Performance and durability properties of self-compacting mortars with electric arc furnace dust as filler

A. Lozano-Lunar[1], Pedro R. da Silva[2], Jorge de Brito[3], J.I. Álvarez[4], J. M. Fernández [5,+,*] and J. R. Jiménez [1,+,*]

[1] Construction Engineering Area, School of Engineering Science of Belmez, Universidad de Córdoba, Córdoba, Spain.

[2] CERIS, Instituto Superior de Engenharia de Lisboa (ISEL - IPL), R. Conselheiro Emídio Navarro, 1, 1959-001 Lisbon, Portugal

[3] CERIS, Instituto Superior Técnico, Universidade de Lisboa, Av. Rovisco Pais, 1049-001 Lisbon, Portugal.

[4] MIMED Research Group, Department of Chemistry, School of Sciences, University of Navarra, Pamplona, Spain

[5] Department of Inorganic Chemistry and Chemical Engineering, School of Engineering Science of Belmez, Universidad de Córdoba, Spain.

[+]Both authors equally contributed to the paper.

[*]Corresponding authors at: Construction Engineering Area (José Ramón Jiménez), Department of Inorganic Chemistry and Chemical Engineering (José María Fernández). E.P.S. of Belmez, Universidad de Córdoba, Avda. de la Universidad s/n, 14240, Belmez (Córdoba). Spain.

E-mail addresses: jrjimenez@uco.es (J.R. Jiménez), um1feroj@uco.es (J.M. Fernández).

Abstract

This study analyses the feasibility of incorporating electric arc furnace dust (EAFD) in the production of self-compacting mortars from the point of view of durability. Two types of EAFD, previously characterized and classified as hazardous waste, were used. Siliceous filler (powder) was replaced at different ratios (25%, 50%, and 100%, by volume). The following self-compacting mortar properties were measured at different curing times: total heat of hydration, compressive strength, ultrasonic pulse velocity, pore size distribution through mercury intrusion porosimetry, shrinkage and mass loss, water absorption by capillary, water absorption capacity, bulk density, skeletal density, open porosity for water, wettability by contact angle measurement, electrical resistivity, and carbonation depth. Although some of properties linked to the durability of the mortars, such as water absorption by capillarity, wettability or carbonation, were negatively affected by the incorporation of the EAFD, the results show the feasibility of replacing siliceous filler at up to 25% and the valorisation of EAFD as a secondary raw material (SRM). This research demonstrated that EAFD can be used as SRM as an alternative to landfills, thereby contributing to the promotion of the new circular economic model pursued by the European Union.

Keywords:

Electric arc furnace dust; industrial waste; secondary raw material; self-compacting mortars; durability.

1. Introduction

In the search for construction materials that are more cost-competitive than conventional ones, self-compacting concrete (SCC) has been one of the most important progresses in the construction industry. SCC is composed of the same materials (cement, water, aggregates, additives, and mineral additions) as conventional concrete (CC). However, SCC contains greater amounts of cement and mineral additions, as well as smaller coarse aggregates. Changes in the composition modify its behaviour in the fresh and hardened state (Aslani and Nejadi, 2012), therefore, SCC offers advantages over CC, as its placement and compaction can be carried out without vibration energy, segregation, blockage of coarse aggregates, bleeding, or paste exudation, resulting in better surface quality (EFNARC, 2005). Additionally, the increased content of mineral additions results in greater packing of the particles, which allows higher flexural and compressive strengths (Persson, 2001) and superior durability properties (Assié et al., 2007) of SCC relative to CC. Owing to such numerous advantages of SCC, its use has become widespread in recent years in civil works, construction, and the precast industry.

Additionally, Billberg (1999) argues that concrete rheology can only be optimized if the mortar phase is designed for optimum rheological performance, therefore, SCC's performance is generally defined by the behaviour of the self-compacting mortar phase (SCM). Nepomuceno et al. (2012) and da Silva and de Brito (2015) evaluated the SCM phase to predict SCC's properties.

The use of concrete as a construction material is still associated with strongly negative environmental effects (high consumption of natural raw materials, CO_2 emission, large production of waste, etc.) that should be reduced as much as possible (Flatt et al., 2012) to make it a more sustainable material (Weeks et al., 2008).

SCC is more expensive than CC. To overcome this restriction, supplementary materials are used as a partial replacement for fine materials in order to reduce the cost and improve the durability of concrete (Sharma and Khan, 2017). Assié et al. (2007) performed a durability comparative study between a SCC and a CC of equal compressive strength. Despite the higher w/c ratio of SCC, properties such as chloride diffusion, water absorption by capillarity, deterioration against carbon dioxide and ammonium nitrate were equivalent in SCC and CC, whereas SCC was more resistant to oxygen permeation.

The use of waste as secondary raw material (SRM) in different production processes is a new paradigm pursued by the European Union with the aim of implementing the circular economy model (COM(2017)33, 2017). In this context, researchers such as Singh and Siddique (2016), Sharma and Khan (2017), and Si et al. (2018) studied the replacement of natural aggregates with wastes such as iron slag, copper slag, and rubber, respectively. All these researchers pointed out the viability of the partial replacement of wastes as SRM in SCC production.

Spain occupies the third position in European steel production. More than 75% of the steel manufactured is obtained through recycling of scrap as a raw material in electric arc furnaces (Unesid, 2013). One of the wastes generated during the scrap fusion is electric arc furnace dust (EAFD). For each tonne of steel produced, around 15-25 kg of EAFD is generated (Sayadi and Hesami, 2017). This waste is formed by metal volatilization (Zn, Pb, Fe, or Cd) during scrap fusion and subsequent reaction with oxygen and condensation into solid particles. The heavy metal content in EAFD can reach up to 25% by weight in steel operations based on scrap fusion; therefore, the European Waste Catalogue - EWC (2002) classifies it as hazardous waste. Owing to the high pollution risk and its fineness, handling of EAFD is difficult; hence, the most usual and quick way to manage it is to deposit it in landfills after treatment using cement-based materials (Ledesma et al., 2018).

The valuable metals lost, the amount of natural resources (in terms of natural sand, filler, cement, etc.) needed, and the volume of non-useful materials generated through the treatment of EAFD are too high; consequently, landfilling is not an environmentally sustainable option as it is not consistent with the circular economy model.

The use of EAFD to prevent its landfilling has been studied by numerous researchers. Alsheyab and Khedaywi (2013) analysed the possibility of using EAFD as an additive in asphalt concrete mixes for road construction. These authors concluded that the mixes revealed adequate properties for road construction based on the penetration index values obtained. Mymrin et al. (2016) stated that EAFD's use led to ceramic materials with three times the mechanical properties and improved their structures relative to conventional ones; this behaviour was attributed to the very low melting point of the heavy metals. Fares et al. (2016) studied the optimal replacement of cement with EAFD and compared their performance with various supplementary cementing materials. They observed that 3% of EAFD was the maximum content that can partially replace cement with a water/binder ratio of 0.5, showing good performance with respect to properties such as compressive strength or chloride ion penetration. Alsheyab and Khedaywi (2013), Fares et al. (2016) and Mymrin et al. (2016) highlighted the importance of waste management through its use in the construction industry. Although there have been studies on the incorporation of EAFD in construction materials, the development of SCM with EAFD has been ignored until now.

The main objective of this research is to manage and valorise EAFD as SRM to obtain a SCM with a performance comparable to that of mortar produced with conventional aggregates. Particular attention is devoted to properties connected with the applicability and long-term durability behaviour of the new SCM that ensure its safe and feasible use. Up to now, EAFD has been managed through landfilling; therefore, the novelty of this study is the use of two EAFDs as replacement of siliceous filler (SF) in the production of SCM. The several properties studied allowed determining the maximum percentage of EAFD that could be incorporated. This research helps preventing the deposition of EAFD in landfills and reintroduces it into the construction industry as SRM, which is in line with the new circular economy model of the European Union.

2. Materials and methods

2.1. Characterization of mortar component materials

Two EAFD wastes (EAFD1 and EAFD2) were selected from two steelworks located in Guipúzcoa (Spain). The wastes' composition was determined by wavelength dispersion X-ray fluorescence spectrometry (WD XRF) with a power of 4 kW and S4PIONEER equipment. The EAFDs' composition shows that the major oxides were ZnO (38.3% and 34.2%) and Fe_2O_3 (29.9% and 31.8%), respectively, for both wastes (Figure 1).

The main mineralogical phases of the EAFDs were determined by X-ray diffraction (XRD). The equipment used was a Bruker D8 Discover A25 with Cu-Kα radiation and a goniometric scan from 10° to 70° (2-theta) at the speed of 0.00625° min^{-1}/step was performed. The mineralogical identification was carried out by using the database of Joint Committee on Power Diffraction Standard-International - JCPD (1995). Both wastes showed the same mineralogical phases (Table 1): Franklinite ($ZnFe_2O_4$; 22-1012), zincite (ZnO; 36-1451), metallic Mn (33-0887), manganese oxide (MnO; 07-0230), quartz (SiO_2; 33-1161), and lead oxide (PbO_2; 37-0517) (JCPD, 1995).

