Experimental Measurement of Internal Stresses in Polypropylene-Injected Disks

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SYNOPSIS

Disks in polypropylene were injected in a mold with an unbalanced cooling pattern to induce warpage. The internal stresses were determined using IR dichroism for molecular orientation measurement and an original method for thermal stress assessment, on the basis of the analysis of the dilatometrical behavior of microtomed cuts removed from the part. From both experimental results and structural observations with an optical microscope, a simple model is proposed to describe the warpage due to the effect of unbalanced cooling. A constant deformation was observed and cannot be described by our model, but calculated relative deformations of the part geometry are in good agreement with experimental results.

INTRODUCTION

After having been regarded as only good substitute materials, appreciated because of their low cost and easy processibility, thermoplastics have now entered new fields of applications where quality criteria become more and more severe. Those requirements not only concern mechanical, chemical, or thermal properties, but also dimensional precision and stability. But an injected part is characterized mainly by high shrinkage rates and a very low ability for maintaining its shape and geometry, especially when used under severe thermal conditions.

Among the numerous problems presented by injected parts, a convenient solution for warpage has not found yet. Its origins are attributed to the action of internal stresses resulting from processing. Two kinds of stresses are generally considered:

- The residual flow stresses,[1-3] giving rise to molecular orientation (entropic term); and
- The thermal stresses,[1,3] due to heterogeneous shrinkage rates throughout the part during cooling (energetic term).

To study the origin of warpage on molded parts, we injected disks of polypropylene (PP) in a mold with a controlled unbalanced cooling pattern to induce warpage. The internal structure of the part was studied by optical microscopy, then the internal stresses (both components) were determined using IR dichroism (molecular orientation) and a dilatometrical method (thermal stresses) that was developed during this work.

EXPERIMENTAL

1. Mold and Polymer

We used a disk mold whose main characteristics are as follows:

- Diameter: 160 mm
- Thickness: 2 mm
- Centrally gated
- Diameter of the sprue at the base of the part: 8 mm.

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This mold is equipped with five pressure transducers (two located on the sprue and three along one radius of the disk). The temperature measurement inside the mold is achieved by seven thermocouples: Five are located in front of the cited pressure transducers and just under the wall (0.5 mm from the wall) and one in the bulk of each part of the mold.

The mold is mounted on a Billion H90/280 injection machine, whose maximum clamping force is 90 tons. The process is controlled by a microprocessor system Visumat 4000. Disks were injected using an isotactic homopolymeric PP provided by Appryl (PP H3050 MN1–MFI 230°C/2.16 kg = 5).

2. Processing Conditions

The mold thermal regulation was the only parameter to be studied, and other processing conditions remained unchanged throughout the cycles and were set as

- Holding pressure: 60 bars
- Holding time: 10 s
- Melt temperature: 250°C

The value indicated for the holding pressure is the hydraulic pressure set point. It corresponds to a 400 bars pressure on the polymer at the entry to the part. The mold thermal regulation is set to obtain different temperatures on each side. Disks are referred as PP $T1/T2/n$, where $T1$ is the temperature on the stationary half of the mold; $T2$, the temperature on its movable half; and n, the serial number.

To simplify those T' references, we considered only the set temperatures, instead of the true values recorded by the thermocouples in the bulk of each half of the mold when the thermal equilibrium was reached (after about 20 cycles). Table I indicates those true temperatures for each set point.

Table I Correspondence between the True Temperatures and the Set Points

	True T'_1 (°C) Stationary Side	True T'_2 (°C) Movable Side	ΔT (°C)
10/10	21	14	+7
10/30	21	29	−8
10/50	21	52	−29
10/80	21	72	−51
30/10	32	15	+17
50/10	51	15	+36
65/10	63	15	+48
80/10	78	16	+62

The thermal amplitude ΔT between each wall inside the cavity is then defined by

$$\Delta T = T'_1 - T'_2$$

where T'_1 and T'_2 are, respectively, the true temperatures measured in the bulk of the stationary (T'_1) and the movable (T'_2) half of the mold.

