IMPACT OF EXTRUSION AND COMPRESSION MOLDING TEMPERATURES ON FOAM MORPHOLOGY AND TENSILE PROPERTIES OF BIOCOMPOSITES MADE FROM RICE HUSK AND POLYOLEFIN

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Abstract. Polymer foam is known as a lightweight and porous material with various advantages over neat polymer materials such as low density, great acoustic absorption, and low cost. In this study, the solid and azodicarbonamide (ADC)-foamed biocomposites made from rice husk (RH) and polyolefin blend (recycled high-density polyethylene (rHDPE)/recycled polyethylene terephthalate (rPET)) were prepared by extrusion and compression molding. The RH and polyolefin blend composition was fixed at 70/30 wt%. This research focused on the effect of processing temperatures including extrusion temperature profiles (P1: 170-195-190-185 °C, P2: 160-185-180-175 ºC) and compression temperature varied at 195, 200, and 205 °C affected the foam morphology, density, and tensile strength of the biocomposites. By adding ADC foaming agent at 2 parts per hundred resins, the density and tensile strength of the biocomposites generally reduced which aligned with the foam (porous structure) formed as shown in the microscopic structure analyzed by Field Emission Scanning Electron Microscope (FESEM). From this preliminary finding, the best processing temperature of the formulated biocomposite foams was achieved at P2 (extrusion) and 195 ºC (compression) with the highest density reduction (10.63 %) and tensile strength at 10.8 MPa. The research indicated that lightweight biocomposite foam offers potential for sustainable applications such as the packaging industry, formwork, flooring, and automotive industry in the future.

Keywords: Biocomposite foam, azodicarbonamide, density, foam morphology

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1. INTRODUCTION

Environmental awareness has increased interest in the invention of more environmentally friendly and sustainable materials. Global interest in the field of composite materials is currently growing due to waste plastic industrial and agricultural products. Because of their trustworthy features materials, high-density polyethylene (HDPE) and polyethylene terephthalate (PET) have been widely used in the polymer industry particularly in packaging [1]. However, the solid waste of plastics has become severe that led to innovation in the use of recycled materials due to their potential for the recycling process to develop new products. Researchers discovered that the blend of HDPE/PET has the characteristics of the individual polymer components, resulting in less brittle than recycled polyethylene terephthalate (rPET) but stiffer than pure HDPE. In this blend system, the additive of ethylene-glycidyl methacrylate (E-GMA) as compatibilizer was required to improve the compatibility between hydrophilic PET and hydrophobic HDPE. This is accomplished through a strong chemical interaction facilitated by the epoxy ring-opening reaction of E-GMA, along with morphological stabilization achieved through coalescence suppression and a reduction in interfacial tension at the interface. These actions collectively enhance the properties of the resulting blend [1-2].

In striving for green material products, natural fibre such as rice husk (RH), kenaf, hemp, and bamboo has grown in popularity as a biofiller. It strengthens the mechanical and physical performances of the composite when natural fibre is added to the polymer matrix. RH has been utilized extensively due to its low density (90-150 kg/m^3), affordability, and abundance of agro waste. There are several studies using RH with some polymer matrix that can prove the improvement of mechanical properties when compared to neat polymer [2-4]. In the previous study by our research group [5], the formulation of HDPE/PET matrix blend with various RH filler loadings (30-70 wt%) was investigated and 70 wt% RH was reported as the optimum loading, which had the highest tensile modulus and flexural strength. Even the use of natural fibre has been proven to improve the strength, but the density of rice husk is higher than polyethylene thermoplastics (~87-97 kg/m^3). Therefore, using high loading rice husk (70 wt%) results in denser and heavyweight composite products. This is undesirable in many applications, such as automotive, biomedical appliances and sound insulator that require lightweight materials while maintaining mechanical performance, which is the focus of this current study.

