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Influence of calcium carbonate reinforcement on mechanical properties and sustainability of hdpe-recycled plastic composites for industrial applications

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HIGHLIGHTS

- Twelve formulations with HDPE, CaCO₃, and recycled plastic were prepared and tested
- Higher CaCO₃ reduced tensile strength and elongation, increasing brittleness
- Recycled plastics enhanced elongation and impact resistance, improving toughness
- Optimal blends of CaCO₃ and recycled plastics balance strength and sustainability
- The composites are affordable, eco-friendly, and suitable for various industries

Keywords:

High-density polyethylene (HDPE) CaCO₃ Recycled Plastics Mechanical Properties Sustainability

ABSTRACT

Currently, the demand for sustainable materials has led to an interest in polymer composites filled with mineral fillers and recycled materials. High-density polyethylene (HDPE) is a potential matrix for green composite production due to its widespread application, excellent mechanical properties, ease of recycling, and high density. This study investigates the mechanical properties of HDPE composites containing different mix proportions of calcium carbonate (CaCO₃) and recycled plastic. The mechanical performance of composites was evaluated by testing their tensile strength, elongation at break, Young's modulus, hardness, flexural strength, flexural modulus, and impact strength. Inclusion of CaCO3 in the matrix resulted in improved stiffness and rigidity of HDPE, with the Young's modulus increasing significantly to 1517.2 MPa for sample A3. However, with an increase in stiffness, there was a decrease in tensile strength and elongation at break, indicating a transition to a more brittle behavior. Samples containing a higher content of recycled plastic showed an increase in ductility and impact resistance, with sample A8 surpassing all others. The blends A9 to A12, which are the most balanced mixtures of HDPE, CaCO3, and recycled plastic, offered the best compromise in terms of mechanical strength and flexibility. Sample A12 demonstrated good performance in terms of tensile strength (18.71 MPa), elongation at break (90%), Young's modulus (210 MPa), and impact strength (69.9 kJ/m²). The results of the current study support the idea that plastic waste can be recycled and used as components in functional materials, aligning with the sustainable development goals. Recycling plastic reduces landfill waste, conserves resources, and enables the creation of new products, thereby contributing to a more circular economy.

1. Introduction

High-density polyethylene is a thermoplastic polymer known for its superior mechanical strength and chemical resistance [1]. It is widely used in applications such as packaging, coatings, and automotive parts [2]. In this investigation, calcium carbonate was selected as the primary filler due to its widespread industrial use, cost-effectiveness, and ability to enhance specific properties of polymer matrices. Compared to other fillers, CaCO₃ offers advantages including increased stiffness, dimensional stability, and thermal resistance. It is abundantly available, economically sourced, and enhances product opacity and a fine surface finish desirable traits in sectors like packaging and construction. These properties make CaCO₃ a promising additive for creating high-performance, sustainable polymer composites.

HDPE's industrial significance began in the mid-20th century. Although first applied in radar cable insulation during world war II, HDPE was synthesized in 1953 by Erhard Holzkamp and Karl Ziegler, and it was first commercialized in pipe production in 1955. Its ongoing relevance stems from excellent mechanical properties, low plasticization time, ease of processing, and affordability. Other polyethylene variants include medium-density polyethylene (MDPE) and low-density polyethylene (LDPE).

Most studies in the literature investigate polymer composites reinforced with either mineral fillers or recycled plastics separately. However, limited work explores the combined influence of both additives in HDPE-based composites. This study addresses that research gap by systematically evaluating how various proportions of CaCO3 and recycled plastics affect the mechanical performance of HDPE composites. The objective is to harness the reinforcing effects of CaCO3 stiffness and thermal resistance while leveraging the flexibility and sustainability of recycled polymers. HDPE/CaCO3 blends are widely used in various industries due to their improved properties and cost-effectiveness. They are used in the automotive sector for lightweight components, pipe construction, flooring, and panels, as well as packaging for rigid containers, films, and bottles [3, 4]. CaCO₃ can be categorized into natural, ground, precipitated, coated, and uncoated forms. Natural calcium carbonate requires chalk, limestone, or marble for whiteness and brightness. Ground calcium carbonate is produced by grinding natural sources, such as limestone or marble, and is commonly used in the plastics, paints, and paper industries [5, 6]. Precipitated calcium carbonate (PCC) was first commercially manufactured in England in the 1850s and has been used in the production of nylon for the plastic industry [5, 7]. Coated calcium carbonate is coated with agents such as stearic acid to improve dispersion and compatibility with polymer matrices [8]. The particle size of CaCO₃ has emerged as a critical area of research for enhancing polymer composites, especially in HDPE-based nano- and microcomposites [9]. CaCO3 as a filler in HDPE and recycled Plastic blends can improve stiffness and lower costs, but it can also cause defects or negative effects on the material's overall performance. These include reduced impact strength [3, 10], agglomeration and poor dispersion [11], compatibility issues [12], decreased flexibility [13–17] and thermal degradation [18-24]. To address these issues, methods such as optimizing filler loading, enhancing compatibility, and using compatibilizers have been developed [25, 26]. Surface treatments, such as silane coupling agents and stearic acid, can enhance the interaction between the polymer and the infill, while filler loading should be controlled to maintain a balance between cost and mechanical performance [27–29]. Compatibilizers, such as MAPE [30–34] and EVA [35–37], can also help mitigate phase separation between the HDPE matrix and the CaCO3 infill, leading to improved mechanical properties and dispersion. Environmental problems, primarily the growing issue of plastic waste and limited resources, have heightened the global demand for sustainable composite materials. Under this critical scenario, preparing HDPE composites synergistically reinforced with inorganic fillers and made from recycled plastic offers a very promising pathway. The present study elaborates on the formulation, characterization, and testing of new HDPE-based composites that utilize CaCO3 as a mineral filler and recycled plastic to achieve partial substitution of virgin polymer. This research investigates the development of sustainable polymer composites with varying proportions of HDPE, calcium carbonate, and recycled plastics. The primary importance is given to studying how these compositional variations affect both the mechanical and thermal properties of the materials, to improve their performance. The study's significance lies in its dual emphasis on advancing the mechanical properties of materials while promoting environmental consciousness. Through the strategic incorporation of recycled plastic, the research reduces the use of virgin polymers, thereby contributing to the preservation of resources and the prevention of waste buildup. At the same time, the inclusion of CaCO₃ has been proven to increase the material's stiffness and is relatively inexpensive to implement. Hence, the findings are highly useful for practical applications in industries such as packaging, construction, and automotive, which are trending toward lightweight and cost-effective material solutions with an environmentally friendly approach. This paper describes the experimental method, presents the results of material characterization, and discusses the findings in terms of sustainable composite materials.

