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# State-of-the-Art Modification of Plastic Aggregates Using Gamma Irradiation and Its Optimization for Application to Cementitious Composites

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**Copyright:** © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). Abstract: In the past few decades, there have been numerous attempts to add plastic aggregates composed of polymeric materials to cementitious composites, either as an alternative to using natural aggregates or as fillers and fibers. However, the addition of plastic aggregates often results in cementitious composites with lower mechanical performance. In this paper, we attempt to address this issue by applying gamma irradiation technology to restore the mechanical performance. We aimed to determine the optimal gamma irradiation and mixing combinations by comparing the experimental results with information summarizing the recent literature related to the use of gammairradiated plastic aggregates within cementitious composites. To this end, the effects of changes in the physical and chemical properties of plastics due to irradiation with gamma irradiation on the strength of cementitious composites were evaluated using irradiation doses of 25, 50, 75, and 100 kGy and various plastic materials as key parameters. In the compressive strength test, it was found that adding gamma-irradiated plastic increased the compressive strength of the cementitious composites compared to the nonirradiated plastic. This suggests that the irradiation of plastic aggregates with gamma rays is an effective method to recover some of the strength lost when plastic aggregates are added to cementitious composites. In addition, modifications in the microstructure and chemical properties of the gamma-irradiated plastic were analyzed through SEM and FT-IR analysis, which allowed the determination of the strength enhancement mechanism. The results of this study show the possibility of the state-of-the-art performance improvement method for using plastic aggregate as a substitute for natural aggregate, going further from the plastic performance improvement technology for limited materials and radiation dose presented in previous studies.

**Keywords:** lightweight aggregate; plastic aggregate; gamma irradiation; irradiated plastic; compressive strength

# 1. Introduction

1.1. Aggregates of Industrial Byproducts Used as Lightweight Concrete

The recent trends in construction automation indicate that modular, preform, and precast structures are on the rise; however, as a result of the materials used in these structures, they are difficult to transport, lift, and install due to their excessive weight, so the development of lightweight structures is essential. Weight reduction is therefore an important goal in the construction industry. Commonly, the replacement of natural aggregates with lightweight aggregates leads to a decrease in unit weight in cement-based materials (mortar and concrete).

M. Priyanka et al. [1] were able to reduce the unit weight of concrete to 1550 kg/m<sup>3</sup> by replacing 60% of natural aggregates in lightweight expanded clay aggregates (LECAs),

which had a strength of over 25 MPa. They concluded that LECAs can be used as structural lightweight concrete.

C.O. Chin et al. [2] conducted a study on lightweight concrete using lightweight activated carbon manufactured from agricultural waste, namely oil palm kernel shell (OPKS), as a coarse aggregate substitute. It was reported that in the case of concrete mixed with OPKS activated carbon, it was possible to secure higher compressive strength compared to natural coarse aggregates by increasing the bond strength with cement paste due to its rough surface and porous structure.

Various technologies related to artificial lightweight aggregates are being developed for the manufacture of lightweight concrete, but their application to structures is limited due to the inherent properties of lightweight aggregates. By using a lightweight aggregate, the concrete unit weight can be reduced by more than 30% compared to natural aggregates; however, the quality, including the mechanical strength, of the concrete decreases due to bonding with cement paste and material separation.

### 1.2. Plastic as an Additive in Cement-Based Materials

The advantageous properties (low density, high durability, high strength-to-weight ratio, low cost, and ease of manufacture) of plastics have led to their use as valuable construction materials utilized as additives (virgin and recycled waste plastic) in conventional concrete over the past few decades. Firstly, they are used as resin to produce a polymer mortar and concrete by adding water-soluble or dispersed polymers in parallel during mixing. Secondly, they are used as plastic powders in the replacement of binder materials, as plastic fibers used in fiber-reinforced concrete, and as plastic aggregates in the replacement of natural aggregates. The latter method will be the focus of this paper. The physical and mechanical properties of cementitious composites fabricated according to different plastic types, shapes, and substitution ratios related to the use (virgin and waste) of plastics in cementitious composites (mortar and concrete) have been investigated by numerous researchers [3–19]. Table 1 provides information on the types, replacement ratios, and investigated properties of plastics used in cement-based composite materials investigated by existing researchers.

Reference	Plastic Type	Shape	Replacement Types and Ratio	Studied Properties
[3]	PE	Powder	Cement: 15, 25, 50, 75, and 100 vol.%	Flexural strength
[4]	PO + PP + PE	Pellet	<sup>1</sup> FA: 10 and 25 vol.%	Density, compressive strength
[5]	HDPE	Pellet /Flake	<sup>2</sup> CA: 25, 50, and 100 vol.%	Density, compressive and flexural strength
[6]	PO + other types	Irregular	CA/FA: 4.7, 7.5, 9.6, 12.5, and 15 vol.%	Density, bond, compressive and tensile strength
[7]	PP, HDPE and PVC	Irregular	CA/FA: 25, 50, and 75 vol.%	Density, compressive strength, conductivity, acoustic insulation
[8]	Tire rubber	Irregular	FA: 10, 20, and 30 vol.%	Workability, durability, impact resistance, compressive strength
[9]	PET	Pellet /Flake	FA: 5, 10, and 15 vol.%	Density, compressive and flexural strength
[10]	PC	Pellet	FA: 5, 10, and 20 vol.%	Impact resistance, compressive strength
[11]	PET	Flake	CA/FA: 5, 10, 15, and 20 vol.%	Impact resistance, compressive strength
[12]	PE	Pellet /Flake /Fiber	CA: 25, 50, 75, and 100 vol.%	Density, bond, compressive and flexural strength
[13]	PP	Pellet	CA: 10, 20, 30, and 40 vol.%	Durability, compressive, tensile and flexural strength

Table 1. Literature studies related to properties of cementitious composites that contain plastic materials.

