DE GRUYTER J Polym Eng 2018; aop

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Influence of talc and rubber contents on surface replication of polypropylene injection molding application to automotive plastics

https://doi.org/10.1515/polyeng-2018-0251
Received August 8, 2018; accepted November 19, 2018

Abstract: The influence of talc and rubber contents included in polypropylene (PP) on surface replication properties for microstructure was analyzed. By increasing the talc and rubber contents, surface replication on the injection-molded parts has been reduced. From the surface observation, an uneven microstructure surface was formed for a material containing talc during the shrinkage process. It was confirmed that if the PP resin contains talc, then the correlation between glossiness of the mold and glossiness of the molded product was lost and glossiness was decreased. On the contrary, as the rubber content increased, the replication properties improved. At the same time, generation of the streak pattern in the transferred area, which was generated in the base PP, decreased and glossiness increased. Moreover, the fine uneven structure of the nontransferred area, as observed in the talc-containing material, was not observed in the rubber-containing material.

Keywords: automotive plastics; injection molding; microstructure; polypropylene; talc and rubber.

Abbreviations

DMA	dynamic mechanical analyzer
EDX	energy dispersive X-ray spectroscopy
MFR	melt flow rate
PP	polypropylene
SEM	scanning electron microscope

1 Introduction

Currently, polypropylene (PP) is largely used as a resin material in automobiles. With the study proposed by Nomura et al. [1] in 1994, the performance of PP has improved significantly. Nomura et al. developed polymer alloys for improving conventional PP. Based on an inversed sea-island (salami-like) structure, they developed PP materials with high flow ability, high rigidity, and high impact resistance. In addition, the PP materials possessed high crystallinity and low molecular weight. They were able to achieve micro-crystallization of PP by using a PP-compatible elastomer copolymer. The developed PP exhibited an improved strength in comparison with conventional PP. In resin materials that are currently used in automobiles, many bumpers and instrument parts, which are representative resin parts, are often made using PP blended with approximately 10%–20% talc or rubber using this technology. As the scope of the application continues to expand and its global use progresses, the polymerization techniques and blending technologies continue to progress.

With the recent advancements in PP, the use of automobile parts that are made of resin has increased. Many resin parts of automobiles are obtained using injection molding. Microinjection molding technique has also been developed, which uses a micromachined (surface-textured) die in the injection molding for improving its functionality. In addition, ultrafine structures of various patterns on the surface of a molded product can be obtained using microinjection molding. Since its publication in 1996, microinjection molding has been widely studied by many researchers. Many plastic products with a sub-microscale surface structure have been developed for the micromolding technology in injection molding, including optical disks. Such technologies are technically advanced precision transfer molding, following DVD and Blu-ray DVD in recent years. Therefore, there have been many discussions on the surface transferability and moldability [2–5]. From the viewpoint of the relation between a material's characteristics and transferability, there have been discussions on the viscosity of molten resins,

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changes in the modulus of elasticity of cooled resin during molding, and transfer mechanism by deformation of a skin/shear layer [6–8]. Moreover, in general, in injection molding methods, a high transfer rate has been demonstrated to the concave surface with a depth of 36 μm and an arc diameter of 200–300 μm [9]. However, Sato et al. [10] and Kato [11] verified the transfer rate and showed that the limit area of charging resin into a conical concave portion was approximately 10 μm in diameter. For this reason, research was conducted to slow the resin solidification, and on how to complete the transfer before the solidification of the resin. Previously, for improving the transfer, many studies have been reported on ultrahigh-speed injection molding, injection compression molding, vacuuming in the mold, high-temperature molding, CO_2 gas injection molding, and ultrasonic vibration molding, etc. [12–18].