This study aims to investigate the synergistic effects of combining recycled plastic and calcium carbonate in an HDPE-based polymer matrix, addressing current obstacles in cost-effectiveness, environmental sustainability, and material performance.

2. Experimental method

2.1 Materials

A total of twelve HDPE matrix samples were prepared using varying proportions of HDPE, CaCO₃, and recycled plastics. High-density polyethylene (HDPE) HBM 5510 Virgin Granules were supplied by Arya Sasol Petrochemical Company (ASPC), which provided the HDPE with a density of 0.955 g/cm³ and a melt flow index of 0.504 g/10 min at a 5 kg load & 190 °C. The resulting granules have a density of 0.915 g/cm³ and a melt flow index of 5.94 g/10 min at a 5 kg load & 190 °C Figure 1a. Calcium carbonate (CaCO₃) was supplied by Pooya Polymer, Tehran Figure 1b. A variety of recycled plastic materials were collected, mixed, crushed, and processed into granules using injection molding at a local manufacturing facility. Physical condition: natural, solid color Figure 1c.



Figure 1: Materials used: (a) recycled plastic; (b) CaCO₃ filler; (c) HDPE

2.2 The various steps involved in plastic recycling are outlined

To maintain consistency and quality of the material, recycled plastics were prepared in the study through a multi-step procedure, as illustrated in Figure 2 (a-f). The procedure involved collection, washing, drying, shredding, and granulation. Each step was necessary for removing contaminants and ensuring uniform particle sizes, which would lead to adequate recycled plastic for composite fabrication. **Collection of Plastic Waste:** Figure 2 a, shows the collection of plastic waste, such as plastic films or packaging, which are used as raw materials for recycling. **Shredding and Sorting:** Figure 2 b, shows the plastic waste is being shredded into small pieces, thereby preparing it for the next stage in the recycling process. **Extrusion Process:** Extrusion, depicted in Figure 2 c, is another way in which the shredded and melted plastic is forced into molds to achieve a new shape or form for a given product. **Cooling and Solidification:** In this Figure 2 d, the water is utilized to cool and solidify the plastic while it retains its new form. **Granulating:** In Figure 2 e, the granulating phase is shown, where the extruded plastics are turned into small pellets, easier to handle and immediately ready for manufacturing into new plastic products. **Final Product:** The final product-small plastic pellets that will be further processed into new products- is shown in Figure 2 f.



Figure 2: Procedure of recycled plastic preparation

2.3 Processing

The set of twelve composite samples was prepared by melt mixing HDPE with varying ratios of calcium carbonate and recycled plastics, followed by compression molding. Incorporating CaCO₃ and recycled plastics at varying levels of 0, 25, 50, and 75% by weight, these twelve formulations comprise virgin HDPE for the remainder.

The rationale behind opting for these levels was twofold. The first is that it establishes a broad yet systematically stepped set of intermediate points to observe the gradual changes in composite properties, from zero (pure HDPE) to the heavily filled (75% filler). These intervals, on the other hand, are very common in the current literature as well as in industrial practice, serving as benchmark levels that permit meaningful comparisons and performance mappings. A 25% increment offers a fair balance between resolution and the number of experiments required so that clear trends in mechanical behavior can be discerned. All samples were processed using a single-screw extruder [38, 39]. At 200 °C, the mixing was carried out under a screw rotation speed of 50 rpm and a mixing duration of 10 minutes [26], as illustrated in Figure 3. These parameters were established in consideration of a preliminary trial to ensure adequate dispersion of the fillers while preventing thermal degradation of the recycled plastics. No change was allowed in process conditions between samples. Hence, uniform processing conditions were maintained across all formulations, allowing only the composite compositions to affect the mechanical properties investigated, as shown in Table 1.

Sample No.	HDPE (%)	CaCO ₃ (%)	Recycled Plastics (%)
A1	75	25	0
A2	50	50	0
A3	25	75	0
A4	100	0	0
A5	0	25	75
A6	0	50	50
A7	0	75	25
A8	0	0	100
A9	50	10	40
A10	50	15	35
A11	50	20	30
A12	50	0	50

Table 1: Formulation and labeling of HDPE blends with recycled plastic and CaCO₃ fillers

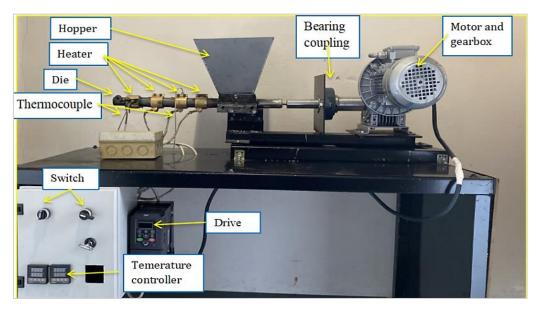


Figure 3: Single screw extruder, a homemade machine

2.4 Testing

To ensure the accuracy and repeatability of the mechanical test results, each test was typically run three times for all composite samples. Three identical specimens were created and tested for each formulation under standardized conditions. Figure 4 (a-d) displays the specimens prepared for (a) tensile testing, (b) hardness testing, (c) impact testing, and (d) flexural testing. The average values and standard deviations were then calculated and reported to manifest the consistency and variability of the data, respectively.

Before use in such testing, the instruments were calibrated according to the guidelines provided by the manufacturers, and environmental conditions were maintained under control throughout the process to minimize other influences. No outlier points or disturbances worthy of mention were observed in any of the repeat evaluations, which adds strength to the credibility of the attained results.

