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Mechanical Behaviour Analysis of Polyester Polymer Mortars Modified with Recycled GFRP Waste Materials

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Abstract**—**In this study the effect of incorporation of recycled glass-fibre reinforced polymer (GFRP) waste materials, obtained by means of milling processes, on mechanical behaviour of polyester polymer mortars was assessed. For this purpose, different contents of recycled GFRP waste powder and fibres, with distinct size gradings, were incorporated into polyester based mortars as sand aggregates and filler replacements. Flexural and compressive loading capacities were evaluated and found better than unmodified polymer mortars. GFRP modified polyester based mortars also show a less brittle behaviour, with retention of some loading capacity after peak load. Obtained results highlight the high potential of recycled GFRP waste materials as efficient and sustainable reinforcement and admixture for polymer concrete and mortars composites, constituting an emergent waste management solution.

Keywords**—**GFRP waste, Mechanical behaviour, Polymer mortars, Recyclability.

I. INTRODUCTION

GLASS and carbon fibre-reinforced polymers (GFRP and CFRP), nowadays commonly used in the construction, transportation and automobile sectors, have been considered inherently difficult to recycle due to both: cross-linked nature of thermoset resins, which cannot be remoulded, and complex composition of the composite itself, which includes glass/carbon fibres, organic matrix and different types of inorganic fillers [1]. Presently, most of the FRP waste is landfilled leading to negative environmental impacts and supplementary added costs. With an increasing awareness of environmental matters and the subsequent desire to save resources, recycling would convert an expensive waste disposal into a profitable reusable material [2].

At present, there are three main methods to recycle FRP thermostable materials: (a) incineration, with partial energy recovery due to the heat generated during organic part combustion; (b) thermal and/or chemical recycling, such as solvolysis, pyrolisis and similar thermal decomposition processes, with glass or carbon fibre recovering; and (c) mechanical recycling or size reduction, involving breaking-down of the composite by shredding, crushing, milling, or other similar mechanical processes; the resulting scrap pieces can then be segregated by sieving into fibrous products (richer in fibres) or powdered products (richer in resin), that makes de material suitable as reinforcement or filler in new composite products. [3]-[5].

Mechanical recycling has important advantages over the previous ones: there is no atmospheric pollution by gas emission, a much simpler equipment is required as compared with ovens necessary for thermal recycling processes, and does not require the use of chemical solvents with subsequent environmental impacts [3]. Providing feasible market outlets exist, mechanical recycling is the favoured recovered technique, at least for relatively clean waste materials. Mechanical recycling of FRP waste remains however hindered by the scarceness of viable end-use applications.

The need for valuable applications for FRP waste materials, and the sequent pressure on the development of new economically viable markets for the recyclates, have driven over the last 15 years a relative great amount of research work on recycling techniques and related potential applications [2], [4], [6]-[9]. Several potential uses for ground FRP waste have been investigated. Reinforcing filler for artificial wood [10], HDPE (high density polyethylene) plastic lumber [11], rubber pavement blocks [12], bulk (BMC) and sheet (SMC) moulding compounds [4]-[5], or dense bitumen macadam [13]; reinforcement for wood particleboard [9], and core material for textile sandwich structures [14] were some of the foreseen potential recycling applications.

Nevertheless the wide scope of potential applications among composite materials, the most extensive research work has been carried out on Portland cement concrete, in which grinded FRP waste scrap has been incorporated either as reinforcement, aggregate or filler replacement [15]-[20]. Reported added values, besides environmental benefits, as function of specific mix formulation and design, comprise slight increase on mechanical properties, lower permeability with subsequent improved durability, a less drying shrinkage and a global cost reduction of raw materials. Potential applications of FRP waste in concrete include pre-cast paving slabs, roof tiles, wall panels, paving blocks and architectural

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cladding materials [15]-[18].

Regardless of the relative large amount of research work undertaken recycled FRP waste in cementitious based concretes, few studies have been reported until now, on the effect of the incorporation of these sort of organic/inorganic wastes on polymer concrete materials [21]-[22].

Polymer concrete (PC) materials have gained an increasing research interest due to their wide range of possible applications in civil construction [23]-[25]. In this class of materials, a thermoset resin is used as binder of natural or artificial aggregates, replacing the paste of Portland cement/water of conventional hydraulic concretes. The most commonly resins used as matrix binders have been unsaturated polyesters, acrylics and epoxies systems [26].

