

# Cement-based mortars production applying mining residues treated with an electro-based technology and a thermal treatment: Technical and economic effects

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## Abstract

The combination of treatments may improve the extraction of toxic and critical elements from secondary resources, produce changes in materials' physico-chemical properties and improvements on the sustainability of strategies. Mining residues were treated with an electro-dialytic process alone or coupled with a thermal procedure, and reused in cement-based mortars by replacing 10, 25 and 50% of the cement binder content. This study demonstrated the technical and economic feasibility of mortars produced with treated mining residues, namely in applications where flexural strength is key factor. The mechanical properties of the mortars corroborated their compatibility with bedding mortars and fired brick masonry walls.

**Keywords:** Construction product; Mortar; Mining waste; Electro-dialytic process; Thermal treatment; Cost efficiency

## 1 Introduction

Cement industry is facing challenges on supply energy costs, greenhouse gases emissions and alternative pozzolanic materials to thermal plant fly ashes [1]. Cement production is responsible for a majority of the environmental impacts of cementitious products production, namely when compared with the impact of other components, as aggregates and supplementary cementitious materials, and procedures as mixing and transport [2]. Cost/eco-efficient constituents to cement will promote civil engineering practices to become more sustainable and move towards circular economy principles [3].

Research on mining by-products reuse in the construction sector has been conducted during the past decades [4]. Mining industries provide materials to build infrastructures and instruments of daily use [5]. However, mining activities are one of the most disturbing practices for the engaged ecosystems. Their negative impacts are caused by voluminous mining residues disposal and acid mine drainage. Acid mine drainage often results from rock minerals/ore deposits exposure to water and/or oxygen. This facilitates the mobilization of present toxic chemical species and increase their concentrations in water supplies and food chains [6].


The reuse of mining residues in cement composites encourages waste management alternatives to landfill, the balance of mining sites and the minimization of primary raw materials needs [7]. Studies have been conducted on mining resources incorporation in cementitious composites (e.g. mortar, concrete), for binder and aggregate partial replacement in building materials. Alkali activated products [8] and pozzolanic materials [9] were successfully produced with tungsten mining residues. The durability of conventional products may be enhanced in terms of resistance to abrasion and acid attack due to a lower water absorption [10].

Mechanical and structural performance of mortars is dependent on the properties of the components involved in the formulation, as well as the need for repair interventions [11]. The effectiveness of mortars maintenance is mainly affected by compressive and flexural strengths, water vapour permeability, water absorption by capillarity action and drying [12].

To enlarge the application of mining residues within the construction sector, several strategies could enhance durability properties of composites produced with mining residues.

The electro-dialytic (ED) process is defined as a treatment that promotes the removal of substances from polluted substrates, based on the application of a direct low-level current intensity [13]. This treatment demonstrated potential to improve the removal/recovery of substances from mining residues, namely arsenic (35%) and tungsten (22%) [14]. After the ED treatment, mining residues also proved their viability as cement replacer in mortars [15].

The pozzolanic properties of mining residues, alone or coupled with other treatments could be improved [16]. Amorphous silica ( $\text{SiO}_2$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) compounds present in fine graded materials, when combined with  $\text{Ca}(\text{OH})_2$  and water generate calcium silicates and aluminates hydrates by pozzolanic reaction [17].

The reactivity of materials in cementitious composites is affected by:  (1). [Instruction: Number (1) is not related to equation (1). Thus, it does not need any hyperlink. Thank you] content of amorphous  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$ ; (2) temperature of the thermal treatment; (3) period at high temperature and cycle of heat treatment; (4) content and type of clays; (5) particle size distribution and specific surface and (6) water and  $\text{Ca}(\text{OH})_2$  availability for the chemical reactions involved [12].

Heating leads to the loss of water from the structure of the minerals and to the disruption of  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  crystal structure. For most clayish materials, high amounts of residues in the amorphous state are produced during controlled heating intervals, from 600 °C to 900 °C [18]. Particularly, tungsten mine sludge was subject to 950 °C to improve its pozzolanic activity [9].

The application of treatments to mining residues, before its reuse as construction materials, can empower circular economy of elements of interest, as critical raw materials. Also, it may leverage the sustainability of secondary resources, once it allows a prior removal of harmful compounds. This contributes to decrease the disposal of residues in mining sites and a safer reuse of mining residues in several applications (e.g. mortars and concretes). The thermal treatment may also enhance the pozzolanic reactivity of the residues, improving durability properties of construction products, although the curing conditions may influence mortars and concrete performance.