Ham et al. [19] visualized filling patterns of V grooves using ultrahigh-speed injection molding. They reported on the impact of injection rate on transcription. In addition, changes in the transfer rate by arranging stamper's groove and effects of filling rate and pressure in the mold, etc., has been studied in detail. Ito et al. conducted research on the adhesion between the resin and the mold wall using micro structure processing technology [20–26]. Various studies on microinjection molding have been reported. Many researches have been performed using amorphous materials. In recent years, studies on crystalline materials have increased as well [27, 28]. Jiang et al. [29] obtained PP microgears and evaluated their surface mechanical properties by conducting structural analysis and nanoindentation. Furthermore, Maksims et al. [30] evaluated the heat transfer coefficient (HTC value) of PP and mold in micro-injection molding by conducting experiments and simulations. Ito et al. [31] and Taki et al. [32] reported the internal structure of PP and the effect of crystallization on transcription. In addition, Takai et al. [33] evaluated the influence of PP molecular structure and viscosity properties on fine transfer and reported that the low strain fluidity is due to transfer rate. Although some reports based on the fine transferability by injection molding of crystalline resin materials including PP have been reported, there are no reports on the influence of talc or rubber-blended PP materials on the fine transfer of PP.

On the contrary, there is an increase in the need for matt finish for resin surface with a reduced glossiness to enhance the texture of the product. In addition, a study was reported on the visual influence of the glossiness [33]. To obtain a product surface with low glossiness, it is necessary to precisely transfer a fine structure measuring a few micrometers due to unevenness of a mold surface. It can be easily expected that the talc or rubber components at microscale blended in PP affect the microtransfer. Therefore, understanding the influence of the talc or rubber components blended in PP on the surface transfer not only can improve texture improvement but also can reduce transfer failure in mass production and application to advanced decorativeness and functionalities.

In this study, we focused on the microtransfer of PP resin materials containing talc or rubber, which are widely used in automobiles. We used pristine PP as a base PP, and resin materials with various contents of talc component and/or rubber component were varied to study the influence of respective components on the transfer and to investigate the influence of the glossiness on the resin surface.

2 Materials and methods

2.1 Material

We used the base PP, which is PP before adding the talc and rubber components (NEWCON, NBX03HRS manufactured by Japan Polypropylene Corporation, Marunouchi, chiyodaku, Tokyo, Japan), talc (MAT-725TP manufactured by Shiraishi Calcium Kaisha, Ltd., Nakanoshima, Kitaku, Osaka, Japan), and rubber (LC170 manufactured by LG Chem, Ltd., Yeoui-daero, Yeongdeungpo-gu, Seoul, Korea) as test samples. Keeping the base PP before adding the talc and/or rubber components as the standard, we obtained a sample by adding 5%–20% talc only and a sample by adding 5%–20% rubber component only.

2.2 Mold

To study transferability around the gate and flow end that can occur in actual molded products, we used a test piece (320 × 80 mm and $t = 2.5$ mm), as shown in Figure 1, and evaluated the transferability at the position that was 300 mm away from the gate. In actual bumper injection molding for automobiles, the flow length from one gate is often approximately 300 mm. In this study, we selected and evaluated the region where filling pressure is low. The measurement site was designed as a nesting type to enable the evaluation by exchanging the plurality of nesting patterns. The material of the nesting mold (HPM38 manufactured by Hitachi Steel Co., Ltd., Minato-ku, Tokyo, Japan) and fine machining was performed in a range of 13 × 13 mm toward the center of the nesting. For fine machining of the

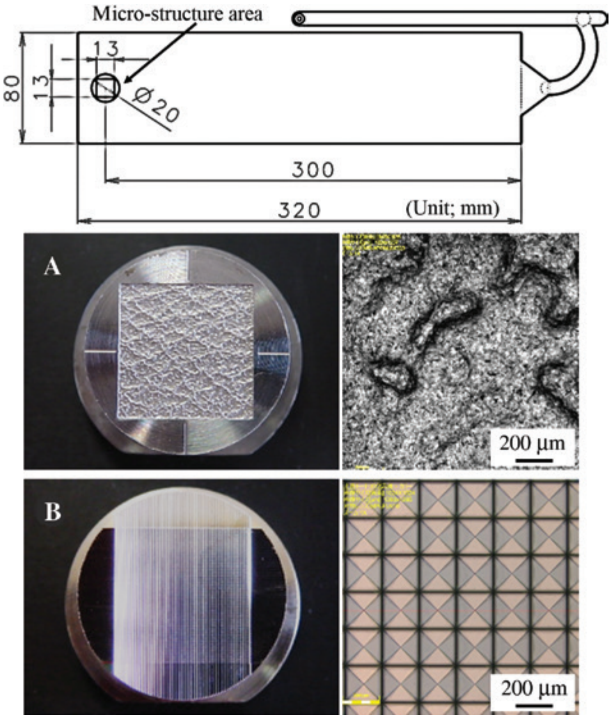

Figure 1: Shape of molded samples and pictures of mold insert with microstructure surface produced. (A) Edging process. (B) Ultra-precision cutting process.

