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INVESTIGATIONS ON INJECTION MOLDING CONDITIONS FOR MAKING FLAX FIBER-HIGH DENSITY POLYETHYLENE BIOCOMPOSITES

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Abstract

Oilseed flax fiber, a very large renewable resource in Western Canada, has been recognized as a potential replacement for glass fiber to reinforce plastic. Develop flax fiber reinforced composites-innovative lightweight, strong and environmental friendly materials – to be used in the plastics industry will not only contribute to the economic growth, but also benefit to the environment. In authors' previous work, flax fiber-HDPE biocomposites had been developed via injection molding which is an important processing method with the characteristic rapid production rate. In this paper, the injection processing conditions including injection temperature, pressure, and cooling time were discussed for flax fiber-HDPE biocomposites. It was found that injection temperature had a significant influence on mechanical properties of the biocomposites compared with injection pressure. The processing temperature of lower than 195°C was recommended for making biocomposites. To estimate the minimum cooling time during injection molding, the thermal properties of biocomposites were studied. How the minimum cooling time related with mold, injection, and ejection temperatures were discussed.

Keywords: Flax fiber, fiiber-reinforced biocomposites, high density polyethylene, injection temperature, minimum cooling time

INTRODUCTION

Flax is an annual crop used both for fiber and its edible seed. With the huge amount of oilseed flax production in Western Canadian each year, more flax straw is left in the fields without suitable utilization. Because flax stalk requires a much longer time to degrade than many other agricultural residues, it is burned by farmers and brings the environment problem. Therefore, development of products from flax fiber will benefit Canadian and world seed flax growers and achieve the sustainable development for Canada Agriculture.

Development of natural fiber-reinforced biocomposites has been attracted more attentions in recently years. Flax fiber has higher mechanical strength than polyethylene and most of other plastics. Therefore, adding flax fiber into polyethylene increased the strength of biocomposite, making the composite materials more competitive to market. Besides, flax fiber has low cost, reduced energy consumption and it is a renewable resource. Our research objective is to develop flax fiber-reinforced high density polyethylene biocomposites (HDPE) via injection molding. HDPE is easily molded in mass-production quantities by injection molding. HDPE has very low glass transition temperature (T_g= - 110°C) associated with a good retention of mechanical properties including flexibility and impact resistance at low temperatures (Charrier 1991). It is suitable to be matrix in composites especially in country as cold in winter as Canada. This research will promote and accelerate the application development of using HDPE as matrix in fiber reinforced composites.

Being as an important plastic processing method, injection molding is characterized by rapid production rates. Injection molding operational conditions and the amount of fiber could affect the properties of composite materials (Saint-Martina et al. 2003; Mohanty et al. 2003). However, how injection molding parameters influence the properties of flax fiber-plastic biocomposites had not been revealed. To understand the processing and optimize injected biocomposite properties, this research studied the injection molding process and optimized the operation for manufacturing flax fiber-high density polyethylene biocomposites.

During injection molding, the material may be released from the mold as soon as its outer layer is sufficiently rigid. Cost savings could be realized if the minimum cooling time is controlled and the total molding cycle time is shortened. This paper investigates the minimum cooling time of flax fiber-HDPE biocomposites after knowing the thermal diffusivity of biocomposites. Minimum cooling time is related with the thickness of the molding part, temperature difference between the mold and biocomposite, and mold release temperature which is known as ejection temperature.

MATERIALS AND METHODS

Materials

Oilseed flax fiber grown in Saskatchewan was provided by Biofiber Industries (Canora, SK, Canada). HDPE (injection moulding grade) with melt flow index of 12.5 g/10 min was purchased from Nova Chemicals (Mooretown, ON, Canada). Technical grade chemicals including acrylic acid (Lancaster Synthesis, Inc., Pelham, NH) and sodium hydroxide (EM Industries, Inc., Gibbstown, NJ) were used for fiber surface modifications.

Fiber Surface Modification

Chemical modifications are conducted on flax fiber to improve the fiber-matrix bonding because the hydrophilic nature of natural fibers often causes low interfacial properties between fiber and hydrophobic plastic matrix. Flax fiber was washed with 2% detergent solution and immersed in sodium hydroxide solution for 0.5 h. It was then soaked in 5% acrylic acid solution at 50ºC for 1 h. The fiber was then washed thoroughly with distilled water and dried in an air oven at 70°C for 24 h.