We notice that for a given set point (especially at lower temperatures) the actual temperatures obtained in both halves of the mold are significantly different. This behavior is attributed to the effect of the sprue that has no specific cooling circuit and constitutes a "hot runner" all along the cycle.

3. Dimensional Measurements

Warpage was characterized by the maximum deformation at the far end of the part. Because of their low thickness (2 mm) and the low stiffness of PP, injected disks are very sensitive to creep and any measuring method implying a contact is forbidden as it induces an additional deformation.

We used a laser transducer mounted on the Y axis of a tridimensional measurement machine. This transducer delivers a tension proportional to its distance from the part.

4. Molecular Orientation Measurements

Infrared dichroism measurement leads to the second moment of the global orientation function.[4-9] It allows separate determination of orientation for both crystalline and amorphous zones of the polymer. The absorption of IR radiation for a chemical group in a polymer chain depends on the angle α between the transition moment of the considered vibration with the chain axis and on the angle θ between the chain axis and the orientation direction.

The second order moment of the orientation function $P2\langle\cos\theta\rangle$ is linked to α and θ by

$$P2\langle\cos\theta\rangle = (3\langle\cos\theta\rangle^2 - 1)/2 \qquad (1)$$

or

$$P2\langle\cos\theta\rangle = \frac{2\,\mathrm{cotg}^2\alpha + 2}{2\,\mathrm{cotg}^2\alpha - 1} \times \frac{D-1}{D+2} \qquad (2)$$

where D is the measured dichroic ratio, defined by

$$D = A0/A90 \qquad (3)$$

where $A0$ = absorbance when the polarization is parallel to the reference direction, and $A90$ = ab-

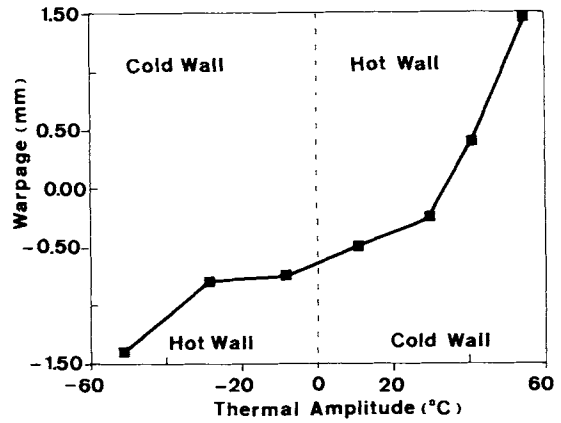

Figure 1 Warpage vs. thermal amplitude.

sorbance when the polarization is normal to the reference direction.

The orientation function $P2\langle\cos\theta\rangle$ varies from -0.5 (perfect perpendicular orientation) to 1 (perfect parallel orientation). FTIR spectra were obtained on a Perkin-Elmer spectrometer 1720X equipped with a rotating polarizer Harrick. Ten coadded interferograms were scanned at 2 cm^{-1} resolution.

RESULTS

1. Dimensional Measurements

Figure 1 shows the evolution of warpage (defined as above) vs. the thermal amplitude ΔT. Experimental results plotted there correspond to measurements at 75 mm away from the center of the disk. A former study showed that the general shape of our parts is a cone (a Chinese hat shape), so that any point located at a given distance from the center has the same altitude. Therefore, we finally considered only one radius to characterize the warpage. The angular position of the part was controlled by the trace left by the pressure transducers.

Data plotted in Figure 1 show

- A rather low deformation of the parts, even for a high thermal amplitude ($\Delta T = 62°C$ or $\Delta T = -51°C$).
- An increasing deformation vs. ΔT. Then, the part warps toward the hot face.
- A residual deformation of the disks, even in the case of an equilibrated cooling pattern.

