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EB Radiation Processing of HDPE/Rice Husk Ash Composites

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Abstract

High-density polyethylene (HDPE) is a valuable material, but its application under certain operational conditions is limited by oxidation resistance. To mitigate this, rice husk ash (RHA), a silica-rich (~95%) byproduct, was incorporated as a reinforcing filler. This study evaluates the effect of electron beam (EB) irradiation, at doses up to 100 kGy, on the properties of HDPE/RHA composites, focusing on mechanical performance and the polymer–filler interface. The results demonstrate that EB irradiation induces crosslinking and enhances interfacial interaction between the HDPE matrix and RHA filler. While the overall tensile strength of neat HDPE tended to decrease with irradiation dose (from 28.5 ± 1.2 MPa to 24.1 ± 1.5 MPa at 100 kGy), the optimization of dose and filler contents produced notable results: A maximum tensile strength of 29.0 ± 1.1 MPa was achieved in the composite containing 5 wt% RHA at 75 kGy. Furthermore, irradiation stabilized the material's behavior, resolving the heterogeneous dispersion observed in non-irradiated samples with low RHA content. Regarding toughness, Izod's impact resistance increased from 3.2 ± 0.2 kJ/m^2 to 3.7 ± 0.3 kJ/m^2 for the 10 wt% RHA composites irradiated at 50 kGy. Statistical analysis (ANOVA, $p < 0.05$) confirmed the significance of these changes. In conclusion, electron beam irradiation is an effective tool for optimizing the mechanical properties and performance uniformity of HDPE/RHA composites, making them promising candidates for applications requiring enhanced durability and consistency, such as food packaging.

Keywords: HDPE; RHA; EB irradiation; crosslinking; mechanical properties

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1. Introduction

High-density polyethylene (HDPE) is a petroleum-derived thermoplastic requiring 1.75 kg of petroleum per kilogram produced. Discovered in the 1950s, HDPE rapidly became the most useful alternative to low-density polyethylene (LDPE). It has a reduced number of branches, and its consistency creates a resistant material for several large-scale utilization purposes. With a predominantly linear structure, HDPE offers higher strength, chemical resistance, and versatility, making it suitable for various industrial and consumer applications [1,2]. Consequently, the development of naturally reinforced materials like composites has become a primary focus in research on various types of green composites [2–4].

Radiation processing, an accelerated energetic procedure used to achieve profound restructuring of polymers, is a promising technique for both recycling and modifying immiscible blends [5], indicating the suitability of these commercial polyolefins for modification in the presence of the second phase. During radiation exposure, the formation of reactive radical species promotes chemical bonding between polyethylene (PE) and polypropylene (PP) macromolecules, significantly improving their compatibility without the need for a third component [6]. This approach is recognized as a universal method for reactive compatibilization in polymeric blends [7–9]. Several studies on gamma-irradiated PP/HDPE blends revealed notable changes in polyethylene behavior [6,10,11]. The irradiation increased impact resistance, although a higher HDPE content (from 10 to 20 phr) reduced this resistance due to particle coalescence at nodule interfaces, leading to earlier macroscopic fractures. The morphological characterization confirms that irradiated blends exhibit better compatibility than non-irradiated ones [12]. FTIR analysis indicated that BDDA-grafted molecules facilitated chemical reactions between PP and HDPE chains, enhancing their interactions and improving blend compatibility [11]. This study provides essential insights into the impact reinforcement mechanisms promoted by electron beam irradiation, contributing to the development of advanced polymeric blends with superior mechanical properties. All these examples demonstrate the potential for preparing more stable blends, including composites with inorganic fillers [13].

Radiation technology enhances polymer properties by the formation of covalent bonds through crosslinking (50–200 kGy), increasing thermal, chemical, and mechanical resistance [14]. The extension of the lifespan of composites [15] has been frequently studied. Nezafat et al. [16] investigated the effect of crosslinking by EB exposures that modify the HDPE morphology simultaneously with the improvement of material thermal properties, especially decreasing melting temperatures. Alfaro et al. [9] previously investigated the effects of high EB radiation doses (0–500 kGy) on HDPE/RHA composites, while Ortiz et al. [17] applied an EB treatment on HDPE at 250 kGy, when the polymer matrix was loaded with 20% of RHA to obtain high-performance composites. High-yield EB exposure enables the large-scale production of crosslinked polymers for applications such as cables, packaging films, pipes, foams, and automotive parts [14]. EB polymerization, the radiochemical process usually associated with the intimate modification of polymers by irradiation techniques, is a fast, solvent-free method used in food and medical packaging without initiators [18]. The quantification of radiation effects concerning the mechanisms of continuous EB polymerization is a necessary background in industrial applications [19,20].

RH is a major byproduct of rice processing, and every 0.25 kg of RH is generated by 1 kg of raw rice. Its hard surface, high silica content, and low biodegradability make it difficult to reuse [9]. Unused husk is often stored or burned, causing greenhouse gas emissions, pollution, and health issues such as bioaccumulation and cancer. Despite its limited use as a fertilizer or animal bedding, other sustainable solutions are needed for more intelligent utilization [9,21]. RH, which has efficiently extended the usage of composites over recent years, can be incorporated into polymers for various industrial applications [3,4,9,22]. As it is rich in silica (87–97% SiO_2) [22], rice husk is valuable for high-performance products like fibers, paper, and fertilizers [2]. The extraction of silica via the sol–gel method yields about 20 grades of amorphous nanosilica and mesosilica. Their purity is confirmed by two complementary methods: XRD and FTIR; in contrast, SEM shows spherical mesosilica and irregular nanosilica [22]. Both kinds of SiO_2 exhibit thermal stability, and their particle stability is verified through zeta potential analysis [2,23,24].

Because the characterization of radiation-processed HDPE/RHA composites is not comprehensively described, this study presents the evolution of the corresponding HDPE composites with varying concentrations of RHA after their irradiation with EB at different doses. The aim of this paper is to present a new beneficial material with enhanced mechanical properties.

2. Materials and Methods

2.1. Materials

The polymer material investigated in this study is an HDPE homopolymer with an additive package (Exelene® HDPE 8000UV, Montachem International, Inc., Fort Lauderdale, FL, USA), containing an MFI of 8.0 g/10 min (measured at 190 °C and 2.16 kg load) and a density of 0.961 g/cm^3. RHA (95% silica-based filler) was purchased from Camil® Alimentos, São Paulo, Brazil, as the grade obtained by the moving great reactor method ("MG" type, black tone). This material is currently produced in Brazil [25], and it is usually considered agricultural waste obtained from the rice husk combustion process. The composition of ash is presented in Table 1, previously obtained by the XRF analysis, while in Table 2, some main specific characteristics of RHA are presented.

Table 1. The composition of RHA powder [25].

Concentration (wt%)						
SiO_2	Al_2O_3	Fe_2O_3	K_2O	CaO	Cl	P_2O_5
90.02	0.08	0.01	0.81	0.00	0.03	0.34
MgO	SO_3	TiO_2	ZnO	MnO	LOI *	TC **
ND ***	0.07	ND ***	0.01	0.00	9.88	8.63

* LOI: Loss on ignition; ** TC: total carbon; *** ND: not detected.

Table 2. The main initial specific characteristics of RHA [25] are listed.

Specific Weight (g cm^{-3})	Surface Area (m^2 g^{-1})	Mean Particle Diameter (Retained Fraction, µm)
2.20	39.27	19.74

2.2. Composites Preparation

The HDPE/RHA composites were produced by the extrusion of HDPE-based materials after the addition of RHA as a filler into the HDPE matrix (Figure 1).