In the present work, mortars were produced by replacing cement binder contents by mining residues from Panasqueira mine, previously treated with the ED process alone or coupled with a thermal procedure. The replacement of cement was 0% for the Control, and 10, 25 and 50% (wt%) for the modified mortars. The characterisation of the mortars fresh and hardened state, after 28 days of cure, was assessed. These properties were also compared with the results from Almeida et al. [15], regarding the economic and technical feasibility of applying a complementary thermal treatment intended to increase pozzolanic reactivity. A cost-efficiency analysis of mortars produced with Panasqueira mining residues was carried out.

## 2 Experimental

### 2.1 Materials

Cement-based mortars were prepared with mining waste mud collected from the Panasqueira mine sludge circuit (Covilhã, Portugal, 40°10'11"N, 7°45'24"W). The sample was dried in a fume hood for 48 h, at 20 °C. Panasqueira mine produces 90,000 t of tungsten per year [19] and the mud waste generated is composed by particles below 2 mm [20].

Panasqueira mining residues were tested to replace CEM II/BL 32.5 N (Secil, Portugal), classified based on EN 197-1 [21]. A river siliceous sand was used as aggregate, with particles between 0.5 and 2.0 mm, similar to the CEN reference sand [22] as shown in Table S1 (in appendix A). Tap water was used as mixing water in mortar formulations.

## 2.2 Electrolytic and thermal treatment of mining residues

The ED treatment was primary applied for the separation of tungsten and arsenic from Panasqueira mining residues [14]. Mining residues were treated in a two-compartment ED acryl XT reactor (RIAS A/S, Roskilde, Denmark). The diameter of the reactor was 80 mm, with 50 mm length for the electrolyte compartment and 100 mm for the sample section. An anion exchange membrane AR204SZRA, MKIII, Blank (Ionics, USA) divided the two compartments. Two electrodes (Ti/MMO Permascand wire) with a length of 50 mm and a diameter of 3 mm (Grønvold & Karnov A/S, Denmark) were applied in each compartment. Constant current was maintained inside the reactor (50 mA) with a power supply E3612A (Hewlett Packard, Palo Alto, USA). Also, a magnetic stirrer was placed into the sample compartment. A suspension of 39 g of mining residues with 345 mL of deionized water and 5 mL of choline chloride/malonic acid (CAS 67-48-1; ≥99%, Sigma-Aldrich, Germany) was set in the cathode compartment. In the anode, 250 mL of 0.01 M NaNO<sub>3</sub> were added as anolyte. Ten experiments were operated for 4 days.

Following the ED process, a thermal treatment was performed at 950 °C [23]. The oven furnace was set in the selected temperature and the sample was placed inside for 1 h. After, the sample was cooled down for 24 h in a fume hood, at 20 °C.

## 2.3 Mortar formulation and samples curing

Mortars production was carried out based on EN 196-1 guidelines [22]. The volumetric proportion of binder, aggregate and water used was 1:3:0.5. This proportion ensured an adequate filling of the sand volume by the binder paste. Also, the water content was kept constant in all formulations to guarantee a proper comparison between the conventional production and the use of mining residues and its influence on workability. A Control mortar was produced only with cement. The remain mortars were produced replacing 10, 25 and 50% of the cement weight content by mining residues submitted exclusively to the ED treatment or coupled with the thermal procedure. Table 1 presents the mortars' code, materials loose bulk density, mass proportion and contents of materials. Loose bulk densities of the materials used for mortars formulation were determined by the quotient of the weight of a recipient with known volume, filled with each uncompacted material (volume in kg/m<sup>3</sup>).

**Table 1**

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Materials' loose bulk density, percentual weight and contents of binder, aggregate and water, and mass proportions of mortars formulation.