Reference	Plastic Type	Shape	Replacement Types and Ratio	Studied Properties
[14]	PET	Pellet /Flake	FA: 5, 10, and 15 vol.%	Compressive, tensile and flexural strength
[15]	Various plastic mixed waste	Powder	Cement: 5, 10, 15, 20, and 25 vol.%	Compressive, tensile and flexural strength
[16]	LDPE + red dune sand	Irregular	CA: 25, 50, 75, and 100 vol.%	Density, compressive, tensile and flexural strength
[17]	PET	Flake	FA: 5, 10, and 15 vol.%	Density, compressive, tensile and flexural strength
[18]	PS and LDPE	Pellet	FA: 10, 30, and 50 vol.%	Density, compressive strength
[19]	PET	Fiber	Mixed in concrete: 0.25 vol.%	Density, compressive, tensile and flexural strength

Table 1. Cont.

<sup>1</sup> FA: fine aggregate; <sup>2</sup> CA: coarse aggregate.

From the viewpoint of reducing the weight of concrete, most of the studies reported in the literature suggest that the unit weight of cementitious composites decreases with an increase in the replacement rate of plastic aggregates. Conversely, the compressive strength, splitting tensile strength, and flexural strength decrease with an increase in the replacement rate of plastic aggregates. In addition, it was confirmed that in the manufacture of cementitious composites containing plastics, plastics of a single material were mostly used as aggregates, centering on PET of relatively excellent quality. The replacement rate compared to natural aggregates was investigated in various ways and ranged from 5 to 100%, but 25 and 50% were commonly used in most studies.

#### 1.3. Modification of Plastic Materials Using Gamma Irradiation

The focus of research in the literature described in the previous section was on recycling plastic wastes, rather than treating virgin plastics, to achieve optimal performance before and after mixing them into cementitious composites. Therefore, there is a need for a review that examines potential innovative methods of treating waste plastic before and/or after inclusion within cementitious composites. One literature review has reported that various treatments are used to improve the properties of cementitious composites containing plastics [20]. Plastic aggregates are inherently smooth and hydrophobic and characterized by poor bond formation in the interfacial transition zone (ITZ) between plastic aggregates and cement paste, dissimilarly to natural aggregates; consequently, the inclusion of plastic aggregates in cementitious composites results in decreased compressive strength [4,14,17,21-27]. Therefore, the properties of cementitious composites with the incorporation of plastic aggregates depend significantly upon the treatment applied to the plastic particles [21,28,29]. The type, shape, and surface texture of the plastic used in the cementitious composite are important in relation to the performance of cementitious composites containing plastics. Therefore, it may be hypothesized that the poor bond strength and higher air content between the aggregate and cement paste within cementitious composites containing plastic aggregates are key factors determining their lower performance.

Commonly, chemical surface modifications such as the use of mineral admixtures along with alkaline bleaching (bleaching with NaOH) have been reported to improve the bond properties between plastic particles and the cement matrix [21,30]. It is hypothe-sized that the reaction between plastics and oxidizing chemicals creates certain chemical substances that modify the polymer surfaces of functional groups that may eventually be involved in the cementitious reaction. Thus, a partial chemical bond between cement paste and plastic particles could be possible.

Gamma-ray treatment is attracting attention as a new method to modify the physical and chemical properties of plastic materials without adding or heating special additives and without the release of harmful chemical byproducts. In recent decades, interest in polymer treatment through gamma irradiation has steadily increased as polymer materials have been modified and improved with respect to their chemical, physical, structural, and optical properties. Research related to the improvement of durability and corrosion resistance and the surface hardening of plastics through crosslinking, curing, grafting, and decomposition, including the processing of polymer materials using gamma irradiation emitted from cobalt-60, has been reported. Upon gamma-ray irradiation, chemical reactions occur, as shown in Figures 1 and 2; these reactions change the physical and chemical properties of the plastic. When plastic is irradiated with gamma rays, various chemical reactions occur, as shown in Figure 3. Among these, the most important for improving the physical properties of plastics are crosslinking and chain scission [31].

Ionization	Ρ	>	P+ + e
Excitation	Ρ	>	Ρ*

Figure 1. Major reactions in polymer materials by gamma irradiation.

Recombination	P⁺ + e⁻	)	P* (neutralization)
Free radical formation	P* P*	)	$R_1 \bullet + R_2 \bullet$ (main chain: chain scission) $R_3 \bullet + H \bullet$ (side chain: crosslinking)
Radical recombination	$R_3 \bullet + R_3 \bullet$	)	R-R
Oxidation	$R \bullet + O_2$ $R_1 O O \bullet + R_2 H$ R O O H $R O \bullet + R_3 H$ $2 R O \bullet$ $R_1 \bullet + R_2 O$	···› ···› ···›	R00• $R_100H + R_2$ • R0• + H0•, R00• + R0• + H <sub>2</sub> 0 R0H + R <sub>3</sub> • RCH0 + R•, RCOR + R• $R_10R_2$

Figure 2. Secondary reactions in polymer materials by gamma irradiation.



