

Development and Characterization of Innovative Composite Materials for Construction

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Abstract

The paper aims to develop and characterize innovative composite materials for use in construction, with a focus on sustainability and improving mechanical properties. The study investigates the integration of recycled plastic waste, such as polyethylene terephthalate (PET), low-density polyethylene (LDPE), and polypropylene (PP), into a clay and bentonite based ceramic matrix, with a view to obtaining environmentally friendly and efficient materials for structural applications. The composites were sintered at 1100 °C to ensure structural stability and the formation of ceramic phases, a process that significantly influences the final properties of the materials. The surfaces of the sintered materials were examined by microscopy to investigate the microstructure and the distribution and interaction between the polymer and ceramic phases. In addition, X-ray diffraction (XRD) analyses were performed to identify the crystalline phases formed after sintering and their influence on the properties of the material. Motivated by the need to reduce the environmental impact of conventional building materials and to recycle plastic waste, the research contributes to the development of sustainable solutions, promoting the circular economy and reducing the consumption of natural resources. The results obtained offer valuable insights into the compatibility between recycled polymer components and the ceramic matrix, as well as the potential of these composites for use in modern construction applications.

Keywords

ceramic particles, polymeric composites, polyethylene terephthalate (PET), low-density polyethylene (LDPE), polypropylene (PP)

1. Introduction

Plastics have become essential to the global economy since the 1950s, with their use expanding rapidly due to low production costs and versatility in areas such as food, textiles, construction, and transportation [1, 2]. The production of these materials is largely based on non-renewable resources, particularly crude oil, and growing demand suggests an increase in the consumption of these raw materials in the coming decades. In 2023, global plastic production reached approximately 413–415 million tons per year, marking an increase from the 368 million tons reported for 2019. Data shows that production continues to grow, and the trend is expected to reach approximately 450 million tons by 2025 [3].

However, the global rate of plastic waste management remains insufficient: only about 10% of this waste is recycled, while the rest is incinerated, landfilled, or released into the environment [4]. The inefficient management of this waste highlights the need to develop advanced and sustainable recycling technologies, at least until fully biodegradable and economically viable alternatives can replace conventional plastics [1].

In this context, the integration of recycled plastic into building materials is becoming a promising direction. The incorporation of recycled polymer fibers into ceramic matrices, such as clays, offers the opportunity to transform waste into a useful material, while contributing to reducing environmental impact and improving the properties of composites [5, 6]. Studies analyzing the effects of combining these two types of materials remain limited. Existing research on building materials obtained from mixtures of clay and plastic consistently shows improvements in mechanical properties, as well as a significant reduction in water absorption. For example, Salahuddin et al. [7] manufactured bricks from plastic waste (HDPE and LDPE) mixed with artificial sand and quarry dust at approximately 200 °C. The results showed that these bricks are lightweight, cost-effective, and have low water absorption, highlighting their potential as a sustainable alternative to conventional materials. Koppula et al. [8] and Gounden et al. [9] produced bricks by combining high-density polyethylene (HDPE) with quartz sand

and bitumen, achieving compressive strength values significantly higher than those of traditional bricks. The conclusions of these studies indicate that HDPE waste can be effectively reused to manufacture eco-friendly bricks made from plastic and sand, directly contributing to reducing the amount of plastic disposed of in the environment [8,9]. In a similar study, Ashraf et al. [10] made "Lego" type bricks using recycled polyethylene (PE) and polyethylene terephthalate (PET) waste, achieving optimal compressive strength values depending on the mixing proportions of the plastics. Deraman et al. [11] investigated the use of PET waste as a partial replacement for sand in cement and sand bricks, and the results showed that it improves thermal conductivity (0.581 W/mK) while maintaining mechanical properties within ASTM standards (C129-11 for compressive strength), demonstrating the potential of recycled PET as a sustainable material for the construction industry.

