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Review Article

Problems and Constraints Involved in the Injection Moulding Process of Biocomposites: A Review

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Abstract: Biocomposite materials have revolutionized the engineering field, predominantly in manufacturing applications, where these materials were widely used as an alternative to the metal-based structural body or component. Since conventional materials have caused various pollution problems to the ecosystem, innovative efforts have been taken to develop the based product on natural materials composites but presently are not fully explored yet. Most issues raised are due to the lack of expertise in addressing some of the problems and constraints related to preparing feedstock materials involved in the injection moulding technique. The injectability process of the biocomposite material significantly depends on the early stage of feedstock preparation. The preparation steps should be considering the detailed characterization of the composite material, the effectiveness of the mixing method, the identification of the feedstock's flowability properties, and the appropriate processing parameters used. This review manuscript has a significant contribution to researchers interested in furthering the application of biocomposite-based materials, particularly in plastics injection moulding technology for manufacturing and processing engineering.

Keywords: Natural fibre; polymer materials; biocomposite; injection moulding; processing parameter; engineering components

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1. Introduction

The development of metal and synthetic based materials in recent engineering tools and compartment have caused environmental pollution problems to the natural ecosystem (Yu *et al.*, 2013). These non-degradable materials with uncontrolled manufacturing processes have led to excessive carbon dioxide (CO_2) emissions. Furthermore, the weakness of the heavyweight tractors and vehicle efficiency in managing the consumption of fuels was also contributing to an increase in the pollution rate. A study reported that a 25% reduction in vehicle weight could save 250 million barrels of fuel a year, indirectly reducing the CO_2 gas emission rate of 220 billion pounds (lbs.) (Pandey *et al.*, 2010; Ahmad *et al.*, 2015). This situation has pushed most engineering industries to implement renewable materials as a component (e.g., interior and exterior), which are more lightweight, environmentally friendly, and saving the cost-effectively (Pickering *et al.*, 2016).

To date, the production of biocomposites has prompted manufacturing engineers to extend the potential of these renewable materials (Zini & Scandola, 2011). Biocomposite has been applied as an alternative to previously developed metal and synthetic composite materials in vehicle structural bodies or components. The use of natural reinforcement materials such as plant fibres (e.g., flax, jute, kenaf, sisal, sugar palm) in polymer composites has been evaluated as a practical approach in producing high-quality composite materials (Mohd Radzuan *et al.*, 2019). These low-density plant fibres have high strength properties and are readily available at low cost, even non-toxic extract resources. The combination of both materials is feasible to produce biocomposite with unique mechanical and durability strength properties which close comparable to any individual component (Da Silva *et al.*, 2016). Natural-based composites' purpose is to reduce the reliance on petroleum and metal products since they were listed as the shortage resources (Salazar *et al.*, 2011; Faruk *et al.*, 2012; Al-Oqla & Sapuan, 2014).

Generally, the performance of the mechanical and physical strengths of most biocomposite is slightly different. It depends on the type of polymer matrix and natural fibres used and the form of reinforcement bonded between them by using specific processing techniques (Ku *et al.*, 2011). According to Väisänen *et al.* (2017) and Noor Azammi *et al.* (2018), considerations such as fiber-matrix bonding compatibility and fiber distribution uniformity are essential for improving the tensile strength of reinforced composites. The high loading of fibres with the appropriate aspect ratio was also concerned with Young's modulus properties' highest achievement.

Meanwhile, the appropriate selection of processing techniques is significantly relying on the type of polymer matrix used. The suitable manufacturing process will transform the composite material to the desired shape without affecting the finished product. Among the manufacturing process, such as compression moulding and hot pressing, plastic injection moulding is one of the central processing techniques used in the mass production of most biocomposite materials (Tholibon *et al.*, 2019). A complex geometrical engineering component with precise dimension and high surface finished can be produced in this technique. Furthermore, a short processing time cycle can be achieved as this method can produce products in bulk quantities (Ho *et al.*, 2012).

