TECHNICAL REVIEW



Industrialization of automotive glazing by polycarbonate and hard-coating

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Abstract

The automobile industry has a strong need for weight savings in order to meet the regulations regarding the issue of the global environment. TEIJIN LIMITED started the development of plastic glazing in 1998, not only the functional materials (polycarbonate resin and hard-coating (HC) liquid) but also the processing technology, and succeeded in the industrialization of the plastic glazing production of limited parts in 2006. Now, our mass-produced glazing products are being supplied to automotive manufacturers. To promote business expansion in plastic glazing, we clarified the deterioration mechanism of plastic glazing used in the outdoors and established the plasma chemical vapor deposition (CVD) coating method. We design and provide "the new driving experience" with our plastic glazing.

Introduction

The ability to meet the new challenge of producing lighter weight automotive bodies, a product of stricter global fuel economy regulations in the automotive industry in response to the recent worsening problem of global climate change, will influence the competitiveness of automotive manufacturers and materials manufacturers in the global market moving forward.

In response to the demand for lighter weight automotive bodies, as metal and glass are being replaced by lightweight plastics, the move in Europe toward the use of polycarbonate (PC), an engineering plastic with superior transparency, impact resistance, and heat resistance, in automotive glazing is picking up steam [1]. However, the relative cost allowance and long-term reliability of PC (durability, weather resistance, and abrasion resistance) compared to glass are bottlenecks to its market penetration.

Teijin started the development of PC automotive glazing in 1998. At that time, plastic glazing technology was still immature, and there were virtually no specialist parts

⊠ Toshiaki Hotaka t.hotaka@teijin.co.jp manufacturers. Therefore, Teijin started development on both its own materials and processing technologies and has been working to promote the penetration of plastic glazing by proposing it to automotive manufacturers. To date, via the key technologies presented below, Teijin has solved issues including the following: (1) Creating large automotive plastic glazing of 1 m^2 or larger; (2) achieving reliability in the adhesion of plastic glazing to metal bodies on the automotive assembly line; and (3) achieving longterm reliability because of sufficient weather resistance.

Outline of the development

In this paper, new materials technology, molding process technology, and surface function addition technology for plastic glazing that make new driving experiences possible through pursuit of emotional design, integration, and eco-friendliness unachievable with metal or glass are presented.

Problem

- 1. Replacing glass with plastics in large automotive glazing of 1 m^2 or larger
- 2. Securing reliable adhesion between plastic glazing and metal bodies on the automotive assembly line
- 3. Securing long-term reliability because of sufficient weather resistance

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Solution

- 1. Development of heat-shielding PC with improved heat resistance during molding
- 2. Development of a PC alloy that can be directly bonded to a metal body with a urethane adhesive
- Development of thermoset silicone hard coat for automotive glazing
- 4. Development of the world's largest 4-axis parallelism control 2-component injection press-molding technology (3D ultralow distortion-molding technology)
- 5. Development and industrialization of world's first 3dimensional dual-side flow coating technology
- 6. Development of longevity prediction technology for automotive plastic glazing

Results and discussion

New materials technology that solved plastic glazing issues

Developments to date have led to the following materials technologies: (1) PC for automotive glazing material: ultraviolet and heat-shielding PC obtained through improved molding heat resistance at the additive level as well as proprietary dispersion technology of special inorganic particulates; (2) PC for automotive glazing of 2-component window frames: PC alloy with good appearance and low linear expansion that can be directly bonded with a urethane adhesive to a metal body on the automotive glazing: thermoset silicone hard coat for automotive glazing: thermoset silicone hard coat with improved weather resistance and abrasion resistance, which are weaknesses of PC.

Solution 1: Development of heat-shielding PC with improved heat resistance during molding

PC with improved molding heat resistance

PC has been used in quarter windows and sunroofs in Europe since ~2000 for reasons including lighter weight automotive bodies that leverage transparency, impact resistance and heat resistance; greater freedom in design; and integration and modularization of peripheral parts. PC with added weather resistance, which is widely used in headlamp lenses, has been used in these applications.

A key problem with using this type of typical weatherresistant PC in large automotive glazing applications such as panoramic roofs or back windows of over 1 m^2 has been the lack of sufficient molding heat resistance to withstand the harsh thermal history in large injection molding equipment. In other words, because the injection volumes of PC glazing material are a magnitude of order greater than those of headlamp lens material, the molding temperatures will be higher, and the molding cycles will be longer. Thus, due to the extremely harsh conditions in cylinders, further improvement in molding heat resistance was needed.

Therefore, to improve heat resistance at the additive level, a PC composition that can withstand large-scale molding was established. In the development of PC for automotive glazing, a new weather-resistant grade that markedly improves molding heat resistance and makes both stable quality and mass production possible even under harsh molding conditions was established without harming the qualities (transparency, heat resistance, impact resistance) particular to PC [2]. In particular, because PC glazing depends on its design surface, minimization of the silver streaking that accompanies PC degradation is desired, so composition design with improved molding heat resistance is implemented on all additives added to PC, including stabilizers, mold-release agents, and absorbents.

Specifically, when conventional headlamp lens-grade material is used in retention molding for 10 min at 350 °C, the generation of an uneven shear flow pattern was observed under polarized light. Using a cross-sectional observation method to analyze this uneven flow led to the generation of subtle flow disturbances caused by bulk PC degradation byproducts. When this uneven flow grows, it leads to an undesirable phenomenon known as "splay or silver streaking during molding". We have defined this uneven flow as the precursor to inferior molding heat resistance.

