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Recycling polypropylene and polyethylene wastes in production of polyester based polymer mortars

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HIGHLIGHTS

- Polymer mortars with waste polymers were produced.
- Improvements of 82% on the flexural deformation were obtained with waste polypropylene.
- Improvements of 30% on the compressive deformation were obtained with waste polyethylene.
- Improvements of 27% on the compressive strength were obtained with waste polypropylene.
- Structural modifications on waste particles were related to mechanical strength.

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ABSTRACT

Although polymers were originally seen as an option to replace other materials due to their physico-chemical properties and low cost, they have become an environmental problem due to poor final disposal practices. For these reasons, in the presented investigation, impact of waste polypropylene (wPP) from bottle caps and waste polyethylene (wPE) from bottles, on mechanical characteristics of polymer mortars were observed. Mortars were produced with unsaturated polyester resin at 20% ratio and silica sand at 80% ratio. Sand was replaced with the wastes at three different ratios as 1, 2 and 3 wt% and sizes as 0.71, 1.4 and 2.38 mm. Observations exhibited improvement about 27% for compressive strength when adding wPP particles; 30% on the compressive deformation (adding wPE) as well as 82% on the flexural deflection (adding wPP). Nevertheless, flexural strength and the elasticity modulus of polymer mortars decline by depending on the increment in content and sizes of the wastes used. That is highly connected to morphologies observed by SEM of the fractures polymer mortars.

1. Introduction

Currently one of the greatest challenges facing researchers is to strike a balance among the construction industry needs, environmental protection and the human health. Synthetic polymers as polypropylene (PP) and polyethylene (PE) can make possible this balance. Polyethylene has wide acceptance due its high chemical resistance, non-toxicity, excellent electrical properties and weight lightness; while polypropylene has high compressive and tensile strength, excellent dielectric properties and resistance to acids and alkalis. Due to their high production and consumption, full attention is done for their collection, handling and final disposal.

Recycling polymers include mechanical and chemical technologies. In the mechanical, several processes are involved, as melt filtration, separation, froth flotation, magnetic density separation and X-ray detection. The chemical recycling includes chemolysis, pyrolysis, fluid catalytic cracking, hydrogen techniques (hydrocracking and IH^2 process), KDV process and gasification [1].

Several studies have been carried out on the waste or recycled polypropylene (PP) and polyethylene (PE). In the case of polypropylene: I) recycling of polypropylene (rPP) causes embrittlement, similar tensile stress (yield strength and elastic modulus) as those for virgin PP and mixtures of them. However, the impact strength values of rPP are lower than those for virgin PP. Such

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results show that rPP has been evaluated for applications requiring tensile resistance with low impact strength [2]; II) waste polypropylene (wPP) from bottle caps was blended with polyethylene terephthalate (PET) from water bottles, for producing filaments for 3D printing. Addition of the wPP increased the elongation to failure, decreased the tensile strength and the crystallinity, and the glass transition (Tg) was changed to the temperature elevated for mixes respect to neat recycled PET [3]; III) grinded polypropylene waste (from bottle caps) was blended with lignin powder (byproduct from pulp industry) by melt spinning at 170 °C, for producing textile fibers. They had 170–250 μm diameter and improved mechanical properties [4]; IV) recycled polypropylene (from bottle cap cores), have less weight and cheap honeycomb pattern to structural practices. They were blended with aluminum skins and epoxy resin for producing sustainable sandwich panels. The composites had enhanced strength and stiffness and were studied by Analysis of Variance (ANOVA) [5]; V) recycled polypropylene (rPP) from post-consumer containers was blended with elastomers and calcium carbonate as compatibilizers, for producing composites with enhance mechanical properties. As it is known, miscibility and compatibility of polypropylene are important parameters for producing consumer packaging [6].

