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High-Temperature Piezoelectric Film Ultrasonic Transducers by a Sol-Gel Spray Technique and Their Application to Process Monitoring of Polymer Injection Molding

Makiko Kobayashi, Member, IEEE, Yuu Ono, Member, IEEE, Cheng-Kuei Jen, Senior Member, IEEE, and Chin-Chi Cheng

Abstract—Thick-film (90 µm) piezoelectric ceramic high-temperature ultrasonic transducers (HTUTs) have been successfully deposited on metallic substrates by a sol-gel spray technique. The gel is composed of fine powders of bismuth titanate dispersed in a lead-zirconate-titanate solution. The films with desired thickness have been obtained through multilayer coating approach. Piezoelectricity is achieved using the corona discharge poling method. The center frequencies of ultrasonic signals generated by these HTUTs are around 10 MHz and their signal-to-noise ratio (SNR) is more than 30 dB in pulse-echo mode at 500 °C. The main advantages of these new HTUTs are that they 1) are applicable at temperatures higher than 500 °C, 2) are miniature, 3) can be coated on flat and curved surfaces, 4) do not need ultrasonic couplant, 5) can be operated at low and medium megahertz frequency range with sufficient frequency bandwidth, and 6) have sufficient piezoelectric strength and SNR. The ability of the HTUTs to monitor the polymer injection molding process in real-time at the mold insert of the machine is demonstrated.

Index Terms—High-temperature ultrasonic transducer (HTUT), polymer injection molding, process monitoring, sol-gel spray technique.

I. INTRODUCTION

POLYMER injection molding is the process of forcing melted polymer into a mold cavity having an unique shape for a designed production parts. Injection molding is often used in mass production and prototyping for various kinds of polymeric products. Recently, miniaturization is the demanding trend in the field of mass production of low-cost micro- and

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nanosystems. For miniaturized devices, micromolding of polymeric materials becomes one of the alternatives to replace the present expensive serial fabrication methods associated with silicon substrates. The micromolding process has the ability to mold microchannels with an extreme precision that is a key to fabricate disposable, miniature and diagnostic lab-on-a-chip devices. However, polymers will need to be selected and modified in order to meet the requirements of narrow process windows involved with their flow, solidification, and microstructure development for the recent advanced parts. Because of this complexity, real-time process monitoring is requested to improve the quality of the molded part and optimize the process.

Ultrasonic method may be one of the candidates because of its ability to probe the properties of polymers within the barrel and mold during molding processes [1]–[4]. It is our intention to monitor the entire molding process from feed hopper to the part exit. Therefore, the preferred requirements of ultrasonic sensors for real-time monitoring of the injection molding are that ultrasonic transducers (UTs) 1) are applicable at temperatures higher than 200 °C, 2) are miniature, 3) can be coated on curved barrel surfaces and flat mold inserts, 4) do not need ultrasonic couplant, 5) can be operated at low and medium megahertz frequency range with sufficient frequency bandwidth, and 6) have sufficient piezoelectric strength and signal-to-noise ratio (SNR).

Thick (> 40 μ m) piezoelectric ceramic films can be made by the technologies of jet printing [5], screen printing [6], dipping [7], tape casting [8], etc. However, they may not meet all the above mentioned requirements. Here, an attractive sol-gel spray technique is used, which was firstly developed at Queen's University [9]. In this technique, the piezoelectric particles are dispersed in the sol-gel solution for producing thick piezoelectric film [10]–[12]. The spray can be carried out by an air gun at room temperature and it is simple and inexpensive. In our previous works [10]-[12], it was demonstrated that piezoelectric powders such as lead-zirconate-titanate (PZT) ceramics or lithium tantalate crystals could be dispersed into PZT or alumina solutions to achieve the piezoelectricity. In this paper, we focus on the bismuth titanate (BIT) powder dispersed into PZT solutions because of the high Curie temperature, 675 °C, and reasonable piezoelectric strength of BIT. Convenient electric poling and top electrode deposition methods will be employed. Also, the real-time monitoring of the polymer injection molding process using these newly developed sol-gel high-temperature (HT) UTs will be investigated.



Fig. 1. Flowchart showing the fabrication steps of high-temperature ceramic film ultrasonic transducers by a sol-gel spray technique.

