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Case study

Polymer waste materials as fillers in polymer mortars: experimental and finite elements simulation

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ABSTRACT

Serious environmental problems are due to large amounts of polymer waste, which are mostly thrown into landfills. As we known, polymer composites has been used to produce a variety of products like acid tanks, manholes, drains, highway median barriers, and so forth. One option is to use waste polymers as aggregates in polymer composites. In this work, waste polymers (PET, polycarbonate and automotive tires), partially replaced silica sand in polyester based mortar. Waste particles (0.7–2.36 mm), in concentrations of 1, 2 and 3% by weight, were used. The polymer mortar specimens were subjected to compressive and flexural tests, and the elasticity modulus was calculated. In addition, mechanical values were calculated by Finite Element Method (FEM), and compared with experimental data. Surface morphology and degree of crystallinity of waste particles were analyzed by SEM and XRD techniques, respectively. The results show improvement on the mechanical strength (up to 20%) for polymer mortar with waste PET; but lower mechanical values when adding polycarbonate or tire particles, compared to control mortar. These mechanical results can be related to the crystallinity degree, because PET particles shown higher crystallinity than those for polycarbonate and tire particles. This work is an alternative to reduce environmental contamination through to use waste polymers as fillers in polymer mortars.

1. Introduction

At the present, life without polymer materials is difficult, not only for its usefulness but also economic importance. Despite of its undeniable daily usefulness, polymers become waste and cause environmental problems in water, air and soil, as well as in health. Investigations of waste polymer materials as fillers in composite materials have been carried

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out. Modifications of the mechanical properties, as well as density, water absorption and chemical attack resistance has been obtained [1–3]. For example, high density Polyethylene (HDPE) aggregates were used as partial replacements of natural sand (0, 15, 30, 45 and 60%), to produce lightweight mortars. The results showed increased on the ductility but reduction of the dynamic modulus of elasticity, for mortar with 60% HDPE [4]. In another study, the effects of the sand-to-polyethylene terephthalate (PET) ratio were studied, as well as curing conditions on the physical and mechanical properties of mortar with recycled PET. It was observed that as the sand-to-PET ratio increased, the compressive and flexural strength increased [5]. An overview of published investigations regarding the use of waste plastic in concrete is presented in the reference [6].

Due to the damages caused by composite materials to the environment, novel low-cost and efficient technologies are searched, mainly those involving recycling of waste materials. Recycled materials like non-biodegradable polymers has been used as fillers in composite materials.

Polymer mortar is a composite material which results from polymerization of a monomer/mineral aggregate mixture. In the case of the polymer, the most common are polyester or epoxy resins [7,8]. Some studies are concerning to polymer blends of different waste materials as polycarbonate (PC), polyethylene terephthalate (PET), automotive tires and others. For example, (waste polycarbonate + waste PET) blends, were elaborated and molded by compression and injection, for to obtain granules. The results shown improvements on the mechanical properties, which are attributed to the transesterification process between waste polycarbonate and waste PET, which occurs in the molten state [9]. In other study, mechanical properties of recycled polycarbonate/butylene terephthalate (rPC/PBT) blends were evaluated. The result showed improvement on both tensile strength and tensile modulus [10].

Blends of polypropylene (PP) and recycled PET fibers (PP/rPET), as reinforcing material were elaborated. Mechanical behavior was evaluated by tensile, flexural, impact and fatigue tests, and thermal behavior by HDT (Heat Deflection Temperature). Improvements of the strength and the elasticity modulus, as well as HDT were obtained; while impact strength, strain at break, and fatigue decreased [11].

Waste tires have been used in polymer composites, for example in the (rubber-PET-HDPE) blend. The results showed improvement on the interface properties due to functionalization of PET with polyethylene glycol (PEG), and SDS (sodium dodecyl sulphate). Blends with 45% PET-SDS showed highest values of tensile strength (1.56 N/mm^2), impact strength (43.72 kJ/m^2), and compressive strength (158.78 N/mm^2) [12].

In cement mortars, effects of polymers as coarse aggregates on the fresh and harden properties had been investigated. For example, PAC mortar (PET Aggregate Mortar) had a compressive strength value of 30.3 MPa, when adding 20% of PET and using 0.42 w/c ratio. This value was 9% lower than NAC mortar (Natural Aggregate Mortar), however, PAC mortar has significantly high workability (1.8 cm slump). Therefore, PAC mortar with low w/c ratio and high workability can be used as structural mortar member [13]. In other work, waste rubber tire was used as partial replacement of fine aggregate in mortars with three w/c ratios. Two different rubbers were used: a) rubber ash (RA), and b) rubber ash with rubber fibers (RA + RF). The results show that flexural strength of mortar decreases when increasing RA concentration, whereas same property increases when RA + RF concentration increases. In general terms, the abrasion resistance, carbonation depth, modulus of elasticity and chloride ion penetration are affected by addition of rubber ash and rubber fibers [14].