In the case of Polyethylene, some studies show that: I) recycling of post-consumer HDPE was carried out by cone-plate rheometry technique. During the process, the post-consumer HDPE displayed cross-linking, then 0.2% antioxidant was added for to stabilize the process. Recycling was studied in terms of the molar mass, molar mass distribution and catalytic residues [7]; II) waste polyethylene (wPE) was pyrolyze by using molten $AlCl_3$-NaCl eutectic salt as catalyst that decline first temperature with 200 °C for pyrolysis, and increase the reaction rate. The heavy oil components in liquid materials were decreased and no olefins were produced. Moreover, char taken away from the rest had porous bodies [8]; III) deterioration of LDPE was made by prodegradant (oxo-degradable) additive (1–50 wt%) and exposing to a Xenon arc chamber to arrest generation of hydro peroxides. Processability and mechanical strength of the blends decreased and the matrix properties decrease when the prodegradant content increases in the neat LDPE. The elasticity, plasticity and tensile properties decrease because of breakage and cross-linking of polymer chains [9]; IV) branched PE (LDPE or LLDPE) from an industrial landfill was characterized. The existence of organic like PP and cellulose and inorganic like soil minerals, salts and metal particles pollutants were shown. Then, PE-based recyclates were produced by a solventless ball-milling treatment, and their tensile, impact, morphological, and thermal analyses were assessed. A four times of increment in final extension and 20% improvement for tensile strength were obtained [10].

One of the most successful techniques for polymer reusing is based on to use polymers as fillers in composite materials used in the construction industry. Polypropylene and polyethylene have been used for such purpose. Some examples of such composites with waste or recycled polypropylene, as follows: I) waste polypropylene (wPP) from bottle caps of juices and soft drinks were added (5–20 wt%) to concrete, called ecofriendly. The compressive, split tensile and flexural strength increase with the increase of bottle caps concentration [11]; II) recycled polypropylene fibers (rPP) added to mortars produced higher elasticity modulus, lower tensile strength, less cracks on its surface, as well as a more ductile failure mode [12]; III) waste polypropylene (wPP) carpet fibers were added (up to 1.25 wt%) to fiber reinforced mortar. The fibers were 30 mm length, which increasing the tensile strength but decreasing compressive strength (up to 16% when using gravity method and up to 18% by pumping method) [13]; IV) Addition of polypropylene fibers (1–2 vol%) to recycled aggregate mortar improve the compressive strength, split tensile and shear strength [14]; V) Polypropylene and a plasticizer were added to polymer concrete. The results show highest compressive strength (29 MPa), lowest electrical resistivity (160 O × m), and 0.31 W/mK thermal conductivity [15].

In the case of composites with polyethylene (PE), similar results have been obtained. For example: I) high density polyethylene (HDPE) from post-consumer packaging was added to virgin resin, at 25–100 wt% concentrations, and then polymer blends were made by extrusion and injection molding. The HDPE waste produced an electrical insulation system with low voltage (up to 600 V) when adding 75% of virgin resin [16]; II) waste cross-linked polyethylene (wXLPE) was added to mortars, having three particle sizes:<2 mm (fine), 2–8 mm (middle) and 8–16 mm (-coarse). The results show higher water permeability, moderated reduction of strength and shrinkage in addition to the reduction of unit weight, when increasing the concentration and size of the waste polyethylene (wPE) [17]; III) waste low density polyethylene (wLDPE) (1.5–10 wt%), pine wood wastes, and coupling agent (maleic anhydride, MA) were used for produce composites; which having higher deformations and lower elasticity modulus values, as well as thermal stability (up to 220 °C) [18]; IV) high density polyethylene waste (wHDPE) was added to unfired clay bricks, at 1–20 wt% concentrations. Outcomes exhibit reduction of compressive strength by depending on concentration and size of wHDPE was increasing. However, porosity and capillarity coefficient increase [19], V) high density polyethylene waste (wHDPE) obtained from packages, including bottle and food crates, was mixed (10–80 vol%) with cement for to produce plastic cement. The results show improvement on the ductility and the workability, but the density decrease (15%). However, such results lead to produce lightweight plastic cement [20]; VI) waste polyethylene (wPE) was added to oriented strand board panels (OSB) at 10–50 wt% concentrations. Improvements on the thickness swelling, dimensional stability, screw withdrawal resistance, and the water absorption were observed on the OSB panels. However, for high wPE content the moisture content decrease from 5.79 to 3.34%. Moreover, the elasticity modulus, the strength and rupture modulus decrease when wPE increasing [21].