Practically, the development of natural materials-based composites has not been fully discovered. It is due to the lack of expertise in addressing some of the problems and constraints related to processing, particularly involving injection moulding techniques. Thus, this paper summarizes the previous studies on the injection moulding process of natural fibre reinforced polymer composites materials. It focuses primarily on the constraints and feasibility of injection and several other processing factors affecting the biocomposite's performance in producing more sustainable engineering components comparable to other synthetic-based composites materials.

2. Fibre-matrix Reinforcement

Interfacial bonding plays a vital role in forming a composite where both fibre and matrix phases are interacting through adhesion called reinforcement. Its mechanism performance critically depends on the effectiveness of fiber-matrix load transfer in light of the suitability type of reinforcement formation between them (Shubhra *et al.*, 2013). Besides, composites can be classified according to the type of reinforcement in the form of short fibre (filler and particle), long fibre (continuous and discontinuous), and mat (woven and non-woven). The differential in size and geometry of each type of fibre will determine the appropriate processing using the efficient moulding technique. The type of reinforcement formed by this technique will produce different reinforcing composites in terms of their mechanical and physical strength. Therefore, it is not practical to do a comparative study among the reinforced composite with different processing techniques. (Koronis *et al.*, 2013).

The short fibre promising benefits and potential reinforcement in biocomposite development. Typically, this short fibre can be obtained with two different reinforcing forms extracted from the skin and core found in most plants. According to the aspect ratio measurements, both types would be identified in various shapes and sizes (Azaman *et al.*, 2015). Kwon *et al.* (2014) have studied how the aspect ratio of kenaf filler and corn husk form may influence molded polymer composites' mechanical strength properties. Fillers with lengths (211.6–528.2 µm) and diameters (48.6–39.0 µm) significantly have high aspect ratio values to play as a controlling factor in enhancing the mechanical strength characteristics the composite. It contrasts with a husk that has relatively low aspect ratios due to the smaller size and geometry. However, theoretically claims that composite mechanical properties could increase with the increase of filler loading. The increasing loading size of more minor fillers (<500 µm) will provide more and greater surface area, and the latter that stress transferred will occur more efficiently via excellent interfacial bonding and may result in better reinforcement (Ismail, 2004).

Most of the short fibre reinforced composites are adapted to be formed through the plastic injection moulding process (Chaitanya *et al.*, 2017). There is a lack of research progress on developing this type of reinforced composites due to the presumption about the poor characteristic of short fibre reinforcement with non-existent bonding mechanisms that might occur in polymer composites. However, some researchers reported that short fibre might offer the high stiffness strength characteristic to the reinforced composites even have similarities and comparable to other synthetic fibre composites (Azaman *et al.*, 2015). Table 1 summarizes the research done on the processing of short fibre reinforced composites through injection moulding. The research records show that the short fiber study can be utilized as reinforcement in polymer composite.

Table 1. Previous works on the utilization of short fibre reinforcement for biocomposite by using an injection moulding process.

Size of fibre	Fibre	Matrix	Remarks	Author(s)
5 mm	Oil Palm EFB	Polyethylene (PE)	• The highest mechanical strength properties of EFB/PE composites were obtained at optimal processing parameters at holding pressure of 80 bars and injection temperature of 150 °C	Megat-Yusoff *et al.* (2011)
6 mm	Ramie	Polypropylene (PP)	• The mechanical strength and modulus properties of ramie/PP composite increase with increasing fibre load • At 30 wt.% fibre content, the tensile and bending strength recorded at 67 MPa and 80 MPa, respectively	Feng *et al.* (2011)
-	Kenaf & Sisal	Polypropylene (PP)	• The mechanical strength of kenaf/PP composites produced higher Young's modulus strength compared to sisal/PP composites • The tensile strength of kenaf/PP composites showed an increment in the amount of reinforced material compared to sisal/PP composites	Phiri *et al.* (2014)
5 mm	Kenaf	Polypropylene (PP)	• Improvement of the flexural strength of kenaf/PP composite 30 wt.% fibre loading • The stiffness strength of kenaf/PP increase with increasing tensile	Chaitanya & Singh (2016)

