POSSIBILITIES FOR CONSERVING NATURAL RESOURCES AND THE ENVIRONMENT THROUGH THE USE OF RECYCLED WASTE AGGREGATES AS A SUBSTITUTE FOR NATURAL AGGREGATES IN CEMENTITIOUS COMPOSITES

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Abstract

The conservation of the environment and the protection of natural resources are urgent and current challenges. The objective of this experimental investigation was to evaluate the potential use of aggregates derived from recycled glass waste, blast furnace slag, recycled brick waste aggregates and recycled electronic waste aggregates (textolite) as replacements for natural aggregates in cement-based composites. The experimental tests aimed to investigate how the replacement of natural aggregates with recycled waste aggregates affects various physico-mechanical parameters, including density, compressive strength, flexural strength, abrasion resistance and capillary water absorption. This investigation also included detailed microstructural analysis using optical microscopy, SEM, EDX and XRD techniques. The aim of the research was to explore the potential for soil conservation by reducing the amount of waste to be disposed of, and at the same time to conserve natural resources by identifying alternatives using recycled materials, thereby contributing to the implementation of the circular economy concept. The results of the research confirmed this potential; however, depending on the nature of the recycled aggregates, there are influences on the physico-mechanical performance of the cement composite that can be seen at the microstructural level.

Keywords: Waste recycling; Cementitious composites; Environmental conservation; Conservation of natural resources

Introduction

There is currently a critical global need to find ways of conserving the environment by reducing the considerable amount of waste generated by various activities. At the same time, it is equally important to explore ways of conserving natural resources. In this context, the construction sector has a significant impact on both the production of waste and the consumption of natural resources. In the EU-28 in 2014, construction contributed 34.7% of the
total, followed by the extractive industry (28.2%), manufacturing (10.2%), water and waste (9.1%) and households (8.3%). The remaining 9.5% was generated by other economic activities, mainly services (3.9%) and energy (3.7%) [1]. In 2020, Romania reported 283,831.313 tonnes of glass waste, 304,856.548 tonnes of plastic waste, 93,786.442 tonnes of discarded electrical and electronic equipment, 19,830.607 tonnes of non-construction mineral waste and 1,167,699.165 tonnes of construction and demolition waste [2]. Statistical reports indicate that by 2020, the majority of waste in Romania comes from the extractive and construction sectors. Therefore, in order to protect the environment and natural resources, actions in these sectors are most likely to yield significant results. Regarding the expenditure and investments recorded in 2021-2022 for the preservation and protection of the environment and natural resources in Romania, 21.3% of the total expenditure and investments were related to the extractive industry and 3.6% to the construction industry. In addition, 88.8% of the total expenditure was allocated to activities related to the collection, treatment and disposal of waste and the recovery of recyclable materials [2–5].

A current pressing global concern is the need for sustainable constructions with minimal environmental impact and increased durability. The need for sustainable construction with minimal environmental impact and increased durability is a pressing global concern. Ongoing research in this area focuses on identifying opportunities to develop innovative construction materials with improved performance. One such development direction is the creation of cementitious composites that enable the recycling of waste and industrial by-products while maintaining or improving performance characteristics. Information in scientific literature provides strong evidence for the desirability of recycling and using waste as an aggregate. This approach not only serves to reduce the use of primary resources, but also has the advantage of reducing the volume of waste and promoting the implementation of circular economy principles [6–10]. Nonetheless, in many cases, the use of recycled waste aggregates is restricted due to their negative impact on the performance of cementitious composites, requiring an analysis of acceptable limits [11]. Many studies in the literature have highlighted the extensive range of recycled aggregates that are compatible with the cementitious binder matrix, showcasing variations in properties such as density, water absorption, mechanical strength, freeze-thaw resistance, and resistance to chemical agents. This diversity contributes to a significant variability in the properties of the composite material [11–37].

Some studies suggest that the water absorption of recycled brick waste aggregates can be up to 25 times higher than that of gravel or 10 times higher than that of sand. This has a direct effect on the water available for cement particle hydration and the formation of its hydration-hydrolysis compounds, which affects the workability of the composite in its fresh state and, consequently, the physical-mechanical performance indicators of the solidified composite [14, 23, 30, 34, 38, 39]. For this reason, the scientific literature recommends either determining the additional water required to maintain the workability of the composite or preparing recycled aggregates from brick waste by soaking them in water for 24 hours prior to use. Another suggestion is to determine the additional water required to maintain workability consistency [23, 30, 35, 40]. According to the literature, the compressive strength of cementitious composites where natural aggregates are replaced by recycled aggregates from brick waste can be reduced by up to 35%. This reduction becomes more significant as the amount of recycled aggregates used increases [30]. At the same time, other bibliographic references indicate a marginal decline in this parameter when keeping the recycled aggregate within a maximum limit of 15%. The literature on the use of recycled glass waste aggregates suggests that their size and concentration in the cement matrix are the key factors affecting the performance of the composite [41–53].

