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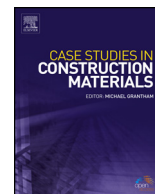
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Case study

Mechanical properties of mortar containing waste plastic (PVC) as aggregate partial replacement

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ABSTRACT

The purpose of this work is the reuse of polyvinyl chloride (PVC) deriving from waste electrical and electronic equipment (WEEE) used as a partial substitute for the mineral aggregate to produce lightened mortars. PVC was recovered from copper electrical cables, ground and used as replacement of mineral aggregate in 5, 10, 15 and 20 % vol. in mortar. A thermal characterization of the starting material was carried out to understand its composition. The mortar samples were mechanically tested both using class G cement and ordinary Portland cement. The results showed a worsening of the mechanical properties of around 50 % for only 5 % in volume of sand substituted with PVC waste. A likely explanation to this phenomenon was found in the mechanical characteristics of the PVC used and to its poor adhesion with the matrix, that resulted in the creation of porosity. However, the mortar prepared contributes to the conservation of natural resources and maintains mechanical properties adequate for the use in non-structural applications (e.g. screed or substrate).

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1. Introduction

Waste electrical and electronic equipment (WEEE) represent a peculiar type of waste coming from everyday devices. They are made up of a heterogeneous mix of materials with various additives [1] and, generally, contain traces of compounds hazardous to human health [2]. Currently new methods of recycling this typology of waste materials are under investigation, but their inhomogeneity and composition, especially of the polymeric component [3], strongly limits their possibility of being reused and recycled. Plastic materials derived from electronic waste, thus, are generally destined to landfilling. Usually PVC is used in many different applications, from piping to window profiles in its rigid form, and can be progressively additivated, up to almost 60 % of the final composition, in order to obtain a more flexible material [1], ideal for cables insulation and electronic wires. Once its life cycle comes to an end, although, most of the PVC is destined to landfilling [4], but little is known to what extent the additives are mobilised and how they contribute to landfill leachate and gases [5]. In particular, while some processes and techniques were found to give a second life to rigid PVC [1,6,7], the flexible PVC used for electronic equipment hardly finds an alternative to waste disposal. In fact, this PVC contains as additives numerous substances that may prove hazardous to human health [1], like lead and cadmium stabilizers, or plasticizers such as di(2-ethylhexyl)phthalate (DEHP) [4], which are linked to health or environmental risks. That, combined with the hazards of chloride-containing compounds, limits its possibilities of recycling [5]. Besides, in the European Union, in 2000, 3,6 million

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tons of waste polyvinylchloride were produced, with a predicted growth of 6,4 million tons in 2020 [4]. Of this quantity, around 60% is made up by flexible PVC. These numbers alone, especially if inserted in the wider framework constituted by all the remaining WEEE waste, suggest its environmental impact and underline the necessity of finding ways to re-use this material in an alternative way.

The use of polymeric waste as substituent of the aggregate in mortar and concrete seems a valid solution to combine two most current necessities: a re-use technique that limits the reprocessing, thus preventing any hazardous substance to leach or any degradation to take place, and at the same time allows large volumes of recycling [8,9]. The construction sector has continued its development over the last decades thanks to the boost provided by developing countries, with an increasing demand for cement and aggregates to obviate the requests of the growing markets. In 2015, cement production in the European Union amounted to 167 million tons, placing the EU as the third largest producer behind India and China, whose combined production matches 2,6 billion tons per year, contributing to the 57.3% of the global production in 2015 [10]. Concrete manufacturing is responsible of the impoverishment of natural resources and raw materials the planet provides, like limestone, clay and coal, whose exploitation results in deforestation, and top-soil loss [11,12]. The substitution of aggregate with waste materials would then combine two advantages, not only decreasing the amount of landfilled plastic materials, but also preserving the natural resources and the environment [13,14]. The use of plastics in concrete would, in this scenario, decrease the amount of inert aggregate used.

The substitution of aggregate with polymeric waste in general [15–19], and PVC waste specifically [20,21], was already explored in literature. Data are quite scattered in terms of what component is substituted (fine [22,23] or coarse [24,25] aggregates, both [26] or even cement [27]), substituted quantities (%weight [27,28] or %vol [22,23,25,26,29]) and type and size of both the substitute and the substituent. A common factor seems to be the substitution of aggregate with rigid PVC, generally deriving from piping or fixtures [22–25,28–31]. As a rule, the mechanical properties of the PVC waste containing concrete decrease with respect to the ordinary concrete [22,29–31], in line with what was observed more generally for polymeric waste containing concrete [15]. At the authors' knowledge, the effects of flexible PVC in mortars or concretes have not been studied in the literature.