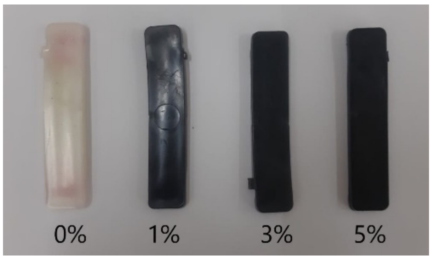


Figure 1. Izod specimens of HDPE/RHA composites, with 0, 1, 3, and 5 wt% of RHA filler.

The HDPE/RHA composites were produced by the extrusion of HDPE-based materials after the addition of RHA as a filler into the HDPE matrix (Figure 1). The ASTM D4976 [26] standard was used to determine the extrusion processing parameters of the composites. The RHA was incorporated in three different weight percentages, 1, 3, and 5 wt%, relative to the mass of HDPE. The average thicknesses were 4.45 ± 0.15 mm. This thickness is appropriate for the application of ASTM D638 [27], because the double-face exposures were applied. After the extrusion process using a co-rotating twin-screw extruder (AX Plásticos Máquinas Técnicas, Diadema, Brazil), the resulting HDPE/RHA composite pellets were subjected to injection molding (Battenfeld®, Battenfeld Cincinnati, Wien, Austria) to prepare the specimens following ASTM D638 and ASTM D256 [28] standards; the preparation was carried out for the characterization of thermal properties, crystallinity degree, gel fraction, and mechanical properties.

The HDPE/RHA composite samples with varying proportions of RHA were irradiated at three doses (50, 75, and 100 kGy) (Table 3). This process was performed inside a Dynamitron® JOB 188 model EB accelerator (Radiation Dynamics, Inc., Edgewood, NY, USA) with a maximum energy of 1.5 MeV, a dose rate of 20 kGy min^{-1} at room temperature, and a maximum current of 25 mA; this process was carried out at the Radiation Technology Center of IPEN-CNEN.

Table 3. HDPE/RHA composite formulation.

RHA (%)	0 kGy	50 kGy	75 kGy	100 kGy
0	0/0	0/50	0/75	0/100
1	1/0	1/50	1/75	1/100
3	3/0	3/50	3/75	3/100
5	5/0	5/50	5/75	5/100

2.3. Measurements

2.3.1. Differential Scanning Calorimetry (DSC)

The thermal analysis was conducted using a Mettler Toledo® simultaneous TGA/DSC thermogravimetric analyzer (Mettler Toledo, Columbus, OH, USA) under a controlled nitrogen atmosphere with a flow rate of 65.0 $\text{mL}\cdot\text{min}^{-1}$ to prevent oxidation during the measurements. Samples of approximately 10 mg were subjected to a heating–cooling–reheating cycle, with heating and cooling rates of 10 $^{\circ}\text{C}\cdot\text{min}^{-1}$, allowing a precise determination of the melting temperature in the region corresponding to the main endothermic evolution. The samples were placed in an alumina sample holder and heated to 600 °C. DSC analyses were performed to record thermal transitions, such as the melting temperature of HDPE, and to evaluate the potential interactions with RHA. The resulting data were analyzed to correlate the effects of ionizing radiation and the incorporated RHA phase on the thermal properties and stability of the composites.

2.3.2. Crystallinity Degree

The heat of fusion (ΔH_m) was calculated from the area under the melting peak region, which is estimated at ~133 °C. The crystallinity percentage (Equation (1)) was determined by comparing the sample's heat of ΔH_m with that of a 100% crystalline sample ($\Delta H_m{}^o$), which is 293 J g^{-1} for HDPE following the adapted literature [29–32]:

$$\%\ X_c = \frac{\Delta H_m}{\Delta H_m{}^o} \times 100 \tag{1}$$

where X_c is the degree of crystallinity, ΔH_m is the specific enthalpy of melting, and $\Delta H_m{}^o$ is the specific melting enthalpy for 100% crystalline HDPE.

2.3.3. Gel Fraction

The gel content is tested according to the ASTM D2765-16 standard [33]. The gel content of the samples of HDPE/RHA composites irradiated at different doses was characterized by comparing the initial mass change with the final mass before and after the test, where the samples were immersed in boiled xylene (purity of $\geq$99%) [16,34] for 24 h and dried at room temperature for 12 h. The initial composite samples had their weights around 1 g. Before boiling, these samples were placed in dense mesh copper bags as small chips.

The error of mass determination was ±2 × 10^{-3} g. The gel content (X_g) in percentage was calculated according to Equation (2):

$$X_g = \frac{m_i}{m_0} * 100 \quad (2)$$

where m_0 and m_i are the sample mass before and after extraction, respectively.

2.3.4. Mechanical Tests

The determinations of tensile strength and elongation at break were conducted following the appropriate ASTM standards for each material: ASTM D638-14 (Standard Test Method for Tensile Properties of Plastics, West Conshohocken, PA, USA, 2022) for "dogbone" specimens of solid HDPE and ASTM D882-12 [35] (Standard Test Method for Tensile Properties of Thin Plastic Sheeting, West Conshohocken, PA, USA, 2018) for thin films, which is applicable in the characterization of RHA composites. Ten specimens per experimental condition were tested. The specimen dimensions of 73 × 12 mm, an average thickness of 2.5 ± 0.1 mm, and a gauge length of 50 mm are the experimental parameters. The tests were performed at a speed of 50 mm/min, according to ASTM recommendations. Stress was calculated as the measured force divided by the initial cross-sectional area of the specimen (MPa = N/mm^2), ensuring consistency with ASTM standards. The measurements were performed using an Instron® 5567 universal testing machine equipped with a calibrated 10 kN load cell (Barrus, Bicester, Oxfordshire, United Kingdom). ASTM D638-14 and ASTM D882-12 standards were adjusted with minor adaptations to accommodate the experimental setup with respect to the presented material grades. Tensile properties, including maximum tensile strength and elongation at break, were recorded to evaluate the mechanical behavior of the samples under axial loading conditions.

Izod impact resistance tests were performed in accordance with ASTM D256 (Standard Test Methods for Determining the Izod Pendulum Impact Resistance of Plastics, West Conshohocken, PA, USA, 2025), ensuring the convergence of our results with information from the literature. The tests were accomplished on both notched and unnotched specimens to evaluate the material's ability to absorb the impact energy. For each condition, ten specimens per sample were tested under identical parameters. The specimens had the following dimensions: 2 mm of notch depth, 0.25 mm of notch root radius, and 12 mm of notch width (entire specimen width). All samples had a sample thickness of 2.5 ± 0.1 mm. The impact tests were carried out at the UNIGEL® laboratory, following standard procedures to ensure the reproducibility and reliability of the results. The comparison between our notched and unnotched specimens provided insights into the material's toughness and its sensitivity to crack propagation.

To determine the Shore D hardness, HDPE/RHA composites were tested after being subjected to different doses of ionizing EB radiation. The test followed the ASTM D2240 standard (Standard Test Method for Rubber Property—Durometer Hardness, West Conshohocken, PA, USA, 2021 [36]) using a Shore D durometer with a 30° conical indenter and a calibrated spring. Each sample was placed on a flat surface. The readings were recorded 5 s after the load was applied. At least five measurements were taken at different points on each sample to minimize local variations. The final hardness was expressed as the average of the obtained values, allowing the evaluation of the effects brought by incorporated RHA and ionizing radiation on the mechanical properties of these composites.