Research has suggested that recycled glass aggregates with particle sizes of less than 150 µm can react with portlandite (\( \text{CH} \cdot \text{Ca(OH)}_2 \)) during the cement hydration process to form calcium silicate hydrate (C-S-H), potentially resulting in increased mechanical strength [7, 41,
On the other hand, incorporating coarser glass aggregates into the cement mixture may lead to a significant alkali-silica reaction (ASR), which can reduce the strength and durability of the concrete [45, 46]. Furthermore, studies indicate that the incorporation of waste glass as an aggregate generally results in reduced water absorption in the composite, improved abrasion resistance and the replacement of up to 20% of natural aggregates with recycled glass aggregates can potentially increase compressive strength, as the glass granules meet the requirement for high fineness. With regard to the effects of the use of blast furnace slag, research studies in the literature indicate that this industrial by-product has become a commonly used material for partial replacement of Portland cement. This substitution results in a reduction in early strength but improves late strength and increases the durability of the concrete [43, 52, 55]. As an alternative to natural aggregates in cement-based composites, some studies have shown limited effects on composite performance, while others have documented mechanical advantages, including a potential increase in compressive strength of up to 60% under certain conditions. There is also the prospect of reduced density and improved durability [56–62].

Other studies have shown that in some cases the replacement of up to 30% of the natural aggregates with these wastes does not result in a significant change in compressive strength. In other cases, even total replacement of aggregates, although initially (at 2 and 7 days) resulting in a slight reduction in compressive strength, subsequent testing at 28 days or more after casting has shown improved compressive strength [63, 64]. However, alternative studies suggest that as the proportion of slag replacing natural aggregates increases, the apparent density of the composite decreases and open porosity increases, as evidenced by increased water absorption. This is undesirable as it increases the susceptibility to chemical agents penetrating the composite. For example, replacing 90% of the sand with slag can increase capillary water absorption by more than 30% [57].

In general, slag reactivity and its effect on composite performance increases with higher levels of CaO, Na₂O and K₂O and decreases with lower levels of SiO₂, FeO/Fe₂O₃, TiO₂ and MnO/Mn₂O₃. A favourable result is associated with a high CaO/SiO₂ ratio, and an increased Al₂O₃ content contributes positively to hydration heat generation and promotes higher early strength [65–70]. This behaviour has been attributed to the ability of Al-containing phases (e.g. ettringite) to better fill the voids due to their high molar mass, thus increasing the compressive strength of the composite. X-ray diffraction (XRD) analyses in conjunction with scanning electron microscopy (SEM) reveal that the predominant product of the Portland cement hydration process is calcium silicate hydrate (C-S-H). In addition, portlandite (CH), the second most abundant product in the hydrated paste of Portland cement, is observed to be present in the form of relatively large crystalline structures with a plate-like morphology [71]. The presence of AFt (ettringite) and AFm (monosulphate hydrate) phases depends on the amount of tricalcium aluminolate (C₃A) and ferrite in the original cement. Ettringite (AFT) occurs as prismatic crystals resembling needles, which later transform into monosulphate hydrate (AFm) and adopt a hexagonal plate-like crystal structure. In addition, X-ray analysis reveals the presence of unhydrated cement clinker (C3S and C2S). Further investigations suggest that similar crystalline products are expected in the slag composite, including the identification of hydrated calcium alumina silicate (CSAH). However, the intensity of the peak associated with portlandite (CH) decreases with increasing Al₂O₃ content [65, 72, 73].