The rise of the use of plastic products introduces the industry to the foaming technology to develop lightweight plastic items in order to reduce product weight, conserve resources, and reduce production costs [6]. To form porous material, chemical foaming agents such as azodicarbonamide (ADC) [7], sodium bicarbonate [8] and zinc carbonate [9] can be added into the composite formulation. Commonly, ADC has been chosen to form a bubbly gas inside the structure of the biocomposite due to its great efficacy in producing foaming gas, the ease of dispersion, affordable and non-toxicity. ADC is a chemical foaming agent with a decomposition temperature of 230 °C. Several studies proved the use of ADC with optimal extrusion and compression temperatures and amount produced great cell morphology with negligible changes in mechanical performance [6,10]. Researchers found that with the addition of an activator like ZnO, the decomposition temperature of ADC was reduced [11]. In this regard, the processing temperature can be a major factor in forming the size and distribution of the cell foam, which is greatly dependent upon the thermal decomposition of the foaming agent. As in consequence, it influences the resultant mechanical performance of the biocomposite foams. Recent studies [3-6, 12] have shown a well-distributed and smaller cell foam size leads to better tensile properties due to the uniform distribution and deformation of tensile stress.

Numerous research works have been conducted on foaming biocomposite using ADC [3-6,11-13]. However, the processing parameters for each formulation still vary and need to be determined at the early stage to confirm the success of the foaming process, especially considering the high filler loading (70 wt%) utilized in this research, a rarity in other studies. The processing temperatures play a vital role in determining the quality of foamed composites. Extrusion involves the early-stage mixing of the fibre-matrix which can subsequently affect the cellular structure of the foam during the compression molding. In this study, foaming occurs during hot pressing which achieved the decomposition temperature of ADC and resulting in a regulated cell structure in the created biocomposite under these circumstances. This work aims to investigate the effect of processing temperatures (extrusion and compression temperatures) on the morphological structure, density, and tensile properties of the RH/polyolefin biocomposites.

2. MATERIALS AND METHODS

2.1 Materials

Rice husk (RH; 100-mesh) was used as filler while the polymer matrix blend consisted of recycled high-density polyethylene (rHDPE; melt flow index of 0.72 g/10 min at 190 ºC, 2.16 kg load) and recycled polyethylene terephthalate (rPET; intrinsic viscosity of 0.68 dL/g). Ethylene-glycidyl methacrylate copolymer (E-GMA; Lotader AX8840 commercial name; melt flow index of 5 g/10 min at 190 ºC, 2.16 kg load) was used as polymer blend compatibilizer. RH, rHDPE, rPET and E-GMA supplied by factory namely BioComposites Extrusion Sdn. Bhd, Malaysia. Maleic anhydride polyethylene (MAPE; melting peak temperature of 135.2 ºC) manufactured by Sigma Aldrich, USA was used as a coupling agent for the biocomposite. Namely Azodicarbonamide (ADC; decomposition temperature of 230 ºC) supplied by Sigma Aldrich (Malaysia) was served as an exothermic foaming agent and zinc oxide (ZnO) was used as a foaming agent decomposition stabilizer (Supplier: R&M Chemicals, Malaysia).

2.2 Preparation of Biocomposite Foam

The preparation of RH/rHDPE-rPET biocomposite foam is shown in Figure 1. The raw materials (RH, rHDPE and rPET) were oven-dried overnight at 90 ºC. Using a co-rotating twin screw extruder (Thermo Prism TSE16PC) (manufacturer: MS Instruments Sdn. Bhd (Subang Jaya, Malaysia), a polymer matrix blend of rHDPE and rPET (75/25 wt%) was mixed with a 5 phr (part per hundred resin) E-GMA compatibilizer with a rotor speed of 30 rpm and extrusion temperatures of 190-270 °C [1]. The pre-extruded polymer blend was called a polyolefin waste blend. The biocomposite foam was prepared by mixing RH, polyolefin waste blend (70/30 wt%), MAPE (3 phc; part per hundred composite), ADC (2 phr; part per hundred resin) and ZnO (1 phr; part per hundred resin). The mixture was extruded at two distinct temperatures profiles: 175, 180, 185, 170 °C (P1) and 165, 170, 175, 160 °C (P2). Both temperatures were lower than the ADC decomposition temperature.

Following extrusion, the 3 mm thickness dumbbell-shaped samples were compression molded (hot pressing) at three distinct temperatures; 195, 200, and 205 °C for 15 minutes to form composite panels. The compression temperatures investigated were nearly close to the decomposition temperature of ADC that cause foaming. This investigation was carried out by the one-variable-at-a-time method. The solid biocomposite without ADC and ZnO was prepared to act as a control sample (CS).