The aim of this work is to develop and characterize sustainable composite materials for construction by integrating recycled plastic waste (PET, LDPE, and PP) into a ceramic matrix based on clay and bentonite, evaluating their structural, microstructural, and phase behavior after sintering at 1100 °C.

2. Materials and Methods

2.1. Materials

The matrix, the basic component that binds all the components together, will consist of bentonite and clay. The reinforcing elements were selected to improve the mechanical and physical properties of the matrix. For this purpose, we used recycled thermoplastic materials, namely: polyethylene terephthalate (PET) – obtained from bottles (Figure 1), polypropylene (PP) – obtained from food containers (Figure 2), and low-density polyethylene (LDPE) – obtained from food bags (Figure 3).



Fig. 1. Short fibres of PP



Fig. 2. Short fibres of PET



Fig. 3. Short fibres of LDPE

The selected materials, including PET, PP, and LDPE, possess essential properties such as mechanical strength, flexibility, and durability, while also being highly recyclable. This selection was motivated by the dual objective of integrating recycled materials and optimizing the mechanical properties of the composite. If favourable results are obtained, the use of these plastics could facilitate the replacement of traditional construction materials such as iron or fiberglass.

Bentonite and clay are natural materials with a fine structure, widely used in industrial and construction applications due to their functional properties. Bentonite is a very fine clay, formed of lamellae, characterized by its high-water absorption capacity, high viscosity when mixed with water, and ion exchange ability, which facilitates the formation of bonds in composites. Due to these properties, it is used as a sealing agent, binder, or absorbent [5].

Overall, these materials are essential in the production of ceramics and bricks, civil and engineering construction, agriculture, the cosmetics and oil industries, and are valued for their versatility and physical-chemical properties.

Polyethylene terephthalate (PET) is a thermoplastic polyester with a long polymer chain, which gives it high mechanical strength. It is transparent, lightweight, dimensionally stable, and has good resistance to breakage, impact, and chemical environments, making it widely used in the food industry. PET is easy to recycle, and in ground form it can be used as a reinforcing material, helping to reduce resource consumption and CO₂ emissions.

Polypropylene (PP) is an important polymer due to its superior heat resistance to polyethylene (melting point ~175°C) and good chemical resistance. Although it can be affected by solvents and UV

radiation, its behaviour can be improved with additives. It is tough, lightweight, and versatile, being processed by injection, extrusion, or into film and fibres. PP is frequently used in composite materials, both as a matrix and as a filler.

Low-density polyethylene (LDPE) is a very versatile material used in packaging (bags, films), the food, cosmetics, electrical, toy, and household container industries. It is notable for its flexibility, moisture resistance, and food compatibility.

2.2. Methods

First, a base material consisting of 70% bentonite and 30% clay was prepared. A small amount of water was added to this mixture and mixed thoroughly until homogeneous. Using a mold, samples with a diameter of 30 mm and a thickness of 15 mm were made from the material obtained. The samples obtained were left to dry at room temperature for 3 days. During drying, cracks and deformations appeared on some samples.

To produce fibre-reinforced composites from recyclable plastics, 4% (PE, PET, or LPDE) was added to the base material. The composites thus obtained are shown in Figures 4, 5, and 6.



Fig. 4. Bentonite and clay composite reinforced with PP fibres



Fig. 5. Bentonite and clay composite reinforced with PET fibres



Fig. 6. Bentonite and clay composite reinforced with LDPE fibres

The samples were sintered in a Nabertherm furnace at 10 °C per minute up to 1100 °C for 2 hours. Once the furnace reached the selected temperature, it was held for 30 minutes. After holding, the composite materials were cooled in the furnace. During sintering, the cracks enlarged, and the samples deteriorated significantly.