EU regulations have been in place since February 2003 to encourage the gathering and recycling of waste electrical and electronic equipment (WEEE) [74]. Nevertheless, the extent of waste collection, recycling, and recovery remains inadequate. Research suggests that printed circuit board waste, when processed into non-metallic products and incorporated into various binder matrices, can, in certain scenarios, provide superior mechanical properties and durability compared to conventional materials and fillers [75–80]. This waste comprises an electric-insulating material, namely a non-metallic powder consisting of layers of ground fabric impregnated with synthetic resins, including glass fibres, commonly known as textolite powder.
However, there is still a lack of research into the use of these wastes in cementitious matrices. Premur et al. [80] observed a significant decrease in the compressive strength of cementitious composites when 5-20% of non-metallic waste from WEEE recycling was replaced with sand, limiting the feasible substitution of natural aggregates with fine particles (sand) to 10%. This limitation was also suggested by other reports in the literature [75, 87–92]. On the other hand, Vishnu Priyan et al. have demonstrated that 5% (mass percentage relative to the amount of cement) of WEEE waste, when used as dispersed reinforcing fibres, contributes to the enhancement of the mechanical strength and durability performance of the cementitious composite [93, 94]. Despite ongoing debates, all studies in this field point to two general conclusions: firstly, a waste composition analysis of non-metallic waste from WEEE recycling is imperative for each specific case, taking into account residues that may affect the health of users; secondly, preliminary research is required for each case of cementitious composite and for each type of waste generally referred to as "textolite waste" in order to understand its influence on composite performance.

On the basis of what has been reported, it can be said that, given the primary objective of protecting the environment and natural resources, and implicitly of protecting the soil by reducing the areas required for waste disposal, a possible direction of development is their recycling and reuse as a substitute for natural aggregates in cement composites intended for the development and/or conservation of the built heritage. The scientific literature supports the possibility of substituting natural aggregates with aggregates derived from recycled waste, indicating the possibility of obtaining satisfactory results for cement composites, with the mention that preliminary studies are necessary on a case-by-case basis due to the significant heterogeneity of the characteristics of recycled aggregates. In addition, the need for a continuous contribution to the current state of knowledge in this field through the development of research programmes in this area of interest is emphasised.

Therefore, the aim of this study was to analyse the possibility of protecting the environment by reusing waste and conserving mineral resources, in particular natural aggregates, by studying the impact of their partial substitution with aggregates derived from four types of locally recycled waste: recycled glass aggregate (RGA), recycled ceramic brick aggregate (RBA), granulated blast furnace slag (GBA) and recycled textolite aggregate (RTA), on the performance of cement composites intended for construction.

Experimental part

For the purpose of the research, various types of cementitious composites were designed, prepared and analysed. Firstly, a reference cementitious composite (R1) was prepared, consisting of cement, natural aggregates, superplasticiser and water. Subsequently, in order to analyse the influence of the partial substitution of natural aggregates with aggregates derived from recycled waste on the physico-mechanical performance, a series of compositions (R2-R5) were prepared, introducing aggregates derived from recycled waste such as glass, bricks, blast furnace slag or recycled textolite waste with WEEE as the primary source.

Materials and Methods

Raw materials

The following raw materials were selected for the preparation of the cement composites Portland cement CEM I 52.5 R, natural aggregates (NA) with particle sizes of 0/4 mm and 4/8 mm, recycled glass aggregates (RGA) with particle sizes of 0/4 mm and 4/8 mm, recycled ceramic brick aggregates (RBA) with particle size of 0/4 mm, granulated blast furnace slag (GBA) with particle size of 0/2 mm or recycled textolite waste aggregates (RTA) with particle size of 0/2 mm, together with MasterEase 5009 superplasticiser and water. All recycled waste aggregates were sourced locally. The characteristics of these raw materials are as follows:
- commercially available CEM I 52.5 R Portland cement with a minimum Portland clinker content of 95% and a compressive strength of between 52.5 N/mm² and 62.5 N/mm² at 28 days;
- the commercially available superplasticizer / high water reducer MasterEase 5009;
- natural aggregates as well as aggregates derived from recycled characterised through the particle size distribution curve, figure 1, according to EN 14582, the bulk density and intergranular porosity according to EN ISO 15586, the true mass and the water absorption coefficient according to EPA 7000B:2007, as shown in figure 2.

