### Accepted Manuscript

Design of bespoke lightweight cement mortars containing waste expanded polystyrene by experimental statistical methods

V. Ferrándiz-Mas, L.A. Sarabia, M.C. Ortiz, C.R. Cheeseman, E. García-Alcocel

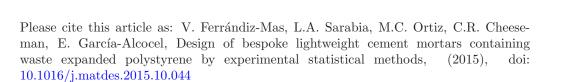
 PII:
 S0264-1275(15)30622-5

 DOI:
 doi: 10.1016/j.matdes.2015.10.044

 Reference:
 JMADE 783

To appear in:

Received date:23 March 2015Revised date:17 July 2015Accepted date:11 October 2015



This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.



### Design of bespoke lightweight cement mortars containing waste

### expanded polystyrene by experimental statistical methods

V. Ferrándiz-Mas<sup>\* a,d,1</sup> L.A. Sarabia<sup>b</sup>, M.C. Ortiz<sup>c</sup>, C.R. Cheeseman<sup>d</sup>, E. García-Alcocel<sup>a</sup> <sup>a</sup>Architectural Construction Department, University of Alicante, Carretera de San Vicente del Raspeig s/n, 03690 San Vicente del Raspeig, Alicante (Spain) <sup>b</sup>Department of Mathematics and Computation, Faculty of Sciences, University of Burgos, Plaza Plaza

Misael Banuelos s/n, 09001 Burgos (Spain)

<sup>c</sup>Department of Chemistry, Faculty of Sciences, University of Burgos, Plaza Plaza Misael Banuelos s/n, 09001 Burgos (Spain)

<sup>d</sup>Department of Civil and Environmental Engineering, Imperial College London, South Kensington Campus, London SW7 2AZ, United Kingdom

#### Abstract

This work assesses the reuse of waste expanded polystyrene (EPS) to obtain lightweight cement mortars. The factors and interactions which affect the properties of these mortars were studied by ad-hoc designs based on the d-optimal criterion. This method allow multiple factors to be modified simultaneously, which reduces the number of experiments compared with classical design. Four factors were studied at several levels: EPS type (two levels), EPS content (two levels), admixtures mix (three levels) and cement type (three levels). Two types of aggregate were also studied. The workability, air content, compressive strength, adhesive strength, bulk density and capillary absorption were experimentally tested. The effect of factors and interactions on the properties was modelled and analysed. The results demonstrate how the factors and synergistic interactions can be manipulated to manufacture lightweight mortars which satisfy the relevant EU standards. These mortars contain up to 60% of waste EPS, low amounts of admixtures and low clinker content CEM III. Sustainable mortars containing silica sand gave flow table spread values between 168 and 180 ± 4 mm, bulk density between 1280 and 1110 ± 100 kg/m<sup>3</sup>, and C<sub>90</sub> between 0.279 and 0.025 ± 0.07 kg/m<sup>2</sup>·min<sup>0.5</sup>, making them suitable for masonry, plastering and rendering applications.

<sup>&</sup>lt;sup>1</sup> 'Main address': Architectural Construction Department , University of Alicante, Carretera de San Vicente del Raspeig s/n, 03690 San Vicente del Raspeig, Alicante (Spain)

<sup>&#</sup>x27;Present address': Department of Civil and Environmental Engineering, Imperial College London, South Kensington Campus, London SW7 2AZ, United Kingdom

<sup>\*</sup> Corresponding author. Tel.: +44 207 594 6018

E-mail address: v.ferrandiz12@imperial.ac.uk (V.Ferrandiz-Mas)

**Keywords:** lightweight mortar; recycling; expanded polystyrene; fitted design; statistical methods

#### 1. Introduction

Expanded polystyrene (EPS) is a low-density, inert, hydrocarbon thermoplastic that is stable in the presence of most chemicals with the exception of concentrated acids, organic solvents and saturated aliphatic compounds [1]. It is commonly used in a variety of applications because of its low density, high thermal insulation, moisture resistance, durability, acoustic absorption and low thermal conductivity [2]. The amount of waste EPS is increasing due to increasing use in thermal and acoustic insulation, packaging, and reusing and storing food. Therefore, over 30 countries have signed an international agreement to maximize reuse and recycling of EPS [3].

EPS has been recently used as foam core in lightweight structural insulated panels used to protect from the impact of windborne debris [2, 4] or to design thermally insulating composites made with foamed cement pastes [5]. There are also several studies which use EPS as a lightweight aggregate in concrete. In particular, the mechanical properties of such concretes have been characterised and the impact of using EPS with different grain sizes, organic admixtures and other additions such as fly ash and silica fume evaluated [6-8]. Other studies have characterised the mechanical and thermal properties of concrete containing EPS [9, 10]. However, only a limited amount of research has investigated commercial EPS [11] or various types of waste EPS in cement mortars [12-15]. The properties of lightweight cement mortars containing EPS, where Portland cement (CEM I) was replaced by cements with lower clinker (CEM II and CEM III), has recently been reported [16]. Due to the high volume of waste EPS it is important to develop new beneficial reuse applications for this material.

The aim of this research was to assess the reuse of waste EPS to obtain sustainable lightweight mortars with durable properties that can be used for masonry, rendering and plastering applications. Ad-hoc or fitted experimental design was used in this study to analyse the impact of design factors and interactions on the studied properties. The type of waste EPS, EPS content, cement type and the mix of admixtures were the chosen factors, while two aggregate types were also studied. These factors and their interactions determine the final properties of these mortars. The workability, air content, compressive strength, adhesive strength, bulk density and capillary absorption were determined.

With classical experimental design it is possible to know the effect of a factor on the studied property by varying one factor and monitoring the relevant property. Nonetheless, variation in the property is in reality related to a combined process involving multiple factors, rather than a single factor. Ad-hoc design allow multiple factors to be modified simultaneously, which reduces the number of experiments compared with classical experimental design. Moreover, it allows both the simultaneous effect of individual factors and synergic effects, resulting from interactions between factors, to be evaluated. The final result on the property is a combination of the two aforementioned effects. They need to be evaluated together and the interactions

cannot be ignored, unless they are not significant for the studied property. These two effects can be positive, increasing the studied property, or negative, decreasing it. Understanding these effects allows manipulation of the levels of the studied factors to manufacture sustainable lightweight mortars with durable properties. While there is limited research using statistical designs to produce mortars or concrete containing waste materials, such as: full factorial designs [10], standard orthogonal arrays [17] or mixture experimental designs [18], as far as the authors are aware no studies have used fitted factorial designs to assess the impact of four factors and the synergic effect of the interactions on the final properties of lightweight cement mortars.

The results demonstrate how the factors and synergistic interactions can be manipulated to manufacture sustainable lightweight mortars, which contain up to 60% of waste EPS, low amounts of admixtures and low clinker content CEM III, that are suitable for masonry, plastering and rendering applications.

#### 2. Materials and methods

#### 2.1 Materials

Three types of cement were used: Portland cement type CEM I 52.5R, Portland cement containing slag type CEM II/A-S 42.5N and cement type III containing ground granulated blast-furnace slag (GGBS), CEM III/A 42.5N (Holcim Morteros S.A). The chemical composition of the three types of cement, showing major components as oxides determined by x-ray fluorescence (XRF) are shown in Table 1. Two types of aggregate with different grain size and mineralogy were used: standard silica sand with bulk density of 1.77 g/cm3, complying with EU standard EN 196-1:2005 [19] and crushed limestone sand from Foncalent quarry (Alicante, Spain) with a bulk density of 1.85 g/cm3. Figure 1 shows the grain size distribution for both types of sand, measured according to EN 1015-1 [20].The main difference between both types of sand is the amount of fine particles, which determine the water demand of the mortar.

Ground and powdered EPS were supplied by "Asociación Nacional del Poliestireno Expandido" (ANAPE, Madrid, Spain) [1]. The differences between the two types of EPS were mainly related to particle size. Both types were obtained by mechanical grinding and sieving recycled EPS. 100% of the ground EPS (EPS gr) particles passed through a 1 mm sieve and the bulk density was 0.013 g/cm3. Powdered EPS particles (EPS pw) passed through a 0.5 mm sieve and had a slightly higher bulk density of 0.022 g/cm3. An air-entraining agent (A, BASF Rheomix 934), a water retaining admixture (R, Hydroxypropyl methylcellulose TER CELL HPMC 15 MS PF), a superplasticizer (S, BASF Rheomix GT 205 MA) and a dispersible polymer (V, VINNAPAS 5028 E) were also used to make mortar samples.

### 2.2 Preparation of mortars

All mortars were prepared using distilled water and a binder/sand weight ratio of 1:3, following the procedures described in EN 196-1 [19]. Due to the different fines content of the aggregates, and in order to get suitable workability, the water/binder ratio was 0.5 for mortars made with standard silica sand, and 0.6 for mortars made with crushed limestone sand. The EPS was dosed as an addition to the total mortar volume, expressed as the apparent volume of sand (v/v%). Admixtures were added to mortars as a percentage of the weight of cement (w/w%).

### 2.3 Experimental design methodology: Ad-hoc designs. D-optimal criterion

Factor selection was based on previous work [12, 13, 16]. Selected factors were: EPS addition content and type of EPS (to study the influence of particle sizes and shape), cement type and the admixture mix (Table 2). Sand type was not included as a factor, although its influence was investigated. This was for practical reasons, because the type of sand available to manufacture cement mortars varies geographically. Two levels were set for factor A, type of EPS: ground EPS (EPS gr) and powdered EPS (EPS pw) (Figure 2). The factor D, cement type, was set at three levels: CEM I 52.5R, CEM II/A-S 42.5N and CEM III/A 42.5N. The flow table method [21] was used to determine the levels of factors B and C, EPS content and admixtures mix, respectively. The EPS content and admixtures were chosen to obtain a suitable workability for masonry mortars. According to the EU standards [22] the flow table spread should be between 165 mm and 185 mm for these types of mortars. This configuration of factors and levels is the experimental domain, D, at which the four factors will vary. It is important to highlight that D is a discrete set formed by isolated points. That is, it is not a hyper rectangle in the four dimensional space, since not all the prior points belong to it. For instance, a point with an intermediate value between CEM I and CEM II is not part of D. This is comparable for all the factors.