In this work, the possibility of using plastics deriving from PVC wires (flexible PVC) to replace aggregates in mortars is investigated. After the chemical, physical and dimensional characterization of the material, the polymeric waste was added to the paste in substitution of the natural aggregate, and the resulting mortars have been tested for compressive and flexural strength, in order to investigate their mechanical performance in comparison to the one of ordinary concrete, containing only inert aggregate. Stereomicroscopy was then used to analyse the interface between substituent and the cement paste.

2. Materials and methods

2.1. Materials

The PVC waste, collected from a local waste demolition site (Amiat S.p.A.), is the results of the end-of-life line of the copper recovery process from data cables and wires. The material, shown in Fig. 1, is a mix of differently shaped granules with different sizes, irregular and unravelled surface, because of the shredding treatment the copper recovery process requires. The waste bulk density was evaluated through pycnometer (Quantachrome Ultrapyc 1200e). The result obtained for the bulk density of the waste was an average of ten measurements. Granulometry was investigated through manual sieving using sieves of 4 mm, 3 mm, 1.7 mm, 1 mm, 420 μm , 180 μm and 63 μm .

Polymeric waste was thermally characterized through thermogravimetric analysis and differential scanning calorimetry, in order to have a qualitative evaluation of its composition and to understand whether a treatment in temperature may deteriorate the material. The thermo gravimetric analysis was made with a Mettler Toledo model 1600. The analysis was performed in flowing air at 50 mL/min, with a heating ramp of 10 °C/min up to 1000 °C. The apparatus used in this study for differential scanning calorimetry is a Perkin-Elmer Pyris 1 Heat-Flux differential scanning calorimeter, working in Ar atmosphere from 25 °C to 400 °C with a heating ramp of 20 °C/min.

For the mortars preparation, an American Petroleum Institute (API) oil-well cement Class G (Lafarge North America) was used. The use of Class G cement required a curing time of 24 h at 85 °C in 100% relative humidity. Ordinary Portland cement (Calce Cementi Lauriano Cem II/A-LL 42.5R) was also used in this study to compare the mechanical performances of mortar. CEN Standard sand (a natural siliceous sand consisting of rounded particles having a silica content of at least 98%, whose particle size distribution lies within specific limits according to UNI EN 196-1) was used as fine aggregate and was purchase from Société Nouvelle Du Littoral.

Five distinct mortar mixtures were produced with and without PVC waste aggregates. In mortars compositions, the natural sand was progressively substituted by increasing quantities of polymeric waste to realize 0, 5, 10, 15, 20 %_{vol} substitution. The samples will be respectively referred as 0PVC, for 0 %_{vol} of natural sand substituted with PVC waste, and 5PVC, 10PVC, 15PVC and 20PVC, where the number in front of the acronym indicates the percentage in volume of sand substituted with polymeric waste; the quantities used are shown in Table 1. To check the consistency of the results obtained with class G cement, then, type II Portland cement was used to realize mortars with 10 and 15 %_{vol} polymeric substitution and a reference with 0% substituent. These samples are labelled 0PVC28 for the reference sample with no natural aggregate substitution, and 10PVC28 and 15PVC28 for respectively 10 and 15 %_{vol} substitution of sand with polymeric waste; the mix design is shown in Table 2. Constant water to cement ratio (equal to 0.5) was used to produce all the samples.

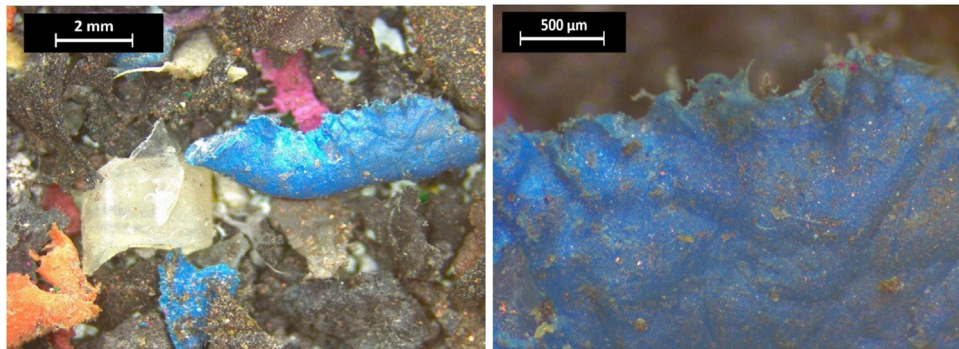


Fig. 1. Stereomicrographs of polyvinyl chloride waste (left) and detail (right) to show the unravelled, irregular surface.

