# RECYCLING OF LOW-DENSITY POLYETHYLENE WASTE TO PRODUCE ECO-FRIENDLY GYPSUM COMPOSITES

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## ABSTRACT
Over 367 million metric tons of plastic were produced worldwide in 2020. Within the most consumed standard plastics, low-density polyethylene (LDPE) is the second resin most demanded in Europe. Far from reducing its consumption, this continues to grow despite environmental impact. Due to the low recycling rate of LDPE waste (~35%), coming mainly from post-consumer packaging and lightweight bags, most parts of this waste end up in the environment. As an alternative to restrained governmental steps, i.e. extra taxes to reduce single-use plastic consumption, new solutions for the recycling and reuse of plastic waste are being developing from different economic sectors. Because of the worldwide significance of the construction sector, which has a large consumption of natural resources and production of waste, this sector plays a critical role within the new models of circular economy. In light of this, and committed to the sustainable development goals adopted by the United Nations, the present work intends to determine the feasibility of using LDPE waste sourced from plastic bags to produce eco-friendly gypsum-based composites. Four replacement levels of plastic waste were chosen for analysis – 0.25%, 0.5%, 1%, 5% by weight of gypsum. The experimental campaign consisted of analysing the resulting compounds based on density, mechanical behaviour (flexural and compressive strength), as well as evaluating their water absorption by capillary action and adsorption by exposing them to continuous moisture. Comparing this to the control material, lighter composites with a decrease of mechanical strength were obtained. However, all the values of flexural and compressive strength exceeded the minimums established by standard. Also, the LDPE-containing composites presented lower water absorption and adsorption capacity (up to ~50% when compared to the control material). So it could be inferred that the waste material can be considered as a viable partial substitute to the current commercial gypsum for the manufacturing of gypsum products, i.e. plasterboards.
*Keywords: LDPE waste, gypsum plaster, mechanical properties, water-resistance properties, eco-friendly material.*

## 1 INTRODUCTION

Plastic waste management stands as one of the main worldwide concerns to minimise the impact of human activities on the global environment. Over 55 million metric tons of plastic were produced in Europe in 2020. However, the amount of plastic post-consumer waste collected was lower than 50% of the total produced, from which 23% was still sent to landfill [1]. Within the most consumed standard plastics, low-density polyethylene (LDPE) is the second resin more demanded in Europe [1]. The lightness, durability, low thermal conductivity, impermeability and low cost are some of the most notable properties that make that far from reducing the consumption of LDPE, this continues to grow despite environmental impact [2].

Due to the low recycling rate of LDPE waste (~35%), coming mainly from post-consumer packaging and lightweight bags (single-use products), most part of this waste end up in the environment. Recently research showed that plastic litter exposure to external mechanical forces, as well as solar radiation may result in the transformation of this waste to microplastics [3]. Thus, because of the high resistance of LDPE to natural degradation,

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considerable damage toward the terrestrial and marine ecosystems is caused [4], [5]. In an attempt to look for effective solutions to reduce environmental pollution derived from single-use plastics, restrained governmental steps, i.e. extra taxes added to the consumption of this type of products, have been stablished [6]. Nevertheless, there is a need to find suitable ways to reuse or recycle plastic, primarily those with a low recycling rate in comparison with their production (i.e. low density polyethylene), from different economic sectors. Because of the worldwide significance of construction sector, which means, great consume of natural resources and production of waste, this sector plays a critical role within the new models of circular economy.

In this line, numerous research on the recycling of plastic waste to produce new construction materials for a variety of sustainable building applications, have been conducted by architectural and civil engineering sectors.

Related to the use of recycled LDPE waste to develop new building materials, interesting studies were published in concrete manufacture field. The influence of shredded LDPE from plastic bags used as partial substitution of fine aggregate on the behaviour of concrete was analysed by Jain et al. [7]. As a result, the workability, density, mechanical strength and modulus of elasticity of concrete samples decreased with increase in the plastic waste content, while penetrability to water rose. Also, Poonyakan et al. [8] found a significant reduction of thermal conductivity of LDPE-containing concrete composites at 30% volume fraction of LDPE content, when compared with the plain concrete.