Figure 3. Various chemical reactions of polymer materials by gamma irradiation.

As shown in Figure 4, by irradiating the plastic with gamma rays, free radicals generated inside the plastic combine with each other to form a three-dimensional network structure; this is called a crosslinking reaction [32]. By increasing the free radical polymerization reaction or network chain, crosslinking occurs between adjacent molecules, thereby rendering the plastic structure dense. Crosslinking polymers by gamma irradiation has the capability of significantly improving their mechanical properties, such as elastic modulus, toughness, strength, and hardness, compared to virgin plastics [33]. The inclusion of these plastic aggregates crosslinked by gamma irradiation into cementitious composites leads to improved mechanical properties [34].



Figure 4. Schematic diagram of crosslinking reaction according to gamma irradiation.

On the other hand, as shown in Figure 5, free radicals are generated inside the plastic chain cut by gamma irradiation, and the molecular weight is reduced; this is called a chain scission reaction. In the case of the oxidation reaction, free radicals react with oxygen to generate various functional groups containing oxygen, such as C–O, C=O, COO, and COOH. Chain scission and oxidation reactions induced by gamma irradiation modify the surface of a highly inert plastic to generate various hydrophilic functional groups on the surface, giving adhesiveness and hydrophilicity, thus making it possible to improve the bonding strength with cement when applied to cementitious composites. In addition, it has been shown that the crystallinity of the plastic is increased due to the chain scission reaction, giving the plastic material a higher modulus, toughness, stiffness, strength, and hardness [35].



Figure 5. Schematic diagram of chain scission reaction according to gamma irradiation.

Although the two mechanisms of crosslinking and chain scission usually occur simultaneously and competitively, one mechanism is more prominent than the other depending on the level of the gamma irradiation dose and the polymer type [36–39].

## 1.4. Cement-Based Material Containing Plastic Particles: The Effects of Gamma Irradiation

In the previous section, the modification mechanism of plastics following gamma irradiation was analyzed. In this section, the effects of modified plastic materials on cementitious composites under various types of plastic aggregates and gamma irradiation doses are described, and a review of the literature is presented in Table 2.

Table 2. Literature studies related to properties of cementitious composites that contain gamma-irradiated plastics.

Reference	Plastic Type	Application Type	γ Irradiation Dose Levels	Major Observations
[40]	Nylon	Fiber	5, 10, 50, and 100 kGy	<ul> <li>Cement composites with 1.5, 2.0, and 2.5 vol.% of gamma-irradiated nylon fibers.</li> <li>In the case of 50 kGy, yield tensile stress was 143.9% higher than that of nonirradiated fiber specimen, and the compressive strength was increased by 292.3% compared to the nonirradiated fiber specimen.</li> </ul>
[41]	PET	Binder (cement substitute)	10 and 100 kGy	• The compressive strength of concrete containing gamma-irradiated plastic increased by 20% compared to nonirradiated concrete.

Reference	Plastic Type	Application Type	γ Irradiation Dose Levels	Major Observations
[42]	PP	Fiber	5, 10, 50, and 100 kGy	<ul> <li>Fibers blended at 0, 1.0, 1.5, and 2.0% in volume with Portland cement, gravel, sand, and water.</li> <li>The highest values of compressive strength were obtained with irradiated fibers at 10 kGy and with 1.5% volume of fiber. The result is 101 MPa, as compared to 35 MPa for simple concrete without fibers.</li> <li>The highest values of compressive strength were found for concrete containing 1.5% in volume of irradiated fibers at 10 kGy. The result is 101 MPa, as compared to 35 MPa for simple concrete without fibers. Thus, there is a compressive strength improvement of 189% with respect to simple concrete.</li> <li>In the case of tensile strength, with 5 kGy dose, the yield tensile stress was 92.1 MPa, which was 14% higher than that of the nonirradiated fiber specimen. On the other hand, the tensile strength decreased at higher irradiation doses.</li> </ul>
[34]	PP	Fiber	5, 10, 50, 100, and 150 kGy	<ul> <li>The compressive strength of concrete containing 1.5% in volume of irradiated fibers at 50 kGy was measured to be 19% higher than nonirradiated concrete.</li> <li>Above a certain irradiation dose (50 kGy), the compressive strength decreased.</li> </ul>
[43]	Tire rubber	Fiber	50 and 100 kGy	<ul> <li>Compared to concrete without fibers, the compressive strength was increased by up to 26% when containing 1.2% in volume of irradiated fibers at 50 kGy.</li> <li>The flexural strength was increased by 48% compared to nonirradiated concrete when containing 1.2% in volume of irradiated fibers at 100 kGy.</li> </ul>
[44]	SAE <sup>1</sup> + NCC <sup>2</sup>	Binder (cement substitute)	10, 20, 30, and 50 kGy	<ul> <li>Compressive strength increased with up to 2~8% of SAE-NCC content and decreased thereafter; compressive strength increased with gamma irradiation dose up to 30 kGy and then decreased.</li> <li>The highest values of compressive strength were found for concrete containing 8% in volume of SAE-NCC binder irradiated at 30 kGy dose.</li> </ul>
[36]	PET	Fine aggregate	100, 150, and 200 kGy	• The highest values of compressive strength were found when fine aggregate was replaced with PET flakes irradiated at 100 kGy dose with 1% in volume of the fine aggregate volume, and it was increased by 34% compared to nonirradiated concrete.
[20]	PET, PC, and tire rubber	Fine aggregate	100 and 200 kGy	<ul> <li>The compressive strength of mortar according to the type of polymer was highest in the order of PET, PC, and tire rubber.</li> <li>The compressive strength of the mortar according to the irradiation dose increased as the dose increased up to the 200 kGy dose.</li> </ul>