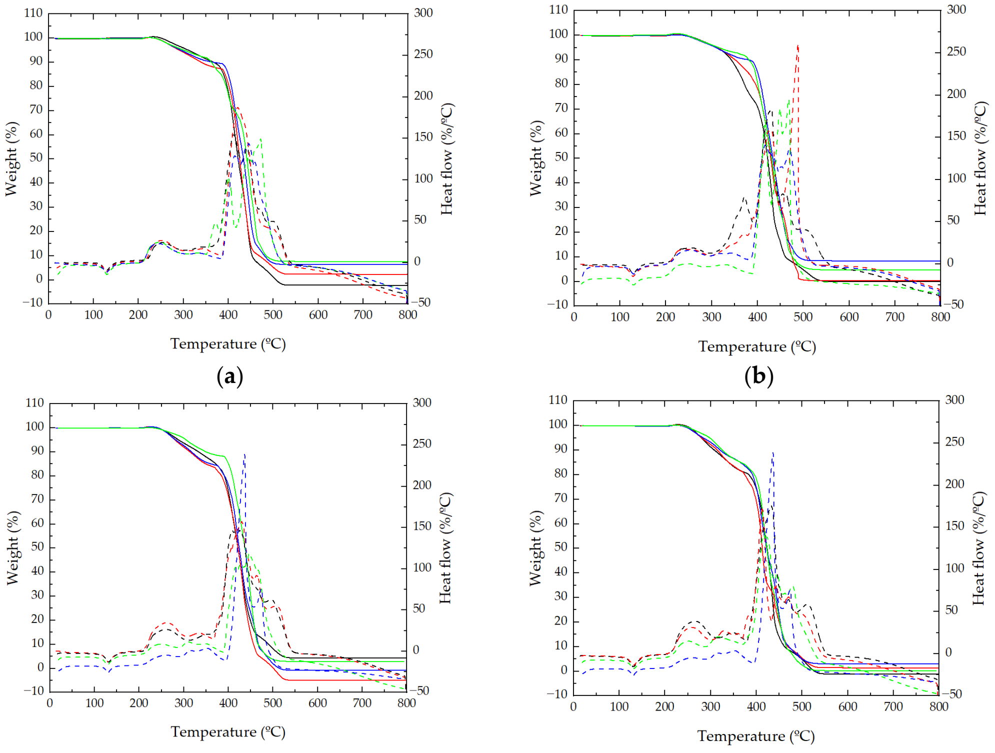
2.4. Statistical Analysis

The standard errors are calculated. One- and Two-way ANOVA were performed, and if the null hypothesis was rejected, Tukey's tests were used to compare the means at a significance level of a *p*-value threshold of 0.05 (95%).

3. Results

3.1. Simultaneous TGA/DSC Analysis and Crystallinity Degree

The results obtained from the DSC analysis are presented in Figure 2. The thermograms obtained for all groups show similar profiles, which are characterized by a single endothermic transition attributed to the melting of HDPE. This detail proves that the thermal behavior is consistent with the literature for the HDPE matrix [22]. The presence of a single, well-defined melting peak without the appearance of multiple transitions or secondary shoulders suggests that the polymer keeps its homogeneity during preparation and irradiation. It is also demonstrated that the investigated composites do not undergo significant degradation or do not generate secondary phases even after their radiation exposure. The data analysis allows the determination of the melting peak temperature (T_m) and the enthalpy change associated with the phase transition (ΔH), for which its values are listed in Table 4. The glass transition temperature of neat HDPE and its composites was not detected, indicating that it likely occurs below 30 °C.



(**a**) (**b**) (**c**) (**d**)

Figure 2. TGA/DSC curves recorded on irradiated HDPE/RHA composites. Loading: (-----) 0 wt%; (-----) 1 wt%; (-----) 3 wt%; (-----) 5 wt%. Dose: (**a**) 0 kGy; (**b**) 50 kGy; (**c**) 75 kGy; (**d**) 100 kGy.

The thermal investigations were performed immediately after the end of radiation treatments, avoiding structural changes due to decay of short-lived radicals.

The average melting temperature of the samples remains around ~120 °C across all analyzed groups, presenting minimal variations [37]. The gel content increases with increasing irradiation doses [37,38]. However, the degree of crystallinity, crystallization temperature, and melting point remain largely unchanged among the samples (131.6–132.7 °C) [16,31,38,39]. After the application of extrusion and injection processes on the composite specimens, the standard samples (unirradiated and free of filler) reached ~50% crystallinity. Thus, they were used as standard specimens for all further comparisons. This indicates that the HDPE matrix preserves its good thermal stability even after exposure to EB medium doses such as 100 kGy. The consistent melting temperature suggests that no significant changes occurred in the crystalline regions of the polymer structure, which would typically affect DSC thermal behavior [39].

Table 4. Fusion temperature, heat flow, and crystallinity degree of the HDPE/RHA samples.

Samples (%/kGy) *	T_f (°C)	ΔH_f (J g^{-1})	% X_c
0/0	132.41	146.5	50
1/0	119.84	106.5	36.35
3/0	116.81	112.2	38.29
5/0	117.53	110.3	37.64
0/50	115.81	112.8	38.50
1/50	116.29	103.1	35.18
3/50	116.51	90.92	31.03
5/50	116.37	141.1	48.15
0/75	117.59	115.4	39.38
1/75	116.60	133.9	45.70
3/75	120.16	105.8	36.11
5/75	115.94	106.1	36.21
0/100	116.82	131.5	44.88
1/100	118.01	134.6	45.94
3/100	118.56	132.6	45.25
5/100	116.03	116.2	39.66

* Here, wt% is the RHA percent, and kGy is the dose applied.

It may be observed that during the degradation stage between 350 and 500 °C, the incorporated RHA underwent active pyrolysis, with ~55% of its average mass lost in the non-irradiated samples. The value is higher compared to the 48% mass loss reported for ~20 wt% RHA in other studies [2,3,6] and also higher than the 44% mass loss observed during thermal degradation in [16]. The heat flow (%/°C) curve shows the highest peak at ~436 °C (3/75 and 3/100) and a shoulder peak at ~487 °C (1/50), indicating the maximum degradation of the RHA and HDPE matrix, respectively. Comprehensive data on how irradiation dose influences the crystalline structure of irradiated HDPE have been reported elsewhere [8,32,38].