Cement type CEMI 42.5R and SF were used. These materials were named CEM and SF and were supplied by SECIL Company (Lisbon, Portugal) and Minas Carmina Palau Saverdera (Gerona, Spain), respectively. These suppliers also provided the CEM and SF compositions. The aggregates used were two natural sands with size ranges 0/2 mm and

0/4 mm (NS-0/2 and NS-0/4, respectively), and their composition was determined by an energy dispersive X-ray analyser coupled with a scanning electron microscope JEOL JSM-6300 by using 20 kV acceleration potential and 15 mm distance. The chemical composition of CEM, SF, NS-0/2, and NS-0/4 is shown in Figure 2. Calcium oxide (CaO) was the main component of the CEM, which contained lower percentages of quartz, iron oxide (Fe_2O_3), and sulphur oxide (SO_3). SF was 100% quartz. The main component of both sands was quartz and other minor components such as aluminium oxide (Al_2O_3), potassium oxide (K_2O), and iron oxide (Fe_2O_3) were also present. BASF company provided the superplasticizer additive MasterEase 5025 (Sp), which is a high-range water reducer based on Poly-Aryl-Ethers polymers.

The specific surface areas of EAFD1, EAFD2, and SF were analysed by the Brunauer-Emmett-Teller method (BET) with Micromeritics ASAP 2010 equipment. The values were 3.70 m^2/g, 4.60 m^2/g, and 0.25 m^2/g, for EAFD1, EAFD2, and SF, respectively. SECIL Company provided the specific surface area of CEM (0.35 m^2/g).

The real particle densities of EAFD1, EAFD2, SF, and CEM, determined in accordance with UNE 80103:2013 (AENOR, 2017), were 3.84 g/cm^3, 3.81 g/cm^3, 2.60 g/cm^3, and 3.14 g/cm^3, respectively. Regarding the aggregates, their real particle density and water absorption were calculated according to UNE-EN 1097-6:2014 (AENOR, 2017) as 2.54 g/cm^3 and 0.41% for NS-0/2 and 2.62 g/cm^3 and 0.46% for NS-0/4, respectively.

Figure 3 shows the particle size distributions of EAFD, CEM, and SF measured with a Mastersizer S (Malvern Instruments) device, with ethanol as the dispersant of the samples. The particle sizes of CEM and SF ranged between 0.06 μm and 100 μm, with a higher percentage of particles sized around 20 μm present in both materials. Both EAFD1 and EAFD2 showed bimodal particle size distributions, with a maximum size concentrated around 0.3 μm for both wastes and another one at 3 μm for EAFD1 and 2.5 μm for EAFD2.

The particle size distribution of the sands was tested in accordance with UNE-EN 933-1:2012 (AENOR, 2017). Table 2 shows the particle size distribution of NS-0/2 and NS-0/4 in terms of mass. The maximum size was 2 mm and 4 mm for NS-0/2 and NS-0/4, respectively.

2.2. Mortar design

The SCMs were designed by the Nepomuceno method (2005). This method classifies the cement and mineral additions as "powders" and sand as “fine aggregates”. Consequently, this research establishes CEM, SF, EAFD1, and EAFD2 as "powders" and NS-0/2 and NS-0/4 as “fine aggregates”. The Nepomuceno method was carried out using a ratio of the absolute volumes of the powder materials and fine aggregates (V_p/V_s) of 0.70. A reference mortar (SCRM) was mixed with 50% SF and 50% CEM (by volume). Then, SF was replaced with EAFD using volume replacement ratios (f_{Ad}) of 25%, 50%, and 100% to prepare SCMs with waste. These mortars were named SCM-EAFDi-j, where i is the EAFD type (i = 1, 2) and j is the percentage replacement of SF with EAFD (j = 25, 50, 100). Self-compactability was controlled, as outlined by EFNARC (2005), through the relative spread area (G_m). The process consisted of iteratively adjusting the ratio, in absolute volume, between water and the powder materials (V_w/V_p) and the ratio, in percentage, between the amounts of superplasticizer and powder materials (Sp/p%). G_m was calculated according to equation (1) proposed by EFNARC (2005) and Nepomuceno et al. (2012). D_m was defined as the slump-flow average of the recorded diameters and the values were made to be within the range 270 ± 30 mm, which is suitable for a SCM (Kanadasan et al., 2018). D_0 was defined as the initial diameter of the base of the used cone.

$$G_m = \left(\frac{Dm}{D_0}\right)^2 - 1 \tag{1}$$

Table 3 shows the nomenclature, mix proportions (kg/m^3 of mortar), composition percentages by volume (%) of the SCMs, water/cement ratio (w/c), EAFD/cement ratio, and self-compactability parameters (V_w/V_p, Sp/p%, and G_m) of the mortars. All dry materials were mixed for 30 s without water. Then, 80% of the water was added and mixed for 2.5 min. Finally, the remaining 20% of the water with the diluted Sp was added. This paste was then mixed for 1 additional minute.

V_w/V_p and Sp/p% were fixed at 0.84 and 0.60, respectively, in SCRM. When EAFD was incorporated in the mortars, the water and Sp amounts were higher (Table 3), owing to the EAFD having a finer particle size distribution (Figure 3) and larger specific surface areas (3.70 m^2/g and 4.60 m^2/g for EAFD1 and EAFD2, respectively) than the replaced SF (0.25 m^2/g).

In SCM-EAFD1, V_w/V_p and Sp/p% increased slightly relative to SCRM. However, the use of EAFD2 in mortars caused the auto-compactability parameters V_w/V_p and Sp/p% to increase to 0.98, 1.12, and 1.40 for V_w/V_p, and 0.70, 0.80, and 1.00 for Sp/p% in the cases of 25%, 50%, and 100%, respectively. This phenomenon could be attributed to the differences in the particle size distribution of the two EAFDs. In Figure 3, the highest percentage volume of the particles was observed in the 0.05-1 μm range for EAFD2 (50.9% in EAFD2 *versus* 41.5% in EAFD1). The greater number of fine particles in EAFD2 may have caused a larger surface to hydrate. The higher EAFD2 specific surface area compared to that of EAFD1 may also have encouraged this behaviour.

The G_m values decreased when the EAFD content increased even though V_w/V_p and Sp/p% increased. The G_m values recorded were considered to be acceptable because they were within the range 270 ± 30 mm. In addition, the mixes did not visually show aggregate segregation or water exudation (Table 3).

For each mix, 15 prismatic specimens (40 mm × 40 mm × 160 mm) were prepared. The specimens were stored in a climatic chamber (95 ± 5% relative humidity and 20 ± 2 ºC temperature) for 24 h. Then, the specimens were demoulded and cured under the following conditions: 95 ± 5% of relative humidity and 20 ± 2 ºC of temperature. However, SCM-EAFD2 mortars could not be demoulded after 24 h due to the greater amount of water in them (Table 3). SCM-EAFD2-25 was demoulded after 3 days of ageing. SCM-EAFD2-50 and SCM-EAFD2-100 were demoulded after 6 days.

2.3. Mortar characterization

A study of the total heat of hydration by isothermal calorimetry was carried out to analyse the influence of EAFD in the hydration of cement. The evolution curves of the heats of mortars were determined by using a TAM Air conduction calorimeter for 15 days at 25 ºC. This test was carried out in SCRM, with 25% and 100% replacements of SF with EAFD.

The compressive strengths of the mortars were evaluated according to UNE-EN 1015-11:2000 (AENOR, 2017) after 7, 28, and 56 days by using three specimens for each age tested. The values were expressed as an average of the three replicates. Electron microscopy (SEM) was carried out on some mortars to show how the EAFD addition influenced the microstructure of hydrated phases. This technique was performance by scanning electron microscope JEOL JMS-7800.

The ultrasonic pulse velocity (UPV) was calculated according to UNE-EN 12504-4:2006 (AENOR, 2017) on three specimens. The measurement was made directly, by placing probes on opposite sides of the specimen.

The values of mercury intrusion porosimetry (MIP) were obtained using a Micromeritics AutoPore IV 9500 equipment at the pressure range 0.0015 to 207 MPa. This test was used to analyse the volume of pores and its distribution. It was carried out on cut off samples of volume 1 cm^3 that were extracted from the central area of the mortar specimens after 91 days. Before cutting the cubes, the specimens were dried in an oven at 100 °C until the mass became constant. The MIP values were recorded for SCRM and SCM-EAFD1-25, SCM-EAFD1-100, SCM-EAFD2-25, and SCM-EAFD2-100.

The shrinkage and mass loss of mortars were determined by measuring as a function of time the variation in the length and weight of the three specimens immediately after demoulding, These specimens were cured in conditions of 65 ± 5% relative humidity and 20 ± 2 ºC temperature in accordance with UNE 83831:2010 EX (AENOR, 2017).

The water absorption by capillarity coefficient was determined on three specimens in accordance with UNE-EN 1015-18:2003 (AENOR, 2017).

Additionally, the water absorption capacity was measured according to UNE 83980:2014 (AENOR, 2017) on three specimens. This procedure yielded the amount of water absorbed after immersion until mass became constant (without saturation) and the amount of water absorbed after immersion and vacuuming until mass became constant (with saturation). In accordance with UNE 83980:2014 (AENOR, 2017), the bulk and skeletal densities and the accessible porosity for water of three specimens were determined. Bulk density refers to the relationship between the mass of the specimen and its apparent volume (i.e. the volume limited by the outer surface of the specimen, including any pores). Skeletal density, on the other hand, refers to the relation between the mass of the specimen and its solid volume (including inaccessible pores). Accessible porosity for water refers to the ratio between the volume of pores accessible to water and the apparent volume of the specimen.