Table 2. Cont.

<sup>1</sup> SAE: styrene–acrylic ester; <sup>2</sup> NCC: nanocalcined clay.

Based on the information contained in the reviewed literature, the mechanism behind the performance improvement in plastic materials by gamma irradiation is confirmed, and it can be concluded that the mechanical properties of cementitious composites are improved. One study reported that it was possible to modify the smooth surface of the plastic aggregate to a rough surface as the gamma irradiation dose increased, which improved the mechanical strength of concrete. This results from the fact that gamma irradiation leads to an improvement in bond strength between the cement paste and the plastic aggregate due to the rough particle surface formed by modifying the molecular structure of the polymer. From these observations, it can be concluded that gamma irradiation is a suitable technique for improving the performance of polymeric materials in composites and could be applied to mortar and concrete.

However, in most cases, studies were mainly conducted on mortar or concrete with polymers added as fibers or fillers, and few studies were on cement composites with plastics added as aggregates. Martinez-Barrera et al. [20,36] conducted a study on mortar and concrete containing irradiated polymers as aggregates, but mostly high doses of gamma irradiation were used, and the types of polymers were also limited. Therefore, it is necessary to optimize the gamma irradiation dose for various plastic types by increasing the dose step by step from an initial low dose and to then analyze the changes in characteristics associated with the various dose levels that are tested.

# 2. Experimental

# 2.1. Materials

For preparing the specimens, ordinary Portland cement (OPC) was used, and the chemical compositions and physical properties are shown in Table 3. ISO standard sand was used as an aggregate, and there were a total of five types of plastic fine aggregates used as a substitute for sand, as shown in Figure 6; these were obtained from the Korea Plastic Single Material Association. From the viewpoint of studying the inherent properties of raw plastics, we focused on plastic aggregates in which the plastic had not undergone secondary processing. The plastics used in this study were in the form of pellets or flakes with a diameter of 4–6 mm, as shown in Figure 7, and the basic properties are shown in Table 4.

Table 3. Chemical compositions and physical properties of cement.

Chemical Composition (%)						Blaine	Density	
SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	SO <sub>3</sub>	K <sub>2</sub> O	(cm <sup>2</sup> /g)	(g/cm <sup>3</sup> )
21.9	5.2	3.6	63.2	2.0	2.2	1.0	3480	3.15



Figure 6. Plastics for gamma irradiation and cement composite: (a) PP; (b) PE; (c) PC; (d) ABS; (e) PET.



**Figure 7.** Image of the plastic aggregates showing their shape and size (numbers on the ruler indicate cm): (a) pellet; (b) flake.

Plastic Type	РР	PE	PC	ABS	PET
Density (g/cm <sup>3</sup> )	0.92	0.94	1.20	1.04	1.37
Tensile strength (MPa)	24~27	18~20	57	37	26
Elongation (%)	300	200	105	54	50~150
Flexural modulus (MPa)	1200~1250	815	2244	2198	2800~3100

Table 4. Physical properties of plastic aggregates.

### 2.2. Gamma Irradiation Procedure

For the optimization of the surface modification of plastic aggregates through gamma irradiation, the gamma irradiation doses were set at four levels of 25, 50, 75, and 100 kGy, with reference to previous studies [20,34,36,40–44] that considered the effect of gamma irradiation on the strength properties of cementitious composites at various doses. Irradiation was provided by a cobalt-60 gamma irradiator of automatic pallet type, manufactured by the Nordion Corporation in Canada and located at the Greenpia Technology Corporation in Korea. Plastic aggregates were gamma-irradiated in air at room temperature.

#### 2.3. Characterization of Plastic Aggregates before and after Irradiation

In this study, FT-IR and SEM were used to determine the changes in chemical composition and surface shape morphology of the plastic surfaces resulting from gamma irradiation in order to conduct an analysis of the internal microstructure according to the type of plastic and the gamma irradiation dose. In addition, we aimed to characterize the complex mechanisms by which aspects of the plastic material composition change due to gamma irradiation, contributing to the enhancement of adhesion to cement paste and enhancement of the strength of cement composites.