To identify curvatures in the response as an effect of changing the level of a given factor, it is necessary that the factor for which a non-linear response is expected has at least three levels. In this way, the possible existence of interactions between factors is analysed. As a consequence the equation which describes the fitted design proposed to relate the experimental response, "y", with the 4 factors is the so called presence-absence model, which is described by the Equation 1:

$$y = \beta_{0} + \beta_{A1}x_{A1} + \beta_{A2}x_{A2} + \beta_{B1}x_{B1} + \beta_{B2}x_{B2} + \beta_{B3}x_{B3} + \beta_{C1}x_{C1} + \beta_{C2}x_{C2} + \beta_{C3}x_{C3} + \beta_{D1}x_{D1} + \beta_{D2}x_{D2} + \beta_{D3}x_{D3} + \beta_{A1B1}x_{A1}x_{B1} + \beta_{A1B2}x_{A1}x_{B2} + \beta_{A1B3}x_{A1}x_{B3} + \beta_{A2B1}x_{A2}x_{B1} + \beta_{A2B2}x_{A2}x_{B2} + \beta_{A2B3}x_{A2}x_{B3} + \dots$$

$$+ \beta_{C1D1}x_{C1}x_{D1} + \beta_{C1D2}x_{C1}x_{D2} + \beta_{C1D3}x_{C1}x_{D3} + \dots + \beta_{C3D1}x_{C3}x_{D1} + \beta_{C3D2}x_{C3}x_{D2} + \beta_{C3D3}x_{C3}x_{D3} + \mathcal{E}$$
(1)

where xij (i = A, ..., D indicates the factor and j=1,.., 2 or 3 indicates the level) are binary variables equal to 1 when the i-th factor is at the j-th level, and 0 in any other case.  $\beta_0$  is the intercept and the 56  $\beta_{ij}$  are the coefficients of the model;  $\varepsilon$  is a random variable which follows a normal distribution with zero mean and constant standard deviation  $\sigma$ . The first eleven terms

describe the main effects, for instance:  $\beta_{B2}$  is the effect caused by setting the factor B at the level 2 (50% EPS content). The following 45 terms describe the effect of the interactions. For example,  $\beta_{C3D2}$  is the combined effect of set the mix of admixtures in 0.4A/0.1R/0.5S/6V and use CEM III. Further details about the procedure used can be found in the literature [23].

Once the levels for each factor have been decided, the D-optimal experimental design or fitted design [24] obtained using NemrodW [25] is transformed into the experimental work plan in Table 3, which shows the composition of the 36 mortars made to test each of the properties. This design allows very accurate estimation of the effects, since the variance inflation factors obtained were lower than 2.1.

Since the type of sand was not included as a factor in the experimental design, the experimental plan is the same independently of the type of sand used to manufacture the mortar. In this way, the 36 mortars given for the experimental work plan (Table 3) were made for each type of aggregate (silica sand and crushed limestone sand) to assess the influence of the type of aggregate on the properties of cement mortars containing waste EPS. Therefore, the design of the mortar can be optimised depending of the mineralogy of the available sand.

To help the reader with the interpretation of the results, six control mortars were made for each type of cement and sand, without EPS and admixtures. These data are included at the beginning of each subsection in Section 3, Results and Discussion.

### 2.4 Mortar characterisation

### 2.4.1 Workability and air content

The flow table method [21] was used to test the workability of the mortars and to determine the amount of waste EPS and admixtures to add to the mortars. This test is a measure of the fluidity and moisture content in the fresh mortar. The content of EPS and admixtures were chosen to achieve a suitable workability to use for masonry and rendering applications [22]. That is a flow table spread of  $175 \pm 10$  mm for mortars with a bulk density above 1200 Kg/cm3, and 160 ± 10 mm for mortars with bulk density between 600 and 1200 Kg/cm3. The air content in fresh mortars was tested according to EN 1015-7:1999 [26]. This property is related to the mortar workability and the capacity of the cement paste for give cohesion to the composite.

#### 2.4.2 Compressive strength, bulk density and adhesive strength

Three 4x4x16 cm specimens for each mix given in the experimental work plan were produced to test compressive strength. The samples were cured under water for 28 days at  $20 \pm 2^{\circ}$ C temperature and then tested using an hydraulic press (OMADISA 34.120.31) following the standard EN 196-1: 2005 [19]. Each compressive strength value was obtained from the average value of six tests. Dry bulk density of hardened mortars was determined according to EN 1015-10:2000 [27] using three resulting portions from the mechanical test for each mortar.

Adhesive strength of hardened mortar was tested following the standard EN 1015-12 [28] to measure of the proper functioning of the in-service mortar. To do the test, samples were made using a ceramic substrate with dimensions of 70x23x3 cm and water absorption coefficient of  $0.672\pm0.033$  kg/m<sup>2</sup>·min where a 10 ± 1 mm layer of mortar was applied. A sample was made for each mortar as in the working plan of Table 3. Five adhesive strength values were obtained from each sample after 28 days curing at the conditions specified in EN 1015-12 [28] and using an adhesive strength tester (KN-10 Neurtek).

#### 2.4.3 Capillary water absorption

Capillary water absorption of mortars was determined according to EN 1015-18:2003 [29]. Three specimens of 4x4x16 cm were made for each of the mortars shown in Table 3. The specimens were kept in moulds for 2 days and subsequently cured underwater for 5 days. After curing, specimens were cut in half, and dried in an oven at a temperature of  $65 \pm 2$  °C. After drying, the lateral sides of each specimen were sealed with an Epoxy waterproof paint (Acrilastic PX-03) to restrict water flow along the longitudinal axis. The water flux through the specimen was measured by partial immersion of the samples at a depth of 5 mm. The gain in water mass was measured by weighing the samples after 10 and 90 minutes of submersion. The capillary absorption coefficient, C<sub>90</sub>, was estimated from the slope following the equation  $W= a + C \cdot t^{1/2}$ , where W (kg/m<sup>2</sup>) is the capillary absorption, a (kg/m<sup>2</sup>) is the initial absorption, C (kg/m<sup>2</sup> min<sup>0.5</sup>) is the capillary absorption coefficient and t (min) is the absorption time, using the equation:  $C_{90} = 0.1(M2 - M1)$ , where M1 is the weight of the specimen after 10 min of testing, and M2 is the weight of specimen after 90 min of testing according to EN 1015-18:2003 [29].

### 2.4.4 Scanning electron microscopy (SEM)

The microstructure of selected samples was studied by examining fracture surfaces using scanning electronic microscopy (SEM, Hitachi S-3000N with BRUKER X-Flash 3001 detector). This method allows to visualise the differences between the two types of EPS used in this research. Figure 2a shows some EPS pw particles completely incorporated in a cement mortar sample, while Figure 2b shows the same for EPS gr particles. In Figure 2a it is possible to see how EPS pw particles have lost the typical honeycomb structure, characteristic of commercial EPS pearls, probably due to the grinding process. These EPS pw particles were covered by cement paste, and it is very difficult to distinguish the interface between this EPS and the cement paste. However, in Figure 2b, EPS gr particles are very easy to identify, as well as the cement paste-EPS interface. EPS gr particles maintain the characteristic honeycomb structure, because the grinding process is less intensive in this case.

#### 3. Results and discussion

#### 3.1 Individual factors and interactions

Table 4 shows that the chosen fitted design was significant, with p-values < 0.05 for all the properties, apart from the adhesive strength. Nonetheless, the model was used to make predictions of the adhesive strength of mortars made with silica sand, because it was significant to a 0.10 level, which is an acceptable level of significance for engineering applications. Nevertheless, the proposed model does not describe the experimental data for mortars made with crashed limestone sand. In this case, the model was not significant, since the p-value was 0.48.

Regarding the coefficient of determination  $R^2$ , Table 4 shows that the fitted design explains 96.8% of the workability data, 92.5% of the air content, 94.4% of the compressive strength, 84.4% of the adhesive strength, 93.2% of the bulk density and 92.8% of capillary water absorption data, for mortars made with silica sand. For mortars made with crushed limestone sand, the model explains 98.8% of the workability data, 95.5% of air content, 90.8% of compressive strength, 96.3% of bulk density, 95.2% of capillary absorption and 72.9% of the adhesive strength data. The last value highlights that the model is not suitable for explaining the adhesive strength of mortars made with waste EPS and crushed limestone sand.

Concerning the residual standard deviation  $S_{yx}$ , no significant differences were detected when the type of sand was changed, except for the air content and the capillary absorption (Table 4). In these cases,  $S_{yx}$  for mortars made with crushed limestone sand was lower than for mortars made with silica sand. This could be due to the higher amount of water in mortars made with crushed limestone sand, which produces more homogeneous mortars with less variation in properties. It is worth noting that, since the model is not valid for adhesive strength, it is not valid for making predictions about  $S_{yx}$  for this property.

