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RESEARCH ARTICLE

PHYSICO-MECHANICAL AND MORPHOLOGICAL ANALYSES OF ACTIVATED CARBON/CLAY REINFORCED RECYCLED POLYPROPYLENE COMPOSITES

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Abstract. Plastic waste poses a significant environmental challenge due to its non-biodegradable nature and increasing global waste volume. Recycled polypropylene/clay (rPP/clay) composites typically exhibit inferior mechanical properties compared to virgin materials, limiting their practical applications. However, rPP/clay composites are still being investigated further due to their potential to enhance interaction with natural materials by improving their hydrophilic nature, making them valuable in construction applications. This study aims to enhance the properties of rPP/clay composites by incorporating activated carbon (AC). The rPP/clay/AC composites were prepared using a single screw extruder with varying AC contents (1 wt%, 7 wt%, 15 wt%, and 20 wt%). The physical and mechanical properties were evaluated, including density, flexural strength, flexural modulus, hardness, and morphological analysis. Results indicated that composite density varied with AC content, achieving a balance between mechanical strength and density. The composite with 7 wt% AC exhibited the highest flexural strength (63.04 MPa) and modulus (2.95 GPa), enhancing stiffness and resistance to bending. The added AC reinforced the polymer matrix, supporting a higher load-bearing capacity before failure. Morphological analysis showed no rupture under the flexural test for the composite with 7 wt% AC, indicating strong interfacial bonding and uniform distribution of AC within the polymer matrix. The FESEM images revealed a shear-yielding mechanism, contributing to the material's enhanced toughness. At 20 wt% AC, the hardness reached 75.83, the highest value observed, indicating that higher AC percentages improve hardness, making the composite more resistant to indentation and surface deformation. These results suggest that AC/clay-reinforced plastic waste composites could be valuable for diverse applications, contributing to waste reduction and recycling efforts aligned with sustainable development goals.

Keywords: Recycled polypropylene, clay, activated carbon, plastic composite aggregates.

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1. INTRODUCTION

Plastic waste aggregate (PWA) and plastic composite aggregate (PCA) have emerged as innovative solutions to address the growing issue of plastic waste disposal while contributing to sustainable construction practices [1]. PWA refers to aggregates made from plastic waste, either in its pure form or blended with other materials, while PCA consists of plastic waste combined with reinforcements like natural fibers, minerals, or fillers [2,3]. These aggregates have been incorporated into construction materials such as lightweight concrete and paving blocks due to their potential to enhance durability, reduce weight, and improve sustainability [4,5]. PWAs and PCAs are significant as they mitigate environmental concerns associated with plastic waste and offer a cost-effective and energy-efficient alternative to traditional construction materials [3].

Recycled polypropylene (rPP) is a widely used thermoplastic polymer derived from post-consumer or industrial waste. As a construction material, rPP is valued for its lightweight nature, high impact resistance, and chemical durability. Previous studies have demonstrated that rPP, when used in composite form, can enhance the mechanical properties, including compressive and flexural properties [6]. On the other hand, rPP acts as a good binding material, which also has a lot of scope in the construction fields. Reinforcing the rPP with fillers like wood, glass fibers, other plastic blends further can enhance the mechanical properties [7]. Moreover, rPP demonstrates promising characteristics, its hydrophobic nature and limited compatibility with other materials pose challenges that require further investigation. These limitations can be addressed by improving rPP's properties by adding natural fillers and reinforcements, such as clay and AC. While previous studies have demonstrated the benefits of using natural fillers in polymer composites, there is a lack of research focusing on the combined effects of clay and AC as hybrid fillers in rPP matrices. Previous research has shown that hybrid reinforcements combine two or more different types of fillers are more effective as they offer a range of properties that cannot be achieved with a single type of reinforcement [8].