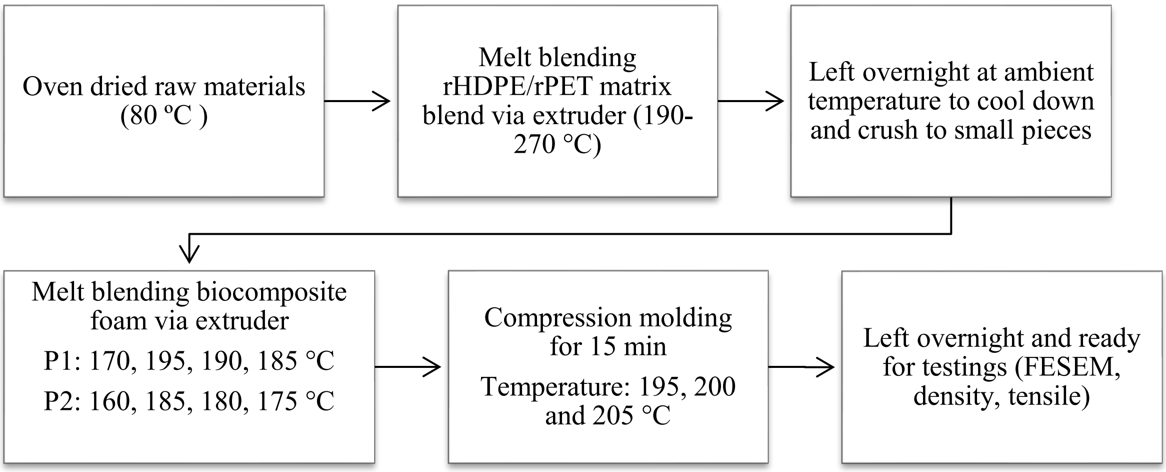

Figure 1: Preparation of RH/rHDPE-rPET biocomposite foam

2.4 Bio-Composite Foams Characterization

The foam morphology and behaviour of the biocomposite foams were studied by a field emission scanning electron microscope (FESEM) (model LEO 1450 VP) (manufacturer: Carl Zeiss (Malaysia)) that ran on an accelerating voltage of 15 kV. The density of solid biocomposite foams was determined using the gravimetric method, with mass over volume calculated for a minimum of six samples for each parameter.

The sample dimensions were measured with a vernier calliper with a precision of two decimals, and the weight was measured with an accuracy of four decimals. The tensile strength of foams was tested using a universal testing machine model Testometric M350-10CT (manufacturer: UK) based on ASTM D638-03 (type I) [3]. A 5 KN load cell and a crosshead speed of 5 mm/min were used.

3. RESULTS AND DISCUSSIONS

3.1 Cells Characterization

The cell foam was categorized via the cell structure (open or closed foamed cells and their distribution), shape (perfect circle, oval, collapsed cells), and size (big or small size, and the uniformity of the cell size) [14-15]. The foamed biocomposites prepared in this study exhibit closed cells with consistent cell size and well-distributed, but there are some variations due to the different processing temperatures (Figures 2 and 3). In comparison with the control sample (Figure 2), the biocomposite foam (Figure 3) showed porous structures with varying cell sizes on their surface morphology.

Notably, biocomposite foam with higher extrusion temperature (P1) exhibits larger and non-uniform cell distribution compared to P2 which obviously can be seen in Figure 3, respectively. This result can be supported by the research of Abu Hassan et al. [6] who proved that the temperature lower than ADC decomposition was preferable in poly (lactic acid) /kenaf biocomposite foam. The extrusion temperature of P1 (170, 195, 190, 185 ℃) is higher and closer to the thermal decomposition of ADC. This adversely affects the cellular morphology in the foamed biocomposites as can be seen by non-uniform cell shape and distribution, and denser structure when compared to P2. This is owing to the previous study that stated higher temperature lowered the viscosity and caused a subsequent result in fewer cells and a larger cell size [16]. The morphology of biocomposite foam pressed with 195 ℃ (Figure 3(f)) formed consistent cell size and better distribution than 205 ℃ (Figure 3(b)) and 200 ℃ (Figure 3(d)).