			strength and flexural modulus compared to pure PP	
5 mm (bast filler) 106 µm (core filler)	Kenaf	Polylactic-acid (PLA)	• The feedstock mixture of core filled composite is more homogeneous and compatible compared to bast filled composite • The surface modification process successfully reduces the hydrophilic properties of the core and thus enhances the fibre-matrix reinforcement	Kawahara *et al.* (2017)

The selection of matrix materials for biocomposites is limited depending on the natural fibers' thermal decomposition temperature. The high processing temperatures will affect a whole biocomposite processing since most of the natural fibres have low thermal resistance and begin to degrade beyond 200°C (Summerscales *et al.*, 2010). Hence, the low-melting-point of the thermoplastic matrix that is suitably used in the development of biocomposites such as polypropylene (PP), polyethylene (PE), polystyrene (PS), and polyvinyl chloride have many advantages. Moreover, thermoplastic polymers would definitely reduce the processing costs, improve design flexibility, and are easily molded into complex components (Yan *et al.*, 2014).

3. Injection Moulding Process for Biocomposite Materials

Plastic injection moulding refers to the compaction or injection of melting mixture materials into the desired mould shape subsequently, followed by specific processing steps shown in Figure 1 (Ho *et al.*, 2012). Problems and constraints often occur during the injection process due to the different physical and mechanical characteristics of biocomposites compared to synthetic composites. Some of the challenges encountered during the injection moulding process are closely related to the feedstock materials' preparation process (Sun *et al.*, 2010; Rahman *et al.*, 2015). The preparation should consider each material component's sensitivity characteristics towards moisture resistance and temperature stability, which are influenced by the size and amount of fibre used (Ku *et al.*, 2011). Furthermore, the increased use of fibre loading leads to agglomeration and voids in the moulded composite. It happened when the fillers were not completely wet and covered by the polymer matrix, resulting in poor interfacial bonding of fibre-matrix (Chaitanya *et al*., 2017; Mehdikhani *et al*., 2019).

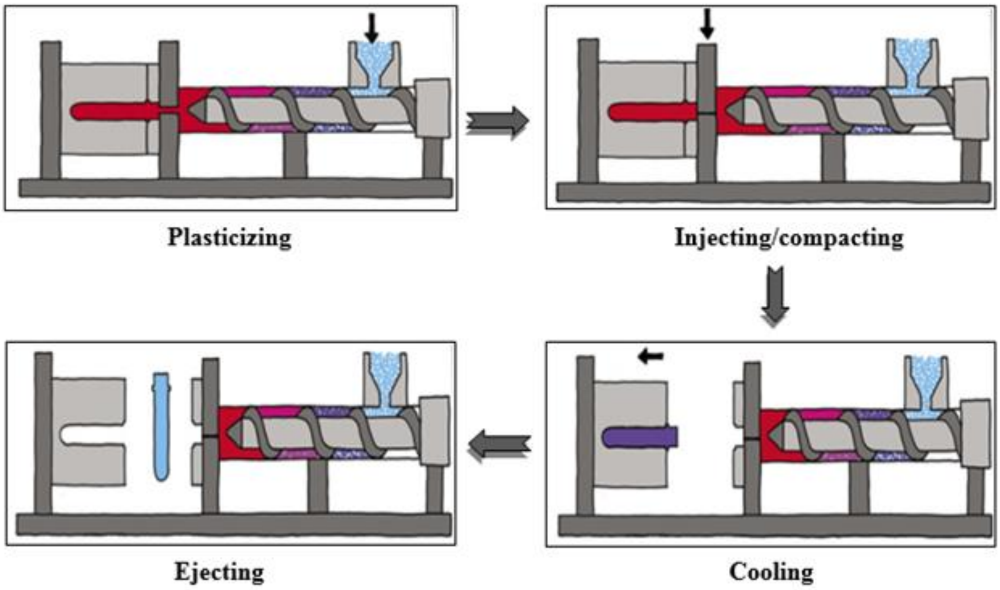

Figure 1. The steps of the plastic injection moulding process (Ho *et al.*, 2012).