The last remark indicates that warpage is not only due to internal stresses but also can be related to other causes. Various authors reported such behavior, which can be attributed to a technological factor implying a deformation of the part during ejection (see Fig. 2). However, it is almost impossible to quantify the forces applied on the part during the opening of the mold and the ejection, as the forces clamping the disk inside the cavity depend on the part's dimensions, which vary with the mold temperature (the shrinkage increases with the mold temperature).

2. Analysis of the Internal Structure of the Parts

Microtomed cuts (25 μm thick) are removed from disks along a radius, normal to the plane of the part. The observation of those cuts under an optical microscope with polarized light revealed a well-known layered structure, characterized in our case by a black line appearing in the core zone. Its position varies with the thermal amplitude ΔT and has been used to characterize the observed structural dissymmetry.

Two different techniques were used for measuring the exact position of the black line from the edge of the part:

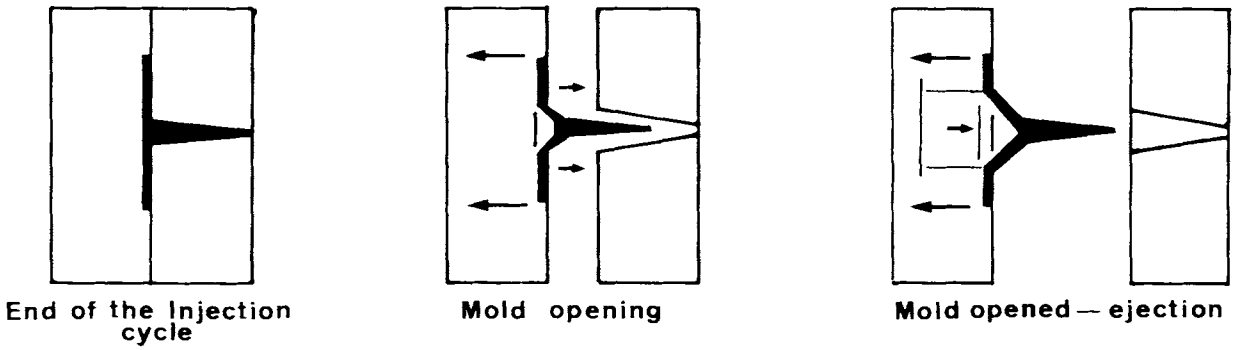

Figure 2 Applied deflection of the part due to ejection.

- one using an optical micrometer mounted on a Zeiss microscope;
- one using an image analysis system mounted on the microscope through a color video camera.

Thus, we quantified the structural dissymmetry by means of a coefficient δe defined as

$$\delta e = \tfrac{1}{2} - L/H$$

where L is the distance between the hot face of the part and the black line, and H, the half-thickness of the part (1 mm).

Figure 3 shows the change of δe measured at three different distances from the gate (30, 45, and 60 mm) vs. the thermal amplitude ΔT.

We observe

- an increasing structural dissymmetry with ΔT (parabolic shape); and
- a lower sensitivity to ΔT for measurements carried out far away from the center of the part.

This can be attributed to the lower melt temperature and also to thermal disturbances caused by geometrical factors (end of the cavity). A calculation, based on the resolution of the heat transfer equation in a stationary case, confirms a lower effect of a thermal amplitude in the mold on a polymer at a lower temperature.

3. Molecular Orientation Measurements

Warpage is a consequence of internal stresses from which we can distinguish molecular orientation (entropic origin) and thermal stresses (energetic term) resulting from heterogeneous shrinkage throughout the part's thickness during cooling.

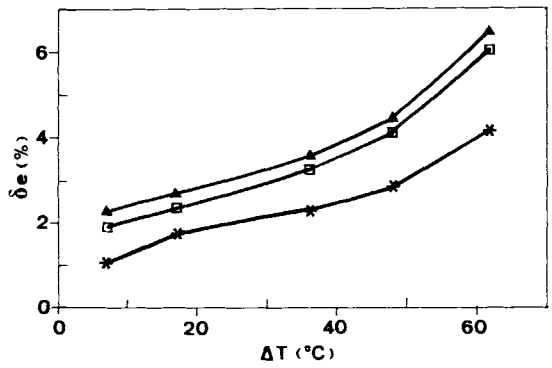

Figure 3 Variation of δe vs. the thermal amplitude inside the mold. Distance from the gate: (▲) 30 mm; (□) 45 mm; (*) 60 mm.