Similarly, the influence of adding fine LDPE waste in cement composites, as partial replacement of sand, on density, mechanical strength and durability properties of plastic cement mortars was evaluated by several studies [9], [10]. Based on the prediction model for cement composites made with LDPE waste designed by Ohemeng and Ekolu [11], the LDPE-containing mortars' flexural and compressive strength were adversely affected with the increase of percentage of plastic waste. However, the standard strength requirements were satisfied by composites with LDPE replacement levels of up to 60% by volume. Furthermore, the possible application of this new cement-based materials with LDPE addition to manufacture paver bricks was researched by Reddy et al. [12].

On the other hand, encouraging alternative eco-friendly solutions to commercial gypsum conglomerate by proposing new recycling ways for diverse type of plastic residues without efficient management at present, have been published. Lightweight materials with enhanced thermal properties made with gypsum and plastics residues from CDW (EPS and XPS, respectively) were developed by San-Antonio-González et al. [13], [14]. In this line, Pedreño-Rojas et al. [15] verified the feasibility of using PC waste to produce lighter and mechanically improved gypsum composites, by using up to a 60wt% PC waste addition. Moreover, the improved water-resistant properties of gypsum composites with the incorporation of plastic waste from rejected cables was analysed by Vidales-Barriguete et al. [16]. Other interesting works sought to find a solution to LDPE plastic pollution and reduce the amount of raw material needed to make enhanced mechanical and thermal LDPE-laterite-gypsum bricks by using reprocessed LDPE, calcium phosphate (CaP), phosphogypsum residues and biochar was presented by Azeko et al. [17].

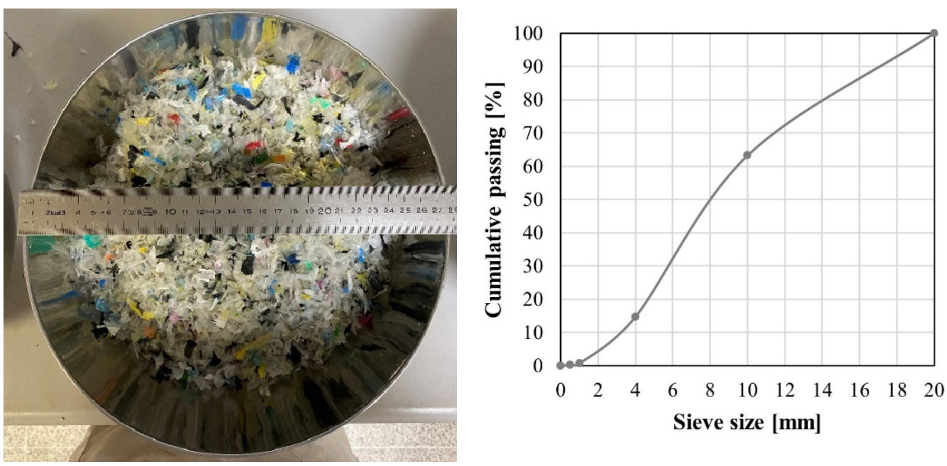
In spite of several studies have evaluated the influence of LDPE waste addition on physico–mechanical and thermal behaviour of concretes and cement-based composites, few research have been developed on the use of LDPE waste as partial replacement to gypsum binder matrixes Therefore, committed to the Sustainable Development Goals adopted by the United Nations, the present work intend to determine the feasibility of using LDPE waste sourced from plastic bags to produce eco-friendly gypsum-based composites. The experimental campaign planed in this work consisted of analysing the resulting compounds based on density, mechanical behaviour (flexural and compressive strength), as well as, evaluating their water absorption by capillary action and adsorption by exposing them to continuous moisture.

## 2 MATERIALS AND METHODS

### 2.1 Materials

The gypsum composites developed in this work were prepared by using the following materials:

- Controlled setting gypsum for construction (B1), according to the standard EN 13279-1 [18].
- Regular tap water in accordance with Council Directive 98/83/EC.
- Low density polyethylene (LDPE) waste aggregate (Fig. 1(a)), obtained from discarded lightweight plastic bags. This residue was collected, processed, and provided by “Reciclados La Red” recycled plant from Seville (Spain). The recycled plastics bags were crushed by using the recycling company’s industrial equipment with an output of particles measuring up to ~20 mm (particle size distribution of the ground LDPE waste in Fig. 1(b)).




(a) (b)

Figure 1: LDPE waste used. (a) LDPE waste particles ready to be mixed; and (b) Particle size distribution.