II. FABRICATION

A flowchart of the fabrication procedure of a high-temperature ultrasonic transducer (HTUT) by a sol-gel spray technique is shown in Fig. 1. The piezoelectric BIT powder was purchased with an average diameter of 50 μ m and dispersed into PZT solution by ball milling method to achieve the gel (step S1). The final diameter of the BIT powder was estimated to be less than 1 μ m. An air gun was then used to spray the sol-gel BIT/PZT composite directly onto metallic substrates [9]-[12] having a flat or curved surface as shown in Fig. 2(a) (step S2). With this technique, the BIT/PZT films can easily be produced at desired locations through a shadow mask made of even paperboard. After spray coating, thermal treatments such as drying, firing and annealing were carried out at temperatures of 90 °C, 430 °C, and 650 °C, respectively, with the optimal time duration (step S3). Multiple layers were made in order to reach the desired thickness by repeating the steps S2 and S3 (step S4).

The films were then electrically poled using a corona discharging technique [13], [14] as shown in Fig. 2(b) (step S5). The corona poling method was chosen because it can pole the thick piezoelectric ceramic film of a large area and on curved surfaces. A high positive voltage supplied from a high-voltage power supply was applied to a needle which was located several centimeters above the film coated on the metal substrate serving as the ground electrode. The poling time was several minutes. The distance and voltage were optimized for different film thickness and geometries. The temperature of the substrate was between 200 °C and 400 °C during poling.

Finally a top electrode was fabricated on the BIT/PZT film as shown in Fig. 2(c) (step S6). We used a silver paste to form the top electrode at room temperature instead of vacuum sputtering reported in [9], [10]. This convenient approach makes the selection of electrode size, which is the sensor size, simple. The silver paste was tested and its operating temperature could be



Fig. 2. Setup for fabrication of ceramic film ultrasonic transducers by a sol-gel spray technique. (a) Setup for sol-gel spray. (b) Setup for corona poling. (c) A 90- μ m-thick BIT/PZT film transducer deposited on steel substrate.

above 500 °C. The optimum diameter of the top electrode for this UT in order to obtain the highest signal strength was around 10 mm [15]. The measured density and dielectric constant of the film were 4 g/cm³ and 90, respectively.

III. ULTRASONIC PERFORMANCE

To provide the desired center frequency in the range of 3.6-30 MHz, $40-200 \ \mu m$ thick films were produced onto various shapes of substrates such as planar and curved surfaces. This frequency range is commonly preferred for nondestructive evaluation of polymers and metals because of its sufficient ranging resolution and acceptable ultrasonic attenuation in



Fig. 3. (a) Longitudinal wave ultrasonic signals reflected at the substrate-air interface and (b) frequency spectrum of the L¹ echo, measured at 500 °C. Lⁿ(n = 1, 2, 3...) denotes *n*th roundtrip echo in the substrate.

such materials. Here, we only present one HTUT's performance fabricated onto a 50.8-mm-long, 25.4-mm-wide, and 12.7-mm-thick steel substrate shown in Fig. 2(c). The thickness of the BIT/PZT film was 90 μ m and the top electrode diameter was 10 mm. The HTUT in Fig. 2(c) was placed on an electric hot plate and heated. The temperature was measured on the top surface of the substrate, 20 mm apart from the top electrode, by a thermocouple. Fig. 3(a) presents an ultrasonic longitudinal wave signals reflected at the substrate-air interface measured in pulse-echo technique at a temperature of 500 °C. Lⁿ denotes nth roundtrip echo propagating through thickness direction in the substrate. The SNR for the first roundtrip echo L^1 , was 34 dB. Fig. 3(b) shows frequency spectrum of the L^1 echo. The center frequency was 8.5 MHz and the 6-dB bandwidth was 4 MHz. Such performance is sufficient for monitoring of polymer injection molding processes, for which the melt temperatures are usually less than 400 °C [16].

IV. MONITORING OF INJECTION MOLDING PROCESS

The developed BIT/PZT film HTUTs were applied to monitoring of polymer injection molding process. Injection molding is the process of forcing melted polymer into a mold cavity having an unique shape for production parts designed. Fig. 4 shows a typical cycle of the injection molding: (i) the polymer melt is injected into the cavity of a mold through a gate; (ii) the cavity is completely filled with the material and the additional melt is forced into the cavity under high pressure until the gate is frozen in order to compensate for the shrinkage due to the continuous cooling. Then, the part is further cooled until it is sufficiently solidified; (iii) the mold is opened and the part is detached from the immobile mold; and (iv) the part is ejected



Fig. 4. Cycle of the injection molding process: (i) filling; (ii) packing, holding and cooling; (iii) mold open; (iv) part ejection.