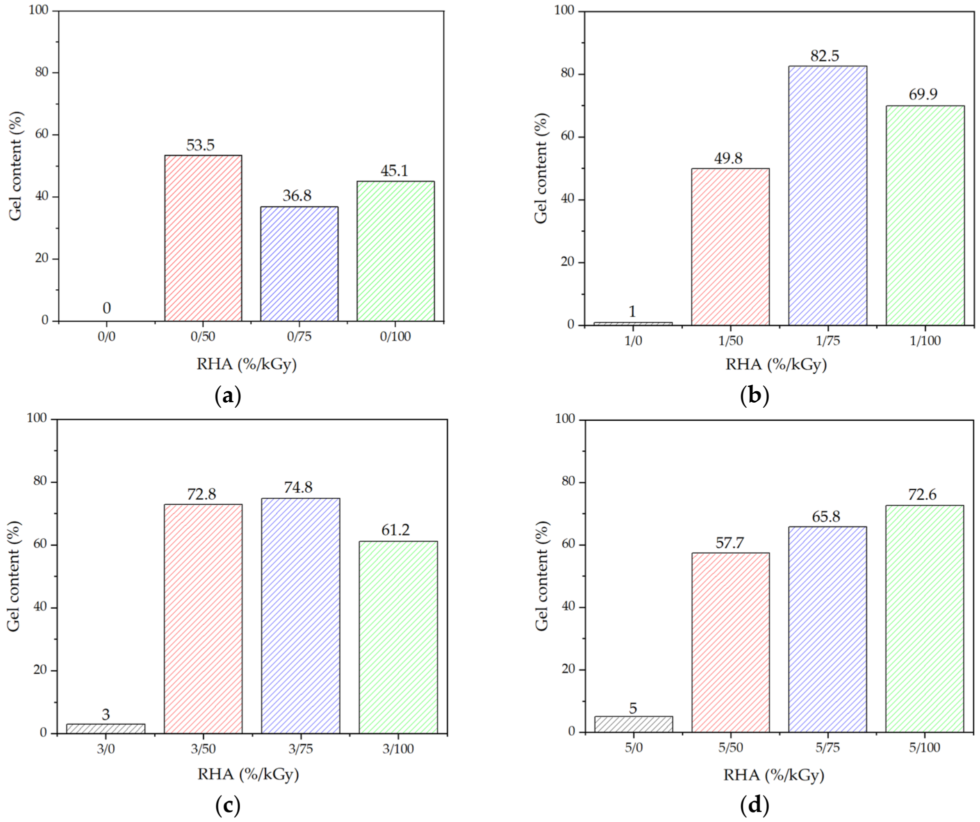
Regarding the addition of RHA at concentrations of 1, 3, and 5 wt%, no significant changes were observed in the values of melting temperatures. This similarity in thermal stability is due to the low filler content [4,22] relative to the polymer fraction, because HDPE represents the dominant phase. The good dispersion of the filler and the absence of aggressive chemical interactions also contribute to the stability of composites [4,22].

DSC results further show that all samples exhibited nearly identical crystallization temperatures, indicating the preservation of a single homogeneous phase during heating. This remark suggests that the incorporation of RHA did not significantly affect the thermal transition temperatures of the polymer matrix. This behavior is consistent with previous findings on the thermal characterization of naturally filled HDPE composites [40].

Additionally, the relatively stable melting enthalpy values indicate that the crystallinity degree of the matrix (% X_c) was not substantially affected by irradiation [32,38]. Some changes during the crystallization of the polymer due to the molecular interactions between the polymer and the surface of RHA occur [4]. Accordingly, the degree of crystallinity directly influences mechanical, thermal, and barrier properties. These results are favorable, opening new paths for the application of polymer/RHA composites. These results suggest that the modifications induced by high-energy irradiation did not compromise the structural integrity of composites among the samples evaluated.

3.2. Gel Fraction

Figure 3 presents the results for the degree of crosslinking of the studied samples as a function of the applied dose, including neat HDPE, for which its null value indicates that neat polyethylene contains no crosslinked content. The increasing dose causes the formation of higher gel content in the processed composites, although the rate of gel formation per unit dose decreases at higher doses [40]. However, beyond a certain dose, crosslinking is completed, causing the gel content to approach its maximum limit, as observed in Figure 3.



(**a**) (**b**) (**c**) (**d**)

Figure 3. Histograms showing the gel content in HDPE/RHA composites as a function of irradiation dose. Irradiation doses: (black) 0 wt%; (red) 50 kGy; (blue) 75 kGy; (green) 100 kGy. Filler loading: (**a**) 0 wt%; (**b**) 1 wt%; (**c**) 3 wt%; (**d**) 5 wt%.

It may be observed that the nonirradiated samples exhibit 100% solubility, because the entire mass was dissolved during the experiment. This solubilization indicates the absence of crosslinking in the initial polymer's structure. For irradiated samples, the degree of crosslinking is determined based on the residual mass after the test. This feature reflects that the accumulated material fraction results from the portion that effectively underwent crosslinking. While the gel content rises with an increase in dose, the degree of crystallinity, crystallization temperature, and melting point remain essentially constant [16,38,39], in good agreement with the thermal results presented in Table 4.

These results show that the degree of crosslinking varied according to the radiation dose and the filler concentration. In Figure 3a, which depicts the comportment of neat HDPE, the highest crosslinking degree is observed in the sample irradiated at 50 kGy (53.5%). This suggests that irradiation promotes a large extent of crosslinking and enhances the material's resistance. However, at 100 kGy, although crosslinking still occurs, polymer chain degradation begins, possibly due to the predominance of radiation-induced chain scission, which compromises the structural integrity of the material and increases this parameter up to 45.1% for the cured phase. Nezafat et al. [16] evaluated the behavior of HDPE under EB treatment in air at room temperature, obtaining a degradation degree of 44%. Khonakdar et al. [24] obtained a crosslinking ratio of 37% using a dose of 100 kGy, while Navratil et al. [37] achieved 60% for the crosslinking of HDPE via exposure at 165 kGy.

In composites containing RHA (Figure 3b,c), another trend may be observed. The highest degree of crosslinking was attained in composites irradiated at 75 kGy (82.5% and 74.8%, respectively), suggesting that these specific doses are the most efficient in promoting HDPE crosslinking in the presence of ash filler. This picture may be related to the interaction between the polymer matrix and the filler, which can influence the progress of degradation by screening free radicals against their oxidation.

These results demonstrate the significant influence of both factors: the radiation dose and the incorporation of filler on the final structure of the EB-processed material. This information suggests the optimal dose range for the maximization of crosslinking without compromising the stability of the polymer matrix. These results allow manufacturers to promote this kind of composite, especially for medical wear, because radiation sterilization does not introduce significant changes in morphological structures. In addition, this treatment may be successfully applied to recycle polymers by radiation processing.