The initial applications of PC, in the late 50's, were the production of building cladding and cultured marble, but its excellent properties rapidly widespread its application fields. Its rapid curing, excellent bond to concrete and steel reinforcement, high strength and durability made it a very attractive repair material. As a mortar (PM) it can be placed with thickness less than 10 mm. Overlays in PC, for bridge surfaces and floors, have also become widely used because of the ability to use thin layers, fast curing time, very low permeability, and high resistance to chemical and frost attack. Precast components are another excellent use of the material. The high strength to weight ratio, good damping properties, moldability and ability to form complex shapes make PC and PMs particularly suitable for these applications [23]-[30].

However, currently, the main asset of PC materials over conventional concretes is their great ability for incorporating recycled waste products, mainly owned to hermetic nature of resin matrix. Recycling and waste encapsulation constitute nowadays the new and emerging branch market for PCs. Most of the successful applications reported involved either industrial by-products or end-life products. Industrial wastes, such as fly ash, slag, wood ships, cork powder and cork granulates, tire rubber, plastic ships from used polyethylene and polyvinyl chloride films, as well as plastic granulates proceeding from milled waste electrical cables, have been successfully used for replacing or partially substituting the filler and aggregate components in PC materials [21],[22],. [31]-[33].

One of the main advantages in using a cementless concrete as host material for GFRP waste incorporation is the insurance that no chemical incompatibility due to alkali-silica reactivity occurs. Under this point of view, and following the above studies, the aim of the present work is to explore a potential waste management solution for GFRP waste (scarp, by-products and end-life products) as reinforcement, aggregate or filler replacement for polymer based mortars. For this purpose, different contents of recycled GFRP waste powder and fibres, with distinct size grading, were incorporated into polyester based mortars as sand aggregates and filler replacements. Added value of recycling solution was assess by means of flexural and compressive loading capacity of GFRP admixed mortars with regard to unmodified plain polymer mortars.

The applied waste material was supplied by *ALTO – Perfis Pultrudidos Lda.*, and it was proceeding from the shredding of the leftovers (edges and small pieces) resultant from the cutting and assembly processes of pultrusion profiles. Currently, these leftovers, jointly with unfinished products and scrap resulting from pultrusion manufacturing process, are landfilled, with an estimated cost of 80€ per ton., which result for this company on an average cost of 3500€ per year.

Thus, besides the evident environmental benefits, a viable and feasible solution for these wastes would also conduct to significant economic advantages.

II. EXPERIMENTS AND PROCEDURES

Detailed experiments were carried out on the use of grinded GFRP pultrusion waste in polymer based mortars. Mix design of plain formulation was in accordance with previous studies carried out by Ribeiro and Ribeiro et al. [27]-[34]. Polymer mortar specimens were prepared by mixing an unsaturated polyester resin with different sand aggregates/GFRP waste ratios. Processed GFRP wastes, with two different size gradings, were used as a partial substitute for sand aggregates at the proportion of 4% and 8% (w/w). Plain or control mortar specimens were also cast and tested in order to compare mechanical and functional properties over those obtained with GFRP waste admixed mortars.

A. Binder Matrix and Sand Aggregates

Commercially available unsaturated polyester resin, with the trade name of AROPOL FS3992 supplied by *Ashland*, was used as binder. AROPOL FS3992 is a rigid, high reactivity and low viscosity resin, with a styrene content of 42%, generally used in pultrusion processes, though it can also be used in bulk and sheet moulding compound applications. This resin system is the same applied as matrix in the manufacturing process of GFRP pultrusion profiles produced by *ALTO*, and its application in this study as polymer mortar binder was justified in order to prevent possible incompatibilities with GFRP waste admixtures. Polymerization process of resin system was induced by cobalt octoate (0.5 phr), as promoter, and 50% methyl ethyl ketone peroxide solution (2 phr), as initiator. Typical physical properties of cured resin, supplied by the manufacturer, are presented in Table I.