Table 1

Mix design for the PVC composites prepared with class G cement.

| Name of sample | Cement (g) | Water (g) | Sand (g) | PVC (g) |
|----------------|------------|-----------|----------|---------|
| 0PVC | 71.2 | 35.6 | 213.7 | 0 |
| 5PVC | 71.2 | 35.6 | 203.0 | 5.8 |
| 10PVC | 71.2 | 35.6 | 192.3 | 11.5 |
| 15PVC | 71.2 | 35.6 | 181.7 | 17.3 |
| 20PVC | 71.2 | 35.6 | 171.0 | 23.1 |

Table 2

Mix design for the PVC composite prepared with ordinary Portland cement.

| Name of sample | Cement | Water (g) | Sand | PVC |
|----------------|--------|-----------|-------|------|
| 0PVC28 | 71.2 | 35.6 | 213.7 | 0 |
| 10PVC28 | 71.2 | 35.6 | 192.3 | 11.5 |
| 15PVC28 | 71.2 | 35.6 | 181.7 | 17.3 |

2.2. Preparation of specimens

The mixing and preparation were done following the guidelines of UNI EN 196–1: first water and cement were mixed at low speed for 30 s. The aggregate, a mix of natural sand and shredded polyvinyl chloride in the correct proportions, was then progressively added during the following 30 s, with additional 30 s mixing of all the components at higher speed. The paste was manually blended for the next 30 s, and then left standing for additional 60 s. At the end of this resting time, the paste was finalized with 60 s mixing at high speed. The slurry was then poured in suitable moulds of 20 × 20 × 80 mm. A representative image of the different prepared samples is shown in Fig. 2

The samples were then treated accordingly to the requirements of the cement used: mortars realized with class G cement were left in the oven at 100 % of relative humidity for 24 h at a temperature of 85 °C, while the mortars realized with Portland cement had a 28-days curing time at a temperature of 20 °C in 100 % relative humidity.

2.3. Mechanical tests and optical characterization

To evaluate the influence of PVC substitution level, and thus the concreteness of the hypothesis and the feasibility of using such waste as substituent of the natural aggregate in mortars and concretes, density evaluations, compressive tests and 3-point flexural tests were done.

Density was evaluated geometrically. Samples were polished to obtained flat, perpendicular surfaces. Width, length and height were estimated with a caliper, and the samples weighted on a RADWAG PS 510/C/1 analytical balance.

Flexural analysis was performed with a single-column Zwick-Line z050 having a maximum cell load of 1 kN, with a pre-load of 5 N and a test speed of 0.01 mm/min. One of the two halves obtained by flexural tests was used to evaluate the compressive strength. Compressive tests were performed using the same machine but with a cell load of 50 kN, a pre-load of

