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Research Article

Reducing the Sink Marks of a Crystalline Polymer using External Gas-Assisted Injection Molding

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External gas-assisted injection molding (EGAIM) has been used to reduce the sink marks of amorphous polymer products, but that of crystalline polymer products has not yet been reported. EGAIM of a crystalline polymer product was investigated in this study, and the influences of process parameters on the sink marks were discussed based on experiments. An isotactic polypropylene (iPP) product was fabricated by EGAIM under different process conditions. A uniform design was applied as an experimental design to investigate the influences of the process parameters on the sink marks. A regression equation was established to describe the quantitative relationship between the important parameters and sink marks in which a data-processing method was applied to determine the optimal value of F_α at significant level α to reduce the possibility of omission of some important parameters. The results show that EGAIM was effective in reducing the sink marks in these iPP products, and the most important parameters were the cooling time, gas pressure, and gas time. This study also provides the quantitative relationship between the important parameters and sink marks as reference for the research of EGAIM on crystalline polymer.

1. Introduction

External gas-assisted injection molding (EGAIM) is an unconventional molding process that efficiently fabricates products with high precision and high surface quality, especially in significantly reducing sink marks [1–3]. A sink mark is usually defined as "an unwanted depression or dimple on the surface of a molding due to localized shrinkage." [4, 5]. The distinct localized shrinkage commonly results from a local thick-wall structure, such as ribs, bosses, and other similar structures [3, 6], which is also greatly affected by the process parameters in injection molding [7–9], including the melt temperature [10–12], packing pressure [13, 14], packing time [15], cooling time [16], injection pressure [17], and injection speed [18].

EGAIM is more complicated than the conventional injection molding (CIM) because of the introduction of gas. Gas with certain pressure is injected between the cavity surface of the mold and the solidified polymer formed during filling, after the cavity was filled by polymer. The gas pressure is maintained until the cooling process is terminated. The pressure on the solidified polymer pushes it to move and deform, which compensate the polymer shrinkage caused by temperature decrease during packing and cooling and reduce the sink marks.

The advantages and complexity of EGAIM have attracted the attention of researchers. Chen et al. [19] investigated the packing effects of EGAIM on the shrinkage and sink marks of plastic parts (ABS) under various rib designs and compared them with those of CIM. The results showed that EGAIM could further reduce the part shrinkage when the gas pressure and gas-packing time were both increased. Su et al. [20] also reduced the ghost marks in plastic parts of PA using EGAIM. Moreover, the quality of the parts was improved by increasing the mold temperature, melt temperature, injection speed, and pressure, although some limitations were experienced. Jiang et al. [21] discussed the relationship between the process parameters and part quality of ABS using the single-factor test method and developed a physical model that described the influence of gas-melting interaction on the sink marks [22].

The aforementioned studies focused on amorphous polymer products and did not explore the application of EGAIM to crystalline polymer products. Crystalline polymers are an important component in industrial production [23], aerospace [24], and pharmaceutical production [25]. In addition, the shrinkage of crystalline polymers is usually more obvious than that of amorphous polymers [26], and the sink marks of crystalline polymer products are usually larger than those of amorphous polymer products. Therefore, investigating