Once the suitability of the proposed fitted design has been proved, the next step is to analyse the effect of changing the levels of the factors, as well as their interactions, for the mortar properties, using the presence-absence model (Equation 1). If all the binary variables are zero all the effects of the factors are absent, and then the model adopts the value  $b_0$ , which is the estimation of  $\beta_0$ . Hence, the value for the studied property is obtained, regarding which the effect of set each factor to a given level is assessed. It should be remembered that once the value of the binary variables is fixed, for example  $x_{A1} = 1$  and  $x_{B2} = 1$ , that is to use powdered EPS at 50% content, then the value of the interactions between both factors is also fixed. For example,  $x_{A1} x_{B2} = 1$ , but  $x_{A2} x_{B2} = 0$  and so on. Since the estimation of  $S_{yx}$  is known, together with the estimated values of the coefficients of the model ( $b_{A1}$ ,  $b_{A2}$ ,  $b_{B1}$ ,  $b_{B2}$ ,  $b_{B3}$ , ...,  $b_{C3D3}$ ), then their significance is known, that is, if each coefficient is different to zero at a level of significance of 0.05. The latest make possible to know the factors, levels and their interactions, which due to be different to zero have a significant influence in the studied property.

Figures 3 to 5 show the graphical analysis of the effect of changing the levels of the factors on the different studied responses. For each response or property, the coefficients of the fitted design are shown beside a bar, with the sign (positive or negative). The positive coefficients

make the value of the property higher when the factor or interaction is at the corresponding level, while the negative coefficients reduce the value of the property. Each coefficient is identified by the corresponding subscript in the Equation 1: A1, A2, B1, B2, B3, C1, C2, C3, D1, D2 and D3 for the factors, and A1-B1, A1-B2,...., C3-D3 for the interactions. In order to distinguish between the coefficients which are significantly different to zero and the coefficients that can be consider null and therefore without effect in the property, two vertical broken lines are added to the graphs. These vertical lines mark the limits of the critical region of the significance test to a level of 0.05 (H<sub>0</sub>: the coefficient is zero, versus H<sub>a</sub>: the coefficient is different to zero). Each coefficient has a different standard deviation. The bar which represents each coefficient is a standardised value, which is the coefficient divided by the standard deviation. As a result, the length of the bars is not the value of the coefficient, but is proportional to them. For instance, in Figure 3a the coefficient A1 is -7.5 (significant) and the coefficient D1 is -11.0 (also significant), although the bars that indicate that they are significantly different to zero are almost of the same length. This tool allows visual establishment of the 56 coefficients of each model which are significant as well as the sign of each effect. Hence, it is possible to predict which levels should be chosen for each factor, depending on the effect to be achieved in a specific property, allowing the design of bespoke lightweight mortars.

#### 3.2 Workability and air content

The significant factors for the workability of mortars containing EPS made with silica sand were: A1, A2, B1, B3, C1, C3, D1, D2 and D3 (Figure 3a). Thus, all the studied factors had a significant influence on the workability, although factors at levels B2 and C2 were not significant. It is also evident that the workability was reduced when powdered EPS was used (A1), when the EPS content increased (B3), using the mix with the lowest amount of admixtures (C3) and the cement with the highest amount of clinker (D1). The significant interactions were: B2-C2, B2-C3, C2-D3, C3-D2 and C3-D3. The B-C interactions show that there is a relationship between the EPS content and the mix of admixtures used. In this case, when a 50% EPS (B2) was used, it would not be the same to use a 0.4/0.1/0.5/6 mix of admixtures (C2) than a 0.3/0.1/0.4/6 (C3) mix, since they have an opposite effect on the workability. This justifies the use of admixtures to guarantee a suitable workability for these mortars. The same happens between the mix of admixtures and the type of cement used (interactions C-D). For the mix of admixtures with the lowest content of air-entraining agent, C3 (0.3A/0.1R/0.4S/6V), a contrary effect on workability was observed depending on whether CEM II (D2) or CEM III (D3) was used. For CEM II, the workability increases by 4 mm relative to the average value, as the coefficient for the interaction C3-C2 shows (Figure 3a). Conversely, if CEM III is used, the workability decreases by 5 mm (C3-D3), since this interaction has a negative value. The A factor (type of EPS) does not have significant interactions with any of the other factors (B, C or D). That implies that the effect on the workability of the type of EPS is independent of the EPS content (B), the mix of admixtures (C) and the type of cement (D).

From the information in Figure 3a, it is possible to choose the levels of each factor to achieve the desirable workability for masonry, rendering and plaster mortars of 175 ± 10 mm [30, 31]. The flow table spread values for control mortars made with silica sand were 190  $\pm$  1 mm, 207  $\pm$ 1 mm and 201 ± 2 mm for CEM I, CEM II and CEM III respectively. Therefore, of these three control mortars are suitable for these applications. Control mortars values can be used by the reader as a guide to understand better the effect of changing the level of the different factors in the mortars. For example, if the objective is to increase the workability of the mortars with silica sand, where the fitted b<sub>0</sub> value was 182 mm, the levels with the highest positive significant coefficient should be chosen when the significant factors are considered (that is: A2, B1, C1 and D3). Subsequently, it should be checked if any of the significant interactions involve the chosen levels. This is not the case, as Figure 3a shows, so there is no conflict between the levels that maximise the significant factors and those which maximise the interactions. As a result, the values  $x_{A2} = x_{B1} = x_{C1} = x_{D3} = 1$  are assigned to the binary variables of the model of the Equation 1, with the remaining values set to zero. Hence, the non-zero terms correspond with these binary variables and their products, which are associated with the interactions: A2-B1, A2-C1, A2-D3, B1-C1, B1-D3 and C1-D3. Taking this into account, the Equation 1 turns into Equation 2:

 $y = \beta_0 + \beta_{A2}X_{A2} + \beta_{B1}X_{B1} + \beta_{C1}X_{C1} + \beta_{D3}X_{D3} + \beta_{A2B1}X_{A2}X_{B1} + \beta_{A2C1}X_{A2}X_{C1} + \beta_{A2D3}X_{A2}X_{D3} + \beta_{B1C1}X_{B1}X_{C1} + \beta_{B1D3}X_{B1}X_{D3} + \beta_{C1D3}X_{C1}X_{D3} = 181.9 + 7.5 + 4.6 + 2.4 + 7.0 - 0.2 + 0.6 - 0.2 + 0.8 - 1.3 + 1.1 = 204.2 \text{ mm}$ (Equation 2)

For the aforementioned conditions, a flow table spread value of  $204 \pm 4$  mm is obtained. That is, the highest workability can be achieved using EPS gr (A2), 40% content of EPS (B1), admixtures mix of 0.8A/0.1R/0.8S/6V (C1) and CEM III (D3). However, if this combination of materials is used, the obtained mortar would be too fluid to achieve the desirable workability for masonry, rendering and plaster mortars of  $175 \pm 10$  mm [22]. Therefore, as the independent term of the fitted design (b<sub>0</sub>) is 182 mm, within the acceptable range for this property, a combination of factor levels that generate a less fluid mortar is required.

This can be achieved in several ways. First, it is possible to use the same EPS content, but change the EPS type from ground to powdered EPS. This entails working with the factors A1, B1, C1 and D3. In this way, an increased flow table spread of 189 ±4 mm is achieved. Consequently, the resultant mortar is less fluid and closer to the required workability value. The influence of the geometry and particle size of the waste EPS on the workability of the mortars made with silica sand is clearly demonstrated, because the use of EPS pw reduces workability by 10.8% compared to the EPS gr.

Other modifications are required to achieve mortars which comply with the standards [22]. Using EPS pw (A1), 60% of EPS (B3), the mix of admixtures 0.4A/0.1R/0.5S/6V (C2) and CEM II (D2) the model predicts a value for the flow table spread of 173 mm, as this combination of factors

reduces the workability by 9 mm. Therefore, it is possible to increase the EPS content and decrease the amount of admixtures, by using cement with an intermediate amount of clinker. Another way to produce the required workability is to use EPS pw (A1), 60% of EPS (B3) and the 0.3A/0.1R/0.4S/6V mix of admixtures (C3) and CEM III (D3). This would reduce the workability by 14 mm, giving a final value of  $168 \pm 4$  mm. The advantage of this option is that a workable mortar is achieved using the highest amount of EPS, the lowest amount of admixtures and the cement with the lowest amount of clinker, thus giving a more sustainable mortar.

The significant factors for the workability of EPS mortars made with crushed limestone sand (Figure 3b) were: A1, A2, B1, B3, C1, C3, D1 and D3, i.e. all except B2, C2 and D2. Therefore, while the change in the type of sand does not change the factors which influence workability, three individual factors at three levels (B2, C2 and D2) had no impact for mortars made with crushed limestone sand. The workability is reduced using EPS pw (A1), increasing the amount of EPS (B3), using the mix with the lowest amount of admixtures (C3) and with the cement with the highest amount of clinker (D1). The A-C interactions (type of EPS with mix of admixtures), B-C (EPS content with mix of admixtures) and B-D (EPS content with type of cement) are not significant. However, the A-B interactions (type of EPS with EPS content), A-D (type of EPS with cement type) and C-D (mix of admixtures with cement type) are significant.

For mortars made with crushed limestone sand the flow table spread values for control mortars were 195 ± 1 mm, 205 ± 1 mm and 200 ± 3 mm for CEM I, CEM II and CEM III respectively. These are all above the recommended value of 175 ± 10 mm [22]. When limestone sand was used, the model assigned a  $b_0$  value of 202 mm for mortars with EPS. This means the mortar is too fluid, due to the use of a 0.6 water/binder ratio for mortars with crushed limestone sand. This choice was based on previous work [16], which demonstrated that mortars made with crushed limestone sand need a higher amount of water to give a suitable workability, due to the high fines content of this sand (Figure 1). That is the case for mortars with no admixtures and no EPS. However, the present study shows that the use of admixtures and cements with lower clinker content, can achieve a suitable workability for masonry, plastering and rendering mortars without increasing the water content. Most of the mortars made with limestone sand were fluid, and therefore it is important to reduce workability to an acceptable value. To maximise the reduction in the workability, A1, B3, C3 and D1 should be chosen. Using EPS pw, the highest EPS content, the lowest amount of admixtures and cement with higher clinker content will cause a reduction of 22.6% relative to b<sub>0</sub>. The flow table spread value decreases by 46 mm, to a predicted value of 156 mm. However, this equates to a dry and non-workable mortar.