Perspective of the conducted investigation is to show impacts produced on mechanical characteristics of polymer mortars using unsaturated polyester resin (UPR), when adding waste polypropylene (wPP) obtained from beverage-bottle caps and waste polyethylene (wPE) obtained from bottles. Such effects are studied in terms of the concentration and particle sizes of both waste polymers.

2. Materials and methods

2.1. Materials and production of polymer mortar

Prismatic specimens were produced by blending the unsaturated polyester resin (UPR), silica sand and waste polyethylene (or polypropylene) particles. Control specimen contained polyester resin at 20% ratio and silica sand at 80% ratio, which was replaced at 1–3 wt%, by waste polyethylene (or polypropylene) particles, as it is shown in the Table 1.

Table 1
Mixture ratios.

Specimen	UPR (%)	Silica sand (%)	Waste PE (or PP) (%)
wPC-0	20	80	0
wPC-1	20	79	1
wPC-2	20	78	2
wPC-3	20	77	3

Characteristics of the resin are presented in Table 2. The resin was obtained from Grupo Químico Industrial, a local company located at Toluca México, marketed under the code MR-300/75C.

The silica sand with an average diameter of 150 μm (mesh 100) was provided by GOSA, a local company (Tlalnepantla, Mexico). This was dried at 80 °C by 24 h to removal moistur, due to the high humidity and low temperatures, where the laboratory is located. The waste polypropylene (wPP) was obtained from beverage-bottles caps and waste polyethylene (wPE) from bottles. They were washed and dried at 25 °C by 24 h, and are shown in Fig. 1.

The waste polymer particles were subjected to a sieving process, by using three different sieves: mesh 7 (2.8 mm), mesh 14 (1.4 mm), and mesh 25 (0.71 mm). The particles passing through the mesh 7 were retained on the mesh 14. Such particles (2.8 mm) were named by us as "large particles" (LP). Then, the particles passing through the mesh 14 were retained on the mesh 25, called by us "medium particles" (MP) and, finally, the particles passing through the mesh 25, the "small particles" (SP) were obtained. The particles size distribution for waste polymers are shown in Fig. 2.

The mechanical properties of waste polymer particles are presented in the Table 3, according to the specifications reported by the company Syrus Distribution. The mortar specimens were produced in 40 × 40 × 160 mm prismatic steel molds, as it is shown in Fig. 3. After curing, the mortars were placed at room temperature by 24 h, and then were subjected to post-curing process consisting of heat to 60 °C by 2 h. For each batch six specimens were produced.

Table 2
Polyester resin characteristics.

Property	Value
Viscosity, cPs	100–200
Gel time, min	6–8
Curing time, min	16
Exothermic temperature, °C	145–163
Density, g/cm^3	1.09–1.11
Stability at 105 °C, h	4

2.2. Test methods

Flexural tests were done on the prismatic specimens shown in Fig. 3, at 1 mm/min rate, according to CPT PCM-8 standard, while the compression test were made with the broken pieces obtained after bending test, at 1.25 mm/min rate, following the ASTM C-39 M-01 standard. Both tests were made by using an Universal

