Effect of Fillers and Processing Parameters on the Shrinkage of Injected Molding Polyamide 66

Fitri Ayu Radini, Muhammad Ghozali, Onny Ujianto, and Agus Haryono

***Abstract*—The in-flow and cross-flow shrinkage of injected molding polyamide 66 (PA66) could be effectively reduced by optimizing glass fiber content and molding condition. The chosen molding conditions were melt temperature, cooling time and cooling temperature. The filler effect between glass fiber and clay on PA66 shrinkage also being compared. The result showed that glass fiber can effectively reduce the in-flow and cross-flow shrinkage by giving 54% to 86% reduction of shrinkage percentage. The optimum shrinkage was predicted sequentially at glass fiber content, melting temperature, cooling time and cooling temperature for in-flow are 45%; 285 °C; 45 seconds; 90 °C and for cross-flow are 45%; 285 °C; 35 seconds; 60 °C. The verification for cross-flow shrinkage was 0.41%, while the predicted value was 0.39%. The optimum cross-flow molding condition was applied to PA66/clay composite giving 28% to 51% reduction for 1% to 5% clay and 23% to 77% reduction for 15 % to 45 % clay. The shrinkage reduction by clay was lower than glass fiber, however, the addition of clay giving isotropic shrinkage that leads to less warpage.**

***Index Terms*—Glass fiber, clay, polyamide 66, shrinkage, injection molding.**

I. INTRODUCTION

An injected molded Polyamide 66 (PA66) product as semicrystalline polymer has higher tendencies to shrinkage. This shrinkage behaviour is a disadvantage particularly for product that need tight dimension tolerances [1]-[3]. Therefore, it is important to investigate the significant factor affected shrinkage in PA66 product in order to achieved optimum stability.

Shrinkage in injection molding can be controlled by variation in some factors as type of filler, level of filler and molding conditions (melt temperature, injection pressure and speed, holding pressure and time, cooling temperature and time) [1], [3], [4]. Glass fiber (GF) is type of filler that commonly used in semicrystalline polymer composite to reduce shrinkage. However, glass fiber tend to give anisotropic shrinkage in composite product [1], [4]-[7]. Clay is one of promising alternative for filler in polymer composite, because of its advantages to increase polymer composite properties with lower content [4], [8], [9].

This research investigated the significant factor that affected PA66 shrinkage both at in-flow and cross-flow direction. The chosen factor were glass fiber percentage, melting temperature, cooling temperature and time. The minimum in-flow and cross-flow shrinkage percentage were also been investigated. The molding condition that resulted minimum shrinkage percentage was used to process PA66/clay composite. This was done to compare the effectiveness of glass fiber and clay on shrinkage of PA66.

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II. EXPERIMENTAL DETAILS

A. Materials

A Commercial PA66 Kopla KDP1000 from Korea was purchased from local supplier in Tangerang, Indonesia. Two types of filler used as filler comparison, namely an industrial grade short GF supplied by the same supplier as PA66, and ogano-modified clay Cloisite 20A produced by Southern Clay.

B. Composite Preparation

The PA66/GF and PA66/clay composite samples were injected using a Battenfeld BA 400 CDC injection molding machine. PA66 was dried in atmospheric oven at 80 °C for 4 hours, while clay was dried at the same condition for 72 hours before injection. The mold used in this research was ASTM D955-08 type D2. PA66/GF was produced according to four factors and three level of Box-Behnken Design of Experiment (DoE), shown in Table I. The optimum process conditions and GF composition to produce the lowest shrinkage were predicted and verified from generated model. The optimum process condition of PA66/GF was applied to PA66/clay and neat PA66 sample preparation. The clay compositions were varied to 1, 5, 10, 15, 30 and 45 wt%.

TABLE I: DOE BOUNDARIES FOR PA66/GF COMPOSITES

Factor	Unit	Low	Mid	High
% Glass Fiber	%	15	30	45
Melt Temp	°C	265	275	285
Cooling Time	Sec	15	30	45
Cooling Temp	°C	60	75	90

C. Characterizations

The length and width of three specimens from each run of PA66/GF and PA66/clay composites were measured using a digital calliper, 168 hours after production. The shrinkage percentage on in-flow and cross-flow direction was calculated according to equation 1 and 2.

