JOURNAL OF APPLIED POLYMER SCIENCE VOL. 18, PP. 3581-3592 (1974)

Stress Birefringence Patterns in Molten Polymers During the Mold-Filling and Cooling Process

CHANG DAE HAN, *Department of Chemical Engineering, Polytechnic Institute of New York, Brooklyn, New York 11201*

Synopsis

An experimental study has been carried out to better understand the phenomenon of stress buildup during the mold-filling process in the injection molding operation. For the study, a rectangular mold with two glass windows was constructed, so that stress birefringence patterns of molten polymers flowing into the mold could be photographed with the aid of a polariscope. As a feeding system, a 1-in. extruder was used attached to the mold with a 2-ft length of stainless steel tubing having a relief valve. In this way, the injection pressure (and injection velocity) was carefully controlled to ensure that the glass windows would not be damaged. The development of stress birefringence patterns during the mold-filling process was recorded on a movie film. It was observed that, in isothermal operation, when flow stopped after the mold was filled, stresses relaxed immediately because of the very slow cooling of the mold by ambient air. However, it was observed that, as cooling proceeded, stresses were gradually built up again in the mold. It was possible, therefore, to determine the residual stress in the mold, which originates from the cooling process alone.

INTRODUCTION

Injection molding is one of the major methods by which thermoplastics are fabricated. Despite its technologic importance, relatively little basic work has been done to elucidate a number of interesting and yet complex phenomena encountered in the process. This may be attributed, in part, to the complexity of many steps involved in the injection molding process: mold filling, packing, cooling, etc. Considering the viscoelastic nature of thermoplastics, one can easily appreciate the difficulties inherent in attempting a rigorous solution of the problem of a hot, non-Newtonian (viscoelastic), compressible liquid flowing through a geometrically complex flow channel the walls of which are much colder than the liquid. Perhaps one may rephrase most problems involved with injection molding as "unsteady-state, nonisothermal flow of viscoelastic polymer melts" in a complex flow channel. It is little wonder that, in the past, injection molding has been left to develop as an empirical art for the most part.

In the injection molding operation, the mold is first closed, and then the set amount of polymer introduced ahead of the plunger. The plunger moves forward and forces the polymer into the heating zone, at the same time forcing already heated polymer out of the cylinder section and into the mold. Pressure is maintained on the plunger for some time after the mold has been filled to permit buildup of adequate pressure in the mold. Cooling water is circulated through channels in the mold so as to keep the mold cavity walls at a temperature which is usually between room temperature and the softening temperature of the polymer. Thus, the hot polymer begins to cool as it enters the mold. When it is cooled to a state of sufficient rigidity, the mold is opened and the piece is removed.

Some of the important variables in the operation of an injection molding machine are: (a) the pressure applied by the plunger, (b) the temperature of the heating section, (c) the temperature of the mold, (d) the plunger-forward time, (e) the mold-closed time, and (f) the mold-open time. Adjustment of these variables is, in general, very complicated. Among the variables which are of importance in determining the quality of the mold piece are the pressure, temperature, and density of the polymer in the mold just before the mold is opened. In general, we wish to know the value of these quantities as functions of time after the mold has been filled.

In principle, the temperature distribution in the molded piece may be computed as a function of time when one knows the geometry of the mold, the temperature at which the polymer enters the mold, and the mold temperature. However, in practice, a rigorous theory is very difficult to develop, because of the geometrically complex shape of a mold and of the rheologically complex nature of a polymer being molded. Note that a molding of good quality is one free from warpage, sinks, bubbles, scoring, cracks, etc. This can be realized only when the correct amount of plastic is injected into the mold at the proper temperature and pressure.