The Finite Element Method (FEM) is a general numerical method and a versatile tool for modeling and simulation of a wide range of engineering problems. By using FEM, it is possible to obtain approximate solutions in space to initial-value and boundary-value problems, including time-dependent processes.

On the basic premise of FEM, based on a region of the solution, it is possible to model it analytically by replacing it with an array of discrete elements. This allows to reduce an infinite number of unknowns of the problem with a finite number of unknowns. On the other hand, parameter conditions can be controlled by FEM (elastic parameters, viscosity, density, temperature, etc.), of each one of the elements or in groups of them, according to constitutive equations.

Finite elements employ preprocessed mesh generation, which enables the model to fully capture the spatial discontinuities of highly inhomogeneous materials. It also allows complex, nonlinear tensile relationships to be incorporated into the analysis. In addition, FEM provide a fast and accurate prediction of the deformation behavior, that is, stress and strain of structural objects subjected to different loads (static, cyclic and nonlinear), geometries and contact conditions. FEM has been incorporated in some commercial software packages and open source codes (e.g., ABAQUS, ANSYS, Palmyra and OOF), and it is widely used to evaluate the mechanical properties of polymer composites [15,16].

Several investigations have been carried out to simulate composite materials. ANSYS 11 program was used to investigate fracture behavior of continuous double steel-mortar composite beams; such numerical model was in a good agreement when comparing to experimental results [17]. Another finite element work was to evaluate the nonlinear properties of the interface between a hollow core slab and topping concrete. The simulation showed comparable results with the experimental data. Results demonstrated that finite element procedures are adequate for composite materials, with an acceptable accuracy [18].

The aim of this work is to investigate the mechanical properties of polymer mortars elaborated with polyester resin, silica sand and waste particulate polymers (PET, polycarbonate, and automotive tire rubber). Such particles partially replace the silica sand in the mixture, in concentrations of 1, 2 and 3% by weight. In addition, calculated data of mechanical properties by using FEM were obtained, and after these were comparing with those obtained experimental.

2. Materials and methods

2.1. Materials and preparations

Polymer mortars were prepared by mixing silica sand, unsaturated polyester resin and waste polymer (PET, polycarbonate or tire rubber particles). The silica sand was provided by a local company GOSA (Tlalnepantla, Mexico) with grain size of 250 μm (Mesh 60). The resin was pre-accelerated by the manufacturer, and methyl ethyl ketone peroxide (MEKP), as initiator was added for initiating the free-radical polymerization process, by using 1 mL/100 g of resin. The resin was provided by company Grupo Químico Industrial and marketed under the code MR-300/75C, its properties are shown in the Table 1.

Waste PET beverage bottles were collected from landfills, while waste polycarbonate was obtained from carcasses of discarded TVs, and the waste automotive tire rubber from collection centers. Polycarbonate, PET and tire rubber were subjected to a process of grinding, washing and drying. Subsequently, the particles were separated by a sieving process. Images of waste particles are shown in Fig. 1.

PET, polycarbonate and tire rubber particles were used as partial replacement of silica sand in the mortars. Concentrations of 1, 2, and 3% in weight were used, as it is shown in the Table 2.

Polymer mortar specimens were elaborated and compacted in a steel mold (40 × 40 × 160 mm), six specimens for each type of concrete were obtained. The specimens were initially cured at room temperature for 24 h; but according to the literature, after curing process some gel composition is present (sometimes up to 5%), and it is convenient a post-curing process for total polymerization, thus specimens were post-cured at 60 °C for 2 h.

2.2. Test methods

Compressive and flexural tests were conducted using a Controls ^{TM} Universal Testing Machine with a load cell of 30 tons. The compressive testing at a loading rate of 1.25 mm/min was done, and three-point bending testing was at a rate of 1 mm/min.

Morphological analysis of the waste polymer particles was carried out by using a Scanning Electron Microscopy JEOL, model JSM-6510LV in the secondary electron mode at 20 keV.

Table 1
Polyester resin properties.