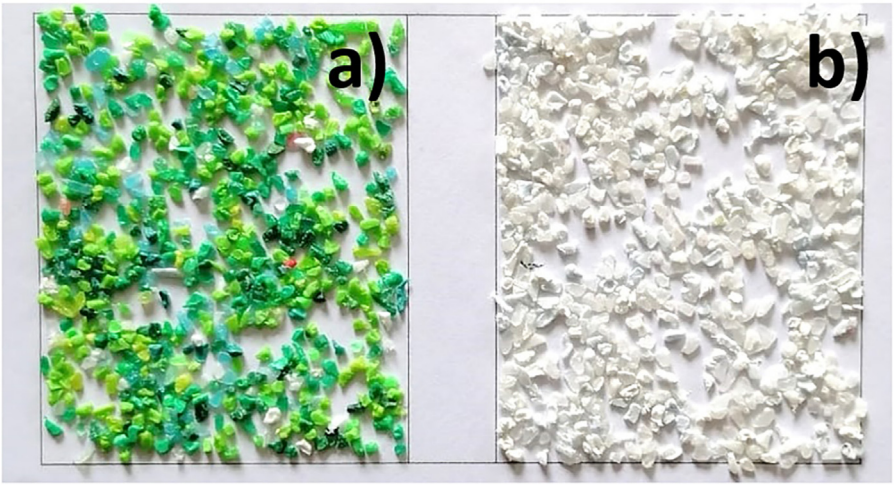

Fig. 1. Waste particles: a) polypropylene (1.4 mm) and b) polyethylene (1.4 mm).

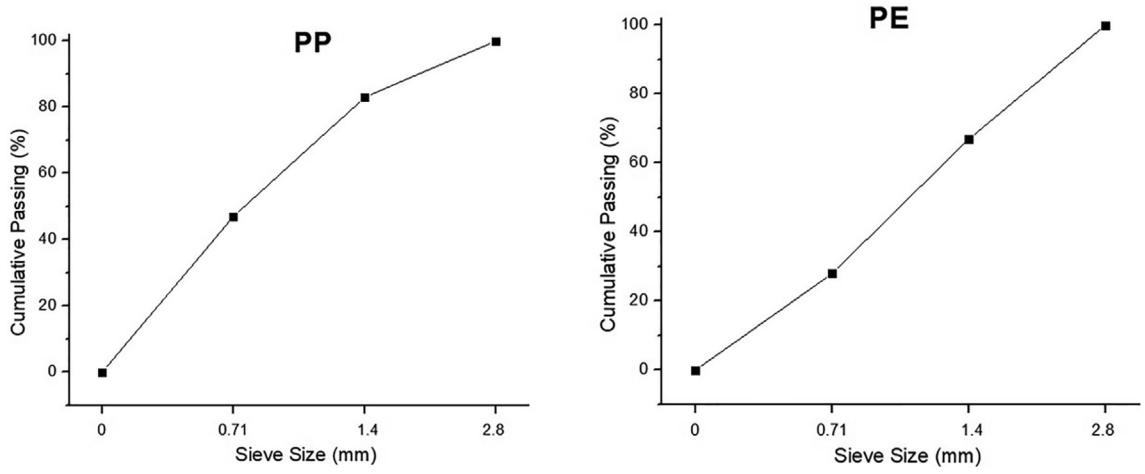

Fig. 2. Particles size distribution of waste PE and PP.

Table 3
Mechanical properties of polyethylene and polypropylene particles.

Property	Polyethylene	Polypropylene
Tensile Strength, MPa	26	38
Tensile Modulus, GPa	1.1	1.9
Flexural Modulus, GPa	1.2	1.7
Elongation (Yield), %	9–18	10–12

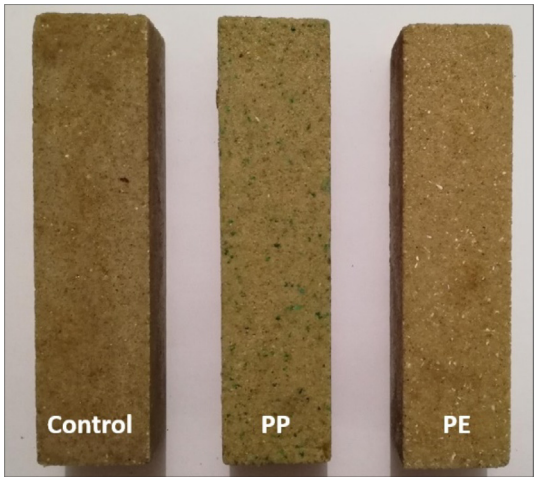

Fig. 3. Control polymer mortars with waste polypropylene (wPP) and polyethylene particles (wPE).

machine model 70-S17C2 (Cernusco, Italy), with a 3,000 kN load cell. Moreover, the surfaces of the fracture zones after compression and flexural tests were studied by Scanning Electron Microscopy (SEM) with a maximum resolution of 5.0 nm, in secondary electron mode at 20 keV.