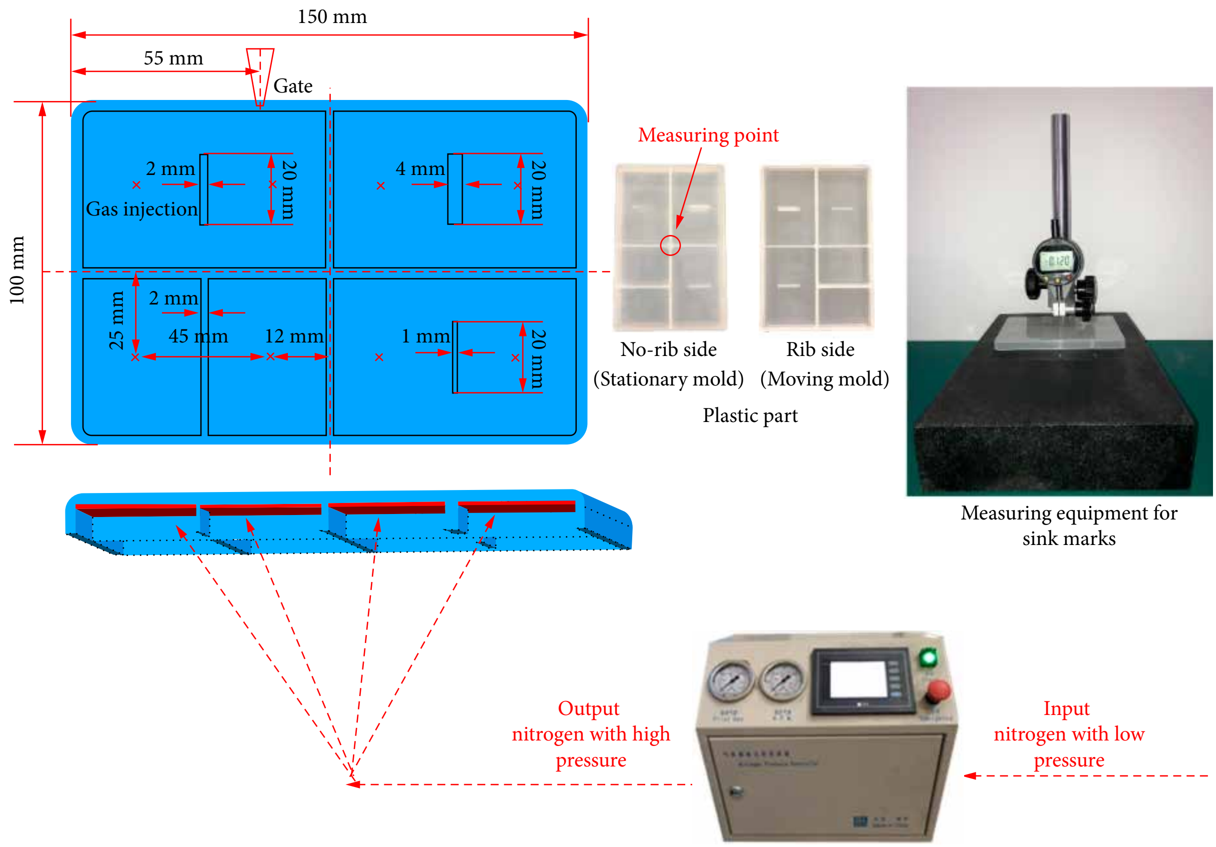

Figure 1: The equipment of EGAIM and measuring instruments.

the application of EGAIM to crystalline polymer products is important. In the present study, an iPP product was manufactured using EGAIM, and the influences of the process parameters on the sink marks of the iPP products were discussed. A regression equation was established to investigate the quantitative influence based on the experimental results in which a data-processing method was applied to determine the optimal value of F_α at significant level α to reduce the possibility of omission of some important parameters. To verify the reduction of the sink marks of these iPP products by EGAIM, experiments with or without gas were carried out under an optimal condition, as calculated by the regression equation.

2. Experiments

A semi-crystalline iPP (T30S, Zhenhai Branch of Sinopec Corp., China) product was used in this study, and its parameters were as follows: melt flow index of 2.5 g/10 min, melting point of 167°C, density of 0.91 g/cm^3, and isotactic index of more than 94%. An injection molding machine (MA3800/2250, Haitian International Holdings Ltd., China) was used to manufacture the products with dimensions of 150 mm × 100 mm × 3 mm. After filling at a later time (delay time), an inert gas (N_2) with certain pressure was injected between the solidified polymer and cavity surface of the moving mold using a nitrogen pressure controller (C8-01, Beijing Chn-Top Machinery Group Co., Ltd., China). The sink marks of the crystalline polymer products were measured by a dial indicator (A0-12.7, Shanghai Siwei Instrument Manufacturing Co., Ltd., China), which has a measurement range of 12.7 mm and accuracy of 0.001 mm. The locations of gate, gas injections and measuring point are shown in Figure 1.