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$$S_w = \frac{(W_m - W_s)}{W_m} \times 100 \quad (1)$$

$$S_l = \frac{(L_m - L_s)}{L_m} \times 100 \quad (2)$$

where:

S_w	:	Cross – Flow Shrinkage, %	S_l	:	In – Flow Shrinkage, %
W_m	:	The Width of Mold	L_m	:	The Length of Mold
W_s	:	The Width of Specimen	L_s	:	The Length of Specimen

Analysis on the effect of each factor on shrinkage percentage was done according to statistical model using Minitab software. The model was developed from collected shrinkage percentage data and regressed using polynomial equation as follows:

$$y = \beta_0 + \sum \beta_i x_i + \sum \beta_{ii} x_i^2 + \sum \beta_{ij} x_i x_j \quad (3)$$

where y is the model response, x_i and x_j are independent factor, and βo, βi, βii, and βij are the coefficients for the constant, linear, quadratic terms, and interactions, respectively. The model was developed by only considering significance factor (p-value < 0.05). The less p-value suggests that the effect of term is more significant to influence shrinkage percentage.

III. Results and Discussions

A. Shrinkage Measurements of PA 66/GF Composites

TABLE II: Shrinkage Percentage of PA66/GF Composites

Sample	Glass Fiber (%)	Melt Temp (°C)	Cooling Time (second)	Cooling Temp (°C)	In-Flow (%)	Cross-flow (%)
1	45	275	30	90	0.26	0.55
2	30	275	45	90	0.39	0.59
3	45	275	30	60	0.34	0.49
4	15	275	15	75	0.42	0.68
5	45	265	30	75	0.28	0.43
6	30	285	15	75	0.33	0.50
7	30	285	30	60	0.37	0.54
8	30	275	30	75	0.36	0.60
9	15	265	30	75	0.54	0.63
10	30	275	15	60	0.38	0.60
11	30	265	45	75	0.50	0.78
12	30	275	30	75	0.32	0.60
13	30	265	15	75	0.58	0.86
14	30	285	30	90	0.35	0.51
15	30	275	30	75	0.34	0.59
16	45	285	30	75	0.21	0.37
17	30	275	15	90	0.37	0.65
18	45	275	15	75	0.27	0.50
19	30	285	45	75	0.30	0.47
20	15	275	45	75	0.41	0.60
21	30	265	30	90	0.34	0.45
22	15	285	30	75	0.44	0.59
23	15	275	30	90	0.54	0.75
24	30	275	45	60	0.40	0.60
25	45	275	45	75	0.26	0.40
26	30	265	30	60	0.37	0.57
27	15	275	30	60	0.43	0.66
Control	0	285	35	60	1.70	2.20

Table II shows shrinkage percentage for all PA66/GF composite samples. The table shows that all samples with the addition of GF have lower shrinkage than control sample. For all condition, the lowest in-flow and cross-flow shrinkage percentage are 0.21% and 0.37%, giving 85% and 80% reduction of shrinkage percentage compare to neat PA66. While the highest in-flow and cross-flow shrinkage percentage are 0.58% and 0.86%, giving 61% and 54% reduction of shrinkage percentage compare to neat PA66. These suggest that addition of GF has succesfully reduced the shrinkage of PA66.

Statistical model was developed from in-flow and cross-flow shrinkage percentage of PA66/GF composites. Table III shows only the best model developed using Minitab Software. It is show that % GF and melt temperature are the significant factors that influence shrinkage for in-flow (p-value 0.000 and 0.004, respectively) and cross-flow (p-value 0.000 and 0.016) direction. The other factors, cooling time and cooling temperature, have higher p-value (p-value > 0.05) at in-flow and cross-flow direction of shrinkage, suggest insignificant effect of these parameters. These insignificant parameter result are in accordance with findings reported by Zhao, *et al.* [10].

The interacted factor also giving no significant effect for in-flow and cross-flow direction of shrinkage since its p-value higher than 0.05, which are 0.094 and 0.156.

