Research Article

Hybrid Biocomposites Based on Used Coffee Grounds and Epoxy Resin: Mechanical Properties and Fire Resistance

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Studies on using biomaterials hybridized with other materials to produce biomaterials have been paid more attention due to their low cost, abundance, renewability, and degradability. Therefore, these materials are ecofriendly and nontoxic to humans. A large number of used coffee grounds (SCGs) are often discarded and replacements are necessary for dealing with environmental problems. This work developed sustainable materials by reusing SCGs. Used coffee grounds were mixed with epoxy resin at different amounts: 30 wt %, 40 wt %, 50 wt %, and 60 wt %. SCGs were treated with 0.5 N NaOH, at SCGs/NaOH ratio of 1 : 2. SEM images showed that the material with 30 wt % SCGs has good compatibility without phase division on the SCGs-epoxy interface. Results of mechanical properties of epoxy composites with 30 wt % SCGs are as follows: tensile strength of 44.81 ± 10 MPa, flexural strength of 80.07 ± 0.16 MPa, compressive strength of 112.56 ± 0.11 MPa, and Izod strength and impact of 8.21 ± 0.19 kJ/m². In terms of flame-retardant properties, the oxygen index is limited to 20.8% ± 0.20 and the burning rate according to UL94HB is 27.02 ± 0.29 mm/min. The obtained results indicate that it is possible to produce biohybrid composites from epoxy resin and SCGs. This work offers an ecofriendly alternative method to use the waste of the coffee industry. It contributes to improvements of the general characteristics of composites such as mechanical, thermal, and flame-retardant properties. This work proved that SCGs have a high potential to be used in a wide range of composite materials for civil engineering applications.

1. Introduction

Coffee is one of the most important agricultural products sold worldwide [1]. World coffee exports reached 11.94 million bags in March 2021, compared with 11.66 million bags in March 2020 [2]. Nearly 50% of coffee production worldwide is processed to make instant coffee, producing about 6 million tons of spent coffee grounds (SCGs) per year [3]. As such, coffee waste poses a high risk of pollution when released into the environment unless it is exploited for other purposes. However, as this residue is derived from coffee beans, it is expected to have similar properties to these beans and thus could be exploited for industrial applications [4]. In this context, several possibilities have been proposed to exploit these coffee grounds to be reused in different applications. For example, Garcia-Garcia et al. and colleagues fabricated wood plastic composite (WPC) with a polypropylene substrate, using 20% by weight of used coffee grounds. The obtained results show that the addition of both treated and untreated SCGs to one polypropylene promotes a slight decrease in flexural strength, and the flexural modulus increases [5]. Conventional coffee-making techniques produce large quantities of used coffee grounds rich in lignocellulose and valuable biologically active substances. Used coffee grounds, an underutilized source of biomass, are potential raw materials for sugar industry and a promising extracted bioactive source. Used coffee powder from SCGs contains a large number of organic matter compounds (fatty acids, amino acids, polyphenols, minerals, and polysaccharides) to justify its valence [6]. Scully et al. reported that
coffee grounds are a promising source of industrially important sugars and polyphenols [7]. Coffee grounds can be considered a green, biodegradable material. The idea of recovering used coffee grounds by introducing them as reinforcements and fillers for polymer materials is likely to be widely studied. Some studies have used different polymers such as polypropylene (PP), polylactide (PLA), and polyvinyl alcohol (PVA). Research results show that when adding used coffee grounds to epoxy resin, some mechanical properties are slightly reduced but not much, and some other mechanical properties are also significantly improved. Coffee grounds can be said as the best material for composites [8, 9]. Suaduang et al. studied the inclusion of coffee grounds in poly lactic acid at the concentrations of 5 wt.%, 7.5 wt.%, and 10 wt.%. The results show that the elongation at break is improved with increasing coffee grounds content [10]. Tarazona et al. used 30 wt % and 35 wt % coffee grounds in making green composites based on epoxy resin, with 35 wt % coffee grounds for best mechanical properties [11]. In addition, to increase the applicability of coffee grounds, coffee grounds have been treated with alkalis at different concentrations. This method can change the structure and, under optimal conditions, does not break the basic structure [12]. To improve the mechanical properties and fire retardancy of epoxy materials, environmentally friendly additives have been studied. Additive nanoclay, multilalled carbon nanotubes are incorporated into epoxy resin to improve mechanical properties while increasing fire retardancy [13–15]. Besides, fly ash waste from thermal power plants is also recycled by using it as an additive to composite materials to improve fire resistance [16–19]. The study of mechanical properties of fiber-reinforced composites is of great interest to many scientists. Mostafa et al. studied the tensile properties of fiber/fabric-reinforced composites. One of the research results only shows that the degree of residual stress induced in the composite components does not only depend on the degree of pretreatment of the fiber but also on the elastic properties of the composite [20]. Therefore, the initial fiber treatment before processing is very important. The structure of composite materials is also studied through tensile and thermal testing [21]. Recent research activities show that biological compounds such as sisal fibers [22], lemon and lime peels [23], cellulose and silk [24], and various fillers obtained from the outermost skins of onions, potatoes, and carrots [25] are an approach to creating biomaterials with desired specific properties. Banana fiber and eggshell powder obtained from agricultural and postconsumer waste are used as biomaterial fillers to reinforce concrete [26]. In addition, chicken waste (femur and beak) and fish bones were used as an adsorbent to remove Cd²⁺ from aqueous solutions [27]. Saberian et al. had an overview study on recycling used coffee grounds as building materials. They concluded that SCGs have a high potential for use as composites in building materials in a wide range of civil engineering applications. However, research is limited and there is a lack of evidence of successful practical applications in the field of building materials [28].