Biocomposite Preparation and Injection molding conditions

The dried fiber was ground through 2 mm screen and mixed with HDPE at 30% fiber content by mass of composite. The mixture of fiber and HDPE was fed into the twin-screw extruder (Ramsey, NJ, USA) at screw speed of 150 rpm and barrel to die temperatures of 90, 120, 130, 140, 160ºC from first to fifth zone as shown in Figure 1. The extrudates after extrusion were pelletized using a grinding mill (Retsch GmbH 5657 HAAN, West Germany) and then processed through the injection moulding machine (Battenfeld, Meinerzhagen, Germany, Figure 2) to biocomposite samples.

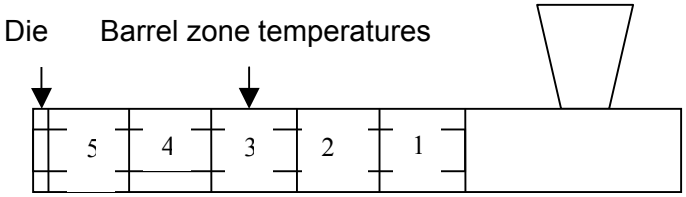

Figure 1. Cross-sectional view of the extruder with five separate heating zones.

Figure 2. Injection molding machine (Battenfeld Maschinen, Germany) at Northern Alberta Institute Technology (NAIT).

During injection moulding, two injection temperatures and two injection pressures were selected for all biocomposites with three different fiber mass content (10, 20, and 30%). Two set of Injection temperatures are D=166ºC, C=182ºC, B=188ºC and A=188ºC and D=177ºC, C=190ºC,

B=200ºC and A=200ºC (refer to Figure 3). The two levels of the injection pressure were 4.8 MPa (700 psi) and 6.9 MPa (1000 psi).

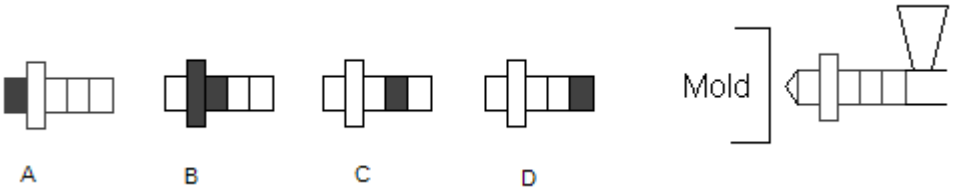

Figure 3. Temperatures setting at injection moulding machine (Battenfeld, Maschinen, Germany).

Tensile Property Testing

Tensile test was conducted using ASTM standard test method D638 (ASTM 1997a). The tests were conducted using an Instron model 1122 testing machine (Instron, Canton, MA, USA) at a crosshead speed of 5 mm/min. Each test was repeated five times. The force at break was recorded and the tensile strength was calculated.

Water absorption of biocomposites

Water absorption test was conducted according to ASTM standard test method D570 (ASTM 1998). The 24 h water immersion method was chosen. Three specimens were tested and the results were presented as average of tested specimens.

Thermal properties and cooling time of biocomposites

The thermal diffusivity (α, m^2/s) of a material is a function of its thermal conductivity (*k,* W/m°C), specific heat capacity (C_p, kJ/kg°C), and density (ρ,kg/m^3) and can be calculated from equation 1:

$$\alpha = \frac{k}{\rho C_p} \qquad (1)$$

The thermal diffusivity of flax fiber-HDPE biocomposites (before sending to injection molding) were determined by measuring the thermal conductivity (k), specific heat capacity (C_p), and density (ρ). The thermal conductivity was measured by Line-source method (Lobo and Cohen 1995), the specific heat capacity was measured by using a TG-DSC111 thermal analysis system (Setaram Scientific & Industrial Equipment, Caluire, France). The details of methods were given in previous paper (Li et al. 2008).

The minimum cooling time (t_c) in injection molding is estimated from equation 2.

$$t_c = \frac{(2h)^2}{\alpha \pi^2} \ln\left[\frac{4}{\pi}\left(\frac{T_m - T_w}{T_e - T_w}\right)\right] \qquad (2)$$

Where:

h = half of the thickness of material inside the mold (m),

T_w = mold wall temperature (°C),

T_m = melted material temperature (°C), and

T_e = ejection temperature (°C).