mold surface, two types of fabrication methods were used. For the purpose of understanding the relation between fine transfer and the glossiness in the actual molding, leather grain patterns that are widely used in automobiles were reproduced in pattern 1. To accurately evaluate the transferred and nontransferred portions, pyramid shapes were obtained in pattern 2 using ultrafine machining because there is a possibility that unintended shapes may be formed by edge processing.

As shown in Figure 1A in the leather grain pattern of pattern 1, the fabrication was done for a one-dimensional structure of approximately 200- and 100-µm depths and a fine uneven two-dimensional structure of approximately 5–20 µm. Surface roughness value (Ra) of the two-dimensional structure was varied between 0.05 and 0.20 µm in 10 intervals to change the glossiness of the mold from 9.7 to 1.6. When Ra was increased by sandblasting, a fine uneven structure was formed. As a result, diffused reflection of light increased and the glossiness decreased. On the contrary, when Ra was reduced using glass bead blasting, that is, resulting in a smoother surface, the fine structure of the mold surface was reduced. As a result, diffused reflection of light decreased and the glossiness increased.

In pattern 2 that assumed the reproduction of the representative shape and dimensions of the leather grain pattern, as shown in Figure 1B, a pattern shape of a quadrangular pyramid with a side 200 µm and a height of 100 µm was obtained using ultra-precision cutting and nickel electroforming.

2.3 Injection molding machine and molding conditions

We used the 110T injection molding machine (FN2000-25A manufactured by Nissei Plastic Industrial Co., Ltd., Nagano, Japan) with the following conditions: maximum injection speed of 100 mm/s, maximum injection pressure of 220 MPa, clamping force of 1098 kN, screw diameter of 35 mm. Assuming molding conditions of general automobile resin parts and setting the speed value to 50 mm/s, the dwelling pressure value to 20 MPa, the resin temperature value to 200°C, and the mold temperature value to 40°C, materials with different contents of the talc and rubber components were molded under the fixed molding conditions. Injection conditions were set like the average conditions in actual bumper injection molding.

2.4 Measurement and evaluation

Using the mold of pattern 1, where the fine fabrication of the mold surface was carried out by edging and using the base PP before adding the talc and rubber components as a standard, we evaluated the relation between the glossiness of the mold and that of the molded product, when only the talc component was added and when only the rubber component was added, using a gloss meter (GM-268 Plus manufactured by Konica Minolta Corp., Tokyo, Japan). At the same time, the transfer state of the mold shape onto the resin was measured by analyzing the fine shapes of the mold and the molded product with a coherence scanning interferometer (New View 7300 manufactured by Zygo Corp., Berwyn, PA, USA), laser microscope (LEXT OLS4100 manufactured by OLYMPUS Corp., Tokyo, Japan), and scanning electron microscope (SEM TM3030Plus manufactured by Hitachi High-Technologies Corp., Tokyo, Japan).

For pattern 2, where the quadrangular pyramid was fabricated using ultra-precision cutting to evaluate transferred and nontransferred portions, a shape measurement was performed using the coherence scanning interferometer and the laser microscope, and the surface state of the nontransferred portion was evaluated using the SEM.