3. Results and discussion

The sintered samples clearly show the mineralogical phases in the composition of clays and bentonites, namely: SiO_2 , SiC , Fe_2O_3 , and Al_2O_3 (Figures 7, 8, 9, and 10) and slight traces of PP (8) and LDPE (Figure 10) fibres. The observation of visible traces of LDPE and the very small amount of PET on the surface of the samples under the digital microscope can be attributed to differences in the thermal stability and degradation rate of the polymers; LDPE decomposes rapidly at relatively low temperatures, leaving visible residues, while PET, which is more thermally stable, degrades more uniformly, generating minimal traces in the ceramic matrix.

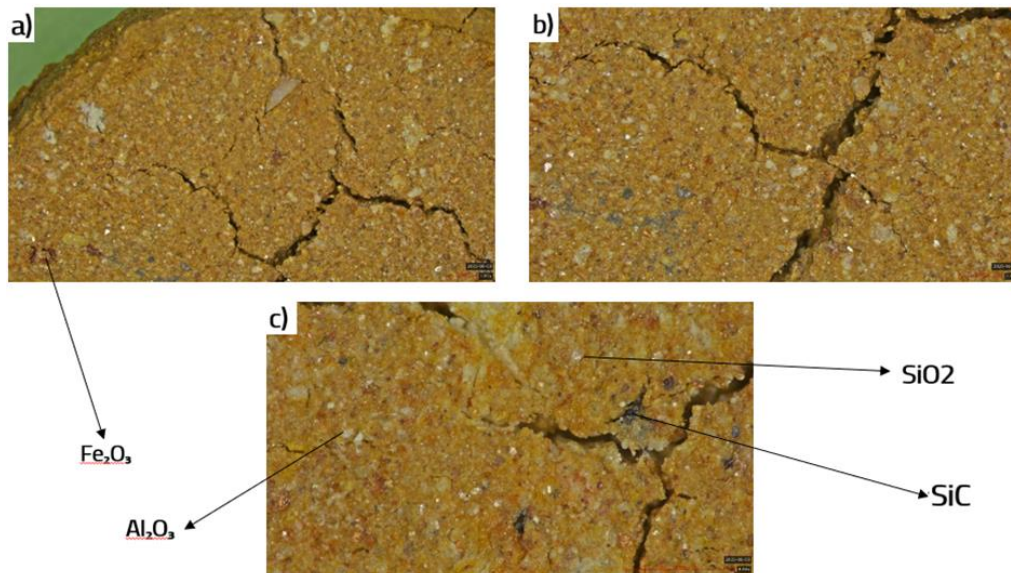


Fig. 7. Surface appearance of the base material at different magnifications: a) $\times 1$; b) $\times 1.6$; c) $\times 4$