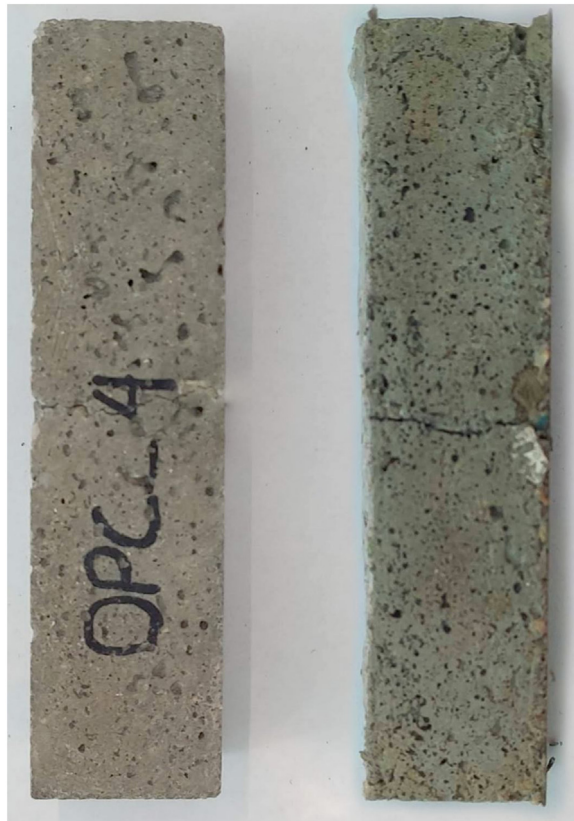


Fig. 2. Different prepared samples: OPC specimen (left), 15PVC specimen (right).

30 N and a test speed of 0.5 mm/min. Each result of compressive and flexural strength was an average of at least four measurements.

Stereomicroscope Leica EZ4 W was used to investigate the interface adhesion between the polymeric aggregate and the cement paste.

3. Results and discussion

3.1. Characterization of the PVC and specimens

As shown in Fig. 1, the polyvinyl chloride waste has a dirty, irregular surface, because of the shredding and mixing procedure required to recover copper. This process leaves on the surface some copper powder. PVC particles recovered have different size and dimension and the cumulative granulometric distribution is shown in Fig. 3. Four mm is considered the superior dimensional limit for fine aggregate, used for mortar preparation. The graph shows, together with the granulometric distribution of the shredded PVC, in black-square, the Fuller curve, in blue-star. This curve represents the ideal granulometric distribution of the aggregate to obtain the maximum density, and minimum porosity, in the final material. For an aggregate size above 2 mm, the two curves have net similarities. The difference between the two distributions is, on the other hand, more pronounced for smaller sizes of the aggregate, with more than 30 % of PVC having a size comprised between 1 and 1,7 mm, with respect to the about 10 % dictated by the Fuller curve. This discrepancy determines a non-ideal packing of the aggregate leading probably to the formation of porosity. Porosity strongly contributes to the mechanical behaviour of the material [32,33].

PVC mean particle density was estimated to be 1.4 g/cm³, in line with analogous waste used in literature [34]. The fineness modulus of PVC is 3.4 while for standard sand used is 3.0. The thermal stability of the PVC used, measured by thermogravimetric analysis (TGA), is shown in Fig. 4a. The first degradation, starting at 240 °C, with a weight loss around 45 %, is typically due to the release of HCl and phthalates, together with CO₂, probably resulting from the reaction between HCl and gypsum, a common filler used in the PVC for wires and cables [35]. A second degradation phase starts at 408 °C, leading to a 26 % loss in weight corresponding to the thermal cracking and decomposition of the PVC backbone, with the formation of water and carbon dioxide [36]. The third weight loss, in the range 600–750 °C, is likely linked to the thermal degradation of carbonate fillers with formation of CO₂ [35].

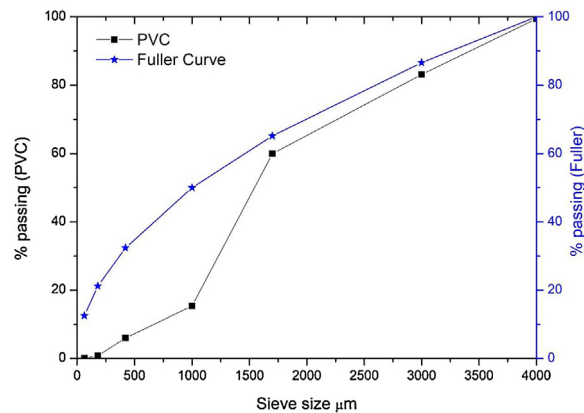


Fig. 3. Cumulative granulometric distribution of polymeric waste (in black-square) and Fuller curve (in blue-star). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