Properties	Value
Brookfield Viscosity, cPs	100–200
Gel time, min	6–8
Curing time, min	16
Exothermic temperature, °C	145–163
Specific weight, (lb/gal)	9.10–9.30
Stability at 105 °C, h	4

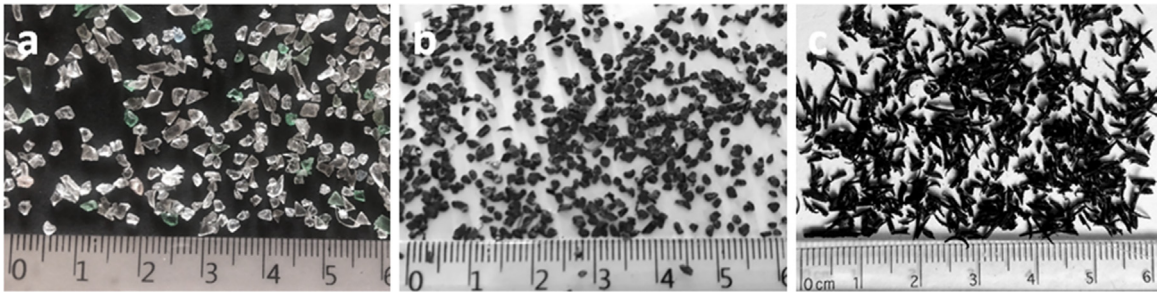

Fig. 1. Waste polymer particles: a) PET, b) polycarbonate and c) Tire rubber.

Table 2
Polymer mortar components.

Lot (code)	Resin (%)	Sand (%)	Waste polymer particles (%)
PC	20	80	0
PC-1	20	79	1
PC-2	20	78	2
PC-3	20	77	3

X-ray diffraction spectra of the waste polymer particles were collected on a BRUKER D8 ADVANCE diffractometer coupled to a copper-anode X-ray tube (Cu-Kα radiation). The test conditions were: tube power 30 kV, Window 5°–65°, and speed of 1°/min.

2.3. Finite element model

The commercial finite element package ANSYS 16.0 was used for simulating polymer mortar. This software has proven its reliability in many benchmark studies, and it was considered suitable for the current task. ANSYS is used for both linear and nonlinear analysis of static and dynamic problems. For solving of nonlinear problems, ANSYS uses the Newton–Raphson method, in which the total load is divided into series of load increments, until the final convergence of solution.

For modelling materials like concrete, ANSYS proposes the use the finite element SOLID186. This element is a higher order 3-D 20-node that exhibits quadratic displacement behavior. The element is defined by 20 nodes having three degrees of freedom per node. The element supports plasticity, hyper-elasticity, creep, stress stiffening, large deflection, and large strain capabilities.

Polymer mortar was modeled on base of a bilinear and multilinear strain hardening yield stress-strain curve. Testing data were used for calibration of the FE model and validation of the adopted parameters for all materials.

3. Results and discussion

3.1. Compressive strength

Fig. 2 shows the compressive strength values of polymer mortar specimens. The highest values were obtained for polymer mortar with PET, followed by those with polycarbonate and finally for mortars with tire rubber particles. In the case of polymer mortar with PET particles, the highest value (78 MPa), was obtained when adding 1 wt% of PET particles, which means a maximum improvement of 20%, respect to polymer mortar without PET particles (called control mortar). Such increment is attributed to the compatibility between polyester resin and waste PET.

Different behavior was observed for mortar with polycarbonate, whose compressive strength values are similar to control mortar, which can be attributed to the similar mechanical performance of both materials, polycarbonate and silica sand. According to the literature, polycarbonate has lower compressive strength and modulus of elasticity than that for polyester resin.

In the case of polymer mortar with tire rubber, a well-defined behavior is obtained: compressive strength values decrease when increasing tire rubber concentration. Such behavior is attributed to the tire rubber properties, in particular to the cross-linked structure, generated during the vulcanization process; such structure restricts the movement of rubber molecular chains. Thus, limited interaction forces between tire rubber particles and polyester resin matrix are present, resulting in a decrease of the polymer composite performance. It is well known that toughness increase and the elasticity modulus decrease, when a rubber material is added to a thermoset resin [19].

