



Circularity and Sustainability of Bio-Based Polymer/Natural Fiber Reinforced Composite

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Abstract. Amidst growing concerns about sustainability of composite materials, the renewed push towards adoption of bio-based polymer/natural fiber reinforced composites (Bio-composite) are gaining increasing demands for various applications which are being the environmental and eco-friendly alternative to synthetic composite materials. Hence, the bio based composite development should be integrated in the circular economy (CE) model to ensure a sustainable production that leads to the conception of closed loops in which resources are in the circulation of production and consumption. However, ironically, the environmental sustainability of composite materials itself is still a challenge, due to the difficulty of recycling and reusing its components when the products reach the end of their useful life. In this context, a holistic attainment of sustainability makes it imperative to adapt sustainable practices not only for raw materials but at every stage of the product. Hence, this work provides a detailed exploration of the appropriate processing of natural fiber reinforced bio-polymer composites and an insight on using recycled bio based composite constituents which could lead to a reduction in material waste and environmental footprints.

Keywords: Bio-based polymer · Circular economy · Liquid composite molding · Natural fiber reinforced composite · Sustainability

1 Introduction

A circular economy is an effective technology for the recycling and reuse that impact of resource consumption and waste on the environment. This led to the sustainable growth and creation of alternative closed loops in which resources are in the circulation of production and consumption. As the circular economy goes beyond the linear model based on the principle: extract-use-dispose model, to create circular systems that gain the maximum value by reducing and recirculating natural resources [1]. The circular economy advocates sustainable development, promoting the necessity to strike a balance between environmental and economic values. In this context of sustainable development, there is great potential for an increase in the demand for renewable and bio-based polymers and the focus on the use of natural fibers have offered opportunities to produce highly durable natural fibers reinforced composite into new products.

Natural fiber reinforced composites are growing demand in the global sustainability and energy efficiency because as the result of biodegradability, excellent stiffness to weight ratio, low abrasion, low-cost composite with synthetic fibers such as glass or carbon fibers [2, 3] and environmental friendly reinforcement to produce sustainable composite for a wide range of applications of sports, automotive and building materials [4, 5]. This demand can be met by recycling of bio-based polymer and renewable resource as shown in Fig. 1 shows the recyclability for biocomposites.

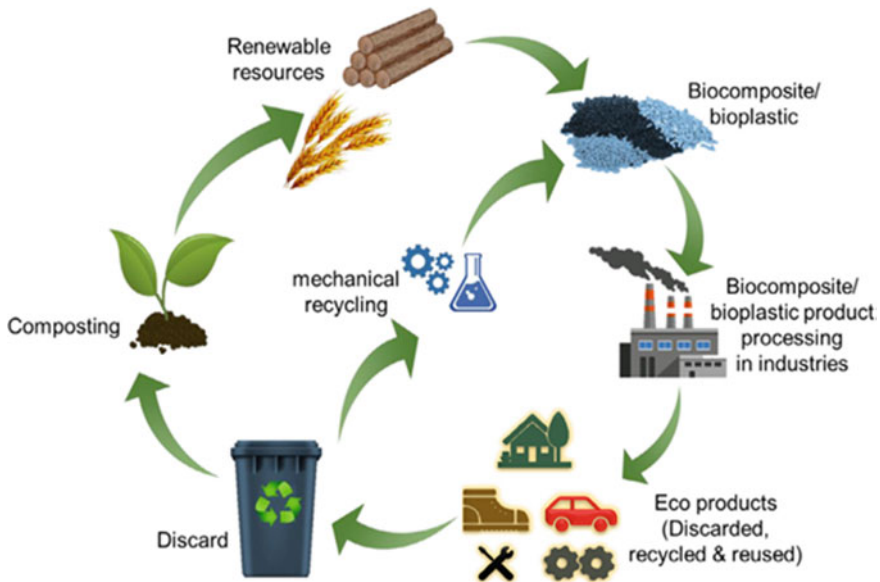


Fig. 1 A flow diagram of recycling natural fiber composites toward a circular economy in which future manufacturing processes will be designed to minimize and remove waste from the system. Figure adapted from Ref. [6]