However, the inherent brown color of SCG places a limit on the color of the composite product and thus Li et al. studied the debrowning process of coffee grounds and reinforced polylactic acid (PLA) resins [29], in addition to researching and using used coffee grounds to make multifunctional materials on plastic substrates such as epoxy and polylactic acid (PLA). They used coffee grounds as a versatile green energy source. Extraction of biodiesel from coffee grounds has been studied to significantly increase the profitability of biodiesel production, especially for the formation of glycerol carbonate. Coffee grounds after extraction of oil are an ideal material to make garden fertilizer, a raw material for ethanol, and a type of fuel [25, 30].

There is a need for research and development of new green composites to meet the growing industry demand for structural applications. In this study, used coffee grounds at 30 wt %, 40% wt %, 50 wt %, and 60 wt % concentrations were added to epoxy resin to form hybrid green composites. Mechanical properties such as tensile strength, flexural strength, compressive strength, and Izod impact strength were investigated. The SEM method was used to study the morphology and structure of the materials. Fire resistance was investigated by LOI and UL94-HB methods.

2. Materials and Methods

2.1. Materials

(i) Epikote 240 epoxy (EP 240) was obtained from bisphenol F, of Shell Chemicals (USA) with 24.6% epoxy content, equivalent to epoxy group 185–196, and viscosity 0.7 ÷ 1.1 Pa s at 25°C (see Table 1).

(ii) Curing agent used was diethylenetriamine (DETA), from Sigma-Aldrich; chemical formula of DETA is H₂N(CH₂)₂NH(CH₂)₂NH₂, with MW of 103 g mol⁻¹ and specific gravity at 25°C: 0.95 g/cm³.

(iii) Spent coffee grounds (SCGs) were collected from coffee shops in Hanoi City, Vietnam. Coffee was produced by Trung Nguyen Coffee Company, Vietnam.

(iv) NaOH was obtained from Sigma-Aldrich, Vietnam.

2.2. Methods

2.2.1. Sample Preparation. SCGs were washed with distilled water (7 to 10 washes) at room temperature to achieve a neutral pH level. The SCGs were then dried in a drying oven at 80°C for 8 hours. The SCGs were then sieved to obtain less than 50 μm in size and selected SCGs were stored for later use in all studies.