3.1. Compounding Process

Compounding becomes an essential mixing step in producing homogeneous feedstock materials before the injection process could proceed. Nevertheless, no clear parameters have been identified in determining the optimal level and homogeneity regarding the mixing processes. Different types of a mixture of biocomposite materials will achieve different degrees of homogeneity. It depends on several factors, including the mixing machine used, the mixing technique, the type, size, or geometry of the fibres, and the mixture's composition ratio (Lu *et al.*, 2004). For biocomposites, compounding is generally divided into one-step (continuous mixture, such as extruder) and two-step (batch mixture, such as two-roll mill and rotor mixer) processes illustrated in Figure 2. Both mixing methods have been adopted in most biocomposites manufacturing industries involving plastic injection moulding techniques (Clemons, 2002).

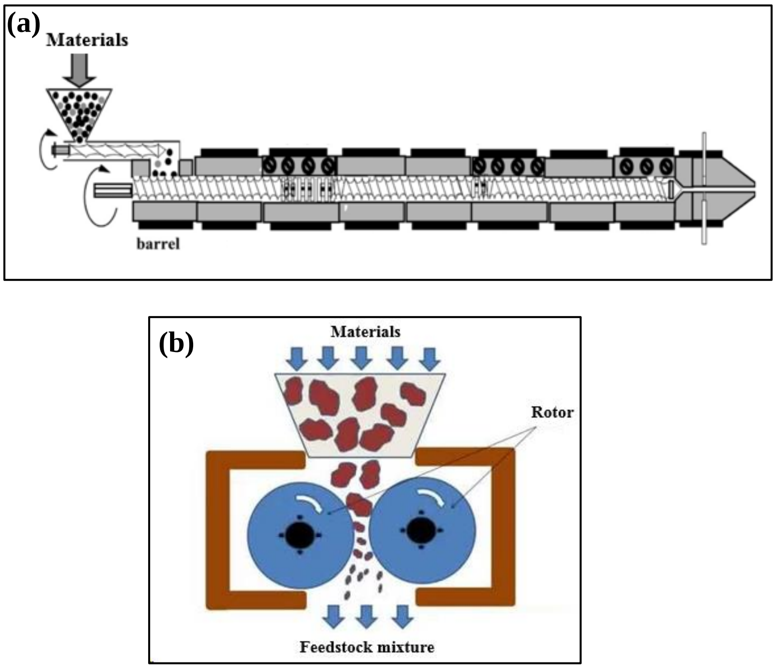

Figure 2. The compounding: (a) extruder for continuous mixture; and (b) rotor mixer for the batch mixture (Barbas *et al.*, 2013).

The study reported that fibres extracted from the feedstock with two-roll mill mixers have the highest aspect ratio and lowest standard deviation characteristics and even are uniformly dispersed compared to the fibers' geometry extracted from the extrusion mixture (Sun *et al.*, 2010). Due to a high shear mechanism present by the friction force between the feedstock and rotating rotors, it optimizes the mixture performance by improving the degree of compatibility and surface wetting (Rozman *et al.*, 2011; El-Shekeil *et al.*, 2013).

Besides, parameters used during the mixing process also affected the homogeneity of the mixture. The setting parameters involved in the mixing process, such as temperature, time, and mixing speed, are closely related to the type of reinforcement material and the matrix used. A previous study determined that the mixing temperature in a range between 180°C up to 200°C was the significant value to be considered an optimum parameter (El-Shekeil *et al.*, 2011). From the observations, the mixture of fibres and polymer matrix will not mix well if the temperature used is below the melting point of the matrix. Most natural fibres begin to decompose when the blending temperature exceeds 200°C (Feldmann, 2016).