To study the efficiency of EGAIM on the reduction of sink marks in the crystalline polymer products, almost all of the process parameters were considered, including the material temperature, injection pressure, injection speed, packing pressure, packing time, cooling time, gas pressure, gas time, and delay time. The uniform design focuses on the uniform distribution of test points within the test range to obtain the largest available information with the least number of tests. Thus, the design is especially suitable for a multi-factor test, and the system model is completely unknown [29]. A uniform design is chosen to design the experiments using uniform table remarking $U_{30}(3^1 \times 5^1 \times 6^2 \times 10^5)$. The levels and parameters are listed in Table 1, where X_1–X_9 represent the material temperature, injection pressure, injection speed, packing pressure, packing time, cooling time, gas pressure, gas time, and delay time, respectively. The nine parameters were selected referring some studies of the sink marks [10–18].

3. Data-Processing Method

Regression analysis is a set of statistical processes that estimates the relationship between the parameters and results. More specifically, regression analysis helps us understand how the

Table 1: Uniform table remarking $U_{30}(3^1 \times 5^1 \times 6^2 \times 10^5)$ in EGAIM.

Number	Parameter X								
	X_1 (°C)	X_2 (MPa)	X_3 (mm/s)	X_4 (MPa)	X_5 (s)	X_6 (s)	X_7 (MPa)	X_8 (s)	X_9 (s)
1	180	65	25	10	1	10	0	5	0
2	200	70	30	20	2	15	1	10	0.5
3	220	75	35	30	3	20	2	15	1
4	—	80	40	40	4	25	3	20	1.5
5	—	85	45	50	5	30	4	25	2
6	—	90	50	60	—	35	5	30	2.5
7	—	95	55	70	—	40	6	—	—
8	—	100	60	80	—	45	7	—	—
9	—	105	65	90	—	50	8	—	—
10	—	110	70	100	—	55	9	—	—

Table 2: Uniform table.

Number N_j	Parameter X_l				Result Y
	X_1	X_2	$\cdots$	X_P	$Y = X_{P+1}$
N_1	X_{11}	X_{12}	$\cdots$	X_{1P}	$Y = X_{1(P+1)}$
N_2	X_{21}	X_{22}	$\cdots$	X_{2P}	$Y = X_{2(P+1)}$
$\cdots$	$\cdots$	$\cdots$	$\cdots$	$\cdots$	$\cdots$
N_n	X_{n1}	X_{n2}	$\cdots$	X_{nP}	$Y = X_{n(P+1)}$

typical values of parameters change when any one of the results is varied [28]. One of the most important steps in regression analysis is the determination of optimal value F_α at significant level α, which directly determines whether parameters are introduced into the regression equation. In general, significant level $\alpha = 0.05$ is commonly used in statistics [29]. However, some important parameters are sometimes omitted in injection molding. Thus, a data-processing method was applied in this study to establish the optimal value of F_α at significant level α, which would reduce the possibility of omission of some important parameters. The data in the uniform table need to be dealt with by this processing method.

A uniform table was designed using a uniform design to reduce the number of experiments. The uniform table consists of N_j $(j = 1, 2, \ldots, n)$ experiments, X_l $(l = 1, 2, \ldots, P)$ parameters, and Y (recorded as X_j, where $j = P + 1$) results. The value of parameter X_l in the jth experiment is X_{jl}, and result Y is recorded as $X_{j(P+1)}$, as listed in Table 2.