Kaolin clay, a naturally occurring mineral rich in kaolinite, is widely recognized for its diverse industrial applications due to its exceptional properties and adaptability. In construction, its calcined form, metakaolin, is prominently utilized as a pozzolanic material in the production of geopolymer concrete, where it enhances durability, strength, and resistance to chemical attacks [9,10]. Beyond construction, kaolin clay is widely valued in the plastics industry as a high-performance filler. Its unique characteristics, such as lightweight, smooth surface finish, and cost efficiency, make it an attractive choice for manufacturers [10]. Moreover, kaolin clay contributes to the mechanical enhancement of polymer composites by improving their stiffness, strength, and impact resistance. This dual functionality across industries underscores its importance as a material with diverse applications and significant benefits. When combined with PCA, kaolin can improve the interfacial bonding between the plastic and cement matrix, resulting in better strength and reduced permeability. Previous studies have shown that kaolin-reinforced PCA composites exhibit increased compressive strength and reduced porosity [3]. However, drawbacks such as limited compatibility with hydrophobic plastics and the brittle nature of kaolin-modified composites highlight the need for optimization.

Activated carbon (AC) is a stable carbonaceous material characterized by its well-defined hierarchical microporous structure, high surface area, large pore volume, and tunable surface chemical properties [11-13]. The pores on its surface can enhance the contact area with polymers, creating a strong bond between the filler and the matrix. Using AC as a filler can inhibit crack propagation, thereby increasing the composite's toughness [13]. Additionally, AC-based composites are notable for their unique surface properties, excellent thermal and mechanical stability, and superior electrical performance [14,15]. Depending on the desired material properties and application, AC can be combined with various organic or inorganic materials [16]. Not only does AC serve as an effective filler in plastic composites, but the inclusion of AC as a filler in concrete influences the properties of concrete by enhancing its thermal stability and reducing water absorption, thereby improving durability and longevity. Previous studies show that adding AC to the concrete improves the concrete's flexural strength and compressive strength [17], higher workability, density, compressive strength, splitting tensile strength and resistance to water penetration when optimal amount is added [18]. However, there is a lack of research on using AC as a hybrid filler with clay in plastic composite aggregates for concrete application.

Despite these benefits, current research on rPP composites has typically investigated single filler systems, leaving the synergistic effects of hybrid fillers specifically clay and AC underexplored. Moreover, prior studies have largely focused on standard filler contents, with several identifying 8 wt% AC as the optimum level for mechanical improvement [19]. However, there is little insight into how higher AC contents influence composite performance, particularly when coupled with kaolin clay.

In addition, while processing conditions such as mixing temperature and shear rate are known to affect filler dispersion and composite morphology, these parameters are rarely studied in tandem with hybrid filler optimization. Therefore, this study aims to fill these critical gaps by evaluating the physico-mechanical and morphological properties of rPP composites reinforced with varying concentrations of clay and AC beyond the commonly used 7 wt%. Additionally, it investigates the effect of controlled processing parameters, specifically temperature and mixing speed, to optimize performance. The outcomes of this work are expected to contribute to the development of next-generation sustainable composites for structural applications.

2. MATERIALS AND METHODS

2.1 Raw Materials and Preparation

Recycled polypropylene (melting point: 130 °C; density: 0.92 g/cm^3) was obtained from San Miguel Yamamura Plastic Film Sdn. Bhd. Metakaolin clay was supplied by Edutech Supply & Service, and AC was sourced from R&M Chemicals. The recycled polypropylene (rPP) pellets were manually blended with 1% clay and varying weight percentages of AC as shown in Table 1. This rPP/clay/AC-X blend, where X represents the percentage of AC, was processed using a Cincinnati Extrusion GCE 30T single screw extruder at 185 °C and a screw speed of 50 rpm, then cut using a palletizer. The resulting pellets were dried in an oven at 80 °C for 24 hours. These dried pellets were then compressed using a hot press (Laboratory Press Model GT7014-A) at a molding temperature of 200 °C and a pressure of 140 kg/cm^2. The hot press process took 60 minutes, including 15 minutes each for preheating and cooling and 30 minutes for compression. Finally, all samples were left at room temperature for at least 24 hours to be conditioned before being subjected to further testing and analysis.