TABLE III: Model Coefficient and p-Value of Shrinkage of PA66/GF Composites

Term	In-Flow		Term	Cross-Flow	
	Coef.	P-val.		Coef.	P-val.
Constant	0.374	0.000	**Constant**	0.555	0.002
%GF	-0.096	0.000	**%GF**	-0.098	0.000
Melt Temp	-0.050	0.004	**Melt Temp**	-0.062	0.016
C. Time	-0.008	0.608	**C. Time**	-0.031	0.211
C. Temp (CT)	-0.005	0.755	**C. Temp (CT)**	0.003	0.887
%GF * CT	-0.046	0.094	**%GF * CT**	0.047	0.156

The effect of GF content and melting temperature on in-flow and cross-flow shrinkage are shown in Fig. 1(a) and Fig. 1(b). Since cooling time and cooling temperature have no significant effect, further analysis will be done according to one setting of cooling time and temperature, namely at mid-setting.

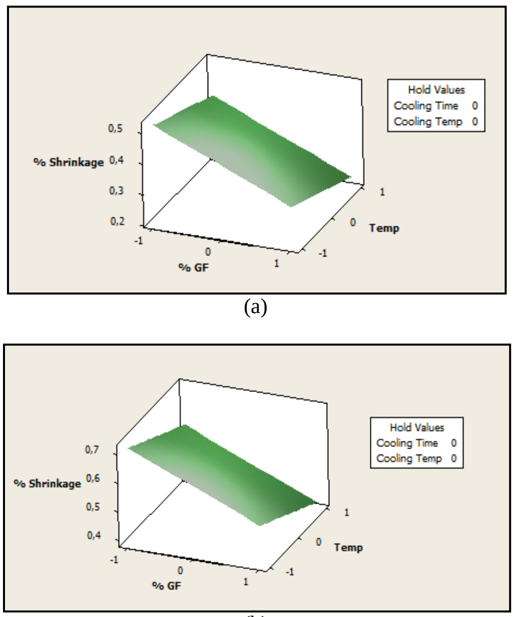

Fig. 1. Effect of significant factors on in-flow shrinkage (a), and cross-flow shrinkage (b) of PA66/GF composites.

Fig. 1(a) and Fig. 1(b) shows that shrinkage percentage is lower as melting temperature increase. This happens for in-flow and cross-flow direction. This result is correspond with other research [3], [11]-[13]. At low melting temperature, shrinkage is higher due to less effective material packing caused by lower melt flow rate of polymer. At higher melting temperature, material flow easily resulting more material in mold cavity to compensating shrinkage [1], [2], [14].

Shrinkage percentage for in-flow and cross-flow direction is also reduced by higher GF content as shown in Fig. 1. The low CTE (coefficients of thermal expansion) of GF will cause GF to contract less as the temperature changes. Since GF compensate some volume of polymer, this controlled contraction is contributed in lowering shrinkage of composite. This decrease is proportional with GF content [1], [4], [6], [8].

As can be seen in Fig. 2, at overall level (low,medium and high) of GF content and melting temperature, the shrinkage percentage of in-flow direction was lower than cross-flow direction. This shrinkage differential may be due to molded-in stress and differential orientation. Minimizing molded-in stress reduces differential shrinkage. One of the common causes of molded-in stress is lower melt temperature [1]. While the differential orientation is caused by the GF orientation that tend to orient in the flow direction when processing and cooling. This oriented fiber will prevent the in-flow direction to shrinking, while the cross-flow direction will shrink more to compensate the volume of injected part [1], [4].