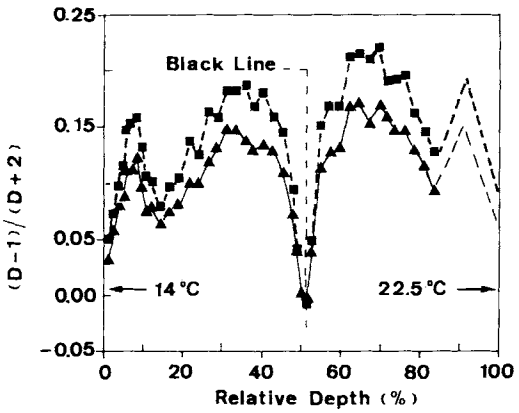

Figure 4 Orientation of amorphous (▲) and crystalline (■) phasis in PP 10/10 from infrared measurements.

In the first step, we observed a structural dissymmetry inside the warped disks, increasing with the thermal amplitude applied in the mold. Molecular orientation was studied using the infrared dichroism technique. Measurements were carried out on microtomed cuts removed throughout the thickness of the part on samples taken at 40 mm from the gate, using an FTIR spectrometer and a polarizer.

We studied in particular, two different absorption bands at 973 cm^{-1} (pure amorphous band) and 998 cm^{-1} (pure crystalline band). The literature[10,11] gives a value of $\alpha = 18°$ for the 998 cm^{-1} band, whereas no value was found in the case of the amorphous band.

Measurements were carried out on microtomed cuts (average thickness 25 microns), removed from the thickness of the part at 40 mm from the gate. Both amorphous and crystalline phases show a similar profile of orientation (Fig. 4), which can be described as follows:

- an increasing orientation from the wall to a depth of about 150 microns, attributed to the filling stage of the mold;
- a zone of relaxation (about 150 microns thick), attributed to the quenching effect of the packing pressure immediately following the end of the filling stage;
- an increasing orientation during the holding phase; and
- a thermal relaxation following the end of the holding stage and leading to an isotropical state of orientation ($P2\langle \cos\theta \rangle = 0$).

Figure 5 shows the different profiles of orientation measured using the 998 cm^{-1} band in four different cases. We observe very similar behavior in all cases,

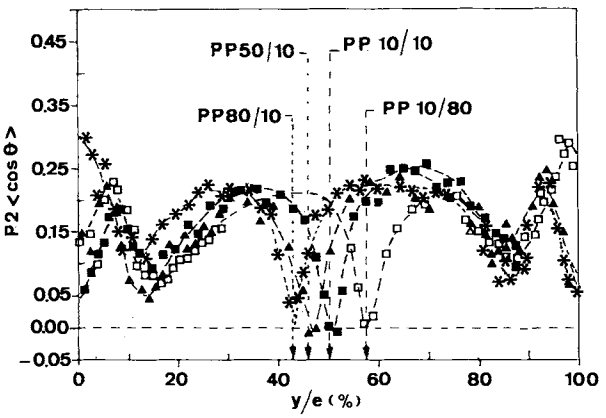

Figure 5 Influence of ΔT inside the mold on orientation of the crystalline phasis.

except in the core region, where the location of the isotropical orientation point is shifted toward the hot face. Then, for the highest temperatures applied to the mold (T = 80°C), the orientation factor is maximum on the wall and does not increase during the filling stage. The calculated interface temperature T_i in that case gives T_i = 92°C, while, because of the very high cooling rate in that region, the crystallization temperature is about 85°C. Therefore, the polymer does not solidify immediately on the wall and is oriented by the flow during the filling stage.