Mortar code	Binder		Aggregate		Water		Mass proportion			
	CEM II/BL 32.5 N (1000 kg/m <sup>3</sup> )	Mining residues – Electrolytic (1003 kg/m <sup>3</sup> ) & thermal treatment (970 kg/m <sup>3</sup> )	Sand (1540 kg/m <sup>3</sup> )	Tap water (1000 kg/m <sup>3</sup> )	Cement:residue:sand					
	Wt (%)	Wt (g)	Wt (%)	Wt (g)	Wt (%)	Wt (g)	Binder:sand:water			
Control	100	432	–	–					1:4.63:0.77	1:0:4.63
E10	90	389	10	42					1:4.64:0.77	1:0.11:5.14
E25	75	324	25	105					1:4.66:0.78	1:0.32:6.17
E50	50	216	50	210	100	2000	100	333.3	1:4.69:0.78	1:0.97:9.27
ET10	90	389	10	44					1:4.62:0.77	1:0.11:5.15
ET25	75	324	25	111					1:4.60:0.77	1:0.34:6.17
ET50	50	216	50	222					1:4.57:0.76	1:1.03:9.26

Notation: Control - Reference mortar; E – mortars with electrolytic treated mining residues; ET – mortars with electrolytic and thermal treated mining residues. E10, E25 and E50 mortars are also analysed in [15] in comparison to mortars using the raw mining residue (without any treatment).

Mortars were produced in triplicate in 40 mm × 40 mm × 160 mm metallic moulds. For each formulation, circular samples with 90 mm diameter and 20 mm width were also produced. Samples were kept inside polyethylene closed bags at 20 °C and demoulding was carried out after 48 h (based on EN 196-1 [22]), to guarantee setting before the curing conditions. Mortars were cured horizontally in a tap water bath for the next 26 consecutive days at 20 °C. The samples were then dried at 60 °C until constant weight.

## 2.4 Mortars' testing campaign

### 2.4.1 Fresh state

Flow consistency tests (EN 1015-3 [24]) and wet bulk density (EN 1015-6 [25]) were performed. To measure the slump, a flow table test was used while for the bulk density a mould (volume known) was weighted and filled with the mortar specimen.

### 2.4.2 Thermal conductivity

Circular samples were stabilized in a climatic chamber for 24 h (20 °C, 75% RH). The determinations were carried out inside the chamber with an Isomet 2104 Heat Transfer Analyzer (Applied Precision, Slovakia) equipped with a contact probe API [Instruction: Please, correct the code to API 210412. Thank you]210412 (Ø = 60 mm) and operation range of 0.04–0.3 W/(m.K).

### 2.4.3 Dry bulk density and dynamic modulus of elasticity

Based on EN 1015-10/A1 [26], dry bulk density was calculated by the quotient of the mass (digital scale precision = 0.001 g) by the volume (digital caliper precision = 0.01 mm) of the prismatic samples. Using a Zeus Resonance Meter ZMR 001 equipment, the dynamic modulus of elasticity (DME) was determined based on EN [Instruction: Please, correct the standard, to EN 14146. Thank you]14146 [27]. Considering the mass and volume of each sample, the response to an induced vibration signal along samples was determined. Four nondestructive tests were performed, resulting in twelve tests per sample.

### 2.4.4 Flexural and compressive strength

A Zwick/Rowell Z050 equipment was used to determine flexural and compressive strengths according to EN 1015-11 [28]. A three-point bending test was carried out with the prismatic samples, with two steel supporting rollers separated in 100 mm regarding flexural strength determination. The loading was progressively applied at a constant

rate of  $50 \pm 10$  N/s. Compressive strength tests were performed with one half of each sample resulting from the previous flexure tests. The loading was applied at a constant ratio of  $2400 \pm 200$  N/s.

### 2.4.5 Capillarity absorption and drying

Capillarity absorption (EN 1015-18 [29]) and drying (EN 16322 [30]) tests were conducted at  $20^\circ\text{C}$  and 65% RH. The assays were performed on cubic specimens (40 mm) cut from one half of each sample resulting from the flexure tests. Cubes lateral faces were waterproofed with Sikagard 570 W (Sika, Portugal). Specimens were placed on a plastic grid, into a plastic box, where the cut surface was maintained in contact with water (5 mm). The capillarity absorption curve was determined by weighing the samples until an asymptotic value was achieved, in mass increase per area in contact with water by square root of time. Both capillarity absorption coefficient and asymptotic values were calculated.

The drying capacity was tested immediately after capillarity tests inverting and moving the cubes to a waterproof surface and weighing the samples until constant weight. From mortars' drying curves (mass decrease per drying area), rates of first (by time) and second (by square root of time) drying phases were calculated by the negative slope of the initial and intermediate linear segment, respectively. The drying index (DI) was determined based on EN 16322 [30].