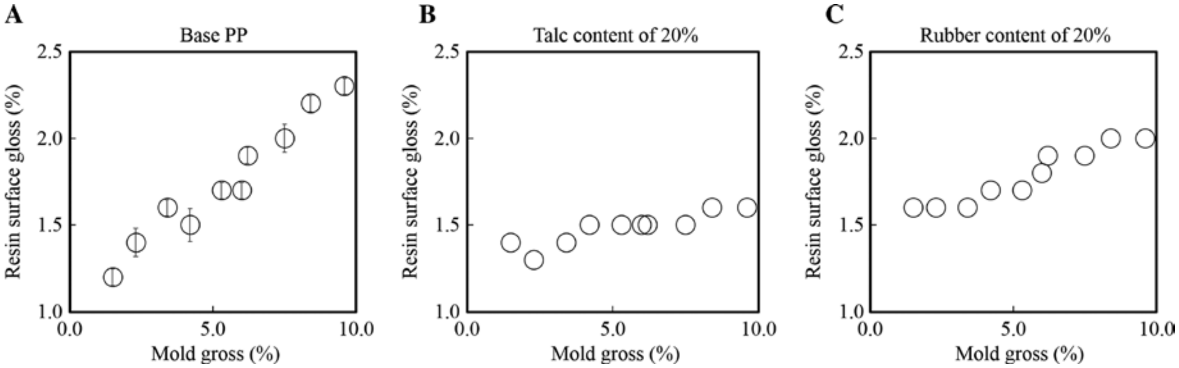

Figure 2: Relation between the metallic mold gross and the resin surface gross. (A) Base PP, (B) PP + talc content, (C) PP + rubber content.

2.5 Viscoelasticity measurement

To evaluate the flowability of the talc and rubber materials containing the base PP, a viscoelastic analyzer (MCR301 manufactured by Anton Paar Corp., Graz, Austria) and a dynamic mechanical analyzer (DMA) (RSA3; TA Instruments-Waters LLC, New Castle County, DE, USA) were used for measurement, evaluating the influence of adding the talc and rubber components to the base PP on shear viscosity and dynamic viscoelasticity.

2.6 Component analysis by energy dispersive X-ray spectroscopy (EDX)

To evaluate the influence of the talc and rubber components on the surface condition of the nontransferred portion, the nontransferred portion was evaluated using a component analyzer energy dispersive X-ray spectroscopy (EDX) (TM3030Plus manufactured by Hitachi High-Technologies Corp.) in combination with the SEM analysis of the nontransferred portion.

3 Results and discussion

3.1 Measurement results of glossiness of the mold fabricated by edging and glossiness of products

We molded using a nesting mold, changing the glossiness of base PP by edging, sandblasting, and glass bead blasting. The molded products were obtained using continuous molding, and measurements were taken using the molded products in the stable range after 10 shots as samples. As shown by the measurement results in Figure 2A, the glossiness of the products changed linearly with a change in glossiness of the mold. This is thought to be possible because the fine uneven structure of the mold was transferred to the surface of the molded product and thereby the glossiness was uniformly changed. Accordingly, we can suggest that, in the case of the base PP, the glossiness of the product may be controlled by the glossiness of the mold.

3.2 Influence of the talc content on glossiness

Figure 2B shows the influence of the talc content on the glossiness of the product. Using a material containing 20% of talc in the base PP, a similar molding experiment was conducted. As a result, the glossiness of the product decreased with an increase in the talc content, and the relation of the glossiness of the molded products with a change in the glossiness of the mold was lost. Figure 3

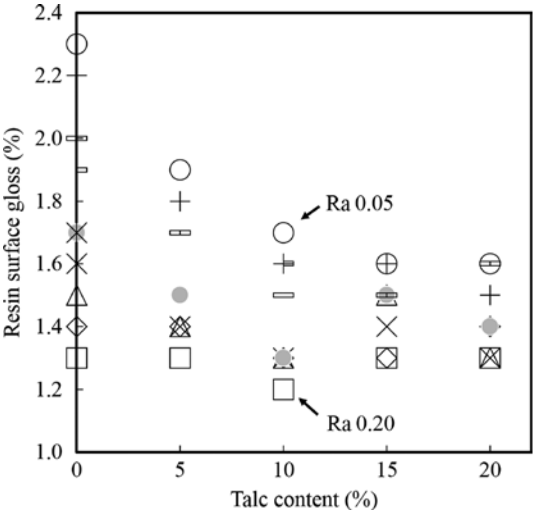

Figure 3: Relation between the material content and the resin surface gross.