An acceptable flow table spread value of 185 mm is obtained when A1, B3, C3 and D2 are chosen. This produces a reduction of 8 % in relation to  $b_0$ . Similarly, if A1, B3, C3 y D3 are chosen, the fitted design gives a value of 180 mm, a reduction of 11% relative to  $b_0$ . This illustrates the importance of studying all interactions. If interactions were not studied, D3 would not be a suitable choice, because it increases the workability. However, when the interactions between the factors are considered, especially A1-D3 and B3-D3 interactions, it is possible to

achieve a reduction in workability. This option leads to a sustainable mortar with a suitable workability (180  $\pm$  4 mm), the highest amount of waste EPS, the lowest amount of admixtures and the cement with the lowest content of clinker (EPS pw, 60% EPS, 0.3A/0.1R/0.4S/6V and CEM III).

Air content is linked with workability and compressive strength, and allows an assessment of whether the mortar is homogeneous. A high air content is associated with high workability and low compressive strength. The standards do not specify a value for the air content, but in practice, a value between 20-30% is accepted for manufacturers for commercial masonry, plastering and rendering mortars. Previous work has demonstrated that the presence of EPS increases the air content in mortars [16].

Figure 3c shows the impact of air content on mortars made with silica sand. The air content for control mortars made with silica sand were 7.0 %, 5.5 % and 5.0 % for CEM I, CEM II and CEM III, respectively. All of them are too low to satisfy the recommended range of 20-30%. The fitted  $b_0$  value was 31 %. The coefficients C1, C3, D1 and D2, as well as interactions A1-C1 and A2-C1 were significant in this case. To increase the air content, A1, C1 and D2 should be chosen. Thus, the highest values for air content could be obtained using the factors: A1, B1, C1 and D2 (EPS pw, 40% EPS, 0.8A/0.1R/0.8S/6P and CEM II). In this case, the fitted value is  $41 \pm 2\%$ , which equates to an increase  $b_0$  value by 29 %. Conversely, if the objective is reduce the air content, then A2, B3, C3 and D1 should be chosen. That means EPS gr, 60% EPS, 0.3A/0.1R/0.4S/6P mix of admixtures and CEM I, which gives 25 % ± 2% air content or 19 %reduction compared with  $b_0$ . To manufacture the most sustainable mortar, factors A1, B3, C3 and D3 (EPS pw, 60% EPS, 0.3A/0.1R/0.4S/6V mix of admixtures and CEM III) or A2, B3, C3 and D3 (EPS gr, 60% EPS content, 0.3A/0.1R/0.4S/6V, and CEM III) should be selected. These options give an air content of  $27 \pm 2\%$  and  $29 \pm 2\%$  respectively. That proves there is no significant difference between the EPS type in terms of the air content (Figure 3c). The influence of the type of admixtures is highlighted using the following combinations of factors and levels: A1, B3, C1 and D3 (EPS pw, 60% EPS content, 0.8A/0.1R/0.8S/6V and CEM III) as well as A1, B3, C3 and D3 (EPS pw, 60% EPS, 0.3A/0.1R/0.4S/6V and CEM III) where the fitted model air content is  $35 \pm 2\%$  and  $27 \pm 2\%$  respectively. These values are significantly different and consistent with the amount of air entraining agent in the mix. It should be noted that the levels of the factor D (type of cement) is dramatically different for CEM I (coefficient= -3.4) to CEM II (+3.8) and CEM III (-0.4). This would not be apparent without considering three levels for this factor.

The air content of control mortars made with crushed limestone sand were 3.8 %, 2.8% and 4.0% for CEM I, CEM II and CEM III, respectively. As for the control mortars made with silica sand, these values are not enough to satisfy the recommended values for masonry, rendering and plastering applications, of 20-30%. When mortars were made with crushed limestone sand (Figure 3d) the significant factors were: C1, C2, C3, D1 and D2 and none of the interactions were significant. Hence, the air content solely depends on the admixture mix and the cement

type. The b<sub>0</sub> value fitted by the model was 29 %, within the acceptable interval for commercial mortars (20-30%). However, it is still possible to decrease the air content slightly, to obtain an intermediate value for this property. Using EPS pw, the highest EPS content (60%), the mix with the lowest amount of admixtures and the cement with the highest amount of clinker (A1, B3, C3 and D1) it is possible to reduce the air content to  $21 \pm 2$  %. Applying sustainable criteria leads to mortars with the highest waste EPS content, the lowest amount of admixtures, and cement with the lowest clinker content. The combination A1, B3, C3 and D3 and also A2, B3, C3 and D3 makes this possible, with predicted values of  $26 \pm 2$  % and  $28 \pm 2$  % respectively.

#### 3.3 Compressive and adhesive strength

The compressive strength of control mortars with silica sand at 28 days curing time were  $45.6 \pm 3.7$  MPa,  $43.9 \pm 3.0$  MPa and  $44.6 \pm 2.7$  MPa for CEM I, CEM II and CEM III, respectively.

Figure 4a shows the significant factors for compressive strength after 28 days curing time for mortars with silica sand were: A1, A2, B2, C1, C2, C3, D2 and D3. Hence, while all the factors influence this property, the levels B1, B3 and D1 were not significant. In addition, the significant interactions were: A-C (EPS type with admixture mix), B-D (EPS content with cement type) and C-D (admixture mix with cement type). The fitted  $b_0$  value for compressive strength was 6.6 MPa (Table S6 in the SM). To increase the compressive strength, the combination A1, B1, C1 and D2 is appropriate. The significant interactions for this combination are B1-D2 and C1-D2, and in both cases they increase the compressive strength. That involves working with EPS pw (A1), 40% EPS content (B1), an admixture mix 0.8A/0.1R/0.8S/6V (C1) and CEM II (D2). This combination increases by 5.0 MPa the  $b_0$  value, giving a value for compressive strength of 11.6 ± 1.1 MPa. Increasing the EPS content from B1 to B3 in the previous combination, gives a value for compressive strength of  $11.7 \pm 1.1$  MPa, which is not significantly different to the mortar with the lowest amount of waste EPS. Both mortars can be classified as M10, with respect to the standard for masonry mortars EN 998-2 [31] and as CSIV following the EN 998-1 for rendering and plastering mortars [30]. Once again, by choosing the highest EPS content, the lowest amount of admixtures and lower clinker content (A1, B3, C3 and D3) it is possible to manufacture more sustainable mortars. The compressive strength in this case is  $5.7 \pm 1.1$  MPa. This mortar can be classified as M5 with respect to the standard for masonry mortars [31] and CSIII regarding the standard for rendering and plastering mortars EN 998-1 [30].

The model also shows that, when EPS gr (A2) is used, the compressive strength is reduced, although interactions also need to be considered. The combination of factors and levels A2, B1, C1 and D2, and since B1 and B3 are not significant levels, the combination A2, B3, C1 and D2, produce compressive strength values of  $13.0 \pm 1.1$  MPa and  $12.2 \pm 1.1$  MPa, respectively. These mortars are classified as M10 masonry mortars [31] and CSIV with respect to the rendering and plastering standard [30]. If an intermediate amount of waste EPS is used, namely 50% (B2), the model predicts that the compressive strength decreases by 39.2 %, to 7.9  $\pm$  1.1MPa, compared to mortars containing 40% or 60% EPS.

Manufacturing a sustainable mortar can be achieved using A2, B1, C2 and D2 ( $7.3 \pm 1.1$  MPa), A2, B1, C1 and D3 ( $7.0 \pm 1.1$  MPa) or A2, B3, C2 and D2 ( $6.8 \pm 1.1$  MPa). These values are not significantly different to the fitted b<sub>0</sub> (6.6 MPa). These mortars are classified as CSIII, CSIV for rendering and plastering mortars [30] and M5 for masonry mortars [31]. The combination A2, B3, C3 and D3 gives a compressive strength of  $0.9 \pm 1.1$  MPa, which is not suitable for any of the studied applications and illustrates those interactions which decrease the compressive strength.

Control mortars made with crushed limestone sand had compressive strength values of  $46.6 \pm 3.0$  MPa,  $42.6 \pm 2.0$  MPa and  $50.0 \pm 1.6$  MPa, for CEM I, CEM II and CEM III, respectively. The significant factors were: A1, A2, C1, C3, D1 and D2, and none of the interactions were significant (Figure 4b). Thus, it can be concluded that the content of EPS does not determine the compressive strength. Comparing Figure 4b with Figure 4a, it is observed that the change in the levels in the mix of admixtures (C) and the type of cement (D), have a different effect on the compressive strength depending on the type of sand used. This was not observed for the fresh properties, workability and air content, and this shows that the mineralogy and particle size distribution of the two sand types strongly influence compressive strength.

The fitted b<sub>0</sub> value for the compressive strength of mortars made with crushed limestone sand was 7.4 MPa. To obtain the highest possible compressive strength for this type of mortars, the combination A1, B1, C3 and D1 should be chosen. Using 40% EPS pw, the admixtures 0.3A/0.1R/0.4S/6V and CEM I will give a value of  $11.4 \pm 1.0$  MPa, which is an increase of 4 MPa above the b<sub>0</sub> value. It is possible to work with a higher content of EPS without decreasing the compressive strength, since the EPS content had no significant effect at any of the levels. A possible combination to achieve this it is A1, B3, C3 and D1 (11.3  $\pm 1.0$  MPa). If obtaining a mortar with the lowest amount of admixtures and clinker is prioritised, the combination giving the highest compressive strength values would be A1, B2, C3 and D3 (50% EPS pw, 0.3A/0.1R/0.4S/6V and CEM III), providing a value of 10.4  $\pm 1.0$  MPa. Since the EPS content is not significant (B), using different EPS contents leads to similar compressive strength values. For A1, B1, C3 and D3 the model predicts a value of 10.2  $\pm 1.0$  MPa and for A1, B3, C3 and D3 the value is 10.2  $\pm 1.0$  MPa. All the above mentioned mortars can be classified as CSIV rendering and plastering mortars [30] and M10 masonry mortars [31].