As described above, in PC glazing, PC experiences extremely harsh conditions inside the cylinder or hot runner of the molding machine; thus, it is essential to improve PC molding heat resistance. Using thermogravimetric analysis to measure the weight reduction at $350 \text{ }^\circ\text{C} \times 30$ min as an index, optimal composition design was performed on all additives added to PC, including stabilizers, mold-release agents and ultraviolet absorbents (UVA). This design resolved the uneven shear flow pattern that occurred during molding at $350 \text{ }^\circ\text{C} \times 10$ min retention, and a new PC glazing material that resolved the subtle flow disturbance from bulk PC degradation byproducts even under morphological observation was established.

PC with heat-shielding function

To create even greater customer value based on the previously described weather-resistant grade, heat-shielding PC with improved moisture and heat resistance was developed by using compounding technology, which enables the dispersal of special inorganic particles absorbing infrared radiation into PC without any damage to transparency. Specifically, when heat absorbents such as lanthanum hexaboride (LaB₆) are dispersed into PC and exposed to heat and humid conditions, the heat absorbents become inactive, and the heat-shielding performance drops significantly. In addressing this problem, it was discovered that the heat absorbent coating on the particle surface, the coating thickness and trace impurities in the PC have an influence on this. Heat and moisture resistance were improved through appropriate use of a silicone coating, a thicker coating thickness and control of the PC trace impurities to <70 ppm [3].

Furthermore, by using dispersion technology with cesium-doped tungsten to maintain the natural transparency of PC, the world's highest level of performance with an increased solar heat gain coefficient was developed [4]. In preparation for future environmental regulations, the Panlite® heat-shielding grade AM-1100ZV series has been released to the market. By increasing the solar heat gain coefficient while minimizing the reduction in visible light transmission, the AM-1100ZV series has created customer value by allowing the transmission of light while blocking heat. Because PC glazing with the AM-1100ZV series allows bright light to enter but can suppress a rise in cabin temperature from sunlight, it contributes to improved fuel efficiency and power usage by increasing air conditioning efficiency and minimizing power consumption. It is already being used in PC glazing for railway cars. The Panlite® heat-shielding grade is being used not only in PC glazing but also in sheet film applications that require heat-shielding performance (Fig. 1).

Solution 2: Development of a PC alloy that can be directly bonded to a metal body with a urethane adhesive

PC for automotive glazing of 2-component window frames

One fundamental problem with using plastics as a glazing material to replace glass is the method of affixing plastic glazing to a metal automotive body frame. Here, a fixation method of bolting would typically not be very appropriate due to the high-stress concentration. Adhesion through direct glazing is a typical method in automotive glass glazing, in which case the issue of stress concentration is unlikely to arise. A direct glazing system is typically a urethane adhesion system, and it is used together with a special primer to increase adhesion between glass and a urethane sealant. However, if a glass-based direct glazing system is used with typical PC, there will be situations when the PC glazing does not achieve firm adhesion with the metal material. Thus, there was a need to improve the chemical resistance of PC to urethane sealants and to further improve the heat and moisture resistance of adhesive



Fig. 1 Panlite[®] heat-shielding grade AM-1100ZV series (T_v : visible light transmittance, T_{TS} : total solar transmittance)

strength. Therefore, a PC alloy for use as a window frame material that could be used as it is in the glass window adhesion system already employed on automotive assembly lines was developed [5].

Specifically, in addition to improving chemical resistance to urethane sealants through compositing of polyethylene terephthalate, heat and moisture resistance were improved by optimizing the stabilizers and glass fiber binder, and a PC alloy was released to the market with a proprietary composition design to strongly bond with materials having a low linear coefficient of expansion (such as metal). This PC alloy is a superior material in maintaining adhesive strength, particularly in hot and moist environments. Because stable adhesive strength is achieved even under harsh usage conditions in automotive applications, this alloy is already being used as an automotive PC glazing window frame material for various models in Europe, such as Daimler, Honda, SEAT and Smart. In addition, it is established as the de facto standard in plastic glazing applications.

Furthermore, this PC alloy has recently been used for the quarter window in the new DS5 from Citroën (France) and in the quarter window of the new CLIO from Renault (France). For the DS5 quarter window, by integrating transparent glazing parts and a pillar cover with a high gloss black surface made of a special grade of PC alloy into one part by 2-component injection molding, a complex design that could not be formed with glass was achieved. At the same time added value was created in the form of superior aerodynamics. Compared to using glass in the quarter window, using plastics not only improves freedom in molding characteristics but also can reduce the window weight by ~20%. (Lighter weight and superior design).

The quarter window used in the DS5 was jointly developed with Freeglass (Germany), one of the world's leading companies in PC glazing. To date, Teijin's PC glazing materials have been used in various car models from Daimler, Honda, Porsche, SEAT and Smart, and Freeglass is a partner in bringing these materials to the global market. By further strengthening ties with Freeglass through a relationship since the late 1990s in the development of new PC glazing technologies and materials and by working with customers from the prototype and development stages on various materials, Teijin not only will achieve lighter weights but also, by achieving performance on par with glass, will create customer value in pursuit of emotional design and eco-friendliness not achievable with glass. Furthermore, it will demonstrate a new driving experience to consumers, who are the customers of our customers, by attaining the joy and exceptional value that makes customers say "Wow!". In this way, Teijin aims to accelerate the expansion of plastic glazing.