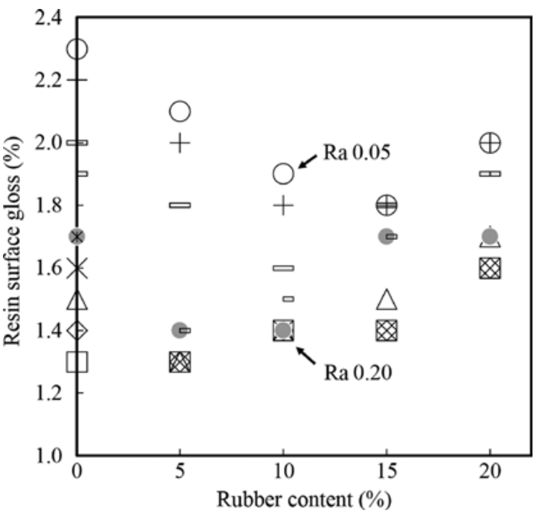

Figure 4: Relation between the material content and the resin surface gross.

shows the relation between the talc content and the glossiness of the product. As shown in the figure, when the glossiness of the mold was as high as 9.7 (Ra = 0.05 μm), the glossiness decreased as the talc content increased. However, when the mold had a low glossiness value of 1.6 (Ra = 0.20 μm), the glossiness of the products hardly changed even when the talc content increased. On the other hand, the minimum value of the gloss of the resin surface occurred at the talc content of 10%. This is presumed to be a range of variations because the gloss value was averaged and the standard edition of the whole was 3 σ at a value of approximately 0.13.

3.3 Influence of the rubber content on glossiness

Figure 2C shows the influence of the rubber content on glossiness of the product. Using a material containing 20% rubber per base PP, a similar molding experiment was conducted. Being different from the talc-containing PP, the relation between the glossiness of the mold and the glossiness of the product was not lost. Figure 4 shows the relation between the rubber content and the glossiness of the product. When the mold had a low glossiness value of 1.6 (Ra = 0.20 μm), the glossiness of the rubber-based product increased as the rubber content increased, which is different to the result obtained for the talc-based product. When the mold had a high glossiness value of 9.7 (Ra = 0.05 μm), the glossiness of the product decreased with increase of the rubber content up to 15%. However, the glossiness of the product increased with a further increase of the rubber content of 20%. This is shown in detail in the experiment in Section 3.7. The inclusion of rubber at 15% or more tends to greatly improve transferability. It is presumed that the improvement of transferability is a factor that increased the glossiness.

3.4 Measurement results of the surface shape of the mold and molded products

For the purpose of evaluating the transferability of the mold and the products, the surface of the molded product, which was the same as the portion of the mold surface having glossiness of 9.7 (Ra = 0.05 μm), was measured using a coherence scanning interferometer. The results are shown in Figure 5. For the molded products, the samples of the base PP with 20% of talc and 20% of rubber were evaluated. Approximately 200 μm of the representative one-dimensional structure was transferred for the base PP. On the contrary, the transfer in the one-dimensional structure significantly decreased for the molded product containing 20% of talc. In addition, approximately 5–20 μm of the fine uneven structure was widely observed in the nontransferred area, which was not observed in the base PP sample. On the contrary, the one-dimensional structure was accurately transferred for the molded product

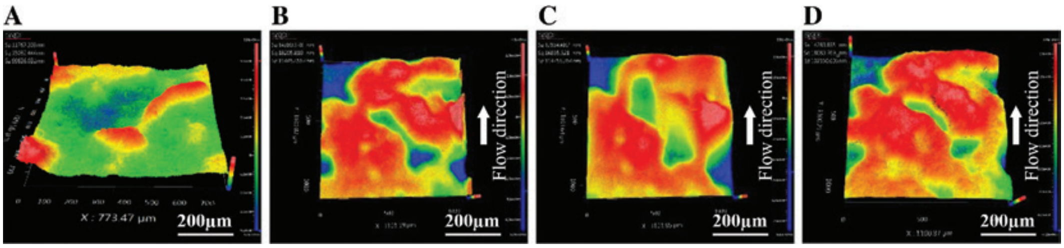

Figure 5: Observed surface temperatures distribution and shape. (A) The mold surface, (B) base PP, (C) PP + talc content 20%, (D) PP + rubber content 20%.

containing 20% of rubber, which indicates good transferability even in comparison with the base PP.