Wettability is the ability of a surface to be moistened by a liquid (Di Mundo et al., 2018). This property was determined in the mortars by measuring the contact angle (CA) between a drop of water and its solid surface over time. The contact angle was measured in test pieces of 1 cm^3 extracted from the centre of the mortar specimen, on a plane surface area. A device for contact angle determination OCA 15 EC (Dataphysics) was used including an electronic syringe unit ESr-N and the software module SCA 20.

The electrical resistivity of the three specimens was determined according to technical recommendation RILEM TC 154-EMC (Polder et al., 2000). The method involved the application of 60 V (direct current) to the specimen and the measurement of the reading in milliamperes (mA) of the current produced between the two measuring electrodes placed in parallel in contact with the two opposite faces of the specimen.

The three specimens for the carbonation depth test were previously cured up to 21 days of age at 95 ± 5% relative humidity and 20 ± 2 ºC temperature. Then, they were stored in a dry chamber at 60 ± 5% relative humidity and 20 ± 2 ºC temperature up to 28 days of age. After this period, the specimens were introduced in a carbonation chamber under the conditions of relative humidity 55-65%, temperature 23 ± 3 ºC, and CO_2 concentration 5% ± 0.1%. The depth of carbonation was measured after 7, 14, 28, and 56 days of exposure in the carbonation chamber in accordance with UNE-EN 13295:2005 (AENOR, 2017) after the spraying with phenolphthalein pH indicator. The non-carbonated area showed a purple-red colour in agreement with its high alkaline pH, whereas the carbonated area showed no colour modification due to its lower pH.

In a previous research on self-compacting mortars containing EAFD as a substitute for SF, the leaching behaviour in monolithic and granular state was evaluated (Lozano-Lunar et al., 2019). The environmental classification of the self-compacting mortars, according to their leaching behaviour, was compared to the established "Non-hazardous" limit of the European Directive 2003/33/EC (2003). In the monolithic state, all self-compacting mortars registered metal release below the "Non-hazardous" limit. In the granular state, the mortar with 25% EAFD2 instead of SF was environmentally safe, keeping all the elements encapsulated due to the self-compacting matrix. In any case, the main objective of this research is the study of performance and durability properties of self-compacting mortars with electric arc furnace dust as filler.

3. Results and discussion

3.1. Total heat of hydration

Total heat of hydration can be ascribed to the heat release owing to the formation of hydrated cement compounds. The assessment of the effect of the EAFD's on the heat of hydration allowed us to know if these waste materials substantially interfered with the normal hydration of the cement. Figure 4 shows the cumulative total heat of hydration of the mortars over time (360 h) after the isothermal calorimetry test. It is observed that the highest total heat production was achieved for SCRM (70.93 J/g).

SCM-EAFD1-25 exhibited total heat values of 44.47 J/g. It was recorded that SCM-EAFD1-25 showed total heat values of around 27 J/g after 7 days (168 h), even though an upward change in the slope of the curve was observed over a greater heat range. This change in the slope suggests a delay in the hydration of cement associated with a possible heat generation. This attenuated slope can be correlated with a longer induction period and with a slow heat release during the hydration of the compounds, which are indicative of a delay in the hydration. The SCM-EAFD1-100 mortar showed a curve in which the upward progression in the total heat production was not observed over time. Therefore, the hydration of cement in this mortar was not sufficient to be considered as a cementitious matrix and its use in SCM is not recommended since the heat curve of hydration not evolved as for the rest of mortars.

SCM-EAFD2-25 showed a total heat evolution curve with a similar tendency to that of SCRM, reaching a value of 66.70 J/g after 360 h. SCM-EAFD2-100 showed a steeper slope, with values of 70.83 J/g after 360 h. This suggests that the hydration of the mortars with EAFD2 was approximately equal to that of SCRM. Weeks et al. (2008) observed a similar trend in mortars with Zn production slag as a substitute of natural sand. These researchers obtained total heat values similar to those achieved by their reference mortar, with values between 100 and 120 J/g for all the mortars, except in the case of 20% replacement, which was attributed to a measurement error during the test.

3.2. Compressive strength

The compressive strength of the SCMs is shown in Table 4. It increased for all the SCMs with curing time. The corresponding values for SCRM reached 48.3 MPa, 62.7 MPa, and 66.2 MPa after 7, 28, and 56 days, respectively.

After 7 days, SCM-EAFD1 exhibited very low compressive strengths (1.0 MPa, 3.3 MPa, and 4.6 MPa for SCM-EAFD1-25, SCM-EAFD1-50, and SCM-EAFD1-100, respectively) compared to those of SCRM. This mechanical behaviour cannot be related to the w/c ratio because the variation was not significant when EAFD1 was incorporated in the mixes (Table 3). However, SCM-EAFD1-25 gained compressive strength with values of 22.4 MPa and 48.2 MPa after 28 and 56 days due to a delay in the cement hydration. This delay in cementitious mixes with EAFD has been related to the high concentration of Zn in this waste (Figure 1) that promotes the formation of a protective cover on the cement particles, which prevents common cement hydration (Balderas et al., 2001; de Souza et al., 2010; Fares et al., 2016). Despite the similar composition, the wastes registered clearly differentiated behaviour when incorporated in self-compacting mortars for the same percentage of replacement and that may be related to small differences in their composition.

The different heat of hydration behaviour and subsequent compressive strengths of mortars with EAFD1 and EAFD2 could be attributed to the different volume of fine particles in both EAFD (41.52% in EAFD1 *versus* 50.92% in EAFD2) and greater specific surface area of EAFD2 (3.70 m^2/g for EAFD1 and 4.60 m^2/g for EAFD2). Due to these physical characteristics, EAFD2 could improve the nucleation points enough to encourage the formation of hydration products (Moon et al., 2017; Thongsanitgarn et al., 2014).

The pH registered by EAFD1 (9.6) and EAFD2 (13.3) could play an important role in total heat of hydration and mechanical behaviour. The pH value of EAFD2, closest to the alkaline pH formed during the hydration of the cement (≈ 12), could favour normal hydration of cement and, therefore, compressive strengths closer to SCRM´s. The pH values registered in EAFD played an important role in cement-based materials performance the previous research (Ledesma et al., 2018).

The compressive strength behaviour of SCM-EAFD1-25 agreed with the release of its heat of hydration. After 7 days (168 h), the value of total heat of hydration was around of 27 J/g and, afterwards, it increased up to 44.47 J/g.

Researchers such as de Castellote et al. (2004) and de Vargas et al. (2006) also observed a gain in compressive strength over time, although EAFD caused a delay in the hydration of cement. Castellote et al. (2004) obtained a reference mortar with compressive strengths of 25 MPa and 38 MPa after 7 and 28 days, respectively, and the mortars with EAFD/cement ratio equal to 0.86 did not register compressive strength values until after 12 days, with values lower than 3 MPa. However, after 28 days, these mortars showed a value of approximately 35 MPa. De Vargas et al. (2006) added up to 25% EAFD to cementitious pastes. These researchers observed a gain in compressive strength of the 25% paste, registering a value of 1 MPa after 3 days and 42 MPa after 7 days, while the reference mortar had values of approximately 48 MPa and 50 MPa after 3 and 7 days, respectively. These researchers also carried out a setting time study of the cementitious pastes based on the hydration heat. They observed that the 25% paste did not release heat for 56 h, justifying the low compressive strength observed after 3 days.

The compressive strengths of the mortars with 50% and 100% EAFD1 did not exceed 7 MPa at any curing time. The higher content of heavy metals in SCM-EAFD1-50 and SCM-EAFD1-100 could promote a greater delay in the hydration of cement, thereby preventing any gain in the compressive strength before 56 days. Based on the results of compressive strength, the use of EAFD1 is recommended up to a replacement value of 25%.

The compressive strength of the mortars with EAFD2 was recorded because cement hydration in samples bearing EAFD2 was possible (Figure 4). It decreased relative to SCRM when the EAFD2 content increased. This could be associated with the greater contents of heavy metals such as Zn and Pb in SCM-EAFD2 and the higher w/c ratio of EAFD2 (Table 3). The behaviour of SCM-EAFD2 agreed with the results of Ledesma et al. (2017). These researchers investigated EAFD in conventional mortars with a filler and limestone aggregate by using cylindrical specimens and also experimentally determined that the compressive strengths decreased when the EAFD/cement ratio increased, as a consequence of the greater amounts of heavy metals in the mortar and higher w/c ratio.

Note that the compressive strength of SCM-EAFD2-25 reached a value of 65.3 MPa that was very close to that of SCRM (66.2 MPa) after 56 days, indicating the feasibility of using up to 25% EAFD2 with an insignificant loss of the compressive strength (1.4%).

The hydration microstructure of SCRM and SCM-EAFD1-100 was analyzed by SEM and mapping analysis of elemental composition (Figure 5). These samples corresponded to the mortars with extreme behaviour shown in Figure 4.