Fig. 1. Graphical representation of the particle size distribution of the used aggregates

Fig. 2. Graphical representations of the a) Bulk density (pp) and actual density (pb) of aggregates; b) Bulk intergranular porosity (v) of aggregates; c) Water absorption capacity (WA24) of aggregates

In addition, the oxide composition of the furnace slag (GBA) was determined by X-ray fluorescence analysis (XRF), table 1. The textolite waste was analysed for residual metal content, table 2, according to ISO 11047.
Table 1. Characterisation of blast furnace slag (GBA)

<table>
<thead>
<tr>
<th></th>
<th>SiO₂</th>
<th>Al₂O₃</th>
<th>Fe₂O₃</th>
<th>CaO</th>
<th>MgO</th>
<th>SO₃</th>
<th>Na₂O</th>
<th>K₂O</th>
<th>P₂O₅</th>
<th>TiO₂</th>
<th>Cr₂O₃</th>
<th>MnO₂</th>
<th>P.C.</th>
</tr>
</thead>
<tbody>
<tr>
<td>%</td>
<td>30.20</td>
<td>10.05</td>
<td>14.70</td>
<td>4.05</td>
<td>0.20</td>
<td>0.38</td>
<td>&lt;0.52</td>
<td>&lt;0.05</td>
<td>2.15</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 2. Characterisation of waste textolite (RTA)

<table>
<thead>
<tr>
<th></th>
<th>As</th>
<th>Ba</th>
<th>Cd</th>
<th>Cr</th>
<th>Cu</th>
<th>Hg</th>
<th>Mo</th>
<th>Ni</th>
<th>Pb</th>
<th>Sb</th>
<th>Se</th>
<th>Zn</th>
</tr>
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<tbody>
<tr>
<td>Mg/kg</td>
<td>2.85</td>
<td>603.8</td>
<td>&lt;0.03</td>
<td>18.2</td>
<td>1064</td>
<td>&lt;0.003</td>
<td>&lt;3.0</td>
<td>&lt;0.20</td>
<td>&lt;0.30</td>
<td>&lt;0.5</td>
<td>&lt;0.015</td>
<td>12.59</td>
</tr>
</tbody>
</table>

Note: "<" represents values below the limit of determination of the method.

Manufacturing of cementitious composites

The recipes were designed based on the Romanian Standard NE 012-1 "Standard for the production and execution of works in concrete, reinforced concrete and prestressed concrete - Part 1: Production of concrete", for a concrete class C20/25, characterised by a water-cement ratio of 0.6, as shown in Table 3.

Table 3. Control recipe (R1)

<table>
<thead>
<tr>
<th>Recipe code</th>
<th>Design class</th>
<th>Water/cement ratio</th>
<th>Cement (kg/m³)</th>
<th>Natural aggregates, cumulative (kg/m³)</th>
<th>Natural aggregates sort 0/4 mm (% of total aggregates)</th>
<th>Natural aggregates sort 4/8 mm (% of total aggregates)</th>
<th>MasterEase 5009 superplasticizer additive (% mass ratio to cement quantity)</th>
</tr>
</thead>
<tbody>
<tr>
<td>R1</td>
<td>C 20/25</td>
<td>0.6</td>
<td>366</td>
<td>1577</td>
<td>70</td>
<td>30</td>
<td>0.5</td>
</tr>
</tbody>
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Subsequently, recipes were developed in which natural aggregates were replaced in varying proportions by recycled waste aggregates, as described in figure 3. The criteria for determining aggregate proportions were guided by the requirement that each cumulative distribution curve of the aggregates be within the favourable, or at least usable, range specified by the design standard, as illustrated in figure 4.

Fig. 3. Graphical representation of the distribution of types of aggregates used in cementitious composites recipes in which natural aggregates have been replaced by recycled waste aggregates, compared to the control recipe R1

*Note: Although the mixing water was replenished during preparation when ceramic brick aggregates, textolite aggregates and slag aggregates were used, the recalculated water/cement ratio remained within the limits of 0.60-0.65 while maintaining a constant workability indicator.
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Fig. 4. Graphical representation of the overall aggregate mix design according to the guidelines of the NE 012-1 standard - exemplified for a) the reference mix R1, b) the blend with natural aggregates substituted by recycled glass waste aggregates R2-1.