Correlation coefficient r_{ij} accurately describes the reliability between the parameters and results. The value of r_{ij} is positively correlated with the reliability between the parameters and results, as expressed in Equation. (1) [30]

$$r_{ij} = \frac{l_{ij}}{\sqrt{l_{ii}l_{jj}}} = \frac{\sum_{k=1}^{n}\left[\left(X_{ki} - \frac{1}{n}\sum_{k=1}^{n}X_{ki}\right)\cdot\left(X_{kj} - \frac{1}{n}\sum_{k=1}^{n}X_{kj}\right)\right]}{\sqrt{\sum_{k=1}^{n}\left(X_{ki} - \overline{X}_i\right)^2 \cdot \sum_{k=1}^{n}\left(X_{kj} - \overline{X}_j\right)^2}};$$
$$i = 1, 2, \ldots, P + 1,\ j = 1, 2, \ldots, n, \tag{1}$$

where l_{ij} and l_{ii} are the sum of the products of the mean deviation and sum of squares of the deviation from the mean, respectively. $\overline{X}_i$ is the average value of X_{jl} $(l = 1, 2, \ldots, P+1, j = 1, 2, \ldots, n)$. The important parameters were selected by comparing the values of correlation coefficient r_{ij}.

The r_{ij} forms the matrix of the correlation coefficients, as expressed in Equation. (2) [31].

$$\left[r_{ij}\right] = \begin{bmatrix} r_{11} & r_{12} & \cdots & r_{1P} & r_{1(P+1)} \\ r_{21} & r_{22} & \cdots & r_{2P} & r_{2(P+1)} \\ r_{31} & r_{32} & \cdots & r_{3P} & r_{3(P+1)} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ r_{n1} & r_{n2} & \cdots & r_{nP} & r_{n(P+1)} \end{bmatrix} \tag{2}$$

Generally, F should be calculated for every parameter in the regression analysis and is compared with pre-determined F_α at significant level α to determine whether the parameter is retained or not. In this study, the important parameters were obtained by comparing the correlation coefficients that should be included in the regression analysis. If the commonly used significant level, i.e., $\alpha = 0.05$, in the statistics is applied to the regression analysis in EGAIM, some important parameters might be omitted. Thus, this study introduced a method for modifying significant level α and corresponding F_α.

In this work, the data processing can be summarized by the following steps.

Step 1. A set of significant level α in a certain range and corresponding F_α are obtained using the critical value table of the F distribution.

Step 2. The F_{X_l} values of the important parameters are calculated using Equation (3) [32],where subscript X_l represents the selected important parameters using the comparison of the correlation coefficients.

$$F_{X_l} = \frac{\left[\frac{\left(r_{i(P+1)}\right)^2}{r_{ii}}\right] \times (n-3)}{r_{ii} - \left[\frac{\left(r_{i(P+1)}\right)^2}{r_{ii}}\right]};\ i = 1, 2, \ldots, P + 1 \tag{3}$$

Step 3. Minimum F_{X_l} is compared with F_α using Equation (4). If minimum F_{X_l} is less than F_α, F_α cannot ensure that the important parameters are included in the regression equation. If minimum F_{X_l} is larger than or equal to F_α, F_α is suitable for the regression analysis.

Table 3: Values of the sink marks of the crystalline polymer products.