PC for automotive bodies

In addition, to fully explore automotive plastic body materials capable of new designs, Panlite® AM-9937F, which is a plastic with high dimensional accuracy, moldability and an appealing appearance that enables not only lighter weight but also designs that cannot be achieved with conventional metal materials, was developed [6]. As momentum towards protecting the global environment grows, including through prevention of global warming and reduction in fossil fuel resource use, in the automotive industry, lighter weight automotive bodies and improved fuel efficiency have become major issues in consideration of the environment. Thus, there is a move toward the use of PC automotive parts including automotive glazing, frame materials and body materials as a replacement for glass and metal to reduce body weight. To date, various types of plastics, primarily PC, have been used as replacement materials for metal in automotive parts such as door handles and wheel caps.

As time goes by and the move toward even lighter weight bodies continues, there is growing customer demand for the use of plastics in large parts such as back doors and fenders. However, with polymer alloy plastics, which are a mixture of polyester resin and PC that has been used as a metal substitute in automobiles, there was a problem with achieving both the dimensional accuracy and moldability required in large parts along with appealing appearance characteristics (smooth surface, decreased color unevenness, etc.). The newly developed Panlite® AM-9937F blends a special fine fiber as a filler into a base of a polymer alloy of PC and a polyester resin to preserve characteristics such as high heat resistance, moldability, design, impact resistance and rust prevention while achieving both high dimensional accuracy and appealing appearance that were not possible with conventional plastics. As a result, automotive parts with designs that had been difficult to obtained using metal parts can now be manufactured. Because of recognition of this excellent characteristic, Panlite® AM-9937F is being used as the material for luggage door garnish in the Lexus HS250h premium sedan from Toyota.

By leveraging these characteristics and using this plastic as the material in places that to date have used steel sheets, such as the outer panel in automobiles, weight reductions of ~20% versus steel sheets can be expected. Moving forward, by leveraging various high-performance characteristics such as impact resistance and dimensional accuracy required in a replacement material for metal, as well as moldability only achievable in plastics, this new material will not only meet the demand for light weight and higher functionality but also help broaden options in automobile design. Teijin will strive to expand existing foreign and domestic markets while taking on the challenge of developing a new market segment.

Solution 3: Development of thermoset silicone hard coat for automotive glazing

In realizing PC automotive glazing, there is another problem with improving weather resistance and scratch resistance. The technology that forms an acrylic or silicone hard coat on a PC surface has been developed as a method for improving scratch resistance [7], and the following two methods are the current mainstream approaches.

- (1) Formation of a silicone hard layer through application and curing of silicone hard coat
- (2) Formation of a coating layer comparable to glass by plasma chemical vapor deposition (CVD)

In the formation of a silicone layer, after applying hydrolyzed and condensed alkoxysilane sol (hard coat), the sol-gel thermosetting technique at ~100–140 °C is used. Furthermore, to improve weather resistance, certain methods were employed, such as adding an UVA to the hard coat layer and to the primer layer between the hard coat and PC. In addition, to suppress deterioration from water and heat, durability of the hard coat against, for example, water and heat is required. To improve these qualities, a technique was developed by using a self-cross-linking acrylic resin that utilizes an acrylic resin having an alkoxysilane group and by using a thermoset acrylic resin reacted with a cross-linking agent by introducing functional groups to the acrylic resin.

Teijin's silicone hard coat

In addition to hardness and weather resistance, because a hard coating (HC) for automotive glazing requires Fig. 2 Layer structure of Teijin's silicone hard coat DMT series



superior heat resistance, water resistance and toughness against elastic stress, an HC that could persist throughout the service life of automobiles has not been established until now. To be able to blend the maximum amount of a triazine-based UVA with superior durability and heat resistance into the HC, the copolymerization composition ratio of the multicomponent copolymer thermoset acrylic resin was uniquely adjusted to optimize their compatibility. By adding inorganic metal particles into a thermoset silicone HC on top of the primer coat, the DMT series that satisfies the above characteristics was developed [8]. This lineup possesses particularly strong heat resistance compared to that of conventional HC and can even withstand use at 130 °C (Fig. 2).

The DMT series consists of two layers of HC. The top layer (hard coat layer) is a hard coat formed via the sol-gel method using colloidal silica and alkyl alkoxysilane that add abrasion resistance. When forming this type of coat, because volumetric shrinkage occurs due to the condensation reaction during thermosetting, stress forms between the substrate and primer layer, so cracks will often form in the coating layer. Cracking can be suppressed by increasing the ratio of alkyl components, but this procedure reduces abrasion resistance. Therefore, the silanol group condensation state was quantified by using ²⁹Si NMR spectroscopy at the sol state, and by applying this technique to industrially manage the scope of usage of a coating that offers both abrasion resistance and crack suppression, the product was successfully commercialized. In addition, by using this technology, high heat resistance was achieved by suppressing the level of unreacted silanol residues in the coating after the sol-gel reaction was completed [9].

Furthermore, for industrialization as a coating liquid, stability over a long time period is required. By blending the alcohol amine choline, used as a curing catalyst, in acetate form into the coating liquid, a sol of pH 5 to 6 was converted into a stable buffer solution. Thus, a coating liquid that could withstand continuous use in a dip tank even over a 6-month period was developed.