3. Results and discussion

3.1. Compressive strength

Strengths for polymer mortars with waste particles were taken account in respect to factors: I) particle size and II) concentration. In order to see the comparison among polymer mortar specimens and control specimen which does not contain wastes, in each graph, a dotted line was placed, beginning from control mortar value. Then, the values above or below of this are observed.

Fig. 4 shows compressive strengths for control specimen as well as for specimens with waste polyethylene (or polypropylene) particles. For control specimen, compressive strength was 55 MPa. Following the parameters: I) according to the particle size, values gradually reduce while increasing of grain size, independently of waste polymer (wPP or wPE) addition.

II) In point of particle content, strengths reduce when adding more particles. Specimens containing 1 wt% of waste have higher strengths when compared to control specimen. However, for 2 and 3 wt%, the values are lower than this. The highest strengths were observed on specimens containing 1 wt% wPP particles of 71 mm size (SP). In the case of wPP, the highest value was 70 MPa that presents 27% increment when compared to control specimen, while for mortars with wPE an improvement of 16% was obtained. Such difference on the improvements can be related with the higher values of tensile strength and tensile modulus by the polypropylene respect to those for polyethylene, reported by manufacturer.

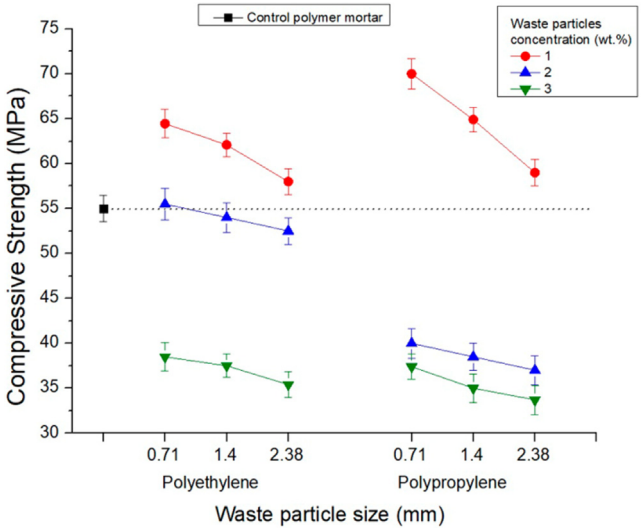

Fig. 4. Compressive strengths of polymer mortars.

3.2. Flexural strength

Flexural strengths for control specimen as well as for specimens with polyethylene (or polypropylene) waste particles are shown in Fig. 5. For control polymer mortars the flexural strength is 18 MPa. Following the parameters: I) according to grain size, flexural strengths decrease when the waste particle size increase, independently of the added waste polymer (wPE or wPP); II) Respect to the particulate concentration, strengths reduce when incorporating more waste concentrations.

Effect of both particle size and particulate concentration on the flexural strength is more evident, compared to compressive strength, because with exception of a couple of values, they are less than control. Only the specimen containing 1 wt% of wastes of 0.71 mm (SP) presents slightly higher strengths when compared to control specimen.

As it is known, flexural test is different to the compressive test; in the case of flexural, the applied forces are made in one direction on three different points. The flexural values decrease due to the poor adhesion between mortar matrix and the waste polymer par-