It is assumed that the material's original temperature (T_m) is the injection temperature set on the injection machine. The thickness ($2h$) of the material being molded is 0.004 m; then the minimum cooling time at a certain mold temperature or ejection temperature (mold open temperature) can be predicted.

RESULTS AND DISCUSSION

Effect of Processing Conditions on Tensile Properties of Biocomposites

The biocomposites (10, 20, and 30% wt.fiber) were manufactured at two injection temperatures and two injection pressures. Their tensile strengths were then measured. The highest tensile strength was found in biocomposite with 30% fiber and processed at low injection temperature (166-188ºC) and low injection pressure (4.8 MPa). But low injection pressure and high injection pressure had no significant difference on their influence to strengths. SPSS (SPSS Inc., Chicago, IL) statistical analysis showed that the tensile strength was significantly dependent on fiber content and injection temperature. Among the three factors, the factor with the most impact was fiber content, followed by injection temperature; the factor with the least impact was injection pressure.

If the injection temperature uses the Zone A temperature (refer to Figure 3), the relationship between biocomposite tensile strength (σ_t, MPa) and processing variables (fiber content F, % and injection temperature-zone A, T, ºC) can be developed. The regression model is shown in equation 3 with a R^2 of 0.8748. Then the surface curve according to equation 3 is plotted in Figure 4 using Matlab (The MathWorks, Inc. Natick, MA).

$$\sigma_t = 28.9516 + 0.2531\ F + 0.0049\ F^2 - 0.0002\ T^2 - 0.0017\ FT \qquad (3)$$

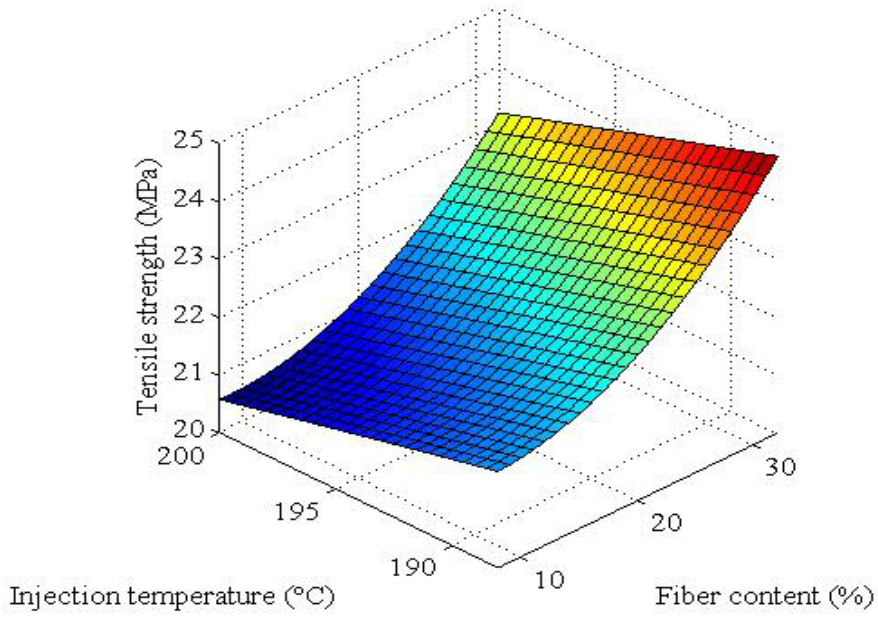

Figure 4. Estimated tensile strength of flax fiber-HDPE biocomposites as affected by fiber content and injection temperature.

It was found that low injection temperature (188ºC) resulted in higher tensile strength than high injection temperature (200ºC) regardless of fiber content (10 to 30%). It was also emphasized by Fung and co-researchers (2003) that high injection moulding temperature (210ºC) caused the

thermal depolymerization of hemicellulose and the glycosidic linkages of cellulose. Therefore, the high temperature will cause the fiber loss and reduce the reinforcement effect.

Effect of Processing Conditions on Water Absorption of Biocomposites

When fiber content increased, the water absorption of biocomposites increased as well. Since higher fiber content is desired in biocomposites to achieve good mechanical properties, decreasing water absorption by controlling injection temperature and pressure are important.