The primer layer is formed between the PC and hard coat layer (top layer), and in addition to the function of making these two layers adhere together, weather resistance is added by blending UVA. From early on, Teijin has focused on the superior durability of triazine-based UVA as an organic UVA; however, triazine-based UVA has poor compatibility with the acrylic resin used as the primer layer of silicone HC. When a durability test was conducted on the coating film, there was a problem with phase separation, which resulted in whitening of the entire film. To address this problem, various acrylic monomers having different side chains were investigated, and stable dispersion of triazine-based UVA in an acrylic resin-based primer layer was achieved by optimizing the solubility parameter of the acrylic resin. This optimization approximately doubled the weather resistance in the resulting HC composition compared to a composition that conventional benzophenone-based uses UVA or benzotriazole-based UVA.

In addition, in automotive plastic glazing applications, weather resistance as well as high heat resistance is required. For the acrylic resin used for the primer layer, it was difficult to attain the required heat resistance with the thermoplastic resin state. Therefore, by developing a thermoset acrylic resin coating blended with an isocyanate-based cross-linking agent with blocked functional groups to maintain the long-term stability of the coating, a primer coating that possesses high heat resistance and can withstand several years of use as a coating was developed.

In addition, to further increase the weather resistance of the DMT200 series, UVA is used in the top layer to a degree that does not affect the abrasion resistance.

By using this kind of proprietary technology, characteristics that can be useful in automotive glazing applications are added. Not only is the DMT series used in HC applications for plastic glazing, but it is also being used in sheet film applications that require weather resistance and abrasion resistance (Table 1).

Teijin's silicone dry HC technology

Teijin is working to make a commercially viable dry HC technology for automotive glazing by plasma CVD [10–13]. CVD is a technology that initiates a chemical reaction between a decomposed raw material and a carrier gas in the gas phase under plasma conditions and deposits the reaction product(s) on the substrate surface. Because it is possible to form a coating at low temperatures, this technology can also be applied to plastic substrates. The DMT200 α series, developed by Teijin using this technology for automotive glazing applications, is garnering attention from automotive manufacturers, as it possesses surface hardness on par with glass that can comply with the abrasion resistance required by new safety standards (UNECE regulation 43 on glazing materials) applied to new vehicles from July 2017.

In addition, the weather resistance is approximately doubled by creating a dense CVD layer with increased gas barrier performance, which prevents the deterioration of the hard coat layer and PC substrate by oxygen and water (Fig. 3). These HC characteristics were achieved through multistage coating steps that leverage qualities of a parallel plate plasma system that can control coating layer forming conditions uniformly and with high accuracy, as well as through an advanced gradient structure that establishes a good balance among abrasion resistance, durability, and heat resistance.

In December 2016, a pilot plant that enables CVD processing on 3-dimensional substrates of actual product size $(1300 \text{ mm} \times 800 \text{ mm})$ was installed at the Matsuyama R&D Center in Ehime Prefecture (Fig. 4). Moving forward, Teijin aims to establish mass production technology for CVD plastic glazing to bring this technology to the market.

Solution 6: Development of longevity prediction technology for automotive plastic glazing

"Exactly how many years will plastic glazing last?"

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		Condition/method	DMT200a (CVD)	DMT200 (WET) European Union Standard (ECE R43, rigid plastic)	American Standard (FMVSS, ANSI AS2)
Optical property -Transparent	Light Transmission	Light source; A	% >88	>88	>70	>70
-PC substrate (5mmt)	Haze	Light source; A	% <0.6	<0.6	I	No requirement (<1% only for AS4a)
Scratch Resistance	Taber abrasion Δ Haze	ASTM D1044; For Exterior CS-10F, 1000cycle	% 0.5~1.5	5~7	<2	< 2 (both sides)
		ASTM D1044; For Interior CS-10F, 100 cycle	% 0.4	0.4	<4	I
	Pencil hardness	JIS K5600	F~H	F∼H	I	1
Durability -Appearance	Boiling water resistance	Dipping 3 h	Excellent	Excellent	I	1
-Adhesion	Heat resistance	110 °C, 1000 h	Excellent	Excellent	Ι	I
	Heat cycle	-30 °C, 1 h and 100 °C, 1 h interval 15 min each	Excellent	Excellent	I	I
	Weathering (Super Xenon Weather Meter)	JIS K7350-2, 180 W/m ² (Exposure >10 years equivalent)	6500 MJ/m ²	3800 MJ/m ²	500 MJ/m ²	306 MJ/m ²



Fig. 3 Optical property changes of DMT200 α during a weathering test. (Xenon weather meter, irradiance of 180 W/m²@300-400 nm, black panel temperature of 63 °C, light for 102 min, Light + spray for 18 min)



Fig. 4 Pilot plant for plasma chemical vapor deposition (CVD)

Presently, the use of plastics in automotive glazing is still a technology in its infancy, so compared to the lifespans of metal and glass materials that have thousands of years of history, the ability to predict the lifespan of plastics will determine whether automotive plastic glazing will widely penetrate the market. Therefore, lifespan prediction technology of plastic glazing was investigated by comparing accumulated outdoor exposed parts and parts taken from the actual market (periodically collected from the market) with samples subjected to an accelerated weathering testing. The results showed that by grinding away the silicone hard coat layer, the ratio of UVA remaining in the primer layer could be directly quantified via infrared (IR) spectroscopy and Raman spectroscopy. In this way, the degradation mechanism during the weathering test could be understood, and technology for predicting the longevity of plastic glazing, which was previously difficult, has been established.

