RESEARCH ARTICLE



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Elaeocarpus ganitrus (rudraksha) seeds as a potential sustainable reinforcement for polymer matrix composites

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Abstract

Creating environmentally friendly materials like polymer matrix composites (PMCs) enhanced with natural fillers has grown in recent decades. It supports the United Nations sustainable development goals (SDGs) by reducing the reliance on non-renewable resources, minimizing waste generation, and promoting sustainable land and water management practices. This investigation assesses the viability of employing Elaeocarpus ganitrus (rudraksha) seeds as a filler (rudraksha seed filler [RSF] in environmentally sustainable composite materials. Various loading concentrations and sizes of RSFs were incorporated by hand lay-up into polymer epoxy resin (ER). The effect of filler on the mechanical properties of composites was observed. Thermal properties of RSF/epoxy composites were investigated using thermogravimetric analysis (TGA) and Fourier transform infrared (FTIR). The 10 wt% 100-mesh filler had the maximum tensile strength (TS) at 42.30 MPa and flexural strength (FS) at 87.4 MPa, surpassing other filler sizes and loading concentrations. Meanwhile, the highest impact properties have been achieved with 20 wt% of RSF with 0.436 J. The results of this study highlight the significant impact of filler type and concentration on the mechanical properties of the material. These findings offer valuable insights that may be applied to various industrial contexts. Furthermore, incorporating 10% up to 30 wt% RSF in epoxy composites has brought more thermal stability than virgin epoxy. It revealed that the RSFs possess higher decomposition temperatures than the epoxy matrix. RSF has a potential sustainable reinforcement in polymer composites and holds great promise for addressing environmental challenges and aligning with several SDGs.

Highlights

• Interest in eco-friendly materials, like polymer composites with natural reinforcements, has been increasing in recent decades. Research International Collaboration, Grant/Award Number: 1831-Int-KSln-KLPPM/UNTAR/XI/2021

- Rudraksha sees filler is lightweight with low moisture, high carbon content, and some mineral elements.
- The addition of Rudraksha seed filler has enhanced the tensile, flexural, and impact properties of the composites.
- Rudraksha seed filler possess higher decomposition temperatures than the epoxy matrix.
- Rudraksha seed filler has a promising future as a sustainable friction material for automotive brake pads.

KEYWORDS

Elaeocarpus ganitrus, natural filler, rudraksha, sustainable development goals, sustainable materials

INTRODUCTION 1 1

Polymer composites, composed of a polymer matrix and reinforcing fillers, are frequently utilized in the construction, packaging, consumer products, automotive, and aerospace industries, among other areas.^{1,2} Due to their inherent characteristics, such as those of glass fiber and carbon fiber, common synthetic fillers have restrictions on their impact on the environment and the availability of resources. Moreover, investigating natural fillers as reinforcement in polymer composites has progressed in engineering and material science.^{3,4} Examining the mechanical, thermal, and morphological characteristics of natural-filled composites, as well as improving processing methods to ensure even dispersion and effective load transfer inside the composite structure, have been the main study areas.5,6

Filler size and filler loading are two important variables that profoundly affect the characteristics and performance of polymer composites. For composite materials to be properly tailored to meet application requirements, it is essential to understand the mechanisms by which these constituents interact.^{1,7,8} Furthermore, the size of the filler particles has an impact on the composite's mechanical properties. Fillers that are nanoscale in size have a high surface area-to-volume ratio, which improves molecular reinforcement. As a result, the composite has an increased modulus, stiffness, and tensile strength (TS). Micron-sized fillers can patch cracks in polymer matrixes, enhancing the material's mechanical characteristics and interfacial adhesion. Even though particles may not offer the same level of reinforcement, macro-sized fillers can provide the composite with certain mechanical properties like toughness or ductility.^{7,9-11} Additionally, the amount of filler in the composite has a significant impact on its mechanical properties as well. The mechanical properties of the composite regularly

become stronger since filler dispersion and reinforcement work well at lower filler loadings. Lower filler loadings effectively disseminate the fillers throughout the matrix, which increases stiffness and strength due to the particles' improved capacity to support loads. High filler loading, on the other hand, might induce agglomeration and insufficient interfacial bonding, reducing the material's strength and impact resistance.^{12–14}

Numerous studies have been conducted to determine the effects of natural filler size and loading on the tensile properties of epoxy composites. The tensile properties of composites have been discovered to be significantly influenced by the size and loading of fillers. TS and tensile modulus (TM) generally rise as filler content does.^{15,16} As filler loading rises, tensile strain tends to climb.¹⁷ Additionally, the outcomes reveal that the sizing of the fillers can enhance the composites' mechanical properties.¹⁸ It is important to consider, though, that there might be a point at which additional filler loading increases could lead to a decline in tensile properties.¹⁹ Cionita and coworkers examined how filler affects the flexural properties of an epoxy resin (ER)-reinforced composite made of untreated palm kernel cake filler (UPKCF). The results suggest that the maximum flexural strength (FS) and flexural modulus (FM) peak at 30% filler content before dropping to a minimum at 40% filler content as the filler load concentration increases. With 40% filler loading, the UPKCF/ER composites in FS and FM were lowered by 39% and 23%, respectively. This can be caused by deterioration in the interfacial adhesion between the hydrophobic epoxy matrix polymer and the hydrophilic UPKCF.¹

An investigation carried out by Verma et al. (2020) explores the potential of pyrolyzed carbon black, obtained from used tires, as a filler in epoxy resin composites. The study focuses particularly on the utilization of carbon black in coating materials. The primary aim of this research endeavor is to examine the effects of varying

proportions of carbon infill on the composites' physical, mechanical, and thermal properties. The evaluation of the composites' mechanical properties was conducted through a variety of experiments. Note that the UTS and percentage elongation were determined to be the tensile properties. As a result of the addition of carbon black infill, the UTS increased, according to the study's findings. This observation implies that the enhanced strength can be attributed to the reinforcing effect of the carbon particles. The implementation of carbon black infill led to a progressive increase in the compression parameters, such as the ultimate compressive strength and strain at failure. As the quantity of filler increased, the flexural properties, particularly the flexural strength and flexural modulus, demonstrated a decreasing trend. Additionally, a decrease in both impact energy and intensity was noted in correlation with the increase in filler concentration. With an increase in filler quantity, the hardness of the composites demonstrated a progressive rise. The mechanical properties exhibited by these composites provide significant insights regarding the potential applications and performance attributes of coating materials.²⁰

On the other hand, Marichelvam et al. (2021) investigate the automotive application of a hybrid palm sheath and sugarcane fiber-reinforced ER composite. The objective of the research was to evaluate the performance of the composite material and ascertain the most effective composition for fabricating a dashboard to decrease the weight of the vehicle and improve its fuel economy. The mechanical properties testing findings revealed that the composite material possessing the most favorable mechanical properties was the one consisting of palm and bagasse fiber in a weight proportion of 60:40. In particular, the material exhibited the following characteristics: TS of 19.80 MPa, Young's modulus of 0.953 GPa, an FS of 28.79 MPa, an impact strength of 2 kJ/m^2 , and a hardness value of 38.02 HD. Based on these findings, the optimal sample for the construction of an automobile dashboard was determined.²¹