Pretreatment of spent coffee grounds: SCGs were soaked with NaOH 0.5 N (SCG/NaOH 0.5 N ratio = 1 : 2) for 5 hours at 50°C. Then, each sample of the treated SCGs was dried at 80°C in a drying cell for 8 hours and stored in a glass bottle for use in the next analysis [12].

Dried SCGs were added to the epoxy resin at the following mass percent: 30 wt %, 40 wt %, 50 wt %, and 60 wt % SCGs. The mixture was mechanically stirred at 1500 rpm at 80°C for 90 min. The mixture is allowed to cool at room temperature; then, a DETA curing agent (50 grams of epoxy resin...
Table 1: Properties of epoxy Epikote 240 resin.

| Properties             | Test method | Unit  | Value   |
|------------------------|-------------|-------|---------|
| Epoxy group content    | SMS 2026    | mmol/kg | 5100–5400 |
| Epoxy molar mass*      | g           |       | 185–196 |
| Viscosity at 25°C      | ASTM D445   | Pa.s  | 0.7–1.1 |
| Colour                 | ASTM D1209  | Pt-Co | 200 max |
| Density at 25°C        | ASTM D4052  | kg/L  | 1.12    |

*No. of grams of resin containing 1 g equivalent of epoxide (weight per equivalent, WFE, is an alternative term).**1 Pa.s = 10 poise.

requires 12 grams of DETA) to the mixture. Stir the mixture at 300 rpm for about 7–10 minutes and then pour the mold. To cure at room temperature for 24 hours, dry at 80°C for 3 hours. One week later, remove the mold to get the product. The product is stored in hygroscopic plastic containers and measured for properties. A summary of the pretreatment process of SCG and the fabrication of epoxy composites is presented in Figure 1.

2.2.2. Analysis

(i) Fire-Retardant Evaluation Method.

(i) Limiting oxygen index (LOI) according to JIS K720 standard (Japan): the sample bars used for the test were 150 × 6.5 × 3 mm³. The average values of the five specimens were reported.

(ii) The horizontal burning tests (UL-94HB): standard bar specimens are to be 125 ± 5 mm long by 13.0 ± 0.5 mm wide, provided in the minimum thickness of 3.0 (~0.0 + 0.2) mm (ASTM D635-12). The average values of the five specimens were reported.

The UL 94 flame-retardant and oxygen limit tests are conducted at the Polymer Materials Research Center, Hanoi University of Technology, Vietnam.

(ii) Method for Determining Mechanical Properties.

(i) Tensile strength was determined according to ISO 527-1993 standard on an INSTRON 5582-100 kN machine (USA) with a tensile speed 5 mm/min, temperature 25°C, and humidity 75%. The average values of the five specimens were reported.

(ii) The flexural strength was determined according to ISO 178-1993 on an INSTRON 5582-100 kN machine (USA) with a bending speed of 5 mm/min, a temperature of 25°C, and a humidity of 75%. The average values of the five specimens were reported.

(iii) Compressive strength was determined according to ISO 604–1993 standard on an INSTRON 5582-100 kN machine (USA), with a compression speed 5 mm/min and temperature of 25°C. The average values of the five specimens were reported.

(iv) Izod impact strength was determined according to the ASTM D265 standard on the Tinius Olsen machine (USA), measured at Research Center for Polymer Materials, Hanoi University of Science and Technology. The average values of the five specimens were reported.

(v) The morphology of the samples was carried out with a scanning electron microscope (SEM, SU3800, HITACHI, Japan), measured at Materials Room 1, Faculty of Mechanical Engineering Technology, Hanoi University of Industry, Vietnam.

3. Results and Discussion

3.1. Morphology. The structural morphology of the hybrid SCGs/epoxy composites was evaluated by SEM and is presented in Figure 2.