HC loses its viability because of cracking and delamination that occurs due to ultraviolet, oxygen and moisture exposure in actual use environments as well as in accelerated weathering tests. Of these factors, the first focus is on t_{fail} , the time when the HC delaminates, and D_{fail} , the amount of ultraviolet radiation.

$$t_{\text{fail}} = \frac{1}{k} \log_{10} \left[\frac{10kD_{\text{fail}} + T_0 - 1}{T_0} \right]$$
(1)

Using formula (1) [14], the UVA decrease coefficient k is obtained. (T_0 is initial light transmittance.)

Using the UVA decrease coefficient in the primer layer of DMT250 aged in a xenon weather meter until delamination and of DMT250 after three years of outdoor exposure testing in Arizona, a predicted lifespan of DMT250 in Arizona of ~5 years was derived. (However, UV radiation equivalent to 1 year in Arizona = $3 \text{ MJ/m}^2@340 \text{ nm}$). The predicted lifespan of DMT200, with its improved weather resistance compared to that of DMT250, was approximately 10 years using the same evaluation method. The validity of these predicted values is currently being examined.

In addition, it was discovered that UVA reduction behavior during this evaluation followed two patterns. These patterns were differentiated as initial degradation and aged degradation. Research is being conducted on whether maintenance of UVA performance during initial degradation, when UVA reduction is particularly rapid, leads to increased weather resistance. In addition, a profile analysis of the UVA amount in the depth direction using Raman spectroscopy was conducted by crosssection cutting of the HC film. The results showed that although the UVA residue was uniform in the depth direction in the initial state, after weather testing, the UVA residue was 0 near the primer surface (near the top-layer interface), and some of the UVA even remained near the PC substrate. From this finding, it was discovered that the amount of UVA residue in the primer layer was dependent on coating thickness. On the basis of this knowledge, Teijin aims to further improve HC durability in an effort to achieve product value and reliability no less than those of glass.

New processing technology that solves plastic glazing problems

Regarding the processing technologies that have been developed to date, a framework has been established for the production of plastic glazing products with the following characteristics.

 Development and industrialization of the world's largest two-component rotary injection press-molding technology with 4-axis parallelism control capable of Fig. 5 Outline of world's largest injection press machine



3-dimensional ultralow distortion molding of 1 m^2 or larger that had been difficult to achieve with conventional technology

b. Development and industrialization of 3-dimensional dual-side coating technology that had been difficult to achieve with conventional technology.

Solution 4: Development of the world's largest 4axis parallelism control 2-component injection press-molding technology (3D ultralow distortionmolding technology)

Despite the progress in the replacement of metal and glass with lightweight plastics to create lighter weight vehicles, PC panoramic roofs commercialized with conventional injection-molding technology have a limit in area of $<1 \text{ m}^2$. For this reason, the development of a processing technique that can handle even larger glazing parts is desired. The higher cost of PC automotive glazing than glass glazing is a bottleneck to market penetration. Moving forward, truly spreading lighter weight vehicles via PC products will absolutely require the establishment of commercialization technology (glass replacement) and cost-cutting technology (glass and metal replacement, rationalization of assembly process) from window and body integration.

In 2003, Teijin installed an injection press-molding machine with 4-axis parallelism control with the world's

strongest pressing force of 3400 tons (33,540 kN) capable of large and fast plastic molding at a high speed of 35 mm per second (Fig. 5). This machine has significantly reduced problems with distortion and warping that occur in large-scale molding of parts such as panoramic roofs.

In addition, to meet the demand for larger sizes and high added value in automotive applications, major modifications were made to the molding machine in 2006. Teijin developed the world's largest 4-axis parallelism control ultralarge 2-component rotating injection press-molding machine and achieved integrated molding of a 1.7 m^2 scale 2-component panoramic roof + body panel.

Then, in 2013, Teijin successfully industrialized technology with superior visibility by reducing transparent distortion without optical distortion (the view seen through the car window is not distorted) that enables 3-dimensional thick ultralow distortion molding of 1 m^2 or larger that had been difficult to achieve with conventional technology.

Development of injection press-molding technology

Scale-up technology of glass replacement (injection press molding technology) Having superior impact resistance along with the ability to withstand breaking even if stepped on by an elephant, PC is an ideal plastic for replacing glass, but it requires surface hard coat treatment for abrasion and weather resistance. However, as treatment of PC with surface HC causes a drastic reduction in impact resistance, producing windows that will not break requires highmolecular-weight PC from the perspective of solvent resistance.

However, because the moldability of PC decreases as the molecular weight increases, there had been a limit on obtaining sizes of just below 1 m^2 with injection molding. In addition, with injection molding, residual stress is formed easily due to PC being forcefully injected into closed molds with injection pressure. Because cracking occurs easily when surface hard coat treatment is applied, annealing is needed. In other words, for PC automotive glazing, the biggest issues were how to mold large-size and large surface area parts with highmolecular-weight PC and how to uniformly mold parts without distortion (residual stress).

To solve the problems described above, since 1999, Teijin has been developing key technology toward the use of plastic in automotive glazing with a focus on injection press molding [15]. With automotive glazing, because the entire product is the design surface, the gate must be positioned on the product edge. However, with injection press molding, because plastic is filled into the mold in a slightly (several mm) opened state, the unbalanced load causes mold parallelism to become disrupted, and due to the large size of the mold (tens of tons), the mold will slip under its own weight. Thus, injection press molding did not become a production technology due to product optical quality as well as mold galling and damage.