### 2.4.6 Mineralogical characterisation

The mineralogical characterisation of mortars was carried out through an X-ray microdiffractometer (BRUKER Discovery, Germany). The global and fine fractions of samples were analysed. Test specimens were prepared from the samples used for compression test. For the global fraction analysis 4 g of mortar were milled and sieved at 0.106 mm, while for the fine fraction analysis 2 g of the binder paste were milled and sieved also at 0.106 mm. The pastes were obtained by previously removing sand particles of each mortar.

Thermogravimetric and differential thermal analysis (TGA/DTA) of global fractions were performed in a Setaram TGA92 equipment in argon atmosphere (3 L/h) and with platinum–rhodium crucible of 50  $\mu\text{L}$  capacity. The heating rate was  $10^\circ\text{C}/\text{min}$ , from  $20^\circ\text{C}$  to  $1000^\circ\text{C}$ .

### 2.4.7 Pore size distribution

For pore size distribution, a mercury porosimeter Micromeritics Autopore II was used. Samples with around  $1.7\text{ cm}^3$  were collected from mortars and stabilized in an oven for 24 h at  $40^\circ\text{C}$ .

### 2.4.8 Cost-efficiency analysis

The cost-efficiency analysis was performed using a simplified methodology. Cost effectiveness factor (CEF) of mortars was determined applying Eq. (1) [31]:

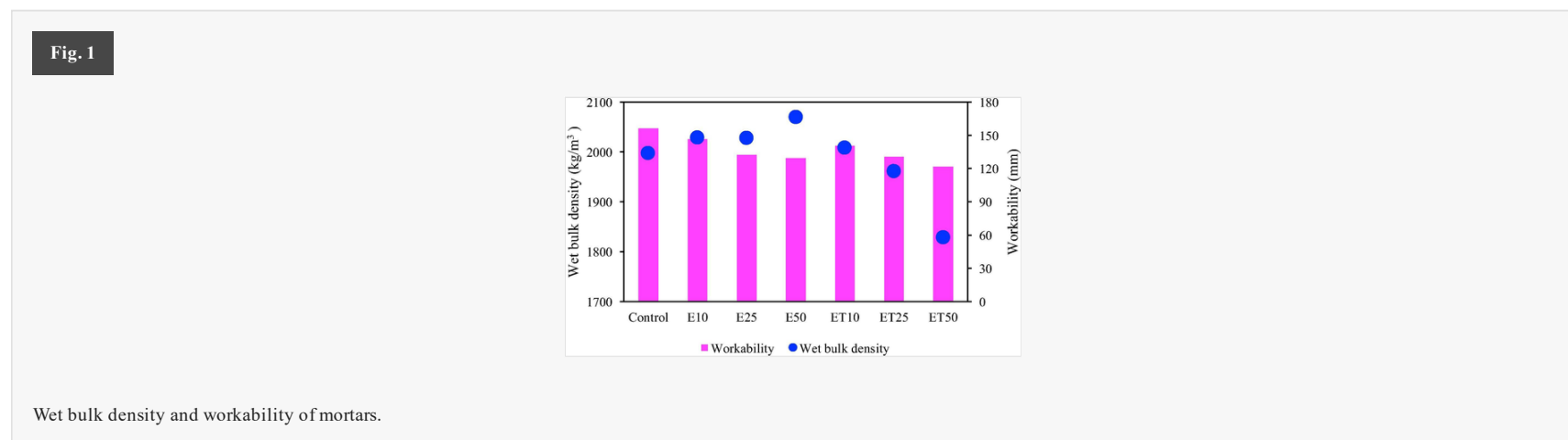
$$CEF_{f/c} = \frac{F_{f/c}}{C} \times 100 \quad (1)$$

where  $CEF_{f/c}$  is the ratio of the flexural ( $F_f$ ) or compressive strength ( $F_c$ ) of mortars to the total material cost per  $\text{m}^3$  ( $C$ ). Local prices of construction materials were used to determine cement cost: 0.12 €/kg CEM II/BL 32.5 N [32]. Sand and water costs were not considered since they are a constant in all mortars. The costs of mining residues resulting from the ED treatment were considered null, once a residue from the mining industry is being reused after a secondary recovery of critical raw materials and removal of harmful elements. When mining residues are submitted to a thermal process, there is an increased cost related to the energy requirements. In this sense, an extra cost of 0.05 €/kg was assumed for ET samples.