In SCRM (Figure 5 a)) Portlandite particles ($Ca(OH)_2$) were clearly observed according to the Ca atom mapping and the Si and Al absence in the same zones, which rules out that these particles are calcium silicates or aluminates. The SEM results for SCRM agreed with those obtained by Giraldo and Tobón (2006). In SCM-EAFD1-100 (Figure 5 b)), Portlandite ($Ca(OH)_2$) formation was not registered, related to its low heat of hydration (Figure 4) and compressive strength (Table 4), being in agreement with the mortar setting studies with EAFD carried out by Ledesma et al. (2017) and Lozano-Lunar et al. (2019).

3.3. Ultrasonic pulse velocity (UPV)

The UPV results of SCM-EAFD1 and SCM-EAFD2 after 7, 28, and 56 days of age are shown in Figures 6 and 7, respectively. The values of UPV are useful to monitor the intended level of packing of the mortar. In accordance with Whitehurst (1951), concrete with a density of approximately 2400 kg/m^3 can be classified as “very poor”, “poor”, “good”, and “excellent” based on UPV values of 0-2000 m/s, 2000-3500 m/s, 3500-4500 m/s, and above 4500 m/s, respectively. Since the performance of a SCC is generally defined by the behaviour of the mortar phase, the comparison between SCM and SCC is applicable. For this reason, the classification ranges of mortar quality are included in Figures 6 and 7.

As the curing time increased, the UPV values of all mortars also increased. The speed of the ultrasonic impulses for SCRM registered values of 5000 m/s, 5049 m/s, and 5097 m/s after 7, 28, and 56 days of age. The UPV value for SCRM indicates a mix of high compactness; therefore, it is expected that the UPV does not increase significantly with the increase in curing time (independently, the other properties show a more significant change). Şahmaran et al. (2006) obtained similar values (4687 m/s and 4813 m/s after 28 and 56 days, respectively) to those obtained in this study for their reference SCM fabricated with cement CEMI/42.5R and river sand.

Figure 6 shows that SCM-EAFD1-25 decreased in UPV value (2079 m/s) with respect to SCRM (5000 m/s) after 7 days. Subsequently, SCM-EAFD1-25 experienced increases to 3856 m/s and 4211 m/s after 28 and 56 days of age. After 56 days, the classification of SCM-EAFD1-25 was "good". However, although the UPV values of the SCM-EAFD1-50 and SCM-EAFD1-100 mortars slightly increased as curing time evolved, they remained around 2500 m/s at all ages, resulting in the classification of "poor".

In the SCM-EAFD2 mortars (Figure 7), a decrease in the speed of the impulses with respect to SCRM was observed as the replacement ratio increased. It should be noted that SCM-EAFD2-25, despite reduced speed of propagation, maintained UPV values (4508 m/s) very close to those of SCRM after 56 days (5000 m/s). These values allowed SCM-EAFD2-25 to be classified as "excellent".

The decrease in UPV relative to SCRM implies a mortar of worse quality with the increase in the EAFD content, since generally high velocities are associated with significant mechanical strength. The study of UPV in SCMs with EAFD has not been previously carried out as part of other studies that included EAFD in the compositions of mortars or concrete. Nevertheless, the relationship between mechanical behaviour and UPV has been widely studied by researchers such as Hamid et al. (2010), Tennich et al. (2015), and Sharma and Khan (2017) for the replacement of cement or natural aggregate with silica fume, marble waste, marble tile waste, or copper slag in the production of high-performance concrete or SCC. Furthermore, Şahmaran et al. (2006), Le at al. (2017), and Kanadasan et al. (2018) studied the replacement of cement or natural aggregate with fly ash, limestone powder, brick powder crushed from clay bricks, kaolinite, stainless steel slag sand, or palm oil clinker in the production of SCMs that are conventional or have high fluidity. All of them observed a good correlation between the UPV values and the mechanical strengths experimentally determined for their materials.

Figure 8 shows the correlation between the UPV and compressive strength values for the mortars investigated in this research. For short curing times, the slope is greater than those corresponding to 28 and 56 days of age, which indicates that the replacement effect is more relevant for 7 days.

3.4. Mercury intrusion porosimetry (MIP)

The MIP values of the mortars are shown in Figure 9. Figure 9 a) shows the graph of cumulative mercury intrusion values *versus* pore diameter for the intrusion (solid line) and extrusion (dotted line) cycles. The cumulative mercury intrusion values increased with the amount of EAFD incorporated, with the highest values being recorded for the mortars with 100% EAFD. Figure 9 b) exhibits the mercury intrusion difference values as a function of pore diameter, i.e. the frequency of pore size. The cumulative and differential intrusion values were consistent with each other, and more intrusions were observed as the contents of EAFD1 and EAFD2 increased.

The classification of pore sizes in hydrated cement pastes is included in Figure 9, according to Silva et al. (2001). Pores with sizes lower than 0.01 μm were named "gel porosity", those between 0.01 μm and 0.05 μm "medium capillaries", those between 0.05 μm and 10 μm "large capillaries", and those larger than 10 μm "large capillaries and entrained air". In capillaries considered as "gel porosity", mercury does not fully penetrate (Abbaszadeh and Modarres, 2017) and these pores are related to C-S-H phases. “Medium capillaries” influence properties such as shrinkage since the water contained in these pores is due to surface tension and when it is removed can cause contraction of the system (Mehta and Monteiro, 2017). "Large capillaries" have been reported to be detrimental to properties such as compressive strength and water permeability (Silva et al., 2001; Mehta and Monteiro, 2017; Ortega-López et al., 2018).

In Figure 9 b), it is observed that all mortars showed mercury intrusion curves with mainly two maximums of the pore diameter (Cook and Hover, 1999). One was observed at around 100 µm; these pores are related to the air contained in the mixes (Mehta and Monteiro, 2017). The second sharp peak (corresponding to maximum frequency of the pore size) varies its location depending on the mortar. Between 100 µm and 0.3 µm, the mortars revealed very similar intrusion volumes. SCM-EAFD1-100 exhibited an intrusion volume of up to 1.38 mL/g approximately in "large capillaries" (around 0.2 µm). This phenomenon is consistent with the values shown in Table 4, which suggests that the compressive strength of SCM-EAFD1-100 is not comparable to that of SCRM. For around 0.06 µm (Figure 9 b)) in the "large capillaries", both SCRM and SCM-EAFD1-25 showed maximum intrusion volumes; the volume was higher in the mortar with EAFD1 (0.33 mL/g *versus* 0.18 mL/g for SCRM). Once again, the compressive strength values recorded in these mortars (Table 4) were consistent with those recorded for mercury intrusion. Regarding the mortars with EAFD2, SCM-EAFD2-25 showed an intrusion volume in "large capillaries" that was slightly lower than that of SCRM (0.12 mL/g for SCM-EAFD2-25 *versus* 0.18 mL/g for SCRM). This small difference may be considered negligible, which agrees with the distribution of the "large capillaries" of SCM-EAFD2-25 with a similar compressive strength (Table 4) to SCRM.

The sharp peaks indicate mercury intrusion in the mortar specimen through the pores connected to the surface (Garboczi, 1990; Cook and Hover, 1999). However, SCM-EAFD2-25 exhibited a different behaviour. Its mercury intrusion curve (Figure 9 b)) revealed rounded peaks at 0.007 μm and 0.04 μm, indicating a different intrusion mechanism. Researchers such as Cook and Hover (1999) pointed out two possible reasons for this behaviour. On the one hand, the rounded peaks may appear due to the intrusion of mercury into a material phase with a wide distribution of smaller pores. On the other hand, during the hydration of cement, the interconnected pores can be totally isolated, leading to the disappearance of the sharp peak. However, if there is a pore volume that is still blocked, the mercury may access the pore by destroying the blockages and facilitate the appearance of the rounded mercury intrusion peaks.

Although SCM-EAFD2-100 registered the minimum intrusion volume in the "large capillaries" (0.06 mL/g), in "medium capillaries" (0.01 μm and 0.05 μm) the intrusion reached a value of 0.92 mL/g, with a maximum between 0.01 μm and 0.02 μm. The rest of the mortars revealed lower intrusion volumes in “medium capillaries”: 0.19 mL/g for SCRM, 0.38 mL/g for SCM-EAFD1-25, 0.11 mL/g for SCM-EAFD1-100, and 0.30 mL/g for SCM-EAFD2- 25.

Regarding "gel porosity", it is observed that the SCM-EAFD1-100 mortar did not register any value (0.00 mL/g), which is in accordance with the consideration of a non-cementitious matrix (the absence of C-S-H phases in agreement with the negligible release of heat during hydration, as showed in Figure 4). The remaining values were: 0.01 mL/g, 0.08 mL/g, 0.13 mL/g, and 0.24 mL/g, for SCRM, SCM-EAFD1-25, SCM-EAFD2-25, and SCM-EAFD2-100, respectively.

Table 5 shows the values obtained from the MIP test, according to the total volume of mercury intrusion, threshold diameter, median pore diameter, bulk density, skeletal density, and porosity. The total volume of mercury intrusion increases with EAFD content, with values of 0.42 mL/g, 0.82 mL/g, 1.56 mL/g, 0.61 mL/g, and 1.27 mL/g for SCRM, SCM-EAFD1-25, SCM-EAFD1-100, SCM-EAFD2-25, and SCM-EAFD2-100, respectively. It was observed that EAFD1 produced a greater intrusion than EAFD2.