The results showed that when the fiber content in biocomposite was 30% and the injection temperature was low (D=166ºC, C=182ºC, B=188ºC and A=188ºC), the average water absorption of biocomposite was 0.22%; but at the same fiber content, when injection temperature increases (D=177ºC, C=190ºC, B=200ºC and A=200ºC), the average biocomposite water absorption increased to 0.54%. This indicates that it is very important to control the injection temperature to decrease the biocomposite water absorption when the fiber content is higher.

SPSS statistical analysis also showed that fiber content and injection temperature significantly influenced the biocomposite water absorption, while injection pressure did not. The regression model of biocomposite water absorption with fiber content (*F*, %) and injection temperature (*T*, ºC) was developed as equation 4 with a high R^2 (0.9641).

$$M\% = 1.0034 - 0.2143\,F - 5.5975\times10^{-5}\,F^2 - 2.9689\times10^{-5}\,T^2 + 0.0012\,FT \qquad (4)$$

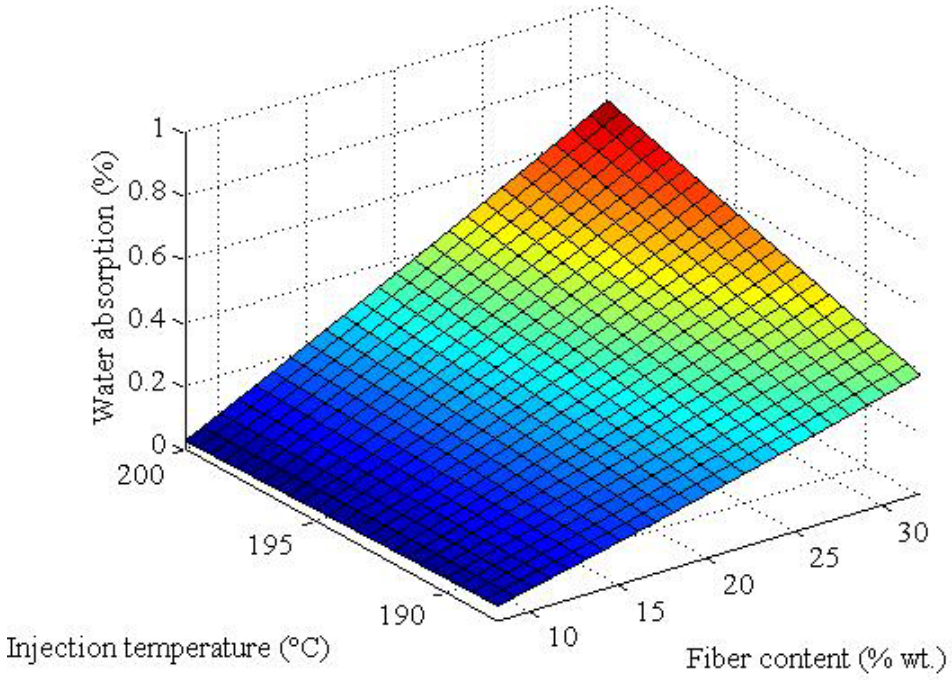

Figure 5. Estimated water absorption of flax fiber-HDPE biocomposites as affected by fiber mass content and injection temperature (Zone A temperature in Figure 3).

The surface curve according to equation 4 is plotted in Figure 5. It showed that when the biocomposite included less than 20% flax fiber, changing the injection temperature did not result in a big difference on the biocomposite water absorption. But when the fiber content increased to 20-30%, the biocomposite water absorption was obviously influenced by injection temperature. There was an increase of water absorption when injection temperature was higher

than 195°C. This was because at higher temperature, fiber degradation occurred, thus more pores were formed between the fiber and matrix interface, which gave paths for water to enter the biocomposites. Therefore, injection temperature (zone A- at where the material is injected into the injection mold) is recommended below 195°C for manufacturing flax fiber-HDPE biocomposites.

Thermal Diffusivity and Minimum Cooling Time of Biocomposites

The thermal diffusivity of biocomposites was measured before the biocomposites sending to injection molding. It was found that fiber content had a significant influence on the thermal diffusivity of biocomposites. The thermal diffusivity of the biocomposites decreased with fiber content. The influence of temperature on thermal diffusivity was not as obvious as fiber content. The linear regression equation fitted to the measured thermal diffusivity (α, m^2/s) of biocomposite was developed by SPSS stepwise method as given in equation 5:

$$\alpha = 2.06 \times 10^{-7} - 1.32 \times 10^{-7} F - 1.38 \times 10^{-7} T \quad (5)$$

where F = fiber content (0 to 30% wt.),
T = temperature (170 to 200°C), and $R^2 = 0.9799$.