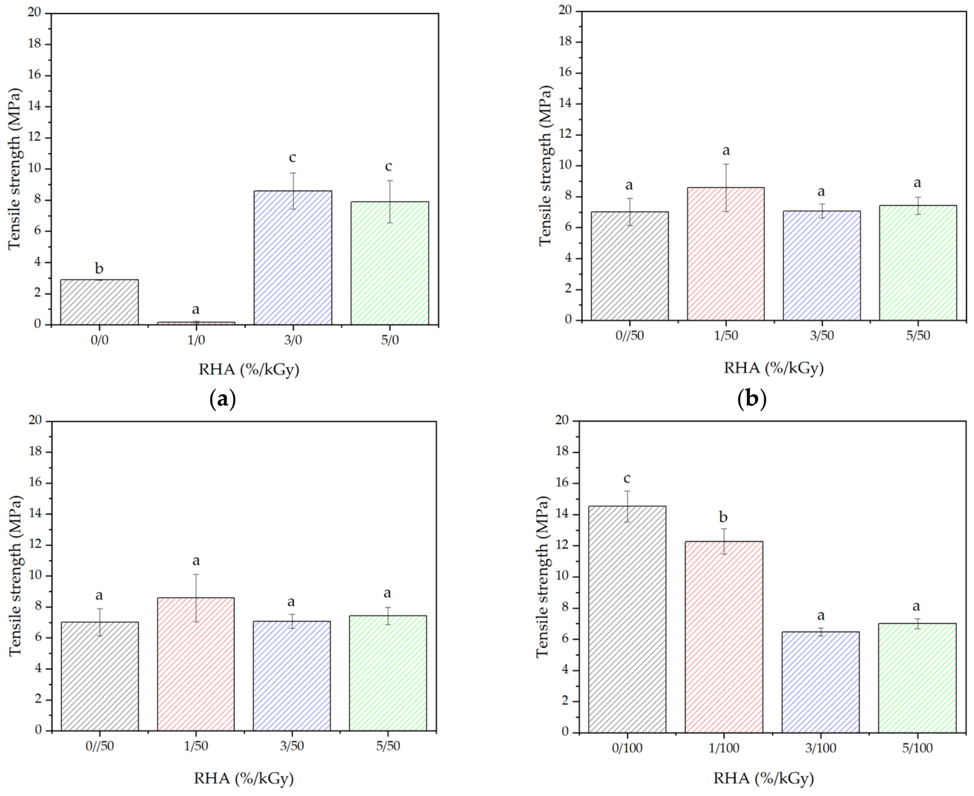
3.3. Mechanical Tests

Figure 4 presents the results of the tensile test performed on the groups subjected to different irradiation doses: 0, 50, 75, and 100 kGy. The inclusion of RHA in the HDPE matrix greatly increased the tensile strength at break [4,17]. The neat HDPE (Figure 4a) sample presents a tensile strength of 3 MPa. This property increases to 8.50 MPa and 7.80 MPa for unirradiated HDPE samples loaded with 3 wt% and 5 wt% RHA, respectively, due to the incorporation of RHA into the HDPE matrix. The low interfacial adhesion between the filler and polymer can lead to the formation of microcracks, facilitating crack propagation during tests [3]. These results indicate that the EB exposure has a positive effect on tensile strength, significantly enhancing their ability to withstand stress before breaking [17]. This effect can be attributed to the radiation-induced crosslinking process [17,32,41], when the formation of covalent bonds between polymer chains reinforces the molecular structure, reduces chain mobility, and increases the internal cohesion of materials.

These findings highlight the potential of EB irradiation as an effective strategy for modifying and optimizing the mechanical properties of polymer composites. Strengthening the polymer matrix by crosslinking can expand the applications of these materials, making them more resistant and durable in various industrial purposes without compromising their structural integrity. Further studies could be conducted to investigate the long-term behavior of these irradiated composites, as well as their performance under different processing and usage conditions.

This investigation indicates a significant increase in the tensile strength at break of HDPE upon the addition of RHA. Improvements of approximately 250% for HDPE reinforced with 3% RHA and about 300% for composites containing 5% RHA were observed. This increase is expected because the addition of 3 and 5% filler allows crosslinking to occur between the polymer and the filler, thus enhancing tensile strength [4,42].

The increase in the tensile strength of irradiated HDPE-based composites (Figure 4) is caused by the crosslinking of the major component. The increasing number of covalent bonds during EB exposure induces the enhanced strength of the polymer phase. Cao and collaborators [43] observed a general increase in the tensile strength of the composites compared to neat HDPE, and this is similarly observed in the present study. It was found that all neat ash composites exhibit significantly higher tensile strength and flexural modulus after their irradiation compared to neat HDPE.



(**a**) (**b**)

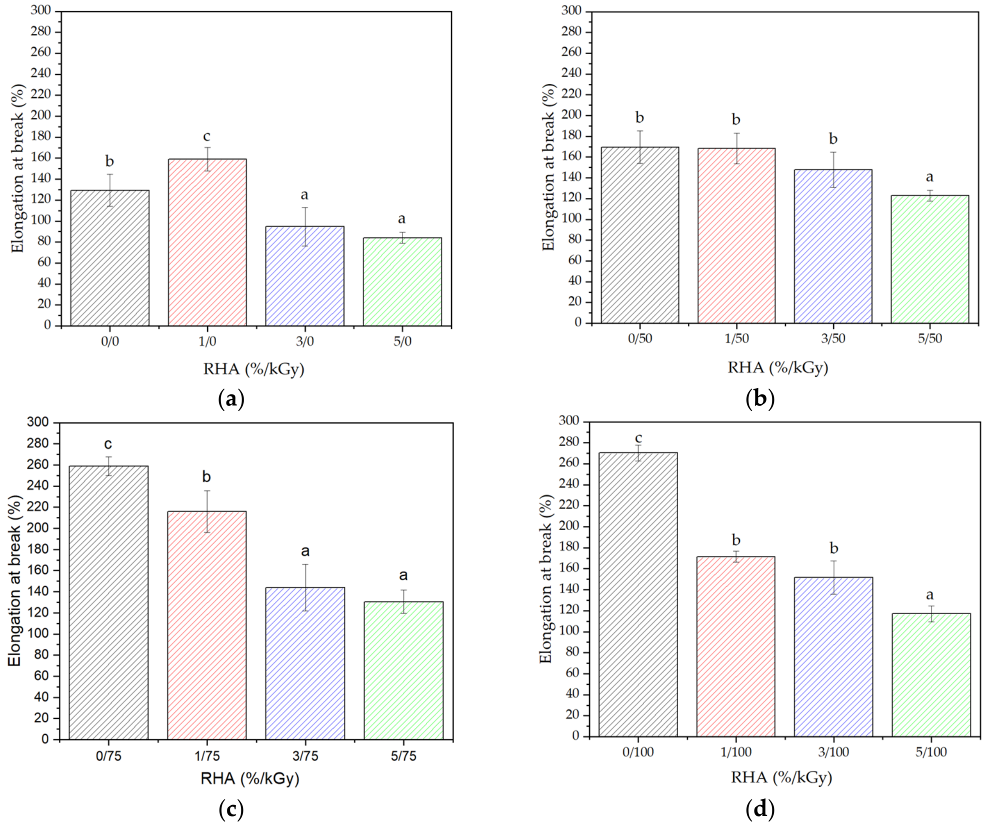
(**c**) (**d**)

Figure 4. Tensile strength of HDPE/RHA specimens as a function of RHA filler loading. Filler contents: (black) 0 wt%; (red) 1 wt%; (blue) 3 wt%; (green) 5 wt%. Irradiation doses: (**a**) 0 kGy; (**b**) 50 kGy; (**c**) 75 kGy; (**d**) 100 kGy.

The values of elongation at break obtained from the tensile tests are presented in Figure 5a–d. A detailed analysis of these data reveals that all irradiated sample groups show a significant increase in their elongation at break, with an average improvement of about 20% in relation to the nonirradiated group of composites. This behavior suggests that the exposure to EB induces structural modifications in the polymer matrix, promoting a competition between crosslinking and controlled chain scission [16].

The crosslinking phenomenon significantly contributes to the formation of a more cohesive and elastic polymer network, providing materials with greater deformation capacity before rupture. This effect was particularly evident in samples irradiated at 50 and 75 kGy. These irradiation degrees enhance ductility without significantly compromising the mechanical strength of the studied composites. The simultaneous development of crosslinking and controlled chain scission may directly influence the reorganization of the molecular structure, allowing greater segmental mobility without the excessive modification in the material's embrittlement. It was proven [41] that the elongation at break increases at a certain gamma dose of 50 kGy, followed by a gradual decrease as the dose increases until fracture occurs.

It is worth noting that moderate irradiation doses improve mechanical properties. At higher doses, for example, 100 kGy, the contribution of degradation would partially compromise the structural integrity of the composite due to molecular fragmentation during radiolysis. This effect has been reported in previous studies, which indicate that beyond certain dose thresholds, degradation outweighs the benefits of crosslinking, leading to reduced mechanical strength and increased brittleness [44,45]. These results highlight the importance of irradiation as an effective technique for modifying the mechanical properties of polymers, enabling the controlled optimization of the flexibility and strength of composites. Furthermore, they highlight the need for precise control of applied dose to prevent degradation mechanisms from surpassing the desired property enhancements.