At the hot-pressing temperature, it is believed to trigger the decomposition of ADC, thereby producing decomposition gases. In this case, a compression temperature of 195 ℃ is found to produce better foam to minimize the density of the biocomposite and maintain the mechanical effectiveness. This temperature seems to be optimal for ADC decomposition, ensuring stable cell growth and favouring the production of well-distributed and homogeneous foams. This is supported by the report of a previous study [17] which found that the presence of ZnO can reduce the ADC decomposition temperature by 40 °C (from 230 °C).

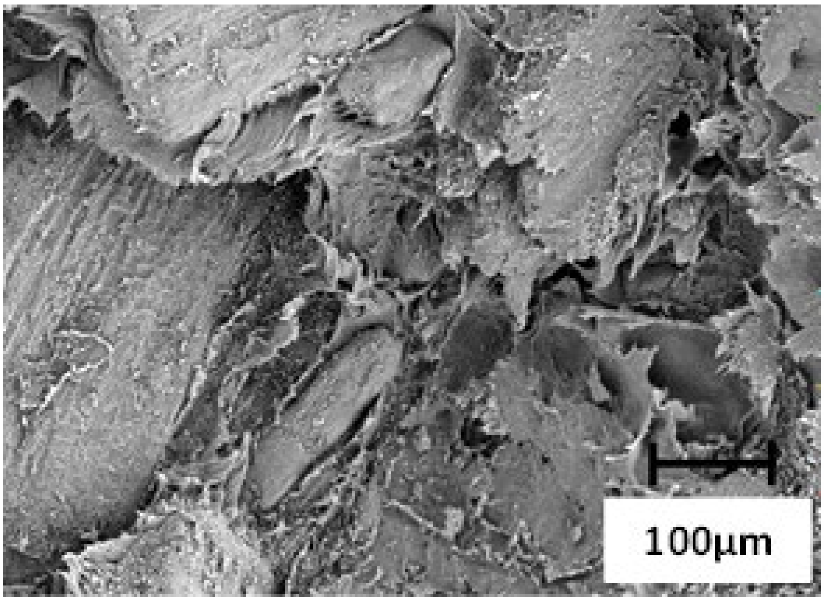

Figure 2: FESEM image of RH/rHDPE-rPET biocomposite control sample (P1/HP195ºC)