Fig. 3 shows the XRD diffraction pattern of PET, polycarbonate and tire rubber particles. For PET particles, the main diffraction peak is located at 2θ = 26° corresponding to the lattice plane (100) [20]. For polycarbonate particles, the pattern shows a main peak at 2θ = 19°, while the peaks at 2θ = 28° y 37° can be attributed to impurities of the sample [21,22]. For tire rubber particles, peaks are detected at approximately 2θ = 20° attributed to the Styrene Butadiene Rubber (SBR) present in the tires [23,24]. Peaks located at 32°, 34° and 37° can be attributed to impurities.

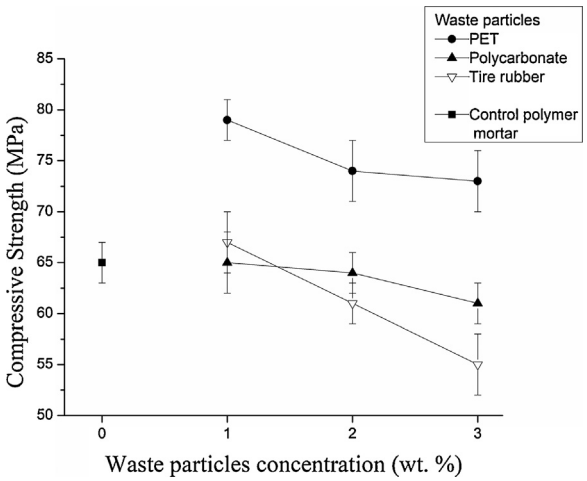

Fig. 2. Compressive strength of polymer mortar with waste particles.

The crystallinity index, Ic, was calculated by using the equation:

$$Ic = 100x\frac{l_a - l_{am}}{l_a} \quad (1)$$

where *la* is the maximum intensity of the main peak, and l_{am} is the intensity attributed to amorphous phase. The main peak was located at 2θ = 26°, 19° and 20° for PET, polycarbonate and tire rubber, respectively; while the amorphous peak at 10.6°, 10.8° and 10.9°, respectively. The crystallinity index was: a) 79% for PET, b) 60% for polycarbonate, and c) 52% for tire rubber particles.

It is well known that response of materials to applied forces depends on: a) structural arrangement of atoms or molecules, and b) type and number of imperfections in the material, which is related to crystallinity property. Thus, after loading a deformation is originated, and can be related with the crystallinity of the material, where higher crystallinity means more stiffness. Thus, compressive strength values of polymer mortars can be related to the crystallinity of waste polymer particles.

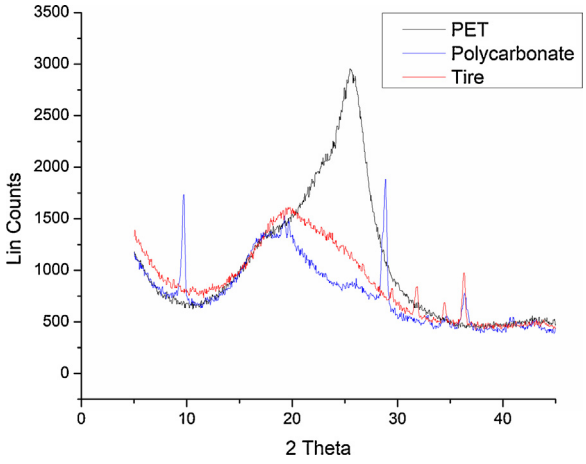

Fig. 3. X-ray diffraction of waste polymers.