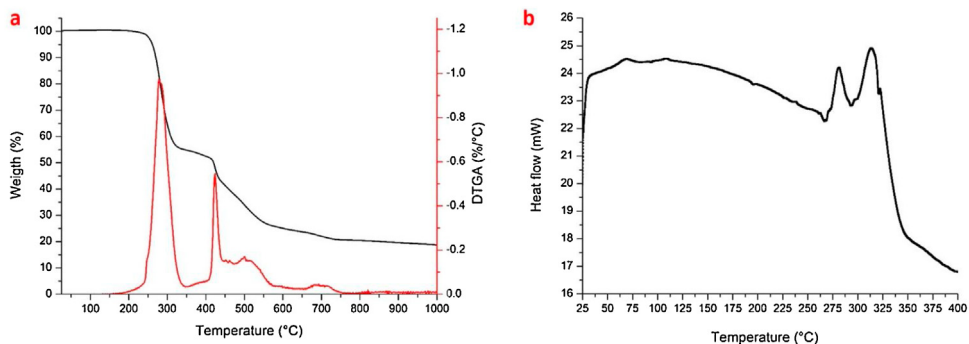


Fig. 4. a) Thermogravimetric analysis of polyvinyl chloride waste performed in air with a heating ramp of 10 °C/min. b) Differential scanning calorimetry of PVC in Ar atmosphere from 25 °C to 400 °C with a heating ramp of 20 °C/min.

Differential scanning calorimetry (Fig. 4b) delivers results that are in line with those obtained for the thermogravimetric analysis. The single peak centred at around 300 °C in the TGA is the combination of two peaks, that appear more clearly in the DSC analysis. These two peaks, at 281 °C and 313 °C respectively, were similarly observed on analogous PVC waste in literature [36].

Densities of the mortar test specimens are presented in Fig. 5. The lower specific gravity of the polymeric substituent with respect to the natural aggregate determines a decrease in density of the final mortars, proportional to the amount of polymer substituted.

3.2. Mechanical properties

The samples were tested for 3-point flexural strength, in order to evaluate the correlation between their resistance and the quantity of polymer waste substituted to the natural aggregate. Results of the flexural tests conducted on class G cement mortars are shown in Fig. 6a. The flexural strength decreases with an increase in plastic content, but the correlation between the polymeric volume fraction and flexural strength is not linear. For 5 % substitution, the loss in flexural strength is around 30 %, and reaches almost half the initial value at 15 % vol of PVC.

The trend obtained for flexural strength is confirmed by the results of compressive tests, shown in Fig. 6b. Compressive strength decreased with the increase in substitution, but again there is no linear correlation. At low percentages of substitution, compressive strength presents a considerable drop compared to the drop in flexural strength, above 50 % of its original value for only 5 % in volume of sand substituted with PVC waste. The ulterior loss for higher polymeric content, on the other hand, seems to be minor for substitutions up to 15 %. Class G cement is used for oil-wells, but it is not common in the building sector. To evaluate how the substituent would impact mortars realized with cements commonly used for mortars and concrete, samples of the same dimension and polymer content, but made with ordinary Portland cement, were produced. Results obtained for flexural tests on Portland cement mortars are shown in violet in Fig. 5c, in comparison to those collected for class G cement mortars, in yellow. The behavioral trend of the two is analogous, but mortars realized with 28-days cement showed a smaller loss in mechanical properties with respect to those done with class G cement. The same

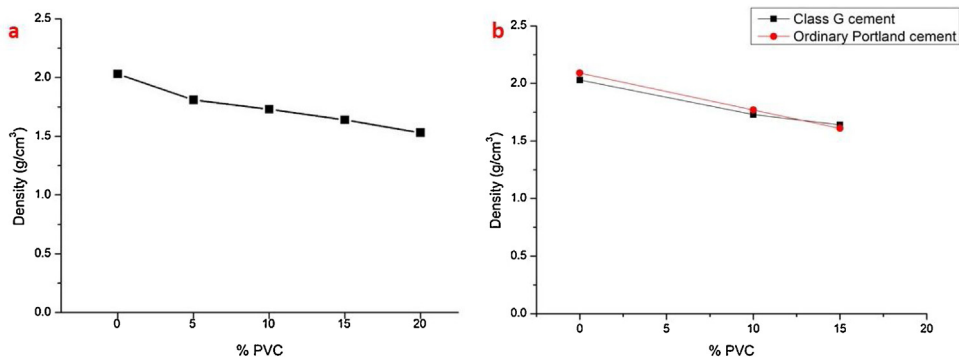


Fig. 5. Variation of mortar specimens density in relation to the percentage, in volume, of natural aggregate substituted with PVC waste using class G cement (a) and comparison between class G and ordinary Portland cement (b).