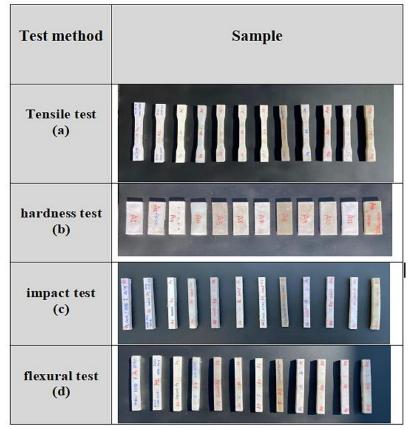


Figure 4: Test specimens: (a) tensile test specimens, (b) hardness test specimens (c) impact test specimens, (d) flexural test specimens

2.4.1 Tensile test

The tensile test was performed on an ASTM D638-22 type I dogbone specimen at a constant strain rate of 50 mm/min. Tensile properties of the samples were measured in a (XHG-50 RING STIFFNESS TESTER). The apparatus was calibrated to a capacity of 50 kN, and axial displacement was measured using a clip-on extensometer with a gauge length of 50 mm, as shown in Figure 5.

2.4.2 Flexural test

The flexural test was conducted using a GOTECH universal tester (AI-3000) with a maximum loading capacity of 5 kN, as shown in Figure 6. Small specimens measuring 127 mm by 12.7 mm by 3.2 mm underwent three-point bending with a span of 51.2 mm. According to ASTM D790-15, these specimens were tested flatwise on the support span, with a span-to-depth ratio of 16:1. The test was conducted at a crosshead speed of 2.0 mm/min. The tests were conducted at a temperature of 20 °C.





Figure 5: Universal testing machine for tensile test

Figure 6: Universal testing machine for flexural testing

2.4.3 Charpy Impact Test

Impact strength was assessed using notched specimens in a Charpy impact tester (Model XJJD-50 Series) used for Charpy impact tests depicted in Figure 7. ASTM D6110-18 conducted tests. Heat was applied to the test samples, which measured 127 mm x 12.7 mm x 4 mm (length x width x thickness), and they were arranged into the specimen holder. The samples were V-notched at the midpoint of the $10 \times 63.5 \text{ mm}^2$ surface with a 2 mm depth notch inclined at 45° .

2.4.4 Hardness Test

Hardness measurement was conducted using a Shore D durometer hardness tester, as shown in Figure 8. The test was conducted in accordance with the requirements of ASTM D2240-15 standard. The test samples, measuring 72 mm x 32 mm x 6 mm (length x width x thickness), were placed in the specimen holder. The range was 0.5 Shore D, while accuracy was 1-100 Shore D. A minimum of five points were tested per sample, and average results were computed.

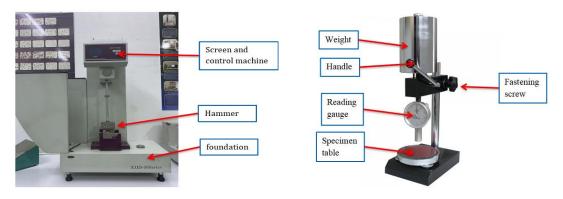


Figure 7: Digital image of the Charpy Impact testing machine

Figure 8: Shore D Hardness Tester

3. Results and discussion

3.1 Analysis of tensile strength of hybrid blends of HDPE, CaCO₃, and recycled plastics: interplay and material performance

The tensile test data are summarized in Table 2, where the variation in tensile strength observed across the different samples is evident, along with their corresponding Standard Deviations (SD), which show the influence of the variation in proportions of

HDPE, CaCO₃, and recycled plastic. The pure HDPE sample (A4) without any fillers showed the highest tensile strength of 23.8 MPa, while the smallest tensile strength observed was 4.06 MPa in sample A7, which agrees with the published values for unmodified HDPE in the literature [1, 2, 9, 40, 41]. This corroborates the expected behavior of pure HDPE, which is generally characterized by high tensile strength due to its inherent material properties.

A significant reduction in tensile strength at high CaCO₃ loading is a well-documented phenomenon in polymer composites and can be attributed to several key factors. Firstly, excessive filler content often leads to poor dispersion and increased agglomeration of CaCO₃ particles within the polymer matrix. These agglomerates act as stress concentrators, initiating microcracks under mechanical loading and thus lowering the composite's overall strength [40, 42, 43]. Secondly, high filler concentrations reduce the volume of the load-bearing polymer phase, limiting the matrix's ability to transfer stress across the composite structure efficiently [44]. Moreover, when the filler—matrix interfacial adhesion is insufficient—which is common with untreated or non-reactively treated CaCO₃ the stress transfer becomes even more ineffective, further weakening the material [10]. Lastly, high CaCO₃ content increases the brittleness of the composite, leading to reduced energy absorption and premature failure under tensile load [45].

Sample No.	Tensile strength (MPa)±SD	Young's modulus (MPa)±SD	Elongation at break (%)±SD
A1	15.13 ± 0.82	770 ± 10	47.5 ± 4.77
A2	9.99 ± 0.98	820 ± 40	17.33 ± 2.52
A3	5.72 ± 0.63	860 ± 20	8.46 ± 0.87
A4	23.81 ± 1.64	440 ± 40	435 ± 33.51
A5	9.06 ± 1.10	250 ± 50	111.33 ± 19.50
A6	6.86 ± 0.95	310 ± 30	30.99 ± 7.85
A7	4.07 ± 0.31	360 ± 40	9.47 ± 1.41
A8	14.15 ± 0.78	120 ± 20	604 ± 49.27
A9	16.03 ± 1.56	260 ± 50	46.79 ± 3.35
A10	14.59 ± 1.69	306.67 ± 20	45.91 ± 1.86
A11	14.19 ± 1.04	326.67 ± 20	44.57 ± 2.21
A12	18.72 ± 0.30	210 ± 10	89.60 ± 1.15

Table 2: Mechanical Properties of HDPE-Based blend Samples with Varying Filler Ratios

Figure 9 a showed that the tensile strength decreased to 15.12 MPa with an increase in CaCO₃, as in A1. The material still exhibits acceptable tensile properties; however, its performance does decline. Though not without sacrifice.

The mechanical properties of sample A2, in particular, the tensile strength of 9.98 MPa, are lower than that of pure HDPE, suggesting that the addition of CaCO₃ causes a decrease in tensile strength. The weakening effect of CaCO₃ filler on the polymer matrix, which may result in a weaker bond with the HDPE molecules and a lower stress tolerance [45, 46].

