Influence of Talc Concentration on Angular Deformation in Injection Molded Parts of Talc and Rubber Filled Polypropylene

Tadayoshi TAKAHARA*1,*2 / Kiyohito KOYAMA*2

Polypropylene (PP), filled with talc and rubber, is one of the most popular resins used for automotive plastic parts. The influence of talc concentration in PP compounds on the angular deformation of L-shaped specimens is discussed.

The experimental and analytical results are as follows:
1) Angular deformation increases sharply with increasing talc concentration until approximately 10 wt% of talc. The angle reaches a maximum at 10 wt% of talc, and then decreases above the critical concentration.
2) The shrinkage in the in-plane direction of the plate specimens shows a decrease until the talc concentration reaches approximately 10 wt%, and then becomes constant above the critical concentration. On the other hand, the shrinkage in the thickness direction increases sharply until talc concentration approaches 5%, and then decreases gradually above that concentration.
3) The strains for each sample are calculated using Hooke’s Law. In this case, the force that induces angular deformation is assumed to be the difference of shrinkage between the thickness and in-plane directions. The tendencies of calculated strains are quite qualitatively consistent with measured angular deformations.
4) A deformation angle peak can be explained by the difference of shrinkage in the thickness and in-plane directions, which increase sharply until 5 wt% talc concentration. Above this concentration, shrinkage will remain constant despite an increasing flexural modulus.

Key words: Warpage/Angular Deformation/Injection Molding/Polypropylene/Shrinkage/Talc

1. Introduction

Plastic parts are increasingly being used for automotive vehicles to achieve weight reduction, with the consideration of environment protection by decreasing carbon dioxide emissions and improving fuel efficiency. The most popular polymer material used for automotive plastic parts is polypropylene, usually reinforced with talc to improve stiffness, heat-resistance and to obtain lower shrinkage, and filled with rubber to improve impact-resistance. For high quality automotive parts, it is important to prevent warpage resulting from the injection molding process. Almost no detailed studies have been, however, made so far at warpage of injection molded parts of polypropylene compounds containing talc and thermoplastic rubber excepting our previous studies*1-3.

In those papers we focused on two points. First is the shrinkage anisotropy: the difference of shrinkage in the thickness and in-plane directions. It was found that the deformations of molded parts is complex enough to analyze owing to multiple factors, such as thermal shrinkage, elastic recovery, orientation of molecules and/or fillers, and crystallinity in the case of semi-crystalline polymer like polypropylene. The shrinkage anisotropy is caused as a macroscopic phenomenon by these complexly intertwined factors*4. Second point is the three-dimensional shape. Authors focused on the angular deformation of the three-dimensional shape, using the specimen with an L-shaped cross section. We found that the buckling deformation occurs on three-dimensional shapes, only.

Most studies on deformation*5-7 or shrinkage anisotropy*6,8 for injection molded plastics have been limited to the plane shape so far. A few studies*9-12 focused on three-dimensional shapes. Michii et al.10 reported the influence of fiber orientation by using a ribbed plane shape specimen made of glass fiber reinforced PBT. Kobayashi et al.11 reported the effect of the difference inside from outside of the corner, using a S-shaped specimen made of
Table 1 Composition of materials

<table>
<thead>
<tr>
<th>Talc wt%</th>
<th>Resin wt%</th>
<th>Detail composition of resin</th>
</tr>
</thead>
</table>
|         | Polypropylene wt% | rubber wt% | rubber ratio (% of rubber 
|         | (cavity side) | (cavity side) | (PP + rubber) |
| 0       | 100       | 80 | 20 | 20 |
| 2.5     | 97.5      | 78 | 19.5 | 20 |
| 5       | 95        | 76.5 | 18.5 | 20 |
| 10      | 90        | 72 | 18 | 20 |
| 20      | 80        | 64 | 16 | 20 |
| 30      | 70        | 56 | 14 | 20 |

Table 2 Molding conditions (set-up value)

