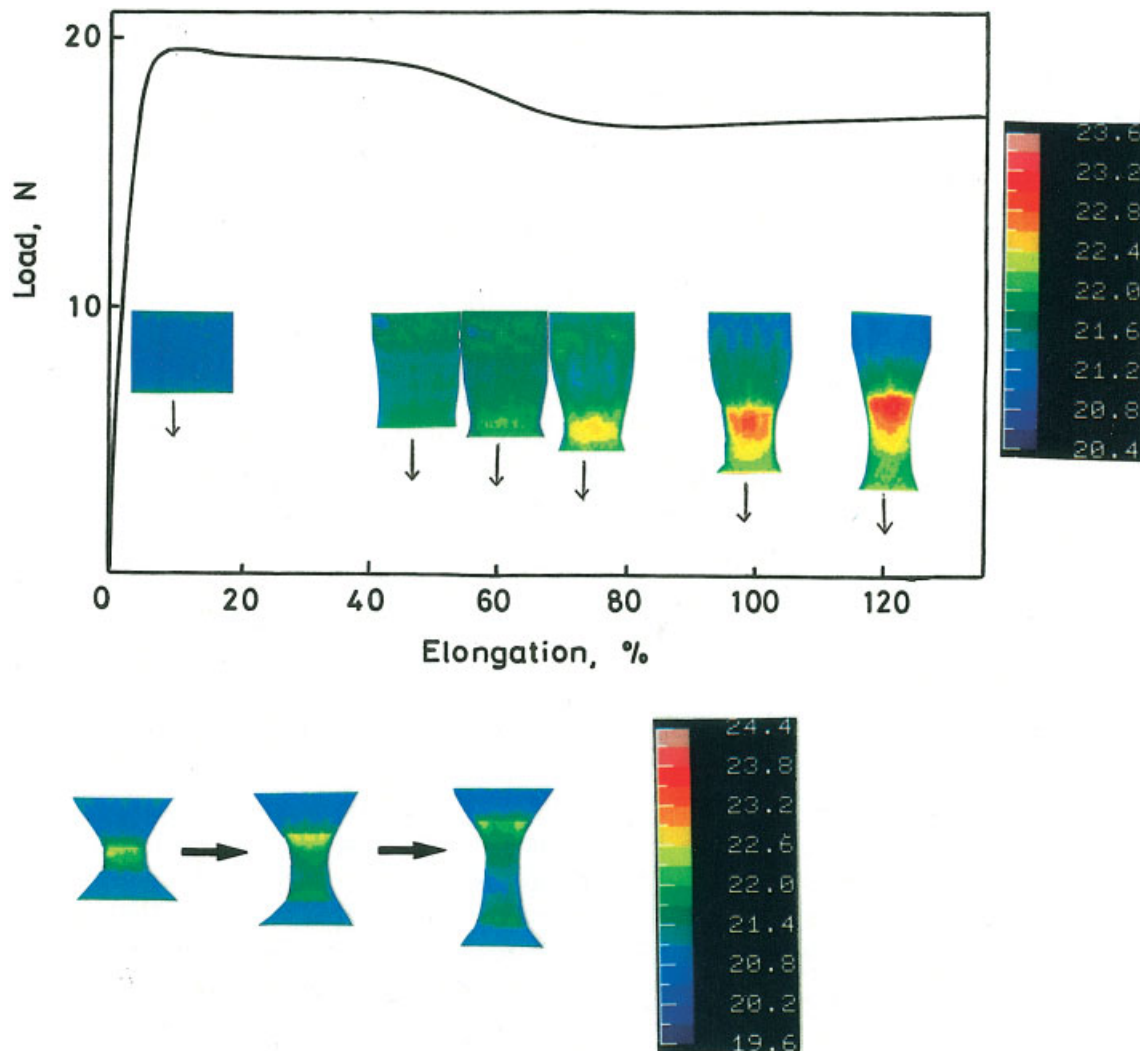


Figure 4 (a) Load/elongation relationship and some selected temperature distribution images from the drawing of PE film. (b) Selected temperature distribution images from the drawing of different PE film; From left to right, at 35, 70, and 100% elongation. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

upper end of the specimen. Further, the length of the undrawn portion was ~ 41 mm and the remaining portion of 9 mm was drawn to ~ 60 mm length during the necking period of 6 s. Thus, ~ 1.5 mm in original length or 1.5/50 mm of the original mass was drawn within 1 s. Since heat generated at the necking front during the necking was propagating, the heat within this period was able to be roughly estimated from the rise of temperature at the necking front with the assumption of heat capacity being $1.93 \text{ J/g K}^{\circ}$ and knowing the drawn mass. The temperature rise accompanying the necking was 15°C and the estimated heat within 0.25 s was 28 mJ. On the other hand, the mechanical work done during the period was calculated as 150 mJ from the load (75 N) and the elongation (2 mm). During the necking period, the generated heat and mechanical work were nearly constant and thus the exchange rate from mechanical work to the estimated heat was constant at $\sim 19\%$.