The cement composites were prepared by mixing dry raw materials with water and adding the superplasticiser MasterEase 5009 using an ELE type paddle mixer. For all prepared compositions, the quantity of cement was kept constant, and the amount of superplasticizer used was 0.5%, by weight, relative to the amount of cement. The raw materials were dosed by precise weighing using a KERN FKB 36K0.1 scale with an accuracy of 0.1g. Due to the specific water absorption characteristics of the different types of recycled waste aggregates, which can affect the overall water/cement ratio of the composite and, consequently, its workability and subsequent physico-mechanical properties. In order to reduce their influence on the water available for the hydration-hydrolysis reactions of the cement particles, the water content of the mixture was determined for each case, taking into account the effective water/cement ratio. The consistency of the mix (EN 1015-3) was used as a reference indicator, with the condition that it should be kept constant and similar to the control, i.e. the flow diameter should be within the range of 175±10mm (EN 1015-2). In this context, the partial replacement of natural aggregates with recycled brick aggregates resulted in a shift of the water/cement ratio to 0.64, as seen in the mix with the maximum substitution of natural aggregates with recycled brick aggregates (R3-3). In the case of replacing NA with GBA, the water/cement ratio reached a maximum value of 0.625 (R4-3), while in the case of replacing NA with RTA, the water/cement ratio peaked at 0.65 (R5-4). In the case of RGA use, the water/cement ratio remained unchanged, corresponding to that of the control mix with an w/c ratio of 0.6.

Analysis of the physical-mechanical performances of cementitious composites

The samples were tested 28 days after casting. For each type of cementitious composite, the following performance indicators were determined and analysed: density in the hardened state, according to the standard method specified in EN 1015-10, compressive and flexural strength, according to the standard method specified in EN 1015-11, using an ADR Auto 250/25 compression and flexure testing machine with an accuracy of 0.01 kN, abrasion resistance, according to the standard method specified in EN 1338 and capillary water absorption, according to the standard method specified in EN 1015-18. The samples were weighed using a KERN FKB 36K0.1 balance with an accuracy of 0.1g and their dimensions measured using an electronic calliper gauge with an accuracy of 0.01mm.

The testing was carried out under laboratory conditions at 23°C and 60% relative humidity, with the results for each monitored parameter and composite expressed as the arithmetic mean of the individual values.
Macro and microstructural analysis of cementitious composites

The macrostructural study of cementitious composites involved an initial visual assessment without optical magnification equipment, focusing on ensuring the uniform distribution of aggregates within the cementitious binder matrix. A microscopic analysis was then carried out using a LEICA SAPO optical microscope to observe the distribution at the macroporosity level.

SEM images were obtained using a JEOL/JSM 5600 - LV scanning electron microscope (JEOL Ltd, Tokyo, Japan) operating in SEI mode at an acceleration voltage of 15kV. In order to enhance electrical conductivity for electron microscopy analysis, a gold coating was applied to the samples through plasma sputtering as an integral step in the preparation procedure.

Results and discussion

Macrostructural analysis of cementitious composites

Illustrative photographic images of cementitious composites in a fractured state are shown in figure 5. A homogeneous distribution of natural and recycled waste aggregates in the cementitious binder matrix is observed, with no segregation zones, but with a clear influence of the type of recycled aggregate on the texture of the composite.

![Illustrative photographic images of cementitious composites in a fractured state](image)

Fig. 5. Optical microscopy images depicting the changes and macroporosity of the composites as a result of the replacement of natural aggregates with recycled waste aggregates

The optical microscopy images shown in figure 6 illustrate the changes in the macroporosity of the composites as a result of the replacement of natural aggregates with recycled waste aggregates. In the reference composite, pores within the range of 0.40 to 1.01mm were identified in the binder matrix, with the binder seamlessly integrating the aggregate granules. The replacement of natural aggregates (NA) with recycled glass aggregates (RGA) resulted in an increase in the number of pores, accompanied by a reduction in their dimensions (typically between 0.27 and 0.50mm, rarely exceeding 0.8mm). In addition, both AN and RGA granules were effectively embedded in the binder matrix. When recycled brick aggregates (RBA) were used as a replacement for NA, their successful integration into the binder matrix was evident, with an evenly distributed array of pores. These pores were more abundant compared to the control, although presenting smaller dimensions (0.109 to 0.335mm). The use of GBA as a replacement aggregate yielded similar results, with a uniform distribution of pores in the binder matrix, which were more widespread and ranged in size from 0.107 to 0.500mm. However, there was a notable difference when using RTA: a lack of contact and a relatively uneven distribution of RTA in the binder matrix could be observed at the interface between RTA and the cement binder. Porosity was also unevenly distributed, with pore sizes varying significantly and diameters ranging up to almost 1.0mm.