TABLE I
PHYSICAL PROPERTIES OF CURED RESIN (AROPOL FS3992)

Physical Properties	Method	Value
Heat Deflection Temperature **(ºC)**	ASTM D-648	95
Tensile Strength (MPa)	ASTM D-638	60
Flexural strength (MPa)	ASTM D-790	110
Barkoll Hardness	ASTM D-2583	45
Elongation at Break (%)	ASTM D-638	3.2

A siliceous foundry sand, with a rather uniform particle size with an average diameter of 245 μm, was used as fine aggregate. Foundry sand is a generic term to denote sand with a high-grade of silica (> 99.0%). This silica sand is extracted/processed by *Sibelco, Lda*, and has been commercialised by *Fundipor* under the commercial name SP55. Particle size distribution of SP55 foundry sand is represented ahead in this paper in Fig. 2, jointly with particle size distributions of milled GFRP waste materials.

B. GFRP Pulruded Waste Admixtures

Grinded GFRP pultruded waste supplied by *ALTO, Perfis Pultrudidos, Lda* was applied in this study as admixture and partial sand replacement. GFRP waste was further processed by milling on a heavy-duty Cutting Mill laboratory unit (Type SM2000, Retsch). Two different size gradings of milled GFRP waste were obtained using bottom sieves with different perforation sizes inside the grinding chamber. Perforation size of bottom sieve determines the fineness of the material being comminuted. Obtained recycled products, a mix of powdered and fibrous material with different quantities of varying length of glass fibres, hereinafter designated by coarse (CPW) and fine (FPW) pultrusion waste, are show in Fig. 1.

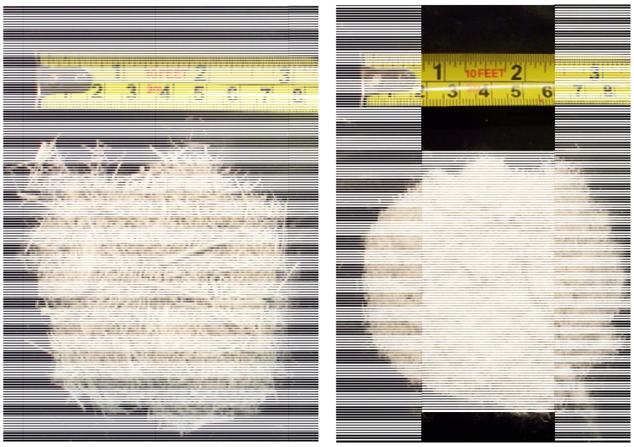

Fig. 1 Milled GFRP pultrusion waste: CPW -coarse pultrusion waste (in the left), and FPW -fine pultrusion waste (in the right).

GFRP wastes were characterized with report to organic and inorganic fraction contents and particle size distribution.

Burning tests carried out at ‘Fire Laboratory’ of INEGI (Inst. of Mechanical Eng. and Industrial Management), on five random samples, revealed an average inorganic material content of 71% (v/v), corresponding to glass fibre and calcium carbonate fractions, and an average resin content of 29% (v/v).

Particle size distribution obtained by sieving process of both types of recycled wastes, in accordance with EN 933-1:1997 [35], are presented in Fig. 2. Particle size distribution of filler fractions (< 74 μm), were further evaluated by laser diffraction technique using a Particle Size Analyser Laboratory Unit (Malvern Mastersizer 2000G). Obtained results are presented in Fig. 3.

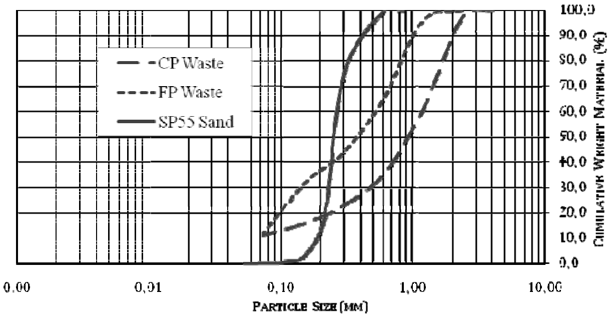

Fig. 2 Particle size distributions of GFRP waste and sand aggregates.