The measurements of orientation in the injected parts reveal a complex distribution throughout the thickness and confirm the observations made by optical microscopy (location of the "black line"). However, even if molecular orientation can be regarded as a term of internal stresses, it has actually no direct effect on the warpage, because it is only a term of "frozen entropy." To complete the study of the origin of warpage, we measured the thermal stresses.

4. Thermal Stress Measurements

Thermal stresses are generated by heterogeneous shrinkage rates throughout the part's thickness during cooling. Several experimental methods were applied to plastic parts to measure them: layer removal, hole drilling, etc.

In the present work, we developed an original method for determining the thermal stress profile inside the part, based on dilatometrical behavior.

Figure 6 shows the different steps of the study of an injected disk:

- Samples of 10 × 5 × 2 mm are removed at 40 mm from the gate to confirm results from optical microscopy and IR measurements as well.
- These samples are microtomed into cuts of 10 × 5 × 0.025 mm.
- A sample of 2 × 0.5 × 0.025 mm is taken on each microtomed slice and is heated in a hot stage between 30 and 100°C at 5°C/mn, under an optical microscope equipped with a video camera.
- An image analysis system computes the dilatometric behavior of the sample (see Fig. 6).

Figure 7 shows the dilatometric results obtained with a given sample submitted several times to the same thermal treatment from 30 to 100°C at a rate of 5°C/mn. We observe a typical relaxation between the first and the second heating, whereas the dilatometric behavior is strictly the same between the second treatment and the following ones.

When the relaxation occurs (typically when the temperature reaches 80°C), both curves from the first and the second heatings are linear and parallel, so that we can measure the constant term σ related to the relaxed term of stress.

Finally, we can express the stress σ by

$$\sigma = E \cdot \varepsilon / (1 - \nu) \qquad (4)$$

where E is the Young's modulus at 80°C; ν, the Poisson coefficient; and ε, the measured thermal strain.

This principle was applied to microtomed cuts removed from the thickness of an injected disk. The effect of cutting and of the thermal treatment on the polymer structure were studied and revealed no influence on the calculated stress.

Figure 8 shows the thermal stress profile obtained on a PP50/10 disk with E = 1300 MPa and ν = 0.3. We observe a parabolic profile, with compressive terms in the skin region and tensile terms in the core region. Then, the maximum of the curve is located exactly on the position of the "black line," which was used to characterize the structural dissymmetry inside the part.

5. Calculation of Warpage

On the basis of the previous experimental results, we propose a simple model to describe the warpage observed on our disks injected in a nonequilibrated mold. We assume that

- warpage is only due to thermal stresses; and
- the thermal stress profile is parabolic and maximum on the exact location of the "black line"