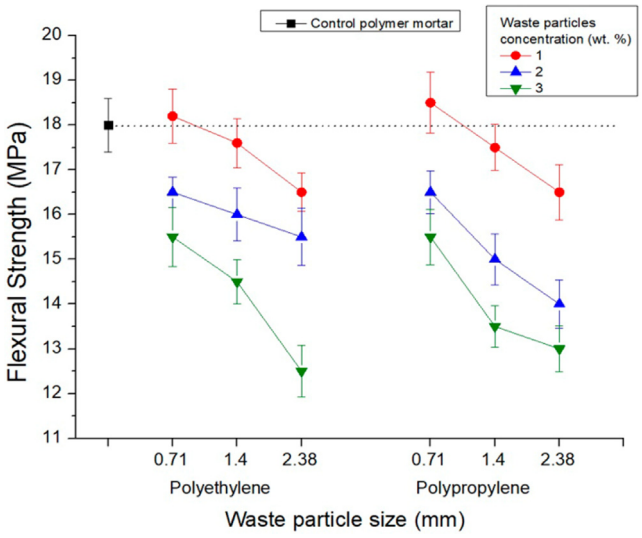

Fig. 5. Flexural strengths of polymer mortars.

ticles, which producing cracks propagation and in consequence fracture.

3.3. Compressive modulus of elasticity

Fig. 6 exhibits compressive elasticity modulus for control specimen as well as for specimens containing waste polyethylene (or polypropylene) particles. For control polymer mortars the value is 18 GPa. Following the parameters: I) according to the particle size, the elasticity modulus decrease when the waste particle size increase (for both kind of waste particles); similar behavior was obtained for compressive and flexural strength. II) In point of particulate concentration, modulus reduces when adding more concentration of waste particles. As mentioned previously increment of both particle size and particulate concentration are detrimental for the compressive and flexural strength. However, such increments produce a more ductile mortar, corroborated by the elasticity modulus measurements.

However, specimens containing 1 wt% of waste PE particles, with 0.71 mm (SP), and 1.4 mm (MP) sizes presented higher compressive elasticity modulus when compared to control specimen. Notorious are low strength specimens containing 2 and 3 wt% of wPP particles, especially those mortars with 3 wt% of wPE particles, which are up to 35% less that value for control specimen. Such specimens are more ductile.

3.4. Ultimate compressive strain

Compressive strain values for control specimen as well as for polymer mortar specimens with waste polyethylene (or polypropylene) particles are shown in Fig. 7. For control polymer mortar compressive strain is 0.036 mm/mm, which is lower than all values obtained for polymer mortars with waste particles. Moreover, up to 30% improvement on strain values were observed when adding wPE, and to 25% when adding wPP.

According to the parameters: I) in the case of the particle size, the polymer mortars with wPE show highest values when adding 1.4 mm (MP) particles, but for higher size the strains decrease. While, for mortars with wPP the strains increase when particle sizes increase. Differences on both types of mortars can be related with the fact that the PP particles have lower tensile strength than those for PE, according to the data reported by the manufacturer; II) In the case of particulate concentration, the deformation gradually decrease when content of waste particles increase.

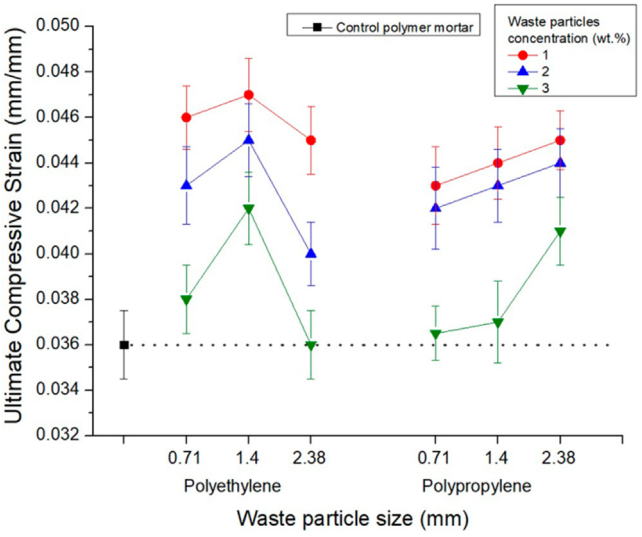

Fig. 7. Compressive strains of polymer mortars.