As another result, the selected temperature distribution images with the load–elongation curve for PE are shown in Figure 4. At the beginning of stretching, the load steeply increased and reached the maximum. With further stretching up to $\sim 50\%$ elongation, the load gradually decreased and the observed uniform increase of the temperature suggested heat generation throughout the whole specimen. After passing $\sim 50\%$ elongation, the extent of falling load increased and the lower part of the specimen began to narrow. This indicated the beginning of the necking in PE. The part showed a slightly high temperature in light green color. That is, the heat generation seemed to concentrate in the narrow part. At $\sim 75\%$ elongation, the load falling decreased and the narrowed zone expanded upward, i.e., the necking was propagating. After the load decreased to a constant level, the temperature distribution image gradually became constant, although the high temperature part shown in red at the center of the necked part was moving upward.

Beyond ~95% elongation, the nearly constant temperature distribution moved upward with the propagation of the necking front. The distribution showed the stable necking front in red color and a gradual decay of the temperature by thermal conduction and radiation to the surroundings in yellow and thin green color. Compared with the necking front shown in the high temperature portion of the PP, the one in PE had a fairly wide area and the extent of temperature increase was low. Furthermore, its center was not located at the upper neck end but appeared to remain inside of the necked zone below the upper end. Thus, the rearrangement of molecular and crystal structure during the drawing of PE seemed to occur extensively. The length of the undrawn portion was ~30 mm and the remaining portion of ~20 mm was drawn during the necking period of ~3.5 s, thus, ~5.6 mm in original length was drawn within a second. The maximum temperature during the drawing was far lower than the melting temperature of 142°C.⁸ Thus, partial melting must not occur during the necking period in either PP or PE.

Heat generation and the exchange ratio from mechanical work to heat in PE were also estimated in the same way as for PP, with the assumption of heat capacity being 2.3 J/g k.⁸ For example, the specimen was uniformly expanded at ~35% elongation before the start of necking. The area was defined on the specimen and a rise of the average temperature within 0.25 s was determined to be 0.75°C. The heat was estimated from the original mass of the specimen and from the temperature rise to be 61 mJ. The mechanical work done during the period was estimated as 38 mJ from the load of 19 N at 35% elongation and the elongation of 2 mm during 0.25 s. On the other hand, at 100% elongation during the stable necking period, heat generation only occurred at the necking front. The heat within 0.25 s was roughly estimated from the temperature rise at the necking front and the estimated drawn mass. The determined temperature rise during the necking was 1.5°C and the estimated heat was 3.5 mJ, while the mechanical work at this period was 34 mJ. As a result, the exchange rate from mechanical work to heat changed from 160% before necking to 10% during the necking.

Another series of temperature distribution images during the drawing are also shown in Figure 4(b) for PE film with different specifications. At the beginning of the necking with ~35% elongation, the high temperature part appeared over the necked zone and the fairly higher temperature part colored in yellow was located at the upper neck end. While the necking was propagating at ~70 and 100% elongation, the high temperature part was distributed at both ends of the necked zone and a higher temperature part colored in yellow was located at the upper neck end. Thus, the molecular and crystal rearrangement occurred at the necking front in this PE film.

These results suggested that a somewhat different necking mechanism should be considered between different PE films if the specifications were different.

CONCLUSIONS

Heat generation during the drawing of polymer films was successfully measured as a series of temperature distribution images by use of thermography. For the polymers drawn without necking, the uniform heat generation throughout the specimen during the entire drawing process indicated their uniform drawing. On the other hand, for PP and PE as the representative polymers that exhibited the necking phenomenon, their heat generation at one side of both ends of the necked zone and supplementary heat generation at the other ends during the necking process suggested that necking actually occurred at the necking front, i.e., the end of the necked zone. Heat, however, was extensively generated at the necked zone for the different PE film, showing a somewhat different necking mechanism. From the temperature rise, the generated heat within some period was able to be roughly estimated and thus the estimated heat led to the thermodynamic investigation, i.e., the relation between mechanical work and thermal energy by the use of thermography. Further thermodynamic studies using thermography for various polymer films are expected to clarify more precisely the polymer drawing mechanism with the help of other methods such as X-ray diffraction and morphology changes.

The author thanks Prof. J.L. Davis, Department of Engineering Professional Department, University of Wisconsin-Madison, for his critical reading of the manuscript and Dr. T. Taga for his useful comments. The author also thanks Laboratory of Wood Processing and Laboratory of Biomaterial Design in Graduate School of Agriculture, Kyoto University for permission to use the thermal imaging system.

References

- Nielsen, L. E. *Mechanical Properties of Polymers*; Reinhold: New York, 1962; Chapter 5.
- Marshall, I.; Thompson, A. B. *Proc R Soc London Ser A* 1954, 221, 541.
- Oono, R. *Sen-i Gakkaishi* 1993, 49, 654.
- Muller, F. H. In *Rheology, Theory and Applications*, Vol. 5; Eirich, F. R., Ed.; Academic Press: New York, 1969; p 417.
- Yamauchi, T.; Murakami, K. *Jpn Tappi* 1992, 46, 70.
- Yamauchi, T.; Okumura, S.; Noguchi, M. *J Mater Sci* 1993, 28, 4549.
- Baumeister, T., Ed. *Standard Handbook for Mechanical Engineers*, 7th ed.; McGraw-Hill: New York, 1952; p 4.
- Roff, W. J.; Scott, J. R. *Fibers, Films, Plastics and Rubbers, A Handbook of Common Polymers*; Butterworths: London, 1971.
- Yamauchi, T.; Tanaka, A. *J Appl Polym Sci* 1994, 53, 1125.
- Billmeyer, F. W., Jr. *Textbook of Polymer Science*; Interscience: New York, 1962; p 164.