Table 1: Formulation table for rPP/clay/AC composite

Sample	rPP (wt%)	Kaolin clay (wt%)	Activated Carbon (wt%)
rPP/clay/AC0	99	1	0
rPP/clay/AC1	98	1	1
rPP/clay/AC7	92	1	7
rPP/clay/AC15	84	1	15
rPP/clay/AC20	79	1	20

2.2 Density Measurement Test

The density of the rPP/clay/AC composite samples was measured using an Electronic Densimeter (MD-MD3002, AlfaMirage, Japan). These measurements followed ASTM D792 standards and were based on the Archimedes principle. For accuracy, at least five measurements were taken for each sample to determine the average value.

2.3 Flexural Testing

The flexural properties were assessed using a Shimadzu AGS-X Series Universal Testing Machine in accordance with ASTM D790 standards. The tests were conducted at a 1.28 mm/min crosshead speed and room temperature. The average values were calculated from five samples.

2.4 Hardness Testing

A durometer Shore D hardness tester (Teclock Model GS-702G) evaluatedthe hardness of the rBOPP-based compounds in accordance with ASTM D2240. The hardness of the 6 mm thick sample was measured using the Durometer Hardness Teclock Model GS-702G (Shore D). To achieve the 6 mm thickness, two 3 mm samples were stacked together. Each sample was tested five times, and the average value was calculated to ensure greater confidence in the data.

2.5 Field-Emission Scanning Electron Microscopy (FESEM)

FESEM using a Zeiss EVA 50 instrument was employed to examine the fracture morphology of the flexural test samples. The FESEM analysis was conducted at an accelerating voltage of 5 kV in secondary electron mode with 100x, 300x and 1000x magnification. Before FESEM observation, the sample surfaces were cut and gold-coated to improve electron reflection during imaging.

3. RESULTS AND DISCUSSION

3.1 Density

The density measurements of rPP/clay/AC composites as shown in Table 2 demonstrate a significant influence of AC content on the overall material density. An initial increase in density was observed with increasing AC concentrations, reaching a maximum of 0.9580 g/cm³ for the composite containing 15 wt% AC. This enhancement can be attributed to AC's relatively higher intrinsic density than the base rPP/clay matrix and its ability to occupy voids, leading to improved packing efficiency within the composite structure. Correspondingly, the relative density difference increased incrementally, peaking at 0.0533 for the 15% AC sample. However, at 20 wt% AC, the density decreased slightly to 0.9305 g/cm³, with a reduced relative density difference of 0.0230. This reduction is likely due to the agglomeration of AC particles at higher loadings, which disrupts uniform dispersion and creates microvoids within the matrix. Such particle clustering diminishes interfacial bonding and packing density, resulting in a less compact structure. These observations are corroborated by morphological analyses, which revealed increased void content in composites with elevated AC levels. This behavior aligns with prior research on filler-reinforced composites, where excessive filler addition often leads to matrix heterogeneity and deteriorated physical and mechanical performance [20]. While moderate AC incorporation enhances the composite's density and structural compactness attributes desirable for lightweight, high-strength construction applications exceeding the optimal filler threshold compromises these advantages. These results are consistent with established findings in hybrid composite systems, emphasizing the critical role of filler dispersion and content balance in optimizing material performance [21].

Table 2: The density of rPP/clay/AC composite with the percentage difference

Sample	Density (g/cm^3)	Relative Density
rPP/clay/AC0	0.9095	0
rPP/clay/AC1	0.9164	0.0075
rPP/clay/AC7	0.9537	0.0485
rPP/clay/AC15	0.9580	0.0533
rPP/clayAC20	0.9305	0.0230

3.2 Flexural Strength

The graphs in Figure 1 illustrate the flexural strength of rPP/clay composites with varying percentages of AC, revealing significant trends in the mechanical properties of these materials. Initially, the flexural strength increased with AC, peaking at approximately 63.04 MPa for the rPP/clay/AC7 sample containing 7% AC. The peak flexural strength at 7% AC indicates an optimal balance between filler content and matrix integrity. This enhancement is due to the reinforcing effect of AC, which improves load-bearing capacity by acting as fillers that enhance stress transfer and reduce crack propagation within the matrix. The composite containing 7% AC-treated clay exhibits good filler dispersion and minor micro-cracking, indicating an effective interfacial interaction between the clay and the polymer matrix. The relatively uniform filler distribution enhances the stress transfer from the rPP matrix to the filler, resulting in improved resistance to flexural stress. However, the flexural strength decreases beyond 7% AC content, reaching about 54.69 MPa at 20% AC. This decline is likely due to the filler agglomeration, void formation, and poor interfacial adhesion that creates stress concentration points, as seen in Figure 6. These defects reduce the composite's ability to bear flexural loads, leading to brittle failure under bending.