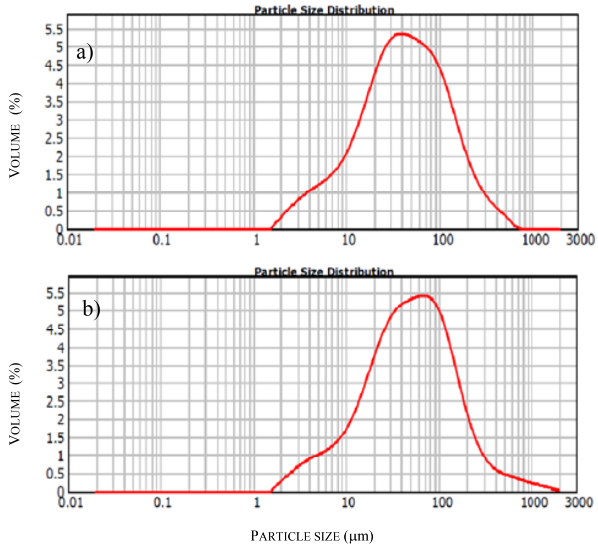

Fig. 3 Laser diffraction measurements of particle size distribution - filler fraction- of a) FP and b) CP waste materials.

C. Mix Design and Casting Process

Four different GFRP waste admixed mortar formulations were analysed, varying the type and content of GFRP powder and fibre mix waste. Experimental trials are presented in Table II. The following notation was adopted: CP or FP accounts for the type of GFRP waste and the sequent number for the weight content of waste admixture. Control or plain polyester polymer mortars were also (P-0) investigated for comparative analysis purposes.

TABLE II
Mix Proportions (w/w) of Raw Materials of Polyester Polymer Mortar Formulations

Experimental Trials	Resin (%)	Foundry Sand (%)	GFRP Pultrusion Waste	
			Fine (%)	Coarse (%)
P-0 (Ref.)	20	80	-	-
FP-4	20	76	4	-
FP-8	20	72	8	-
CP-4	20	76	-	4
CP-8	20	72	-	8

Polymer mortars, with binder formulations and mix proportions specified in Table II, were mixed and casted into standard prismatic moulds (40x40x160 mm^3), as illustrated in Fig. 4, according to RILEM recommendation CPT PC-2:1995 [36].

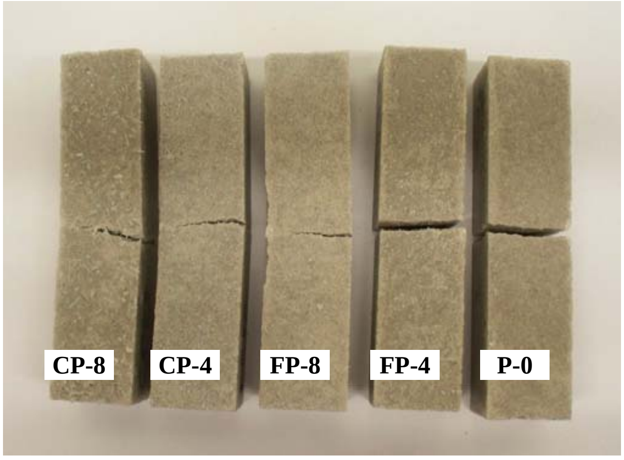

Fig. 4 Polymer mortar specimens (one of each formulation) after being tested in flexural.

Before manufacturing process, sand aggregates and GFRP wastes were previously dried in an oven until constant weigh, in order to prevent eventual inhibition of polymerization process due to moisture presence.

For each formulation, six prismatic specimens were casted. All test specimens were allowed to cure 24 hours at 30ºC / 50% RH, and then post-cured at 80ºC for 3 hours, before being tested in bending and compression at room temperature.

D. Testing Procedures

Prismatic polymer mortar specimens were tested in three-point bending up to failure at the loading rate of 1 mm.min $^{-1}$, with a span length of 100 mm, according to RILEM CPT PCM-8 standard test method [37]. The specifications of this standard, in terms of specimen geometry and span length, are similar to those specified in ASTM C348-08, standard test method for flexural strength of hydraulic cement mortars [38]. Despite the very low value of span to specimen thickness ratio, shear effect is disregarded and it is not considered. Mortar is assumed as an isotropic material and the theory of plane cross-section is used.

One of the two leftover parts, of each broken specimen in bending, were tested afterwards in compression at the loading rate of 1.25 mm.min^{-1}, following the procedure described in UNE 83821:1992 test standard [39]. Both, flexural and compressive testing set-ups, are presented in Fig. 5.