Despite the fact that exact solutions of many injection molding problems are difficult to obtain, much insight into the process may be gained by applying some carefully chosen experimental techniques and by using plausible approximations and idealizations in a theoretical study. In the early 1950's and 1960's, several researchers made very important contributions to this end, notably Spencer and Gilmore[1-3] and Ballman and his co-workers.[4-6] Spencer and Gilmore[3] made a flow visualization study involved with mold-filling processes and the flow pattern as affected by fill time, temperature, and pressure, using a glass-window mold. Very recently, a similar experiment was reported by White and Dee.[7]

Wales et al.[8] made an attempt to compare, as a measure of the molecular orientation, the birefringence of injection-molded articles and the flow birefringence of molten polymers during steady, isothermal shear flow. They reported that the birefringence of the molded article (plate in their case) was found to be relatable to the magnitude of the shear stress at the cavity wall during the mold-filling process, independent of the temperature employed in the flow birefringence experiment. In reference to the observation made that the flow birefringence was found to be related to the wall shear stress in isothermal flow, they concluded that the orientation of the injection-molded articles is dominated by the shear stresses during the mold-filling process regardless of the temperature and of the macroscopic rate of deformation.

Very recently, the author carried out an experimental study of flow birefringence in complex flow channels, simulating the mold-filling and cooling process, in the injection molding of polystyrene and high-density polyethylene. In his experiment, the author was able to follow the changes in the stress birefringence patterns as a hot melt was injected into a rectangular mold and subsequently cooled by ambient air. In this paper, we shall discuss some of the representative results of the flow birefringence study and discuss the results in terms of the rheological properties concerned and the molding variables.

EXPERIMENTAL

Apparatus

The apparatus consists of three major parts: (a) the polymer melt feed system, (b) the mold, and (c) the optical system, as shown schematically in Figure 1.

As the polymer melt feed system, a 1-in. extruder was used, which was connected to the inlet of the mold by a 2-ft length of stainless steel tubing (0.5 in. I.D.). The tubing had a relief valve for the purpose of protecting the glass windows installed on either side of the mold from a rapid buildup of pressure in the mold at normal screw speeds. This rather long feed line was used to sufficiently reduce all the spiral motions present in the melt created by the screw in the extruder, and also to ensure that the melt being injected into the mold was at uniform temperature. The mold was placed on an optical bench on which a polariscope was attached. It should be noted that the tubing section through which polymer passes was covered

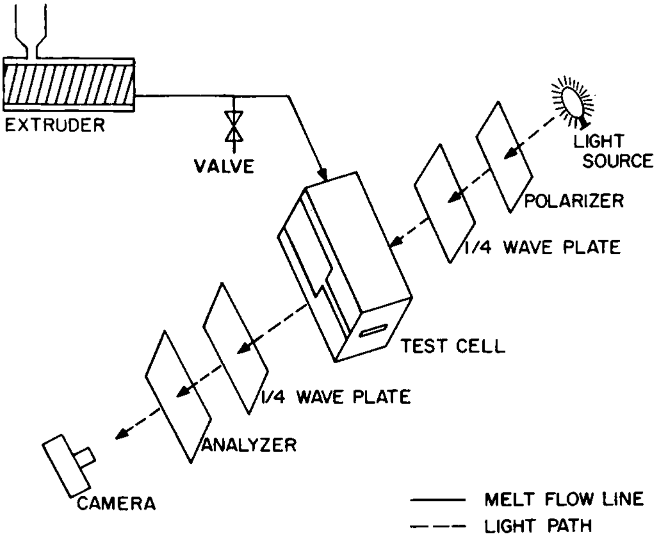


Fig. 1. Schematic diagram of apparatus.

with nichrome wire heaters and thermally insulated with asbestos. The heating system was controlled by a thermistor-regulated temperature controller.

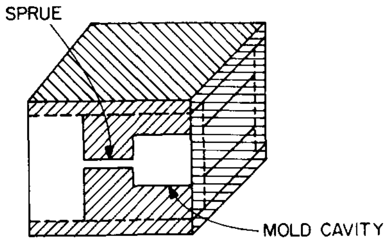
The main components of the optical system were a light source, interference filter, diffusion screen, polarizer, quarter wave plates, analyzer, and a camera. The optical system used in the present study was the same as that described in an earlier paper by Han and Drexler.[9]

Design of the Molds

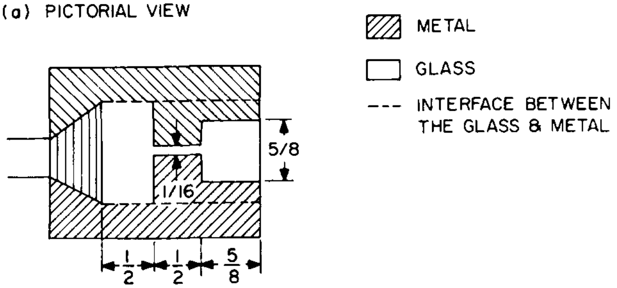
A rectangular mold with a small gate in the front was designed and built for use in this study. Figure 2 shows views of the rectangular mold used.