It is important to highlight that mortars made with crushed limestone sand can achieve the same strength as those made with silica sand. However, when using limestone, it is possible to make mortars using the lowest admixture (C3) and the lowest clinker content (D3). Therefore, the influence of sand type on the compressive strength is shown again. Further, the negative effect of ground EPS on this property is reflected in combinations of factors and levels when the EPS type changes. For example, A1, B1, C3 and D1 gives a compressive strength of  $11.4 \pm 1.0$  MPa and A2, B1, C3 and D1 gives  $8.1 \pm 1.0$  MPa, which is a reduction of 29%. Mortars made with waste ground EPS can be classified as CSIV for rendering and plastering applications [30] and M5 for masonry applications [31].

An important property for mortar durability is the adhesive strength. Control mortars made with silica sand had values of  $0.50 \pm 0.22$  MPa,  $0.44 \pm 0.04$  MPa and  $0.50 \pm 0.08$  MPa for CEM I, CEM II and CEM III, respectively. Figure 4c shows that C1 and D3 were the only factors which significantly impact the adhesive strength of mortars made with silica sand. However, significant interactions were: B-D (EPS content with cement type) and C-D (admixture mix with cement type). Consequently, the only factor that did not influence the adhesive strength is EPS type (A). In addition, working with CEM III (D3) reduced the adhesive strength. Hence, to avoid reducing the adhesive strength this cement type should not be used. Furthermore, the negative interaction C1-D3 (0.8A/0.1R/0.4S/6V and CEM III) also decreases the adhesive strength by 0.12 MPa. In addition, predicted negative interactions when 50% EPS and CEM II are used at the same time (B2-D2), and between the mix of admixtures 0.3A/0.1R/0.4S/6V and CEM I (C3-D1) have a similar effect. These interactions decrease the adhesive strength by 0.08 MPa (B2-D2) and 0.10 MPa (C3-D1). Only the significant interaction C1-D1 increases this property. For example, the combination A2, B1, C1 and D1 (40% EPS gr, 0.8A/0.1R/0.8S/6V for the mix admixtures and CEMI) provides a value of 0.71 ± 0.10 MPa, an increase of 42% with respect to the fitted  $b_0$ , 0.50MPa. A very similar value (0.72 ± 0.10 MPa) is obtained with the combination A2, B3, C1 and D1. This allows manufacture of mortars with suitable values for adhesive strength but containing the highest amount of EPS. However, it is not possible to significantly increase the adhesive strength if lower amounts of admixtures or cements with low clinker content are used. That is the case for the combinations: A2, B1, C1 and D2 ( $0.66 \pm 0.1$  MPa); A2, B3, C1 and D2 (0.65 ± 0.1 MPa); or A2, B3, C3 and D2 (0.63 ±0.10 MPa).

The highest adhesive strength values are obtained using a high amount of admixtures (C1) and cement with a high clinker content (D1). Under these conditions, the EPS type (A1 or A2) can be altered or the highest EPS content used (B3), without reducing the adhesive strength. More sustainable mortars, with a lower amount of admixtures (C3) or cement with lower clinker content (D2), have lower adhesive strength. Cements with the lowest amount of clinker (D3) reduce the adhesive strength. As an example, combinations A2, B1, C1 and D3 or A2, B3, C3 and D3 give values of  $0.45 \pm 0.10$  MPa and  $0.44 \pm 0.10$  MPa, respectively.

Masonry mortars require an adhesive strength > 0.15 MPa [31], while plastering and rendering mortars require a minimum adhesive strength of 0.30 MPa [30]. Hence, it is possible to make sustainable mortars with a high EPS content and cements with minimum clinker content, which satisfy the relevant standards.

The fitted equation was not significant for mortars made with crushed limestone sand (p-value = 0.476 > 0.05). For this reason, the factors and interactions are not discussed further for these mortars.

#### 3.4 Bulk density and capillary absorption

Comparing Figures 4a and 4b, with Figures 5a and 5b, it is observed that the tendencies between compressive strength and bulk density are the same for both types of sand, since the same significant factors apply. This is consistent with the similar physicochemical properties of these materials, and this confirms the validity of the fitted design chosen in this research.

Figure 5a shows the factors and interactions for the bulk density of mortars made with standard silica sand at 28 days curing time. The values for the bulk density of control mortars with silica sand at 28 days curing time were 2090  $\pm$  100 kg/m<sup>3</sup>, 2070  $\pm$  300 kg/m<sup>3</sup> and 2050 kg/m<sup>3</sup>, for CEM I, CEM II and CEM III, respectively. All the factors are significant, except for B3 (60% EPS content). The significant interactions were between EPS type and the mix of admixtures (A-C), the EPS content and the cement type (B-D) and between the admixture mix with the highest amount of admixtures and CEM III (C1-D3). The maximum bulk density of lightweight rendering and plastering mortars specified by standard EN 998-1 is 1300 Kg/m<sup>3</sup> [30]. As the fitted b<sub>0</sub> value calculated by the model is 1300 kg/m<sup>3</sup>, to obtain lightweight mortars, factors at levels with a negative coefficient should be chosen, together with interactions that do not increase the bulk density. Sustainable mortars can be made with A2 (EPS gr), B2 or B3 (50% or 60% EPS), C3 (0.3A/0.1R/0.4S/6V) and D3 (CEM III). These combinations have bulk density of 1100 ± 100 kg/m<sup>3</sup>, a decrease of 200 kg/m<sup>3</sup> compared with b<sub>0</sub>. Conversely, to manufacture mortars with higher bulk density and therefore higher compressive strength, the combinations A1, B1, C1 and D2 (1550  $\pm$  100 kg/m<sup>3</sup>) and A2, B1, C1 and D2 (1590  $\pm$  100 kg/m<sup>3</sup>) are appropriate. This involves decreasing the EPS content and increasing the admixtures and clinker content of the mortars.

Control mortars made with crushed limestone sand had a bulk density at 28 days curing time of 2040  $\pm$  200 kg/m<sup>3</sup>, 2020  $\pm$  100 kg/m<sup>3</sup> and 2020  $\pm$  100 kg/m<sup>3</sup>, for CEM I, CEM II and CEM III, respectively. Figure 5b shows the significant factors and interactions for the bulk density at 28 days curing time for mortars containing EPS and crushed limestone sand. These were: A1, A2, C1, C3, D1 and D2. In this case, the EPS content has not influenced this property. The significant interactions were: A-C (EPS type with admixture mix) and C-D (admixture mix with cement type). The combinations A2, B2, C1 and D2 or A2, B3, C3 and D2 provide bulk density values of 1200  $\pm$  100 kg/m<sup>3</sup> and 1300  $\pm$  100 kg/m<sup>3</sup> respectively. Thus, both give important reductions in bulk density relative to the fitted b<sub>0</sub> value of 1400 kg/m<sup>3</sup>. To manufacture the most sustainable mortar, the combination of factors and levels A2, B3, C3 and D3 should be chosen. This gives a predicted bulk density value of 1400  $\pm$  100 kg/m<sup>3</sup>, which is not a lightweight mortar according to the standard for rendering and plastering applications [30]. However, by increasing the amount of admixtures (A2, B3, C1 and D3) a value of 1200  $\pm$  100 kg/m<sup>3</sup> is obtained, which can be considered a lightweight mortar.

The last property studied was the capillary water absorption, which was included to assess mortar durability. According to the standard for rendering and plastering applications, mortars can be classified as  $W_1$  ( $C_{90} < 0.40 \text{ kg/m}^2 \cdot \text{min}^{0.5}$ ) or  $W_2$  ( $C_{90} < 0.20 \text{ kg/m}^2 \cdot \text{min}^{0.5}$ ) depending on

their capillary water absorption coefficient [30]. The C<sub>90</sub> values for control mortars made with silica sand were  $0.18 \pm 0.01 \text{ kg/m}^2 \cdot \text{min}^{0.5}$ ,  $0.18 \pm 0.01 \text{ kg/m}^2 \cdot \text{min}^{0.5}$  and  $0.12 \pm 0.01 \text{ kg/m}^2 \cdot \text{min}^{0.5}$ , for CEM I, CEM II and CEM III, respectively. Hence, all of these control mortars satisfy the desired values for the target applications. However, the EPS tended to decreases C<sub>90</sub>, enhancing the durability of these mortars.

Figure 5c shows that the significant effects for the capillary water absorption coefficient of mortars made with standard silica sand were: A1, A2, C1, C2, C3, D1 and D2. Therefore, all the factors except those related with the EPS content (B), have a significant influence on this property. The significant interactions were: A-C (EPS type with mix of admixtures) and A-D (EPS type with cement type). This highlights the influence of EPS type on the properties of the hydrated cement paste and consequently the capillary absorption [12, 13]. It is useful to know the combination of levels and factor which minimize this capillary water absorption, as the standard EN 998-1 [30] requires values lower than 0.40 kg/m<sup>2</sup>·min<sup>0.5</sup> for mortars classified as  $W_1$  and 0.20 kg/m<sup>2</sup>·min<sup>0.5</sup> for those classified as  $W_2$ . The combination A2, B1, C1 and D1 gives a C<sub>90</sub> value of 0.14 ± 0.07 kg/m<sup>2</sup>·min<sup>0.5</sup>. However, this value is similar to the fitted b<sub>0</sub> value of 0.16 kg/m<sup>2</sup>·min<sup>0.5</sup>, since positive interactions increase the C<sub>90</sub> value, particularly A2-C1 and A2-D1. Combinations of factors which produce sustainable mortars, containing either high EPS content or cement with lower clinker content are A2, B3, C1 and D1 and A2, B3, C1 and D3 respectively. Both mortars have C<sub>90</sub> values of 0.06 ± 0.07 kg/m<sup>2</sup>·min<sup>0.5</sup>. These mortars are classified as  $W_2$  [30].