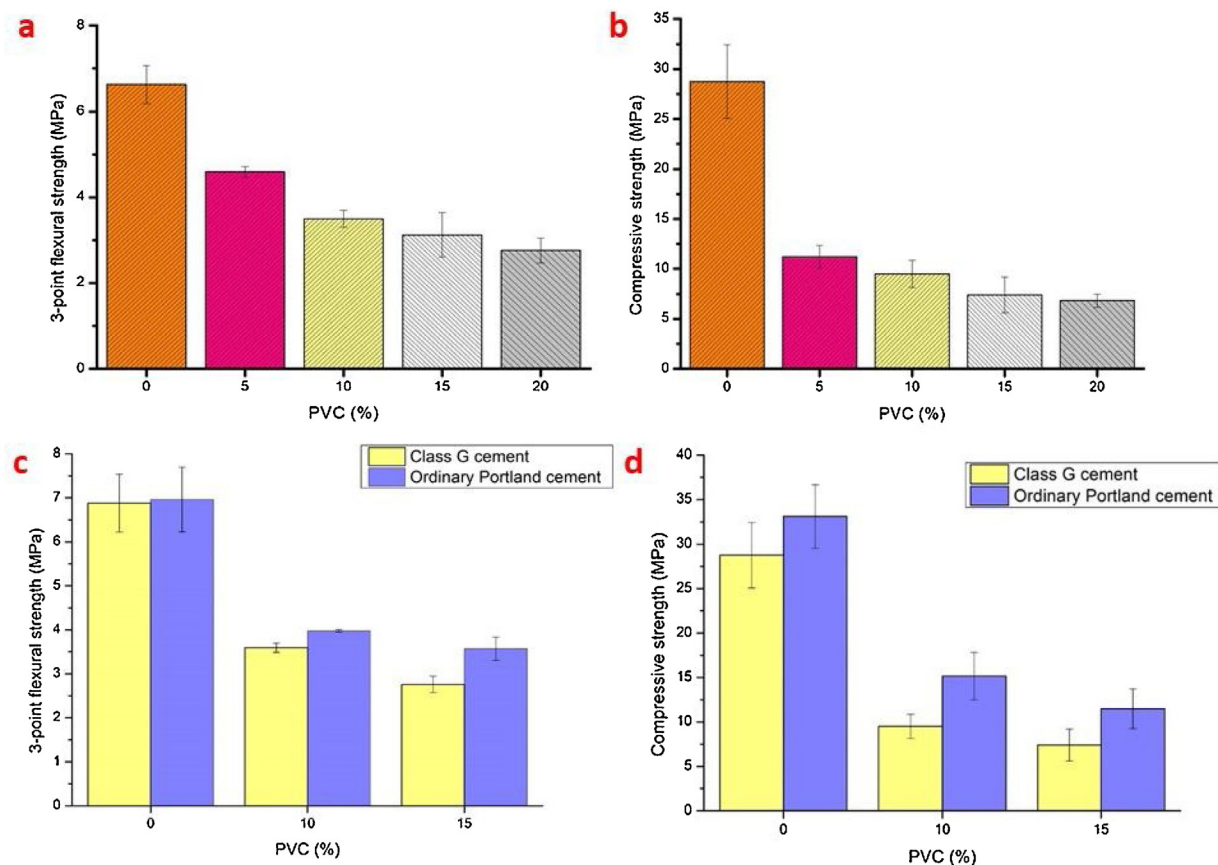


Fig. 6. Variation of mechanical properties in relation to the percentage, in volume, of natural aggregate substituted with PVC: a) 3-point flexural using class G cement. b) Compressive strength using class G cement c) Comparison in the variation of 3-point flexural strength in case of Class G cement (in yellow) and ordinary Portland cement (in violet) d) Comparison in the variation of compressive strength in case of Class G cement (in yellow) and ordinary Portland cement (in violet). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

can be observed for the results of compressive tests (Fig. 5d): again, the trend is analogous to that obtained for class G cement, but the reduction in mechanical properties is less prominent.