(**a**) (**b**) (**c**) (**d**)

Figure 5. Elongation at break of HDPE/RHA specimens as a function of RHA filler loading. Filler contents: (black) 0 wt%; (red) 1 wt%; (blue) 3 wt%; (green) 5 wt%. Irradiation doses: (**a**) 0 kGy; (**b**) 50 kGy; (**c**) 75 kGy; (**d**) 100 kGy.

In Figure 5, a reduction in elongation at break can also be observed for samples where RHA 1 wt.% is present. This aspect is caused by the reduced mobility of HDPE chains in the presence of rice husk ash as reinforcements. In particular, for the other two filler concentrations (3 and 5 wt%), the composites' lower deformability can be observed. This relationship is due to the interaction between HDPE and RHA, which contributes to a decrease in the elongation at break [46]. The graphs illustrate the deformation capacity of the materials under stress, for which their elongation-at-break percentages are dependent on the filler's loading. The observed differences between the overall pictures of each group emphasize the influence of experimental variables related to the material's structuration. The studied mechanical properties reflect how each material behaves under stress conditions. The discrepancy observed in non-irradiated HDPE/RHA composites (native materials) at 0 and 1% RHA is mainly related to particle dispersion and interfacial adhesion in the raw materials in the absence of irradiation.

At low filler concentrations, the RHA particles tend to form micro-agglomerations, resulting in heterogeneous stress distributions during the tensile test. Electron beam irradiation promotes crosslinking and improves the interaction between the matrix and the filler, making the mechanical behavior of the irradiated materials (b–c) more uniform and stable. Furthermore, small variations in local crystallinity degrees and particle dispersion in the non-irradiated composites can significantly affect the mechanical response, especially at low RHA contents, where small differences become more noticeable.

EB irradiation primarily causes the formation of crosslinks predominantly in the amorphous chemical structure regions of HDPE [41]. In the less organized zones of the polymer, this crosslinking process significantly increases the degree of intermolecular bonding. As a result, there is a considerable improvement in the material's properties, especially with respect to its mechanical performance. This strengthening of intermolecular bridges contributes to greater resistance and durability in the HDPE phase. The improvement of its mechanical performance under stress by increasing functional properties is structurally linked to the contribution of radiolysis intermediates. This positive effect of crosslinking can be attributed to the increased cohesion between polymer chains, which gives the material a more stable and resilient structure [42].

Figure 6 presents the results of the Izod impact test for the materials studied, highlighting the influence of irradiation and the loadings of RHA on the toughness of the composites. At the same time, Figure 6a–d correspond to the nonirradiated group and patterns subjected to doses of 50, 75, and 100 kGy. The maximum Izod impact resistance was ~ 50 kJ m^{-2} at 0 kGy and 100 kGy for non-notched samples. In contrast, values around ~ 35 kJ m^{-2} were found for all notched and irradiated samples. The analysis of these results discloses that the impact resistance of neat HDPE increases slightly with the incorporation of RHA.

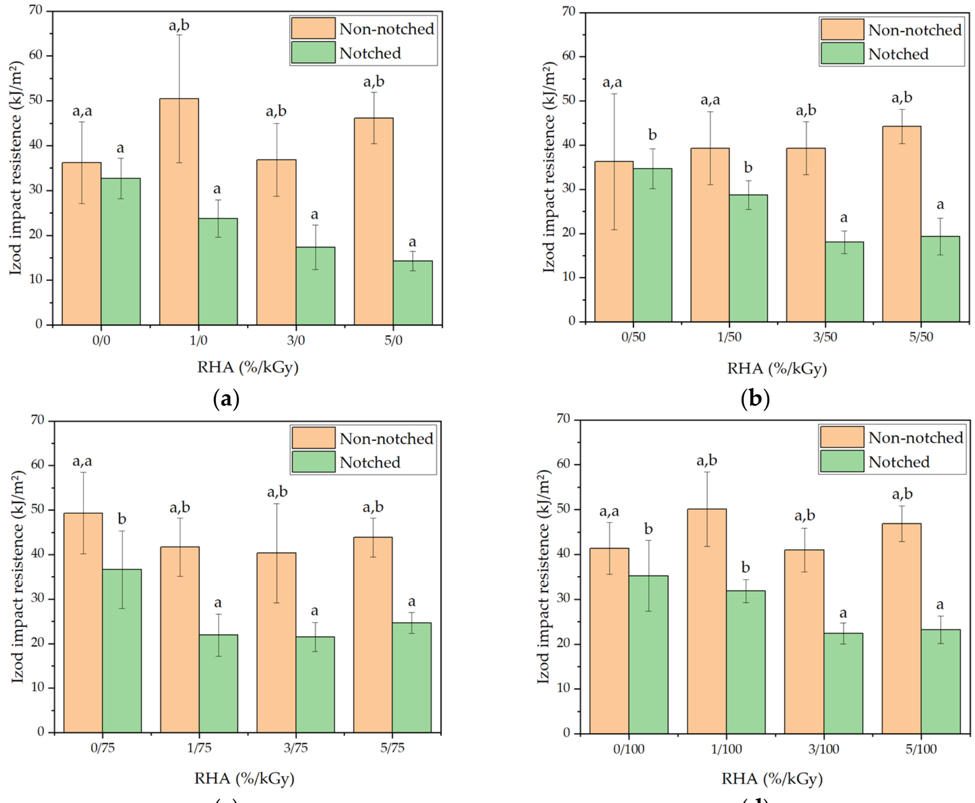


Figure 6. Graphs showing the mechanical response of materials at different irradiation doses. The orange bars represent non-notched specimens, while the green bars represent notched specimens. Each graph corresponds to a specific irradiation dose, ranging from non-irradiated to irradiated materials at different doses: (**a**) 0 kGy, (**b**) 50 kGy, (**c**) 75 kGy, and (**d**) 100 kGy.

This suggests the positive effect of the RHA filler, which is largely composed of silica, on energy dissipation during collision. This behavior can be ascribed to the presence of amorphous silica particles in the RHA, which act as reinforcing agents. It largely emphasizes the energy absorption and reduction in crack propagation in the polymer matrix. Hýlová et al. [47] reported that the increase in indentation hardness at 198 kGy was 4% compared to the non-irradiated neat HDPE, while Ortiz et al. [11] reported 77.3 kJ m (neat HDPE) and 627.3 J m for HDPE/RHA = 80/20 irradiated at 250 kGy.

In the non-irradiated composites, a uniform trend in collision behavior is observed, showing moderate and consistent improvements in their impact resistance relative to the neat HDPE. This information suggests a good dispersion of the filler inside the polymer phase. However, a different tendency may be noticed in the results of the irradiated sample groups, especially at 75 kGy. Under this condition, the neat HDPE exhibits higher impact resistance than its composites. This observation is correlated with the induced structural changes in the polymer matrix, which modulate the filler–matrix interaction and energy dissipation.

The reduction in impact resistance for the irradiated composites may be associated with several factors, such as the formation of microvoids due to the rearrangement of the polymer's morphology, the changes in interfacial adhesion between HDPE and RHA, or the predominance of chain scission over crosslinking. The ionizing radiation generates free radicals in the polymer matrix, representing the fragmentation effect, which are practically involved in the creation of intermolecular bridges that increase the material's stiffness. They may also reduce their impact absorption capacity due to decreased molecular mobility. Additionally, the presence of the filler may interfere with this process, altering the composite's deformation mechanisms.