Number	Parameter X_l									Sink mark Y
	X_1 (°C)	X_2 (MPa)	X_3 (mm/s)	X_4 (MPa)	X_5 (s)	X_6 (s)	X_7 (MPa)	X_8 (s)	X_9 (s)	$Y = X_{10}$ (mm)
1	180	105	40	10	5	35	3	5	1.5	0.264
2	220	110	35	80	2	25	8	10	2	0.155
3	220	70	60	90	3	10	1	30	1.5	0.0723
4	180	75	25	50	2	25	2	20	2	0.121
5	200	100	50	100	1	40	6	30	0.5	0.098
6	200	110	60	50	5	50	7	25	2	0.067
7	180	85	65	90	2	45	9	5	1	0.202
8	200	95	30	40	2	40	0	15	2.5	0.168
9	220	85	35	70	4	45	3	30	2.5	0.143
10	180	90	35	60	5	20	8	30	1	0.127
11	200	110	25	40	3	10	40	25	0	0.040
12	180	90	70	40	1	15	4	10	0.5	0.176
13	220	85	40	30	1	30	90	25	0	0.073
14	220	95	25	100	4	50	5	15	1	0.157
15	220	65	55	50	5	35	70	10	0	0.154
16	180	70	30	20	4	40	6	15	0.5	0.107
17	180	65	45	80	1	35	5	25	2	0.080
18	180	95	70	30	4	30	0	25	1.5	0.213
19	200	80	60	20	2	45	40	30	0	0.066
20	200	80	45	80	5	15	0	15	0.5	0.120
21	220	75	70	60	3	55	6	20	1.5	0.132
22	200	70	40	30	3	55	8	5	1.5	0.208
23	180	100	55	70	3	10	7	15	2.5	0.112
24	220	105	65	10	2	20	5	20	1	0.079
25	180	105	45	90	3	55	10	20	0	0.117
26	200	75	50	10	4	15	9	20	2.5	0.044
27	220	90	50	20	1	50	1	10	2	0.240
28	220	100	55	60	4	25	2	5	0.5	0.167
29	200	65	30	70	1	20	2	5	1	0.172
30	200	80	65	100	5	30	4	10	2.5	0.145

$$\begin{cases} \min F_{X_l} < F_\alpha; \text{unsuitable} \\ \min F_{X_l} \geq F_\alpha; \text{suitable} \end{cases} \tag{4}$$

Step 4. Correspondingly, this condition results in many suitable F_α values based on Step (2), but not all F_α values are optimal F_α. To improve the accuracy, maximum F_α is considered as the optimal value from a large number of suitable F_α values.

The aforementioned data-processing method provides the basis for selecting F_α at significant level α by comparing it with F_{X_l} of the important parameters, which reduces the possibility of omission of some important parameters in the regression equation and improves the accuracy of this equation.

4. Results and Discussion

4.1. Correlation Coefficients of the Parameters. The sink marks of the crystalline polymer products measured by the dial indicator are listed in Table 3.

The matrix of the correlation coefficients is shown in the form of a heat map by combining Equations (1) and (2) to intuitively express the relationship between the parameters and sink marks, as shown in Figure 2. In the heat map, red and blue represent the positive and negative effects, respectively, and the number and shade of the colors indicate the correlation between the abscissa and ordinate. The value of the correlation coefficient between the gas time and sink marks is the largest value, which reaches −0.71. Thus, gas time is the most important parameter in EGAIM, and the negative sign indicates that the relationship between the gas time and sink marks is positive. The longer the gas time is, the smaller is the sink-mark value. The correlation coefficients of the cooling time and gas pressure are also large, which reach 0.32 and −0.23, respectively. Therefore, the gas time, cooling time, and gas pressure are important in the sink marks of the crystalline polymer products. The value of the correlation coefficient between the delay time and gas pressure is also large, which reaches 0.27. Thus, the influence of the delay time and gas pressure should not be neglected. The determination of the important parameters establishes a basis for exploring the