Accordingly, although the one-dimensional structure of a few hundreds of micrometers was accurately transferred in case of the base PP, transferability of the one-dimensional structure seemed to decrease when the base PP contained the talc component. In addition, we can infer that, as the transferability of the one-dimensional structure deteriorated, the portion around the talc component was contracted in the nontransferred portion during the contraction process of the base PP. As a result, a fine uneven structure was formed according to dispersion of the talc particles. Therefore, although transferability worsened as the talc content increased, the fine uneven structure was formed in the nontransferred portion, and thereby, the value of glossiness decreased. On the contrary, as the rubber content increased, transferability of the one- and two-dimensional structures seemed to improve. The rubber contracted similarly to PP in the contraction process of the base PP so that there was no fine uneven structure formed (like in the talc case), and the value of glossiness of the product changed according to the change of the value of glossiness of the mold.

3.5 Ultra-precision cutting mold and transferability measurement results

To accurately separate and evaluate the mold-transferred and nontransferred areas, a mold having a fine pattern with a representative shape of quadrangular pyramid was obtained by ultra-precision cutting. The shape and dimensions of the prepared mold were measured using a laser microscope. It was confirmed that the side and the height of the microstructure were successfully obtained with a precision of ±3 μm with respect to the design values (side of 200 μm and height of 100 μm). Using the mold with this microstructure, we performed molding similar to that using the edging mold and observed the states of the transferred and nontransferred areas.

3.6 Influence of the talc content on transferability

Similar to the edging mold, molding was performed using materials with an increased talc content in the base PP. Figure 6 shows a laser microscope image of the molded products containing the base PP, talc, and the rubber components. Figure 7A and B shows an SEM image of a sample containing the base PP and 20% of talc. As a result of surface observation with the laser microscope and SEM, it was found that a few tens of micrometers of the fine uneven structure were irregularly formed in the nontransferred area. This was not observed in the nontransferred area of the base PP. Furthermore, we conducted a component analysis using EDX built in the SEM to analyze the fine uneven structure. As shown in Figure 8, Si and Ca were widely detected; and we were able to confirm that those components were from the talc material. From this result, we confirmed that the transferability decreased with an inclusion of talc content, and about a few tens of micrometers of the fine uneven structure was formed in such a way that dents were formed around the talc particles. This fine evenness is considered to be formed by a similar mechanism to that of mold with a fine structure made by edging. We can infer that the fine shape of the mold was not directly transferred but the fine uneven structure was formed on the nontransferred area, and thereby, glossiness was kept low. As a result, the fine