The increasing demand for the use of natural resources in biocomposites manufacturing exemplifies the need for a CE in biocomposites that also allows for their recycling and reuse [7]. Although the concept of CE is concerned with reducing the carbon footprint at every stage of a product's life cycle. Adaptation of the CE in biocomposites improves their recyclability, reusability and convert biocomposites waste into useful products, energy or secondary materials, which could lead to a reduction of carbon footprints and bio composites waste landfilling [8].

1.1 Objectives

In the transition toward circular economy practices it is important to have a complete understanding of the impact of recycling and reprocessing on material properties of Natural fiber as they are reused in subsequent product applications. Several research studies have been reported over the past decades. The aim of the present work is to elaborate

on the suitable processing method for bio based natural fiber reinforced composite by Liquid composite molding (LCM) techniques and characterize the materials for determination of durability of the product. Furthermore, provided an overview on the use of recycle constituent into sandwich composite laminate.

2 Materials and Methods

The material studied is based on the bio-based matrix system alternative to fossil based were supplied by bto-epoxy GmbH Amstetten, Austria. The matrix is Epinal b.poxy IR 78.31 with bio-based content of 37.58% and Epinal IH 77.11, a conventional fossil-based hardener. The reinforcement plies used for the composite laminate is a balance woven fabric made of flax fiber known as Amplitex 5042 from Bcomp Ltd, Fribourg, Switzerland. The flax fiber is a twill 4/4 weave with nominal area weight of 500 g/m^2 and fiber density of 1.47. The textile reinforcement was cut into $270 \text{ mm} \times 270 \text{ mm}$ (length and width), the preform stack was composed of 6 layers of textile with nominal thickness of composite of 4 mm which has nearly fiber volume content of 48%. Since the natural fiber have tendency to absorb moisture, the preform are dried in the heating oven at $120 \text{ }^\circ\text{C}$ for 30 min. Bio based natural fiber reinforced composites were processed by vacuum assisted resin infusion (VARI) and resin transfer molding (RTM) (shown in Fig. 2a and b). Vacuum assisted resin infusion is the process in which composite is fabricated using a single sided rigid mold and vacuum bagging where vacuum pressure is applied. The schematic representation of VARI composite manufacturing is depicted in Figure 2a. Composite laminate were fabricated with nominal thickness of 4 mm using the dry virgin preforms composed of six plies were laid on the mold with distribution channel, which has been previously coated with a release agent. Release peel ply is placed over the preform, allowing easy separation from the vacuum bagging and resin distribution media is usually laid over the peel ply to enhance the speed of resin flow. Once the inlet and vent tubes are positioned, the mold is closed by the vacuum bag using the sealant tape (tacky tape). The vacuum bag provides consolidation of plies with the vacuum pressure applied to the vent where inlet is clamped. The mold tool temperature set at $60 \text{ }^\circ\text{C}$ and vacuum pressure of -1 bar is applied. Before infusion, the entire layup is subjected to the vacuum drop test by clamping the vent to check for leakage in the vacuum bagging. The resin hardener mixture of 100:25 (by weight) is degassed in a pressure pot under vacuum to reduce the air bubbles in the mixture. The degassed resin mixture is drawn into the perform by the differential pressure between the vent and the inlet (resin front). Once the preform is fully infused, inlet and vent are clamped, as the resin pressure gradients gradually dissipate and the pressure boundary conditions are maintained until the resin is cured for 180 mins. After the curing, the mold is cooled down to the room temperature and the composite laminate is demolded. Resin transfer molding is a closed mold process where the composite laminates are formed between rigid mold halves. The schematic representation of RTM composite manufacturing is depicted in Figure 2b. The mold carrier used for the RTM test series was a laboratory press, LZT-OK-80-SO from Langzauner (Lambrechten, Austria). Composite panel were fabricated using the dry virgin preforms composed of six plies were placed into the mold cavity. It should be noted that the thickness of the composite was defined by a mold cavity of 4mm. The mold