In 2022, Chandrika et al. investigated the impact strength of sugarcane bagasse ash, madar fiber, and ER composites. Impact energy increased by 22.28% as a result of the 7 wt% increase in the filler material. Furthermore, the impact strength of the madar fiber/ER composites made of sugarcane bagasse ash was 22.28% greater than that of clean madar fiber/ER composites, measuring 17.5 kJ/m² at 7 wt% filler loading. Researchers discovered that composites comprised of madar fiber, ER, and sugarcane bagasse ash were combined to create composites. Since the filler was spread evenly throughout the matrix and the surfaces adhered effectively to one

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another, these composites exhibited a higher impact strength. Hence, the impact strength was decreased due to facile energy absorption and crack avoidance.²²

The filler loading in polymer composites significantly impacts thermal behaviors, as thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) demonstrate. With increased filler content, thermal stability improves since the fillers serve as heat sinks, dispersing heat and delaying degradation. Furthermore, including fillers reduces polymer chain mobility, resulting in a slower TGA degradation rate.²³⁻²⁵ Some fillers can encourage char formation, providing a protective layer during breakdown. Filler loading influences the glass transition temperature (Tg) in DSC, increasing it due to reduced chain motion. Furthermore, fillers influence crystallization behaviors by supplying nucleation sites, modifying crystallinity, and influencing the latent heat of crystallization. Thus, understanding these mechanisms is critical for modifying thermal behaviors in polymer composites for various applications requiring increased thermal stability and specific thermal transitions.^{7,23,26–28}

Jawaid and colleagues investigated the thermal properties of oil palm empty fruit bunch (OPEFB) composites with varied weight fractions of jute fibers. Decomposition of the hybrid composite (OPEFB/jute) started at a higher temperature (263-286°C) than that of the OPEFB composite alone (260°C), and the final degradation temperature ranges from 433 to 463°C. The final decomposition temperature of the composite is increased by hybridizing OPEFB with jute fibers.²⁹ Additionally, the thermal stability of the OPEFB composite improves with increasing jute fiber loading. This is due to jute's significantly superior heat stability compared to OPEFB fibers. As a result of the alkali-treated jute fiber (ATJF) polymer composites' thermal properties from Sajin et al. (2022), the weight loss of the composite occurs between 220 and 330°C due to degradation of the reinforcement material, notably cellulose, and hemicelluloses in ATJF. In contrast, the weight loss occurs at 100°C due to water evaporation from the specimen. In the alkali-treated jute fiber composite (ATJFC) sample, the peak between 340 and 450°C indicates isophthalic polyester (IP) is degrading. In contrast, the weight loss above 460°C indicates that lignin degrades in the ATJF sample. Note that the ATJFC specimen's thermal stability above the processing temperature is indicated by the peak of the DTG curve.³⁰

According to the literature mentioned above, the type, size, loading, surface treatment, and compatibility of the fillers with the polymer matrix may all affect the filler loading's mechanical and thermal properties. The primary objective of this study is to analyze the impact of filler size and filler loading on the mechanical and thermal characteristics of epoxy composites that are

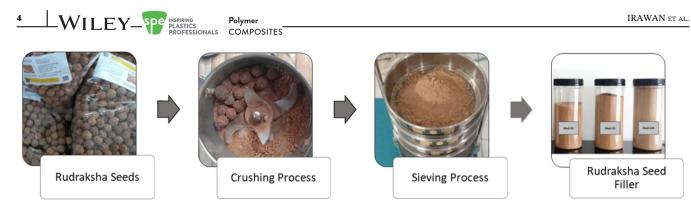


FIGURE 1 Process of rudraksha seeds to rudraksha fillers.

reinforced with a rudraksha seed filler (RSF). Knowledge regarding the properties and practical implementations of rudraksha as a reinforcing agent in polymer composites is noticeably limited. Hence, the scarcity of information highlights the necessity for additional investigation to delve into and clarify the potential of rudraksha to augment the characteristics of polymer composites. The present work encompasses a thorough investigation of the mechanical and thermal characteristics while also introducing a novel use of rudraksha fillers for enhancing the mechanical properties of polymer composites. The incorporation of rudraksha fillers is closely linked to many sustainable development goals (SDGs), underscoring the need for sustainable material development and responsible business practices. The exploration and utilization of rudraksha seeds in an innovative setting hold the potential for researchers and industries to actively contribute toward a future that is both ecologically mindful and socially responsible.

2 | EXPERIMENTAL SETUP

2.1 | Material

Elaeocarpus ganitrus, a species of plant that belongs to the *Elaeocarpaceae* family, is frequently referred to as a rudraksha. Numerous genera and approximately 350 species are spread over Southeast Asia, India, Indonesia, Nepal, China, Fiji, New Zealand, Hawaii, Malaysia, Madagascar, Australia, and New Guinea.³¹ Rudraksha seeds were purchased from a supplier in Semarang, Indonesia. The processing of rudraksha seeds into rudraksha fillers is displayed in Figure 1. Consequently, the seeds were cleaned and rinsed with distilled water before being crushed. Subsequently, the filler was sieved using a sieve shaker with mesh sizes ranging from 30, 50, and 100.

In order to produce the polymer matrix utilized as a binder for the rudraksha filler, it was mixed with the epoxy hardener (651) product from Hexion, United States, in a weight ratio of 3:1. The epoxy and hardener were

TABLE 1 Composition of rudraksha filler, epoxy, and hardener.

	Weight composition, wt%		
Materials	RSF10/ER	RSF20/ER	RSF30/ER
Rudraksha filler	10	20	30
Epoxy resin	152	144	136
Hardener	38	36	34

Abbreviations: ER, epoxy resin; RSF, rudraksha seed filler.

purchased from IZE Solution in Kuala Lumpur, Malaysia. Epikote 828 resin had a viscosity of 12–14 Pa.s at 25°C, a specific gravity of 1.16 at 25°C, a cure duration of 20–24 hours, and a transparent white color.

2.2 | Fabrication of specimen test

The RSF (RSH 30-mesh, 50-mesh, and 100-mesh) with constant loading at 10 wt.%, epoxies, and hardener were blended to produce varying filler sizes for the research study on the influence of filler sizes in epoxy polymer composites. At 100-mesh, filler loadings of 10 wt% (RSF10), 20 wt% (RSF20), and 30 wt% of RSF (RSF30) were employed. Table 1 summarizes the filler, hardener, and epoxy weights for each batch, which had a total weight of 200 g.