Fig. 5. HTUT sensor inserts (right) with and (left) without an electrical connection used for monitoring of injection molding process.

from the cavity of the mobile mold by the ejection pin. Then, the mold is closed and the entire cycle (i)-(iv) is repeated.

A. Experimental Setup

In order to monitor the behavior of the polymer inside the mold cavity during the injection molding process, HTUT sensor inserts were fabricated and then embedded into the mold insert. Mold inserts are commonly used by injection molders, in particular, for multicavity molding processes. Our approach using the sensor inserts in combination with the mold insert is practical for molds of all sizes. Fig. 5 shows the HTUT sensor inserts (right) with and (left) without an electrical connection. Teflon insulated coaxial cables were used for electrical connection between the top electrode and the pulser-receiver used. The maximum operating temperature of this coaxial cable was 200 °C. For applications at temperatures higher than 200 °C, a ceramic insulated coaxial cable can be used as an electrical cable.

Four HTUT sensor inserts (UT1-4) embedded into the mold insert are shown in Fig. 6, which also demonstrates that sensor array configuration is feasible. The mold and mold insert were made of the same steel as those of the HTUT sensor inserts in Fig. 5. The mold insert had dimensions of 76-mm width, 165-mm length, and 21-mm thickness, which was 1 mm thinner







Fig. 6. Mobile mold with four HTUT sensor inserts embedded in the mold insert. (a) Front view (polymer side). (b) Back view (UT side).

than the mobile mold shown in Fig. 6(a) and (b) so that a rectangular part with dimensions of 76-mm width, 165-mm length, and 1-mm thickness could be molded. A hole at the center of the mold insert was for an ejection pin, as seen in Fig. 4(iv). By replacing the mold insert, the shape and dimensions of the molded part can be easily modified to meet the customer's demands. The bottom surfaces of the HTUT sensor inserts were flushed with the mold insert surface (cavity surface), as seen in Fig. 6(a). A distance between the center of the UT1 (UT3) and UT2 (UT4) was 34.9 mm and that of the UT2 and UT3 was 44.5 mm, as shown in Fig. 6(b). All the HTUTs had almost the same ultrasonic performance, which indicated that fabrication of the HTUTs was consistent. The center frequency and the 6-dB bandwidth were 9–11 and 6–8 MHz, respectively.

Fig. 7 presents a cross-sectional view of the mold (mobile and immobile), mold insert and molded part (polymer) with four HTUT sensor inserts (UT1-4). Polymer melt was injected into the cavity of the mold through the gate at the center of the immobile mold. For comparison purpose with ultrasonic data, a temperature and cavity pressure sensor (6190A, Kistler Instrument AG, Winterthur, Switzerland), whose sensing end had circular shape with a diameter of 4 mm and was flushed with the internal surface of the immobile mold, was attached to the immobile mold. This Kistler sensor probing end was facing to the UT1 as shown in Fig. 7. $L^n(n = 1, 2, 3...)$ represents *n*th roundtrip echoes propagating in the HTUT insert and reflected



Fig. 7. Cross-sectional view of the mold (mobile and immobile), mold insert, and molded part (polymer) with four HTUT sensor inserts (UT1-4). L^n and $L_{2n}(n = 1, 2, 3...)$ represent *n*th roundtrip echoes propagating in the HTUT insert and those in the polymer, respectively.

at the insert-polymer interface, and $L_{2n}(n = 1, 2, 3...)$ represents those in the polymer and reflected at the polymer-immobile mold interface.