Another relevant aspect of the effects of radiation on the studied composites is the influence of radiation dose on material stability. At moderate doses (50 kGy), the ratio between crosslinking and degradation may optimize the materials' mechanical behavior, while at higher doses (75–100 kGy), the polymer's structure is slightly degraded, reducing impact resistance. This behavior underscores the complexity of the interaction between the polymer matrix and RHA reinforcement during the irradiation of composites. At the same time, the attained properties indicate the need for further studies to better understand the underlying mechanisms of these changes.

It may be noted that when these results are compared with HDPE composites containing other plant-based fillers, a reduction in impact resistance is evident. The possible correlation with the differing morphological and chemical characteristics of these fillers, such as particle size, porosity, and compatibility with the polymer matrix, may explain this behavior [48].

All injection-molded HDPE/RHA composites exhibit ameliorated impact resistance. This improvement may be attributed to the action of more efficient interfacial adhesion resulting from a well-compacted and consolidated surface in the interphase regions. This statement is also sustained by the action of shear forces during the injection molding process [49].

No significant differences were observed between the HDPE-based composite groups in terms of impact resistance. However, it is worth noting that the sample with the highest concentration of RHA (5%) shows the best impact resistance among the analyzed composites. The highest impact resistance values were recorded in materials irradiated at 50 and 100 kGy, highlighting the combined influence of additive concentration and electron beam irradiation effects on the mechanical properties of the material. These results suggest that the EB irradiation may promote the formation of crosslinks in the polymer matrix, allowing greater energy dissipation capacities during impact.

When the pendulum force was applied in the Izod impact test, it was observed that the presence of reinforcement inhibited the propagation of cracks initiated by the notch previously introduced into the samples and those formed spontaneously during the test.

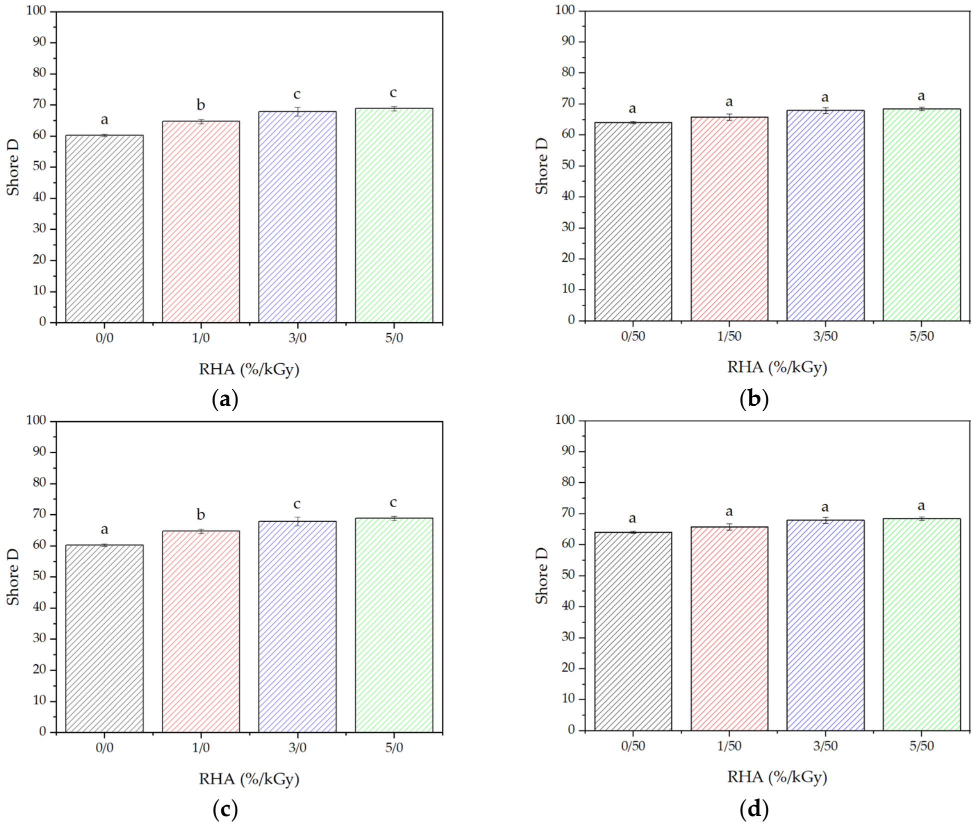
This behavior reinforces the role of the filler in dissipating impact energy, hindering crack advancement. Impact resistance is consistently higher in unnotched samples, where the absence of this stress concentration region allows the material to withstand a higher load before failure. This effect is evident in all analyzed groups because the structural integrity of the polymer matrix is less compromised when no initial discontinuities are present.

Among the studied compositions, the composites with 5 wt% RHA exhibit the highest fracture resistance compared to the other structures. This suggests that this concentration promotes more efficient interactions between the HDPE matrix and the filler. Consequently, better stress distributions result. The addition of RHA into HDPE not only improves impact resistance but also contributes significantly to their structural reinforcement, as evidenced by the results presented in Figure 6. The slight reduction observed in some cases may be ascribed to the structural changes promoted by the presence of the filler, which limits the intermolecular mobility of the functional group's content. Accordingly, the polymer matrix becomes less flexible and more resistant to deformations [50].

In this work, the enhancing effect of EB irradiation is revealed by the formation of crosslinks in the polymer phase and the development of a more rigid and resistant structure. These results demonstrate that the nature of the filler, combined with irradiation treatment, plays a crucial role in modulating the mechanical properties of the composites. They also highlight the importance of properly selecting materials based on the type of reinforcement and structural modification techniques.

EB irradiation at doses higher than 150 kGy does not contribute to the improvement of mechanical properties. This effect is even more evident at doses of 300 kGy, where impact resistance is negatively affected. Thus, at high irradiation doses, polymer chain scission and the formation of structural defects occur, leading to a reduced ability of the composites to dissipate impact energy [51]. For this reason, in this study, irradiation doses of 50, 75, and 100 kGy were used, as they are considered moderate enough to promote the crosslinking of the polymer matrix without inducing chain breakage, thereby contributing to structural reinforcement and improvements in the mechanical properties of the composites.

The hardness results are presented in Figure 7, which shows a comparative analysis among the different studied samples. The hardness of the non-irradiated neat HDPE, as shown in Figure 7a, had an average value of 60 Shore D. This result is in good agreement with the typical values reported in the literature for HDPE [52], which are usually within a range between 60 and 70 Shore D depending on the processing conditions and the molecular structure of the investigated material. Hardness is a fundamental parameter for characterizing the mechanical behavior of polymers. It reflects the material's resistance to penetration and deformation under any applied loads. In the case of neat HDPE, the observed hardness confirms its inherent properties, such as high impact resistance and good stiffness—features that make it widely used in various industrial applications. The incorporation of RHA into the HDPE results in composites with higher hardness compared with neat HDPE. The average Shore D hardness values for the composites ranged from 66 to 68, representing an increase of over 10% relative to the neat material. Navratil et al. [34] obtained a reference value of 65.8 for the neat HDPE specimen, while Zaki et al. [40] measured values of ~48 and ~52 (Shore D) for 0 and 50 kGy doses applied to HDPE. This increase in hardness can be attributed to the developed interaction between the inorganic filler and the polymer, which causes greater material stiffness. RHA acts as a structural reinforcement, reducing the movement of polymer chains and increasing the material's strength against surface deformation. This effect is commonly observed in polymer composites reinforced with mineral fillers or agro-industrial residues, highlighting the potential of RHA as a functional additive [50].