#### 2.4. Preparation of Cement Composite Specimens

Cementitious composite samples were constructed using a basic process in which the components were mixed with a mortar mixer and poured into 50 mm  $\times$  50 mm  $\times$ 50 mm cube molds, a standard for compressive strength testing. A total of 90 specimens were manufactured with the type of aggregate (sand, PP, PE, PC, ABS, and PET), the range of gamma irradiation doses (0, 25, 50, 75, and 100 kGy), and the curing period (3, 7, and 28 days) as variables. The density of ISO standard sand is 2.61 g/cm<sup>3</sup>, and the density of plastic aggregate is different depending on the type. The fine plastic aggregate was replaced with 50% of the volume of ISO standard sand in consideration of factors such as the density, and the effect on the properties of the cementitious composite material according to the plastic type and gamma radiation dose were evaluated. Here, the total volume of the cementitious composites considers the density of plastic aggregates shown in Table 4. A constant water-to-cement ratio of 0.5 was used for preparing all specimens, the mixing procedure was performed according to the KS L ISO 679, and the mix design is shown in Table 5. After 24 h, the specimens were removed from the molds and cured in tubs of water for 3, 7, and 28 days.

Smaainnan	Cement	Water	Fine Aggregate (kg/m <sup>3</sup> )				Vol. Ratio		
Specimen	(kg/m <sup>3</sup> )	(kg/m <sup>3</sup> )	Sand	PP	PE	РС	ABS	РЕТ	(%)
Control			1524	-	-	-	-	-	
PP50				268.6	-	-	-	-	
PE50	509	254		-	273.0	-	-	-	FO
PC50	508	234	762	-	-	350.3	-	-	50
ABS50				-	-	-	303.6	-	
PET50				-	-	-	-	400.3	

Table 5. Mix design of cementitious composites.

#### 2.5. Physical and Mechanical Tests

The unit weight of cementitious composites according to the type of plastic aggregate was measured using the KS F 2462 standard. The compressive strength tests of the cementitious composites were performed on cube-shaped specimens of 50 mm<sup>3</sup> at three curing periods of 3, 7, and 28 days using an Instron Universal Testing Machine (UTM) of capacity 1000 kN and according to the KS L ISO 679 standard. This procedure was carried out for each of the six specimens of the 30 cementitious–plastic composite combinations.

#### 3. Results and Discussion

## 3.1. Morphological Characterization by SEM Analysis

The morphological characterization of gamma-irradiated plastic particles was performed as follows: First, specimens were cut with a Struers model Secotom-50 micrometer that produces very thin and uniform cuts, and then they were vacuum-coated with platinum in Hitachi model JP/E-1010. Finally, the surfaces were analyzed by scanning electron microscopy (SEM) in a JEOL model JSM-6500F machine in the secondary-electron mode. The results of analyzing the morphological surface shape changes in the five types of plastic aggregates irradiated at each gamma-ray dose using SEM are shown in Figure 8.



Figure 8. SEM images of polypropylene (PP) aggregate: (a) 0 kGy; (b) 25 kGy; (c) 50 kGy; (d) 75 kGy; (e) 100 kGy.

All cases of raw plastic aggregates that were not gamma-irradiated had a smooth surface, as shown in Figures 8a, 9a, 10a, 11a and 12a, respectively. However, surface degradation began when gamma irradiation was applied. A rough surface was obtained at 25 kGy. For higher doses, a rougher surface with segregated particles and cracks was obtained. These morphological changes are a consequence of the scission of polymer chains inside plastic aggregates. Moreover, such morphologies develop more contact points and greater superficial area on the plastic particles, and a more compact surface is obtained, as shown in Figure 13. In summary, the plastic particles modified by gamma irradiation improve the interfacial bond strength with the component surface of the cementitious composite, contributing to having higher compressive strength values as a result of the physical interaction.



**Figure 9.** SEM images of polyethylene (PE) aggregate: (**a**) 0 kGy; (**b**) 25 kGy; (**c**) 50 kGy; (**d**) 75 kGy; (**e**) 100 kGy.



**Figure 10.** SEM images of polycarbonate (PC) aggregate: (**a**) 0 kGy; (**b**) 25 kGy; (**c**) 50 kGy; (**d**) 75 kGy; (**e**) 100 kGy.



**Figure 11.** SEM images of acrylonitrile butadiene styrene (ABS) aggregate: (**a**) 0 kGy; (**b**) 25 kGy; (**c**) 50 kGy; (**d**) 75 kGy; (**e**) 100 kGy.



**Figure 12.** SEM images of polyethylene terephthalate (PET) aggregate: (**a**) 0 kGy; (**b**) 25 kGy; (**c**) 50 kGy; (**d**) 75 kGy; (**e**) 100 kGy.



Figure 13. SEM images of cement.

## 3.2. Chemical Characterization by FT-IR Analysis

Chemical characterization of gamma-irradiated plastic particles was analyzed by using FT-IR microscope, in a BRUKER model VERTEX 80v coupled to HYPERION 2000 machine. FT-IR analysis was performed to analyze the chemical properties of the plastic aggregates as a result of the effects of gamma irradiation. Referring to the existing literature [45], the absorption bands for the FT-IR spectra are summarized in Table 6 and were used to analyze the properties of each plastic. Figures 14–18 show a comparison of FT-IR spectra changes according to the gamma irradiation dose for each plastic type. The symbols assigned to the absorption bands used for identification by plastic types in Table 6 are matched and displayed in Figures 14–18. The absorption band peak in the spectra for each plastic type was found to be mostly similar to the vibrational characteristics of the absorption band peak of each plastic material described in the existing literature.

Table 6. List of important vibration modes and mode assignments for the FT-IR spectra [45].