A significant turning point in the behavior of the tensile strength can be observed when the CaCO₃ content exceeds 50%. Sample A3 shows a marked reduction in tensile strength, dropping to 5.71 MPa. This represents a considerable drop from the tensile strength of pure HDPE (23.8 MPa), illustrating the negative impact that higher CaCO₃ content has on the material's strength. Interestingly, sample A7 exhibited an even lower tensile strength of 4.06 MPa. The drastic reduction seen in these samples highlights the weakening effect of combining large proportions of CaCO3 and recycled plastic, which may disrupt the overall structural integrity of the composite. In contrast, sample A8, which was made entirely from recycled plastics, exhibited a tensile strength of 14.15 MPa. This result is consistent with the expected strength of recycled plastics. On the other hand, the samples that were composed of recycled plastic and CaCO3, such as A5, A6, and A7, exhibited significantly lower stress resistance. These samples, which incorporated substantial amounts of recycled plastics and CaCO3, generally showed poorer stress resistance. The use of recycled plastic and CaCO₃ will cause a reduction in the tensile strength of the material, particularly when used in large quantities [40, 47, 48]. Upon examining samples A9 to A12, which are composite materials containing a mix of HDPE, CaCO₃, and recycled plastics, we observe relatively higher stress values than those seen in the earlier samples. Sample A9 achieved a tensile strength of 16.03 MPa. Similarly, sample A10 demonstrated a tensile strength of 14.59 MPa, and sample A11 yielded a value of 14.19 MPa. Sample A12 manifested 18.72 MPa of tensile strength, which is lower than pure HDPE (A4: 23.81 MPa) but considerably higher than CaCO₃-filled blends. This implied that the tensile strength in the HDPE matrix remained fairly high as recycled plastic was added without CaCO₃. These results indicate that the inclusion of both HDPE and recycled plastics, along with a small amount of CaCO₃, leads to a balanced material with moderate stress resistance [48].

Overall, the stress resistance of blend materials made from varying combinations of HDPE, CaCO₃, and recycled plastic generally reflects a trade-off between strength and flexibility. The data in Figure 9 b, concerning the Young's modulus of various samples, show that the Young's modulus values measured for the 12 samples follow a trend that links the stiffness of a material more closely with its composition, specifically related to calcium carbonate and recycled plastics. An extreme stiffness of 860 MPa was recorded for sample A3, showing that the calcium carbonate reinforcement gave the highest rigidity. On the other hand, the lowest value of 120 MPa was obtained by sample A8, which consisted of 100% recycled plastic, indicating maximum flexibility. This value is in stark contrast to a pure HDPE sample A4, which has a Young's modulus of 440 MPa.

Samples A2 and A1 exhibited slightly less stiffness than sample A3, with values of 820 and 770 MPa, respectively. This indicates that the presence of calcium carbonate in these samples was the primary factor in increasing rigidity. The recycled material has a lower Young's modulus, resulting from the loss of some mechanical properties caused by the recycling process.

On the contrary, CaCO₃ reinforced the mechanical properties, giving maximum stiffness to the material by acting as a filler that strengthened the bond between the polymer chains. Young's modulus, due to its properties, makes the material more rigid and resistant to deformation. Samples A5, A6, and A7 demonstrated an increasing trend in Young's modulus as the CaCO₃ content in the samples rose, with measured values of 250, 310, and 360 MPa, respectively. These results indicate that the stiffness of the material improves with an increase in CaCO₃ content in the mixture.

Samples A9 to A12, which are composites formed in different proportions of HDPE, calcium carbonate, and recycled plastics, exhibit intermediate values of Young's modulus (260, 306.67, 326.67, and 210 MPa). These samples offer a trade-off between stiffness and flexibility, as the calcium carbonate content increases rigidity, while the recycled plastic somewhat counteracts this effect. Modifications are made in the elongation at break shown in Figure 9 c for different samples. The elongation at break values reveal the ductility and flexibility of the materials. Sample A1 has an elongation at break of 47.5%, indicating a moderate level of flexibility. This value suggests that the material can withstand some stretching before failure. Sample A2, with an elongation at break of 17.33%, exhibits much less flexibility. This lower value suggests that the material is more brittle and might break or crack under even minimal stretching or deformation.

In contrast, sample A3 shows an even lower elongation at break of 8.45%, further indicating poor ductility. Materials with such low elongation at break are typically more prone to brittle failure. They may not perform well in dynamic applications that require some degree of flexibility or adaptability. On the other hand, sample A4 (pure material), with an elongation at break of 435%, offers a good balance between flexibility and strength. While not as flexible as sample A8, which has a higher elongation at break of 604%, A4 still exhibits significant stretchability before failure, making it suitable for applications where moderate flexibility is required without compromising structural integrity. A8, with its higher elongation, is better suited for applications that demand extreme flexibility; A4 is likely to perform better in applications where a balance of flexibility and strength is crucial. While A8 offers superior flexibility, A4 retains more strength, making it a better choice for products that need to withstand both stretching and mechanical stress.

Sample A5 follows with an elongation at break of 111.33%, indicating that it is more flexible than the samples with elongations below 50%. This material would likely be used in applications where a balance between flexibility and strength is needed. With the elongation at break at a mean of 30.99%, sample A6 is more ductile than A2 and A3 but less so than A1 and A5. It is considered a medium-firm material and can perform functions where some elongation is possible, but excessive stretching should be avoided.

Sample A7, with a mean elongation at break of approximately 9.46%, exhibits similar features to sample A3, indicating low ductility and high brittleness, which is less desirable for any application where tensile forces must offer more flexibility. Providing a mean elongation at break of 46.78%, sample A9 is endowed with a moderate degree of ductility, much akin to A1. The material in sample A9 can serve the purpose of some elongation in an application, while retaining sufficient strength to avoid excessive elongation or failure.

In terms of elongation at break, sample A10 has a capacity with a mean value of 45.90%, almost equal to that of sample A9, thus stipulating that both materials exhibit similar behavior based on elongation at break. Sample A11 is given with an elongation at break of 44.6%, slightly less than that of A1, A9, and A10, but showing a reasonable level of flexibility. Although it is not the most ductile, it still offers sufficient stretchability for applications that require a balance between strength and flexibility. Sample A12 exhibited 90% elongation at break, which was significantly lower than that of pure HDPE (A4: 435%).