From Figure 2, it can be seen that when mechanically stirring at 1500 rpm at 80°C, the coffee grounds moved strongly and the impact performance was strong, thus enhancing the penetration into the plastic substrate and increased dispersion. The size of the coffee grounds is reduced. Also, from the morphological structure of the SCGs-epoxy hybrid material (see Figure 2), it is shown that the NaOH-treated coffee grounds give an effective wetting ability to the epoxy resin base, along with the strong surface adhesion between the phase of materials (according to Figure 2, the overall image SEM of the destroyed material surface at 45 resolution). We can also observe microcracking that appears normal to the grain contour, and this harms the compatibility (Figure 2, red circle area and white arrow). However, the crack size is small and the path length is short. The compatibility is not good due to the appearance of phase separation on the epoxy-SCGs interface. This is completely normal because when the surface of SCGs has not been modified much, the compatibility will decrease. Phase separation is highly efficient when the surface of the SCGs is modified and the input content is at the right level [8].

The dispersion of ESCG particles is homogeneous as depicted in Figure 3. On the interface between SCGs–epoxy, the interaction is very good, and wetting and mechanical properties can be improved. However, observing Figure 3(d), there is a separation between epoxy materials and SCGs (two white arrows). However, observing Figure 3(d), there is a separation between epoxy materials and SCGs (two white arrows). This delamination is the result of the force transfer process acting on the coffee grounds filler; the coffee grounds have received the force and are loaded instead of the epoxy base material. The addition of coffee grounds filler changed the path of the crack. Observation on the interface of the crack showed that the addition of coffee grounds filler caused a change in the direction of the crack. The upper adhesive bond fracture type in Figure 3(d) shows a good interaction between the fillers of waste coffee beans powder and epoxy.

Figures 2 and 3 show that with the addition of coffee grounds to the content of 60 wt %, the compatibility is observed on the phase division surface. However, due to the high content, the coffee grounds particles have agglomerated into larger particles than the shape of the coffee grounds (see Figures 2 and 3). There is a phase separation on the coffee grounds-epoxy interface because at the position where force is applied, the amount of object is different. This can lead to deterioration of mechanical properties.
When the coffee grounds content was reduced to 50%, although no agglomeration of coffee grounds was observed, large cracks were still clearly visible on the SCGs-epoxy interface (Figures 4(a) and 4(b)). At the same time, because the content of coffee grounds is still high, the viscosity of the mixture is large, leading to the existence of air bubbles (holes in Figures 4(b) and 4(c)). In a certain region, epoxy-SCGs still exist without any cracks (Figure 4(d)).

From the results of the morphology and structure (SEM image, Figure 5), the fracture surface of the SCGs-epoxy hybrid material, the cracks, and agglomeration (clustering) of the SCGs are almost absent. Cracks are reduced and voids are no longer present (mixed viscosity decreases).

The SEM images in Figure 6 show the difference in structural morphology compared with the SEM images of Figures 2–5. Figure 6(c) shows that the coffee grounds are in
the form of beans with sizes ranging from about 20 to 50 µm, already completely embedded in the plastic mass. The coffee grounds have been separated from each other, not as clumps, and the particles are covered with epoxy resin. Cracks still appear on the fracture surface; however, the fracture trajectory tends to change due to the influence of SCGs. The compatibility is clearly shown in Figure 6(c), and phase separation was not observed. Wetting was performed on the SCGs-epoxy interface. Possibly at 30 wt % SCGs, the best compatibility was achieved compared with 40 wt %, 50 wt %, and 60 wt % concentrations. It is promising this is the rate at which mechanical properties and flame retardancy will be improved [10]. Through SEM images of the fracture surface of the material, it shows that delamination or fracture occurs with the form between the particles and the epoxy-based resin (broken on the epoxy-SCGs phase-divided surface interface).