Plastic Type	Molecular Structure	Absorption Bands (cm <sup>-1</sup> ) for Identification Indicating the Peak Position and Mode Assignments
Polypropylene (PP)	$     \begin{bmatrix}       H & H \\       - C & -C \\       - C & -C \\       - H & -H_3 \\       H & CH_3 \end{bmatrix}_{n} $	2950 (a): C–H stretching 2915 (b): C–H stretching 2838 (c): C–H stretching 1455 (d): CH <sub>2</sub> bending 1377 (e): CH <sub>3</sub> bending 1166 (f): CH bending, CH <sub>3</sub> rocking, C–C stretching 997 (g): CH <sub>3</sub> rocking, CH <sub>3</sub> bending, CH bending 972 (h): CH <sub>3</sub> rocking, C–C stretching 840 (i): CH <sub>2</sub> rocking, C–CH <sub>3</sub> stretching 808 (j): CH <sub>2</sub> rocking, C–C stretching, C–CH stretching
Polyethylene (PE)	$ \begin{array}{c}     H & H \\     -C & -C \\     -C & -C \\     H & H \\     H & H \\     n \end{array} $	2915 (a): C–H stretching 2845 (b): C–H stretching 1472 (c): CH <sub>2</sub> bending 1462 (d): CH <sub>2</sub> bending 730 (e): CH <sub>2</sub> rocking 717 (f): CH <sub>2</sub> rocking
Polycarbonate (PC)	$ \begin{bmatrix} 0 & \xrightarrow{CH_3} & 0 \\ I & \xrightarrow{I} & 0 \\ CH_3 & 0 & -C \\ CH_3 & 0 & -C \end{bmatrix}_n $	2966 (a): C–H stretching 1768 (b): C=O stretching 1503 (c): Aromatic ring stretching 1409 (d): Aromatic ring stretching 1364 (e): CH <sub>3</sub> bending 1186 (f): C–O stretching 1158 (g): C–O stretching 1013 (h): Aromatic CH in-plane bending 828 (i): Aromatic CH out-of-plane bending
Acrylonitrile butadiene styrene (ABS)	$ \begin{array}{c} \begin{pmatrix} H & H \\ C & C \\ H & C \equiv N \end{pmatrix}_{x} \begin{pmatrix} H & H & H & H \\ C & C = C & C \\ H & H \end{pmatrix}_{y} \begin{pmatrix} H & H \\ C & C \\ H & H \end{pmatrix}_{z} $	2922 (a): C–H stretching 1602 (b): Aromatic ring stretching 1494 (c): Aromatic ring stretching 1452 (d): CH <sub>2</sub> bending 966 (e): =C–H bending 759 (f): Aromatic CH out-of-plane bending, =CH bending 698 (g): Aromatic CH out-of-plane bending
Polyethylene terephthalate (PET)	$\left[\begin{array}{c} O \\ O \\ O \end{array} - C \\ O \\$	1713 (a): C=O stretching 1241 (b): C–O stretching 1094 (c): C–O stretching 720 (d): Aromatic CH out-of-plane bending



Figure 14. FT-IR spectra of polypropylene (PP) aggregate with different irradiation doses.



Figure 15. FT-IR spectra of polyethylene (PE) aggregate with different irradiation doses.

As a result of the FT-IR analysis of the PP aggregate, from Figure 14, in the FT-IR spectrum of neat PP, the peaks observed near 1166 and 977 cm<sup>-1</sup> confirm that the PP used in the present case is isotactic. It is observed that the intensity of these peaks continuously decreases with the increase in gamma irradiation dose, and this indicates that these bonds are the result of chain scission by gamma radiation. Furthermore, new characteristic bands at 1650 cm<sup>-1</sup> appear at all the gamma dose levels. These correspond to the carbonyl C=O groups, and the appearance of the C=O group indicates oxidation. This is because the C-H and C-C bonds of PP are the result of scission and consequently the formation of free radicals and ionized species. The reaction of free radicals with oxygen does not allow the polymer chains to crosslink, which leads to chain scission predominating over crosslinking in PP.



Figure 16. FT-IR spectra of polycarbonate (PC) aggregate with different irradiation doses.



Figure 17. FT-IR spectra of acrylonitrile butadiene styrene (ABS) aggregate with different irradiation doses.

As a result of the FT-IR analysis of the PE aggregate, for nonirradiated PE, because the side chains in polymer structure can move freely, the C–C stretching peak at 1462 cm<sup>-1</sup> is wide, whereas for irradiated samples it changes to a sharp peak. In addition, the C=O stretching peak at the 1700 cm<sup>-1</sup> region increases with an increase in the gamma irradiation dose. The increase in these C=O stretching peaks is due to crosslinking by gamma irradiation.

As a result of the FT-IR analysis of the PC aggregate, when gamma-irradiated, the peak at 1768 cm<sup>-1</sup> is suppressed by gamma-ray irradiation, and a new peak is formed at 1768 cm<sup>-1</sup>, which originates from C=O stretching vibration. This causes the C–O single bonds to be easily broken, compared with C=O double bonds, because the bond energy of C–O is lower than that of C=O. In summary, the mechanism of the surface hardness enhancement for gamma-irradiated PC is closely related to the formation of C=O stretching vibration around 1768 cm<sup>-1</sup> and aromatic hydrocarbons.