In order to maintain homogeneity and avoid bubble formation, the hardener was gradually added to the ER in a 3:1 ratio. The mixture was then agitated with a wooden stick. The liquid still had a few tiny bubbles despite efforts to reduce them. Subsequently, the mixture was stirred for 5 minutes while the filler was gradually added in two to three rotations. Prior to pouring the composite mixture into the mold, air bubbles were removed by vacuuming the mixture for 5 minutes at room temperature (25° C). A homemade funnel built from polyvinyl chloride (PVC) paper cone was used to pour the composite material into the mold to distribute it evenly. The

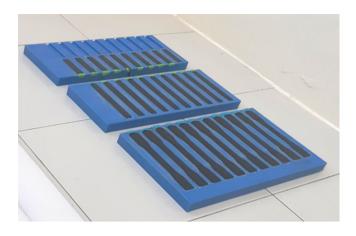


FIGURE 2 Composite specimen test.

composite was then allowed to cure for at least 24 hours at room temperature (Figure 2).

2.3 | Characterization of rudraksha filler

RSF emerges as a new potential filler material for Polymer Matrix Composites (PMCs), and detailed information on its properties is still limited. In the present study, moisture analysis, mass density measurement, x-ray diffraction (XRD) analysis, elemental analysis (EA) with a CHNS(O) analyzer, and inductively coupled plasma mass spectrometry (ICP-MS) were used to evaluate the important properties of RSF.

A moisture analyzer brand AND MX-50, which consists of a weighing and halogen heating device, were used to determine the moisture content using the loss on drying method. 5 g of RSF were weighed and dried at 105°C. The equipment will record the weight loss of moisture until it detects that the weight has remained constant, at which point it will stop automatically. The process takes between 10 and 15 minutes. For this experiment, five measurements with statistical averages were analyzed.

Rudraksha seeds were weighed in air and distilled water to determine their mass density in accordance with ASTM D792 standard.³² A densimeter (Alfa Mirage model MD-300S) was used for this experiment. The measured density was a statistical average value from ten readings. The XRD was used to extract the physical, chemical, and structural properties of the material. Consequently, RSF was analyzed using a Bruker D8 Advance with Ni-filtered Cu-K, radiation (1 = 0.154 18 nrn), 40 kV, and 40 mA. The present method was carried out following procedures similar to those of the previous experiment.³³

EA is an analytical chemistry technique used to evaluate the elemental composition of chemical compounds and their mixtures. The Carbon (C), Hydrogen (H), Nitrogen (N),



Sulfur (S), and Oxygen (O) compositions were determined using the Vario Macro CHNS analyzer. In the present work, the oxygen intake pressure should be between 2.0 and 2.2 bar, and the helium intake pressure should be between 1200 and 1250 bar. The helium flow rate should be 600 mL/ min, and the melt-flow controller (MFC)–thermal conductivity detector (TCD) should also be 600 mL/min. Control combustion tube 1150 and reduction tube 850 were used in the experiment, together with their respective functional software. In this experiment, three samples were weighed and packed in a foil tin. Finally, insert the packing sample on the vario macro cube elementar autosampler. Then, configure the technique based on the type of sample and run the experiment until it is complete.

In this study, it was proposed to measure Sodium (Na), Magnesium (M), Aluminum (Al), Calcium (Ca), Beryllium (Be), and other elements in rudraksha seed with an ICP-MS (ICP-MS 7500, Agilent, US). The procedure for such a technique is similar to that reported by Olalere et al.³⁴ A calibration method was developed using a multi-element standard solution. Five concentrations (0–50 ppm) of the standard solution and a blank of 2% nitric acid (HNO₃) were used to generate a calibration curve. The liquid sample was loaded into the ICP-MS nebulizer and spray chamber. The material was dried, evaporated, atomized, and ionized inside a plasma chamber with several heating zones. The elemental composition of the material was with positively charged ions.

2.4 | Mechanical testing

According to the American Society for Testing and Materials (ASTM D638-14),³⁵ the type 1 tensile test specimen was produced with dimensions of 165 mm overall length and 19 mm width. Instron 3369, a universal testing device with a 50 kN capacity and a crosshead speed of 2 mm/min, was used in this experiment. Note that the flexural testing utilized the same instruments and methods. According to ASTM D790-15,³⁶ 127 mm × 12.7 mm × 3 mm flexural specimens were produced. According to ASTM D256-10,³⁷ the Izod impact test requires a specimen with dimensions of 64 mm × 12.7 mm × 3 mm. For each measurable parameter, a total of seven replicate specimens were evaluated.

2.5 | Scanning electron microscopy

Using a Hitachi TM3030 Plus scanning electron microscope (Hitachi, Tokyo, Japan), the tensile fractures of RSF/ER composites were investigated. Double electrically conductive carbon adhesive tapes were employed to mount the samples

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on the mounting holder for assessments. The samples were initially sputter-coated with a thin palladium coating to prevent surface charge before being mounted on a scanning electron microscopy (SEM) holder. After that, the samples were analyzed under a microscope with 2 K and 5 K times magnification and a 10 kV acceleration tension.

2.6 1 Thermogravimetric analyzer

The Hitachi STA7200 thermogravimetric analyzer (Hitachi, Fukuoka, Japan) was used to assess the thermal stability of ER, RSF, and composite fillers. Each sample comprised 10 mg of powdered material put into an alumina crucible before being heated under strict control in the furnace. The temperature was raised during the experiment at a constant rate of 10°C/min while nitrogen gas flowed at 20 mL/min. Thermal analysis was conducted in the temperature range of 30 to 900°C.

2.7 | Fourier transform infrared spectroscopy

The epoxy and RSF composite samples were run through Thermo Fischer's Nicolet iS50 Fourier Transform Infrared (FTIR) spectroscopic analyzer to investigate the possibilities of forming chemical bonds between the two components. Studies that could be identified in the scientific literature suggest that the IR spectrometer should be set between 4000 and 400 cm^{-1} ¹.

RESULTS AND DISCUSSION 3

Properties of rudraksha 3.1

The mechanical and chemical properties of the RSF, such as moisture content, density, XRD, EA, and ICP-MS, are tabulated in Table 2. The percentage of moisture in the RSF used in this study is around 7.7%. According to earlier studies, fibers made from natural sources typically have moisture contents ranging between 7% and 15%.38,39 It is critical to determine the natural fillers' moisture content before the candidate material is incorporated with epoxy polymer resin to preserve the composite material's performance and effectiveness. Deranged moisture content in the fillers is known to potentially deteriorate the binding efficacy, affecting the composite's material mechanical properties. Additionally, the presence of excess moisture may also interfere with the curing process, affecting the material's consistency in its size and structural stability. Furthermore, excessive water

TABLE 2 Result of rudraksha analysis.