<table>
<thead>
<tr>
<th>conditions</th>
<th>Holding pressure (MPa)</th>
<th>Resin temp. (°C)</th>
<th>Temp. of cooling water (cavity side) (°C)</th>
<th>Injection rate (cm³/sec)</th>
<th>Pressure holding time (sec)</th>
<th>Cooling time (sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A Low holding pressure</td>
<td>20</td>
<td>235</td>
<td>20/20</td>
<td>150</td>
<td>19</td>
<td>20</td>
</tr>
<tr>
<td>B Standard holding pressure</td>
<td>35</td>
<td>235</td>
<td>20/20</td>
<td>150</td>
<td>10</td>
<td>20</td>
</tr>
<tr>
<td>C High holding pressure</td>
<td>50</td>
<td>235</td>
<td>20/20</td>
<td>150</td>
<td>10</td>
<td>20</td>
</tr>
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glass fiber reinforced polyacetal. Yamada et al. focused on the temperature difference between core and cavity using a L-shaped shape specimen made of PA 6. Ammar et al., same as us, focused on shrinkage anisotropy using a double-L-shaped (crank-shaped) specimen made of polypropylene. All of them did not discuss the effect of molding conditions and the material compositions in detail.

Our latest study focused on the effect of injection molding conditions on the poor dimensional stability deformation of 3-dimensional shapes of polypropylene. In this study, the influence of the material compositions, especially talc concentrations will be discussed. The influence of rubber concentration will be published in “Seikei-Kakou” of next month.

First, injection molded specimens with an L-shaped cross section were used to determine the relationships between talc concentration and corner deformation. Secondly, plate specimens were used to determine the correlations between the talc concentration and shrinkage. We performed morphological observations of the cross sections of injection molded specimens by a scanning electron microscope in order to discuss the effect of talc orientation under shear flow on the buckling deformation. Based on these correlations, we will propose a novel mechanism on the angular deformation of injection molded articles from the viewpoint of talc incorporation. Additionally further examinations are carried out for specimens molded under various conditions to confirm the universality of the proposed mechanism.

2. Experimental

2.1 Materials

Ethylene-propylene block co-polymer (MFR = 50 g/10 min for 2.16 kgf at 230°C, flexural modulus 1530 MPa) and ethylene-co-octene co-polymer rubber (EOR, MFR = 9 g/10 min for 2.16 kgf at 230°C, flexural modulus 15 MPa) were used in this study. The densities of polypropylene, rubber and talc are 0.90, 0.87 and 2.7 g/cm³, respectively. As shown in Table 1, six blends were prepared with talc concentrations ranging from 0 to 30% by weight. The weight ratio of polypropylene to rubber was fixed at 80:20.

2.2 Molding conditions

First, molding condition “B” shown in Table 2 was used. And molding condition “A” and “C” were selected to confirm the universality of the novel proposed mechanism.

2.3 Angular deformation test specimens and measurement

The specimens for angular deformation testing were molded by using an electric injection machine (FANUC LTD.; ROBOSHOT S-2000 i 150 B, Maximum clamp force 1500 KN). The geometry and dimensions of the specimen are shown in Figure 1. A fan type gate was set on the edge. The thickness of the specimen was set to be 3 mm, which is typical thickness of automotive plastic parts. The specimens were held at room temperature for about 24 hours to stabilize the dimensions. The corner deformation angle was measured by using the anti-contact RAZAR 3D-measuremet system (PULSTECH INDUSTRIAL CO., LTD.; TDS1623 D). The inner surface of the L-shaped sample was measured to locate the center of the parts. The intersection of these center lines was used to define the corner angle α, as shown in Figure 1. The corner deformation angle φ was calculated using Eq. 1.

\[ φ = 90° - α \]  \hspace{1cm} (1)

For each condition, 2 to 5 specimens were used to ensure
reproducibility.

2.4 Shrinkage specimens and measurement

The plate specimens for shrinkage measurement were molded by using an electric injection machine (TOShiba MACHINE CO., LTD.; IS-130 GN, Maximum clamp force 1280 KN). The geometry and dimensions of the plate specimen are shown in Figure 2. A fan type gate was set on the edge. The thickness of the specimen was 3 mm. The treatment of molded samples until measurement was the same as for the angular deformation specimens.

The shrinkage was measured in three directions, i.e. thickness direction \( S_{\text{t}} \), flow direction in \( S_{\text{w}} \) and transverse direction to flow \( S_{\text{m}} \).

\( S_{\text{w}} \) was calculated by using Eq. 2 as follows:

\[
S_{\text{w}} = \frac{L_{\text{w}} - t}{t_0}
\]

Specimen thicknesses \( t \) was measured by using a micro-meter (Mitutoyo Co.; 406-511) at the points \( T_1 \) and \( T_2 \) shown in Figure 2. Cavity thickness \( t_0 \) was measured by the same method reported in our previous study\(^{10}\). The average value of two points was used.