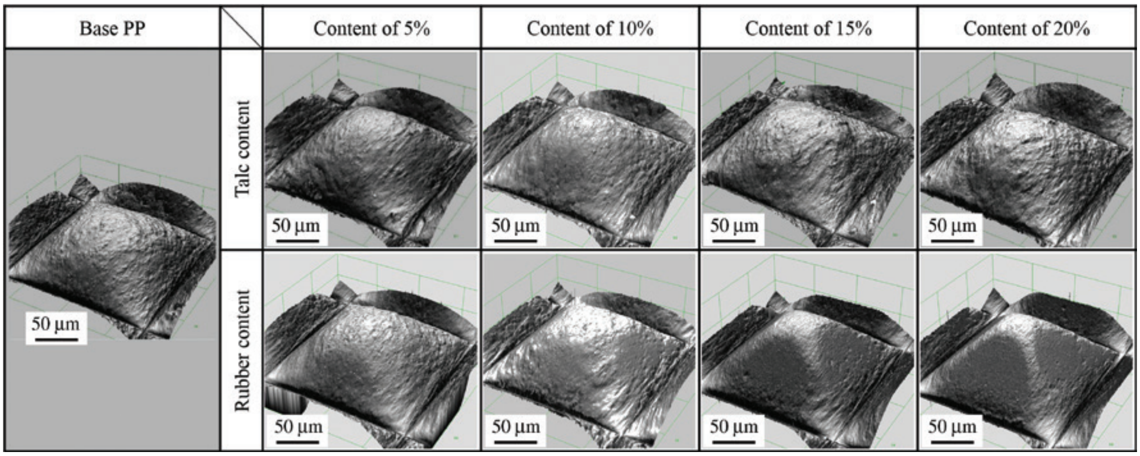

Figure 6: Effect of talc and rubber contents on transcribed surface height shapes.

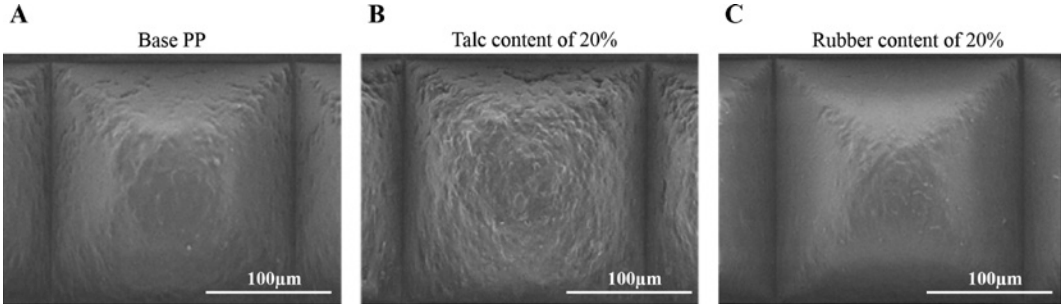

Figure 7: Effect of talc and rubber contents on transcribed shape surface. These pictures were observed by SEM. (A) base PP, (B) PP + talc content 20%, (C) PP + rubber content 20%.

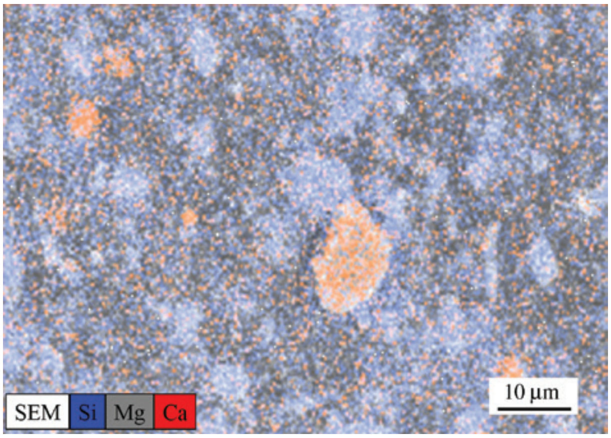

Figure 8: Talc-containing material transcribed shape components distribution. Obtained by EDM.

structure of the mold was not dominant, and thereby, the correlation between the glossiness value of the mold and the glossiness value of the product was lost.

3.7 Influence of the rubber content on transferability

Next, using a material with an increase in the rubber content relative to the base PP, a molding experiment was conducted in a similar manner. The results of the shape analysis obtained using a laser microscope are shown in Figure 6. As can be seen from the figure, in contrast to the talc-containing material, as the rubber content increased, the transferable area increased. Figure 7C shows the comparison of SEM images of the base PP and the molding product sample containing 20% rubber. As can be seen from the figure, even in the nontransferred area, the fine uneven structure, as observed in the talc-containing material, was not formed, and the surface was smooth, similar to that of the base PP. On the contrary, in the transferred area, plural streaks that are a few micrometers deep and looked like creases that are formed in the base PP were observed. However, as the rubber content increased, the smoothness of the surface improved, and at the same time, the generation of the streak pattern decreased. Therefore, we confirmed that the transferability improved in the case when rubber is present. Accordingly, transferability was also improved more than the base PP in the mold with fine structure obtained by edging. In particular, transferability was also improved more in the base PP than in the mold with a fine structure, where the uneven structure was of a few micrometers in size.