More precisely, the mean elongation at break values represent the resulting flexibility levels of the samples, where A4 and A8 are highly ductile; on the other hand, A2, A3, and A7 show very low elongation at break, rendering them susceptible to brittle-type failure. All the other samples lie in the middle with mixed potentials for both flexibility and strength, depending on application requirements [2, 40].

3.2 The influence of HDPE, calcium carbonate, and recycled plastics on composite hardness

In this study, the effect of varying proportions of the components, HDPE, CaCO₃, and recycled plastics, on the hardness of composite materials is explored. Understanding such effects will help determine the best combinations of materials for various industrial applications, as summarized in Table 3.

Significant variations in hardness were observed across the composite samples. The inclusion of recycled plastics generally led to a notable reduction in hardness compared to pure HDPE. This reduction can be attributed to several factors: recycled plastics often contain degraded polymer chains due to previous processing cycles, which decrease the molecular entanglement density and soften the material. Moreover, recycled materials may include LDPE or contaminants, which are inherently more flexible and less crystalline than virgin HDPE, further contributing to a lower surface resistance to indentation [49, 50]. It is worth noting that the recycled material used in this study has a relatively low density of 0.915 g/cm³.

On the other hand, increasing the content of calcium carbonate in the matrix tends to counteract this softening effect by increasing the overall rigidity and surface resistance of the composite. CaCO₃, being a hard and stiff inorganic filler, restricts the mobility of polymer chains and increases the resistance to localized plastic deformation, which manifests as higher hardness values [51, 52]. Additionally, the uniform dispersion of CaCO₃ particles enhances the load distribution under compressive force, thereby reinforcing the composite surface against indentation. This compensatory effect becomes more pronounced at higher filler loadings, effectively offsetting the plasticizing behavior introduced by the recycled content.

It is well known that incorporating a more rigid filler into the polymer matrix results in an increase in the final material's rigidity. This filler performs the function of a nucleating agent, which in turn diminishes the elasticity of the materials and ultimately results in a decrease in the composite's ductility [53].

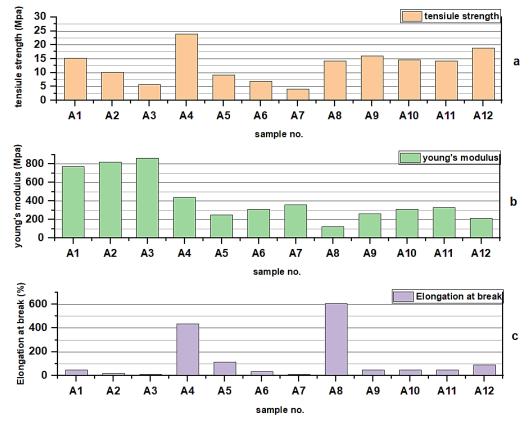


Figure 9: Mechanical properties of HDPE matrix with varying percentages of CaCO3 and recycled plastic (a) Tensile strength, (b) Young's modulus, and (c) Elongation at break

Table 3: Results of Hardness Tests Performed on HDPE Blends with Varying Proportions of Calcium Carbonate and Recycled Plastics

Sample No.	Hardness (Shore D)± SD	
A1	71.00 ± 1.55	
A2	74.16 ± 0.75	
A3	77.33 ± 1.66	
A4	64.16 ± 1.47	
A5	63.33 ± 2.66	
A6	66.00 ± 1.10	
A7	71.00 ± 1.26	
A8	58.33 ± 1.03	
A9	60.58 ± 1.91	
A10	67.91 ± 1.36	
A11	69.96 ± 0.90	
A12	62.41 ± 2.94	

The addition of the filling to the polymer matrix alters the hardness behavior of these polymers. Increased filler concentration leads to a decrease in the free space available to polymer chains, ultimately resulting in a decrease in the material's flexibility. Restricting the motion of the polymer chains would consequently lead to a more rigid structure due to this decrease in free volume. The filler increases rigidity and hardness by restricting the motion and deformation of polymer chains under stress. Therefore, the polymer matrix becomes more rigid, and the composite material becomes less deformable, thus becoming more resistant to wear and mechanical forces. Thus, important behavior in applications where high hardness and durability are required [54]. Additions of CaCO3 into polymers such as HDPE have undergone much research to determine the intended enhancement of material properties, particularly hardness. Calcium carbonate is a crucial filler agent that significantly impacts the mechanical properties of polymer composites. Adding CaCO3 into the polymer matrix can, as a rule, increase the hardness of the matrix material due to the reinforcing effect of the filler [40, 47].

From Figure 10, it can be inferred that sample A3 yielded a hardness test value of 77.33, which was the highest, indicating a rigid and strong combination of materials. Sample A8 is made up of 100% recycled plastics, and its hardness value of 58.33 is the lowest. This value indicates that recycled plastic contributes to the softness and flexibility of the material. Samples A1, A2, and A3 were largely made up of HDPE, with varying amounts of calcium carbonate. These samples presented relatively high

hardness values of 71, 74.16, and 77.33, respectively, indicating that calcium carbonate increases the hardness of the materials significantly [55]. However, A4, which contains 100% HDPE, showed a hardness of 65 [2, 40].

The inclusion of recycled plastics in the samples leads to a noticeable reduction in hardness. For instance, A8 has the lowest hardness of 58.33, emphasizing the softness and flexibility of recycled plastics. Recycled materials tend to be more flexible and less rigid compared to virgin HDPE or filled composites, resulting in lower hardness. The samples A5, A6, and A7, made from recycled plastics with calcium carbonate, showed mean hardness values of 63.33, 66, and 71 Shore D, respectively. These results reveal that recycled plastics exhibit reduced hardness levels; however, the introduction of added reinforcements such as calcium carbonate can partially compensate for this reduction.