\( S_{\text{w}} \) and \( S_{\text{m}} \) were calculated by using Eq. 3 and Eq. 4:

\[
S_{\text{w}} = \frac{L_{\text{w}} - L_{\text{w,md}}}{L_{\text{w}}}
\]

\[
S_{\text{m}} = \frac{L_{\text{m}} - L_{\text{m,md}}}{L_{\text{m}}}
\]

Specimen length was measured with an optical microscope (Nikon Co.; Industrial Microscope Type 2, scale 100 : 1) along the lines L1 to L4 shown in Figure 2. The mold dimensions \( L_{\text{w}} \) were measured previously, which are the same points reported in our previous study\(^{9}\). The average value of four lines was used.

2.5 Observation of cross section microstructure

To examine the effect of morphology of talc orientation on the angular deformation stability, scanning electron microscope (Hitachi, Ltd.; S-4800 and S-3700) observations were carried out for cross section of the specimens.

3. Results and discussion

3.1 Talc concentration effect on angular deformation

Figure 3 shows the correlation between talc concentration and deformation angle in the L-shaped specimens shown in Figure 1. It has been considered so far that the shrinkage anisotropy between the thickness and the in-plane directions increased in proportion to talc concentration, resulting in an increase of corner deformation. However, we should note that deformation angle shows a peak at approximately 10 wt% of talc content. We shall return to this point later.

3.2 Discussion of the tendency of shrinkage

Figure 4 shows the shrinkage in three directions as a function of talc concentration of the plate specimens shown in Figure 2.

The shrinkage in the in-plane directions \( S_{\text{w}} \) and \( S_{\text{md}} \) are lower than those in the thickness direction \( S_{\text{t}} \). Diez-Gutierrez et al.\(^{11}\) reported similar tendencies in linear thermal expansion coefficient not for molding shrinkage. Talc platelets generally have a high aspect ratio.
The elementary platelets are stacked on top of one another. The platelets slide apart during extrusion compounding process and tend to orient parallel to both flow and transverse direction during mold filling due to weak binding forces (Van der Waal’s forces) linking one platelet to its neighbors. These strong orientations in MD and in TD are evident in Figure 5. At center area of thickness of about 0.2-0.3 mm, however, orientation of talc is not parallel to the flow direction. This arrangement of talc can be assigned to a fountain flow effect. From these micrographs, talc orientation is illustrated in Figure 5, schematically. Figure 6 shows the strong orientations of all six talc concentration specimens. These parallel-oriented talc platelets restrict the polymer shrinkage in the in-plane direction, and result in lower shrinkage in the in-plane directions. And $S_{100}$ is slightly higher than $S_{0}$. 

**Fig. 5** SEM images of talc orientation in MD and TD, 20 wt% talc concentration specimen

**Fig. 6** SEM images of talc orientation in MD for each six talc concentrations
One reason is the molecular orientation that restricts the shrinkage in flow direction\textsuperscript{46}. Another reason is the talc shape. Major axis of each talc particle orients to flow direction like glass fibers. This phenomenon is schematically illustrated in Figure 7.

The above is the mechanism of anisotropic shrinkage. Regarding the mechanism of the influence of talc concentration to anisotropic as follows.

As shown in Figure 4, $S_{md}$ and $S_{td}$ gradually decrease with an increase of talc concentration and become almost constant at high talc concentration. On the other hand, $S_{nd}$ increase very sharply until talc concentration of 5 wt %, followed by slow decrease above the concentration.

First, we will discuss variations of $S_{md}$ and $S_{td}$ with talc concentration. The restriction effect of oriented talc decreases with increasing talc concentration, depressing the shrinkage change. Furthermore talc acts as a nucleating agent for crystallization of polypropylene\textsuperscript{17}. This causes the volumetric shrinkage to increase, so that $S_{md}$ increases with increasing talc concentration. It is, however, well known that the enhancement effect of talc on crystallization saturates at small dose of talc. As a result increase of $S_{md}$ is gradually suppressed as talc content increases and shrinkage becomes constant. The total shrinkage in the thickness direction gradually decreases, since the polymer ratio decreases as talc concentration increases. Talc does not shrink, however only polymer material shrink. The substantial shrinkage based on polymer volume $S'_{nd}$ was calculated by using Eq. 5 as follows:

$$S'_{nd} = \frac{S_{nd}}{V_{polymer}}, \quad V_{polymer} = \frac{W_p}{W_t} + \frac{W_p}{0.9} + \frac{W_r}{0.87}$$

$V_{polymer}$ is the volume ratio of polymer material, $W_t$, $W_p$ and $W_r$ are weight percentages of talc, polypropylene and rubber, respectively. Figure 8 shows that the substantial shrinkage $S'_{nd}$ becomes constant. These phenomena corresponding to talc concentration are schematically illustrated in Figure 9.