		5	
Type of analysis	Unit value	Results	
Moisture content	%	7.7 (± 0.2)	
Density	g/cm ³	1.052	
		Carbolite/Carbon (C)	
		Hydrogen (H ₂)	
XRD	-	Nitrogen (N)	
		Oxygen (O ₂)	
Elemental	%	Carbon (C): 48.46	
analysis		Hydrogen (H ₂): 6.37	
		Nitrogen (N): 0.54	
		Sulphur (S): 0.08	
		Oxygen (O ₂): 44.5	
ICP-MS	Parts per million (ppm)	Sodium (Na): 11.86	
		Magnesium (Mg): 99.87	
		Aluminium (Al): 33.43	
		Potassium (K): 216.13	
		Calcium (Ca): 627.48	
		Other materials: Not detected (less than 0.1)	
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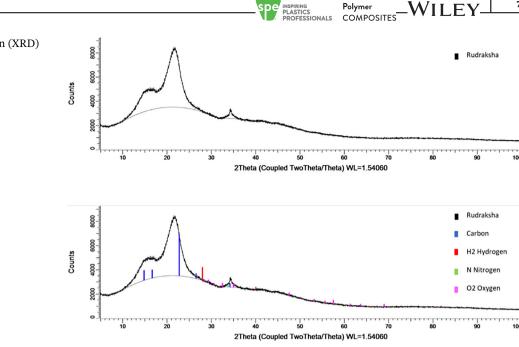
Abbreviations: ICP-MS, inductively coupled plasma mass spectrometry; XRD, x-ray diffraction.

molecules may cause cosmetic damage to the material to an extent, inherently degrading its presentation aesthetically. It is required to dry the filler material at approximately 105°C for 24 hours to ensure that its moisture level is within acceptable range before the mixing can take place.^{40,41}

From analytical data, the rudraksha filler has been reported to have a density of approximately 1.052 g/cm³. Most natural fibers are known to have mass densities between 1.2 and 1.51 g/cm^{3,42} As such, against that of hemp, jute, coir, banana, sisal, kenaf, ramie, pineapple, and cotton fibers, rudraksha has a relatively lighter mass occupying the same volumetric space. This indicates that the RSF has the potential to be used as a lightweight material in the reinforcement of polymers.

Figure 3 illustrates XRD analysis tracing rudraksha chemical elements such as C, H, N, and O. The XRD patterns of activated carbon from rudraksha seeds were mapped to match those of amorphous carbon. They both had two similar broad peaks in the weak diffraction 2 theta range of approximately 23° and 34° (after comparison with the JCPDS 00-050-0926 card). Therefore, the results from the present investigation are consistent with those data reported by other studies that rudraksha

FIGURE 3 X-ray diffraction (XRD) traces of rudraksha.



samples have been found to be broadly peaking at two diffraction angles of 25° and 43° (weak), respectively. This, in turn, suggestively posits the presence of an amorphous carbon element rather than the crystalline carbon.⁴³

Additionally, the XRD results supported an EA that determined the rudraksha's composition to be C (48.46%), H₂ (6.37%), N (0.54%), S (0.08%), and O₂ (44.5%). According to data reported by Gupta (2018), rudraksha has a chemical composition of 50.031% C. 0.95% N, 17.897% H, and 30.53% O.44 This study demonstrated that the chemical compositions of C, H, N, and S are found to be slightly lower than the prior data. In another study, the effects of rudraksha with three, four, and five Mukhi were observed. The findings showed that the content of C and H increased with the introduction of Mukhi rudraksha.⁴⁵ It is noticeable that the origin, type, species, and environment all have an impact on the chemical composition of rudraksha. It is noticeable that the origin, type, species, and environment all have an impact on the chemical composition of rudraksha. Additionally, the findings of the research indicate that rudraksha seeds have significant amounts of carbon and can be classified as carbonaceous materials.

Using an ICP-MS analysis, five different minerals, including sodium, magnesium, aluminum, potassium, and calcium, can be traced in rudraksha. Chlorine, copper, nickel, cobalt, and iron are also commonly found in lower concentrations (less than 0.1 ppm).⁴⁶ Previous research demonstrates that researching the characteristics of rudraksha has the potential for the development of new treatments and medical applications.^{47,48}

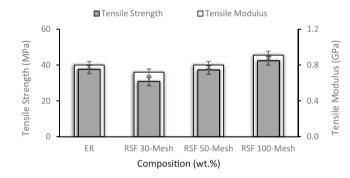


FIGURE 4 Influence filler sizes on tensile properties of rudraksha seed filler (RSF)/epoxy resin (ER) composites.

As reported by the analysis, rudraksha emerges as a significant potential in somewhat broad applications, particularly in the field of polymer composites, owing to its desirable mechanical and chemical characteristics, for instance, the low moisture and high carbon contents, lightweight and the compositional presence of some mineral elements.

3.2 | Tensile properties

Figure 4 illustrates the mechanical properties of TS and TM in polymer composites impacted differently when different filler sizes, such as 30-mesh, 50-mesh, and 100-mesh, were tested. According to the findings, out of the three filler sizes tested, the composite with small particles of 100-mesh had the highest TS with 42.30 MPa. This result can be explained by the larger surface area and effective load transmission of the smaller particles.

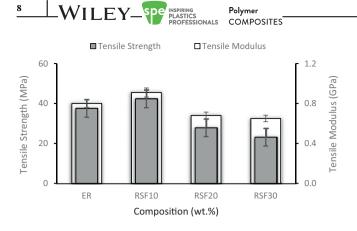


FIGURE 5 Influence filler loading tensile properties of rudraksha seed filler (RSF)/epoxy resin (ER) composites.

This results in improved stress distribution and a reinforced polymer matrix. Note that 30-mesh and 50-mesh were bigger particles with lesser TSs of 30.80 MPa and 37.20 MPa, respectively. This might result from ineffective filler-matrix interactions, agglomeration, or void formation, which leads to stress concentration and reduced overall mechanical properties.49,50 The composite with 100-mesh fine particles displayed the highest TM (0.91 GPa), similar to TS. Moreover, a composite's stiffness and modulus were raised as a result of the small particles' efficient restriction of polymer chain mobility. In contrast, the coarser fillers (30-mesh and 50-mesh) demonstrated lower TM due to their diminished capacity to limit polymer chain motion and less efficient reinforcing.^{51,52} In summary, the comprehensive examination of various filler sizes inside polymer composites yields a nuanced comprehension of the intricate relationship between particle size and mechanical properties. The increased mechanical capabilities seen in the composite containing 100-mesh small particles underscore the significance of optimizing filler qualities in polymer composites. This finding holds potential for use in diverse engineering and structural applications.