Glass windows were installed on either side in order to observe the flow birefringence patterns in the mold during the mold-filling process. The windows cover the entire area of flow field, including the upstream region which extends into the gate. The glass was a strain-free, Vycor glass manufactured by the Corning Glass Company. The pieces were ground, cut, and polished by the Dell Optics Company. The glass plates were bolted to the mold by means of a steel cover plate and were separated from the mold and from the cover plate by an asbestos-filled rubber gasket.

For the injection molding of polymer melt into a hot mold, all metal surfaces of the mold were covered with band electric heaters. The heaters were controlled by a thermistor-regulated temperature controller. When the polymer melt was injected into a cold mold, the electric heaters were disconnected and the cooling was realized by ambient air. In other words, no special provision was made to keep the mold cold.



(a) PICTORIAL VIEW



(b) SIDE VIEW

Fig. 2. Schematic diagram of the mold design.

Experimental Procedure

The experimental procedure used for taking the flow birefringence measurement was straightforward, as described in an earlier paper by Han and Drexler.[9] In the case of injection molding into a cold mold, only the feed system (the extruder and tubing) was preheated to the desired operating temperatures. On the other hand, when an isothermal experiment was carried out, the mold was also preheated together with the feed system. Since separate heating elements were used for the tubing section at the mold, it was possible to maintain the mold temperature at any desired value, lower than the hot melt being injected.

Because of the very viscous nature of the polymer melt, the pressure buildup in the mold was so rapid that when the injection rate was high, the glass windows burst. Therefore, the injection rate was reduced and controlled by the relief valve attached to the 2-ft-long tubing. A motion picture was taken throughout the entire mold-filling period; the flow was discontinued as soon as the mold was completely filled.

Materials

Polymers used for experiment were general-purpose polystyrene (Dow Chemical, Styron 686) and high-density polyethylene (Union Carbide Corp., DMDJ 4309). These polymers were chosen because their viscoelastic properties had been determined in previous studies.[10,11] It is worth mentioning that the transparency of polystyrene permitted us to observe the stress birefringence patterns when the molten polymer froze to room temperature.

RESULTS

When a beam of polarized light is directed at a melt flowing into a mold and the light leaving the liquid is passed through an analyzer (see Fig. 1), it renders visible interference patterns which may be related to the magnitude and direction of the shearing stresses set up by the flowing liquid. When monochromatic light is used, regions of light interference and reinforcement result, giving rise to a pattern of black and white bands known as *isochromatics*.

When white light is used, however, isochromatics appear as bands of the same color. Each band corresponds to a constant value of the maximum shearing stress. It should be noted that the direction of black and white bands represents the orientation of the principal tensile stress. In the past, a number of researchers made use of measurements of birefringence patterns to determine the degree of molecular orientation.[1,5,6]

In order to obtain quantitative information from flow birefringence measurements, one has also to take pictures of isoclinics, which appear as other dark regions, when the maximum shearing stress makes a certain angle with the axis of polarization of the incident light. These isoclinics appear only when plane-polarized light is employed (i.e., when the quarter