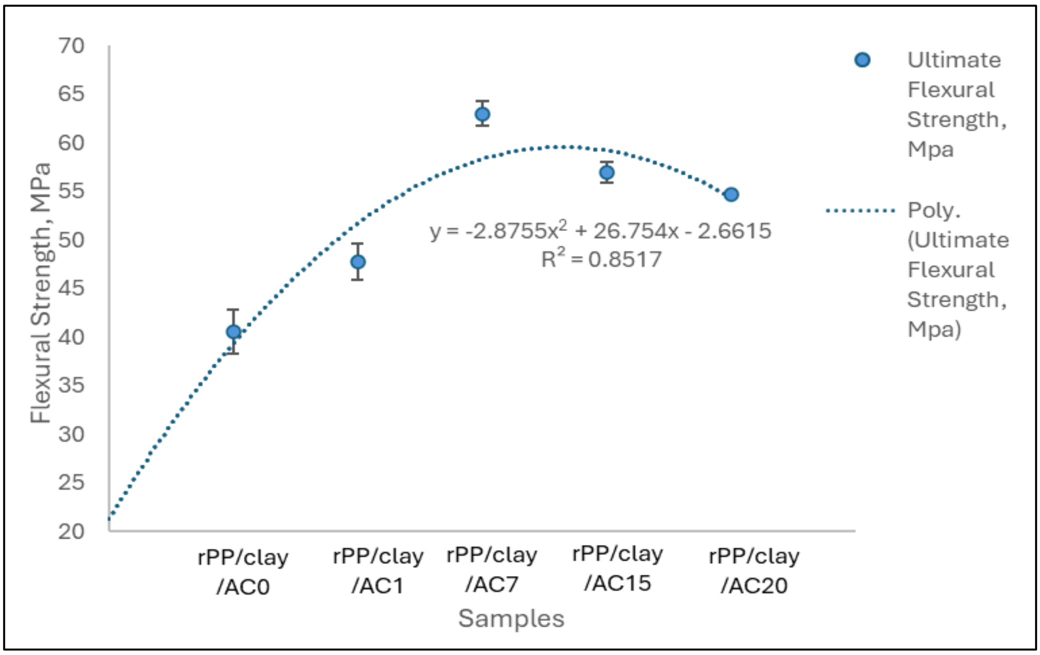

Figure 1: Flexural strength of rPP/clay/AC composite

3.3 Flexural Modulus

The flexural modulus results for rPP/clay composites demonstrate the effect of filler content on the material's stiffness (Figure 2). For rPP/clay/AC7, the flexural modulus is measured at 2.95 GPa, while rPP/clay/AC20 shows a slightly higher modulus of 2.98 GPa. The flexural modulus is a critical property that indicates the material's resistance to bending deformation under applied stress, which depends on the rigidity of the filler and its interaction with the matrix. The higher modulus observed in rPP/clay/AC20 is attributed to the increased content of activated carbon-treated clay, as this filler inherently exhibits high rigidity, contributing to the composite's stiffness. However, the slight increase in flexural modulus for rPP/clay/AC20 does not equate to improved flexural strength.

While a higher filler content enhances stiffness, it also poses challenges related to uniform distribution and defect formation. The morphological defects in rPP/clay/AC20, such as filler agglomeration and voids, reduce the material's ability to distribute and transfer stress efficiently within the matrix. These defects act as stress concentrators, disrupting the uniform stress distribution required for optimal flexural performance. Under bending conditions, the presence of these agglomerations and voids diminishes the overall structural integrity of the composite, leading to a lower flexural strength despite the higher modulus. In contrast, rPP/clay/AC7 achieves a good balance of stiffness and strength due to optimal dispersion, which minimizes morphological flaws and allows effective stress transfer.