The tensile properties of ER reinforced using RSF at different weight concentrations are illustrated in Figure 5. The graph shows virgin ER has a TS and TM of 37.5 MPa and 0.8 GPa, respectively. The TS and TM of ER were increased to 42.30 MPa and 0.91 GPa, respectively, by adding 10 wt.% of RSF. The RSF raises the TS and TM materials by about 12.8%, respectively. Additionally, the epoxy matrix can act as reinforcement with the use of fillers. RSF can enhance load transfer to the filler particles embedded in an epoxy matrix. During loading, stresses are dispersed throughout the composite, and infill particles can help with both distribution and absorption. As a result, the composite's TS may be improved, and the distribution and absorption of stresses during loading may be improved. This may also decrease

the incidence of microcracks and cavities inside the composite. Due to the composite's improved resistance to deformation and failure under applied forces, this can boost TS.

Figure 5 portrays that the TS and TM of composites decrease when the filler concentration rises from 10% to 30%. The TS and TM of the composites are decreased to 23.10 MPa and 0.65 GPa, respectively, when the maximum RSF content of 30% is reached. The possibility that adding filler could lower TS and TM must be considered while designing and optimizing composite materials. Hence, understanding the root causes of this issue and observing the possible mitigation or balance measures are crucial. An increase in the filler content of composites can cause a decrease in their tensile characteristics as a result of the production of agglomerates and clumps of filler particles.^{53,54} Moreover, high filler concentrations can cause the particles to over-compact, reducing the area of the matrix-filler interface and impairing interfacial bonding. This could decrease the stiffness and strength of the filler particles by reducing the load transfer from the matrix to them.²⁰

On the other hand, Shakuntala et al. explored how filler loading affected the mechanical properties of an epoxy composite reinforced with wood apple shells. Composites' TS increased from 5% to 15% of filler content. However, it deteriorated when the filler level was increased to 20%. Therefore, the author concludes that a high filler content makes it difficult for the polymer to reach the fillers' contracting gaps, resulting in inadequate coverage and reduced stress transfer across the filler-resin interface.¹⁴

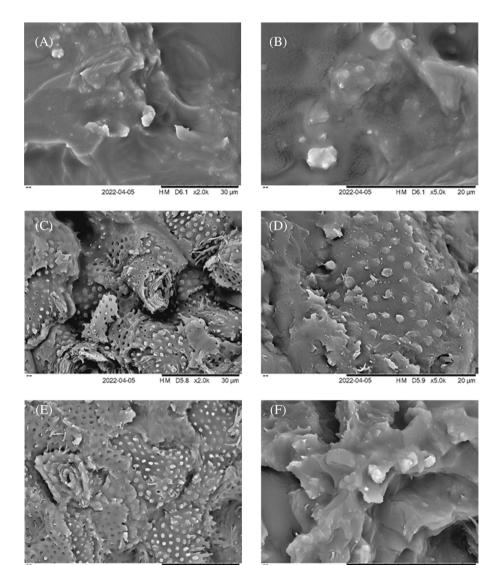
Table 3 compares several filler types investigated in earlier studies and employed in ER. The maximum improvement in TS and TM in composites depends on the filler material concentration, with a weight of 30% being the highest filler loading. However, this study discovered that an RSF10 concentration significantly improved TS and TM. The TS and TM values discovered in this investigation are within the acceptable range suggested by earlier studies. Thus, the TS and TM values can be affected by the type of filler, the matrix, and the proportion between the filler and the matrix.^{20,21}

SEM analysis is a useful tool for examining the microstructure of composites and how filler dispersion affects the mechanical properties of the composite, particularly its TS. Figure 6 displays the fracture surface of RSF loaded with ER at $1000 \times$ and $5000 \times$ magnifications. The distribution of 10% filler within ER is depicted in Figure 6A,B. The amount of porosity and voids in the composite can serve as areas where tension can build up and eventually fail. Therefore, a good filler dispersion can also assist in lowering these characteristics. Furthermore, TABLE 3 Comparison of tensile properties with previous studies.

Filler and matrix	Corresponding filler content (wt%)	Tensile strength (MPa)	Tensile modulus (GPa)	Reference
Coconut shell-epoxy	20	30.60	0.85	55
Wood apple shell– epoxy	15	45.60	_	14
Ipamoea–epoxy	30	23.75	7.20	56
Orange peel–epoxy	20	25.85	_	57
RSF-epoxy	10	42.30	0.91	Current study

Abbreviation: RSF, rudraksha seed filler.

FIGURE 6 Scanning electron microscopy (SEM) images of the tensile fractured surface of rudraksha seed filler (RSF)/epoxy resin (ER) composites; A-B 10 wt%; C-D 20 wt% and E-F 30 wt% of filler.



2022-04-05 HM D5.1 x2.0k 30

22-04-05 HM D5.1 x5.0k 20 µm

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a more uniform stress distribution throughout the composite and higher TS results from achieving homogeneous dispersion of fillers, lowering the likelihood of voids and porosity appearing.^{58–62} A proper filler dispersion in the epoxy matrix can increase the composite's TS by enhancing load transfer to the filler particles. The RSF10 discovered in the TS and TM samples is supported by this investigation.

Figure 6C–F depict the SEM image of the higher filler concentrations (20 and 30 wt%). When filler

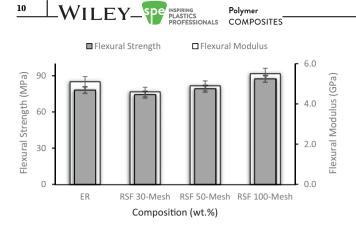


FIGURE 7 Influence filler sizes on flexural properties of rudraksha seed filler (RSF)/epoxy resin (ER) composites.

concentrations in epoxy are raised, particle distribution can become more difficult. Since the mixture has more filler, it could be more difficult to distribute the particles uniformly throughout the matrix. As a result, filler particle agglomeration may happen, resulting in some composite sections having a higher particle concentration than others.^{63–65}

3.3 | Flexural properties

Figure 7 demonstrates the influence of several RSF mesh sizes, specifically 30-mesh, 50-mesh, and 100-mesh. ER has an FS of 78.1 MPa and an FM of 5.1 GPa. TS and TM are marginally reduced when RSF 30 and RSF 50-mesh are added. Surprisingly, adding RSF 100-mesh enhanced TS and TM to 87.4 MPa and 5.5 GPa, respectively. The increase in RSF 100-mesh over RSF 30-mesh is around 18% and 19.5% for TS and TM, respectively. This finding can be due to the smaller particles' greater surface area and improved filler-matrix interaction. The finer particles bridged the gaps in the polymer matrix, resulting in more efficient load transfer and increased resistance to bending forces.^{8,63} Conversely, composites with coarser fillers (30-mesh and 50-mesh) had lower FS, which could be attributed to the existence of voids, agglomeration, reduced interfacial adhesion, and limited load-bearing capability.66,67

Additionally, the fine particle composite (100-mesh) had the highest FM. The finer particles significantly reduced the mobility of the polymer chains, resulting in improved stiffness and rigidity in the composite material. Composites with coarser fillers (30-mesh and 50-mesh), on the other hand, have a lower FM, indicating an inferior resistance to deformation under bending forces.^{29,68}