3.5. Mid-span deflection

Mid-span deflection for control mortar as well as for polymer mortar specimens with polyethylene (or polypropylene) waste particles are shown in Fig. 8. For control polymer mortar the deflection was 0.63 mm, which is lower than all values for polymer mortars with waste particles. Moreover, deformation values are improved up to 66% when adding 3% wPE with 2.38 size, and 82% with addition of 3% wPP with 2.38 mm size.

According to the parameters: I) in the case of the particle size, the displacement increase when particle size increases. II) For particulate content, the values rise when adding more content of waste particles. In summary, high particle size and more particulate concentration produce more deformation in flexion.

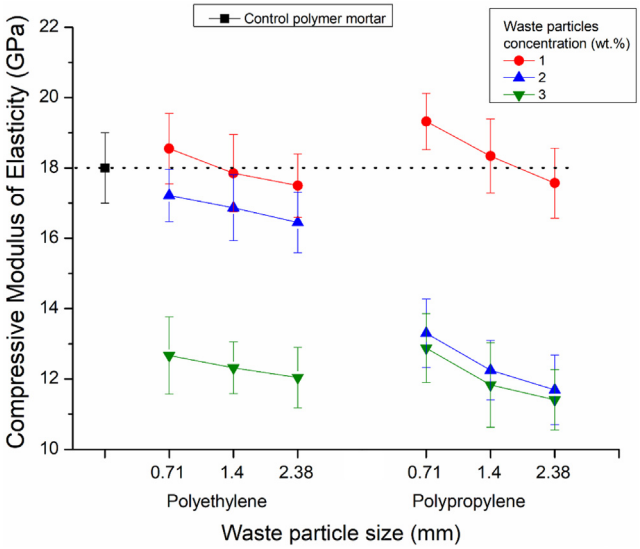

Fig. 6. Compressive modulus of elasticity of polymer mortars.

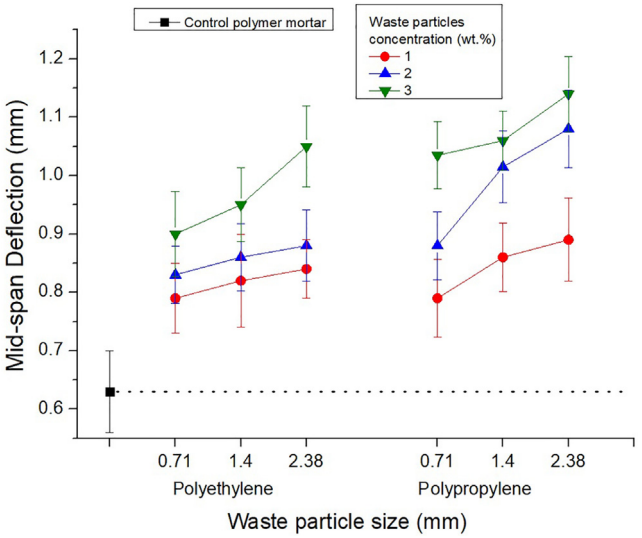

Fig. 8. Mid-span deflection of polymer mortars.

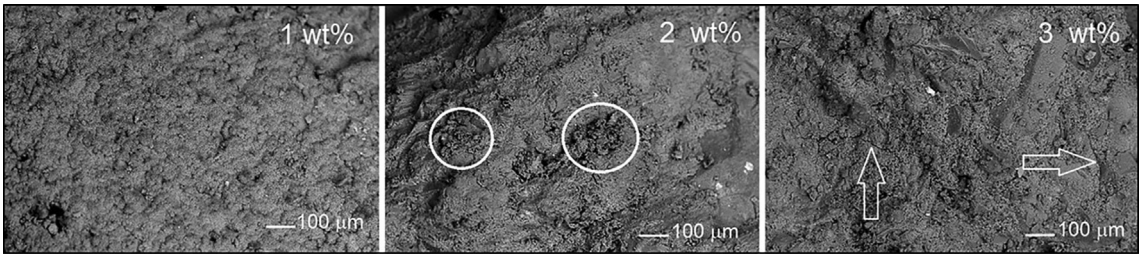

Fig. 9. SEM images of fracture polymer mortars with 1, 2 and 3 wt% of 2.8 mm waste polyethylene particles.