Other combinations, such as A1, B3, C1 and D3, allow classification of these mortars as  $W_2$  according to the standards [30], guaranteeing the durability, as it gives a  $C_{90}$  value of 0.11 ±0.07 kg/m<sup>2</sup>·min<sup>0.5</sup>. The lowest  $C_{90}$  value, 0.01 ±0.07 kg/m<sup>2</sup>·min<sup>0.5</sup>, involves working with the lowest content of EPS pw and the highest amount of admixtures and clinker content, namely A1, B1, C1 and D1.

Comparing Figure 5c with Figure 5d, it can be concluded that the sand type does not change the influence of EPS type and cement type on the capillary water absorption. However, the influence of the mix of admixtures depends on the mineralogy of the sand used.

Control mortars made with crushed limestone sand had  $C_{90}$  values of 0.45 ± 0.08 kg/m<sup>2</sup>·min<sup>0.5</sup>, 0.35 ± 0.06 kg/m<sup>2</sup>·min<sup>0.5</sup> and 0.38 ± 0.05 kg/m<sup>2</sup>·min<sup>0.5</sup> for CEM I, CEM II and CEM III, respectively. These values are higher than those obtained with silica sand. Therefore, it was necessary to prove if the EPS particles lowered the  $C_{90}$  value. The significant factors for capillary water absorption coefficient  $C_{90}$  for mortars made with crushed limestone were: A1, A2, B3, C1, C3, D1, D2 and D3 (Figure 5d). The significant interactions were: A-C (EPS type with mix of admixtures), A-D (EPS type with cement type), B-D (EPS content with cement type) and C-D (admixture mix with cement type). Considering that the fitted b<sub>0</sub> value was 0.08 kg/m<sup>2</sup>·min<sup>0.5</sup> and looking for the highest reduction in the C<sub>90</sub> coefficient, it is possible to obtain a value for C<sub>90</sub> of 0.04 ± 0.02 kg/m<sup>2</sup>·min<sup>0.5</sup> using the combination of factors and levels A2, B2, C3 and D3. The

existence of significant negative interactions means values for  $C_{90}$  of  $0.02 \pm 0.02 \text{ kg/m}^2 \cdot \text{min}^{0.5}$ and  $0.04 \pm 0.02 \text{ kg/m}^2 \cdot \text{min}^{0.5}$  can be achieved using the combinations A1, B1, C3, D3 and A1, B3, C3, D3, respectively. All the mortars manufactured with crushed limestone sand can be classified as W<sub>2</sub> according to the standard for rendering and plastering [30].

#### 4. Conclusions

This study used ad-hoc designs based on the d-optimal criterion to supply a comprehensive mathematical model for the effect of the levels of factors on the properties of lightweight sustainable mortars containing waste EPS. Four factors were studied at several levels: EPS type (A), EPS content (B), admixture mix (C) and cement type (D). The influence in the properties of two types of aggregates was also reported. The following are the key conclusions:

- 1) For mortars made with silica sand, the combination of factors A1B3C3D1 gave the lowest value for flow table spread, of up to 157.3 ± 4 mm. Workability was increased by up to 30% using 40% EPS gr (A2B1), the admixture mix 0.8A/0.1R/0.8F/6V (C1) and either CEM II (D2) or CEM III (D3). However, workability was just increased by 20% if EPS pw instead of EPS gr was used for this composition. This shows that geometry and EPS particle size had a clear influence on the workability of waste EPS mortars. Significant interactions on the workability of mortars made with silica sand were B-C, EPS content and the mix of admixtures, and C-D, the mix of admixtures and cement type. When 50% EPS content was used, workability was reduced by 4.2 mm for the mix 0.4A/0.1R/0.5F/6V, although it was increased by 4.7 mm if the mix with the lowest additives content, 0.3A/0.1R/0.4F/6V was used. In addition, using the lowest content of additives with CEM II increased the workability by 3.5 mm but the use of CEM III reduced it by 4.8 mm. Interactions, such as these, that decrease workability should be avoided. The way that the main factors affect the workability was independent of the type of sand used. When limestone sand was used, significant interactions were observed between the type of EPS and the EPS content (A-B), EPS type and cement type (A-D), and admixture mix and cement type (C-D). One of the most significant was obtained when the highest amount of additives was used (C1). In that case, the use of CEM I increased workability by 11.4 mm, but the use of CEM II or CEM III reduced it by 7.3 and 4.1 mm respectively, highlighting the influence of the cement type on this property.
- 2) For mortars made with silica sand, both types of waste EPS could be increased from 40% to 60% without significantly decreasing the compressive strength. To achieve this, the mix of admixtures 0.8A/0.1R/0.8S/6V and CEM II was needed. Mortars made with EPS pw and gr had a maximum compressive strength of 11.7 ± 1.1 MPa and 13.0 ± 1.1 MPa, respectively. These mortars are classified as M10 and CSIV according to the relevant standards. The use of 50% EPS decreased the compressive strength by 35.7% for EPS pw and 39.2% for EPS gr, compared to mortars containing 40% or 60% EPS. This is reflected by the negative coefficients given by the model for 50% EPS content. While sustainable mortars can be

manufactured with both types of EPS. EPS pw allows the highest EPS content (60%), lowest admixture content (0.3A/0.1R/0.4S/6V) and the cement with higher clinker content (CEM III) to be used. This composition gave a compressive strength of  $5.7 \pm 1.1$  MPa, classified as M5 and CSIII. Mortars made with limestone sand can achieve the same strength as those made with silica sand. However, when using limestone sand, it was possible to manufacture sustainable mortars made with high EPS content (B3), low amount of admixtures (C3) and cement with low clinker content (D3). These mortars are classified as CSIV and M5, which compressive strength values  $\ge 6$  N / mm<sup>2</sup>.

- 3) The capillary water absorption coefficient (C<sub>90</sub>) of mortars containing silica sand was significantly impacted by all factors, except those relating to EPS content; in addition, the interaction between EPS type and admixture mix (A-C), and between type of EPS and cement (A-D) had significant impacts on this property. This revels the influence of the type of EPS on C<sub>90</sub>. When CEM III was used, the use of EPS gr instead of EPS pw decreased C<sub>90</sub> by 45% for the mix with the highest EPS content (B3) and the mix of admixtures C1 (0.8A/0.1R/0.8S/6V). For mortars containing crushed limestone sand all factors were significant as well as most of the interactions, which proves the influence of sand type on this property. Both types of sand allowed production of sustainable mortars, containing either high EPS content or cement with lower clinker content. Moreover, all mortars made with limestone sand are classified as W2 according to the standard EN 998-1, i.e. they have C<sub>90</sub> values ≤ 0.20 kg/m<sup>2</sup>·min<sup>0.5</sup>.
- 4) The chosen fitted design was significant for all the studied properties except for the adhesive strength of mortars containing limestone sand. Ad-hoc designs allow both the simultaneous effect of individual factors and synergic effects, resulting from interactions between factors, to be evaluated. Hence, it is possible to mathematically model the effect of the chosen factors on the studied properties to design sustainable bespoke mortars containing waste EPS for masonry, rendering and plastering applications. Environmentally sustainable cement mortars which satisfy EU standards were manufactured containing up to 60% EPS, low amounts of admixtures, cement with low clinker content and both sand types. These mortars gave values for the spread in the flow table between 168 and 180 ± 4 mm, bulk density between 1280 and 1110 ± 100 kg/m<sup>3</sup>, and C<sub>90</sub> between 0.279 and 0.025 ± 0.07 kg/m<sup>2</sup>·min<sup>0.5</sup>, when they were made with silica sand. Hence, these mortars have appropriate properties to be used commercially.

#### Acknowledgments

The authors wish to thank the Spanish Ministry of Science and Innovation and European Union (FEDER) for project funding (BIA 2007-61170) and the FPI scholarship (BES-2009-012166) awarded to Veronica Ferrandiz Mas which allowed her to develop her doctoral thesis. The

authors also wish to thank Holcim Morteros S.A., for the supply of cement and admixtures necessary to carry out this research. The experimental design was carried out in the Mathematical and Computational Department at Burgos University, thanks to the funding awarded to Veronica Ferrandiz-Mas, for training of researchers in other centres from Alicante University.

### **References:**

[1] <u>http://www.anape.es</u> (Web de la Asociación Nacional del Poliestireno Expandido). Accessed 05/11/2014.

[2] Chen W, Hao H, Hughes D, Shi Y, Cui J, Li Z-X. Static and dynamic mechanical properties of expanded polystyrene. Materials & Design. 2015;69:170-80.

[3] Corinaldesi V, Gnappi G, Moriconi G, Montenero A. Reuse of ground waste glass as aggregate for mortars. Special Issue - VARIREI 2003. 2005;25:197-201.

[4] Chen W, Hao H. Experimental and numerical study of composite lightweight structural insulated panel with expanded polystyrene core against windborne debris impacts. Materials & Design. 2014;60:409-23.

[5] Laukaitis A, Žurauskas R, Kerien≐ J. The effect of foam polystyrene granules on cement composite properties. Cement and Concrete Composites. 2005;27:41-7.