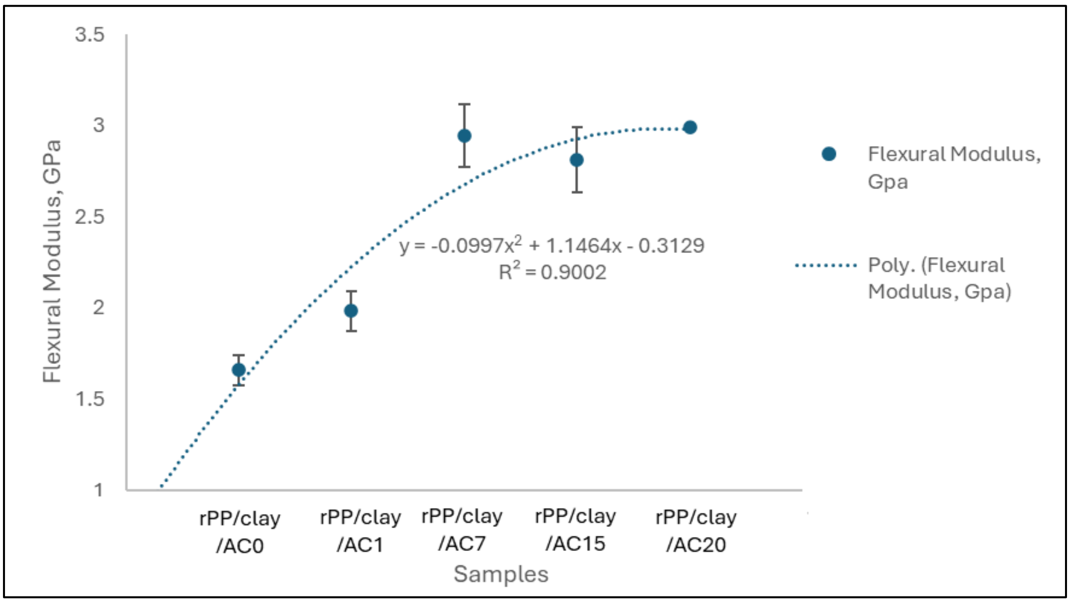

Figure 2: Flexural modulus of rPP/clay/AC composite

3.4 Hardness

Figure 3 shows hardness value of rPP/clay/AC composite. The bar graph shows a general trend of increasing hardness with higher percentages of AC treatment in the clay. The initial sample, rPP/clay/AC0, has the lowest hardness value, around 67.2, indicating that untreated clay reinforcement minimally enhances the rPP matrix. As the AC content increases to 1%, the hardness significantly improves to approximately 72, which can be attributed to the reduced presence of filler-related defects and a smoother microstructure. The polymer matrix likely contributes to the resistance to localized deformation, as the low filler content does not create significant voids or weak points.