Figure 18. FT-IR spectra of polyethylene terephthalate (PET) aggregate with different irradiation doses.

As a result of the FT-IR analysis of the ABS aggregate, it has been confirmed that with the increase in the gamma irradiation dose, there was a decrease in the intensity of the C=O band at around 1768 cm<sup>-1</sup> and C-O band at around 1186 cm<sup>-1</sup>, which is indicative of the chain scission of the surface of PC.

As a result of FT-IR analysis of PET aggregates, it was confirmed that the peaks of the oxygen functional group C=O at 1713, 1241, and 1094 cm<sup>-1</sup> decreased with increasing gamma irradiation dose. Upon gamma irradiation of PET samples, free radicals, ions, and neutral atoms are generated. As a result, it induces a change in PET surface polarity. This is due to the formation of OH and COOH groups during oxidation by the atmosphere. Oxygen in the air reacts with gamma-ray-induced radicals to form C=O bonds. Oxygen released in the H-C=O band can oxidize the surface and improve wettability. In addition, free radicals -C-O-, which improve polarity, can be released. Therefore, gamma irradiation on PET can change the chemical structure of the surface and increase the number of polar groups on the surface, thereby increasing polarity and wettability.

The results of the comprehensive analysis of the FT-IR spectra results for each plastic aggregate are as follows: (a) Depending on the gamma irradiation, the intensity of the identification peak increases or decreases, or a new functional group is formed. Through this, it was confirmed that the chemical structure of the plastic aggregates was changed by gamma irradiation. (b) The functional group on the surface of the aggregate is one of the most important factors for improving the interfacial bonding and mechanical properties between the aggregate and cement paste, and additional analysis and investigation are needed to determine how the functional group improves the structural stability.

#### 3.3. Unit Weight Analysis

The unit weight of concrete depends on the density of the concrete mix composition. Commonly, plastic aggregates have a lower density than natural aggregates. Therefore, the substitution of plastic as an aggregate generally reduces the unit weight regardless of the type and size of substitution.

According to the density analysis of the raw plastics, as shown in Figure 19, the density tended to increase very slightly as most plastic aggregates were irradiated with gam-ma rays. This trend was due to the fact that the internal structure of the plastic became somewhat dense as it was gamma-irradiated. In addition, as is well known, the difference in unit weight for cementitious composites according to the type of plastic depends on the density of the plastic raw material.



Figure 19. Variation in density and unit weight according to gamma irradiation.

#### 3.4. Compressive Strength Analysis

The compressive strength of cementitious composites is considered to be the major parameter determining their suitability for application in the construction field. The purpose of this section is to summarize the compressive strength properties of the cement composites produced according to the main parameters, such as the type of plastic and the gamma irradiation dose. The compressive strength properties at three curing periods of 3, 7, and 28 days were analyzed for cementitious composites containing five types of nonirradiated plastic aggregates in order to first select suitable plastics as aggregates for cementitious composites before using gamma-irradiated plastic aggregates.

In Figure 20, it can be seen that the compressive strength values varied from 16.3 to 22.9 MPa for cementitious composites with nonirradiated plastic aggregates; these were lower than the value of 42.7 MPa obtained for the control specimen, and it was discovered that the use of plastic aggregates as a fine aggregate replacement reduced the compressive strength of the cementitious composites.



Figure 20. Compressive strength of cementitious composites according to plastic type.

The compressive strength of cementitious composites with plastic aggregates depends on the density of the raw plastics. To examine this in detail, the density–stress relationship in raw plastics for each type of plastic was compared, and the results are shown in Figure 21.



Figure 21. Density-stress relationship in cementitious composites by plastic aggregate type.

Since plastic aggregates have different densities, it is necessary to check whether the compressive stress is linearly improved in proportion to the density of the raw plastics. As shown in Figure 21, all plastic aggregates except for PET showed better stress efficiency compared to density than fine aggregates. Among them, PE and ABS showed the best efficiency of compressive stress compared to density because they were the farthest from the trend line formed based on natural sand. Given that the main aim of this study was to reduce the weight of cementitious composites and ensure a certain level of compressive strength, we hypothesized that the use of PE and ABS as optimal aggregates would be the most effective in improving the compressive strength of the cementitious composites.

In order to prevent the reduction in compressive strength while using plastic aggregates as a substitute for fine aggregates, the plastic aggregates were modified using gamma irradiation, and it was necessary to analyze the compressive behavior characteristics of the cementitious composite to which they were applied. Table 7 shows the results of measuring the compressive strength of cementitious composites. As shown in Figure 22, the compressive strength increased linearly and gently as the gamma irradiation dose increased. In particular, when the PE aggregate irradiated with a 75 kGy dose was used, the compressive strength increased by 85% compared to the nonirradiated specimen, and the strength enhancement efficiency by gamma irradiation was the best of all the tested conditions.



Figure 22. Compressive strength of cementitious composites according to irradiation dose.