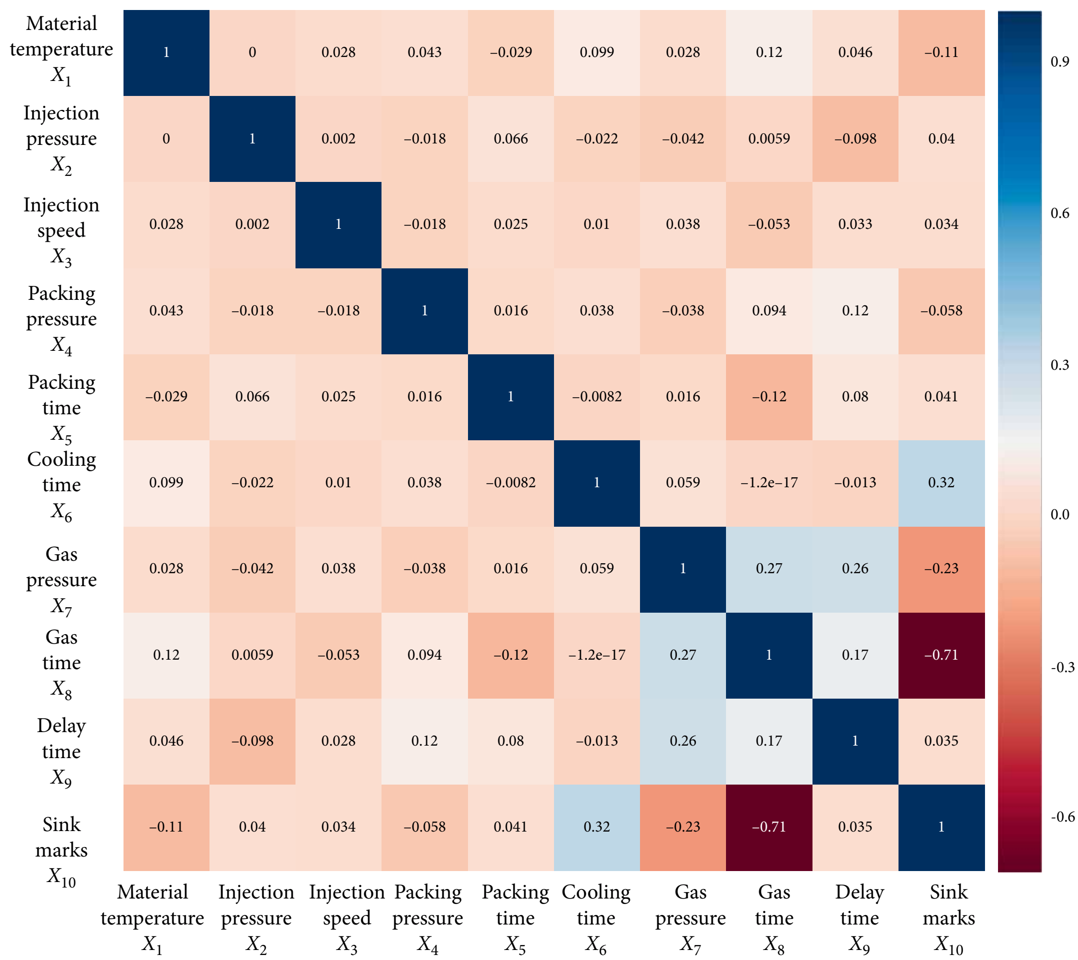

Figure 2: Heat map of the matrix of the correlation coefficients.

quantitative relationship between the parameters and sink marks of the crystalline products in EGAIM.

4.2. Establishment of the Regression Equation. According to the calculation by Equation (3), the F_{X_i} values of the cooling time, gas pressure, and gas time, were 3.19, 1.56, and 28.46, respectively. F_{X_i} of the interaction of the gas pressure and delay time was 2.03. One of the most important steps in considering the regression equation is to determine F_α at significant level α to select the parameters. In general, F_α at significant level α should be defined before exploring the regression equation. However, F_α depends on the preferences of different people. α is generally defined in statistics as 0.05, and $F_\alpha = 2.9$ at significant level $\alpha = 0.05$. If $F_\alpha = 2.9$ is used to investigate the EGAIM parameters, the cooling and gas times are considered in the regression equation because $F_{X_i} > F_\alpha = 2.9$. However, the gas pressure is omitted from the equation, which is unacceptable according to the aforementioned discussion. Significant level α was thus adjusted to $\alpha = 0.3$ to ensure that all important parameters were included into the regression equation. $F_\alpha = 1.4$ at $\alpha = 0.3$. All the F_{X_i} values of the important parameters were larger than F_α, which avoided omitting the important parameters.