[6] Babu DS, Ganesh Babu K, Tiong-Huan W. Effect of polystyrene aggregate size on strength and moisture migration characteristics of lightweight concrete. Cement and Concrete Composites. 2006;28:520-7.

[7] Babu KG, Babu DS. Behaviour of lightweight expanded polystyrene concrete containing silica fume. Cement and Concrete Research. 2003;33:755-62.

[8] Tang WC, Lo Y, Nadeem A. Mechanical and drying shrinkage properties of structural-graded polystyrene aggregate concrete. Cement and Concrete Composites. 2008;30:403-9.

[9] Bouvard D, Chaix JM, Dendievel R, Fazekas A, Létang JM, Peix G, et al. Characterization and simulation of microstructure and properties of EPS lightweight concrete. Cement and Concrete Research. 2007;37:1666-73.

[10] Schackow A, Effting C, Folgueras MV, Güths S, Mendes GA. Mechanical and thermal properties of lightweight concretes with vermiculite and EPS using air-entraining agent. Construction and Building Materials. 2014;57:190-7.

[11] Torres ML, García-Ruiz PA. Lightweight pozzolanic materials used in mortars: Evaluation of their influence on density, mechanical strength and water absorption. Cement and Concrete Composites. 2009;31:114-9.

[12] Ferrándiz-Mas V, García-Alcocel E. Physical and Mechanical characterization of Portland cement mortars made with expanded polystirene particles addition (EPS). Materiales de Construcción. 2012;62:547.

[13] Ferrándiz-Mas V, García-Alcocel E. Durability of expanded polystyrene mortars. Construction and Building Materials. 2013;46:175-82.

[14] Ferrándiz-Mas V, Bond T, García-Alcocel E, Cheeseman CR. Lightweight mortars containing expanded polystyrene and paper sludge ash. Construction and Building Materials. 2014;61:285-92.

[15] de Oliveira LAP, Gomes JPC, Nepomuceno MCS. THE INFLUENCE OF WASTES MATERIALS ON THE RHEOLOGY OF RENDERING MORTARS. Appl Rheol. 2013;23:11.

[16] Ferrándiz-Mas V. "Design of cement mortars with addition of polymeric waste expanded polystyrene (EPS) ". Doctoral Thesis. Research in Sustainable Architecture and Urban Planning Program. University of Alicante, Alicante: Universidad de Alicante; 2013.

[17] Keleştemur O, Yildiz S, Gökçer B, Arici E. Statistical analysis for freeze-thaw resistance of cement mortars containing marble dust and glass fiber. Materials & Design. 2014;60:548-55.
[18] Zaitri R, Bederina M, Bouziani T, Makhloufi Z, Hadjoudja M. Development of high performances concrete based on the addition of grinded dune sand and limestone rock using

the mixture design modelling approach. Construction and Building Materials. 2014;60:8-16. [19] EN 196-1:2005. Methods of testing cement. Part 1: Determination of strength.

[20] BS EN 1015-1:1999. Methods of test for mortar for masonry. Determination of particle size distribution (by sieve analysis).

[21] EN 1015-3:2007. Methods of test for mortar for masonry. Part 3: Determination of consistence of fresh mortar (by flow table).

[22] EN 1015-2 "Methods of test for mortar for masonry. Parte 2: Bulk sampling of mortars and preparation of test mortars".

[23] Lewis D, Mathieu R, Phan-Tan-Luu R. Pharmaceutical Experimental Design. New York: Marcel-Dekker; 1999.

[24] Phan-Than-Luu M, Sergent M. Nonclassical Experimental Designs. In: Brown SD, Tauler R, Walczak B, editors. Comprehensive Chemometrics: Elsevier; 2009. p. 453-99.

[25] Mathieu J, Nony J, Phan Tan Luu R. NemrodW v2007-03, LPRAI, Marseille, France. 2007.[26] EN 1015-7:1999. Methods of test for mortar for masonry. Determination of air content of fresh mortar.

[27] EN 1015-10:1999. Methods of test for mortar for masonry. Determination of dry bulk density of hardened mortar. 1999.

[28] EN 1015-12:2000. Methods of test for mortar for masonry. Determination of adhesive strength of hardened rendering and plastering mortars on substrates.

[29] EN 1015-18:2003. Methods of test for mortar for masonry. Determination of water absorption coefficient due to capillary action of hardened mortar.

[30] EN 998-1:2003 "Specification for mortar for masonry. Part 1: Rendering and plastering mortar".

[31] EN 998-2:2001 "Specification for mortar for masonry. Part 2: Masonry mortar".

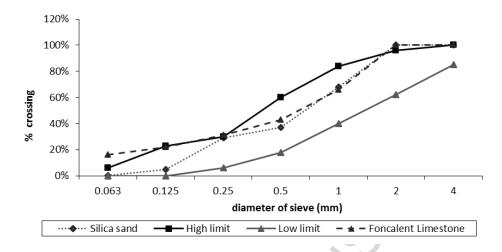


Figure 1 Grain size for the sand used to manufacture the mortars, determined according EN 1015-1:1999

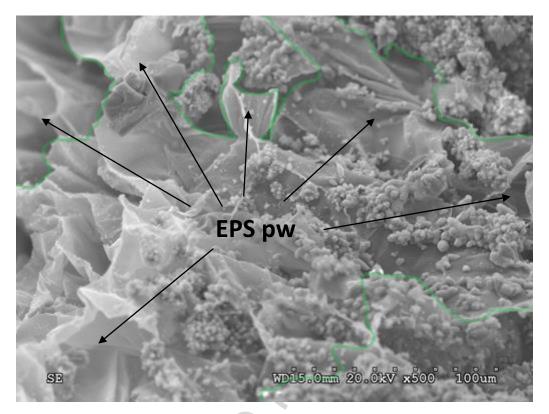


Figure 2a Mortar containing waste powdered EPS (EPS pw), the arrows indicate where the EPS particles are located

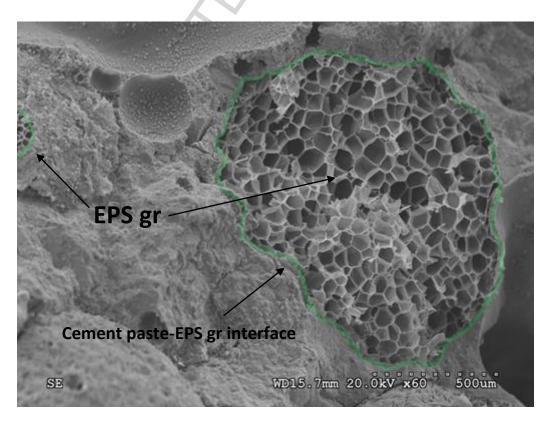
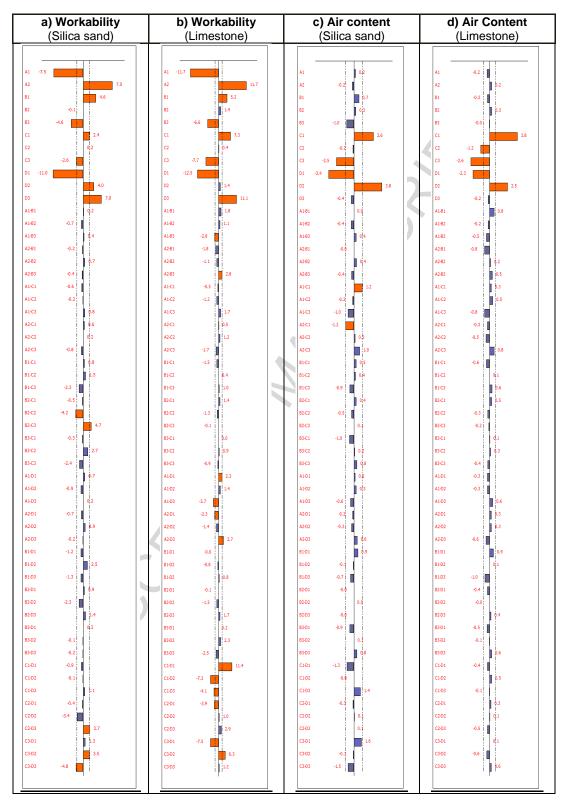
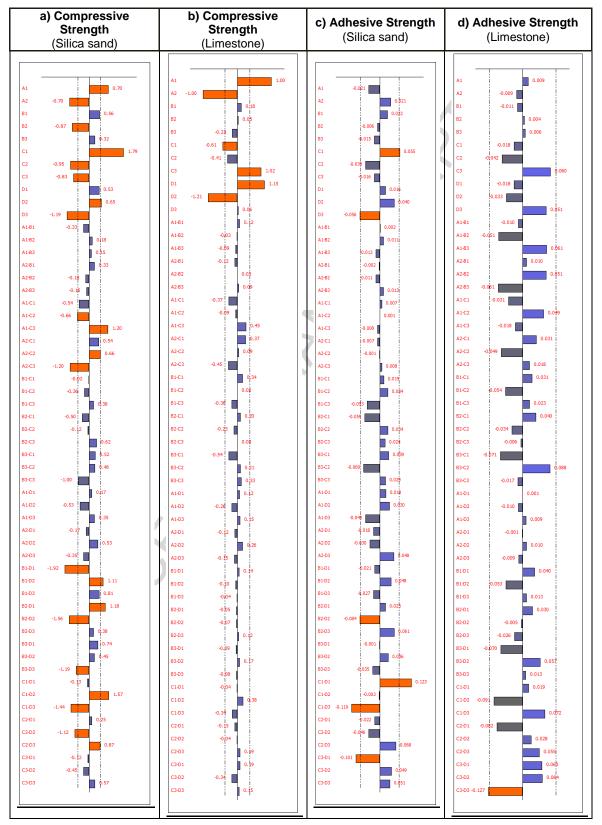