III. RESULTS AND DISCUSSION

Mechanical properties obtained from flexural and compressive tests performed on specimens of all formulations are presented in Table III. Presented values represented average flexural and compressive strengths of six specimens and correspondent standard deviations. Average density of test specimens, with basis on measured weight after curing, is also presented in Table III. In order to assess the effect of GFRP waste admixtures on ductility and stiffness of polymer mortars, typical shapes of obtained load-deflection curves were determined and plotted in Figs. 6 and 7.

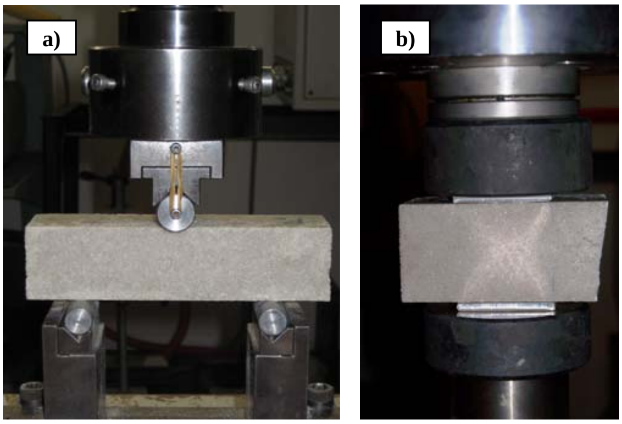

Fig. 5 a) Flexural and b) compressive testing set-ups.

TABLE III
MECHANICAL TEST RESULTS

Test series	Density (g. cm^{-3})	Mechanical Properties	
		Flexural Str. (MPa)	Comp. Str. (MPa)
P-0	1.86	25.17 ± 1.27	76.29 ± 3.26
FP-4	1.83	26.24 ± 1.50	78.05 ± 2.71
FP-8	1.84	27.76 ± 1.61	85.59 ± 2.79
CP-4	1.87	27.53 ± 0.56	83.39 ± 2.60
CP-8	1.87	26.76 ± 1.36	85.70 ± 2.74

A. Effect of GFRP Waste Content

Test results in Table III show that the incorporation of GFRP waste materials on polyester based mortars has an incremental effect on both flexural and compressive strengths of modified mortars, regardless of the GFRP waste type. Apart from flexural load capacity of CPW test series, in which a slight decrease on bending strength occurs for CP-8 formulation, loading capacities of polymer mortars increase with increasing addition of GFRP waste. This effect is more pronounced with regard to compressive behaviour.

Average compressive strength increases of 5.8% and 12,2% corresponding to the addition, respectively, of 4% and 8%,in weight of GFRP waste, were observed with regard to unmodified mortars. The almost linear increase of compressive strength with GFRP waste content, might be attributed to a more continuous particle size distribution of the mix sand/waste particles. The contribution of GFRP waste powder to filler fraction of sand aggregates, leading to an inferior void volume for dry-packed aggregate, has a relevant role in this feature. Aggregate gradation design should aim to produce aggregates mixtures with the maximum bulk density and the minimum voids' content. Generally, this leads to higher strength materials, due to improved aggregate agglomeration.