The worsening in mechanical properties can be explained by considering the difference in mechanical properties between the natural aggregate and the polymeric substituent. The mechanical properties of the substitute itself are much worse with respect to natural aggregate. Most likely PVC does not participate in mechanical performance and behaves like porosities, thus weakening the structure. In favour of this theory, the quantity and percentage of pores increases as a consequence of the use of PVC inside the matrix. The ratio between the mechanical data obtained by compression tests and

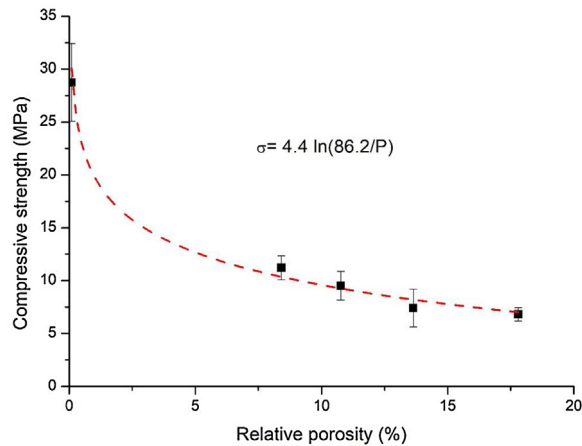


Fig. 7. Correlation between the compressive strength and porosity of the pristine mortar and mortar containing 5, 10, 15, 20 % PVC prepared with class G cement as substitution of mineral aggregate.

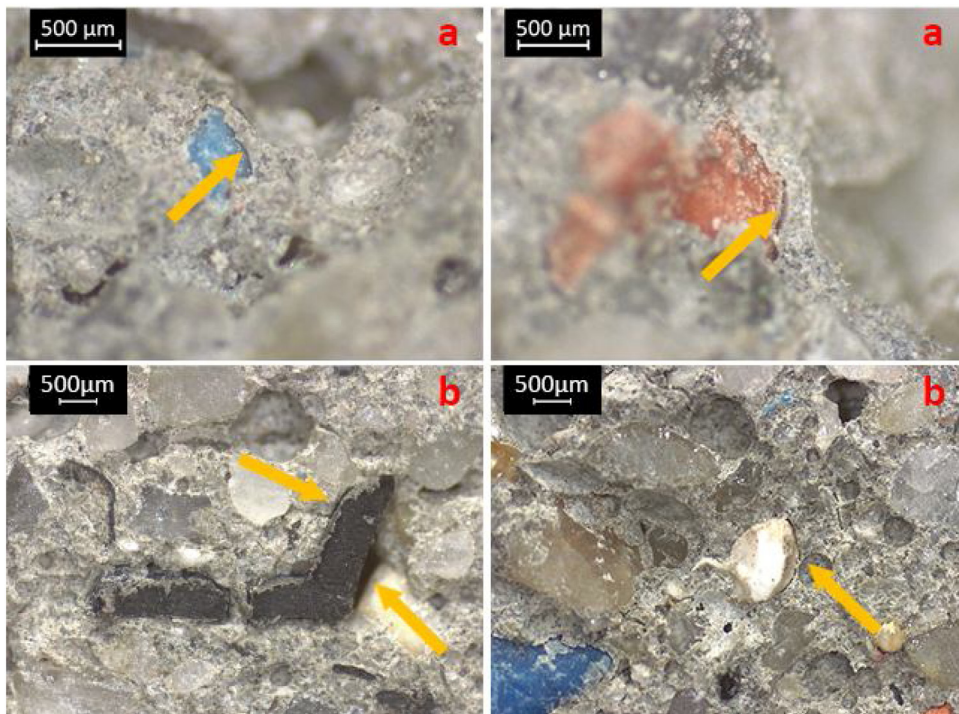


Fig. 8. Stereomicrographs of mortars with class G cement (a) and with ordinary Portland cement (b) containing PVC waste in partial substitution of natural aggregate.

the relative porosity percentages (calculated as the difference between the porosity of the samples with PVC compared to the porosity of the reference sample) follow the Schiller model [37] for porous materials as shown in Fig. 7. Schiller's model is a theoretical approach that deduced a relation between the strength of material to the porosity through the following formula:

$$\sigma = n \ln \frac{P_0}{P}$$

where n is an empirical constant, P_0 is the porosity at zero strength. In this model, a large mechanical loss is expected at small porosity percentages as in our case. The limit of the Schiller model, as explained by Chen et al. [38], are at the extremes: close to 0 % porosity and 100 % porosity.