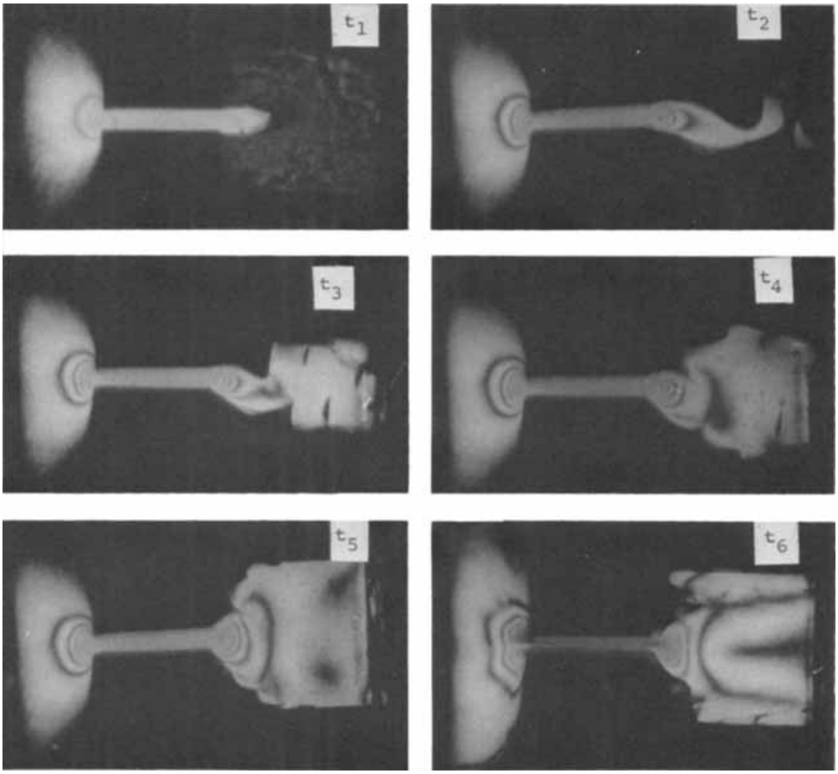


Fig. 3. Stress birefringence patterns of polystyrene at 200°C during the mold-filling process.

wave plates are removed from the optical system shown in Fig. 1). In the present study, however, it was not possible to take pictures of isoclinics simultaneously with isochromatics. Although it is not possible to obtain quantitative information from the flow birefringence measurements taken at unsteady-state flow, the isochromatics provide some important information as to where the greatest stress concentration occurs in the mold and in which direction stresses (and hence molecules) are oriented.

Figure 3 gives a series of isochromatic fringe patterns of polystyrene in sequence as the mold filling proceeds. Note that these pictures were taken under the isothermal injection molding condition. That is, the temperature of the mold was the same as that of the melt being injected. Similar stress patterns were also observed when a hot melt was injected into a cold mold at ambient temperature. Since the mold was not cooled by circulation of cold water, as in the commercial injection molding process, the effect of cooling on the stress patterns during the filling process was not observed. As a result, stresses relaxed immediately right after flow stopped. In other words, the cooling rate was so slow that, at the beginning of the cooling period, little stress was visible when flow stopped. However, as the cooling proceeded, the stresses gradually built up again in the mold.

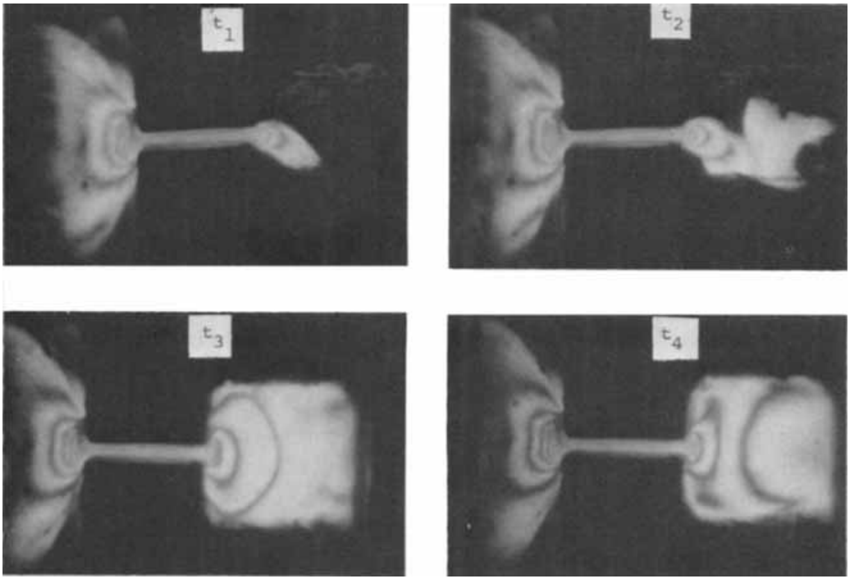


Fig. 4. Stress birefringence patterns of high-density polyethylene at 200°C during the mold-filling process.