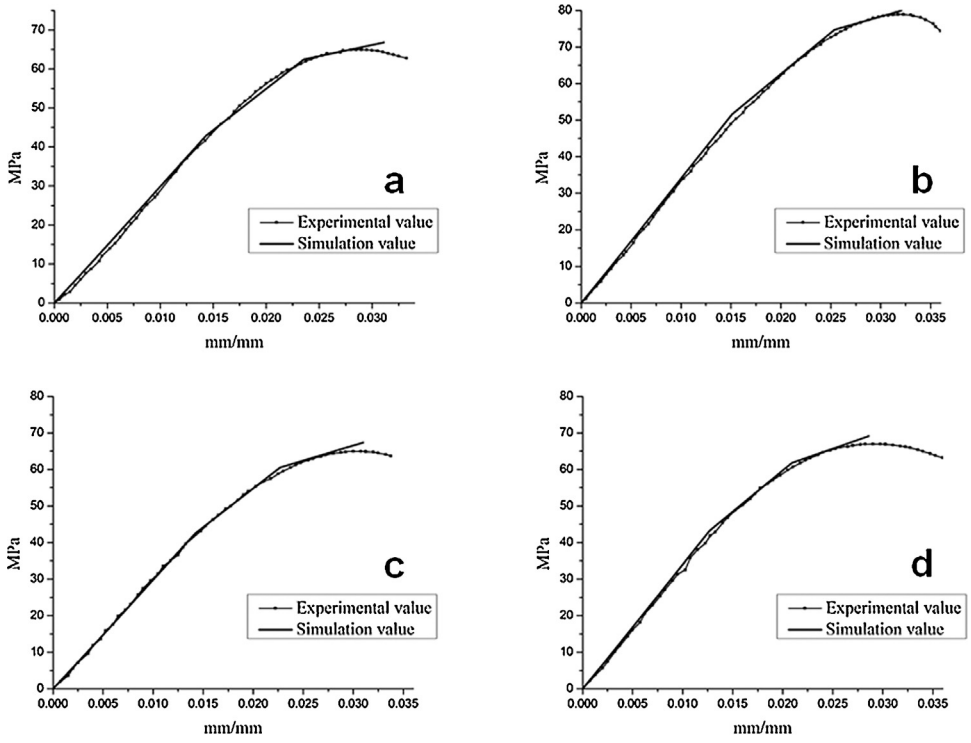

Fig. 4. Stress-strain curves of simulation and experimental results of compression tests for polymer mortar: a) without particles, and with waste b) PET particles, c) polycarbonate particles and d) tire rubber particles.

The stress-strain curves obtained from compression tests and simulated by Finite Element Method (FEM) using ANSYS program, are shown in Fig. 4. The polymer mortar with 1% of waste particles was taken as example. The simulation results showed very good agreement with the experimental data; as it is reported in Fig. 4 and in the Table 3.

According to the Table 3, percentages of difference for compressive strength values are from 1.3 to 1.6, while for young modulus are higher (0.3 to 4.6), and for strain from 7.4 to 12.0. These percentages show that the Finite Element Model and ANSYS program can accurately predict the polymer mortar behavior.

3.2. Compressive strain at ultimate strength

The compressive strain at ultimate strength values are shown in Fig. 5. The deformation of the control mortar is 0.029 mm/mm, this value increases considerably when adding waste PET (up to 20%), or polycarbonate particles (up to 11%). Conversely, diminution of the deformation values were obtained when adding tire rubber particles, up to 20% lower.

Table 3
Comparison of experimental and calculated results for compressive tests.

Polymer mortar	Compressive Strength (MPa)		Compressive modulus of elasticity (GPa)		Strain at ultimate strength (mm/mm)	
	Experimental	Calculated	Experimental	Calculated	Experimental	Calculated
Control	65.0	66.8	2.9	2.8	0.029	0.031
PET	78.8	80.0	3.2	3.2	0.031	0.032
Polycarbonate	64.8	67.3	2.8	2.8	0.029	0.030
Tire rubber	66.9	69.1	3.2	3.2	0.028	0.028

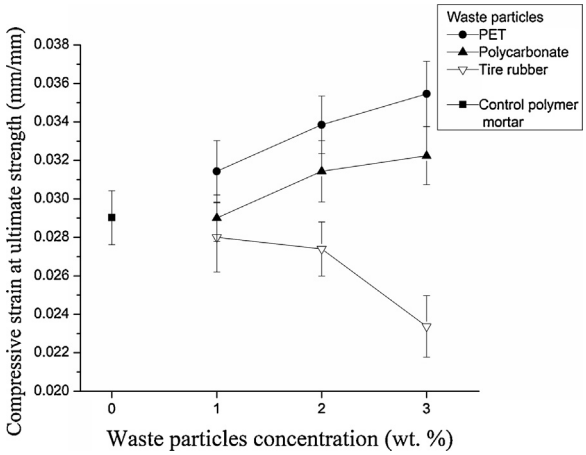

Fig. 5. Compressive strain at ultimate strength of polymer mortar with waste particles.

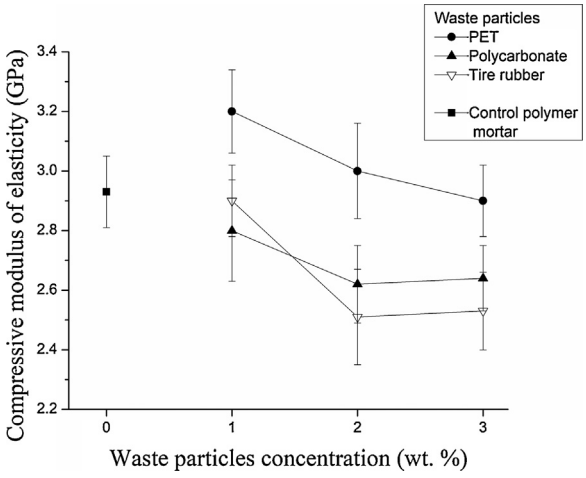

Fig. 6. Compressive modulus of elasticity of polymer mortar with waste particles.