![Illustrative optical microscopy images of cementitious composites with different replacement aggregates](image)
Analysis of the physical-mechanical performance of cementitious composites

The experimental results, as shown in figure 7, highlight some remarkable findings regarding the apparent density of the solid-state composites 28 days after casting. In particular, the replacement of natural aggregates (NA) with recycled glass aggregates (RGA) leads to a slight reduction in the apparent density of the composite. This reduction becomes more pronounced as the proportion of substituted aggregates increases. However, when compared to the control recipe, the reduction in apparent density is not significant and falls within the range of 1.6% - 4.8% of the apparent density of the control recipe R1. It is important to note that this tendency is in line with some findings reported in the literature [20, 21, 23, 26, 30].

In contrast, replacing natural aggregates (NA) with recycled ceramic brick aggregates (RBA) resulted in a greater reduction in the apparent density of the composites, ranging from 13.3% to 20.9% compared to the R1 reference. This reduction is more pronounced with increasing amounts of substituted natural aggregates (NA). In the case of the use of blast-furnace slag aggregate (BFS), no clear trend affecting the bulk density of the cement composite could be identified. The values recorded were close to those of the control sample, possibly with a slight increase in the case of sample R4-1 (substitution of 20% NA by GBA), an observation in line with some reports in the literature [56, 57, 60]. Moreover, the replacement of natural aggregates (NA) with recycled textolite (RTA) resulted in a significant reduction in the indicator analysed, particularly as the amount of recycled textolite increased. This reduction
exceeded 45% compared to the control (R1), particularly in the case of the sample with 40 NA replaced by RTA, which is in line with reports in the literature [75, 80, 87–92].

The compressive strength shown in figure 8 is influenced in a complex way by both the type and quantity of aggregates that replace the natural aggregates. When recycled glass aggregates (RGA) are used, as discussed above, the literature suggests two possible trends in compressive strength evolution. Either an increase could occur if the fineness of the RGA granules facilitates a pozzolanic character, or a decrease could be observed if, due to the particle size distribution (especially large granule sizes), a strong alkali-silica reaction (ASR) is initiated. In the current study, the experimental results were consistent with the latter scenario, indicating a decreasing compressive strength, especially with increasing cumulative amount of RGA.

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As additional confirmation, the minimum reduction in compressive strength (3.8%) was recorded for the composition with the lowest RGA content, regardless of the particle size, i.e. Composition R2-1 (30% RGA fraction 0/4 mm + 15% RGA fraction 4/8 mm). On the other hand, composition R2-2 (40% RGA fraction 0/4 mm + 10% RGA fraction 4/8 mm) suffered a loss of 6%, and even more in comparison with composition R2-3, which lacks natural aggregates (NA), resulting in a loss of compressive strength of more than 27%. This behaviour is considered as a sign of insufficient fine glass particles to allow the manifestation of a pozzolanic character and is consistent with reports in the specialised literature [41–52].

The partial replacement of natural aggregates (NA) with recycled brick aggregates (RBA) resulted in a reduction in compressive strength that increased with the amount of NA replaced, as reported in similar studies in the literature [21, 22]. However, for a replacement of 40% NA by RBA (R3-1 composition), this reduction is acceptable, being only 2.4% compared to the control sample. On the other hand, for other compositions (R3-2 and R3-3), the loss of compressive strength became significant, reaching 15.2% and 25.9% respectively. These situations can be considered challenging from a performance point of view.

The replacement of NA with GBA resulted in an increase in compressive strength compared to the control sample, as expected and in accordance with the literature [52–57]. While this increase was modest for the lower percentages of NA replaced by 20-30% GBA (2% or 6%), when the substitution reached 40% of the initial amount of natural aggregate (NA), the improvement in compressive strength was significant (16.5% compared to the control). When RTA was used as a replacement for NA, there was a significant reduction in compressive strength, reaching 36.5% for 10% RTA (R5-1 recipe) and up to 63.7% for R5-4 (40% RTA), which is in line with references in the scientific literature [75, 80, 87–92].

With regard to the flexural tensile strength, as shown in figure 9, the effect of replacing NA with recycled aggregates shows a somewhat similar pattern to that observed for the compressive strength. Replacing natural aggregates with RGA or RBA resulted in a decrease in this parameter, which became more pronounced as the percentage of substitution increased. Conversely, replacement with GBA led to an increase in this indicator of up to 18% compared to the tensile strength of the control (R1). In particular, the benefit of using GBA was even more pronounced than in the case of compressive strength, with a maximum increase in tensile
strength of over 12% for all samples prepared with GBA. This underlines the microstructural and mineralogical changes inferred from other studies cited in the literature [56–64].

Fig. 9. Graphical representation of the flexural tensile strength of cured composites 28 days after casting.