Figure 4 shows a series of isochromatic fringe patterns of high-density polyethylene during the mold-filling process. These birefringence patterns are very similar to those of polystyrene shown in Figure 3.

It is worth noting that the pictures given in Figures 3 and 4 also show flow patterns (i.e., advancing wave fronts) of a hot melt during the mold-filling process. In other words, the present study provides information as to how a mold is filled by advancing wave fronts of molten polymers and also how stresses develop in the mold during the mold-filling process.

DISCUSSION

Stress Buildup and Orientation During the Mold-Filling Process

When a hot melt flows into a cold mold cavity from the heating zone through the gate, pressure (and hence stress) starts to build up in the mold during the so-called mold-filling process. Subsequently, there will be an abrupt increase in stress during "packaging," that period of the plunger-forward time subsequent to filling. The stresses built in the polymer inside the mold cavity will then relax during the discharge stage. However, the amount of the stress relaxation during the discharge depends on the rate of cooling. For instance, if cooling occurs very fast, there will be less time for the stress to relax, and consequently much of the stress built initially in the melt will remain in the molded article as so-called "frozen-in" stress, which will be discussed later. In 1950, Spencer and Gilmore[1] made some interesting studies which elucidate the origins of residual stresses (or strains) in injection-molded articles, by making observations of the stress birefringence patterns of molded specimens with crossed Polaroids. However, they did not observe how the stresses actually built up and relaxed during the injection molding operation.

The tendency of polymer molecules to orient while flowing leads to "frozen-in" molecular orientation when the flowing plastic is solidified. In the past, there have been a number of studies made on the molecular orientation and the morphology produced in injection moldings.[12–15] In order to study polymer flows through a cooled channel, Ballman and Toor[5] observed the birefringence patterns of simple injection-molded polystyrene specimens and investigated the effect of several process variables on molecular orientation. They postulated a mechanism of nonisothermal plastic flow, based on their observation of birefringence patterns of molded specimens. They suggested that two competing effects come into play in the observed patterns, i.e., molecular orientation induced by shearing forces during flow and relaxation after these forces are reduced when flow stops. They concluded that orientation decreases with increasing (1) mold temperature, (2) cavity thickness, and (3) cylinder temperature. Orientation increases with increasing (1) gate size, (2) ram pressure, and (3) ram forward time.

It is seen in Figures 3 and 4 that the number of fringes increases as the flow rate is increased and as temperature is decreased. This is as expected, because the magnitude of stress increases with flow rate and decreases with temperature. The sensitivity of birefringence patterns to temperature is of particular importance in understanding variations in molecular orientation during the cooling process in injection molding.

Stress Buildup and Orientation During the Cooling Process

Figure 5 gives a series of pictures showing stress birefringence patterns of polystyrene as it progressively cools to a completely frozen state. As mentioned above, when cooling started, there was no stress visible in the mold. However, as the hot melt cooled, stress gradually built up again in the mold. Therefore, the stresses shown in Figure 5 are strictly due to cooling.

It was observed that stresses first built up as the mold cooled off and then relaxed as cooling proceeded further. In other words, there appears to exist an instant during the cooling period at which the stress passes through a maximum. This may be explained as follows. When cooling first starts, the mold surface gets colder than the center of the mold, and hence a temperature gradient develops between the surface and the center. When the temperature gradient reaches a maximum, the stress buildup in the mold will be greatest. Now, as cooling proceeds further, the temperature gradient in the mold will become less, and hence stresses will relax accordingly. When a mold is frozen completely, there still are some stresses left in the mold.

According to Spencer and Gilmore,[1] there are three types of residual stress (or strain) in injection-molded pieces: (1) those accompanying