All the experiments presented in this study were conducted in ultrasonic pulse-echo mode. The ultrasonic data acquisition system was composed of four pulser-receivers (PR35, JSR Ultrasonics, Pittsford, NY, for UT1 and UT2; 5072PR, Panametrics, Inc., Waltham, MA, for UT3 and UT4), two 12-bit dual-channel digitizing boards (CompuScope 12100, Gage Applied Science Inc., Montreal, QC, Canada) with a sampling rate of 50 MHz for each channel and two personal computers. The experiments were carried out using a 150-ton injection molding machine (Engel GmbH, Schwertberg, Austria). A material employed was an injection grade polycarbonate (CALIBRE 200-14, Dow Chemical Co., Midland, MI), which is amorphous thermoplastic. The molding conditions were set as follows: melt temperature was 320 °C; mold temperature was 120 °C; and injection (plunger) speed was 110 mm/s. The total cycle time was about 30 s. The data acquisition rate was 10 Hz, unless particularly mentioned.

B. Results

Fig. 8 shows a typical result of acquired signals with the UT1 during the injection process. Although the signals were acquired during whole cycle (30 s) in time delay range from 4 to 24 μ s covering from the L¹ to L⁵ echoes, only the signals at the beginning (from 4 to 9 s) and the end (from 22 to 27 s) of the process in the time delay range from 4 to 9.6 μ s are shown in Fig. 8. It is noted that the signals presented in Fig. 8 are raw data without any signal processing. One can see the L¹ and L² echoes reflected at the insert-polymer or -air interface, depending on if the polymer existed at the UT location or not. S¹ represents the



Fig. 8. Typical signals measured with the UT1 during injection process.

first roundtrip shear wave echo reflected from the bottom of the HTUT sensor inserts. When the polymer melt arrived at the UT1 location at the process time of 5.8 s, the L_2 and L_4 echoes propagating in the polymer started to appear. At this moment, the L_6 and L_8 echoes were out of the time delay range shown in Fig. 8. At the process time of 25.5 s, the L_2 , L_4 , L_6 and L_8 echoes vanished, which indicates that the molded part was detached from the UT sensor insert.

In order to further investigate the correlation between ultrasonic signals observed and the process cycle, the amplitude values of the L^4 and L_2 echoes with respect to the process time were obtained using the signals measured with the UT1. Here, the L^4 echo was chosen instead of the L^1 echo since, in principle, higher order roundtrip echoes of Lⁿ can lead to higher sensitivity to the UT insert-polymer interface condition because of more reflections at this interface [2]. The results are presented in Fig. 9(a). In order to improve the SNR of amplitude values of these echoes shown in Fig. 9(a), signal processing was conducted as follows: spline interpolation on the acquired raw signals, shown in Fig. 8, to determine the peak amplitude values of the echoes, and moving average of 3-9 points of the amplitude values obtained with respect to the process time. The numbers of the averaging points were carefully chosen not to affect on time resolution for process monitoring. Further improvement of the SNR could be realized if analog and/or digital filtering techniques are used.

Temperature and pressure values measured by the Kistler sensor are also presented in Fig. 9(b). It is noted that the ultrasonic data shown in Fig. 9(a) were acquired with a different cycle from that for the data shown in Fig. 8. Hence, the process time at which the L_2 echo appeared or disappeared in Fig. 9(a) was different from that shown in Fig. 8. At the process time A, the polymer melt arrived at the UT1 and the Kistler sensor locations simultaneously. The amplitude of the L^4 echo suddenly decreased and the L_2 echo started to appear as seen in



Fig. 9. (a) Amplitude variations of L^4 and L_2 echoes measured with UT1 and (b) temperature (solid line) and pressure (dotted line) variation measured with Kistler sensor during injection process. Arrows A, B, and C indicate the time for flow front arrival at UT1 location, mold open, and part ejection, respectively.

Fig. 9(a) due to the fact that a part of the ultrasonic energy was transmitted into the polymer through the UT1 insert-polymer interface and then reflected back at the polymer-immobile mold interface.

At the time A, the pressure and temperature raised up sharply from zero to 40 MPa and from 116 °C to 137 °C, respectively, as shown in Fig. 9(b). It should be noted that the pressure sensor has a threshold in its response, resulting in a slight time delay comparing with the ultrasonic data (\sim 0.1 s with our experimental conditions). In addition, the measured temperature was smaller than the melt temperature of 320 °C since it was not the melt temperature but "contact (or surface)" temperature of the part, which is significantly affected by the interface condition between the sensor and the polymer.