Until talc concentration of 5 wt%, size of specimens in thickness direction decrease because shrinkage in thickness direction $S_{td}$ increases, on the other hand size of specimens in transverse direction increases. Above con-

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**Fig. 7** Schematic illustration of anisotropic shrinkage in three directions

**Fig. 8** Substantial shrinkage based on polymer volume

**Fig. 9** Schematic illustration of anisotropic shrinkage in TD and ND
centration of 5 wt%, shrinkages in both directions become constant. Therefore, until about 5 wt% anisotropic shrinkage increases and above that concentration, that becomes constant.

3.3 Discussion of the reason for the peak in angular deformation

Previous studies\(^1\) showed the mechanism for shrinkage anisotropy of 3-dimensional molded articles: the difference between shrinkage in the thickness and in-plane directions results in angular deformation. This mechanism\(^2\) is illustrated schematically in Figure 10. This illustration shows an example of the cross section of an L-shaped specimen. If the shrinkage in the thickness direction is larger than that in the in-plane direction, the lower left angle of the orange trapezoid area, \(\alpha\), changes to the smaller angle, \(\alpha'\). Thus to keep the connectivity of each trapezoid part, the original shapes must change to the buckling shapes with corner angle less than 90°.

Based on this mechanism, the difference of the shrinkage gives rise to a virtual force (\(\sigma_{\text{aniso}}\)). Eq. 6 shows the relationship between this virtual force (\(\sigma_{\text{aniso}}\)) and the difference of shrinkage.

\[
\sigma_{\text{aniso}} \propto S_{\text{th}} - S_{\text{in}}
\]  

(6)

where \(S_{\text{th}}\) is the shrinkage in the thickness direction (\(S_{\text{th}}\)) and the shrinkage in the plane direction as \(S_{\text{in}}\) (\(S_{\text{th}}\) or \(S_{\text{in}}\)).

Considering the gate position, \(S_{\text{in}}\) is used in place of \(S_{\text{th}}\) in this discussion. Therefore Eq. 6 can be expressed in the form of Eq. 7.

\[
\sigma_{\text{aniso}} \propto S_{\text{th}} - S_{\text{in}}
\]

(7)

Figure 11 shows the calculated shrinkage difference, i.e. \(\sigma_{\text{aniso}}\) using Eq. 7 as a function of talc concentration.

Now we propose daring hypothesis using stress-strain relationship of the molded article in order to discuss the effect of talc on the buckling deformation, since the angular deformation results from the difference of the shrinkage, i.e. virtual force. Using the Hooke’s Law, angular deformation at talc concentration of a wt% can be expressed by Eq. 8.

\[
\varepsilon_c = A \times \frac{\sigma_{\text{aniso}}}{E_c}
\]

(8)

\(\varepsilon_c\) is a strain and from above hypothesis, that is equivalent to an angular deformation. \(E\) is Young’s Modulus, \(\sigma_{\text{aniso}}\) is the force for angular deformation and \(A\) is a proportional constant. In this study, the flexural modulus is used as an equivalent to Young’s modulus. Figure 12 shows the relationship of the flexural modulus plotted against talc concentration. Flexural modulus is measured at ambient temperature, however, the angular deformations occur during the molding process changing
from molten state to ambient state, therefore the stiffness is much softer than measured flexural modulus, however that is proportional to the modulus of all state. Therefore proportional constant $A$ is needed in Eq. 8.

Figure 13 shows the calculated strain using Eq. 8, which qualitatively corresponds to corner deformation angle, where we assumed that the difference of shrinkage from Figure 11 is substituted for virtual force $\sigma_{\text{inert}} \cdot E$, is equal to flexural modulus from Figure 12, and constant $A$ is set as 1. Comparing the calculated strain in Figure 13 to the measured angle in Figure 3, it is clear that the tendency coincides quite well.

Thus, the angular deformation peak corresponding to a certain talc concentration can be explained as follows. At low talc concentration ($< 5$ wt%), as a result of the rapid increase of the difference of shrinkage, the deformation angle increases sharply with increasing talc concentration. Above talc concentration of 5 wt%, the difference of shrinkage, i.e. virtual force $\sigma_{\text{inert}}$ for angular deformation, becomes almost constant, while flexural modulus, the denominator of Eq. 8, increases continuously, so the deformation angle changes degrease.