is closed and temperature was held constant at 60 °C during the injection and curing. The resin mixture in a proportion of 100:25 (by weight) is injected at constant pressure of 6 bar and flows gradually into the mold. Once the resin was observed at outlet, the vent port was closed and cured under constant pressure condition for 180 min. After the curing, the mold tool temperature is cooled to room temperature and composite laminates is demolded. The natural fiber reinforced composite laminates were mechanically and physically characterized and evaluated for tensile properties and density measurements to emphasize on the processes for obtaining longer lasting products.

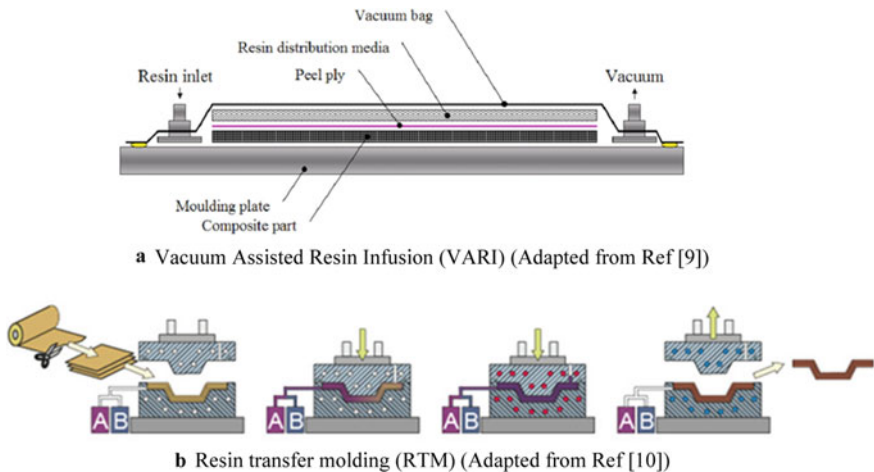


Fig. 2 **a** Vacuum assisted resin infusion (VARI). Adapted from Ref. [9]. **b** Resin transfer molding (RTM). Adapted from Ref. [10]

The specimens for tensile test were prepared according to the standard DIN EN ISO 527–4. The samples were prepared according to specimens type 2 with dimension of 250x25x4 mm with gage length of 150 mm. Glass fibre reinforced composites were used as end tabs. The experiments were performed using Z250, Zwick Roell universal testing machine (Zwick GmbH and Co., Ulm, Germany) equipped with a load cell of 20 kN and test speed of 2 mm/min. With the guidance of an extensometer, the experimental data were collected and processed automatically using testXpert III software (Zwick GmbH and Co., Ulm, Germany). Tensile characteristic was investigated on five specimens of each configuration of composite panel. Density of composite panels were measured by using the Archimedes principle of immersion method according to the standard DIN EN ISO 1183 [11]. Distilled water is used as medium of liquid for the density calibration and the specimens are prepared according to the system specifications. The specimen dimensions are 25 x 25 mm and the samples should “preferably have a mass of at least 1 g. An analytical balance, AG204 Mettler Toledo (Columbus, Ohio, United States) is used for calibration. The densities are measured for at least five specimens per composite panels for in order to get statistically relevant results.

3 Results and Discussion

In this study, the tensile modulus and strength obtained for the bio-based natural fiber reinforced composite laminates are significantly higher in the RTM process than the vacuum infusion process. Figure 3a shows the graph of tensile modulus and strength of bio based natural fiber reinforced composite processed by RTM and VARI method. Result shows that due to lower compaction behavior of the preform, the fiber content in VARI method is varied than RTM method. Table 1 shows the tensile properties, where there is small drop in strength by 7.8% (from 141.84 to 131.14 MPa), modulus by 43% (from 19.53 to 12.52 GPa) between the plates processed by RTM and VARI. However, the composite processed by VARI shows increased strain by 29% than the composite processed by RTM.