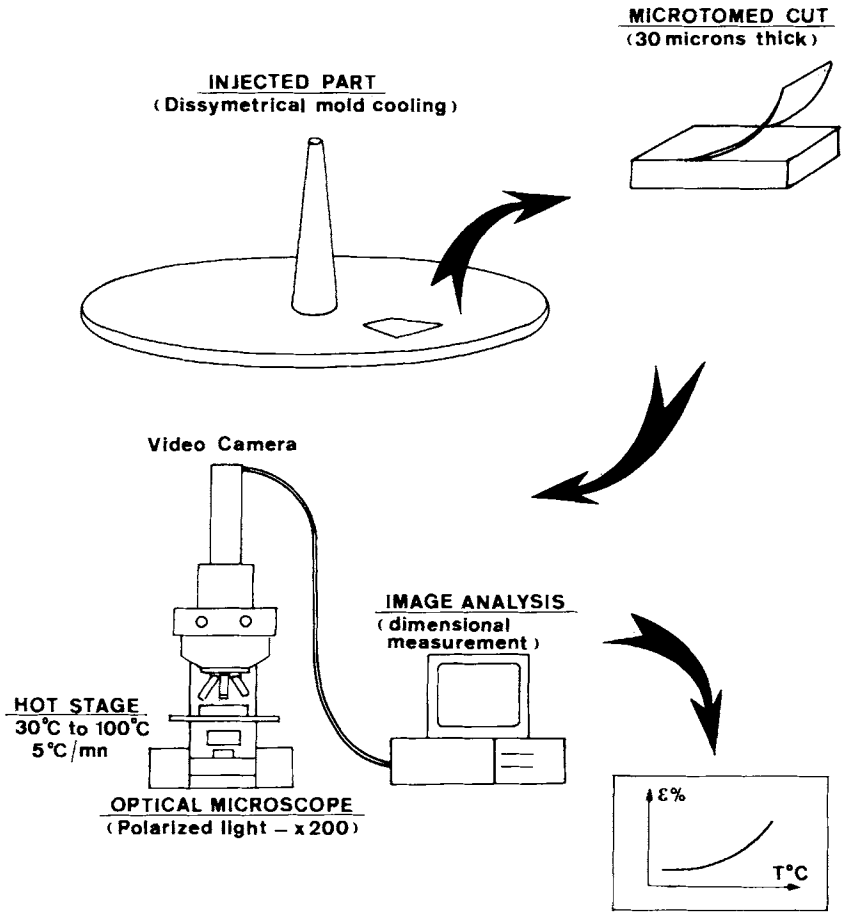

Figure 6 Principle of the analysis of thermal stresses by image analysis.

(see Fig. 9), whose abscissa from the geometrical axis of the part is called "z_n."

Therefore, the internal stress profile can be simply described by

$$\sigma(z) = k_z \cdot z^2 + k_1 \cdot z + k_0$$

$$k_2 = \frac{\sigma_0 - \sigma_1}{(z_0 + z_n)^2}$$

$$k_1 = 2 \cdot (\sigma_1 - \sigma_0) \cdot \frac{z_n}{z_0 + z_n}$$

$$k_0 = \sigma_1 + (\sigma_0 - \sigma_1) \cdot \frac{z_n}{z_0 + z_n}$$

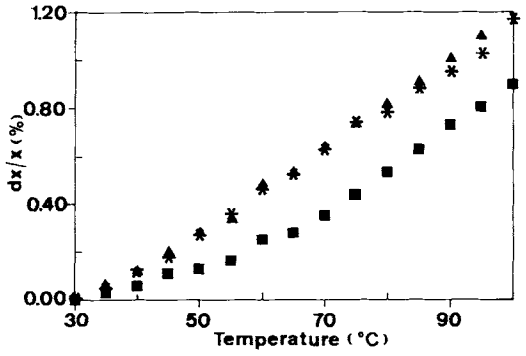

Figure 7 Dilatometrical behavior of a microtomed cut after three thermal treatments. (■) 1st treatment; (▲) 2nd treatment; (*) 3rd treatment.

where σ_0 is the thermal stress on the cold wall of the part; σ_1, the maximum stress located at $z = z_n$; σ_2, thermal stress on the hot wall of the part; and z_0 = half-thickness of the part.

Therefore, σ_2 can be expressed using

$$\sigma_2 = \sigma_1 + (\sigma_0 - \sigma_1) \cdot \frac{z_0 - z_n}{z_0 + z_n}$$

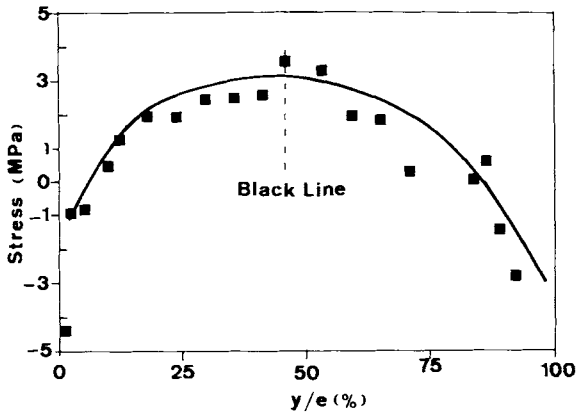

Figure 8 Experimental stress profile in PP 50/10.