3.2. Mechanical Properties. The mechanical properties of SCGs/epoxy materials are presented in Figure 7.

From the results of mechanical properties in Figure 7, it is shown that when increasing the content of SCGs from 30 wt % to 60 wt %, the general mechanical properties (tensile strength, flexural strength, and compressive strength) tend to decrease. This is a common characteristic of materials when using fillers of natural origin (kaolin, talc, or CaCO₃). Only the Izod impact strength increased slightly compared to the rest of the models. Possibly due to the good compatibility on the epoxy-SCGs interface, this interaction is strong enough to increase the impact strength. However, other durability tends to decrease, but not much. This is consistent with the argument in the structural morphology section. In terms of tensile strength, in the presence of SCGs, the tensile strength of the samples decreased especially at 60 wt % SCGs. This can be explained by the low compatibility, adhesion, and dispersion, as well as the uneven geometric structure of the coffee grounds. At the concentrations of 50 wt % and 60 wt % SCGs, there is phase separation on the SCGs-epoxy interface and the association between SCGs and epoxy resins is not high. As the SCGs particles are well dispersed in the epoxy matrix, the overall polymer-SCGs particle interaction has been improved. When increasing from 30 wt % SCGs to 60 wt % SCGs, the mechanical strength, in general, tends to decrease. This is completely consistent with the general rule [8, 10]. Patil et al. studied the inclusion of treated natural fibers (sweet lemons and limes) granules reinforced with 10%, 20%, and 30% volume fraction in epoxy resin, resulting in mechanical properties. Studies show that 30% is the best. At 30% volume fraction (sweet lemon and lime), the tensile strength reached 35.16 MPa and 48.22 MPa, respectively. Meanwhile, the flexural strength of the sweet lemon sample reached 72.11 MPa and the common lemon reached 79.32 MPa.
From Figure 7, it is shown that, for the 30 wt % SCGs sample, the best tensile strength (44.81 MPa) and similarly flexural strength of 80.07 MPa is higher than the remaining samples. This shows that the research results in this work are completely grounded. Used coffee grounds mixed with epoxy at 30 wt % are the most feasible, suitable for composite materials reinforced with biological materials (coffee grounds, lemon, sweet lemon, etc.).

The results of the measurement of mechanical properties in Figure 7 also show that the 30SCG-epoxy sample is a used coffee grounds sample that has not been treated by NaOH with lower mechanical properties than the model that has been treated with 30SCGs-epoxy and other samples. It is clear that the treatment with NaOH aids SCGs to bind the epoxy resin matrix to form a hybrid material. Therefore, the mechanical properties of the samples with treated coffee grounds have much higher mechanical strength.

3.3. Fire-Retardant Property. Flame-retardant properties of SCGs/epoxy materials are shown in Figure 8.
Figure 6: SEM images of SCGs/epoxy (30 wt % SCGs), in different resolutions.

Figure 7: Continued.
From Figure 8, it can be seen that the content of SCGs also affects the fire retardancy of epoxy resin more or less. When the content of SCGs increases, the compatibility decreases leading to an unstable structure and this is the cause of the reduced fire retardancy of the material. Especially at the combined ratio of 30 wt % SCGs, which is a fairly good compatibility ratio, the mechanical properties are high and the flame retardancy is better than other ratios. At the combination of 30 wt % SCGs, the material structure achieved good compatibility (as commented in the structure section via the SEM image above). Looking at the SEM image in Figure 6(c), Coffee grounds are soaked in epoxy resin and surrounded by epoxy resin. It is this compact structure, without holes, that makes the flame-retardant property improve (LOI index reaches 20.8%, burning rate \( V = 27.02 \text{ mm/minute} \), see Figure 8).

From Figure 9, at magnifications of 10.0 k and 20.0 k, we can see that used coffee grounds can exist in small sizes. We observed that the SCGs particles bonded to the epoxy resin very strongly; they formed with the epoxy into a smooth, defect-free substrate. This has had a huge impact on the mechanical properties especially the flame-retardant properties. It is demonstrated that a 30 wt % SCGs blend provides mechanical properties and flame retardancy at the specified level. This may be related to successful oil removal during NaOH treatment. The absence of oil along with reduced particle size may allow for better homogeneity in epoxy, improved surface interaction between SCGs-epoxy [8].
The mechanical properties obtained give similar values to other biosynthetic materials, confirming feasibility of using used coffee composites production facilities [5, 8, 11, 23, 25].

In this article, after selecting 30 wt % of used coffee grounds as appropriate, we included this ratio to process glass fiber-reinforced epoxy composite materials. To evaluate higher applicability, the compatibility of coffee grounds with industrial glass fibers is also considered through SEM images, mechanical properties, and fire retardancy.