Table 4
Mechanical characteristics of polymer mortars with waste PE.

	Control	1 wt% PE			2 wt%PE			3 wt%PE		
		S	M	L	S	M	L	S	M	L
Compressive Strength (MPa)	55	64.4	62.1	58	55.5	54	52.5	38.5	37.5	35.4
Flexural Strength (MPa)	18	18.2	17.6	16.5	16.5	16	15.5	15.5	14.5	12.5
Compressive modulus of elasticity (GPa)	18	18.5	17.8	17.5	17.2	16.8	16.4	12.6	12.3	12.0
Ultimate compressive strain (mm/mm)	0.036	0.004	0.004	0.045	0.043	0.045	0.040	0.038	0.042	0.036
Mid-span deflection (mm)	0.63	0.79	0.82	0.84	0.83	0.86	0.88	0.9	0.95	1.05

Table 5
Mechanical characteristics of polymer mortars with waste PP.

	Control	1 wt% PP			2 wt% PP			3 wt% PP		
		S	M	L	S	M	L	S	M	L
Compressive Strength (MPa)	55	70	64.9	59	40	38.5	37	37.4	35	33.7
Flexural Strength (MPa)	18	18.5	17.5	16.5	16.5	15	14	15.5	13.5	13
Compressive modulus of elasticity (GPa)	18	19.3	18.3	17.5	13.3	12.2	11.6	12.8	11.8	11.4
Ultimate compressive strain (mm/mm)	0.036	0.043	0.044	0.045	0.042	0.043	0.044	0.036	0.037	0.041
Mid-span deflection (mm)	0.63	0.79	0.86	0.89	0.88	1.01	1.08	1.03	1.06	1.14

3.6. Morphology of the fractured surfaces after the mechanical tests

Improvements on both compressive and flexural deformation of specimens can be related with the fractured surfaces after testing. For example, specimens containing 1 wt% waste polyethylene particles of 2.8 mm, show a homogeneous surface (Fig. 9); which is modified by adding 2 wt% particles, now a rougher surface is observed with particulate agglomerations (indicated by circles). For specimens containing 3 wt%, detached particles and some cracks are obtained (indicated by arrows). High concentration of waste particles produces more stress transfers between waste polyethylene particles and the mortar components; such behavior is related to gradual deterioration, shown by the presence of detached particulate and cracks on the fracture zones.

3.7. Summary of the mechanical properties

The Tables 4 and 5 provide a summary of the mechanical properties of both type of polymers mortars. The polymer mortars with the highest values are those with 1% of wPP with 0.71 mm.

4. Conclusions

The impacts of waste particles (polyethylene and polypropylene) on compression and bending characteristics of polymer mortars were studied. The highest compressive strength values were obtained for specimens with 1% particles of 0.71 mm (for both wPP or wPE), which were up to 27% higher than that for control specimens. However, compressive strength values decrease gradually with increasing of both size and concentration of the waste particles. Similar behaviors were obtained for flexural strength and modulus of elasticity. In the case of ultimate compressive strain, two different behaviors were obtained: a) the values increase (up to 30%), when increasing the particle size; but decrease gradually with increase of the particle concentrations. Such behaviors can be related with the detached particles and cracks observed on the fractured surfaces after mechanical tests. The novelty of this work is to use a minimal waste particle concentration (1 wt%) and small particle size (0.71 mm) for to obtain improvements up to 27% on compressive strength or 30% on the deformation, without the need of physical or chemical treatments on the waste polymers; in addition to contribute to recycling of them.

CRediT authorship contribution statement

Miguel Martínez-López: Conceptualization, Methodology, Investigation, Writing - original draft, Writing - review & editing. **Gonzalo Martínez-Barrera:** Methodology, Validation, Investigation. **René Salgado-Delgado:** Formal analysis. **Osman Gencel:** Investigation, Supervision, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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