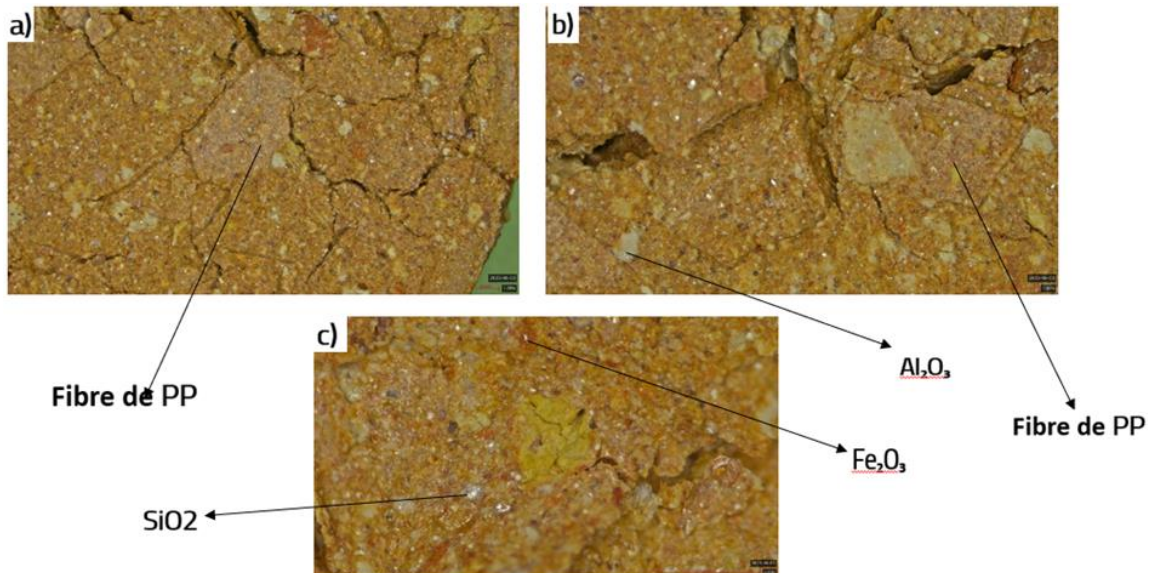


Fig. 8. Surface appearance of PP fibre reinforced composite at different magnifications: a) $\times 1$; b) $\times 1.6$; c) $\times 4$

During sintering, the cracks resulting from the compaction of the materials increased, and for this reason, it was not possible to perform mechanical tests on the samples obtained. The specimens obtained were subjected to the following types of characterization: surface study by digital optical microscopy and X-ray diffraction analysis. Surface analysis by optical microscopy was performed using the LEICA EMSPIRA 3 digital microscope.

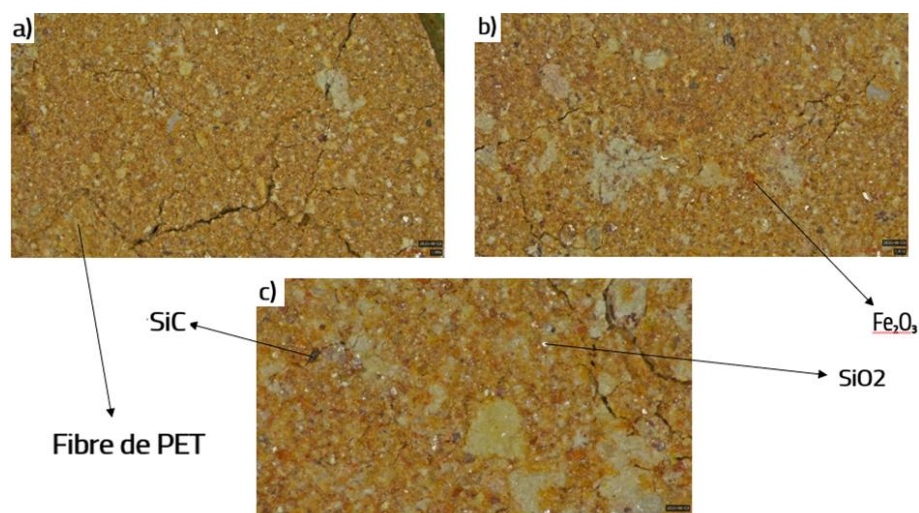


Fig. 9. Surface appearance of PET fibre reinforced composite at different magnifications: a) $\times 1$; b) $\times 1.6$; c) $\times 4$