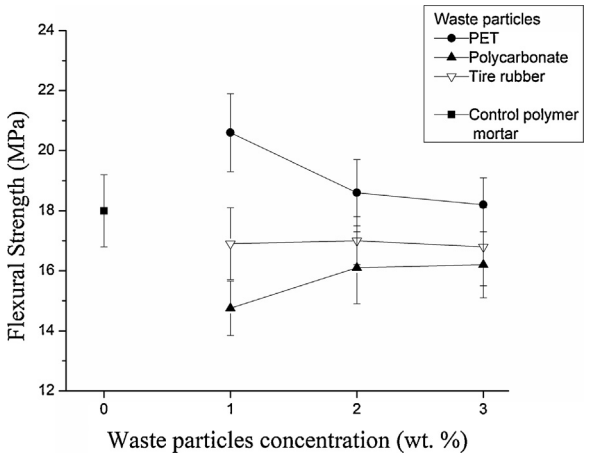

Fig. 7. Flexural strength of polymer mortar with waste particles.

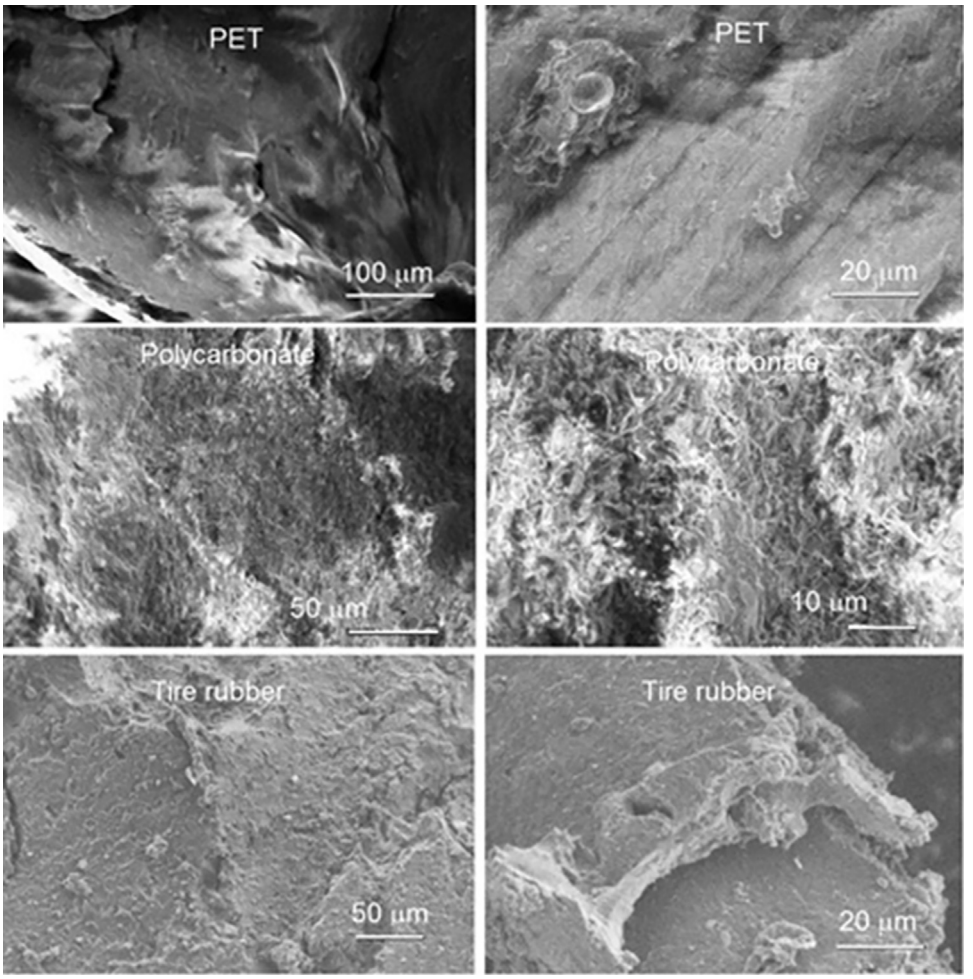

Fig. 8. SEM images of waste polymers.