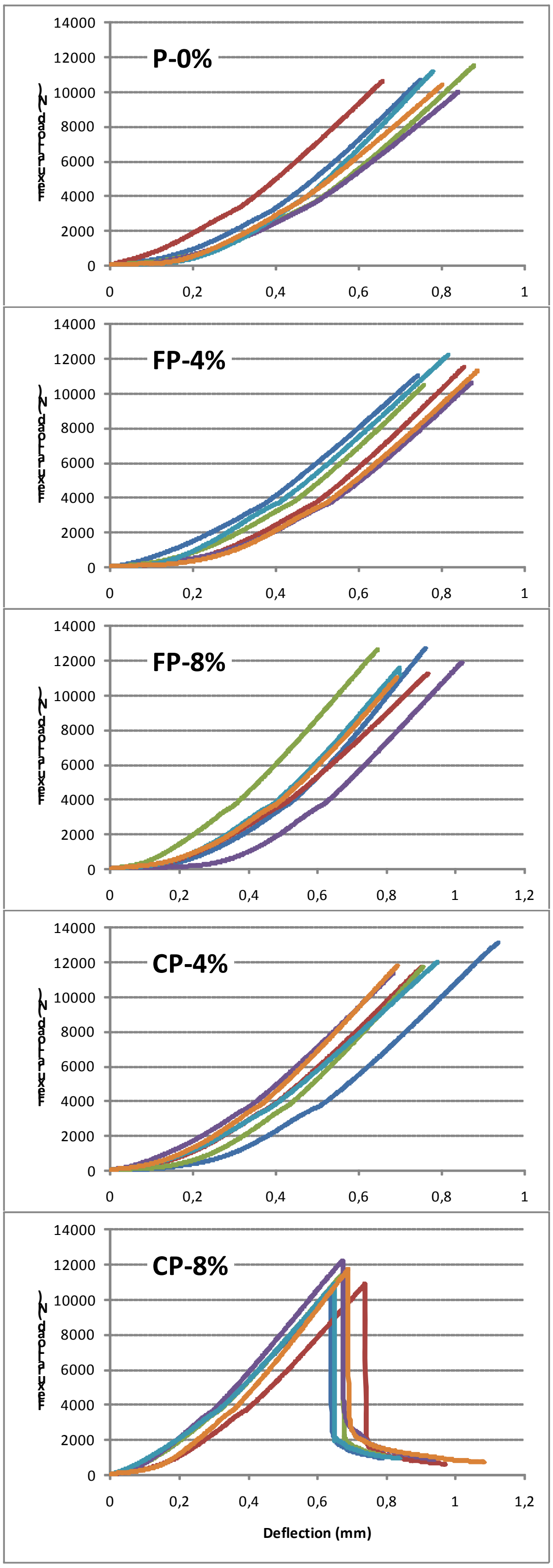

Fig. 6 Flexural load-deflection curves of test specimens.

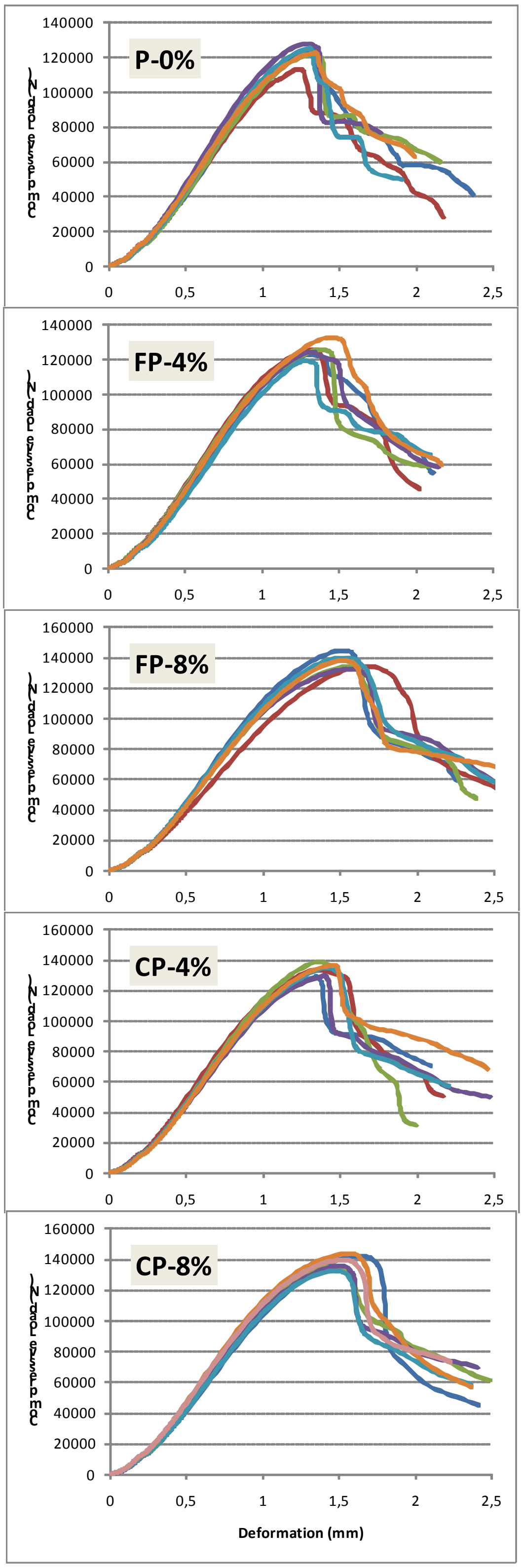

Fig. 7 Compressive load-deflection curves of test specimens.