To obtain the quantitative relationship between the parameters and sink marks, the square term of the parameters and the interaction among the parameters were considered in the regression equation. The regression equation was investigated using the comparison between F_{X_i} and $F_\alpha = 1.4$, as expressed in Equation (5),

$$\begin{aligned} Y = {} & 0.0879 + 0.003025 \times X_6 + 0.02563 \times X_7 - 0.01050 \times \\ & X_5 + 0.1185 \times X_9 + 0.000223 \times X_5^2 - 0.03374 \times X_9^2 \\ & - 0.000514 \times X_7 \times X_8 - 0.00621 \times X_7 \times X_9 \end{aligned} \tag{5}$$

The ratio of variation to the total variation for Equation (5) was described by the important coefficient R^2, which is a measure of the degree of fit. When R^2 more approaches unity, the response model fits the actual data better. The value of R^2 in Equation (5) is 85.37%, which is acceptable.

The regression equation of the sink marks in EGAIM was obtained according to the analysis of the correlation coefficients, which revealed the quantitative relationship between the important parameters and sink marks of the crystalline polymer products in EGAIM.

4.3. Discussion on the Regression Equation. The regression equation explores the influence of important parameters

Table 4: Uniform table remarking $U_{15}(3^1 \times 5^4)$, including the unimportant parameters designed using the uniform design.

Number	Parameter X_l					Result Y
	Material temperature (°C) X_1	Injection pressure (MPa) X_2	Injection speed (mm/s) X_3	Packing pressure (MPa) X_4	Packing time (s) X_5	Sink marks (mm) $Y = X_{10}$
1	180	110	40	80	5	0.116
2	220	110	50	20	4	0.111
3	200	110	60	60	1	0.096
4	180	80	60	20	4	0.096
5	200	100	40	20	2	0.092
6	180	100	70	40	3	0.103
7	220	100	30	100	3	0.102
8	200	80	70	100	4	0.096
9	200	70	50	80	3	0.089
10	200	90	30	40	5	0.096
11	180	90	50	100	1	0.085
12	220	90	70	80	2	0.087
13	220	80	40	40	1	0.091
14	220	70	60	60	5	0.085
15	180	70	30	60	2	0.103

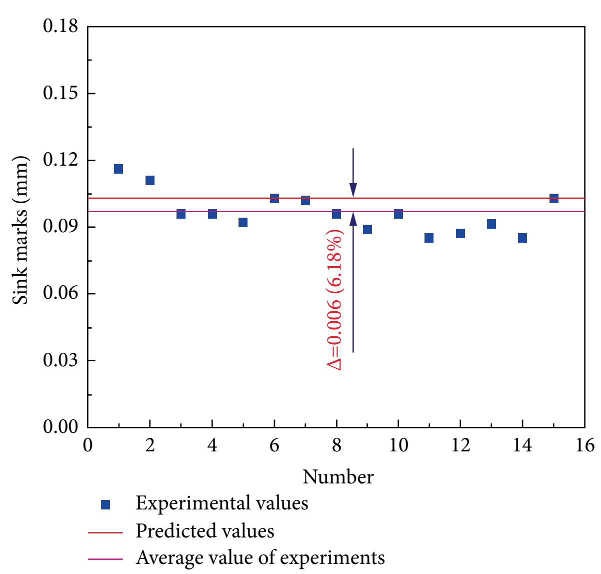

Figure 3: Comparison between the experiments and predictions in the regression equation.

on the sink marks of the crystalline polymer products. The minimum value of the sink marks was obtained by calculating the regression equation, which was equal to 0.103 mm when the cooling time, gas pressure, gas time, and delay time were 55 s, 9 Mpa, 30 s, and 1 s, respectively.