From the structural morphology, mechanical properties and flame retardancy of materials include different SCGs concentrations. 30 wt % SCGs will be selected for further research into fiberglass reinforced composites. The morphological structure of the SCGs/fiberglass/epoxy material is shown in Figure 10. From the SEM image in Figure 10, it is
shown that the bond between the fiber and the epoxy/SCGs resin is relatively good; the compatibility here is very good and there is no flaking (Figure 10(a)). Fiberglass fibers are dislodged from the epoxy matrix and break when an external force is applied to the rough surface (Figure 10(c)). Simultaneously, crack development in the epoxy matrix was prevented by glass fibers (white arrows, Figure 10(d)). Observation of Figure 10(b) shows that the broken glass fiber has a rough surface that shows the damage with more energy consumption. It proves that when the coffee grounds additive is present, the compatibility does not decrease, but rather increases. The epoxy-SCGs-glass fiber interface is strongly bonded and interactive.

In the case of flame cy, SCGs act as a filler of biological origin to solve the problem of a green environment. As for the flame-retardant properties, the research results in this work show that there is almost no effect when mixed at different concentrations. If the application is in the field of building materials with fire resistance, it is possible to mix other flame-retardant additives to have a resonant performance [31].

Figures 11(a) and 11(b) show that the ash after burning has areas of the porous structure (Figure 11(b)); there are dense, continuous areas without pores (Figure 11(a)). This is determined by the fine dispersion states of the SCGs, the role of the binder between the SCGs, and the epoxy that will promote flame retardancy.

From the results of Figure 11(b), it was found that when testing the fire-retardant properties of the reinforced material with glass cloth, it was found that only the epoxy resin and part of the coffee grounds were burned. The glass cloth remains intact (Figure 11(b), red arrow). In this case, the
glass cloth acts as a flame retardant. Therefore, the research on putting used coffee grounds additives into glass fiber-reinforced composites is a prospect in the future, applying to multifunctional structures with high fire resistance or other materials construction material [32].

The flame-retardant properties of glass fiber-reinforced SCGs/epoxy materials are shown in Figure 12. From the results of mechanical properties and flame retardants, it is shown that when 30 wt % of SCGs are combined with fibers glass for processing epoxy-based composites, mechanical properties and flame-retardant properties are kept at the specified level. Composite materials in the presence of SCGs, the mechanical properties, and fire retardant are almost similar to those of epoxy-based composites reinforced with glass fiber. Therefore, this result has scientific significance as it can be used. Coffee waste has partially replaced epoxy resin to make composite materials.

4. Conclusions

In this study, the development of hybrid biocomposites based on epoxy and SCGs was completed, the mass percent of SCGs used were 30 wt %, 40 wt %, 50 wt %, and 60 wt %. The following conclusions are drawn from the current investigation:

(i) Hybrid epoxy biocomposite material developed with used coffee grounds as reinforcement for epoxy resins performs well at 30 wt %.

(ii) Test results of mechanical properties of used coffee grounds hybrid epoxy composite material at 30 wt %, tensile strength 44.81 ± 0.10 MPa, flexural strength 80.07 ± 0.16 MPa, compressive strength 112.56 ± 0.11 MPa, and Izod impact strength of 8.21 ± 0.19 kJ/m².

(iii) Test the flame-retardant properties of the biocomposite hybrid with 30 wt % of used coffee grounds, the limiting oxygen index is 20.8 ± 0.20%, and the rated burning speed according to the UL94-HB method is 27.02 ± 0.29 mm/min. The results show that used coffee grounds do not have much effect on the flame-retardant properties of epoxy resins.

(iv) The surface morphology of the SEM images was revealed when mixing 30 wt % of used coffee grounds (SCGs) with epoxy. The obtained bio-composites had better structural compatibility at other mixing concentrations. On the SCGs-epoxy interface, no phase separation is observed.

(v) This study offers a solution to use coffee grounds as an environmentally friendly material to improve the amount of waste generated by the coffee industry, contributing to improving the general properties of composite materials in terms of mechanical properties.

Data Availability

The experimental data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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