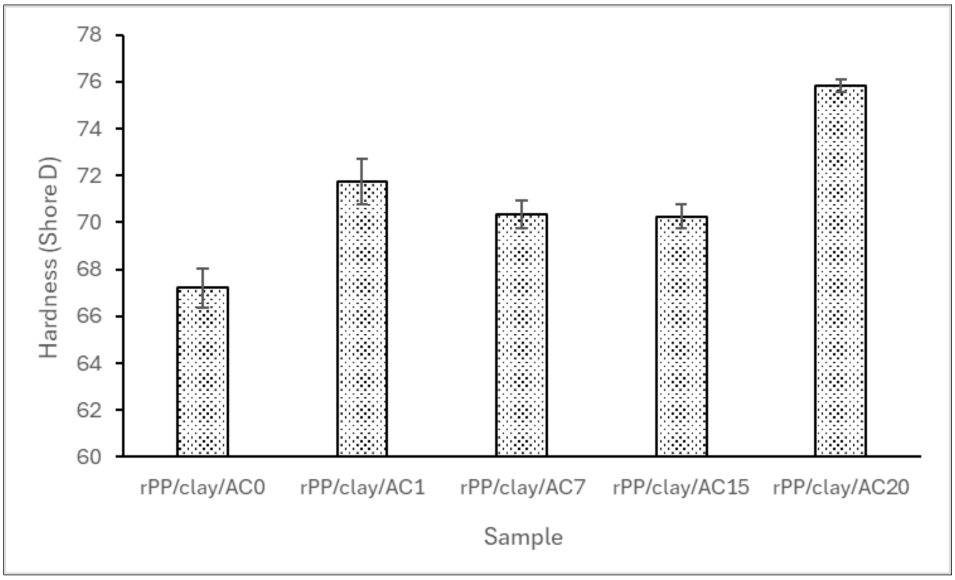

Figure 3: Hardness value of rPP/clay/AC composite

Further increases in AC content to 7% lead to a slight reduction in hardness, possibly due to insufficient dispersion or agglomeration of AC-treated clay within the rPP matrix. However, as the AC content increases to 15%, the hardness stabilizes, remaining comparable to the rPP/clay/AC7 sample. This suggests a threshold where additional AC does not substantially enhance composite hardness, likely due to limitations in polymer-clay interactions or filler saturation. Notably, the sample with 20% AC shows a marked increase in hardness, reaching a peak value of around 75.83. This is related to the high filler content, as AC-treated clay is rigid. However, when the filler content becomes excessive, agglomeration and voids are formed. These agglomerates disrupt uniform stress distribution, thereby reducing the overall flexural strength and modulus, although they may still provide localized resistance to indentation, leading to higher hardness values.

3.5 Morphological Analysis

The FESEM analysis of rPP/clay/AC composites provides valuable insights into their mechanical performance, particularly in hardness, flexural strength, and flexural modulus. Figure 4 illustrates the schematic diagram of the flexural stress distribution of the sample, with the crack indicating the location where the FESEM image was taken, as the sample did not break. Figure 5(a) shows the rPP/clay/AC1 composite's surface at 300x magnification appears relatively smooth, with minor signs of roughness. This suggests that the AC particles are well-dispersed within the rPP/clay matrix, enabling uniform stress distribution during mechanical loading. At 1000x magnification (Figure 5(b)), the finer details reveal adequate bonding between the matrix and the AC particles, with minimal gaps or voids. The tightly integrated phases observed in the rPP/clay/AC1 sample contribute to its mechanical properties, although the lower AC content limits its overall reinforcement effectiveness. While the dispersion is uniform, the 1% AC content does not provide sufficient reinforcement to maximize the composite's stiffness and strength. In comparison, the surface texture of the rPP/clay/AC7 composite in Figure 5(c) and (d) shows notable improvements. At 300x magnification, the structure appears more homogeneous and uniform than rPP/clay/AC1, with better dispersion of AC particles and fewer visible defects or inconsistencies. At 1000x magnification, the microstructure of rPP/clay/AC7 reveals a denser and more compact surface with tightly bonded AC particles and matrix material. The absence of significant voids or cracks indicates excellent interfacial bonding, which enhances the composite's ability to transfer and distribute stress effectively. The uniform reinforcement provided by the 7% AC particles is evident in the denser and more refined microstructure, contributing to the composite's superior mechanical properties, such as higher flexural strength and modulus. In conclusion, well-dispersed fillers improve load transfer and reduce stress concentrations, enhancing strength and modulus [22]. Conversely, poor dispersion or agglomeration can lead to defects and stress concentration points, reducing mechanical performance.

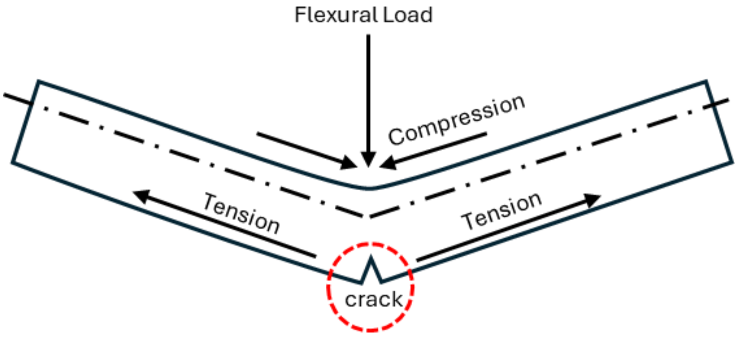

Figure 4: Schematic diagram of flexural stress distribution

As the AC content increases to 15%, the sample exhibits signs of breaking during flexural testing. The rPP/clay/AC15 sample shows intermediate behavior, with noticeable agglomeration and void formation (Figure 6(e) and (f)).