After knowing the thermal diffusivity, the minimum cooling time during injection molding can be estimated according to equation 2. In injection molding, cooling time is the time to cool the molded material to ejection temperature, which typically occupies more than one third of the whole molding cycle (Xu and Kazmer 1999). Therefore, estimating and optimizing cooling time play an important role in injection operations to reduce the cycle time and increase production rates. The minimum cooling time is the duration at which the molded material is being cooled from injection temperature to ejection temperature; thus, higher injection temperature causes longer cooling time and higher ejection temperature leads to shorter cooling time.

It was concluded in previous result that injection temperature of Zone A should be lower than 195°C. If the injection temperature of Zone A is set at 190°C, molded biocomposite thickness is 4 mm, ejection temperature is supposed 100°C, then the minimum cooling time of biocomposites changing with mold temperature is showed as Figure 6.

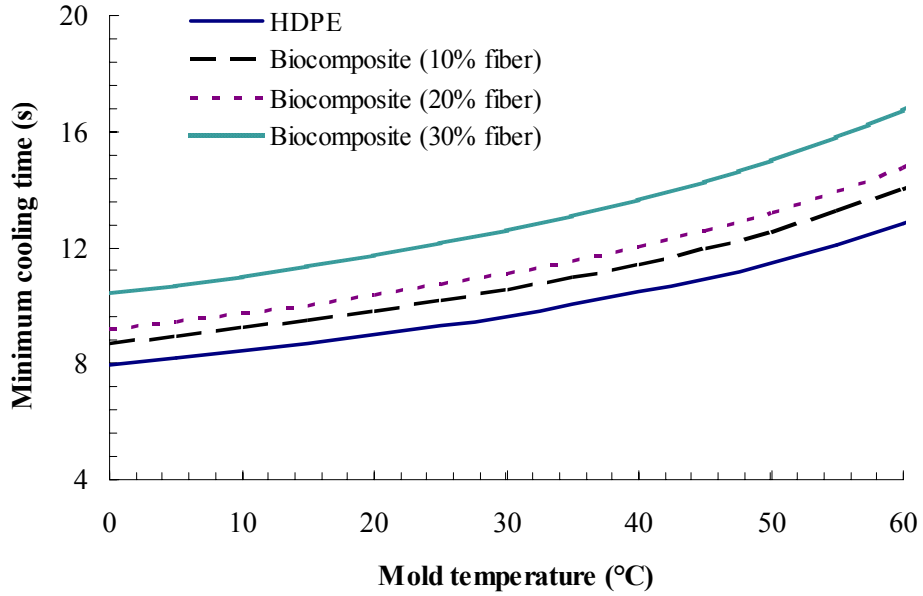

Figure 6. Minimum cooling time versus mold temperature when injection temperature was 190°C.

The cooling time increased with mold temperature. The mold temperature for HDPE ranged from 4 to 38°C (Rosato et al. 2000) or 40°C (Prystay and Garcia-Rejon 1999). Lower mold temperature is usually set to remove the heat quickly from the material and thus achieve short cycles (Rosato et al. 2000). But low mold temperature may also cause problems such as short shots, surface wrinkles or too large dimensions (Creese 1999). So the mold temperature of biocomposites cannot be set too low in order to achieve good product quality. Figure 6 shows that the cooling time of biocomposites increased about 2-3 s when mold temperature increased from 10 to 40°C. Therefore, mold temperature of 40°C is set up for the subsequently analysis. It is also found that it needs longer time to be cooled for biocomposites including higher fiber content.

The ejection temperature is the temperature when the material is cool enough for the mold to open (Isayev 1987). The ejection temperature point should be in a state that the material inside the mold cannot flow, but it is still soft enough to be ejected. There are several ways to determine the ejection temperature. Spring and Williams (1999) investigated three of the most popular methods to determine the ejection temperature of thermoplastic material. They found these methods resulted in very different ejection temperature. One method used the vicat softening temperature test according to ASTM D1525 (ASTM 1997b). One method used the heat deflection temperature as ejection temperature. With this method, Rosato and co-workers (2000) reported that HDPE usually has an ejection temperature of 60 to 90°C. The third method determined ejection temperature by DSC cooling trace: for a semi-crystallization material, it is the temperature after crystallization peak (Spring and Williams 1999). Ejection temperature was around 120°C for HDPE (Rana et al. 1999) since the crystallization temperature of HDPE was near 130°C (Wen et al. 1999).