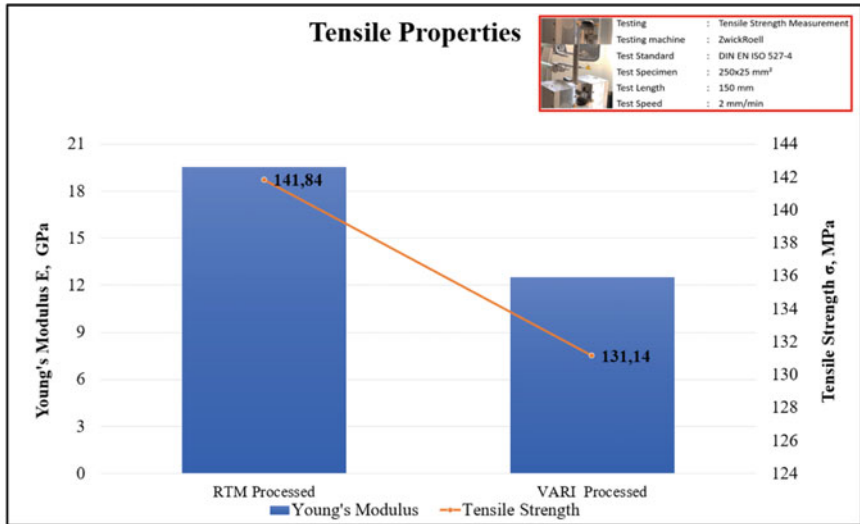
Density is also required to be known for estimating composite weight and evaluate fiber content as well as void content for property predictions. Figure 3b shows the density measurement of composite laminates processed by VARI and RTM method, as the result shows the density properties are significantly scattered over the plates manufactured. However, the average density values in RTM processed laminates have improved density value than the VARI processed, because of the better compaction and packaging of flax fiber that enhance fiber volume content in RTM method. Figure 3b shows the graph of density measurement, where there is a drop-in density measurement by 3.45% (from 1.292 to 1.251 g/cm³) within processes RTM and VARI method.

The composite laminates are analyzed in terms of their physical and mechanical properties to judge their endurance, as the properties are strongly dependent on the processing route. Nevertheless, the properties of flax/bio-based polymer composite laminates have been shown to be very promising in RTM process when compared with VARI process.