Figure 8 illustrates that the amount of filler in a composite affects its mechanical properties, including its FS and FM. This investigation determined that, compared to

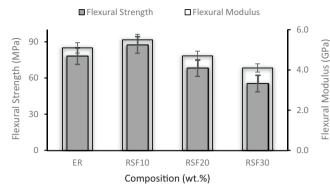


FIGURE 8 Influence filler loadings on flexural properties of rudraksha seed filler (RSF)/epoxy resin (ER) composites.

filler concentrations of RSF20 and RSF30, RSF10 improved these attributes the most. One explanation for this outcome can be the impact of filler loading on the interfacial bonding between the filler and the matrix. According to earlier studies, excessive filler loading can cause agglomeration and reduced interfacial area between the filler and the matrix, leading to a weaker interface and diminished mechanical properties.⁶⁹⁻⁷¹ On the other hand, a lower filler loading enables a more uniform dispersion of the filler particles in the matrix, producing a stronger interface and improved mechanical properties. This finding is consistent with earlier research that evaluated the eggshell filler affected the tensile and flexural characteristics of glass fiber-reinforced polymer composites. Moreover, the increased filler loading causes a decrease in FS and FM. When eggshell filler, ranging from 10% to 40%, is applied, the slope of the flexural stress and strain curve is drastically reduced. This indicates that the modulus falls off as the filler content rises. Compared to samples with 25% and 40% eggshell filler, the composite sample with 10% can tolerate more bending stress.⁷²

The results of this study suggest that the most significant improvements in FS and FM can be obtained in composites with a moderate filler concentration, such as 10 wt%. This study demonstrates the importance of modifying the filler concentration in composite materials for improved mechanical properties. This is consistent with previous research regarding the effect of filler development on mechanical properties. According to most researchers, the sizes and loads of filler can considerably affect the flexural properties of epoxy composites.^{11,19,69,73}

3.4 | Impact strength

As displayed in Figure 9A, the effects of three filler sizes in polymer composites of 30-mesh, 50-mesh, and

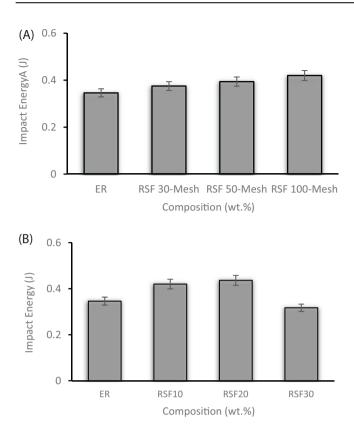


FIGURE 9 (A) Influence filler sizes and (B) filler loadings on the impact strength of rudraksha seed filler (RSF)/epoxy resin (ER) composites.

100-mesh revealed some interesting things about how the impact strength behaved. Furthermore, Figure 9B illustrates that investigating the effect of various filler loadings (10 wt%, 20 wt%, and 30 wt%) provided a better understanding of the composite's reaction to altering filler content. It is clear that the composite with 100-mesh filler particles had the highest impact strength of the three filler sizes tested, measuring 0.420 J, followed by those with 50-mesh and 30-mesh filler particles, measuring 0.394 J and 0.375 J, respectively. There are several important reasons why the fine particle type, specifically the 100-mesh RSF, has better impact performance. To begin with, the increased surface area of tiny particles facilitates enhanced stress distribution inside the composite material under impact loading conditions. The enhanced surface area facilitates enhanced contact with the epoxy matrix, promoting more effective load transmission and dispersion of stress. Additionally, the fact that small particles are less likely to stick together than larger ones means that the RSF is spread out more evenly in the epoxy matrix, which improves its resistance to impact. The enhanced toughness of materials can be attributed to the reduced dimensions of fine particles, which enable them to efficiently absorb and disperse impact energy, hence impeding the spread of cracks. Fine



particles may also have a stronger reinforcing effect on the epoxy matrix because of their smaller size. This makes the matrix stronger and more resilient when it is hit. The occurrence of fewer voids during the curing process, which is linked to the presence of small particles, results in a composite structure that is more compact and uniform. This, in turn, contributes to improved impact performance. In conclusion, better compatibility between the tiny particles and the epoxy matrix leads to stronger adhesion at the filler-matrix interface. This makes it easier to create a cohesive and integrated structure that improves the resistance to impact.^{74,75}

Filler loading of RSF in polymer composites substantially impacted impact strength. Surprisingly, the composite with an RSF 20 wt% filler loading attained the highest impact strength (0.436 J) among the evaluated loadings (RSF 10 wt%, RSF 20 wt%, and RSF 30 wt%). This indicates the presence of optimal filler content, which results in the greatest impact resistance. Lower loadings (10 wt%) may not offer adequate reinforcement, whereas larger loadings (30 wt%) may cause agglomeration or reduced interfacial adhesion, lowering impact strength.^{76,77} The best impact performance was achieved with fine particles (100-mesh) and a 20% filler loading. This indicates how important filler size and loading are for getting the desired impact resistance in the composite.^{1,78–80}

Previous researchers investigated how filler loading affected the impact strength of epoxy composites reinforced with natural filler. The researchers discovered that, up until a certain point, increasing filler loading initially improved impact strength. However, after that point, it lowered impact strength.^{23,69,81,82} Researchers explained this behavior by explaining that the matrix changed from ductile to brittle at greater filler loadings, which caused an impact strength reduction.^{78,83,84} Moreover, the findings of this investigation indicate that composite materials with the greatest impact strength can be produced with a modest filler concentration, such as 20 wt%. Therefore, this study demonstrates how crucial it is to adjust the filler concentration in composite materials to provide the optimum mechanical properties, which is consistent with earlier research on how filler loading influences impact strength.^{3,4}

The results of this investigation challenge accepted theories regarding the relationship between filler size and impact strength in polymer composites. The fact that fine particles outperformed coarse particles in impact resistance demonstrates how different filler size ranges might contribute to various mechanical behaviors and energy-dissipating mechanisms.^{19,73,85} Hence, these findings significantly affect the composite material design, especially where impact resistance is an important consideration.

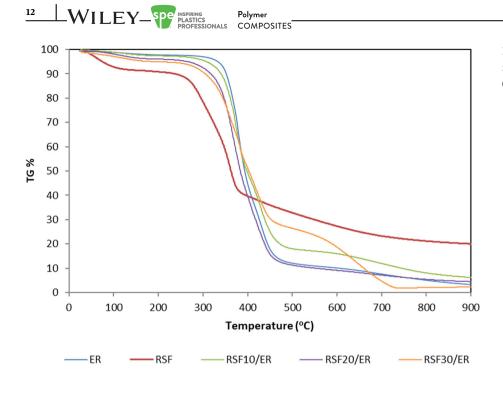


FIGURE 10 TGA curves of rudraksha seed filler (RSF)/epoxy resin (ER) composites.