We demonstrated that the tendency of buckling angular deformation for material composition can be explained by using Eq. 8, assuming that the force for deformation is the difference of shrinkage in thickness and in-plane directions. This is a novel proposed mechanism.

### 3.4 Confirmation of the proposed mechanism in other molding conditions

The universality of the proposed mechanism is confirmed. As a variable of molding conditions, the holding pressure was chosen, because our previous study showed that the holding pressure was the most influential factor. Corner deformation angles of low holding pressure condition (condition “A” shown in Table 2) and high holding pressure condition (condition “C”), are calculated as well as condition “B”. First, shrinkage in 3-dimensions is measured using plate specimen and the difference of shrinkages are calculated by using Eq. 7. Figure 14 shows the measured shrinkage and the calculated shrinkage differences. Next, the strains are calculated by using Eq. 8. Figure 15 shows that, in all three holding pressure conditions, the calculated strains coincide quite well with the measured angles. That is, (1) the peak concentrations almost coincide, (2) the deformation angles increase sharply when the talc concentration approaches a certain value and thereafter the deformation angles decrease slowly, (3) the value of corner deformation angles at the peak in different holding pressure conditions increases in the following order: Low (condition “A”) >
Standard (condition “B”) > High holding pressure (condition “C”).

4. Conclusions

For talc and rubber filled polypropylene, the influence of the talc concentration on the angular deformation was studied and the following conclusions were drawn:

1) In an L-shaped cross section specimen, the corner deformation angle increases sharply at low talc content (<10 wt%), and then decreases after reaching a peak talc concentration of 10 wt%.

2) In plate specimens, shrinkage in the in-plane directions ($S_{in}$ and $S_{on}$) is lower than those in the thickness direction ($S_{on}$). Oriented talc particles restrict the shrinkage of the directions, which are parallel to the major surface of the particles. Shrinkage in the in-plane direction ($S_{in}$ and $S_{on}$) decrease when talc concentration approaches 10 wt%. Above the peak concentration, shrinkage level off with an increase of talc. On the other hand, the shrinkage in the thickness direction ($S_{on}$) increases sharply until 5 wt%, then decrease as increasing talc concentration. Although talc acts as a nucleating agent and induces large volumetric shrinkage, the effect is saturated with a very small amount of concentration. That is a reason why shrinkage increases sharply at lower talc concentration and saturate.

3) The strains for each sample are calculated using Hooke’s Law. In this case, the force that induces angular deformation is assumed to be the difference of shrinkage between in thickness and in-plane directions. The tendencies of calculated strains are quite well consistent with measured angular deformations qualitatively. This is a novel proposed mechanism.

4) A deformation angle peak can be explained by the difference in shrinkage, which increase sharply until 5 wt% talc concentration. Above this concentration, shrinkage will remain constant despite an increasing flexural modulus.

References


タルク・ゴムを含有するポリプロピレン射出成形品におけるタルク含有量の角部閉じ変形への影響

高 原 悠 良・小 山 清 人

タルク・ゴムを含有するポリプロピレンは、最も一般的な自動車用射出成形樹脂材料である。この樹脂で成形した、断面がL字形状の成形品において、そのL字部の角度の閉じ変形量に関するタルク含有量の影響を検討し、以下の結果を得た。

1）角部閉じ変形量は、タルク含有量10 wt%近辺まで、急激に増加し、その後ゆるやかな減少を示す。

2）平板において、3轴方向の成形収縮率とタルク含有量の関係を検討した結果、平面方向の成形収縮率はタルク含有量10 wt%近辺まで減少し、その後もゆるやかではあるが減少を続ける。一方、厚み方向の成形収縮率はタルク含有量5 wt%近辺まで急激に増加するがそれ以降はゆるやかな減少に転じる。

3）厚み方向と平面方向の成形収縮率の差が、角部変形を引き起こす力に相当すると仮定し、Hookeの法則からタルク含有量ごとに歪量を求めた。この歪量は、実測の角部閉じ変形量の傾向と良く一致した。

4）角部閉じ変形量がタルク含有量に対して極大を持つ理由は、成形収縮率の異方性により説明が可能である。すなわち、角部変形を引き起こす力に相当する成形収縮率の異方性は、タルク含有量5 wt%近辺までは増加しその後一定となるが、材料の変形のしにくさに相当する弾性率は一定に増加する傾向があるためである。