### 2.2 Samples preparation

The guidelines set by standard EN 13279-2 [19] were followed to prepare the samples evaluated at this work. Four plastic waste replacement levels were chosen: 0.25%, 0.5%, 1% and 5% by weight of gypsum (wt%). The maximum percentage of LDPE waste was conditioned by the water/gypsum ratio (W/G) of 0.55 fixed for all the mixtures to guarantee a correct workability of the mixtures without using additives.

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A manually procedure was used to prepare the mixes, according to the proportions of material summarized in Table 1. In the first place, LDPE waste aggregate was dry mixed with gypsum powder to avoid a heterogeneous dispersion of the particles through the gypsum matrix. Subsequently, water was gradually added to the rest of blend components and mixed for 2 minutes until a homogeneous state.

Table 1: Mix proportions for each group of six specimens ($40 \times 40 \times 160$ mm$^3$).

| Mix code | Gypsum (g) | Water (g) | W/G ratio | LDPE waste (g) |
|---|---|---|---|---|
| **G/CM** | 1,800 | 990 | 0.55 | – |
| **G/LDPE/0.25** | 1,796 | 988 | 0.55 | 4.5 |
| **G/LDPE/0.5** | 1,791 | 985 | 0.55 | 9 |
| **G/LDPE/1** | 1,782 | 980 | 0.55 | 18 |
| **G/LDPE/5** | 1,710 | 941 | 0.55 | 90 |

After completing this procedure, the mixtures were poured into prismatic moulds of $40 \times 40 \times 160$ mm$^3$ to produce six specimens by serie. A total of 24 samples were prepared, together with the control specimen (G/CM) without plastic waste addition.

### 2.3 Test methods

Once samples were removed from the moulds, these were cured according to conditions stablished by standard EN 13279-2 [19]. Thus, specimens were stored for seven days in a dry chamber at a temperature of 24°C and a relative humidity of 50 ± 1%. Then, they were placed in an oven at 40 ± 2°C to constant mass and cooled to laboratory temperature in a desiccator. Concluded the curing period, the gypsum composites were subjected to the test methods described below:

- *Dry bulk density*: calculated after measuring the dry weight and volume of the test samples, as per standard EN 13279-2 [19].
- *Flexural strength:* based on the procedure explained in EN 13279-2 [19], six samples for mixture were subjected to three-point bending test by applying a progressive centred load with a speed of 10 N/s, until reach the breaking point.
- *Compressive strength*: the 12 broken test sections obtained from the previous flexural strength test were used to carry out the compressive strength test, as per standard EN 13279-2 [19]. A load was applied in the central plane of the sample with a speed of 20N/s, until specimen failure.
  The automatic multi-test press MCO-30 equipment, with a 300 KN load capacity was used to develop both test methods.
- *Water absorption by capillary action:* as determined by standard RILEM TC 25-PEM [20], three samples per serie were placed in a vertical position inside a container, with a distance 10 ± 1 mm of water from the bottom. During a testing period of 10 minutes, the water level in the specimens' surface was measured every minute.
- *Hygroscopicity range:* to developed this non-standardized test, designed by Del Río-Merino [21] in her thesis, samples were undergone to constant moisture, by placing them in a wet chamber for five days at a temperature of 21 ± 2°C and a RH of 72 ± 5%. Afterwards, the difference of samples weight gain was measured.

## 3 RESULTS AND DISCUSSIONS

The resulting data obtained for the experimental campaign to previously described to characterize the new LDPE-containing composites are summed up in Table 2.

Table 2: Summary table with gypsum composites' density and mechanical strength tests results.

| Specimens | Density(kg/m$^3$) (CoV (%)) | Flexural strength (MPa) (CoV (%)) | Compressive strength (Mpa) (CoV (%)) |
|---|---|---|---|
| **G/CM** | 1,205 (1.44) | 3.39 (7.59) | 7.71 (5.61) |
| **G/LDPE/0.25** | 1,195 (0.66) | 2.35 (7.46) | 5.50 (2.13) |
| **G/LDPE/0.5** | 1,190 (0.65) | 2.30 (5.17) | 4.90 (5.00) |
| **G/LDPE/1** | 1,185 (0.57) | 2.17 (5.75) | 4.59 (3.89) |
| **G/LDPE/5** | 1,183 (1.12) | 2.03 (10.05) | 3.02 (5.71) |

### 3.1 Dry bulk density

Regarding to the results observed in the Fig. 2, density values of gypsum-based composited, in all cases, were reduced as the replacement of LDPE waste increased. Compared to reference sample, the most pronounced fall of density was produced when a percentage of 5wt% LDPE waste content (~2% decrease) was added to the mix. However, it should be noted that the highest variation of density values between the control material and the LDPE-containing samples was corresponded to G/LDPE/1 compound. The partial replacement of gypsum binder matrix by LDPE waste with lower density (2650 kg/m$^3$ vs. 920 kg/m$^3$, respectively) could explained the declining trend observed related to dry bulk density.