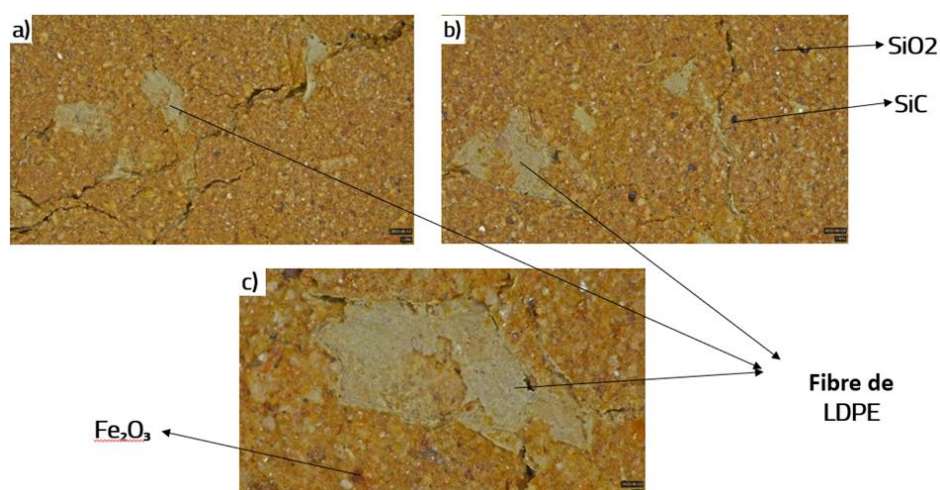


Fig. 10. Surface appearance of LDPE fiber reinforced composite at different magnifications: a) $\times 1$; b) $\times 1.6$; c) $\times 4$

The appearance of the sample surfaces after sintering is shown in the figures below. Both in the case of the base material (Figure 9) and in the case of composites reinforced with plastics (Figures 10, 11, and 12), a monolithic base mass is obtained after sintering.

X-ray diffraction (XRD) analysis was conducted to identify the crystalline phases and determine the structural composition of the specimens. The phase composition was investigated by X-ray diffraction measurements at room temperature. For this purpose, a Bruker D8 Advance diffractometer using Cu-K radiation ($\lambda=1.5418 \text{ \AA}$) was used. The X-ray tube was operated at 40 kV and 40 mA. Motorized slits with an opening of 0.25 mm and a 2.5° Sollers slit were used on the incident beam side, and motorized slits with an opening of 5 mm and an open slit were mounted on the diffracted beam side, mounted on the LYNXEYE XE -T detector operating in 1D mode and high-resolution option. The X-ray diffractograms were recorded in the range $10-50^\circ 2\theta$, with a step size of 0.02° and a counting time of 0.2 s/step.

The X-ray diffractogram analysis was performed in the HighScorePlus 3.0.e software connected to the Crystallography Open Database – COD, version 2024. The quantitative evaluation of the phase composition was performed by refining the experimentally obtained diffractogram using the Pawley method, followed by the relative intensity ratio method.

The samples studied have a phase composition consisting of quartz (COD # 96-900-9667), mullite (COD # 96-900-1332) and cristobalite (COD #96-900-1580) in different proportions highlighted in the

diagrams inserted in the following diffractograms (Figures 11, 12, 13 and 14). It can be observed that the proportions of the mineral phases vary depending on the type of polymer fibres incorporated. In the case of the PP fibre-reinforced composite (Figure 12), the introduction of the polymer material leads to a slight increase in the quartz content and a reduction in the mullite and cristobalite phases. Unlike PP, PET fibres (Figure 13) cause a decrease in the quartz phase, along with an increase in mullite and cristobalite. The addition of LDPE fibres generates significant changes in the composition of the mineralogical phases of the composite (Figure 14), suggesting a different interaction between this type of polymer and the ceramic matrix during sintering.