Meanwhile, the time and mixing speed parameters were selected at 11 min to 15 min and 30 rpm to 50 rpm, respectively. A short time with slow mixing speed will strengthen the wetting mechanism and disperse the composite mixture's fibres. In contrast, biocomposite feedstock that is mixed up at high speeds for an extended period, in turn, will increase the degradation level of the mixture and tend to break down the fibre structure and polymer chains (Väisänen *et al.*, 2017).

3.2. Rheological Behaviour

The flow characteristic of biocomposite materials is an important matter to be determined before the injection process can be adequately performed without any problems and constraints. This flowability can be analyzed based on the melt flow behaviour of the composite materials using a rheological test such as capillary or rotational rheometers (Ogah, 2017). Typically, flow behavior analysis is presented by the relationship of the viscosity, η against the shear rate, $\dot{\gamma}$, which is influenced by the temperature, and load cell variations. In the meantime, the relation between the shear stress variables, τ against the shear rate, $\dot{\gamma}$ will be calculated to determine the flow index of composite materials according to the resulting flow characteristics shown in Figure 3.

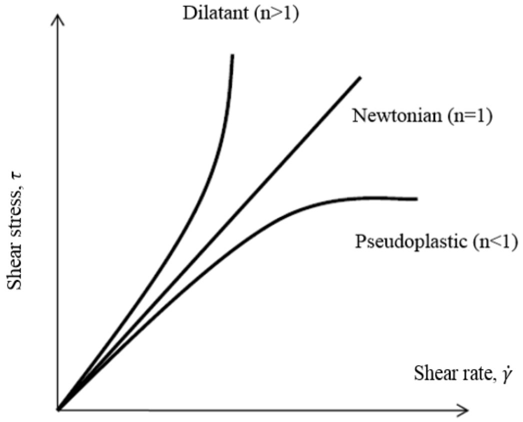

Figure 3. The flow characteristics of a composite material (Ivanov *et al.*, 2001).

Figure 3 illustrates the types of flow generated during the rheological behaviour test, which was classified into non-Newtonian (dilatant and pseudoplastic) and Newtonian flow characteristics. Generally, a polymer-based composite material's flowability is close to the pseudoplastic flow behaviour, which has low viscosity with a shear rate range of 102 s-1 to 106 s-1 (Nanda & Tripathy, 2012). Also, melts with low viscosity are among the biocomposite's appropriate flow properties in meeting the plastic injection moulding process criteria. Therefore, the melt flow index, n for feedstock can be determined through the relation between the viscosity of melts against shear rate changes defined by equation 1:

$$\eta = k\dot{\gamma}^{n-1} \tag{1}$$

The rheological behaviour of biocomposites has been evaluated regarding adding short fibres to polymer composites' injectability process. A study by Lewandowski *et al.* (2016) has shown that the high percentage of the wood filler used (i.e., 30 wt.% to 50 wt.%) would result in a higher viscosity of feedstock melts. However, to obtain results of melts that meet the pseudoplastic flow characteristics, they have increased the shear rate by increasing the load cell over 1000 s^{-1}. Finally, the value of the melt flow index, n, was significantly reduced (n<1) in parallel with the increase of wood filler load between the shear rate range, $\gamma^{\cdot}$ of 10 s-1 to 1000 s^{-1}.