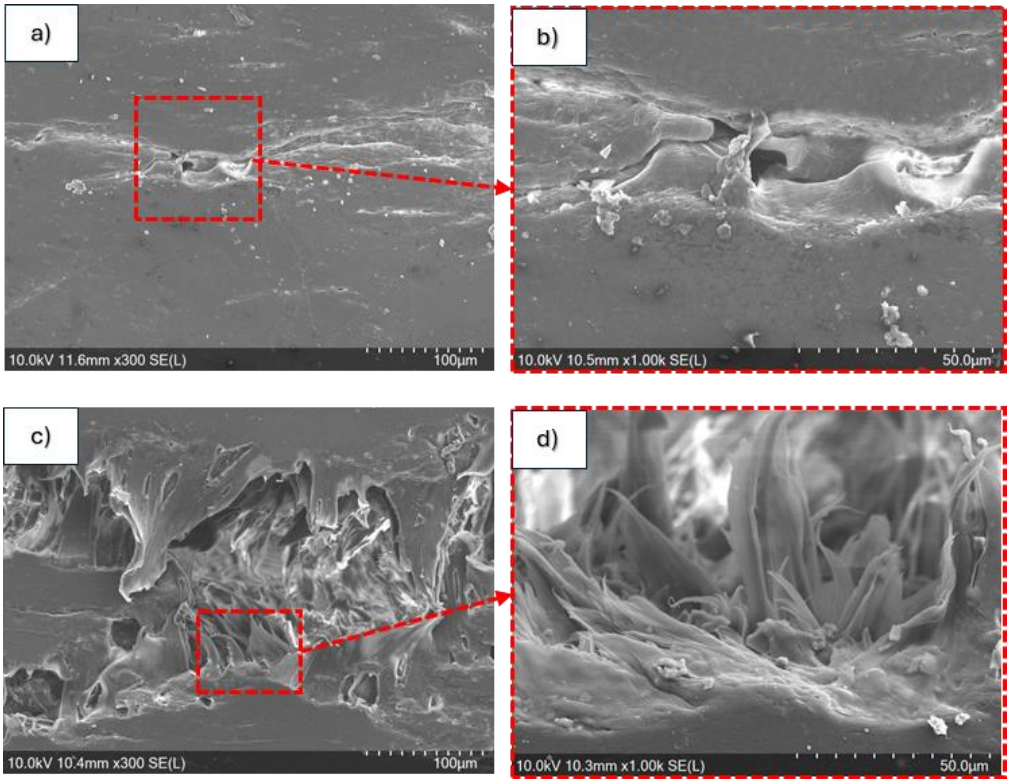

Figure 5: FESEM image of (a) rPP/clay/AC1 in 300x magnification, (b) rPP/clay/AC1 in 1000x magnification, (c) rPP/clay/AC7 in 300x magnification and (d) rPP/clay/AC7 in 1000x magnification