Specimen	Irradiation Dose (kGy)	Compressive Strength (MPa)	Increase Rate Compared to Nonirradiated Specimen (%)
PP0	0	19.09	-
PP25	25	25.09	31.4
PP50	50	25.69	34.8
PP75	75	33.02	73.0
PP100	100	32.55	70.5
PEO	0	21.31	-
PE25	25	24.02	12.7
PE50	50	25.76	20.9
PE75	75	39.43	85.0
PE100	100	32.67	53.3
PC0	0	22.02	_
PC25	25	28.81	30.8
PC50	50	30.3	37.6
PC75	75	32.54	47.8
PC100	100	30.02	36.3
ABS0	0	22.89	_
ABS25	25	28.6	25.0
ABS50	50	29.81	30.2
ABS75	75	34.94	52.6
ABS100	100	32.52	42.1
PET0	0	16.32	-
PET25	25	19.98	22.4
PET50	50	20.42	25.1
PET75	75	23.62	44.7
PET100	100	22.69	39.0

Table 7. Compressive strength for cementitious composites with irradiated plastic aggregates.

Such compressive strength results can be attributed to the effects of factors such as the bond strength between the aggregate and cement paste being enhanced by the physical and chemical changes of the plastic aggregate caused by gamma irradiation, as presented in Sections 3.1 and 3.2. However, when irradiated with a certain dose (100 kGy) or more, the energy supplied by the high dose further deteriorates the surface of the plastic aggregate, and some chain scission occurs, causing the partial destruction of the plastic surface particles, resulting in decomposition; consequently, the strength decreases. Therefore, based on the experimental results collected in this study, it could be suggested that the optimal gamma irradiation dose is 75 kGy, as this was found to be the most effective for increasing strength.

#### 4. Conclusions

This study validates suggestions related to the benefits of using gamma-irradiated plastic aggregates, compared with nonirradiated plastics, to recover some of the lost strength when raw plastic aggregates are added to cementitious composites as a substitute for natural fine aggregates. Based on the detailed review of the presented literature and the test results collected in this study, the following conclusions can be derived:

Analysis of the effect of plastic type on the cementitious composites showed that their compressive strength decreased when plastic aggregate was added, regardless of the type of plastic, and the extent of change depended on the raw plastic density, as reported. In this case, plastic aggregates are inherently smooth and hydrophobic and characterized by poor bond formation in the interfacial transition zone (ITZ) between plastic aggregates and cement paste, dissimilarly to natural aggregates; consequently, the inclusion of plastic aggregates in cementitious composites results in decreased compressive strength.

However, when analyzing the effect of gamma irradiation on the compressive strength of the cement composites, the compressive strength of the cementitious composites increased in proportion to the increase in gamma irradiation dose up to a specific dose (75 kGy). In particular, when the PE aggregate was irradiated with 75 kGy, the compressive strength was increased by 85% compared to the nonirradiated specimen, which was the maximum compressive strength observed in this study. However, it was found that the compressive strength decreased when irradiating with the highest dose (100 kGy). Changes in strength properties of cementitious composites with gamma-irradiated plastic aggregates are justified by SEM and FT-IR analysis.

The SEM analysis results show the roughness of the surface of the irradiated plastic compared to the normal plastic sample. The strength enhancement mechanism through the morphological change of the plastic aggregate according to gamma irradiation observed through SEM is the modification of the essentially smooth surface of raw plastic through gamma irradiation, and as the surface of the plastic aggregate becomes rough, it can be explained that the strength of the cementitious composite is improved by improving the adhesion in the interfacial transition zone (ITZ) between the cement pastes

The mechanism of strength enhancement through the chemical change of plastic aggregates according to gamma irradiation observed through FT-IR can be explained in two ways. First, in the case of crosslinkable plastics such as PE aggregate, the structure of the plastic becomes dense as crosslinking occurs between adjacent molecules, and thus it has superior mechanical properties compared to raw plastic aggregates. Second, as in the case of the PP aggregate, a new functional group is formed due to a chain cleavage reaction to impart adhesiveness and hydrophilicity, thereby improving bonding strength with the cement matrix. In summary, gamma irradiation leads to a more dominant reaction among crosslinking and chain scission depending on the type of plastic; as a result of this reaction, gamma irradiation induces a chemical change in the plastic aggregates. Changes in the chemical structure of plastic aggregates lead to improvement of the interfacial bonding properties between aggregates and cement paste and consequently contribute to the improvement of the strength of cementitious composites.

Based on a comparison with specimens using nonirradiated plastics as an additive, it is clear that irradiation adds the benefit of increased compressive strength. More research needs to be done to find the optimal combination of irradiated plastics and mineral additives for high strength in the use of plastic aggregates.

### Future Work for a Clearer Portrayal of the Mechanism

One important factor in determining the source of the increased strength of cementitious composites containing gamma-irradiated plastic aggregates was the crystallinity of the plastics. The improvement of the compressive strength of the cementitious composites by the addition of irradiated plastic was justified through the analysis of morphological (analyzed by SEM) and chemical changes (analyzed by FT-IR) in this work. However, in addition to this work, in future studies, it is necessary to analyze and present the performance improvement mechanism of cementitious composite materials in more depth using various analysis methods such as analysis of crystallinity (analyzed by XRD) of plastics. This would allow for a clearer portrayal of how the irradiated plastic affects strength by observing the interaction between the irradiated plastic aggregates and cement paste particles and changes in hydration products from a mineralogical point of view. Therefore, further studies on crystallinity analysis are needed before the irradiated plastic aggregates are considered for widespread use in cementitious composite structural elements.

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