The Shore D hardness test was applied to the samples A9 to A12 to determine surface rigidity and resistance to indentation. Some noticeable differences in hardness variations were observed, as summarized in Table 3, which depend upon compositional changes, especially the percentages of CaCO₃ and recycled plastic. Sample A9 registered the lowest average Shore D hardness value of 60.58, presumably owing to the relatively low amount of CaCO₃ (10%) and high amount of recycled plastic (40%). The softer nature of recycled plastic contributed to a lower surface hardness, thereby increasing ductility and decreasing rigidity. Sample A10, which contained 15% CaCO₃ and 35% recycled plastic, imparted further hardness to the composite with an average hardness value of 67.92. The increase in hardness shows the effect of CaCO₃, which makes the composite stiff and resistant to surface deformation. Sample A11 scored the highest average Shore D hardness of 69.97 among the four tested samples. With 20% CaCO₃ and 30% recycled plastic, this mix improved surface hardness because it has a lot of mineral filler, which is strong because of the natural stiffness of calcium carbonate. Sample A12, which contains 50% recycled plastic and 50% HDPE, has yielded an average hardness value of 62.42.

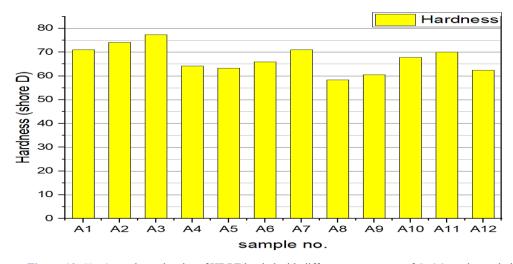


Figure 10: Hardness dependencies of HDPE loaded with different percentages of CaCO3 and recycled plastic

3.3 Effect of high-density polyethylene, calcium carbonate, and recycled plastics on flexural strength and flexural modulus of blended materials

The mechanical properties of blends based on HDPE vary significantly among the samples, depending on the mix of HDPE, CaCO₃, and recycled plastics, as illustrated by the flexural strength and modulus in Table 4.

Sample no.	Flexural Strength (MPa) ± SD	Flexural Modulus (MPa) ± SD	
A1	20.70 ± 0.50	561.84 ± 1.34	
A2	22.99 ± 0.33	743.82 ± 35.94	
A3	23.67 ± 0.11	1517.21 ± 93.00	
A4	20.33 ± 0.33	504.74 ± 46.33	
A5	8.91 ± 0.27	120.47 ± 3.92	
A6	10.76 ± 0.16	149.45 ± 2.59	
A7	13.07 ± 0.21	184.04 ± 5.70	
A8	7.07 ± 2.83	76.78 ± 1.89	
A9	19.68 ± 0.40	324.56 ± 3.38	
A10	20.79 ± 0.57	403.84 ± 1.71	
A11	23.28 ± 0.46	506.93 ± 48.80	
A 12	10.22 ± 0.27	275 27 + 9 90	

Table 4: Bending test rResults of HDPE blends with vVarying proportions of calcium carbonate and recycled plastics

Figures 11a and 11b show that the addition of CaCO₃ to HDPE increases its flexural strength and flexural modulus. At 25% CaCO₃, it enhances the structural integrity of the polymer matrix, thereby improving flexural strength. Strength continues to

increase at 50% CaCO₃, although beyond a certain level, the flexibility of the polymer matrix may start to decrease. At 50% CaCO₃, the flexural modulus is higher due to the additional stiffness; however, excessive filling could result in brittleness of the material and loss of overall toughness, as may be the case at 75% CaCO₃. Flexural strength at 75% may be equal to or slightly lower than that value due to excessive filling. The highest flexural modulus occurs at 75% CaCO₃; however, this may reduce resistance to impact and overall toughness.

Sample A4, which comprised pure HDPE, reported flexural strength values of 20.33 MPa and flexural modulus of 504 MPa. Incorporation of CaCO₃ increased flexural strength and flexural modulus, thus indicating improvements in the performance of the material. Sample A1 showed an average flexural strength (20.70 MPa) and modulus (561.84 MPa), indicating the provision of strength with flexibility by the addition of CaCO₃. The equal part of HDPE and CaCO₃ is A2, which shows increased flexural strength (22.99 MPa) and corresponding modulus (743.82 MPa), indicating more benefits with increasing filler content. A3 shows the maximum performance (23.67 MPa strength, 1517.21 MPa modulus) with 75% CaCO₃, indicating that the higher percentage of CaCO₃ present in the sample leads to increased rigidity and strength. Sample A5, which contains 75% recycled plastics, exhibits the lowest properties (8.91 MPa strength, 120.47 MPa modulus), demonstrating the reduction in integrity of recycled plastic materials. A modest increase is shown in A6, which has a ratio of 50% CaCO₃ and 50% recycled plastics (10.76 MPa strength, 149.45 MPa modulus). In contrast, advanced properties are shown in A7, which consists of more CaCO₃ (75%) and less recycled plastics (25%), compared to the earlier-mentioned mechanical properties (13.07 MPa strength, 184.04 MPa modulus). A8 is composed entirely of recycled ply and shows the lowest performance, highlighting the disadvantages of using recycled materials alone (7.07 MPa, 76.78 MPa).

Samples A9 to A12 exhibited significantly improved mechanical properties when using different ratios of HDPE, CaCO₃, and recycled plastics. Of note is that A9 (50% HDPE, 10% CaCO₃, 40% recycled plastics) yielded a moderately promising performance (19.68 MPa strength, 508.98 MPa modulus) whereas A10 (50% HDPE, 15% CaCO₃, 35% recycled plastics) and A11 (50% HDPE, 20% CaCO₃, 30% recycled plastics) gave an even greater improvement in performance, with A11 being recorded with the highest values (23.28 MPa strength, 744.36 MPa modulus). Sample A12 experienced a flexural modulus of 19.33 MPa, which expresses the almost stiff nature of the sample. Additionally, the flexural strength value was found to be 275.27 MPa, indicating resistance to bending failure. Hence, the calcium carbonate was dispersed well, creating reinforcement within the HDPE matrix. Such performance makes sample A12 an apt candidate for applications requiring rigidity and strength, such as automotive components, construction panels, or rigid packaging materials. Having such a combination means that this composite formulation for A12 strikes a good balance between reinforcement and matrix continuity. An increase in flexural strength and modulus was observed with rising CaCO₃ content up to a certain threshold, typically due to the inherent stiffness and high modulus of calcium carbonate particles, which restrict polymer chain mobility and enhance the composite's ability to resist bending. This improvement is further supported by good filler dispersion and filler—matrix adhesion, which enable effective stress transfer from the matrix to the filler [2, 29, 56, 57].