The internal stress profile leads to an internal moment M_x defined as

$$M_x = \int_{-z_0}^{+z_0} z \cdot \sigma(z) \cdot dz$$

Assuming that the deformation of the part is due only to the action of M_x, we can express the bending radius of the part R as

$$R = \frac{E}{12(1-\nu)} \cdot \frac{e^3}{M_x}$$

where E is the Young's modulus of the polymer; ν, the Poisson's coefficient (= 0.3); and e, the part thickness.

The part deflection is then calculated using the following formula:

$$f = R - \sqrt{R^2 - r^2}$$

where r is the radius of the part.

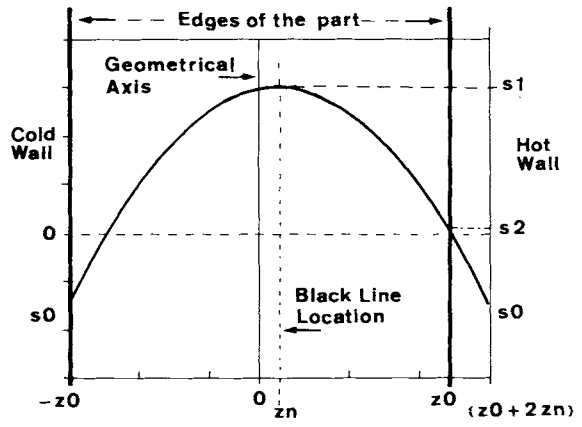

Figure 9 Thermal stress profile assumed for calculation of warpage.

This model cannot describe or explain the residual deformation observed in the case of an equilibriated mold. This defect could be attributed to forces applied on the part during the opening of the mold and ejection.

Figure 10 shows the evolution of the calculated warpage vs. the thermal amplitude applied in the mold. Results are in good agreement with experimental data, from which the constant deformation observed for PP 10/10 is subtracted.

CONCLUSION

Polypropylene disks were injected into a mold with a nonequilibriated cooling pattern to generate warpage. Dimensional measurements revealed an increasing deformation of the parts vs. the thermal amplitude applied in the mold. Then, we observed a residual warpage even in the case of an equilibriated cooling pattern. This defect can be attributed to mechanical deformation during ejection.

Optical microscopy under polarized light showed a structural dissymmetry in the thickness of the part, increasing with an increasing thermal amplitude in the mold. This dissymmetry is characterized by a "black line" in the core region, located at the meeting point of both crystallization fronts coming from each wall in the mold. Its location varies with the thermal conditions applied to the mold and is shifted toward the hot face. Molecular orientation measurements confirmed this dissymmetry and revealed a complex profile inside the part's thickness. An unoriented zone is observed on the location of the "black line."

An original method was set up for measuring the thermal stress profile, responsible for warpage. Based on the study of the dilatometric behavior of microtomed cuts submitted twice to the same thermal treatment, this method has revealed a parabolic

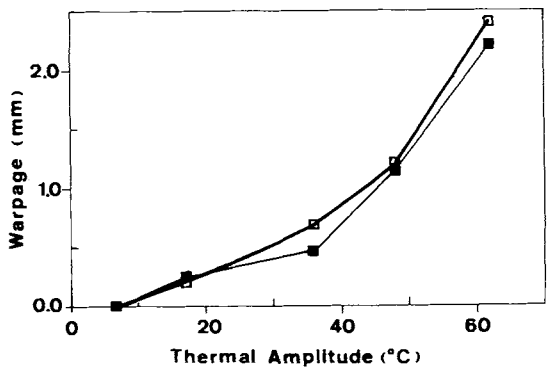

Figure 10 Warpage vs. thermal amplitude: experimental (■) and calculated (□) values.

thermal stress profile in the thickness, with a maximum on the location of the "black line."

On the basis of experimental results, a simple model was developed to calculate the change of warpage vs. the thermal amplitude applied in the mold during processing. Computed results are in good agreement with experimental data.

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