Also, the sensitivity level of melts towards environmental influences such as temperature changes during the injection moulding process can be determined by the activation energy value, E (kJ/mol). This value of the activation energy indicates the degree of dependence of the feedstock material's melting point on temperature change. The lower activation energy value presents a more negligible sensitivity effect on melts from any sudden temperature change during the processing (Sewda & Maiti, 2012; Nanda & Tripathy, 2012). The activation energy value can be obtained using equation 2, where η_0 is the reference viscosity, R is the gas constant (8.314 J/mol K), T is the temperature (Kelvin), and E is the flow activation energy.

$$\eta = \eta_0 \, exp\left(\frac{E}{RT}\right) \tag{2}$$

3.3. Injection Processing Parameters

Processing parameters are one of the most critical factors affecting the smoothness of the injection process of biocomposite. Some of the injection molding techniques parameters that should be considered include injection temperature, injection pressure, injection time, holding pressure holding time, cooling time, and screw rotation speed. In default, these processing parameters will affect a reinforced composite's mechanical and physical strength characteristics (Andrew *et al.*, 2019).

Feldmann (2016) studied the influence of injection temperature on the mechanical strength of cellulose fibres reinforced polypropylene composite. The temperature has been set, ranging from 160°C up to 269°C. Observation reported that the tensile strength and modulus properties of reinforced composite resulting in different results depending on the injection temperature applied. The injection temperature applied up to 250°C is considered too high for most natural composite materials since fibre's limited thermal stability is below 200°C. In contrast, problems and other constraints (e.g., short shot, jetting, sink marks) will arise if polymer composites' processing is carried out at a lower value of temperatures and shear forces. Due to the inefficient flow mechanism of melts into the mould cavity during the injection process (Summerscales *et al.*, 2010).

Other issues were raised related to the processing parameters when the biocomposites melt were being exposed to the formation of residual stress mechanisms. Residual stress is a mechanism of internal stress created by the rapid solidification of polymer melts without external pressure (Singh & Chaitanya, 2015). In general, the characterization of the residual stresses' distribution mechanism will produce stresses on the surface and inner core, as well as compression pressure on the mid-body of the composite. As a result, instant moulding defects such as warpage, shrinkage, and welding line stress will deform on a composite's surface (Chaitanya *et al.*, 2017; Singh & Verma, 2017).

The residual stress mechanism will begin to form on the loose polymer chain under high temperature and pressure conditions during the initial filling and packing stages through the injection molding process. It happened at the end of the injection process, where the polymer composite will experience the shrinkage and warpages affected by the residual stress mechanism. This condition occurred due to the incompatibility effects of the temperature on the polymer composite during the cooling phase (Ho *et al.*, 2012). Each of these processing stages is regulated by several injection parameters, which directly affects the finishing of the moulded polymer composite product. The injection parameters involved include temperature (melting and moulding), time (compaction, cooling, and filling), and pressure (injection and compaction) (Azaman *et al.*, 2015).

4. Conclusions

This paper has examined previous work on the feasibility process of injection moulding of biocomposites materials. Each stage of the process has been clearly defined in terms of the problems and constraints and presented the implementation results of various remedial measures to overcome them. It happened when the natural composite materials are often associated with poor physical resistance to moisture and temperature during the processing. The increases in fibres loads lead to the formation of agglomerations and voids in the moulded composite. It is due to the mixture yields of non-homogeneous feedstocks, with imperfect fibre-matrix impregnation.

Further, it may cause biocomposite materials to fail to meet the plastic injection moulding process's flowability criteria. The injectability of the biocomposite materials significantly depends on the early stage of feedstock preparation. These preparation steps should consider the detailed characterization of the composite material, the effectiveness of the mixing method, and the identification of the feedstock's flowability properties. The optimal processing parameters also play an essential role in producing high mechanical and physical strength performance of composites. The significant influence factor in contributing to constraints and problems is the inappropriate use of processing parameters. As a result, instant moulding defects such as warpage, shrinkage, short shot, sink and burn marks, jetting, and welding line stress will deform on the surface finished of a composite. In conclusion, biocomposites are capable of being injected as advanced polymer composites material. Moreover, it can be developed as an alternative material in producing more sustainable engineering components and comparable to other synthetic-based composites materials.

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