3.8 Viscoelasticity measurement results for the talc- and rubber-containing materials

We conducted a viscoelasticity evaluation to measure a change in flow phenomenon of the melting resin due to the talc and rubber contents. The measurement results are shown in Figures 9 and 10. It has been reported that the transfer of resin mainly takes place at the pressure holding phase, and we focused on the viscosity change in the low-frequency region, where the resin flow rate is at a slow status. In the report of Ito and Takai et al., the influence of low strain viscosity during melting of PP on fine transfer was studied, and it has been reported that the transferability of a resin material with high fluidity at low strain improves. From that, similar consideration is possible in this experiment. As shown in Figure 10, by DMA, PP talc has high storage modulus and loss modulus as a general result, which corresponds to the low strain rate range of the shear viscosity data of Figure 9. From the data on shear viscosity, especially at high strain rates, the viscosity is low and PP talc is considered to have high fluidity due to its low viscosity, whereas at the low strain rate at the final stage of transcription, it can be inferred that the

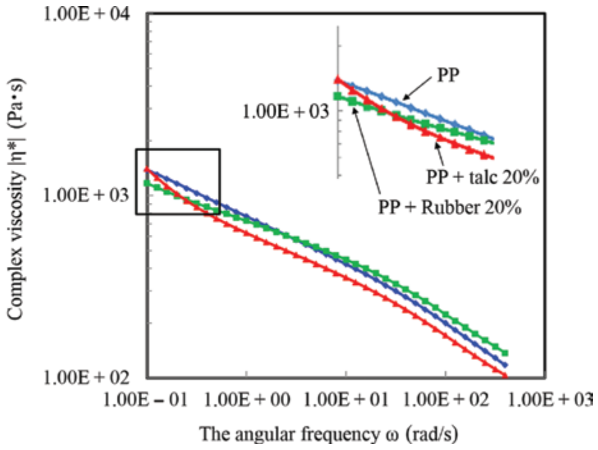

Figure 9: Relation between the complex viscosity and the angular frequency.

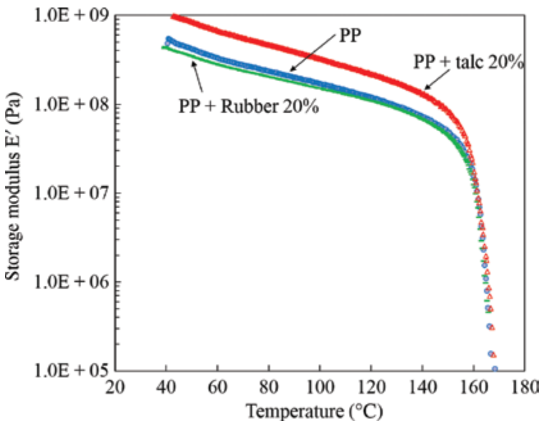

Figure 10: Dynamic mechanical analysis data for PP, talc content 20%, and rubber content 20%.

deformation during release is suppressed or the low strain region has determined the transfer behavior.

On the contrary, in the low-frequency region, viscosity decreased in comparison with that of the base PP as the rubber content increased. This may be because the viscosity of the added rubber component was lower than that of the base PP and viscosity decreased by kneading with the rubber. As for the transferability of rubber, more detailed analysis is required and future additional reports are expected.

4 Conclusions

We qualitatively and quantitatively showed the influence of the talc and rubber content on the transferability and glossiness in a PP resin.

It was confirmed that, if the PP resin contains talc, the correlation between glossiness of the mold and glossiness of the product is lost and glossiness is decreased.

We inferred that the transferability decreased with the addition of talc; and in the nontransferred area, the talc particles scattered near the surface layer formed a few tens of micrometers of fine unevenness as the PP resin contracted, which lowered glossiness.

On the contrary, as the rubber content increased, viscoelasticity decreased and transferability improved. At the same time, generation of the streak pattern in the transferred area, which was generated in the base PP, decreased and glossiness increased. Moreover, the fine uneven structure of the nontransferred area, as observed in the talc-containing material, was not observed in the rubber-containing material.

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