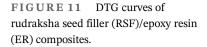
Furthermore, the significance of proper optimization is highlighted by finding the best filler loading (20 wt%) for the highest impact strength. This will help provide the mechanical properties that are required.

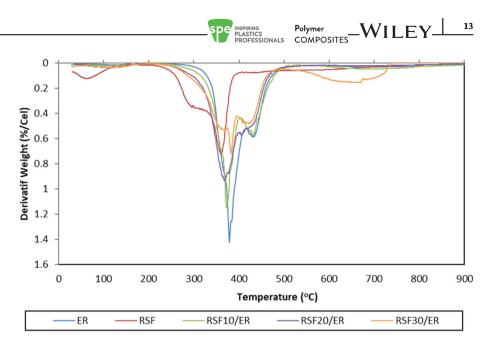
3.5 | Thermogravimetric analysis

Natural filler-reinforced polymer composites are rising in importance in various industries due to their controlled features and superior performances compared to virgin polymers. Fillers included in polymer matrices can substantially impact the resulting composites' thermal properties, making them appropriate for applications requiring greater thermal stability. Additionally, TGA is used in this work to explore the thermal stability of polymer composites with varying filler loadings (RSF 10 wt%, RSF 20 wt%, and RSF 30 wt%) at a constant 100-mesh size. Figures 10 and 11 display the TGA and corresponding derivative thermogravimetric (DTG) derivative curves. The virgin ER experiences an initial weight loss of 1.2% at 100°C, significantly less than the pure RSF, which experiences a weight loss of 7.13%. The evaporation of the water in the RSF fills the cause of this variation.^{86,87} It is clear from analyzing the filler incorporation in the epoxy composite that RSF10 wt% exhibits the least weight loss compared to the other two loading levels. Note that the weight loss for RSF10 is specifically 1.14%, whereas weight losses for RSF20 and RSF30 are 1.9% and 2.7%, respectively. These results reveal that weight loss decreases with filler loading, with RSF10 wt% demonstrating the best thermal stability among the three loading levels. Therefore,

these findings also agree with those observed by other researchers, who reported that the increase in filler content decreased the thermal stability of the composites.^{1,88}

The TGA curve demonstrates that at the highest temperature of 900°C, the char residue for ER was 3.7%. However, the char residue for RSF was substantially higher at 20.03%. This result demonstrates the RSF's significant char-forming capabilities. A big char layer that forms during thermal breakdown indicates that RSF has outstanding thermal stability, as evidenced by the high char residue. Moreover, the char residue of composites containing RSF10 is 10.79%, 4.5% for composites containing RSF20, and 2.33% for composites having RSF30. The findings indicated that a moderate filler content facilitates the formation of a significant char layer during thermal degradation. This is considering that, compared to the composites with the other two loadings, those with RSF10 had greater char residue. Here, the 30 wt% of filler composites had the least char residue. A large amount of filler could prevent a consistent char structure from forming. A perfect filler loading may maximize the char-forming capacity, as evidenced by the composites with RSF20 indicating an intermediate char residue. Furthermore, previous studies have proposed that a lower filler loading can improve the thermal stability of the matrix by slowing down thermal deterioration and increasing char generation during combustion. Conversely, high filler loading may induce matrix deterioration and lower thermal stability values.^{86,89} Hence, these findings have significance for developing polymer composite materials for hightemperature applications where maintaining structural integrity and fire resistance meet critical requirements.





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Figure 11 illustrates the second stage of weight loss of the composite materials, between 200 and 600°C, which was implicated in the chemical composition breakdown in RSF/ER composites. According to Viswanathan et al. (2021), the composition of rudraksha contains 50.031% carbon, 0.95% nitrogen, 17.897% hydrogen, and 30.53% oxygen.⁹⁰ T_{max} is the temperature at which maximal weight loss occurs during degradation and is connected to the maximum decomposition temperature, both of which are essential indicators of the thermal stability of the materials.^{1,91,92} The maximum decomposition temperatures of virgin epoxy and pure RSF were 370°C and 353°C, respectively. Meanwhile, the deterioration peak of RSF10, RSF20, and RSF30 occurred at temperatures of 372.56°C, 375.10°C, and 383.20°C. From the analyzed data obtained, it can be concluded that adding filler has increased the thermal stability of the composites.

3.6 | Fourier-transform infrared spectroscopy

The utilization of Fourier-transform infrared (FTIR) spectroscopy, which can examine the hydrogen-bonded region between 3550 and 3200 cm⁻¹ spectra sweep, as depicted in Figure 12, is crucial for identifying and characterizing chemical compounds. This spectral band focuses mostly on the stretching vibrations of the hydrogen-bonded functional groups, particularly the hydroxyl groups (OH), which are prevalent in various organic compounds. The OH stretch is a strong, broad band often detectable between 3400 and 3300 cm⁻¹, and it offers crucial underlying information on the hydrogen bonding environment and other functional groups in its surroundings. The

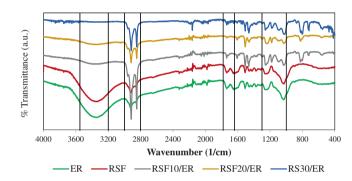


FIGURE 12 Fourier-transform infrared (FTIR) of epoxy, rudraksha filler, and composites.

finding agrees with the data reported in prior work on the utilization of natural filler in composite materials.^{93,94} FTIR spectral analysis further reveals that alcohol groups with a broad and strong intensity (i.e., secondary CH–OH) also showed up at 3328.45 cm^{-1.44}

The stretching vibrations of C—H bonds in organic compounds, such as epoxies, are related to the region between 3000 cm⁻¹ and 2800 cm⁻¹ in an FTIR spectrum.95 The characteristic peak of virgin epoxy is found at 2923 cm^{-1} and 2853 cm^{-1} , respectively. The peak levels of the spectrum for pure rudraksha filler were observed at 2916 cm⁻¹ and 2853 cm⁻¹. Previous studies also reported that sharp and strong intensities of varying functional groups, including alkane, methyl, methylene, ethyl, and alkene, were found in a similar spectrum band to that of rudraksha, at 2925.28 cm^{-1} and 2854.35 cm^{-1} , respectively (Gupta, 2018). Identical characteristics were discovered for epoxy and rudraksha composites made with various concentration loadings. RSF10 has two peaks at 2916 and 2849 cm^{-1} , RSF20 has two peaks at 2918 and 2850 cm^{-1} , and RSF30 has two

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peaks at 2916 and 2848 cm⁻¹, respectively. By analyzing the FTIR data across the 3000–2800 cm⁻¹ spectral band, researchers would be able to further classify the many types of C—H bonds that are present, distinguishing aliphatic and aromatic compounds, and ultimately develop a new body of knowledge concerning the molecular structure and functional groups contained in rudraksha samples. Owing to its versatility, the spectra offer indispensable groundwork for the analysis of a variety of organic compounds, including hydrocarbons, polymers, and other organic functional groups, posing it as a crucial instrument in chemical inquiry and elaborative material characterizations.^{96–98}