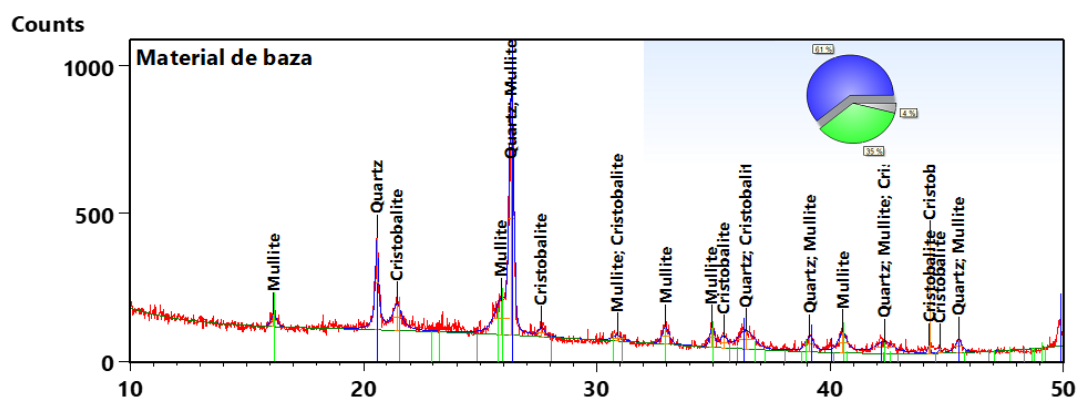


Fig. 11. X-ray diffractogram obtained for the base material (quartz – blue, mullite – green, cristobalite – grey)

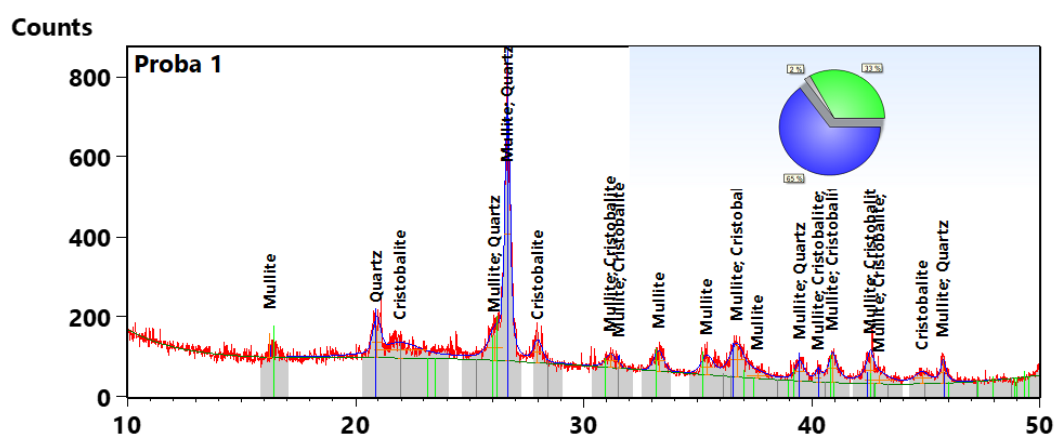


Fig. 12. X-ray diffractogram obtained for the PP fibre-reinforced composite (quartz – blue, mullite – green, cristobalite – grey)

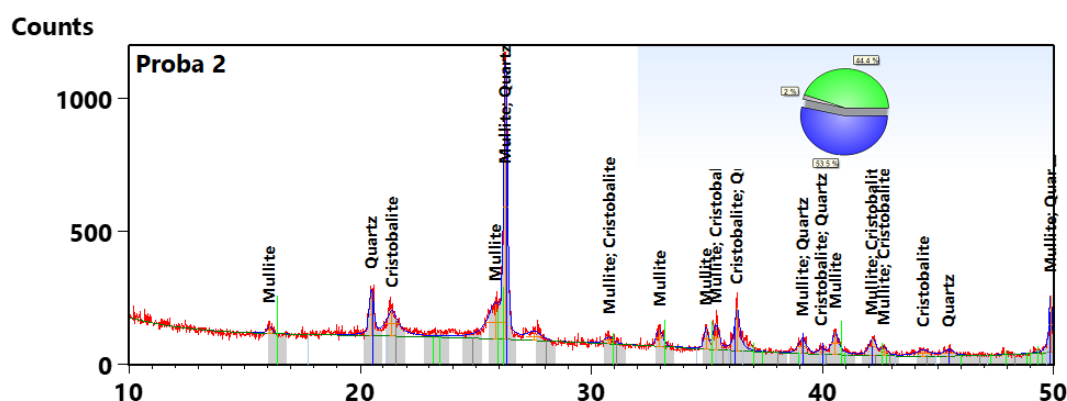


Fig. 13. X-ray diffractogram obtained for the PET fibre-reinforced composite (quartz – blue, mullite – green, cristobalite – grey)