Just after the time A, the amplitude of the L_2 echoes decreased and increased during a few seconds in the time range between 5 and 7 s, which might reflect the variation of ultrasonic attenuation in the polymer because of its solidification. This will be discussed further later. At the process time B, the mold was opened and the molded part was detached from the immobile mold as shown in Fig. 4(iii). Consequently, the amplitude of the L_2 echo increased due to the almost total reflection at the polymer-air interface, and the pressure dropped to zero. In addition, the phase change of 180° was observed on the L_2 echo at the time B since acoustic impedance of the polymer is smaller than that of the steel mold but larger than that of air, resulting in the change of sign of reflection coefficient at this interface from plus to minus. Such phase reverse of the L_2 echo



Fig. 10. Amplitude variation of L^4 echoes measured with UT3 and UT4 showing the time difference (A-A') of flow front arrival at each UT location.

is also a good indication of the part detachment from the immobile mold. At the time C, the amplitude of the L^4 echo recovered to its initial value and the L_2 echo disappeared, indicating that the part was detached from the UT1 insert due to the part ejection as shown in Fig. 4(iv). Hence, the times of the flow front arrival, mold open and part detachment were clearly observed on the ultrasonic echoes as indicated by the arrows A, B, and C, respectively, in Fig. 9(a).

C. Discussion

1) Flow Front Arrival and Flow Speed: During the filling stage, as shown in Fig. 4(i), the flow front advancement and flow front velocity are critical information. The flow front position can be used to control the plunger speed so as to allow smooth transition from the filling to the packing/holding stages, as shown in Fig. 4(ii), to avoid part flashing and mold damage due to high impact. Fig. 10 shows the L⁴ echoes measured with the UT3 and UT4. In this experiment, the signals were acquired every 3 ms (333 Hz) for higher time resolution. As mentioned previously, the amplitude of the L^4 echoes steeply decreased at the time A for UT3 and A' for UT4, indicating flow front arrival at each UT location. Therefore, the average flow front velocity V_{f} between the UT3 and UT4 was calculate to be 306 mm/s by using $V_f = L/\Delta t_f$, where L(= 34.9 mm) was the distance between UT3 and UT4 and $\Delta t_f (= 114 \text{ ms})$ was the time difference between A and A'.

2) Filling Monitoring: Filling completion of the mold with the materials is the most critical requirement for the molding process since the incomplete part must be rejected. Fig. 11 shows one incomplete (#1) and three complete (#2-4) parts molded successively under the same molding conditions. Accidentally the part #1 had defects on both bottom edges as indicated by arrows in Fig. 11. The volume filling rate for the part #1 was calculated to be 99%. Fig. 12 presents the amplitude of the L₂ echoes, obtained during the cycles for the parts #1-4 shown in Fig. 11, with the UT1-4, whose locations are indicated in Fig. 11. Even though the part was filled at UT3 and UT4 locations for the part #1, one can see that the L_2 echoes measured with the UT4 appeared for a few seconds only at the beginning of the cycle. In addition, the amplitude of the L₂ echoes measured with the UT3 gradually decreased to noise level before the mold opened at time B. These are due to the fact



Fig. 11. One incomplete (#1) and three complete (#2–4) parts molded successively under the same molding conditions. Filling rate of part #1 was 99%.



Fig. 12. Amplitude variation of L_2 echoes measured during injection cycles for parts #1–4 shown in Fig. 11.

that the part detachment at the UT3 and UT4 locations occurred before the mold opened because of shrinkage of the part caused by the lack of enough filling pressure. The L_2 echoes measured with the UT2 for the part #1 appeared in the entire time range between A and B; however, the amplitude was a little smaller comparing with those for the part #2–4. It is concluded that the presented ultrasonic method has the ability to monitor the incomplete filling of the part even with filling rate of 99%.

It is noted that the part ejection time C was not observed on the L_2 echoes measured with UT2 and UT3 for the part #2–4, as shown in Fig. 12, since the part detachment at UT2 and UT3 locations had already occurred at time B, and no ultrasonic energy was transmitted and reflected from the part. This is due to that the center of the part along with gate area was weakly pulled toward the immobile mold when the mold was opened at time B, as shown in Fig. 4(iii), resulting in the slight bending of the center area of the part and detachment of this area from the UT2 and UT3 locations which are near the gate in the immobile mold. The gate area was cut from the molded parts after the ejection; hence, it is not seen on the parts shown in Fig. 11.