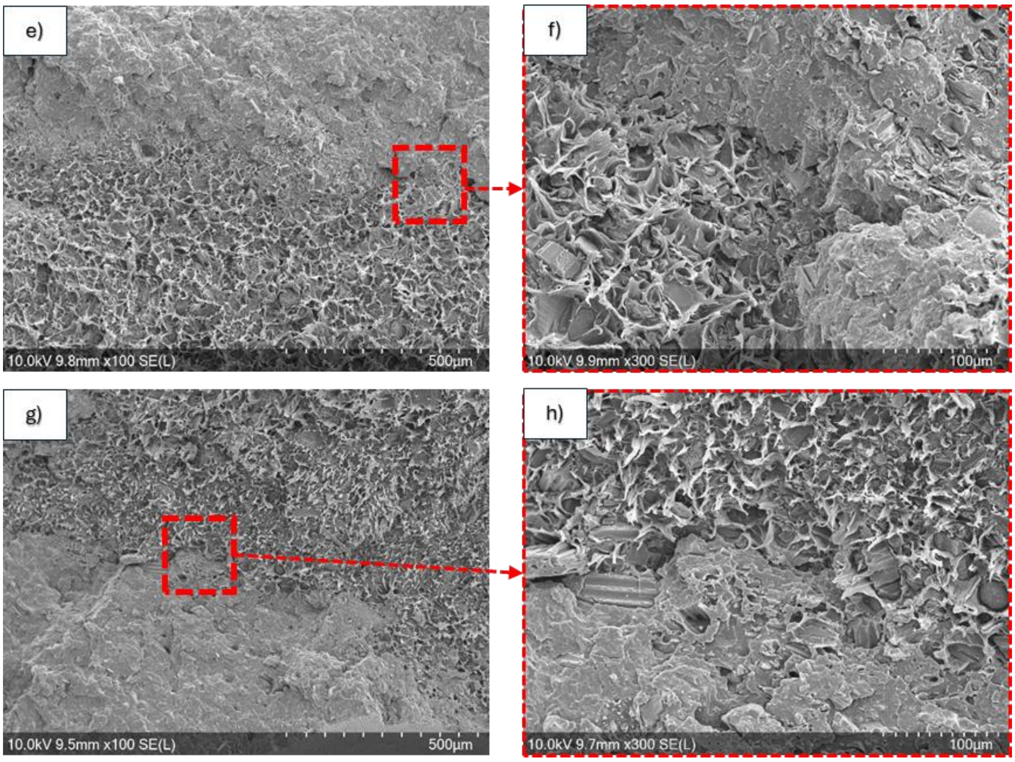

Figure 6: FESEM image of (e) rPP/clay/AC15 in 100x magnification, (f) rPP/clay/AC15 in 300x magnification, (g) rPP/clay/AC20 in 100x magnification and (h) rPP/clay/AC20 in 300x magnificatio

These defects compromise flexural properties and hardness, as the filler content is neither low enough to prevent agglomeration nor optimal for achieving uniform stress distribution. The excessive filler content at 20% AC-treated clay leads to significant agglomeration and voids (Figure 6(g) and (h)), resulting in poor flexural strength and modulus due to stress concentration and inefficient load transfer. Additionally, voids can act as crack initiation sites, reducing the material's strength and durability. However, the rigid nature of the filler contributes to high localized resistance, which explains the higher hardness observed in this sample.

4. CONCLUSIONS

In conclusion, the integration of AC as a filler significantly enhances the mechanical performance of rPP/clay composites, with optimal results at 7 wt% AC due to uniform dispersion, effective stress transfer, and minimal interfacial defects. AC primarily improves polymer matrix bonding through its high surface area and porous structure, enabling mechanical interlocking at the micro- and mesopore levels. This rivet-like interfacial mechanism, coupled with limited chemical bonding between AC's surface functional groups and the polymer, contributes to enhanced tensile and flexural strength. The incorporation of activated carbon into recycled polymer composites enhances mechanical properties by promoting strong interfacial interactions and efficient stress transfer within the material. When derived from renewable or waste-based resources, activated carbon serves as an environmentally sustainable filler option that supports circular economic objectives and reduces reliance on virgin raw materials. These composites not only achieve mechanical performance on par with or exceeding that of conventional virgin polymers but also provide added sustainability benefits, making them suitable candidates for eco-friendly construction applications. Nonetheless, limitations such as reduced ductility and filler agglomeration at higher AC concentrations and the controlled nature of laboratory testing suggest the need for further studies. Future research should assess long-term performance under dynamic, impact, and environmental loading conditions and explore hybrid reinforcement systems to optimize durability and sustainability.

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Author Contributions

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

Disclosure of Conflict of Interest

The authors declare that they have no conflict of interest.

Compliance with Ethical Standards

The work is compliant with ethical standards.

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