To overcome these issues associated with scaling up, in 2003, Teijin and Meiki Co. jointly developed one of the world's largest 4-parallelism horizontal control injection press-molding machines (MDIP2100-DM) at 3400 tons, which was installed in Teijin's Plastics Technical Center (Chiba City), where it has been used in various molding tests and R&D. As a result of pursuing the development of commercialization technology at an actual product level, a 1.3 m² PC panoramic roof capable of reducing weight by ~30% versus glass was successfully developed and established as a commercialized technology.

Parallelism of the molding machine movable platen is preserved by using Meiki's proprietary 4-axis parallelism control technology; thus, even if an unbalanced load occurs, damage to the mold is suppressed. Moreover, not only can product thickness be made uniform, but uniform molding can also be achieved even with high-molecular-weight PC while using approximately one-third to one-fifth of the internal mold pressure versus the typical injection molding technique. Additionally, product sizes that had been difficult to obtain previously were made possible (world's largest size, as of 2003, of 1.3 m^2 was achieved, and 2.2 m^2 was achieved in 2007). In addition, it became possible to obtain optically uniform products without residual stress. This reduction in residual stress also enabled rationalization by an annealing-less process.

Ultralarge injection press-molding technology and reduction in residual stress

Teijin and Meiki have jointly been developing technology to mold automotive panoramic roofs of $\sim 2.0 \text{ m}^2$ in surface area using PC. As a result, MDIP2100-DM, an exclusive injection press-molding machine with 4-axis parallelism control injection press functionality as well as the highest level of mold clamping force in Japan of 3400 tons (33,540 kN), was jointly developed. This machine was also installed in the Plastics Technical Center.

Characteristics of the injection press-molding technique

The typical and widely used injection molding technique, in which a mold is filled with molten resin at high pressure, is characterized by high productivity and the ability to obtain complex shapes such as ribs, bosses, and 3-dimensional curved surfaces.

On the other hand, because the mold is clamped at high force and filled with molten resin at high pressure, the molecule orientation tends to be in the direction of flow. Because molded products will exhibit different shrinkage behavior depending on whether this molecule orientation is aligned in the direction of resin flow or in a perpendicular direction, this phenomenon becomes a cause of warping and stresses in the molded parts. Additionally, because the mold is filled at high pressure, considerable distortion occurs in the interior of the molded parts, which leads to reduced strength.

In contrast, injection press molding was developed as a molding technique that maintains the high productivity of injection molding while incorporating the benefits of compression molding, such as orientation according to high pressure filling and no residual distortion.

Injection press molding is different from injection molding, as the former is a technique in which resin is injected into an open mold that is subsequently closed with clamping force, causing the resin to travel throughout the mold interior. Because this technique does not require resin to be pushed into narrow cavities, the filling pressure can be significantly reduced. Furthermore, because uniform pressure is applied to the entire cavity within the mold from the mold clamping force, molded parts with little distortion can be obtained.

In injection press molding, by simultaneously injecting and pressing according to injection time or fill volume, parts of larger size and surface area than those obtained with Fig. 6 Outline of the 4-axis parallelism control system



conventional injection molding can be molded. In typical injection molding, resin flow within the cavity slows as it travels further from the gate where resin is injected. In contrast, in injection press molding, resin flow generated by the action of mold closing is added; thus, the resin flow does not slow down, and the speed of the flow leading edge can be made constant. As a result, the resin flow length within the cavity can be made significantly longer than that in typical injection molding, enabling the formation of a large projection area and thin-walled molded parts using a low mold clamping force.

In addition, in conventional injection molding, to compensate for the shrinkage when resin solidifies, a procedure known as pressure holding, in which pressure is applied to the resin at the gate to maintain high pressure within the mold, is required. In contrast, in injection press molding, because the same effect as that in pressure holding is achieved by compressing molten resin inside the cavity with mold clamping force, pressure holding is nearly unnecessary. As such, because of reduction in residual distortion at the gate and the absence of the pressure holding procedure, the molding cycle time can be shortened, thereby increasing productivity.

4-Axis parallelism control mechanism

A compound clamping configuration featuring two highspeed transport side cylinders that operate the movable platen and four main cylinders that generate clamping force and pressing force was adopted. This configuration enables a large machine to occupy less space.

The mold clamping force and pressing force are 33,540 kN, while the moving platen and stationary platen that secure the mold are $3000 \text{ mm} \times 3000 \text{ mm}$, all of which are at the highest levels in Japan.

To enable molding of large but thin-walled parts, the maximum press speed was set to 35 mm/s, which is the

fastest speed for a large machine with 33,540 kN of mold clamping force.

When clamping pressure is applied via the four main cylinders, first, the moving platen is moved with the two side cylinder, and then after the main cylinder piston rod is locked with a half nut, the main cylinders are then moved. To prevent malfunction by backlash (play in the screws) of the locking mechanism, a backlash removal device is equipped.

The main feature of the mold clamping mechanism is the presence of a linear scale (position detector) and servo valve for each of the four main cylinders to enable 4-axis parallelism control during the pressing procedure (Fig. 6). This feature makes the four axes controllable in parallel to an accuracy of several microns. This 4-axis parallelism press control enables accurate molding while preserving parallelism even for molded parts with an unbalanced load.

4-Axis parallelism press control features

When attempting to mold a transparent part such as an automotive panoramic roof or back door window, because the gate often cannot be situated near the center of the parts, a side gate from which resin is injected into the side of the parts is commonly used.

In this type of situation, a large unbalanced load will occur on the mold and moving platen, causing them to slant. Mold deformation and guide slanting cause galling, and wall thickness can easily become uneven. In the worst-case scenario, there is even the issue of mold damage.