The threshold pore diameter physically refers to the diameter above which a capillary trajectory cannot be formed in the entire sample (Cui and Cahyadi, 2001). SCRM recorded a threshold pore diameter of 0.15 μm with a total volume of mercury intrusion of 0.42 mL/g (Table 5). EAFD1 produced a small decrease in the threshold pore diameter for 25% substitution (0.10 μm), thus increasing the total volume of mercury intrusion. For 100% EAFD1, the threshold pore diameter increased to 0.25 μm. In general, the smaller the threshold pore diameter, the finer the structure of the pores is considered (Cui and Cahyadi, 2001). In this sense, the mortars with EAFD2 showed the smallest threshold pore diameters (of 0.08 μm for SCM-EAFD2-25 and 0.04 μm for SCM-EAFD2-100).

The values of bulk density, skeletal density or total porosity were observed to be slightly lower than those obtained in the normalized test (Figures 18-21), because mercury does not penetrate gel porosity, while water does (Cook and Hover, 1999; Abbaszadeh and Modarres, 2017). The values of total porosity registered by MIP for the mortars with 25% substitution (EAFD/CEM ratio of approximately 0.30; see Table 3) agreed with those obtained by Castellote et al. (2004) for a matrix with an EAFD/CEM ratio of 0.55 (17.47%).

3.5. Shrinkage and mass loss

The shrinkage and mass loss of the mortars were measured up to a period of 91 days. The results of the total shrinkage and mass loss test for SCM-EAFD1 and SCM-EAFD2 are shown in Figures 10, 11 and 12.

SCRM showed a shrinkage of 1208 μm/m after 28 days, beyond which a change in the trend of the curve of shrinkage was observed that stabilized the results and finally revealed a value of 1390 μm/m after 91 days. The shrinkage curve increased very fast in the first few days, reaching 60–90% after 28 days (Acker and Ulm, 2001). The mass loss of SCRM reached 95.65% after 91 days.

The evolution of the hydration reactions associated with shrinkage depends on the amount of water available in the mortar for the reaction (Alrifai et al., 2013). Thus, the performance of SCM-EAFD agreed with the highest w/c ratio respect to SCRM (Table 3). In addition, the shrinkage recorded in the mortars with EAFD was related to the mechanical behaviour. Mortars with less shrinkage had the highest compressive strength (Table 4). Ledesma et al. (2015) and Torres-Gómez et al. (2016) prepared mortars with 50% each of natural sand and recycled sand from ceramic waste. They related the greater shrinkage of the mortars to their lower compressive strengths and greater w/c ratio relative to the reference mortar.

SCM-EAFD1 (Figure 10) did not show a dosage-pattern response as a function of the EAFD1 content. SCM-EAFD1-25 had the highest shrinkage (1479 μm/m after 91 days). The behaviour of SCM-EAFD1-25 reflected the predominance of drying shrinkage, since the mass loss tendency is the same of shrinkage. SCM-EAFD1-50 experienced the least shrinkage (844 μm/m after 91 days), with values lower than those registered in SCRM. SCM-EAFD1-100 recorded values that were intermediate (1250 μm/m after 91 days) between those of SCM-EAFD1-50 and SCM-EAFD1-25. Up to 25% replacement of EAFD1 by SF retraction occurred. The total shrinkage behaviour of SCM-EAFD1-50 and SCM-EAFD1-100 could be linked to incorrect and/or delayed setting times as a consequence of the EAFD1 incorporation (Figure 4) and therefore, the shrinkage did not evolve as in SCM-EAFD1-25. Figure 11 shows, as a way of example, the evolution of the heat flow of SCRM and SCM-EAFD1-100. For SCRM a second peak in the curve is observed at 10 h approximately (corresponding to the setting heat after the induction period), which is not observed in SCM-EAFD1-100 for 360 h. Additionally, the compressive strengths of SCM-EAFD1-50 and SCM-EAFD1-100 were not in the same range as that of SCRM or SCM-EAFD1-25 (Table 4). Consequently, SCM-EAFD1-50 and SCM-EAFD1-100 could not be considered as cementitious matrixes. Moreover, the mass loss due to water loss reinforced this behaviour because it did not show the same tendency as shrinkage.

The shrinkage of the mortars with EAFD2 is shown in Figure 12. Owing to the demoulding of SCM-EAFD2-25 after 3 days and SCM-EAFD2-50 and SCM-EAFD2-100 after 6 days, their shrinkage was measured immediately after the respective demoulding age. In SCM-EAFD2, total shrinkage was high in the initial curing period and stabilized after 28 days (Acker and Ulm, 2001).

SCM-EAFD2 exhibited greater shrinkage for higher EAFD2 content. This fact could be explained by the greater number of finer particles (Figure 3) when the percentage replacement increased and also the large specific surface area of EAFD compared to SF 3.70 m^2/g, 4.60 m^2/g and 0.25 m^2/g for EAFD1, EAFD2 and SF, respectively. Although shrinkage during the initial period was greater in SCM-EAFD2-25 than in SCM-EAFD2-50, after 14 days, both mortars revealed very similar total shrinkage values (1604 μm/m and 1813 μm/m after 91 days). The largest dimensional change was recorded in the mortar with 100% SF replacement with EAFD2, with a value of 2302 μm/m after 91 days, which represents an increase in shrinkage of approximately 65% as compared with SCRM.

The shrinkage behaviour of SCM-EAFD2-100 agreed with the MIP values (Figure 9 b)). It is known that capillaries smaller than 0.05 μm ("medium capillaries") are more relevant to properties such as shrinkage (Mehta and Monteiro, 2017). In this sense, SCM-EAFD2-100 exhibited a greater total volume of mercury intrusion in "medium capillaries" (0.92 mL/g).

The total volumes of mercury intrusion in the "medium capillaries" of SCRM and the mortars with 25% SF replacement also agreed with their recorded shrinkage. SCRM registered the smallest shrinkage after 91 days (1390 μm/m) and the smallest volume (0.19 mL/g). The mortars with 25% substitution displayed very similar values of the total volume of mercury intrusion in "medium capillaries" (0.38 mL/g for SCM-EAFD1-25 and 0.30 mL/g for SCM-EAFD2-25), thus recording very close shrinkage values after 91 days (1479 μm/m and 1604 μm/m, respectively). In terms of mass loss, it was observed that it increased proportionally with shrinkage. This behaviour indicated that shrinkage in SCM-EAFD2 occurred predominantly through drying of the mortars.

The literature on shrinkage of mortars with EAFD is scarce. Maslehuddin et al. (2011) carried out a shrinkage study by drying in concrete, replacing cement with 2% of EAFD and using a w/c ratio of 0.45. These authors observed 175% higher shrinkage in concrete with EAFD compared to that of the reference concrete. In general, the greater the fine content introduced in mortars, the higher the shrinkage achieved (De Angelis et al., 2002; Silva et al., 2009; Braga et al., 2012), as it is promoted by a greater absorption of fine particles.

3.6. Water absorption by capillarity

The results of water absorption by capillarity after 28 and 56 days are shown in Figures 13 and 14. After 56 days, all mortars revealed slightly decreased water absorption by capillarity, except SCM-EAFD1-25 that showed a strong drop. This phenomenon is related to the evolution of the internal pores of the mortars. After 28 days of curing, the chemical reactions had not yet completed, allowing the pores to be less interconnected;

therefore, the pore structure of the cement pastes became finer with the increase in curing time (Cui and Cahyadi, 2001).

SCRM showed water absorption by capillarity values of 0.10 and 0.09 kg/($m^2 \cdot min^{0.5}$) after 28 and 56 days, respectively. The mortars revealed higher water absorption by capillarity in comparison with SCRM when EAFD1 was incorporated in the mixes (Figure 13). The high capillary coefficients represent the high water absorption rates (Lucas et al., 2016), which is not desired for a construction material such as SCM considered in this study, because it would favour the release of the heavy metals present in EAFD.

It is worth highlighting the behaviour of SCM-EAFD1-25, which had a water absorption by capillarity value six times higher (0.59 kg/($m^2 \cdot min^{0.5}$)) than that of SCRM (0.10 kg/($m^2 \cdot min^{0.5}$)) after 28 days. However, the value of SCM-EAFD1-25 fell to 0.12 kg/($m^2 \cdot min^{0.5}$) after 56 days. This was related to delays in the hydration of cement and the gain of compressive strength of this mortar after 28 days (22.4 MPa) compared to after 56 days (48.2 MPa). Therefore, the change in the slope of the total hydration heat curve (Figure 4) and the delay observed in the development of compressive strength (Table 4) influenced the amounts of water absorbed by capillarity recorded over time.