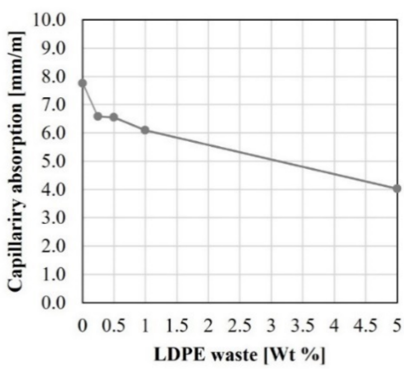
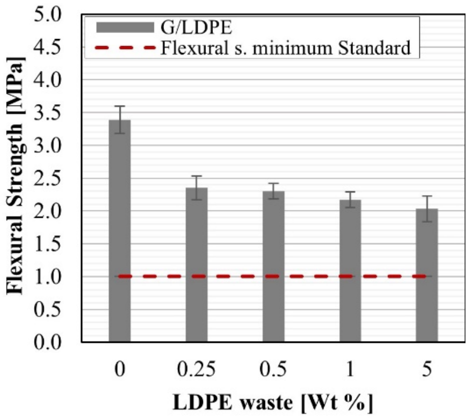


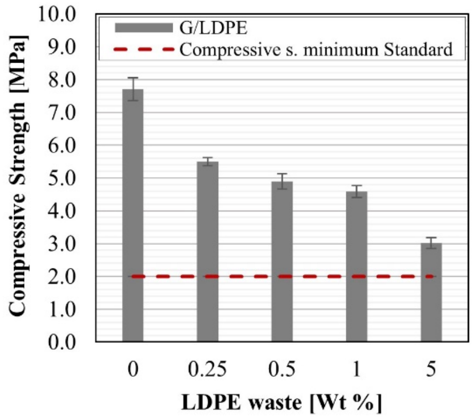

Figure 2: Dry bulk density values (kg/m$^3$) of gypsum/LDPE composites.

### 3.2 Mechanical properties

Expected results were obtained for the flexural and compressive strength tests, as it is shown in Fig. 3. In both cases, the mechanical properties of the new gypsum composites were decreased when the percentage of LDPE addition rose. Concerning to flexural strength, a reduction up to ~40% was reached by composites with a 5wt% LDPE content. At the same way, the higher the percentage of plastic waste aggregate was added to the blends, the lower the compressive strength values were achieved in relation to the reference material. The maximum fall of ~60% was reached by G/LDPE/5 composites. The behaviour presented by new LDPE-containing gypsum composites pointed out that the cohesion at the interfacial transition zone (ITZ) between the LDPE waste particles and the gypsum matrix it is not as strong as that in between the products of hydration and is likely to be a point of failure. Nevertheless, in both flexural and compressive strength tests, all the values were within the minimums 1 MPa and 2 MPa, respectively, stablished by standard EN 13279-2 [19].




(a)




(b)

Figure 3: Flexural and compressive strength values (MPa) of gypsum/LDPE composites.

### 3.3 Water absorption by capillary action

Table 3 showed the data obtained from the water absorption by capillary action test. It can be observed a widespread significant water uptake reduction for the new LDPE-containing composites. The highest decline (up to ~48%) was reached by composites with 5wt% LDPE addition, when compared to the reference gypsum. Concerning to the average water absorption by capillary action (Fig. 4), the lowest value was that of G/LDPE/5, (i.e. 4 mm/min). So, it could be inferred that the increasing presence of LDPE particles imparts an impervious property thereby slowing down the advance of water.