3.4 Impact of Calcium Carbonate and Recycled Plastics on the Mechanical Performance of HDPE Blends

Impact testing is a crucial test for evaluating a material's ability to absorb sudden forces or shocks. It is primarily important in evaluating the toughness of materials, which refers to their ability to absorb energy before fracturing. Impact testing evaluates the performance of materials when exposed to rapid loading and assesses their durability under these conditions.

The impact test calculates the hammer's potential energy when a large pendulum strikes a sample. The procedure involves starting a crack and enlarging it until it breaks. To increase the repeatability of the failure mode, the specimen is often notched. The test can identify brittle-ductile transitions in polymer composites. In impact testing, polyethylene (PE) gains impact strength when calcium carbonate is added [10]. The impact test, as part of this research, aims to assess the mechanical properties of the materials through real-world simulation, thereby determining the points of maximum resilience and failure for these materials. This information will help select the right materials for various uses within the system. Results from this testing are considered crucial in designing each component of a system to withstand the forces it will be subjected to during its operational life. The results obtained are presented and discussed in Table 5.

The influence of CaCO₃ concentration on the impact strength of CaCO₃/HDPE blends. Experimental findings demonstrate that the impact strength of the mixes diminishes as the CaCO₃ component increases. The incorporation of CaCO₃ results in a marginal reduction in impact strength, attributed to the pronounced molecular confinement effect of CaCO₃.

Variations in impact strength were observed across different composite formulations, with samples containing higher proportions of recycled plastics demonstrating enhanced impact energy, despite typically exhibiting lower stiffness and modulus. The mechanism of energy absorption explains this seemingly contradictory behavior during impact loading, which differs fundamentally from that of tensile or flexural loading. Unlike stiffness-driven properties, impact strength is more influenced by a material's ability to deform plastically and dissipate energy rather than resist deformation. Recycled plastics often contain shorter, degraded polymer chains and may comprise more ductile components, such as LDPE and polypropylene (PP), or even minor contaminants from previous use cycles. These characteristics result in a more flexible, less crystalline polymer matrix, allowing greater molecular mobility under sudden loading and thereby enhancing the material's energy absorption capacity before fracture [49, 50]. Moreover, the heterogeneous structure of recycled plastic-rich composites, with their less ordered and more amorphous regions, creates multiple localized yielding zones during impact.

These regions act to dissipate kinetic energy more effectively than in highly crystalline or rigid structures, such as pure HDPE, which tend to fail in a more brittle manner. Changes in hardness are closely linked to the degree of crystallinity; higher crystallinity results in increased molecular packing, leading to greater resistance to deformation. Conversely, reduced crystallinity introduces more amorphous regions, lowering the material's hardness [58]. In contrast, composites with high CaCO₃

content or pure HDPE, while stiffer, tend to be more brittle and less capable of absorbing impact energy, resulting in lower impact strength. The rigid filler particles restrict chain mobility, and at high loading levels, stress concentration around filler particles can cause early crack initiation and propagation under impact [59]. Thus, despite lower stiffness, the increased ductility and plastic deformation potential of recycled plastic-based composites account for their superior impact performance. A noticeable variation was evident in the impact strength of the HDPE-based blends, as shown in Figure 12, with changes in composition.

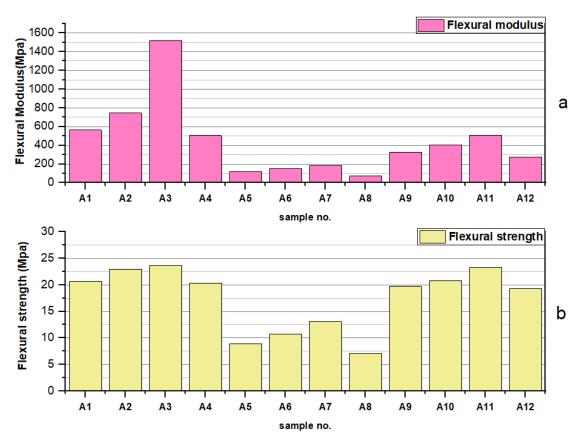


Figure 11: Flexural properties of HDPE composites with varying percentages of CaCO₃ and recycled plastic — (a) Flexural Modulus and (b) Flexural Strength

Table 5: Impact Test Results of HDPE Blends with Varying Proportions of Calcium Carbonate and Recycled Plastics

Sample	Impact Strength $(kJ/m^2) \pm SD$	
A1	24.84 ± 0.18	
A2	23.79 ± 0.38	
A3	7.84 ± 0.42	
A4	29.44 ± 1.20	
A5	57.20 ± 5.26	
A6	28.31 ± 0.47	
A7	15.06 ± 0.69	
A8	76.71 ± 9.15	
A9	44.22 ± 3.22	
A10	43.63 ± 2.49	
A11	39.87 ± 5.06	
A12	69.91 ± 2.48	

The tests revealed a singular trend, with an increase in CaCO₃, in the impact strength testing of the HDPE-based blends. All samples, from A1 to A12, with varying ratios of HDPE to CaCO₃ to recycled plastics, exhibited different levels of impact strength depending on their respective compositions. The concentration of CaCO₃ impinged greatly on the mechanical properties, notably impacting strength-the precarious equilibrium common to rigidity, and toughness, in these blends. Sample A8 consisted entirely of recycled plastics and exhibited the highest impact strength, approximately 73 kJ/m². It is suggested that recycled plastics could provide better energy absorption and flexibility, allowing the material to resist rapid impacts. Such high formation resistance

may result from the inherent ductility of the recycled plastics, as they can deform and absorb the impact energy of a blow more easily than rigid materials.

Conversely, sample A5 exhibited lower impact strength than 55 kJ/m² but still retained its strength. The presence of 25% CaCO₃ means the filler offers rigidity, which reduces the flexibility that recycled plastics typically provide. The toughness of this blend remained reasonably high, as the compromise between rigidity (from CaCO₃) and ductility (from recycled plastics) enabled a significant absorption of energy during impacts. Sample A6 registered an impact strength of 28.31 kJ/m²—A6 with an equal mix of 50% CaCO₃ and 50% recycled plastics. Sample A7 showed an impact strength of 15.05 kJ/m². A7 consisted of 75% CaCO₃ and 25% recycled plastics, resulting in a sample that was significantly stiffer than A6 but less flexible. The high concentration of CaCO₃ reduced the ductility of the material, making it more brittle.