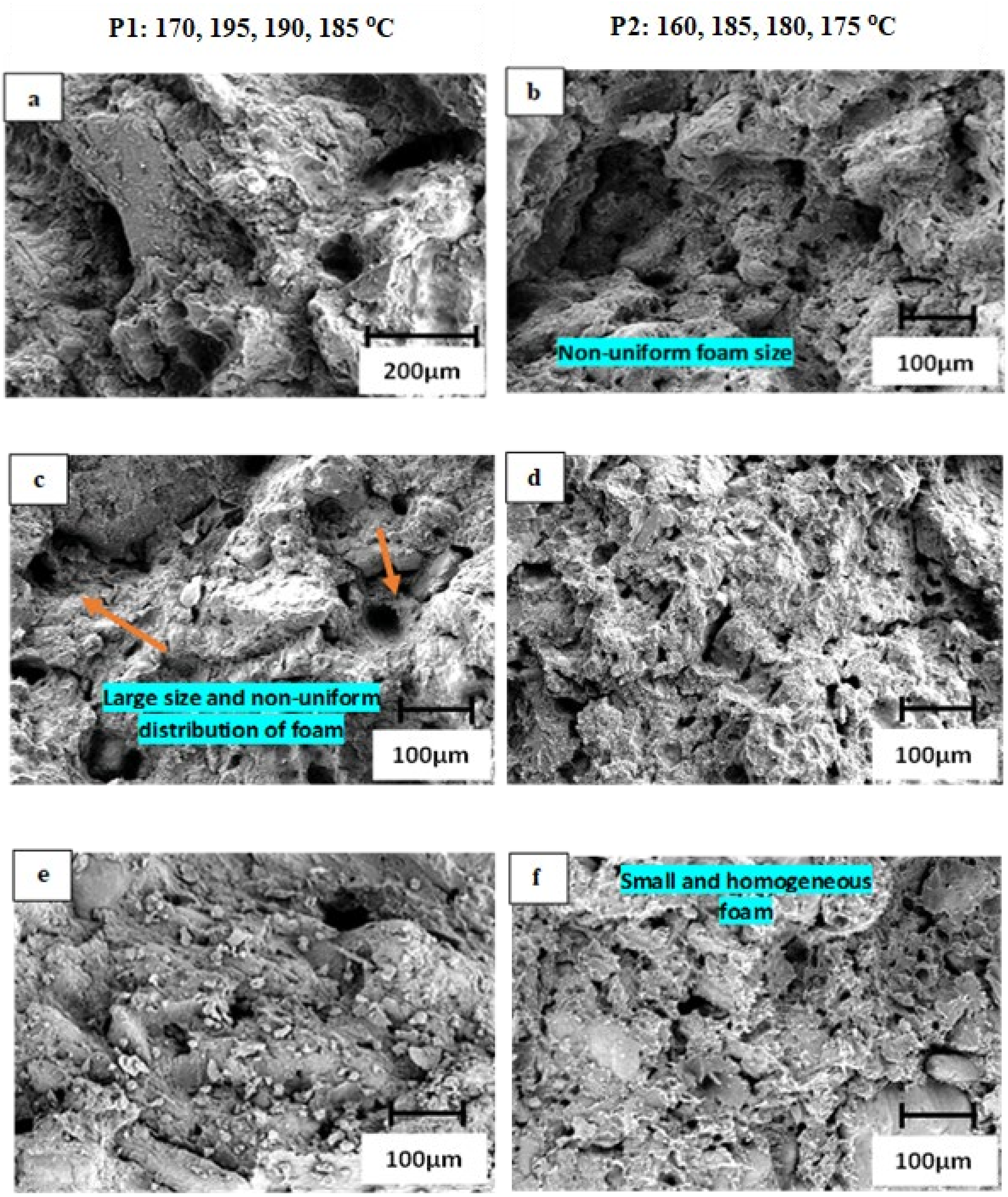

Figure 3: FESEM micrographs of biocomposite foam with different extrusion temperatures: P1 (a, c, e), P2 (b, d, f) and different compression temperatures: (a, b) 205 ºC, (c, d) 200 ºC, (e, f) 195 ºC

3.2 Density

Figure 4 shows the impact of processing temperature on the density of the biocomposite. The density of unfoamed biocomposite was recorded at 1.25 g/cm^3. Biocomposite foam extruded with lower extrusion temperature (P2) generally had lower density than higher temperature (P1). Meanwhile, a compression temperature of 195 ºC produces the lowest density of the sample when compared to 200 °C and 205 °C. From this finding, the highest density reduction is achieved at P2 with 195 °C compression temperature, which is 10.63% reduced from the neat biocomposite. These findings are substantiated by microscopic observations, which demonstrate that the foamed biocomposites extruded and compressed at low temperatures have fewer compact structures than high temperatures (Figure 3(f)). In accordance with the research [18], lower foaming temperature contributes to the biocomposites forming high cell density that leads to the smaller and more uniform cell size of foam.