The deformation increase according PET or polycarbonate increase too; with highest values when adding higher waste particles, is to say, 3 wt. %; thus PET or polycarbonate increase ductility of the polymer mortars. An opposite behavior was found for polymer mortar with tire rubber, where deformation values decrease when increasing concentration of tire rubber. Moreover, increment of the concentration and size of the tire rubber particles allow to increase the voids in the composite and in consequence decrease of the compressive strength and strain.

3.3. Compressive modulus of elasticity

The elasticity modulus values are shown in Fig. 6. These gradually decrease when waste particles are added. Higher values were obtained for polymer mortar with PET particles and lower (up to 14%) for those with tire rubber, respect to control mortar; such behavior is due to lower compressive strength caused by the poor transfer of loads in the polymer mortar.

Diminution of the strength when adding rubber or polycarbonate particles could be also attributed to the weak interfacial bonds between polyester matrix and waste particles. The lower elasticity modulus of waste polymers (PET, polycarbonate and tire rubber) compared to polyester resin value, may cause higher stress concentration and decrease the strength of the composite.

3.4. Flexural strength

The flexural strength values show two different behaviors (Fig. 7): a) increment of values when increasing waste polycarbonate or tire rubber concentration, and b) diminution of values when increasing waste PET concentration. The lowest flexural strength was 20% lower than that for control mortar, corresponding to polymer mortar with waste polycarbonate particles.

Diminution on the flexural strength values can be related to the interaction between waste particles (PET, polycarbonate or tire rubber) and the polymer matrix. Such interactions are depending of the morphology of both resin and waste particles, whose morphologies are shown in Fig. 8. Rough surfaces are observed for PET particles (inclusive for sizes lower than 20 μm). Such roughness facilitates the mechanical anchoring between polymer matrix and PET particles, and in consequence slight increase in flexural strength.

In the case of the waste polycarbonate, different morphological features are observed; which include higher rough, fibrillated elements, detached particles and some cavities. Such characteristics generate lowest flexural strength values, is to

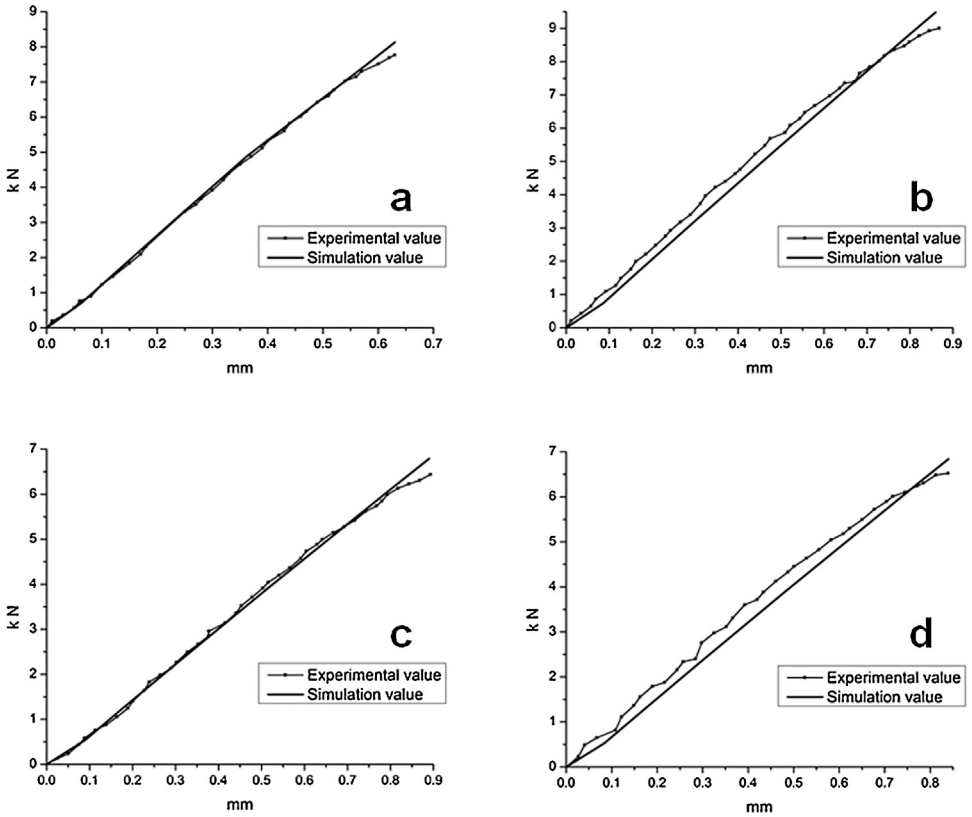

Fig. 9. Experimental and calculated load-displacement curves for polymer mortar: (a) without particles, and with waste particles: (b) PET, (c) polycarbonate, and (d) tire rubber.