Sample A1 showed an impact strength of 24.83 kJ/m². A1 contained 75% HDPE, which was relatively flexible, and 25% CaCO₃, which increased rigidity. However, the inclusion of CaCO₃ enhanced the material's stiffness, resulting in a reduction in toughness compared to pure HDPE, as reflected in its impact strength of 24.83 kJ/m². The material was still able to absorb some impact energy, but it wasn't as flexible as pure HDPE due to the presence of CaCO₃. Sample A2 showed an impact strength of 23.78 kJ/m². The high proportion of CaCO₃ increased rigidity further, but the presence of HDPE mitigated the negative effect on toughness. This blend offered a compromise between rigidity and toughness, as reflected in its slightly lower impact strength of 23.78 kJ/m² compared to A1. The increased stiffness provided by CaCO₃ improved the material's structural integrity. Still, it came at the cost of flexibility, making the material less effective at absorbing high-impact energy than blends with higher HDPE or recycled plastic content. Sample A3, consisting of 75% CaCO3 and 25% HDPE, had the lowest impact strength at around 7 kJ/m². This drastic reduction in impact resistance was attributed mostly to the substantial proportion of CaCO₃, which enhanced rigidity but reduced the material's ability to deform under impact. The presence of a large filler component contributed to the material's brittle behavior, causing it to break more easily under stress. Sample A4, made from pure HDPE, had a moderate impact strength of around 29.44 kJ/m². As expected, HDPE itself provided a balance of strength and flexibility. However, compared to the recycled plastic-based samples, it showed a lower energy absorption capacity, as HDPE is inherently more rigid and less flexible than recycled plastics. This highlighted the role of recycled plastics in enhancing impact resistance, a property that HDPE alone could not match. Samples A9 to A12, which had more balanced compositions of HDPE, CaCO₃, and recycled plastics, exhibited intermediate values of impact strength. Specifically, A9 exhibited an impact strength of around 44.22 kJ/m². This suggested that a relatively low concentration of CaCO₃ contributed to increased rigidity while the higher percentage of recycled plastic maintained decent toughness, offering a good compromise between strength and flexibility. A10 had an impact strength of around 43.63 kJ/m², indicating a slight decrease compared to A9. The increase in CaCO₃ content suggested a further enhancement in rigidity but at the cost of some toughness. This balance showed that as CaCO₃ increased, the material became stiffer but slightly more brittle. A11 showed the lowest impact strength among the balanced samples at 39.87 kJ/m², indicating that the increase in CaCO₃ further reduced impact resistance. However, this sample still exhibited moderate toughness due to the presence of 30% recycled plastic. This demonstrated that while increasing CaCO₃ enhanced stiffness, it also led to a decrease in toughness, which highlighted the importance of maintaining a balance between the filler and the polymer matrix. Sample A12, with an impact strength of 69.90 kJ/m², reflects very high energy absorption capabilities and resistance to sudden fracture. The high toughness makes this composite suitable for applications where resistance to impact loads is needed, such as automotive components, housings for consumer goods, or packaging materials. The experimental results demonstrated that the optimal level of toughness was achieved by using a higher proportion of recycled plastics in conjunction with a lower quantity of CaCO₃ [2, 9, 45, 56, 58].

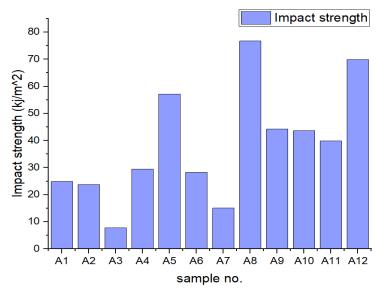


Figure 12: Impact strength dependencies of HDPE loaded with different percentages of CaCO₃ and recycled plastic

4. Conclusion and recommendation

The mechanical properties of HDPE composites with varying degrees of CaCO₃ were analyzed, along with the incorporation of recycled plastics. The key findings included:

- 1) Tensile Strength: The highest tensile strength was exhibited by the pure HDPE. Introducing CaCO₃ at a moderate level (25-50%) gave a balance between the strength and rigidity, whereas anything above 50% resulted in more brittle behavior and loss of tensile strength. The strength of recycled plastics was lower due to degradation during the recycling process.
- 2) Flexural Strength and Modulus: Calcium carbonate improved both flexural strength and modulus, with the optimum values of strength and modulus occurring at 75% by weight of CaCO₃.
- 3) Impact Strength: Recycled materials had higher impact strength, probably due to the additives used to facilitate recycling and the molecular restructuring of polymers during recycling, thereby enhancing energy absorption. Conversely, pure HDPE and samples with higher CaCO₃ content exhibited a low impact strength.
- 4) Hardness: With a rise in the CaCO₃ content, the hardness of the composites also rose, thereby confirming its role as a reinforcing filler.

The article offers recommendations for future research on recycled plastics, identifying three areas for investigation.

- 1) These areas are investigating other types of post-consumer recycled plastics, including PET, PP, and PS, other than the single type investigated in this study.
- 2) Working further on interfacial bonding by compatibilizers or surface-treatment agents such as silane couplers to disperse the filler;
- 3) Investigating scale-up of production processes using twin-screw extruders for better dispersion and homogeneity so that improved reproducible performance of these composites might be obtainable.

Author contributions

Conceptualization, M. Ahmad and Y. Ahmad; data curation, M. Ahmad and Y. Ahmad; formal analysis, M. Ahmad and Y. Ahmad; investigation, M. Ahmad; methodology, M. Ahmad and Y. Ahmad; project administration, Y. Ahmad; resources, M. Ahmad and Y. Ahmad; software, M. Ahmad; supervision, Y. Ahmad; validation, M. Ahmad and Y. Ahmad; visualization, M. Ahmad and Y. Ahmad; writing—original draft preparation, M. Ahmad; writing—review and editing, Y. Ahmad. All authors have read and agreed to the published version of the manuscript.

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Data availability statement

The data that support the findings of this study are available on request from the corresponding author.

Conflicts of interest

The authors declare that there is no conflict of interest.

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