The maximum capillary values were 1.08 and 1.04 kg/($m^2 \cdot min^{0.5}$) after 28 and 56 days, respectively, for the SCM-EAFD1-50 mortar. The SCM-EAFD1-100 mortar showed slightly lower values than SCM-EAFD1-50. For 50% and 100% replacement of SF with EAFD1, the compressive strengths remained below 7 MPa, and could not be considered as cementitious matrices for use in SCC. The greater capillary volume can be attributed to the lower degree of hydration (Silva et al., 2001). The total heat of hydration (Figure 4) and MIP of SCM-EAFD1-100 (Figure 9 b)) corroborated this hypothesis through the low total heat of hydration experimentally obtained and the maximum frequency of the pore size in "large capillaries". The presence of “large capillaries” does not benefit properties like water absorption by capillarity (Ortega-López et al., 2018). Additionally, Mehta and Monteiro (2017) reported that well hydrated pastes with low w/c ratios show pores with sizes between 0.01 μm and 0.05 μm. The maximum frequency of pore size in SCM-EAFD1-100 was outside this range; therefore, SCM-EAFD1-100 is surely not considered a cementitious matrix. Once again, the results obtained in the SCM-EAFD1 mortars point to non-utilization when the percentage replacement of SF with EAFD1 is greater than 25%.

In SCM-EAFD2 (Figure 14), capillarity increased with increasing EAFD2 content, revealing maximum values of 0.21 and 0.19 kg/($m^2 \cdot min^{0,5}$) after 28 and 56 days, respectively, in SCM-EAFD2-100. SCM-EAFD2-25 showed very similar capillarity to SCRM, with values of 0.10 and 0.08 kg/($m^2 \cdot min^{0,5}$) after 28 and 56 days, respectively.

The increase in water absorption by capillarity may also be linked to the greater w/c ratio of the mortars when EAFD was incorporated. Although the w/c ratio in SCM-EAFD2 was higher than that in SCM-EAFD1, the greater number of fine particles of EAFD2 (Figure 3) may have caused a filler effect that alleviated the effect of w/c ratio in the mortars, thereby decreasing the number of pores interconnected and favouring lower coefficients of water absorption by capillarity. In addition, EAFD2 allowed setting of the mortars (Figure 4) and gains in compressive strength (Table 4).

Figure 15 shows a good correlation between water absorption by capillarity of the mortars and their compressive strength. It is observed that water absorption by capillary decreased when compressive strength increased. This suggests that the internal pores were less connected, which favoured a stronger structure, and vice versa.

3.7. Water absorption capacity, bulk and skeletal density and open porosity for water

Water absorption capacity, density and open porosity for water were measured in all mortars after 28 days. However, for SCM-EAFD1-100, the water absorption after immersion and vacuum, bulk density, skeletal density and open porosity for water could not be evaluated. The specimens of SCM-EAFD1-100, previously conditioned at 100 ºC, showed cracks after immersion in water for 48 h and subsequent vacuuming and, therefore, measurement was impossible. This mortar had previously been discarded for not exhibiting sufficient compressive strengths for consideration as SCM.

The water absorption capacity after immersion and after immersion and vacuuming of the mortars are shown in Figures 16 and 17, respectively. SCRM displayed the lowest water absorption values (7.38% and 7.41% after immersion and after immersion and vacuuming, respectively).

All mortars absorbed more water when EAFD was incorporated in the mixes. SCM-EAFD-1 (Figure 16) increased its water absorptions after immersion and after immersion and vacuuming to 11.04% and 11.11% for SCM-EAFD1-25, 12.07% and 12.11% for SCM-EAFD1-50, respectively, and 14.17% for SCM-EAFD1-100. The water absorbed by SCM-EAFD2 (Figure 17) was slightly lower than that measured in SCM-EAFD1. The values recorded after immersion and after immersion and vacuuming were 9.45% and 9.51% for SCM-EAFD2-25, 10.69% and 10.75% for SCM-EAFD2-50, and 14.35% and 14.43% for SCM-EAFD2-100, respectively. The increase in water absorption after immersion indicates that a greater number of pores are filled with water. The saturation of these pores will be greater after subsequent immersion and vacuuming, since the air contained in them is displaced by water.

The bulk density and skeletal density of the mortars are shown in Figures 18 and 19, respectively. SCRM recorded values of 2.17 g/cm^3 and 2.58 g/cm^3 for bulk density and skeletal density, respectively. Bulk density decreased as the amount of EAFD increased. SCM-EAFD1-25, and SCM-EAFD1-50 showed the same bulk density of 2.02 g/cm^3. In SCM-EAFD2, the mortar with 100% replacement had the lowest bulk density of 1.98 g/cm^3. The decrease in bulk density indicates that the number of pores increased with increasing amount of EAFD relative to SCRM. This behaviour is in accordance with that observed for water absorption after immersion and after immersion and vacuuming.

The skeletal density increased with waste content. SCM-EAFD1-50 and SCM-EAFD2-100 exhibited values of 2.67 g/cm^3 and 2.77 g/cm^3, respectively. The skeletal density increased due to the higher real particle density of the wastes compared to that of the replaced SF (3.84 g/cm^3 for EAFD1, 3.81 g/cm^3 for EAFD2, and 2.60 g/cm^3 for SF).

The open porosity for water is the ratio between the volume of pores accessible for water and the volume limited by the external surface of the specimen, including any pores. This property measured after 28 days, is shown in Figures 20 and 21. SCRM had an open porosity for water of 16.06%. The mortars showed a higher open porosity for water as the EAFD content increased, registering values of 22.46% and 24.42% for SCM-EAFD1-25 and SCM-EAFD1-50, respectively. This represents increases in porosity of 39.85% and 52.05% for SCM-EAFD1-25 and SCM-EAFD1-50 relative to SCRM, respectively. Regarding SCM-EAFD2, the values were 19.79%, 22.24%, and 28.56% for SCM-EAFD2-25, SCM-EAFD2-50, and SCM-EAFD2-100, respectively, representing increases of 23.22%, 38.48%, and 77.83% in comparison with SCRM, respectively. The increased open porosity for water may be attributed to the higher w/c ratio in the mortars containing EAFD. Garijo et al. (2018) observed higher values of open porosity in natural hydraulic lime mortars when the w/c ratio increased. In the study of Lucas et al. (2016), the open porosity increased in mortars with replacement of sand with sanitary ware waste due to the increase in w/c ratio.

3.8. Contact angle

Figure 22 shows the recorded contact angle of a drop of water on the surface of the mortars. The mortars exhibited higher hydrophilic surface areas as the percentage of EAFD increased. SCRM displayed the highest contact angle, followed by the mortars with 25% substitution of SF with EAFD. SCM-EAFD1-25 and SCM-EAFD2-25 showed very similar contact angles, close to that of SCRM. For 100% substitution, EAFD2 formed a less hydrophilic surface than EAFD1. SCM-EAFD1-100 registered a very short lifetime of the drop of water on the surface of the mortar as well as a low contact angle, which were related to the large macro porosity of this mortar (Figures 9 b) and 13). This indicates that SCM-EAFD1-100 is a hydrophilic material, whose contact angles rapidly diminish with time (Klein et al., 2012). This behaviour justified the impossibility of measuring the water absorption after immersion and vacuuming, bulk density, skeletal density and open porosity of SCM-EAFD1-100 (Figures 16, 18, and 20).

3.9. Electrical resistivity

The electrical resistivity of the mortars after 7, 28, and 56 days was evaluated. This property characterizes the greater or lesser difficulty of the ion mobility inside the matrix (da Silva, 2013). This property has been used by other researchers to indirectly characterize the durability of concrete by relating it to the chloride transport mechanism (Real et al., 2015). Table 6 shows the values obtained expressed in Ω·m. For all the tested samples, the electrical resistivity decreased relative to that of SCRM (20000 Ω·m, 45000 Ω·m, and 60000 Ω·m after 7, 28, and 56 days, respectively) when the EAFD replacement was higher, achieving the lowest values for 100% replacement (17.5 Ω·m, 44.5 Ω·m, and 76.7 Ω·m after 7, 28 and 56 days, respectively, for SCM-EAFD1-100 and 34.8 Ω·m, 232.5 Ω·m, and 390.6 Ω·m after 7, 28, and 56 days, respectively, for SCM-EAFD2-100). Higher EAFD content in the mortars results in a higher heavy metal content, which can justify a higher conductivity, and, consequently, lower electrical resistivity. When the amount of EAFD increases in the mixes, the w/c ratio increases, which decreases the electrical resistivity (Neville, 1995).

In general, electrical resistivity increased when curing time was higher, w/c ratio decreased, or a reactive mineral such as blast furnace slag, fly ash or silica fume was added (Polder, 2001). Particularly, in cement-based materials, this property can be affected by factors such as the composition and humidity of the mixes, their porosity, the solution contained in the pores, and the number and distribution of these pores (Polder, 2001). In this context, the electrical resistivity values revealed the same trend as properties such as MIP, water absorption by capillarity, open porosity for water, and contact angle, because the higher the w/c ratio, the more porous and interconnected the SCMs were. Therefore, the electrical conductivity of the mortars is higher when the EAFD content increases. Sharma and Khan (2017) analysed the replacement of natural aggregates with copper slag in SCC. These authors observed a decrease in electrical resistivity of concrete for 80% and 100% copper slag and related this behaviour to the greater open porosity for water recorded in the matrices and the high amount of copper slag.

Chloride migration depends mainly on the paste, and not on the aggregate. This was confirmed by Real et al. (2015). The relation between concrete resistivity and rapid chloride migration coefficient was revealed through the relation (2) proposed by Duracrete (2000). Where: A was LN(965:384), ρ were electrical resistivity values of self-compacting mortars and constant B was equal to -1.0027.

$$D_{RCM} = A \cdot \rho^{B} \tag{2}$$

Based on the electrical resistivity values obtained in this research, it is possible to elucidate the migration of chlorides in SCC fabricated based on the mortars considered in this study (Table 7). It is observed that as the EAFD content increases, the rapid chloride migration coefficient will also be higher, which results in a SCC of lower quality.