Table 3: Capillary water absorption test results, measured for 10 min.

| | Capillary absorption by minute (mm) | | | | | | | | | | Total capillary absorption (mm) | Δ Capillary absorption compared to ref. (%) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Specimens | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | | |
| **G/CM** | 39 | 6 | 7 | 5 | 6 | 5 | 3 | 7 | 2 | 2 | 85.3 | – |
| **G/LDPE/0.25** | 28 | 9 | 6 | 6 | 5 | 5 | 4 | 4 | 3 | 3 | 72.3 | –15.23 |
| **G/LDPE/0.5** | 31 | 8 | 6 | 5 | 6 | 4 | 2 | 4 | 4 | 2 | 72.0 | –15.63 |
| **G/LDPE/1** | 25 | 8 | 6 | 5 | 6 | 3 | 5 | 4 | 3 | 3 | 67.0 | –21.48 |
| **G/LDPE/5** | 20 | 3 | 4 | 3 | 3 | 3 | 2 | 2 | 2 | 2 | 44.3 | –48.05 |

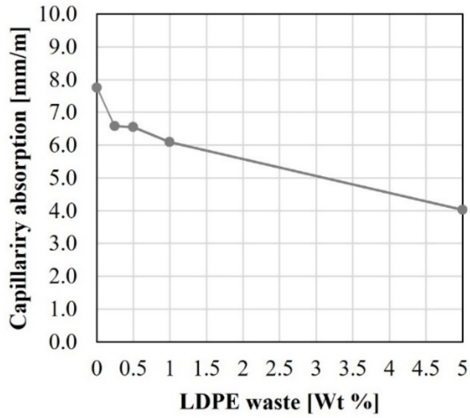



Figure 4: Average water uptake by capillarity (mm/m) of gypsum/LDPE composites.

### 3.4 Hygroscopicity range

After exposing the gypsum composites developed to continuous moderate humidity until 72% RH the results are summarised in Table 4. As it can be observed, the higher the LDPE replacement level was, the lower water adsorption capacity was. Therefore, G/LDPE/5 composite achieved the highest reduction on water adsorption capacity, about 91% lower than the control sample. This trend is similar to that observed in Section 3.3. So, in can be concluded that the growing addition of an impervious material (LDPE waste) lead to an increase of composites' resistance to water movement.

Table 4: Hygroscopicity of the gypsum composites exposed to continuous moderate humidity until 72% RH.

| Specimens | Weight before the test: Dry (g) | Weight after the test: 72% RH (g) | Water adsorption (%) |
|---|---|---|---|
| **G/CM** | 307.61 | 314.18 | +2.14 |
| **G/LDPE/0.25** | 304.72 | 319.27 | +1.99 |
| **G/LDPE/0.5** | 305.87 | 314.35 | +1.78 |
| **G/LDPE/1** | 303.41 | 303.98 | +1.07 |
| **G/LDPE/5** | 302.92 | 303.46 | +0.18 |

## 4 CONCLUSIONS

From above research it can be inferred the feasibility of recycling LDPE waste obtained from lightweight plastic bags as aggregate to partially replace the raw material (i.e. gypsum) in the production of eco-friendly gypsum composites. By considering the results obtained from the characterization tests developed, the following conclusions were drawn:

- No alterations on gypsum mixtures workability were observed after adding of up to 5wt% LDPE waste, which allowed to maintain a 0.55 w/g ratio without adding water reducing admixtures.

- Slight reduction density in dry state of the LDPE-containing gypsums was observed, when compared with the control material (~2% decrease with 5% LDPE waste content).
- Mechanical strength properties showed a widespread decreased with the increasing incorporation of LDPE waste to the gypsum composites. Compared to the control material, the new gypsum-based composites presented a progressive decline trend in flexural and compressive, which is greater in mixtures with a higher level of LDPE waste content (up to ~40% and ~60% decrease, respectively, corresponding to G/LPDE/5 compounds). Nevertheless, all the values were above the minimums indicated in the standard.
- Water absorption and adsorption capacity were reduced after adding impervious LDPE waste to gypsum mixes. This decline observed in both properties, was more noticeable with increasing LDPE waste content (~48% and ~91% reduction, receptively when a proportion of 5wt% LDPE waste was used, compared to control material).

Finally, it can be concluded the viability of producing eco-friendly substitute materials for commercial gypsum used for construction applications included in circular economy strategies (i.e. gypsum drywall), that comply with all the mechanical regulatory requirements and, with improved water-resistance performance, when compared to the plain gypsum without plastic. Likewise, an alternative proposal of waste valorisation to promote the efficient disposition of LDPE waste to facilitate their recycling is presented.

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