In the case of the flexural tensile strength analysis, the use of RTA resulted in the most significant losses in comparison with the control. The reduction recorded reaches a maximum of 34.6% for the sample in which 40% of the natural aggregates are replaced by RTA (R5-4). This observation is in line with what is reported in the literature [75, 80, 87–92].

The observed progression of the quantified wear resistance, shown in figure 10, indicates an improved resistance to abrasion, with mass loss ranging from 23.5% to 40.3% lower in composites containing RGA compared to the control sample. This increase in wear resistance, quantified by reduced mass loss, was greater when the amount of substituted natural aggregates (NA) was lower. The analysis of the behaviour of composites with RGA showed an inconsistency in the evolution of the abrasion resistance indicator. Specifically, compared to the control, the mass loss was lower for the composition with 40% RBA (R3-1), but slightly higher for the compositions in which the natural aggregates were substituted with larger quantities of RBA (R3-2 and R3-3). No clear trend can be established for this type of substituent aggregate. On the other hand, the use of blast-furnace slag (GBA) resulted in a significant improvement in wear resistance, with the performance indicator, mass loss, being 13.7% to 20.8% lower than the control sample. Surprisingly, the use of recovered textolite (RTA) as a replacement for natural aggregates resulted in wear mass loss values comparable to those of the control sample. A possible hypothesis for this behaviour is that although the adhesion between the binder and the recycled textolite aggregate is reduced, the small particle size of textolite and its specific textile nature cause the material to detach from the surface of the test specimen at a finer level, forming more of a powder (as evidenced during testing).

Fig. 10. Graphical representation of the wear resistance of cured composites 28 days after casting, expressed as percentage of mass loss.

In terms of water absorption by capillarity, the experimental results suggested a reduction in this parameter when natural aggregates were replaced by RGA and GBA, and an increase when natural aggregates were replaced by RBA and RTA. In the case of RGA, it is suggested that the reduction in water absorption may be a consequence of its specific
characteristics, namely a lower water absorption of glass aggregates compared to natural aggregates (Fig. 11).

Conversely, there is a reduction in the size of the pores in the binder matrix, as shown by the microscopic examination. When GBA is used, the reduction in water absorption by the composite can be attributed not only to the reduced porosity of the composite matrix, but also to changes in the hydration-hydrolysis compounds of the cement. The increase in water absorption observed in composites containing the other two types of recycled waste aggregates (RBA and RTA) has been attributed either to the higher water absorption of the aggregates, particularly those derived from ceramic brick waste, or to the changes that occur within the composite matrix, characterised by a reduced degree of cohesion between the binder and the aggregate. At the interface between the RTA and the cement binder, there are numerous areas where there is no proper contact, resulting in uneven and variable porosity in terms of distribution and size.

Microstructural analysis of cementitious composites

An SEM analysis was carried out to study the morphological characteristics of the cementitious composites providing an in-depth understanding of their microstructure. The analysis was performed on fractions of the control sample (R1), consisting of ordinary Portland cement paste and the control samples with RGA (R2), and with RTA (R5), after 28 days of maturation are shown, as shown in figure 12.

The selection of these two samples with RGA and RTA for SEM examination was based on their inert properties during the Portland cement hydration process. The main product formed during the hydration of Portland cement is calcium silicate hydrate (C-S-H gel) (Fig. 12a), which significantly influences the mechanical properties of the final material. Calcium hydroxide (CH), which has a plate-like blocky structure, and crystals of portlandite Ca(OH)$_2$, which form chip-like shapes in cavities, were also observed in control sample R1.

The characteristics and coherence of the Interfacial Transition Zone (ITZ) play a crucial role in determining the final properties of cementitious materials. As depicted in figure 12b, the ITZ, located between the aggregate and the main cementitious matrix, has a reduced content of both hydrated and un-hydrated products and a lower porosity compared to the bulk structure. In particular, there is a clear separation between the glass aggregates and the cementitious matrix, with the former being characterised by well-defined sharp edges and a lighter colour, while the latter has prominent, bulky, multi-layered crystal structures. This area is consequently characterized by a reduced bond between the cement matrix and the aggregate, resulting in decreased mechanical strengths due to the presence of a "weakness lines." Similarly, a complete disengagement of the aggregate from the cement matrix is evident in figure 12c, where the ground textolite particles manifest themselves in flake-like forms, either separated or positioned on top of the cement matrix. However, the ITZ of individual aggregates can also show interconnection as shown in figure 12c. In this region, the presence of clustered RTA flakes
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creates areas of reduced physico-mechanical strength as the RTA particles are loosely bonded within the C-S-H matrix.