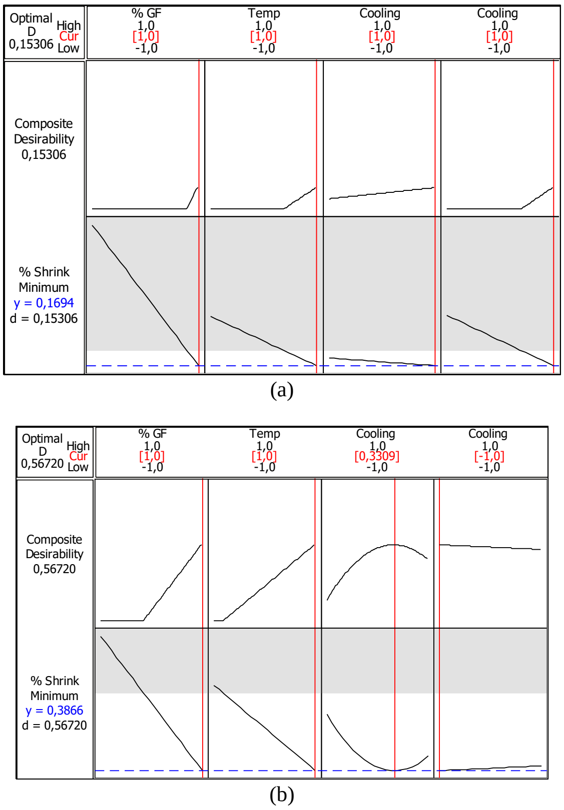

Fig. 2. Optimisation plot for in-flow shrinkage (a), and cross-flow shrinkage (b) of PA66/GF composites.

The optimum conditions to produce the lowest % shrinkage were predicted by response surface analyzer plot (Fig. 2). The plot shows that the optimum conditions for in-flow shrinkage are predicted at (1; 1; 1; 1) in coded unit or 45% GF, 285 °C for melting temperature, 45 second of cooling time and 90 °C for cooling temperature. Applying these conditions would have an in-flow shrinkage of 0.17%. As for cross-flow shrinkage, the lowest value is 0.39% with predicted formula (1 ; 1 ; 0.331 ; -1) in coded unit or 45% GF, 285 °C for melting temperature, 35 second of cooling time and 60 °C for cooling temperature. To validate the model and the predicted result, verification experiment was carried out at optimized conditions of cross-flow shrinkage since cross-flow shrinkage is higher than in-flow shrinkage. The cross-flow shrinkage value of verification sample was 0.41%, giving 78% reduction compare to neat PA66. The ranges of the minimum, optimum predicted, and verification are less than 1 standard deviation of % shrinkage (s.d = 0.11%) for all PA66/GF samples. This suggests that the model is capable of predicting the cross-flow shrinkage percentage.

B. Shrinkage Measurements of PA 66/Clay Composites

The optimum process conditions for cross-flow shrinkage of PA66/GF composite, which are 285 °C for melting temperature, 35 second of cooling time and 60 °C for cooling temperature was used to processed PA66/clay composite. The cross-flow direction was chosen because it had higher shrinkage than in-flow direction.

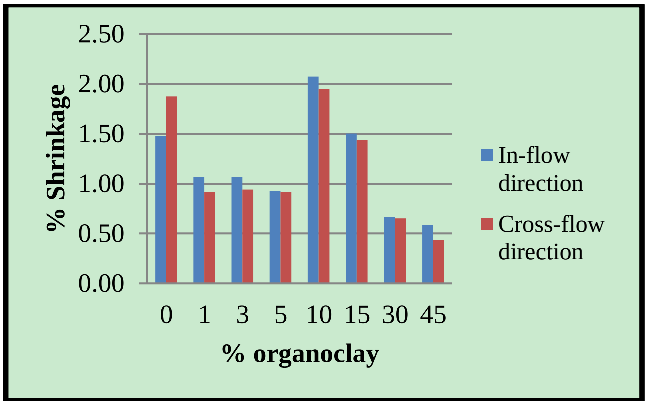

Fig. 3. In-flow and cross-flow shrinkage of PA66/clay composites.

Fig. 3 show the effect of clay content on PA66 shrinkage. Addition of 1% to 5% organoclay resulting 0.93% to 1.07% of in-flow shrinkage and 0.91% to 0.94% of cross-flow shrinkage, giving 37% to 58% reduction on shrinkage percentage compare to neat PA66. This result corresponding with other reseearch, which is organoclay can be used to lower shrinkage of PA66 [4], [8]. At this point, contraction of PA66 matrix is restricted by organoclay which has low CTE and the dispersion of organoclay probably also contributed in shrinkage reduction [8].