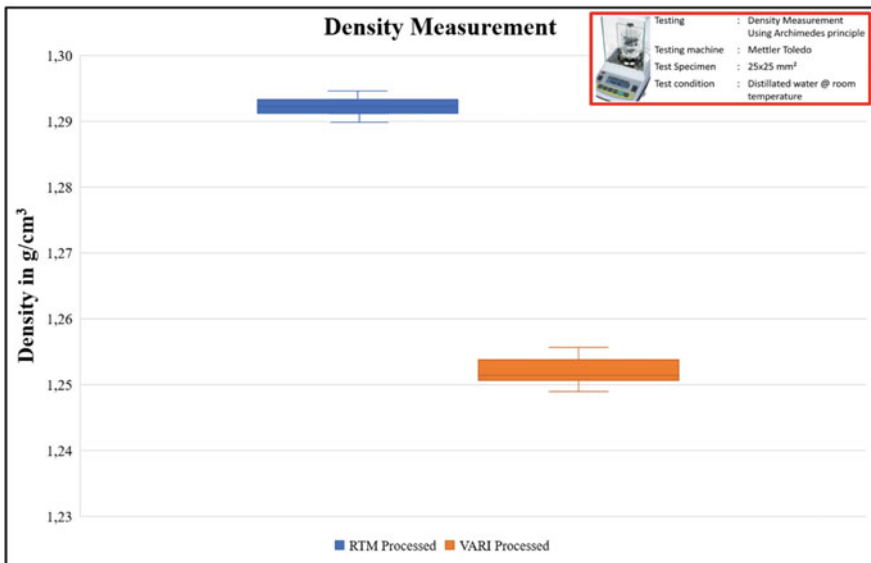
4 Conclusion and Future Outlook

In this paper, we have provided an overview of the most promising processing technique of bio-based polymer natural fiber reinforced composite using appropriate liquid composite molding (LCM). It was observed that the laminates have higher tensile strength/modulus and greater density measurement in RTM process than VARI process, but this difference is due to the compaction behavior of the fabric. However, we found that besides suitable processing methods, the mechanical properties and physical properties are interesting to judge the durability and longevity of products.

To ensure a sustainable production that leads to the conception of closed loops in which resources are in the circulation of production and consumption, the recycled natural fiber composites are utilized. The surplus natural fiber composite are mechanically recycled where the process is distinguished in three main steps, shredding, milling and classifying and shred composite waste into small particles (fibrous and resin rich powder), which is known as recyclates. The shredding process make the laminates into coarsely small fragment. Depending on the particle size and fiber quality, the recyclates can be used either as reinforcing materials or as matrix fillers in new product [11, 12]. The composite waste was shredded by DWZ shredder and the shredded samples are



a



b

Fig. 3 a Tensile properties natural fiber reinforced composite processed by RTM and VARI. b Density measurement of natural fiber reinforced composite processed by RTM and VARI

immersed in liquid nitrogen to maintain a low temperature during milling in universal cutting mill pulverisette 19.

Table 1 Tensile properties of bio-based/natural fiber composite processed RTM and VARI

Process	Tensile strength σ (MPa)	Young's modulus E (GPa)	Ultimate strain ε (%)
RTM	141.84	19.53	0.94
VARI	131.14	12.52	1.26

The samples were cut in different sieve sizes of 4 mm and 10 mm (see in Fig. 4). In the context of developing the circularity concept to reduce the risk of downcycling, the mechanical recyclates can be used as a filler between the raw materials into sandwich laminate as represented in Fig. 5 which results in potential semi structural application. The sandwich natural fiber composite is processed using liquid composite molding (LCM) method for different recyclate particle size.

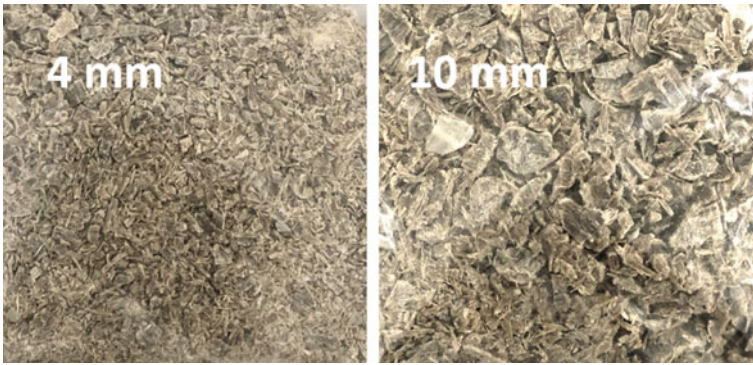


Fig. 4 Recyclate particles (4 mm and 10 mm)

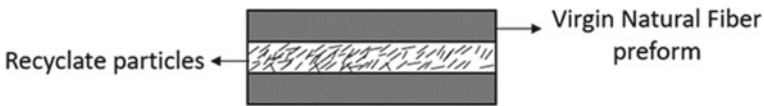


Fig. 5 Sandwich bio-based/natural fiber composite

These studies have focused only on processing the bio-based polymer natural fiber reinforced composite using appropriate liquid composite molding (LCM) technique. In contrast to this, this study looks at the wider picture and desired to expands on the processing of recycled sandwich bio-based/natural fiber composite leads to the conception of closed loops in which resources are in the circulation. To summarize, the bio-based polymer/natural fiber composite ensures reducing the ecological footprints and maintain sustainability as well as leads in potential improvement of semi structural applications.

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