![Image](https://via.placeholder.com/150)

**Fig. 12.** SEM images of the control sample, R1, at a magnification of x2000 (a), the sample containing RGA, R2, at a magnification of x2000 (b) and the sample containing RTA, R5, at a magnification of x2000 (c)

In comparison, as a result of its pozzolanic properties, slag actively participates in the cement hydration process by interacting with water and reacting with calcium hydroxide, thereby increasing the density and cohesion of the cementitious matrix, as shown in figure 13b in comparison with control sample R1. Both images (Fig. 13a and b) show typical hydration products such as C-S-H gel and CH crystals. However, figure 13b shows irregular, angular particles scattered throughout the matrix corresponding to unreacted granulated blast furnace slag grains. Despite these scattered unreacted slag grains, the matrix appears dense and compact, which may explain the increased compressive strength observed in the GBA samples. Because of its oxide composition, the slag actively participates in the formation of specific mineralogical products, influencing both quantitatively and qualitatively the mechanism and products of cement hydration-hydrolysis, and thus indirectly the mechanical strength. In addition, the microstructure of the GBA sample shows some microcracking, a common feature in such cements, resulting from the drying effect induced by the vacuum used for sample analysis.

![Image](https://via.placeholder.com/150)

**Fig. 13.** SEM images of the control sample, R1, at a magnification of x500 (a), and the sample containing GBA, R4, at a magnification of x500 (b).

**Conclusions**

Based on the information presented, a number of trends can be identified regarding the influence of the substitution of natural aggregates with aggregates derived from recycled glass waste (RGA), ceramic brick waste (RBA), blast furnace slag (GBA) or textolite waste derived from waste electrical and electronic equipment (WEEE) (RTA) on the physico-mechanical
performances, which correlate with the influences on the structure of cementitious composites. The density of cementitious composites decreases with increasing substitution of natural aggregates by recycled waste aggregates, particularly at higher substitution rates such as RTA, while the replacement of natural aggregates by granulated blast furnace slag (GBA) increases compressive and flexural strength, whereas other recycled waste aggregates, especially RTA, show a decrease. In particular, GBA significantly improves abrasion resistance, which is typically reduced by the substitution of natural aggregates, and capillary water absorption is influenced by the nature, behaviour of the substitutes and aggregate gradation, resulting in a mixture of decreases and increases.

The comprehensive analysis of the physical and mechanical performances and durability indicators, such as water absorption, revealed the following optimal compositions for waste derived aggregates in cementitious composites: RGA showed superior properties in composition R2-1 with 45% substitution, RBA showed better performance at minimum substitution (R3-1), GBA showed favourable results at maximum substitution (R4-3), and the limited literature on textolite led to a single choice of composition (R5-1) with 10% substitution. These results emphasise the nuanced effect of waste aggregates on composite properties and highlight the need for careful consideration in material selection.

The microstructural study, which includes the identification of characteristic cement hydration compounds and the observation of areas of microcracking, reduced adhesion between the cement matrix and aggregates, and aggregate clusters, corroborates the experimental results in terms of physico-mechanical performances. At the same time, this analysis highlights the influence of the nature of the aggregates on the properties of the composites, emphasising the need for preliminary studies whenever changes are made in the type or dimensions of the aggregates replacing natural ones in cement composites. The experimental results confirm the demonstrated feasibility of partially replacing natural aggregates with recycled waste aggregates in cementitious composites, which, depending on their nature, have different effects on the performance of the composites, and contribute to environmental protection by reducing the negative impact on the soil, either from waste disposal or from changes in the local topography due to quarrying. The research also points to the potential for developing materials that are compatible with original materials and suitable for the restoration of heritage buildings, particularly those made predominantly of ceramic bricks.

Finally, it is appreciated that this analysis contributes to the advancement of knowledge in the field and to the alignment of the results with other reports in the scientific literature. This opportunity arises from the real need to develop and produce eco-innovative building materials adapted to local raw material and waste characteristics, which remains a persistent priority at both national and international levels, given the urgent push for greater implementation of circular economy principles.

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References


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[42] O. Corbu, I.O. Toma, Progress in Sustainability and Durability of Concrete and Mortar Composites, Coatings, 12(7), 2022, Article number: 1024.


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