To address this problem, as described previously, the new molding machine MDIP2100-DM preserves moving platen parallelism through the control of four axes, so even if this type of unbalanced load occurs, there is no fear of mold damage. In addition, product thickness can be made uniform. **Fig. 7** Comparison of the residual stress on molded parts (left: standard injection, right; injection press)



Fig. 8 Flow length comparison (left: standard injection, right; injection press)



Comparison examples of injection press molding and injection molding

Residual stress

To compare the residual stress, a mold for a half-scale back door window of $520 \text{ mm} \times 380 \text{ mm} \times 5 \text{ mm}$ thickness was used to produce model parts by typical injection molding and injection press molding.

In typical injection molding (1300 tons clamping force), high residual stress was observed around the gate, while in injection press molding (600 tons pressing force), extremely low levels of residual stress could be confirmed (Fig. 7). If high residual stress remains in the molded parts, when injection molding is applied to plastic glazing, such as the panoramic roof, strength near the gate, the residual stress is decreased. However, there is no such concern with injection press molding.

Flow characteristics

To compare flow characteristics, the mold for a 1/1 scale panoramic roof of $1300 \text{ mm} \times 1000 \text{ mm} \times 3 \text{ mm}$ thickness was

used together with typical injection molding and injection press molding. In typical injection molding (3400 tons mold clamping force), the flow length was ~230 mm (Fig. 8).

In general, when the flow length is insufficient, it is improved by raising the resin temperature and/or mold temperature to increase the resin fluidity. However, this procedure may lead to an increase in cycle time, and in the worst case, it may cause degradation of resin. Even if increase and/or degradation do not occur, it could be the cause of strength reduction, poor appearance, and deformation.

There is a countermeasure: replacing the resin with a high-fluidity resin, but even with this approach, the flow length will only increase by approximately two times. In contrast, with injection press molding, the mold can be completely filled using common grade resin without changing the resin temperature or mold temperature.

From a materials perspective, to improve fluidity, other characteristics generally need to be sacrificed. However, by using injection press molding, fluidity can be secured while maximizing the potential possessed by the material. Thus, this technique is suited for use in applications, particularly in plastic glazing applications. Moreover, as described previously, the MDIP2100-DM can reduce wall thickness deviation through 4-axis parallelism control, enabling the production of low-distortion and high-strength products.

Verification of light-weighting

Using a 1300 mm × 1000 mm 1:1 scale panoramic roof simulation mold, moldings of various thicknesses were made with Panlite[®] using injection press molding, and the light-weighting effect was evaluated. Compared to the same shape made with glass, which weighs 11 kg, a 5 mm panoramic roof that fulfills practical strength conditions such as surface rigidity when actually attached to the vehicle body weighs 7 kg. Thus, an ~30% weight savings for a size of 1.3 m^2 was verified. Light-weighting the panoramic roof enables the vehicle center of gravity to be lowered. Since driving stability will increase, this effect also contributes to improved fuel efficiency.

HC

PC has superior impact resistance, making it difficult to break. Compared to other transparent plastics, it has higher heat resistance but lower surface hardness, so HC is necessary to make it practical for plastic glazing of panoramic roofs and other parts. To improve the abrasion resistance and weather resistance of Panlite® (PC), Teijin developed the DMT series of proprietary silicone hard coats, which are already available on the market. However, because there is the risk of cracking developing during coating if the residual distortion is large, as with typical injection molding, annealing (process for removing frozen strains through heating) prior to coating is necessary. However, because frozen strains can be significantly reduced with injection press molding, annealing can also be reduced. In this respect, injection press molding is regarded as an indispensable technology to enable plastic glazing for parts such as panoramic roofs.

(3) 2-component molding technology development

In Europe, commercialization of PC automotive glazing is advancing, and the trend is for this process to increase further moving forward. While this type of glazing offers the benefits of lighter weight and greater design freedom than glass, the high cost compared to glass is presently an issue in market penetration. Moreover, in Europe, some vehicle models are produced with a processing technique that integrates the transparent window and black frame into 2-component molding where Teijin's PC (Panlite®) is being used. However, in terms of glass automotive glazing, there are limits to the cost reduction from simply replacing the black coating, so it was essential to develop streamlining technology of the assembly process through modularization via integrated molding.

To address this issue, in 2006, Teijin and Meiki Co. jointly developed a 4-axis parallelism control ultralarge 2-component rotating injection press molding machine (MDIP2100-HR2) that supports modularization [16]. In addition to equipping a second injection device and developing a mold rotation mechanism in order to add 2-component molding to the injection press molding machine already installed, molding of integrated windows and bodies without lowering window optical quality, which is one of the biggest insistence for material manufacturers, became possible by using mold technology independently developed by Teijin. Thus, key technologies were established in modularization, and the world's first successful ultralarge PC panoramic roof of just under 1.7 m^2 was obtained. Additionally, this integration of the body panel enables a reduction in the assembly cost compared to that associated with separate molding and assembly.

Features of the MDIP2100-HR2 2-component injection press-molding machine

Two-component injection press molding is a technique in which two types of resins are injected into a single molding machine to obtain integrated molded parts. Conventionally, the transparent window would be molded first, and after removing the molded parts from the mold, it would then be set in another mold to form the window frame. In contrast, because these steps of removing the molded parts and resetting are eliminated in the MDIP2100-HR2, manufacturing costs can be reduced.