3) Solidification: Ultrasonic velocity and attenuation inside the molded parts are strongly related to the physical properties



Fig. 13. Ultrasonic velocity in the molded part and attenuation of L_{2n} echo obtained using L_2 and L_4 echoes measured with UT2.

of the molded part. Therefore, solidification of the molded part may be monitored with the velocity and attenuation during the process. They can be determined using the time delay and amplitude variations of the L_{2n} echoes propagating in the polymer, as shown in Fig. 8, provided that the echoes have sufficient SNR. The ultrasonic velocity v is determined by

$$v = \frac{2h}{t_4 - t_2} \tag{1}$$

where h is the thickness of the mold cavity measured at the UT location by a micrometer, and t_2 and t_4 are the time delay of the L₂ and L₄ echoes, respectively. It is difficult to estimate the acoustic properties such as acoustic impedance of the UT insert and the mold during the process due to their temperature change, which are necessary to calculate the ultrasonic attenuation in the molded part. Hence, the attenuation α of the amplitude of the L_{2n} echo, which is associated with the ultrasonic attenuation in the molded part, is calculated by

$$\alpha = \frac{10}{h} \log_{10} \left(\frac{A_2}{A_4} \right) \tag{2}$$

where A_2 and A_4 are the amplitude of the L_2 and L_4 echoes, respectively [17], [18]. The spline interpolation and moving average techniques were conducted on the acquired signals to obtain the time delay and amplitude of the echoes used for velocity and attenuation measurements, respectively.

Fig. 13 presents the ultrasonic velocity and the attenuation of the L_{2n} echo obtained at the UT2 location using (1) and (2), respectively. At the process time between 6.3 and 7.2 s, indicated by the dotted lines in Fig. 13, the data are missed, since the ultrasonic attenuation in the polymer was so high due to the solidification [16] that the L_4 echoes didn't have sufficient SNR to determine its time delay and amplitude. The measured velocity inside the polymer varied from 950 to 1600 m/s and the attenuation from 6 to 3 dB/mm due to the variation of the material properties of the part, such as elastic constants, viscosity, and density, indicating a transformation of the polymer from the molten state to solid state [16].

V. CONCLUSION

Thick film (90 μ m) piezoelectric ceramic HTUTs have been successfully deposited on metallic substrates by a sol-gel spray technique. The ball-milled BIT fine powders were dispersed into PZT solution to achieve the gel. The films with desired thickness have been obtained by coating multiple layers on the substrate. Piezoelectricity was achieved using the corona discharge poling method. The metal substrate served as the bottom electrode and the silver paste formed the top one. The center frequency of ultrasonic signals generated by these HTUTs was around 10 MHz and a SNR was more than 30 dB in the pulse-echo mode at 500 °C. The main advantages of these new HTUTs are that they 1) are applicable at temperatures higher than 500 °C, 2) are miniature, 3) can be coated on the flat and curved surfaces, 4) do not need ultrasonic couplant, 5) can be operated at low and medium megahertz frequency range with sufficient frequency bandwidth, and 6) have sufficient piezoelectric strength and SNR.

The developed new HTUTs have been applied to real-time monitoring of polymer injection molding process. The polymer behavior in the mold cavity were monitored using multiple HTUTs sensor inserts embedded in the mold insert at different locations. Mold inserts are commonly used by injection molders, in particular, for multicavity molding processes. Utilization of sensor inserts in the mold insert allow us to monitor the injection process with molds of all sizes. The material employed was injection grade polycarbonate and the molded part had a rectangular shape. Flow front arrival and average flow front velocity of the polymer inside the mold cavity was determined using the amplitude variation of the echo reflected from the bottom of the HTUT sensor inserts. This echo was also used to detect the time of the opening of the mold. The amplitude and/or phase variations of the echoes propagating in the molded part was used to detect the part ejection and to monitor the filling completion of the mold cavity with the polymer. The amplitude of the echoes was sensitive to the part detachment from the mold so that uncompleted part with filling rate of 99% could be detected. The solidification monitoring of the molded part was performed by measuring the ultrasonic velocity and attenuation in the molded part using the multiple echoes propagating in the part. The variation of ultrasonic velocity and attenuation were reflecting the material properties of the part, such as elastic constants, viscosity and density, during the solidification.

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