3.10. Carbonation depth

The carbonation depth was measured in all mortars after 7, 14, 28, and 56 days of exposure. The evolution of carbonation with exposure time (year$^{0.5}$) is shown in Figure 23.

SCRM did not show carbonation at any age. SCM-EAFD1-25 showed rapid carbonation, with the process occurring 7 days faster than in SCM-EAFD2 at any point in time. After 7 days, the carbonation depth of SCM-EAFD1-25 evolved more slowly, with a coefficient of 36.89 mm/year$^{0.5}$ after 56 days.

SCM-EAFD1-50 and SCM-EAFD1-100 recorded complete carbonation after 7 days, which implied that the use of these mortars in the production of reinforced concrete is not recommended. The presence of a large amount of "large capillaries" (Figure 9 b)) justifies the fact that SCM-EAFD1-100 showed complete carbonation after 7 days of exposure.

In SCM-EAFD2, the coefficient of carbonation was higher as the replacement percentage increased, with values of 17.82 and 31.09 mm/year$^{0.5}$ after 56 exposure days in SCM-EAFD2-25 and SCM-EAFD2-50, respectively, and 50.84 mm/year$^{0.5}$ after 28 days in SCM-EAFD2-100. SCM-EAFD2-100 showed complete carbonation after 56 days. Figure 24 shows the carbonation depth of SCM-EAFD1-25 and SCM-EAFD2-25 after 56 days.

The carbonation depth values of the mortars agreed with those of MIP (Figure 9 b)), water absorption by capillarity (Figures 13 and 14), open porosity for water (Figures 20 and 21), and contact angle (Figure 22). When EAFD was incorporated in the mortars, the number of connected and accessible pores increased compared to that in SCRM, which led to faster diffusion of the CO_2 inside the mortar. Song and Kwon (2007) suggested that the volume of pores and the depth of carbonation are proportionally related. In addition, the higher carbonation depths of the mortars with EAFD1 and EAFD2 were also related to the higher w/c ratios of these mortars compared to those of SCRM (Khunthongkeaw et al., 2006).

4. Conclusions

This research analysed, from the point of view of the view of chemical, physical and mechanical properties and durability, the feasibility of fabricating self-compacting mortar (SCM) with two industrial wastes (Electric Arc Furnace Dust: EAFD1 and EAFD2) as replacements of SF at of 25%, 50%, and 100% in volume.

The results showed that up to 25% EAFD1 may be incorporated in the fabrication of SCM, exhibiting a performance that was similar to that of the self-compacting reference mortar (SCRM). However, the incorporation of 50% and 100% EAFD1 was detrimental to the hydration of cement, which was demonstrated by the low heat of hydration, insufficient compressive strength, inadequate ultrasonic pulse velocity and incorrect evolution of shrinkage relative to the SCRM. Therefore, amounts higher than 25% of EAFD1 were discarded as cementitious matrices for use in SCM. Additionally, further analysis of the mortar with 100% EAFD1 confirmed the highest total volume of mercury intrusion compared with that of the SCRM and its negative impact on the absorption of water by capillarity, water absorption capacity, bulk and skeletal density, open porosity for water, contact angle, electrical resistivity, rapid chloride migration coefficient, and depth of carbonation.

Compared to EAFD1, EAFD2 affected to a lower extent the hydration reactions of cement and the properties studied. Although the incorporation of EAFD2 negatively affected the analysis of the durability properties, the mortar with 25% EAFD2 showed a behaviour that was very close to that of the SCRM, guaranteeing the feasibility of producing SCMs containing this waste.

In line with the new circular economy model pursued by the European Union, the results of this research confirmed that using EAFD as secondary raw material (SRM) to produce SCM is a viable alternative to depositing it in landfills.

Acknowledgments

This study was partly supported by the Andalusian Regional Government (Research Groups FQM-391 and TEP-227), the XXI Own Research Plan of the University of Córdoba (2016) – Modality 4.2 (FEDER) and the Project MAT2015-70728-P from Spanish Ministry of Economy and Competitiveness. A. Lozano-Lunar acknowledges the funding from MECD-Spain (http://www.mecd.gob.es/educacion-mecd/; FPU14/05245). The authors also thank the CERIS Research Institute, at IST - Universidade de Lisboa, and the Inorganic Materials and Environment Research Group (University of Navarra), as some of the tests were carried out in their facilities. The authors gratefully acknowledge the support of the Portuguese Foundation for Science and Tech-nology (Fundação para a Ciência e Tecnologia) through the research project PTDC/ECI-CON/29196/2017 (RInoPolyCrete).

References

Abbaszadeh, R., Modarres, A., 2017. Freeze-thaw durability of non-air-entrained roller compacted concrete designed for pavement containing cement kiln dust. Cold Reg. Sci. Technol. 141, 16-27.

Acker, P., Ulm, F.-J., 2001. Creep and shrinkage of concrete: physical origins and practical measurements. Nucl. Eng. Des. 203(2), 143-158.

AENOR, 2017. Asociación Española de Normalización y Certificación, AENOR, Madrid, Spain. Available from: www.aenor.es. (accessed 14 September 2017).

Alrifai, A., Aggoun, S., Kadri, A., Kenai, S., Kadri, E.H., 2013. Paste and mortar studies on the influence of mix design parameters on autogenous shrinkage of self-compacting concrete. Constr. Build. Mater. 47, 969-976.

Alsheyab, M.A.T., Khedaywi, T.S., 2013. Effect of electric arc furnace dust (EAFD) on properties of asphalt cement mixture. Resour. Conserv. Recy. 70, 38-43.

Aslani, F., Nejadi, S., 2012. Mechanical properties of conventional and self-compacting concrete: an analytical study. Constr. Build. Mater. 36, 330-347.

Assié, S., Escadeillas, G., Waller, V., 2007. Estimates of self-compacting concrete 'potential' durability. Constr. Build. Mater. 21(10), 1909-1917.

Balderas, A., Navarro, H., Flores-Velez, L.M., Dominguez, O., 2001. Properties of portland cement pastes incorporating nanometer-sized franklinite particles obtained from electric-arc-furnace dust. J. Am. Ceram. Soc. 84(12), 2909-2913.

Billberg, P., 1999. Fine mortar rheology in mix design of SCC, Proceedings of the first international RILEM symposium on self-compacting concrete, Edited by Å. Skarendahl and Ö. Petersson, Stockholm, 47-58.

Braga, M., de Brito, J., Veiga, R., 2012. Incorporation of fine concrete aggregates in mortars. Constr. Build. Mater. 36(Supplement C), 960-968.

Castellote, M., Menendez, E., Andrade, C., Zuloaga, P., Navarro, M., Ordonez, M., 2004. Radioactively contaminated electric arc furnace dust as an addition to the immobilization mortar in low- and medium-activity repositories. Environ. Sci. Technol. 38(10), 2946-2952.

COM(2017)33, 2017. European Commission, Report from the commission to the European parliament, the council, the European economic and social committee and the committee of the regions on the implementation of the Circular Economy action plan. Available from: http://ec.europa.eu/environment/circular-economy/implementation_report.pdf (accessed 10 October 2018).

Cook, R.A., Hover, K.C., 1999. Mercury porosimetry of hardened cement pastes. Cem. Concr. Res. 29(6), 933-943.

Cui, L., Cahyadi, J.H., 2001. Permeability and pore structure of OPC paste. Cem. Concr. Res. 31(2), 277-282.

da Silva, P.R., 2013. Evaluation of the durability of self-compacting concrete (SCC) (in Portuguese). PhD Thesis in Civil Engineering. Instituto Superior Técnico. Universidade de Lisboa, Portugal.

da Silva, P.R., de Brito, J., 2015. Fresh-state properties of self-compacting mortar and concrete with combined use of limestone filler and fly ash. Mater. Res. Ibero Am. J. 18(5), 1097-1108.

De Angelis, G., Medici, F., Montereali, M.R., Pietrelli, L., 2002. Reuse of residues arising from lead batteries recycle: a feasibility study. Waste Manag. 22(8), 925-930.

de Souza, C.A.C., Machado, A.T., Lima, L., Cardoso, R.J.C., 2010. Stabilization of electric-arc furnace dust in concrete. Mater. Res. Ibero Am. J. 13(4), 513-519.

de Vargas, A.S., Masuero, A.B., Vilela, A.C.F., 2006. Investigations on the use of electric-arc furnace dust (EAFD) in pozzolan-modified Portland cement I (MP) pastes. Cem. Concr. Res. 36(10), 1833-1841.

Di Mundo, R., Petrella, A., Notarnicola, M., 2018. Surface and bulk hydrophobic cement composites by tyre rubber addition. Constr. Build. Mater. 172, 176-184.

Duracrete, 2000. General guidelines for durability design and redesign, Final technical report, the European Union - Brite EuRam III, probabilistic performance based durability design of concrete structures, document B 95-1347/R15. p. 138.

EFNARC, 2005. European Federation of National Associations Representing producers and applicators of specialist building products for Concrete (EFNARC), The European guidelines for self-compacting concrete specification. Production and use. Hampshire, UK. Available from: www.efnarc.org (accessed 25 October 2017).