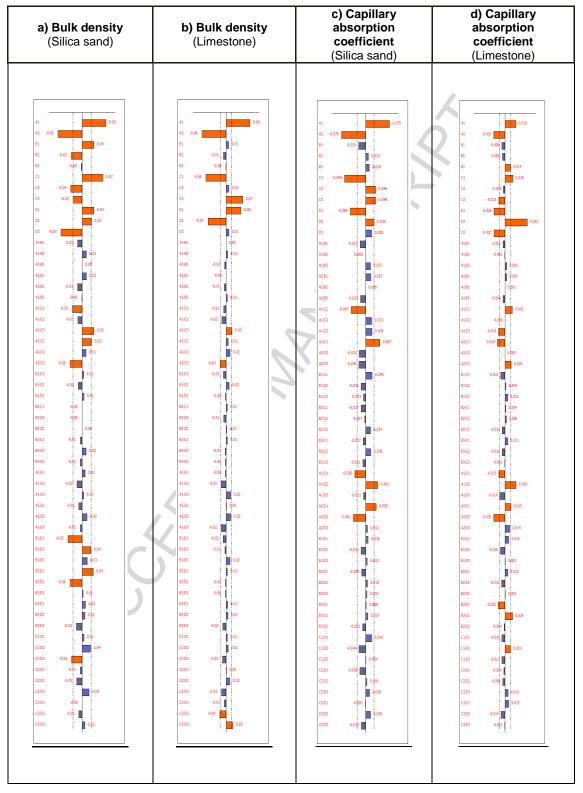
Figure 2b Mortar containing waste ground EPS (EPS gr), the arrows indicate where the EPS particles and the cement paste-EPS interface are located



**Figure 3** Graphic analysis of the effects of the studied experimental factors on the response for workability and air content for mortars made with silica sand and limestone sand. Light orange bars are for significant coefficients (5% significant level); dark blue bars are for the non-significant ones (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article)



**Figure 4** Graphic analysis of the effects of the studied experimental factors on the response for compressive strength (28 days curing time) and adhesive strength for mortars made with silica sand and limestone sand. Light orange bars are for significant coefficients (5% significant level); dark blue bars are for the non-significant ones (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article)



**Figure 5** Graphic analysis of the effects of the studied experimental factors on the response for bulk density (28 days curing time) and the capillary absorption coefficient for mortars made with silica sand and limestone sand. Light orange bars are for significant coefficients (5% significant level); dark blue bars are for the non-significant ones (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article)

%	CaO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	SO <sub>3</sub>	MgO	K <sub>2</sub> O	Na <sub>2</sub> O	TiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	MnO
CEM I	62.05	17.95	4.25	3.21	3.67	2.98	0.78	0.31	0.24	-	-
CEM II/A-S	58.06	20.32	4.56	3.77	3.66	3.06	0.71	0.91	0.40	0.14	0.11
CEM III/A	53.28	23.73	6.33	2.84	3.45	4.42	0.53	0.63	0.40	0.17	0.23

Table 1 Chemical composition for the cement types used to manufacture the mortars, expressed as major oxides determined by X-Ray fluorescence microscopy (XRF)

Factor	Number of Levels	Levels	
EPS type	2	A1	EPS powdered (EPS pw)
	2	A2	EPS ground (EPS gr)
		B1	40
EPS addition content	3	B2	50
		B3	60
		C1	0.8A/0.1R/0.8S/6V
Admixture mix	3	C2	0.4A/0.1R/0.5S/6V
		C3	0.3A/0.1R/0.4S/6V
		D1	CEM I
Cement type	3	D2	CEM II
		D3	CEM III

Table 2 Factors and levels chosen for the proposal experimental design

<section-header>

Nº experience	EPS type	EPS content (v/v%)	Admixtures mix (w/w%)	Cement type
1	EPS pw	40	0.8A/0.1R/0.8S/6V	CEM I
2	EPS gr	40	0.8A/0.1R/0.8S/6V	CEM I
4	EPS gr	50	0.8A/0.1R/0.8S/6V	CEM I
5	EPS pw	60	0.8A/0.1R/0.8S/6V	CEM I
6	EPS gr	60	0.8A/0.1R/0.8S/6V	CEM I
8	EPS gr	40	0.4A/0.1R/0.5S/6V	CEM I
9	EPS pw	50	0.4A/0.1R/0.5S/6V	CEM I
10	EPS gr	50	0.4A/0.1R/0.5S/6V	CEM I
11	EPS pw	60	0.4A/0.1R/0.5S/6V	CEM I
13	EPS pw	40	0.3A/0.1R/0.4S/6V	CEM I
15	EPS pw	50	0.3A/0.1R/0.4S/6V	CEM I
16	EPS gr	50	0.3A/0.1R/0.4S/6V	CEM I
18	EPS gr	60	0.3A/0.1R/0.4S/6V	CEM I
20	EPS gr	40	0.8A/0.1R/0.8S/6V	CEM II
21	EPS pw	50	0.8A/0.1R/0.8S/6V	CEM II
22	EPS gr	50	0.8A/0.1R/0.8S/6V	CEM II
23	EPS pw	60	0.8A/0.1R/0.8S/6V	CEM II
25	EPS pw	40	0.4A/0.1R/0.5S/6V	CEM II
28	EPS gr	50	0.4A/0.1R/0.5S/6V	CEM II
29	EPS pw	60	0.4A/0.1R/0.5S/6V	CEM II
30	EPS gr	60	0.4A/0.1R/0.5S/6V	CEM II
31	EPS pw	40	0.3A/0.1R/0.4S/6V	CEM II
32	EPS gr	40	0.3A/0.1R/0.4S/6V	CEM II
33	EPS pw	50	0.3A/0.1R/0.4S/6V	CEM II
36	EPS gr	60	0.3A/0.1R/0.4S/6V	CEM II
37	EPS pw	40	0.8A/0.1R/0.8S/6V	CEM III
39	EPS pw	50	0.8A/0.1R/0.8S/6V	CEM III
40	EPS gr	50	0.8A/0.1R/0.8S/6V	CEM III
42	EPS gr	60	0.8A/0.1R/0.8S/6V	CEM III
43	EPS pw	40	0.4A/0.1R/0.5S/6V	CEM III
44	EPS gr	40	0.4A/0.1R/0.5S/6V	CEM III
45	EPS pw	50	0.4A/0.1R/0.5S/6V	CEM III
48	EPS gr	60	0.4A/0.1R/0.5S/6V	CEM III
50	EPS gr	40	0.3A/0.1R/0.4S/6V	CEM III
52	EPS gr	50	0.3A/0.1R/0.4S/6V	CEM III
53	EPS pw	60	0.3A/0.1R/0.4S/6V	CEM III

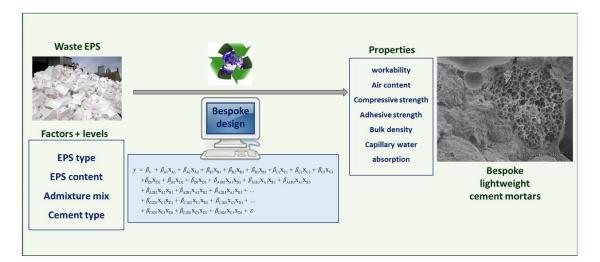
#### Table 3 Experimental plan

Note: EPS in addition of sand, additives in addition of binder A = air-entraining agent (BASF Rheomix 934) R = water retaining additive (Hydroxypropyl methylcellulose TER CELL HPMC 15 MS PF) S = superplastizicer (BASF Rheomix GT 205 MA) V = dispersible polymer (VINNAPAS 5028E)

	Workability		Air content		Compressive Strength		Adhesive Strength		Bulk density		C <sub>90</sub>	
	Silica	Limestone	Silica	Limestone	Silica	Limestone	Silica	Limestone	Silica	Limestone	Silica	Limestone
p- value	1.35·10 <sup>-3</sup>	<1.00.10-4	5.68·10 <sup>-3</sup>	6.51·10 <sup>-4</sup>	1.73·10 <sup>-3</sup>	1.40·10 <sup>-2</sup>	0.10	0.48	4.01·10 <sup>-3</sup>	2.56·10 <sup>-4</sup>	4.93·10 <sup>-3</sup>	8.51·10 <sup>-4</sup>
S <sub>yx</sub>	4.0 mm	4.0 mm	2.0%	1.5%	1.1 MPa	1.0 MPa	0.10 MPa	0.13 MPa	100 kg/m <sup>3</sup>	100 kg/m <sup>3</sup>	0.07 Kg/m²⋅min <sup>0.5</sup>	0.02 Kg/m <sup>2</sup> ·min <sup>0.5</sup>
R <sup>2</sup>	0.968	0.988	0.925	0.955	0.944	0.908	0.844	0.729	0.932	0.963	0.928	0.952

Table 4 p-value, standard deviation ( $S_{yx}$ ) and coefficient of determination ( $R^2$ ), obtained by the model for workability, air content, compressive strength, adhesive strength, bulk density and capillary absorption coefficient ( $C_{90}$ ) for mortars made with silica sand and limestone sand

0.944 0.908



V. Ferrándiz-Mas\*, L.A. Sarabia, M.C. Ortiz, C.R. Cheeseman, E. García-Alcocel

Design of bespoke lightweight cement mortars containing waste expanded polystyrene by experimental statistical methods

#### **Graphical Abstract**

- D-optimal criterion reduced the experiments needed to design lightweight mortars
- Workability increased by using ground waste expanded polystyrene (EPS)
- EPS content had no significant impact on compressive strength of limestone mortars •
- Ground EPS mortars had lower C<sub>90</sub> than powdered EPS mortars, thus better durability