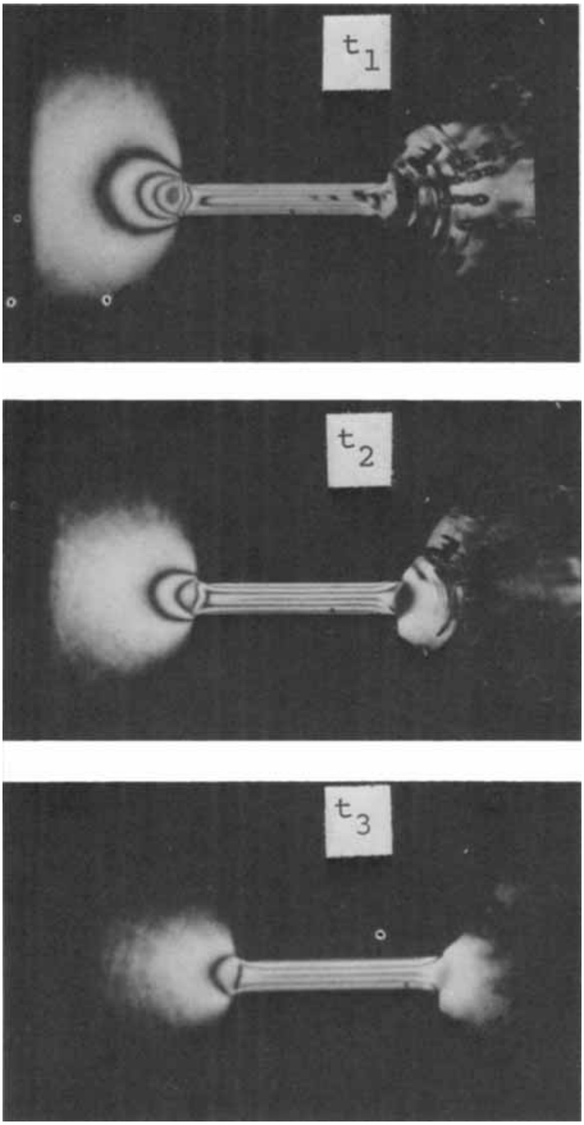


Fig. 5. Stress birefringence patterns of polystyrene during mold cooling.

quenching stresses, (2) frozen-in molecular orientation, and (3) configurational volume strains. Quenching stresses sometimes relieve themselves by producing bubbles or sink marks in the article, and may be otherwise relieved by annealing. Configurational volume strains can be relieved only by annealing (which is frequently impractical), and are not important in many practical cases. Much of the frozen-in orientation present originates during "packing." Considerable reduction in the amount of frozen-in orientation may be effected by minimizing the packing time. Reduction in

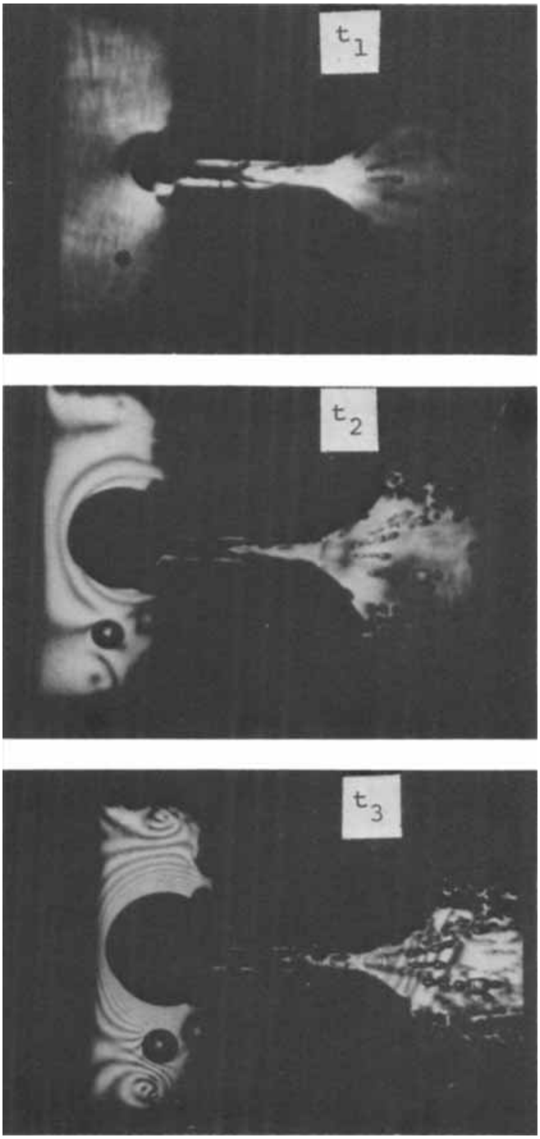


Fig. 6. Stress birefringence patterns of polystyrene during annealing.

the amount of frozen-in orientation reduces the tendency of the moldings to "craze," improves the dimensional stability on heating, and produces more consistent specimens for mechanical testing.