EU, 2003. European Council Decision for the acceptance of waste at landfills. Available from: https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2003:011:0027:0049:EN:PDF (Accessed 18 January 2018).

EWC, 2002. European Waste Catalog. Available from: https://www.boe.es/boe/dias/1999/01/08/pdfs/A00570-00580.pdf (accessed 28 September 2018).

Fares, G., Al-Zaid, R.Z., Fauzi, A., Alhozaimy, A.M., Al-Negheimish, A.I., Khan, M.I., 2016. Performance of optimized electric arc furnace dust-based cementitious matrix compared to conventional supplementary cementitious materials. Constr. Build. Mater. 112, 210-221.

Flatt, R.J., Roussel, N., Cheeseman, C.R., 2012. Concrete: an eco material that needs to be improved. J. Eur. Ceram. Soc. 32(11), 2787-2798.

Garboczi, E.J., 1990. Permeability, diffusivity, and microstructural parameters: A critical review. Cem. Concr. Res. 20(4), 591-601.

Garijo, L., Zhang, X., Ruiz, G., Ortega, J.J., Wu, Z., 2018. The effects of dosage and production process on the mechanical and physical properties of natural hydraulic lime mortars. Constr. Build. Mater. 169, 325-334.

Giraldo, M., Tobón, J., 2006. Mineralogical evolution of Portland cement during hydration process. Dyna 148, 69-81.

Hamid, R., Yusof, K.M., Zain, M.F.M., 2010. A combined ultrasound method applied to high performance concrete with silica fume. Constr. Build. Mater. 24(1), 94-98.

JCPD, 1995. Joint Committee on Power Diffraction Standard-International Centre forDiffraction Data. Swarthmore, PA.

Kanadasan, J., Razak, H.A., Subramaniam, V., 2018. Properties of high flowable mortar containing high volume palm oil clinker (POC) fine for eco-friendly construction. J. Clean. Prod. 170, 1244-1259.

Khunthongkeaw, J., Tangtermsirikul, S., Leelawat, T., 2006. A study on carbonation depth prediction for fly ash concrete. Constr. Build. Mater. 20(9), 744-753.

Klein, N.S., Bachmann, J., Aguado, A., Toralles-Carbonari, B., 2012. Evaluation of the wettability of mortar component granular materials through contact angle measurements. Cem. Concr. Res. 42(12), 1611-1620.

Le, D.H., Sheen, Y.N., Bui, Q.B., 2017. An assessment on volume stabilization of mortar with stainless steel slag sand. Constr. Build. Mater. 155, 200-208.

Ledesma, E.F., Jimenez, J.R., Ayuso, J., Fernandez, J.M., de Brito, J., 2017. Experimental study of the mechanical stabilization of electric arc furnace dust using fluid cement mortars. J. Hazard. Mater. 326, 26-35.

Ledesma, E.F., Jiménez, J.R., Ayuso, J., Fernández, J.M., de Brito, J., 2015. Maximum feasible use of recycled sand from construction and demolition waste for eco-mortar production – Part-I: ceramic masonry waste. J. Clean. Prod. 87, 692-706.

Ledesma, E.F., Lozano-Lunar, A., Ayuso, J., Galvín, A.P., Fernández, J.M., Jiménez, J.R., 2018. The role of pH on leaching of heavy metals and chlorides from electric arc furnace dust in cement-based mortars. Constr. Build. Mater. 183, 365-375.

Lozano-Lunar, A., Raposeiro da Silva, P., de Brito, J., Fernández, J. M., Jiménez, J. R., 2019. Safe use of electric arc furnace dust as secondary raw material in self-compacting mortars production. J. Clean. Prod. 211, 1375-1388. doi:https://doi.org/10.1016/j.jclepro.2018.12.002.

Lucas, J., de Brito, J., Veiga, R., Farinha, C., 2016. The effect of using sanitary ware as aggregates on rendering mortars' performance. Mater. Des. 91(Supplement C), 155-164.

Maslehuddin, M., Awan, F.R., Shameem, M., Ibrahim, M., Ali, M.R., 2011. Effect of electric arc furnace dust on the properties of OPC and blended cement concretes. Constr. Build. Mater. 25(1), 308-312.

Mehta, P.K., Monteiro, P.J., 2017. Concrete: microstructure, properties and materials, McGraw-Hill, New York.

Moon, G.D., Oh, S., Jung, S.H., Choi, Y.C., 2017. Effects of the fineness of limestone powder and cement on the hydration and strength development of PLC concrete. Constr. Build. Mater. 135, 129-136

Mymrin, V., Nagalli, A., Catai, R.E., Izzo, R.L.S., Rose, J., Romano, C.A., 2016. Structure formation processes of composites on the base of hazardous electric arc furnace dust for production of environmentally clean ceramics. J. Clean. Prod. 137, 888-894.

Nepomuceno, M., 2005. Methodology for self-compacting concrete mix design (in Portuguese). PhD thesis in civil engineering, University of Beira Interior, Portugal, 1-799.

Nepomuceno, M., Oliveira, L., Lopes, S.M.R., 2012. Methodology for mix design of the mortar phase of self-compacting concrete using different mineral additions in binary blends of powders. Constr. Build. Mater. 26(1), 317-326.

Neville, A.M., 1995. Properties of concrete. Longman London.

Ortega-López, V., Fuente-Alonso, J.A., Santamaría, A., San-José, J.T., Aragón, Á., 2018. Durability studies on fiber-reinforced EAF slag concrete for pavements. Constr. Build. Mater. 163, 471-481.

Persson, B., 2001. A comparison between mechanical properties of self-compacting concrete and the corresponding properties of normal concrete. Cem. Concr. Res. 31(2), 193-198.

Polder, R., Andrade, C., Elsener, B., Vennesland, Ø., Gulikers, J., Weidert, R., Raupach, M., 2000. Test methods for on site measurement of resistivity of concrete. Mater. Struct. 33(10), 603-611.

Polder, R.B., 2001. Test methods for on site measurement of resistivity of concrete — a RILEM TC-154 technical recommendation. Constr. Build. Mater. 15(2), 125-131.

Real, S., Bogas, J.A., Pontes, J., 2015. Chloride migration in structural lightweight aggregate concrete produced with different binders. Constr. Build. Mater. 98(Supplement C), 425-436.

Sayadi, M., Hesami, S., 2017. Performance evaluation of using electric arc furnace dust in asphalt binder. J. Clean. Prod. 143, 1260-1267.

Sharma, R., Khan, R.A., 2017. Durability assessment of self compacting concrete incorporating copper slag as fine aggregates. Constr. Build. Mater. 155, 617-629.

Si, R., Wang, J., Guo, S., Dai, Q., Han, S., 2018. Evaluation of laboratory performance of self-consolidating concrete with recycled tire rubber. J. Clean. Prod. 180, 823-831.

Silva, D.A., John, V.M., Ribeiro, J.L.D., Roman, H.R., 2001. Pore size distribution of hydrated cement pastes modified with polymers. Cem. Concr. Res. 31(8), 1177-1184.

Silva, J., Brito, J.d., Veiga, R., 2009. Incorporation of fine ceramics in mortars. Constr. Build. Mater. 23(1), 556-564.

Silva, P., de Brito, J., Costa, J., 2011. Viability of two new mixture design methodologies for self-consolidating concrete. Aci Mater. J. 108(6), 579-588.

Singh, G., Siddique, R., 2016. Strength properties and micro-structural analysis of self-compacting concrete made with iron slag, as partial replacement of fine aggregates. Constr. Build. Mater. 127, 144-152.

Song, H.-W., Kwon, S.-J., 2007. Permeability characteristics of carbonated concrete considering capillary pore structure. Cem. Concr. Res. 37(6), 909-915.

Tennich, M., Kallel, A., Ben Ouezdou, M., 2015. Incorporation of fillers from marble and tile wastes in the composition of self-compacting concretes. Constr. Build. Mater. 91(Supplement C), 65-70.

Thongsanitgarn, P., Wongkeo, W., Chaipanich, A., Poon, C.S., 2014. Heat of hydration of Portland high-calcium fly ash cement incorporating limestone powder: Effect of limestone particle size. Constr. Build. Mater. 66, 410-417.

Torres-Gómez, A., Ledesma, E., Otero, R., Fernández, J., Jiménez, J., de Brito, J., 2016. Combined effects of non-conforming fly ash and recycled masonry aggregates on mortar properties. Materials 9(9), 729.

Unesid, 2013. Informe 2013 sobre el reciclaje del acero en la Industria Siderúrgica Española. Available from: https://unesid.org/iris2013/index.html (accessed 30 October 2018).

Weeks, C., Hand, R.J., Sharp, J.H., 2008. Retardation of cement hydration caused by heavy metals present in ISF slag used as aggregate. Cement Concrete Comp. 30(10), 970-978.

Whitehurst, E.A., 1951. Soniscope tests concrete structures. J. Am. Concr. Inst. 47(6), 433-444.

Şahmaran, M., Christianto, H.A., Yaman, İ.Ö., 2006. The effect of chemical admixtures and mineral additives on the properties of self-compacting mortars. Cement Concrete Comp. 28(5), 432-440.