The carbonyl region, also known as the C=O stretching region, refers to the region of the FTIR spectrum between 1780 and 1640 $\text{cm}^{-1.99}$ The information in this spectral band is crucial for understanding compounds with carbonyl functional groups, such as aldehydes, ketones, carboxylic acids, esters, and amides. Previous work has revealed that the pure rudraksha filler content of these functional groups can be identified in the region of the dominant spectrum corresponding to 1746.39 cm^{-1} (Gupta, 2018). The findings from the present work indicate that the spectrum of pure rudraksha filler and the mixture with ER reaches its peak performance level at 1735 cm⁻¹. The absorption peak of virgin epoxy detected at around 1653 cm-1 could represent various functional groups or their respective structural features. The Amide I band, which frequently occurs in the area of similar range and is especially pronounced in proteins as a result of the C=O stretching vibration of the peptide bond, is one critical explanation that may be relevant to this phenomenon as observed by authors.^{100–102}

Respective wavenumbers of 1300 cm^{-1} and 1000 cm^{-1} indicate specific molecular vibrations and functional groups in the material being tested. The vibrations produced by C-H bending in alkanes or C-N stretching in amines fall within the range of 1300 cm^{-1} .¹⁰³ It showed that the peak wavenumber of epoxy was at 1253 and 1030 cm^{-1} , and that of rudraksha was at 1255 and 1030 cm^{-1} , respectively. At a wavenumber of 1240.54 cm⁻¹, a functional group discovered in rudraksha, such as carboxylic acids (COOH), can be identified. Additionally, this region can reveal the presence of specific functional groups, such as the amide III band in proteins. It could be an indicator of vibrations such as C-O stretching in carboxylic acids, esters, and anhydrides, as well as C–C and C–O bending in diverse compounds.^{104–106} The inclusion of rudraksha as a filler in epoxy composites demonstrates a high-level wavenumber of RSF at 1255 and 1029 cm^{-1} , RSF20 at 1236 and 1030 cm⁻¹, and finally RSF30 at 1259 and 1020 cm^{-1} , respectively.

4 | CONCLUSIONS

Elaeocarpus ganitrus (rudraksha) seeds have been investigated as a potential sustainable reinforcement for PMCs, with promising findings. The mechanical and thermal properties of composites with various rudraksha filler sizes and loadings were successfully investigated in the study. Moreover, the results reveal that using small particles improved the mechanical properties of the composites significantly. Notably, when compared to other compositions, the insertion of RSF10 in the epoxy composite resulted in the greatest enhancement in mechanical properties and thermal stability. Finally, including rudraksha as a sustainable reinforcement for polymer composites, particularly with small particles at 10 wt%, demonstrates its promise for enhancing eco-friendly and high-performance materials. According to the findings of the exploration on thermal stability. RSF has a promising future as a sustainable friction material for automotive brake pads. This study adds to the pursuit of sustainable and responsible practices in the materials sector and progress towards several SDGs aimed at constructing a more ecologically conscious and socially responsible future.

AUTHOR CONTRIBUTIONS

The article reflects the collaborative efforts of Agustinus Purna Irawan, Januar Parlaungan Siregar, Tezara Cionita, Deni Fajar Fitrivana, Rusivanto Rusivanto, Jamiluddin Jaafar, Pungkas Prayitno, Rifky Ismail, Athanasius Priharyoto Bayuseno, and Ayub Ahmed Janvekar. Agustinus Purna Irawan led conceptualization and supervision, Januar Parlaungan Siregar handled the original draft and investigation, and Tezara Cionita contributed to methodology and investigation. Deni Fajar Fitriyana conducted formal analysis and data curation, while Rusiyanto Rusiyanto provided support in methodology and resources. Writing, review, and editing were undertaken by Jamiluddin Jaafar and Rifky Ismail, with validation. Pungkas Pravitno contributed to visualization and resources, and Athanasius Priharyoto Bayuseno and Ayub Ahmed Janvekar participated in formal analysis and validation. All authors have approved the published version of the article.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

We ensure that all data are contained within the article.

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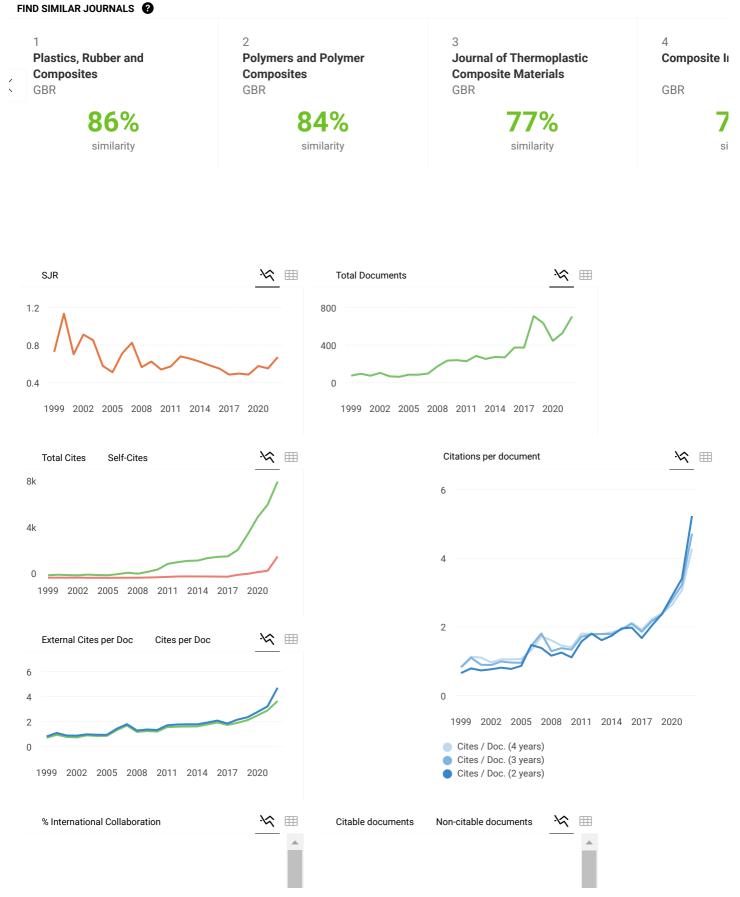
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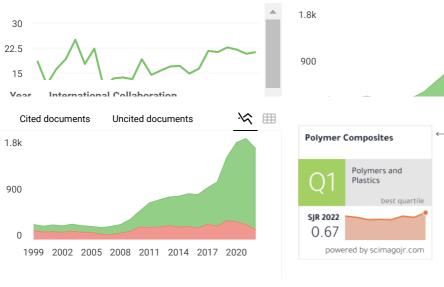
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