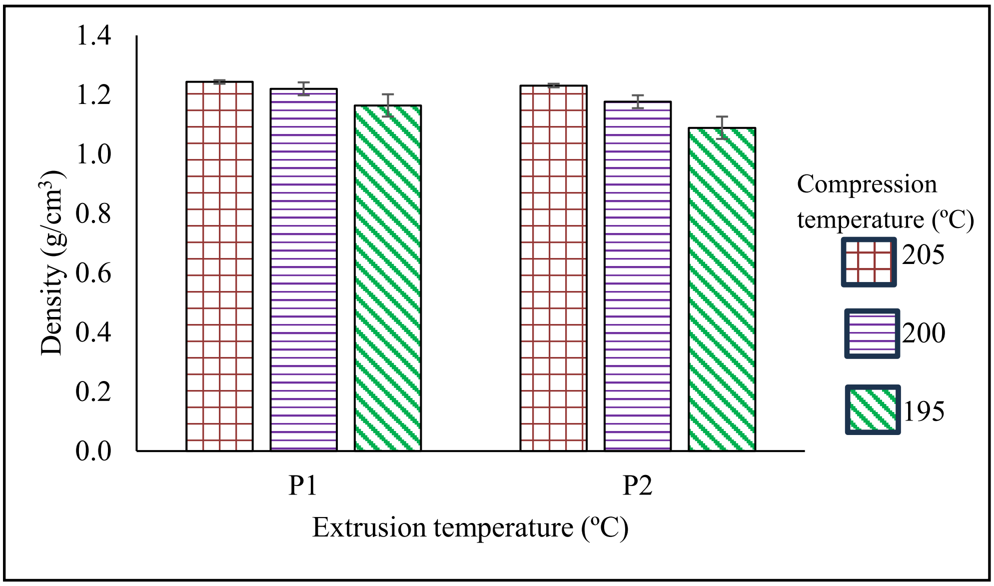

Figure 4: The influence of extrusion and compression press temperatures on the densit y of biocomposite foam. Density of CS: 1.25 ± 0.005 g/cm^3

3.3 Mechanical Tensile Performance

Figures 5 and 6 illustrate the impact of extrusion and compression temperature on the tensile strength (stress yield) and Young's Modulus of biocomposite foams. The unfoamed biocomposites extruded with high temperature (P1) show 11.4 MPa tensile strength and 982.3 MPa of Young's Modulus, whereas the samples extruded with lower temperature (P2) show values of 12 MPa and 1050.5 MPa, respectively. The biocomposite foam compressed with the temperature of 205 ºC has the lowest stress yield and Young Modulus values when compared to 200 ºC and 195 ºC. These results were similarly reported by Abu Hassan et al. [6]. The mechanical properties deteriorated at higher temperatures due to the microstructure that formed non-uniform and collapsed cells. This results in the uneven and inconsistent stress distribution, causing localized stress concentration that might lead to premature failure. The foamed biocomposites with temperature P2 and 195 ºC exhibit a low decrement in both stress yield (13.95 %) and Young's Modulus (19.44 %) compared to the unfoamed biocomposites. This is ascribed to the better dispersion and homogeneous morphology of the foamed cells in biocomposites (Figure 3(f)). Stress is uniformly distributed across the specimen cross-section due to well-distributed, homogenous foam cells. Since the cells in a homogeneous foam morphology are usually consist of smaller cells with high cell density and thick cell walls. Stress is absorbed from one cell to another with efficiency, rendering the structure of the foam deformation inconsequential [19]. As a result, high cell density foams have lower tensile failure and superior mechanical properties [20].