In flexural, this trend, the linear increase of loading capacity with increase addition of GFRP waste, is not so clear. Average increases on bending capacity of 6.8% and 7.5% were found, respectively, for 4% and 8% in weight of GFRP waste additions. It was expected that fibrous fraction of GFRP waste would have a significant reinforcing effect, leading to an higher improvement on flexural behaviour. Although this expected flexural improvement did actually occur for FP test series, in which progressive increases of 4.2% and 10.3% on bending strength were noticed for respectively FP-4 and FP-8 test formulations; a slight decrease on flexural strength was observed for CP test series, when CP waste content was increased from 4% to 8%. In the mixing and casting process of CPW modified mortar specimens, some tendency for the agglomeration of waste fibres was observed, hindering somehow a perfect homogenization of the mixture. This feature, more notorious as higher the CPW content, lead to a non-homogeneous distribution of GFRP waste, and might be a possible explanation for obtained results. Another contributing factor might be the presence of larger particles on CPW recyclate, which tend to be stress raisers, acting as failure initiation sites. This subject should be clarified in posterior study that will focus on microstructure analysis of mortar specimens.

Either in flexural or in compression, a less brittle failure of GFRP admixed mortars was observed. Improved ductility with GFRP waste content is more pronounced in compression than in flexural, with higher retention of load capacity after peck load.

B. Effect of GFRP Waste Type on Mechanical Behaviour

Apart from flexural test results obtained with CP-8 mortar formulation, polymer mortars modified with CPW present improved mechanical behaviour over FPW admixed mortars, for the same waste content. Higher increases in mechanical strength were observed, either in flexural or compression, for 4% addition of CP waste (9.4% and 9.3% increases in flexural and compression strengths, respectively, against 4.2% and 2.3% obtained for the less coarse waste). 8% addition of CPW also leads to higher compressive strength than the same content of FPW.

As shown in Figs.1 and 3b), CP waste presents a wide range of fibre lengths, varying between 25 mm and few micrometers. (Laser diffraction analysis of particle size distribution of CPW filler fractios detected length fibres between at least 74 μm and 2000 μm). Maximum fibre length of FPW is around 5 mm, thus, CPW has an higher reinforcing effect than FPW. This generally leads to improved mechanical behaviour of host material, providing that a good interface bonding is ensured.

In general terms, taking into account the distinct geometric characteristics of FPW and CPW recyclates, it can be stated that whereas FPW acts more like a filler extension for sand aggregate of modified mortar, leading to a less void-volume of resultant material; CPW acts mainly as reinforcing material, conducting to improved mechanical strength and less brittle behaviour of modified mortar.

IV. CONCLUSIONS

Experiments were performed in order to determine the effect of recycled GFRP pultrusion waste admixtures on mechanical behaviour of polyester based mortars. The influence of two different GFRP waste weight contents (4% and 8%) with two distinct size gradings (CPW and FPW) were investigated.

The key findings of the use of GFRP waste recyclates in polymer mortars testing programme are as follows:

- Compressive strength of GFRP waste admixed polymer mortars increases with increasing additions of waste, and with regard to unmodified mortars;
- Flexural strength of GFRP waste admixed polymer mortars also increases with regard to unmodified mortars;
- Different trends were observed for the effect of FPW and CPW recyclates on flexural strength of resultant mortars: whereas flexural strength of FPW admixed mortar increases almost linearly with increasing contents of waste, the addition of higher content (8%) of CPW leads to a lower increase on flexural strength of modified mortar (with reference to unmodified mortars). Non-homogeneous distribution of waste fibres due to agglomeration and tendency of larger particles to be stress raisers might explained this feature;
- For the same waste content, CPW admixed polymer mortars present in general improved mechanical behaviour over polymer mortars modified with FPW, showing a superior reinforcing effect;
- Both types of GFRP waste improve ductility and lead to a less brittle failure of resultant GFRP waste admixed mortars.

The findings of this study showed a viable technological option for improving the quality of GFRP waste filled polymer mortars and recycling GFRP waste as potential construction materials. However, further studies will be necessary and are foreseen, in order to define both: 1) the optimum GFRP waste content that lead to maximum load capacity without compromising workability of the mixture; and 2) efficient methods to prevent fibres agglomeration during mixing and casting processes in order to promote a uniform fibre distribution.

It is expected that the futures studies will confirm the technical and economic viability for commercial exploitation of GFRP waste incorporation in polymer concrete composites.

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