If the injection temperature of Zone A is 190°C, molded biocomposite thickness is 4 mm, mold temperature is set at 40°C, then the minimum cooling time of biocomposites changing with the ejection temperature is showed as Figure 7.

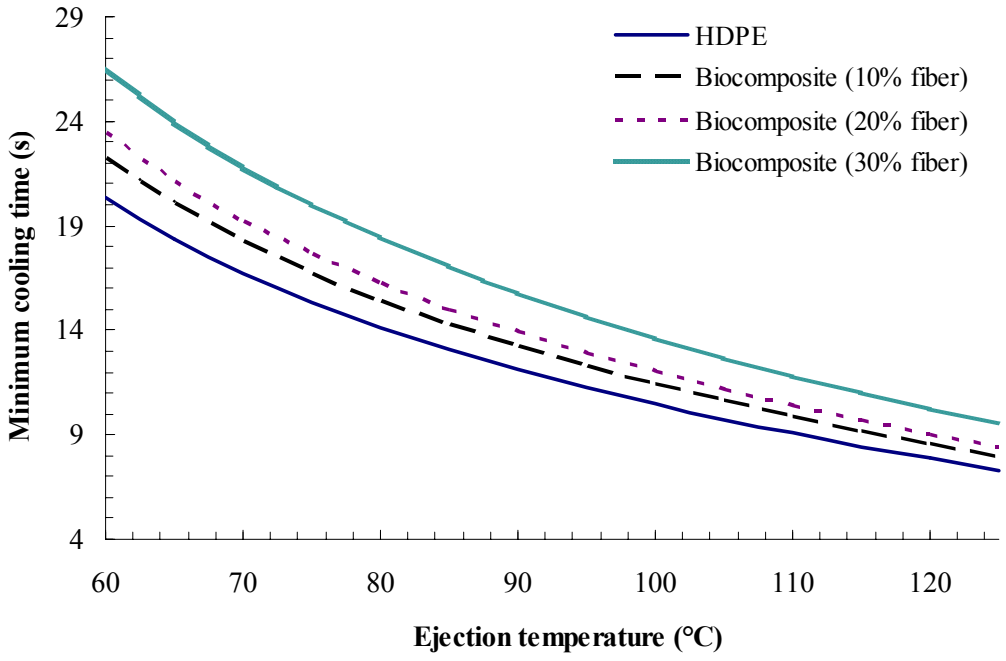

Figure 7. Minimum cooling time versus ejection temperature when injection temperature was set at 190°C.

Figure 7 shows that the cooling time decreases by about 10 s when ejection temperature increases from 60 to 100°C. Such a deviation will play an important role in determining the ejection temperature in industrial applications since a 10 s error in cycle time represents a 20% loss in productivity and significantly greater loss in profitability (Xu and Kazmer 1999). Overall, higher ejection temperature caused shorter cooling time (Morales et al. 2001). Thus, the relatively higher ejection temperature is preferred in injection molding.

CONCLUSIONS

Oilseed flax fiber could be used as reinforcement for composites because it is readily available, environmentally friendly and possesses good mechanical properties. Using injection molding technique, flax fiber and HDPE could be manufactured to biocomposites. The factors including flax fiber content, injection temperature, and injection pressure were investigated on how they affect the tensile properties and water resistance of biocomposite. It was found that compared with injection temperature and pressure, fiber content was the most significant impact factor of influencing the mechanical properties of biocomposites. Injection temperature of lower than 195°C was recommended to achieve better composite tensile strength and water resistance. The minimum cooling time during injection molding was related with biocomposite thermal diffusivity, molded product dimensions, mold temperature, and injection and ejection temperatures. The biocomposite containing more fiber content required longer cooling time. The minimum cooling time increased with the thickness of molded material, mold temperature, and injection temperature. The cooling time decreased with the ejection temperature. The suitable mold and ejection temperature for biocomposites were around 40 and 100°C.

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