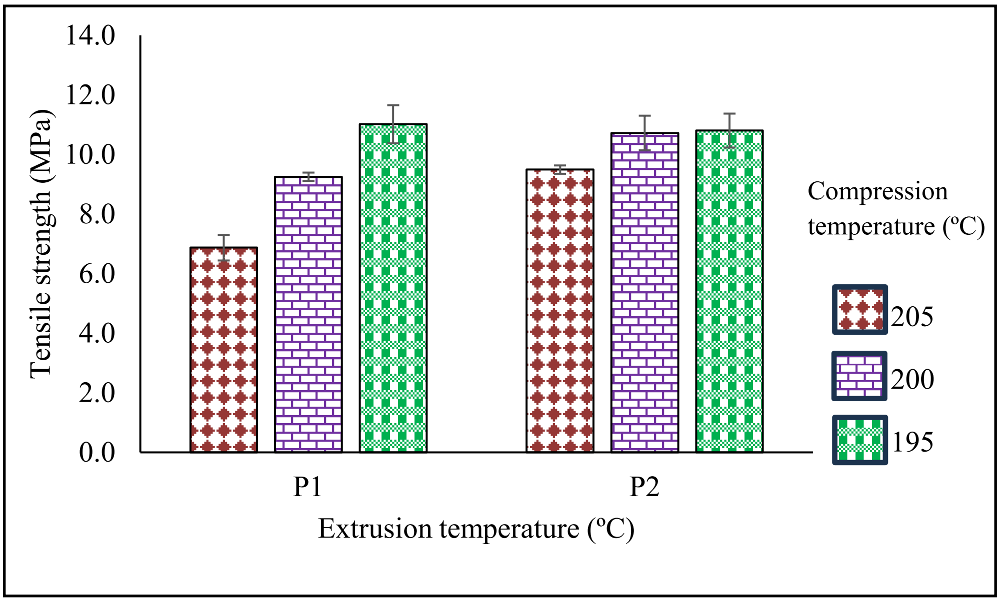

Figure 5: The influence of extrusion and compression temperature on the stress at yield of biocomposite foam. Control sample prepared by P1 and P2: 11.4 ± 0.4 MPa and 12.0 ± 0.8 MPa

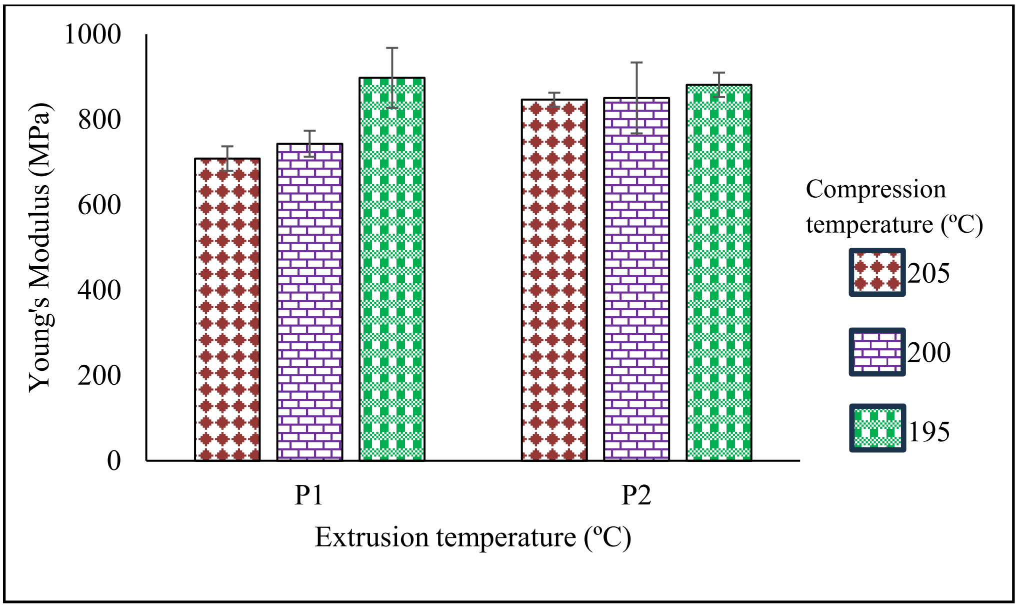

Figure 6: The influence of extrusion and compression temperature on the Young's modulus of biocomposite foams. Control sample prepared by P1 and P2: 982.3 MPa ± 20.1 and 1050.5 ± 47.9 MPa

4. CONCLUSIONS

The research worked on the processing temperatures influencing the foam morphology, density, and mechanical performance of RH/polyolefin blend biocomposites.

The biocomposite foams prepared by extrusion temperatures with P2 (160-185-180-175 ºC) and compression pressing temperature of 195 ºC remarked the homogeneous foam with the highest density reduction and negligible changes of mechanical properties. The biocomposite foam investigated in this study shows potential for versatile applications such as building construction, food packaging, heat and sound insulation, biomedical appliances and the automotive industry. However, the foam properties obtained here have not yet reached a sufficient lower density (1.09 g/cm^3) compared to commercial HDPE (0.93-0.97 g/cm^3) and polypropylene packaging (0.89-0.91 g/cm^3) foam materials. Further work on the development of various loadings of foaming agents with optimized processing temperature might contribute to better foaming quality and low-density product.

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Author contributions

All authors contributed toward data analysis, drafting, and critically revising the paper and agreed to be accountable for all aspects of the work.

Disclosure of conflict of

The authors have no disclosures to declare.

Compliance with ethical standards

The work is compliant with ethical standards.

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