To enable the injection of two types of resins into a single molding machine, in the MDIP2100-HR2, an intermediate platen was installed between the fixed platen and movable platen of the existing MDIP2100-DM. Furthermore, on the side of this machine, a second injection device was installed. The intermediate platen to which the mold is attached has a sliding mechanism for injection press molding, as well as a 180-degree rotating mechanism. By aligning the core together with the other cavity engraved in the shape of a window frame, closing the mold, bringing the injection nozzle into contact with the window frame, and then injecting it with black automotive body-grade Panlite[®], the transparent section is integrated with the panoramic roof.

At the same time, behind this, resin for the next transparent section is injected. After the molding is completed, the mold is opened, and the completed 2-component molded parts are removed. The molded transparent section is left in the core, and the cavity is rotated. The transparent section is then aligned again with the window frame cavity and core, and the window frame portion is molded.

Although there are examples of 2-component molding panoramic roofs made by injection molding if the surface area



Fig. 9 Tommykaira ZZ with Teijin's PC pillar-less front window

is small (<1 m²), large sizes with high surface area have been difficult to achieve. Now, through combination with an injection press molding technique equipped with 4-axis parallelism press control, 2-component molding of ultralarge panoramic roofs of 1.7 m^2 was enabled.

Using the newly developed MDIP2100-HR2 not only reduces the number of parts and processes as well as costs but also enables commercialization of plastic glazing, including shapes such as panoramic roofs that had been difficult to obtain with glass, integrated molding of window frames with peripheral part fittings incorporated, and modularization of panoramic roofs with peripheral parts such as ceiling lights incorporated.

Solution 5: Development of world's first 3-dimensional dual-side coating technology

Teijin developed and installed a 3-dimensional coating machine at Matsuyama R&D Center in Ehime Prefecture in 2007. In addition to using it in product development with customers and in vehicle prototype production, this machine has also been used in developing HCs. In 2013, not only did Teijin add weather resistance and abrasion resistance, but it also successfully industrialized 3-dimensional dual-side coating technology for parts larger than 1 m², which had been difficult to achieve with conventional technology. The newly established technology adds weather and abrasion resistance to PC and provides cabin partition windows with superior through-window visibility and beautiful appearance by using Teijin's proprietary 3-dimensional dual-side flow coating technology

Product development

To go beyond the role of a materials manufacturer and directly offer customers plastic glazing products, in 2006, Teijin began using the materials and processing technologies developed to date to industrialize limited edition models, launching production of the JR Tokai N700 bullet train window and the Toyota Lexus LFA window (in cooperation with Toyota Industries) [17]. In particular, with the bullet train, the conversion of all windows in regular railway cars to plastic resulted in reduced weight and produced excellent heat shielding, which contributed to reduced railcar power consumption. In addition, in 2013, it began industrializing massproduced vehicles, launching production of the partition window in New York City taxis from Nissan Motor. Teijin was the world's first materials manufacturer capable of developing materials from a parts perspective to successfully establish a business for plastic glazing parts.

Furthermore, Teijin announced that it has developed the world's first PC pillar-less automotive front window for use in the Tommykaira ZZ (Fig. 9), a sporty electric vehicle (EV) produced by GLM Co., Ltd., an EV manufacturer launched by Kyoto University. The newly developed PC pillar-less front window has high abrasion resistance and excellent weather resistance and meets new vehicle safety standards applied from July 2017. A weight savings of 36% compared to the combination of a conventional glass window and an A-pillar was realized by making it a pillar-less front window. In addition to contributing to improvements in the running performance of the vehicle, the transparent PC glazing-integrated front window and pillar provide unobstructed sight lines for safer driving and contribute to more enjoyable sightseeing (Fig. 10).

Going beyond the framework of a materials manufacturer, Teijin has become Asia's number one "Plastic Glazing Solution Center" for product development with customers in materials technology (PC and HC), processing technology (ultralarge 4-axis parallelism control 2component injection press molding technology and coating technology), and product size prototyping. In doing so, it has built a thorough support system capable of innovating streamlining of product development processes of customers. In addition, Teijin is working to commercialize lowcost processes and ultrahard surface treatment technology as next-generation plastic glazing technology.

Future plan

Teijin's Mobility Division, the Glazing Department, which aims to quickly create a glazing market, is centralizing strategic planning and marketing functions for automobiles and railcars; materials technology and processing technology development of the Plastics Technical Center (Chiba City, Chiba Prefecture); and surface treatment technology, next-generation materials technology, and technology development for commercializing automotive PC glazing from the R&D Center. By offering a space where joint development with customers can take place from the prototyping and development stages for automobile and railcar parts at actual sizes, Teijin increases its speedy response to customers and the



Fig. 10 Conventional glass front window with an A-pillar (left) and a newly developed pillar-less front window (right)

market and, in this way, contributes to rapid expansion of the market through environmentally friendly technologies such as vehicle body light-weighting. Through its efforts in the mid- and downstream processing business of automobile parts materials, Teijin aims to gain the trust of customers and the market and to increase the added value of materials.

With a focus on high added value in the plastic business, Teijin is leveraging its existing large plastic glazing molding technology as well as HC technology and facilities, including the recently developed plasma CVD technique, to create high added value. In addition, through collaboration with US-based Continental Structural Plastics, one of the world's leading compound material molding manufacturers that became a wholly owned subsidiary in January 2017, Teijin aims to be a total solution provider for automobile manufacturers.

Compliance with ethical standards

Conflict of interest The author declares he has have no conflict of interest.

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