However, shrinkage percentage increased at 10% organoclay content. This might be due to crystallisation effect from adding organoclay into composites. Higher crystallinity would result in higher shrinkage [1], [4], [8], [9]. Then, the shrinkage percentage decreased with the addition of organoclay more than 10%. The lowest in-flow and cross-flow shrinkage was 0.59% and 0.43%, giving 60% and 77% reduction. This lowest value of in-flow and cross-flow shrinkage were achieved by 45% organoclay content.

This probably due to high loading of organoclay that take a large amount of volume replacing shrinking polymer [1], [6].

C. Filler Comparison on PA 66 Shrinkage

The addition of organoclay gives lower shrinkage reduction and higher shrinkage percentage for in-flow and cross-flow direction compare to GF. However, the addition of GF gives anisotropic shrinkage, while the organoclay gives isotropic shrinkage. This anisotropic shrinkage will lead to warpage. The warpage can cause the sample has no flatten surface [1], 4].

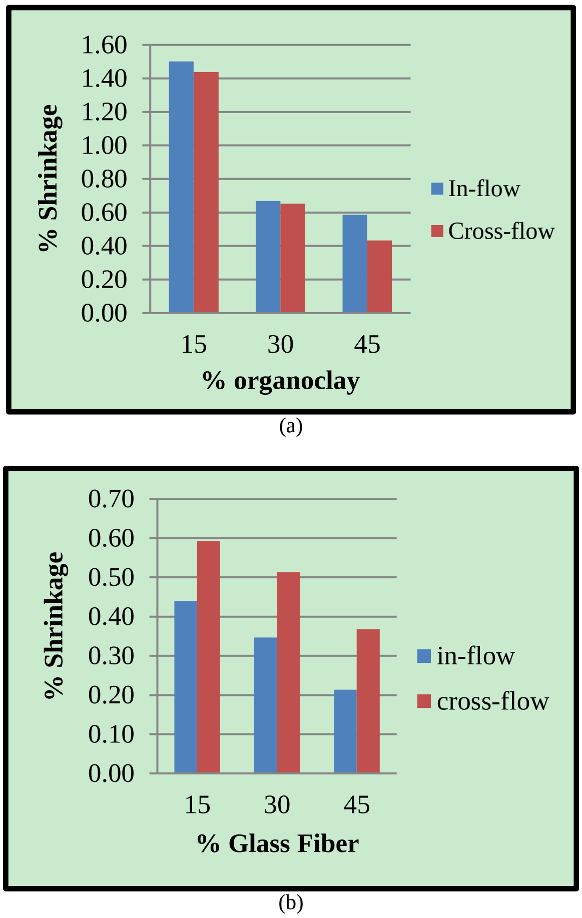

Fig. 4. Comparison of in-flow and cross-flow shrinkage of PA66/clay (a), and PA66/GF fiber (b).

The anisotropic shrinkage of PA66/GF can be seen in Fig. 4 b, where the differential shrinkage between in-flow and cross-flow direction is high. Whereas Fig. 4 a showed isotropic shrinkage of PA66/clay where there is reduction in differential shrinkage. The reduction in differential shrinkage may be related with the aspect ratio of the fillers. Organoclay, as flake filler, has lower aspect ratio (length to width) compare to glass fiber which caused shrinkage reduction proportionally in all direction. Therefore, PA66/clay can be alternative solution for warpage problem in PA66/GF. However, combination both filler probably more advantage for resulting lower shrinkage and warpage in the final molded part [1], [4], [7], [8].

IV. Conclusions

Polyamide 66 (PA66)/ glass fiber (GF) composites were prepared by an injection molding. The effect of GF content and molding condition, namely melt temperature, cooling time and cooling temperature on shrinkage percentage were analyzed. The predicted optimum condition to achieve the minimum cross-flow shrinkage of PA66/GF composite were 45% GF, 285 °C for melting temperature, 35 second of cooling time and 60 °C for cooling temperature. The verification sample of PA66/GF optimum condition resulting 0.41% cross-flow shrinkage with 78% reduction compare to PA66. This reduction might be caused by the low CTE (coefficients of thermal expansion) of GF that cause composite to shrink less. However, this reduction also cause anisotropic shrinkage. The addition of organoclay to PA66 matrix can result isotropic shrinkage although the shrinkage reduction smaller than GF.

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