The viscosity of a molten polymer is a very important property in the study of injection molding, but is not of immediate interest in discussing residual stresses (or strains). This is due to the fact that once a molten polymer fills the mold, there is virtually no bulk flow of the material in the mold. However, due to the high rubber-like elasticity of the material, the stress built in the material does not relax instantaneously, but gives rise to a slow recovery of deformation, which otherwise would occur instantaneously for *inelastic* materials.

If a material is cooled to the softening point while the stress is still acting, the molecular chains are immobilized in the uncoiled state, and we find that frozen orientation is present in the plastic. Residual stresses are commonly found in samples of materials which have been cooled rapidly through their freezing points or hardening ranges. The magnitude of the residual stresses may be reduced by slowing down the process of cooling during the period in which hardening is taking place throughout the body. This annealing procedure decreases the temperature differences present during hardening, and thus reduces the residual stresses. In general, annealing to minimize quenching stresses has little practical application during injection molding. However, annealing of the finished articles may be feasible in some cases.

Figure 6 shows a series of pictures showing stress birefringence patterns of polystyrene as the polymer in the mold cavity was heated up again from the completely frozen state. It is seen that quite a bit of stress which was present in the mold before annealing started is relieved by annealing, consistent with the view expressed by Spencer and Gilmore.[1]

CONCLUSIONS

An experimental result is presented showing the development of stress birefringence patterns in molten polymers during the mold-filling and cooling process during injection molding. It was observed that when the cooling rate was slow, stresses relaxed immediately after mold fill was completed. However, as cooling proceeded, stresses gradually built up again in the mold cavity. This is attributed to nonuniform temperature distributions in the mold during the cooling process. In other words, at the instant the outer surface of the mold cavity reaches the freezing point, the stresses acting can be only hydrostatic. A short time later, a shell of frozen, rigid material has formed, containing within itself a region of hotter, still fluid material. Further cooling of the central region results in a negative hydrostatic stress and corresponding compressive tangential stresses in the rigid shell. This situation persists, under certain conditions, even when thermal equilibrium has been attained, leaving balanced residual stresses in the mold cavity at room temperature. It was observed further that the residual stresses developed during the cooling period, sometimes referred to as "quenching stresses," were relieved to a great extent by annealing.

References

1. R. S. Spencer and G. D. Gilmore, *Mod. Plast.*, **27**, 97 (1950).
2. G. D. Gilmore and R. S. Spencer, *Mod. Plast.*, **28**, 117 (1951).
3. R. S. Spencer and G. D. Gilmore, *J. Colloid Sci.*, **6**, 118 (1951).
4. R. L. Ballman, R. L. Toor, and T. Shusman, *Mod. Plast.*, **37**, 105, 115 (1959).
5. R. L. Ballman and R. L. Toor, *Mod. Plast.*, **60**, 113 (1960).

6. G. B. Jackson and R. L. Ballman, *SPE J.*, **16,** 1147 (1960).
7. J. L. White and H. B. Dee, to be published.
8. J. L. S. Wales, I. J. Van Leeuwen, and R. Van Der Vijgh, *Polym. Eng. Sci.*, **12,** 358 (1972).
9. C. D. Han and L. H. Drexler, *J. Appl. Polym. Sci.*, **17,** 2329 (1973).
10. C. D. Han, *J. Appl. Polym. Sci.*, **15,** 1149 (1971).
11. C. D. Han and T. C. Yu, *J. Appl. Polym. Sci.*, **15,** 2567 (1971).
12. K. J. Cleereman, *SPE J.*, **23,** 43 (1967).
13. I. T. Barrie, *Plast. Polym.*, **37,** 463 (1969).
14. E. S. Clark, *SPE J.*, **23,** 46 (1967).
15. M. B. Kantz, H. D. Newman, and F. H. Stigale, *J. Appl. Polym. Sci.*, **16,** 1249 (1972).

Received April 10, 1974
Revised June 3, 1974