(**a**) (**b**) (**c**) (**d**)

Figure 7. Shore D hardness of HDPE/RHA composites as a function of RHA filler loading. Filler contents: (black) 0 wt%; (red) 1 wt%; (blue) 3 wt%; (green) 5 wt%. Irradiation doses: (**a**) 0 kGy, (**b**) 50 kGy, (**c**) 75 kGy, and (**d**) 100 kGy.

The addition of nanostructured silica as a reinforcement in the HDPE determines a significant increase in Shore D hardness. It was observed that the studied hardness is directly proportional to the silica content, reaching a maximum value with respect to 5 wt%, which improved the material's resistance against plastic deformation. Compared to neat HDPE, this composite showed a 5.65% increase in hardness, demonstrating the positive effect of silica on the material's stiffness [53].

EB irradiation improves the hardness resistance of the composite by promoting structural modifications in the HDPE polymer matrix. The results show an increase of approximately 8 to 10% in Shore D hardness after EB irradiation, indicating significant enhancements in stiffness and resistance to surface deformation.

This effect can be ascribed to the radiation-induced crosslinking, which strengthens the polymer structure, reduces molecular mobility, and makes the material more resistant to impact and wear. Additionally, the reinforcement with RHA further contributes to this increase in hardness by acting as a hardening agent that enhances the cohesion of polymer molecules.

Thus, the present results indicate that the combination of EB irradiation with RHA incorporation introduces a positive impact on the mechanical properties of the studied composite. Accordingly, it becomes a viable alternative for applications requiring greater mechanical strength and durability.

Previous studies have already shown that increasing the filler content in HDPE composites leads to higher Shore D hardness values, which directly result in greater resistance to indentation and mechanical wear. This behavior is associated with the efficient dispersion of the filler in the polymer matrix, which reduces molecular mobility and increases material stiffness [54]. Moreover, the added particles act as a physical barrier against deformation, better distributing the applied stresses and reducing the susceptibility of our composite to surface penetration. This effect is particularly advantageous in applications that demand greater durability and mechanical resistance, such as structural components, coatings subject to impact or abrasion, and the manufacture of fire-retardant materials. Therefore, the incorporation of reinforcement not only improves the hardness properties of composites but also contributes to their long-term mechanical stability and performance [54].

4. Discussion

Radiation processing by ionizing rays is a versatile method by which crosslinkable polymers and their composites attain high structural stability [55]. The nano- and microparticles of silica added to high-performance composites are among the most widely used fillers, improving functional properties and durability when these materials are exposed to extreme energetic inputs like EB, gamma radiation, or high temperatures [56–58]. However, recycling polyethylene in the presence of RHA provides a new route for the preparation of polymer formulations from a natural source of silica [59]. Interest in polymer silica nanocomposites is based on the intimate interaction between the two phases; the structuration of polymers around the silica particles is developed by the formation of strong bridges [60,61], which stabilize the polymer/silica nanocomposites. For irradiated HDPE/RHA samples, the improvement in thermal characteristics is also explained by the reactivity of free radicals involved in the reconstruction of the polymer. Special recommendations have been made [62] for the improvement of the main properties of polymer-based composites, aiming to achieve both enhanced durability and expanded application ranges. The radiation processing of RHA composites exhibits a similar trend, highlighting the practical interest in valorizing natural sources of silica. In this sense, the intermolecular sites of silica particles may immobilize filler particles through covalent bonds [63,64]. During the radiolysis of RHA composites, the materials exhibit suitable morphologies [65], which can be further enhanced by energy transfer and molecular fragmentation. The architectural modifications of irradiated HDPE lead to the restructuring of the polymer phase, the linking of components, and the branching of the macromolecules [66,67]. All of these effects support the measured improvements, including increased material consistency, a higher degree of polymer entanglement around silica particles, and enhanced association at the boundary zones of the composite. According to the radiolytic mechanism of HDPE [42,68,69], the oxygenated products formed during degradation may interfere with the silica particles due to the polarizable fragments. Optimizing the ratio between degradation and crosslinking by irradiation is a key goal that extends the applicability of the material, including the manufacture of medical wear [69]. The valorization of natural sources of silica represents a versatile choice that provides resistant bioactive materials [70]. The addition of silica nanoparticles in the HDPE, followed by high-energy irradiation, makes it possible to generate an improved material with mechanical properties that correspond to market requirements [56]. At the same time, the active surface of nano-SiO_2 particles acts as a crystallization center, and a reduction in melting and crystallization temperatures occurs. Though this treatment induces degradation, the filler particles hinder the propagation of oxidation via the association between organic and inorganic phases [71]. The upgraded HDPE/silica composites, structured during irradiation, open a wide range of applications in innovative food packaging, biomedical implementations, smart electronic textiles, dielectrics and energy storage, and drug delivery [72]. The tuning of functional properties of HDPE/RHA composites renders these nanomaterials suitable for converting polymer waste into products with desirable thermo-mechanical characteristics, and their enhanced properties support long-lasting applications.

5. Conclusions

The effect of radiation-induced crosslinking on HDPE/RHA composites was thoroughly investigated following exposure to EB irradiation. TGA and DSC analyses confirmed that the incorporation of RHA reduced the polymer's crystallinity, while the thermal stability of the HDPE matrix remained robust even after exposure to high EB doses up to 100 kGy with 5 wt% RHA. The melting temperature remained consistently around 120 °C across all analyzed groups, indicating minimal influence of both filler content and irradiation on this characteristic.

A key finding was the direct correlation between increasing radiation doses and higher gel contents, confirming the successful induction of crosslinking within the polymer's structure. Mechanical properties were optimized through a careful balance between filler content and irradiation dose. While the high-dose irradiation of the neat polymer generally led to reduced ductility (as indicated by a decrease in elongation at break), targeted EB irradiation served as an effective tool for property enhancement and stabilization within the composites. The maximum tensile strength of 29.0 ± 1.1 MPa was achieved for the 5 wt% RHA composites irradiated at 75 kGy, a value that is statistically higher than that of the non-irradiated control.

Furthermore, the stabilizing effect of irradiation resolved the mechanical inconsistencies observed in non-irradiated, low-RHA content samples by promoting more uniform particle dispersion and enhanced interfacial interaction. Izod impact resistance also showed a slight but positive increase, rising from 3.2 ± 0.2 kJ/m^2 to 3.7 ± 0.3 kJ/m^2 for the 10 wt% RHA composites irradiated at 50 kGy.

This comprehensive thermal and mechanical characterization provides valuable, quantitative insights into how specific irradiation doses and RHA contents influence performance, contributing to a refined understanding of the potential of EB-irradiated HDPE/RHA composites for engineering applications that demand improved durability and consistent mechanical behavior.

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Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript.

ASTM	American standards for testing and materials
BDDA	1,4-butanediol diacrylate
Dynamitron	Electrostatic particle accelerator
EB	Electron beam
FTIR	Fourier transform infrared spectroscopy
HDPE	High-density polyethylene
H_m	Fusion heat
ΔH	Phase transition enthalpy
ICPE CA	Institute for Electrical Engineering, Advanced Research
IPEN/CNEN	Research Institute for Nuclear Energy/National Commission of Nuclear Energy
kGy	Kilogray
MFI	Melt flow index
SEM	Scanning electron microscopy
SiO_2	Silicon dioxide (silica)
RH	Rice husk
RHA	Rice husk ash
TGA/DSC	Thermogravimetric analysis/differential scanning calorimetry
T_i	Fusion temperature
T_m	Melting temperature
X_c	Crystallinity percentage
X_g	Gel content
XRD	X-ray diffraction
XRF	X-ray fluorescence

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