Table 4
Comparison between calculated and experimental results for flexural tests.

Polymer mortar	Flexural load (kN)		Displacement at ultimate strength (mm)	
	Experimental	Calculated	Experimental	Calculated
Control	7.76	8.13	0.63	0.62
PET	9.00	9.48	0.86	0.86
Polycarbonate	6.44	6.79	0.89	0.89
Tire rubber	6.52	6.84	0.83	0.84

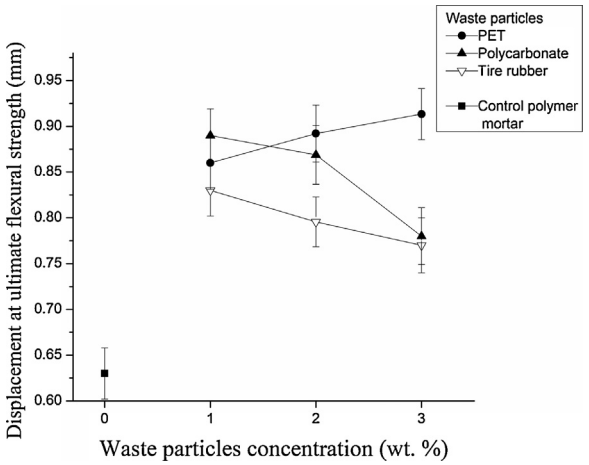

Fig. 10. Displacement at ultimate flexural strength of polymer mortar with waste particles.

say, the particles cannot be full covered by the resin and in consequence some empty spaces are generated, which act as "failures" during the mechanical tests. Finally, for tire rubber particles, more roughness is observed, which allows similar flexural strength values than that for control mortar.

Fig. 9 shows the load-displacement curves of flexural tests of polymer mortar specimens, for both experimental and calculated (by ANSYS program). For control mortar, experimental flexural load was 7.76 and calculated 8.13, which means a 4.7% of difference. In the case of mortars with waste particles (PET, polycarbonate and tire rubber), percentages of difference for failure load were from 4.9 to 5.4. Thus data calculated by Finite Element are in good agreement with those obtained of the experimental results (Table 4). Displacement at ultimate strength was 0.63 for control mortar, and 0.62 for that calculated by ANSYS program, thus a minimal difference was found. Mortars with PET or polycarbonate have the same experimental and calculated values; and for mortar with waste tire rubber a minimal difference was obtained. Thus, ANSYS program is a very adequate tool for to obtain calculated data.

3.5. Displacement at ultimate flexural strength

The displacement at ultimate flexural strength values are show in Fig. 10. For polymer mortar with PET particles, the values gradually increase for higher particles concentrations, but not for polymer mortars with polycarbonate or tire rubber, where the values decrease. The highest value was obtained for polymer mortar with 3 wt. % of PET particles, which means an improvement of 45% respect to control mortar. PET particles generate more ductile polymer mortars, when they are subjected to flexural loads.

4. Conclusions

Effects of polymer waste materials (PET, polycarbonate and tire rubber) on the compressive and flexural properties of polymer mortars were studied. Mechanical features where compared to those for polymer mortar without particles, namely control mortar, and related to the morphology and crystallinity of polymer waste particles. The results showed highest compressive and flexural strength for polymer mortars with 1 wt. % of PET particles. By adding waste PET particles it is possible to obtain the highest elasticity modulus and deformation when they are submitted to compressive load; such results are due to the high crystallinity degree and roughness of PET particles. Conversely, mortars with polycarbonate or tire rubber particles had lower mechanical values (compressive strength, flexural strength and modulus of elasticity) as compared to control mortar. Moreover, regarding the simulation by finite elements, the results showed higher agreement with the

experimental data. The results have been promising, whereby in a second stage waste particles will be modified by chemical treatment or coated with silanes, to improve the interaction with the matrix and thereby increase the mechanical properties.

Conflict of interest

The authors declare that none of them has a direct financial relationship with the commercial trademarks mentioned in this paper that might lead to a conflict of interests for any of the authors.

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