According to the previous research, when the important parameters were defined, the influence of the unimportant parameters on the sink marks of crystalline polymers could be ignored, including the material temperature, injection pressure, injection speed, packing pressure and packing time. When the important parameters were the same as those of the aforementioned values, i.e., the cooling time, gas pressure, gas time, and delay time were 55 s, 9 Mpa, 30 s and 1 s, respectively, and the other unimportant parameters were combined according to the uniform design in a certain range, the sink marks of the crystalline polymer under different unimportant parameters are as those listed in Table 4.

The experimental results are shown in Figure 3, and the values of the sink marks under different unimportant parameters fluctuate from the average value and reach 0.097 mm. The difference between the average and predicted values of the regression equation is 0.006 mm and reaches 6.18%, which indicates that the accuracy of the regression equation and the influence of these parameters on the sink marks of crystalline polymer products are almost negligible. The regression equation establishes the quantitative relationship between the important parameters and sink marks of the crystalline polymer products in EGAIM.

4.4. Reduction of Sink Marks Using EGAIM. The aforementioned research obtained the optimal important parameters for the sink marks, including a cooling time of 55 s, gas pressure of 9 Mpa, gas time of 30 s, and delay time of 1 s. the other unimportant parameters were randomly determined from no. 14 in Table 4, and the material temperature, injection pressure, injection speed, packing pressure, and packing time are 220°C, 70 Mpa, 60 mm/s, 60 Mpa, and 5 s, respectively. The sink marks of the crystalline polymer products with or without gas are compared and shown in Figure 4. Figure 4(a) shows that the sink mark of the crystalline polymer products without the gas is 0.365 mm, whereas that with the gas is only 0.085 mm, as shown in Figure 4(b). The difference in the sink marks of the crystalline polymer products with or without gas is 0.280 mm. Thus, EGAIM significantly reduces the sink marks of the crystalline polymer products. Therefore, EGAIM obviously reduces the sink marks of these iPP products. The results provide sufficient evidence for the application of crystalline polymers in EGAIM.

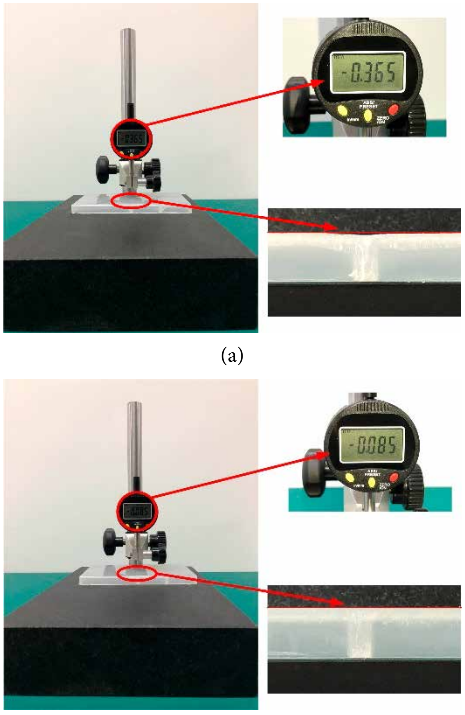

FIGURE 4: The values of sink marks of crystalline polymer products without gas (a) and with gas (b).

5. Conclusion

In this study, the application of EGAIM to an iPP product was investigated by experiment. According to the results of this study, the following conclusions can be drawn. (1) EGAIM can significantly reduce the sink marks of these iPP products. (2) The cooling time, gas pressure, and gas time are important parameters that affect the sink marks of the crystalline polymer products according to the correlation coefficients. (3) The regression equation can correctly predict the sink marks of the crystalline polymer products when the important parameters in EGAIM are constant. In general, this study verifies the effectiveness of EGAIM on reducing the sink marks of crystalline polymer products and provides the quantitative relationship between the important parameters and sink marks of crystalline polymer products in EGAIM.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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