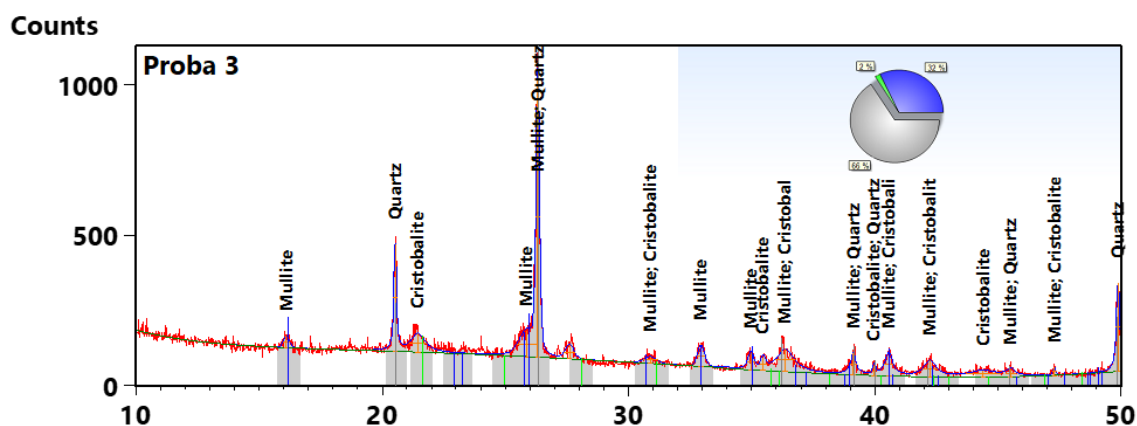


Fig. 14. X-ray diffractogram obtained for the LDPE fibre-reinforced composite (quartz – blue, mullite – green, cristobalite – grey)

The formation of mineralogical phases was significantly influenced by the type of polymer fibres introduced into the matrix. In the case of the PP composite, the rapid degradation of the polymer generated high porosity and low densification, which limited the allotropic transformations of SiO_2 , leading to an increase in the quartz phase and a reduction in mullite and cristobalite. Unlike PP, PET fibres, which have higher thermal stability, favoured sintering reactions, resulting in quartz consumption and increased formation of mullite and cristobalite. In the case of LDPE, the early and intense decomposition of the polymer generated a very uneven microstructure, causing pronounced changes in the composition of the mineral phases. These results show that the nature of the polymer directly influences the evolution of the ceramic phases during sintering.

4. Conclusion

The study explores the use of recycled plastic waste: PET, LDPE, and PP integrated into a clay- and bentonite-based ceramic matrix to obtain environmentally friendly and high-performance composite materials suitable for structural applications.

Digital microscopy analysis highlighted the main mineralogical phases in the matrix—quartz, mullite, and cristobalite—and residual traces of polymer fibres. The differences in the amount of residue observed (more visible for LDPE and almost imperceptible for PET) reflect variations in the thermal stability and degradation mode of each polymer.

XRD analysis showed that the type of polymer fibre significantly influences the composition of the mineralogical phases:

- PP fibres cause an increase in quartz and a decrease in mullite and cristobalite, due to rapid degradation and increased porosity.
- PET fibres, which are more thermally stable, favour the formation of mullite and cristobalite and reduce the quartz content.
- LDPE fibres generate pronounced and uneven changes in phase composition due to early and intense polymer degradation.

The results demonstrate that the nature of the recycled polymer directly influences the evolution of the microstructure and ceramic phases during sintering, highlighting the importance of fibre type selection for optimizing the final properties of composites.

The study confirms the potential of using recycled plastic waste to develop sustainable composite materials that can contribute to reducing the environmental impact of construction and the efficient use of plastic resources.

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