Another important point is the materials adhesion to cement paste. The lack of adhesion at the interface, indicated by yellow arrows in Fig. 8, was detected through stereomicroscopy. The inert used for the ordinary mortars is a polar material

that well combines with the polar characteristics of cement, favouring the surface adhesion and thus the mechanical properties of the material thanks to a better interface. Polymeric waste is made up of organic compounds that differ largely from the composition of the natural aggregate, for instance PVC is weakly polar. These characteristics determine a scarce interface adhesion between the substitute and the cement, and thus contribute to the loss in mechanical properties.

The use of class G cement entails a curing time of 24 h in oven at 85 °C and 100 % humidity. Such conditions may partially impact the surface adhesion of the polymer to cement: the difference in their expansion coefficient may imply the formation of residual stresses at the interface or detachment during cooling. Such phenomena may favour the creation of cracks and reduce the mechanical resistance.

If compared to those present in literature for mortars/concretes containing rigid PVC, our results are characterized by worse final mechanical properties. As already mentioned, the PVC used in this work is polyvinyl chloride deriving from the shredding of electrical cable and wires, and thus a flexible material with lower modulus and lower tensile strength with respect to the rigid PVC derived from pipes and fixtures used in literature. This difference, together with the scarce surface adhesion, could already provide sufficient explanation of the gap with respect to the values obtained in other investigations. If compared to other studies the literature [18,19,39], in which other plastic waste has been used as a substitute for the aggregate, the loss of mechanical properties is higher, due to the different structural nature of the plastic used.

4. Conclusions

This paper investigated the suitability of using flexible PVC derived from waste electric and electronic equipment as substitutive aggregate in the production of mortars. According to the experimental results presented previously, the following conclusions can be drawn:

- Compressive strength diminished with an increase in PVC content. While the loss in resistance is noteworthy starting from low percentages of substitution (around 50 % for 5 % vol PVC substituted to natural aggregate), the trend is much less steep for higher PVC content. Such decrease in mechanical resistance is linked to the lower resistance of the substituent with respect to the natural aggregate. The trend obtained for PVC mortars produced with class G cement was confirmed by the results obtained with the PVC mortars made with Portland cement. In case of normal 28 days cement, however, the loss is less steep.
- Flexural strength diminished with increasing PVC substitution to natural aggregate. Compared to compressive resistance, however, the initial drop is limited, with a loss of 30 % of the initial value for 5 % substitution, and the overall loss less significant. Again, the trend obtained for PVC mortars produced with class G cement was confirmed by the results obtained with the PVC mortars made with Portland cement.
- The use of class G cement delivers reliable preliminary results in a shorter time with respect to Portland cement, correctly describing the behavior of the material while generally being more conservative because of the high temperature curing process typical of class G cement.
- The loss in mechanical properties can be ascribed to two main components. The first is the intrinsic difference between the natural aggregate, ceramic and polar, and the substituent, plastic, weakly polar, and with lower mechanical properties. This difference results necessarily in a worsening of both compressive and flexural strength. The second is due to the lack of adhesion between the cement paste and the polymeric aggregate, as shown by the microscopic investigation, that further compromises the mechanical properties of the final product.

In conclusion, the use of polymeric waste as a replacement of inert aggregate permits the realization of a product which is eco-friendly, contributes to the conservation of natural resources and maintains mechanical properties adequate for the use in non-structural applications.

To guarantee the possibility of legally using the resulting material for more specific, load bearing applications, it is necessary to identify suitable reinforcements or efficient methods of recovering mechanical properties, so to outdo the limits required by local legislations. To reinforce these typologies of composites the use of polymeric or metallic fibers can improve the mechanical properties. Alternatively, expanding oxides (such as MgO) can be dispersed on the surface of the polymer thus improving the interaction with the cement. Finally, ulterior research is necessary to determine how the addition of polymeric material inside the mortar varies the thermal conductivity, and possibly the acoustic properties, of the final product. In this scenario, the product may turn out to be a concrete alternative to current commercial products, for it would be greener while guaranteeing interesting physical properties.

CRedit authorship contribution statement

Alessandra Merlo: Conceptualization, Investigation, Validation, Writing - original draft. **Luca Lavagna:** Methodology, Validation, Formal analysis, Writing - review & editing, Supervision, Data curation. **Daniel Suarez-Riera:** Investigation, Validation, Visualization, Writing - original draft. **Matteo Pavese:** Conceptualization, Supervision, Writing - review & editing, Resources.

Declaration of Competing Interest

The authors report no declarations of interest.

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