## 3 Results and discussion

### 3.1 Fresh state

Both flow table consistency and wet bulk density are presented on Fig. 1. Wet bulk density of E mortars showed higher values when compared to the Control, increasing as the content of mining residues increased. On the other hand, ET mortars demonstrated the inverse trend, since as the content of mining residues increased, the wet bulk density decreased.



Loose bulk density of materials and filler properties may affect mortars wet bulk density [33]. The loose bulk density of the uncompacted materials, presented in Table 1, demonstrated that mining residues treated with only ED are slightly denser ( $1003\text{ kg/m}^3$ ) than cement ( $1000\text{ kg/m}^3$ ), corroborating the higher wet bulk density of mortars prepared with ED mining residues. On the other hand, mining residues treated with both ED and thermal treatment showed a lower bulk density ( $970\text{ kg/m}^3$ ) than cement, promoting a decrease trend on mortars wet bulk density when higher contents of treated mining residues were introduced in the formulations.

Both E and ET mortars reduced their workability, proportionally to the residue incorporation increase. Mining residues particles may absorb more water because of their permeability and surface properties. Due to cement hydration reactions, ET mortars may require more water in sequence  $\text{ET50} > \text{ET25} > \text{ET10}$  to achieve the same consistency of Control. Mining residues crystalline phases and inert characteristics may potentiate the hindering of chemical bonding between hydration products [34].

Generally, the higher incorporation of mining residues reduced mortars plasticity compared to Control, which can be tackled with the addition of a plasticizer as admixture.

### 3.2 Mineralogical properties

The mineralogical composition obtained by XRD of mortars is presented in Table 2. Both global - G - and fine - F (binder enriched) - fraction analysis was conducted, and proportions of the main minerals detected were reported.

Table 2

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Fraction	Experiment													
	Control		E10		E25		E50		ET10		ET25		ET50	
	G	F	G	F	G	F	G	F	G	F	G	F	G	F
Quartz	+++	+/+	+++	+/+	+++	++	+++	+/+	+++	+	+++	+/+	+++	++
Feldspar	++	+/+	++	+	+/+	+/+	+/+	+	++	+	++	+/+	+/+	+/+
Muscovite	+	tc/+	+	+	+/+	+/+	+/+	+/+	+	+	+/+	+/+	+	+/+
Chlorite	–	–	?	?	tc	tc/+	tc	tc/+	?	?	?	?	?	?
Amphibole	tc	tc	tc	tc	?	Tc	tc	tc	?/tc	?/tc	tc	?	?	?
Portlandite	+/+	++	++	+/+	+/+	+++	+	+	++	+/+	++	+/+	+	++
Calcite	+/+	++	+/+	++	+/+	++	+/+	+/+	+/+	++	+/+	++	+/+	+/+
Hydrocalumite	–	–	–	–	–	Tc	tc	tc/+	–	–	–	–	tc	tc/+
Hemicarbonate	–	–	tc	tc	tc/+	+	–	tc	Tc	tc/+	tc	tc/+	–	–
Monosulfoaluminate (AFm)	tc	tc/+	?	tc	–	?	–	–	–	–	–	–	–	–
Ettringite (AFt)	–	tc	tc	tc/+	tc	tc/+	–	tc	Tc	tc/+	tc	tc/+	tc	tc/+
Anhydrous clinker portland silicates	tc	tc/+	tc	tc	tc	Tc	tc	tc	Tc	tc	tc	tc	?/tc	?/tc

Notation: G, Global fraction; F, Fine fraction; +, low proportion; ++, medium proportion; +++, high proportion; tc, traces; ?, doubts on the presence; –, not detected.

The hardened cementitious paste consists in: (1) non-hydrated cement particles, as anhydrous clinker grains; (2) hydrated products (e.g. hydrated calcium silicates and calcium aluminates, hydrated sulfoaluminates and calcium hydroxide); and (3) air voids and capillary pores, that are unfilled spaces between the cement grains [35].

The presence of components as quartz, feldspar, mica (e.g. muscovite) and clay minerals were detected in an average of medium/high proportion. Regarding the effect of mineralogy, higher contents of quartz are effective in adhesive strength features. The silica content of the aggregate is also important on tensile bond strength [36]. Clay minerals are common in aggregates, acting as pore lining rims and pore-filling materials [37].

Calcite was detected in all formulations at similar amounts (low/medium proportion). Calcite comes from the carbonation of calcium hydroxide and from the cement used. In CEM II/BL 32.5 N, the calcite content can vary between 21 and 35 wt% [21]. Calcite has filler properties and affects the distribution of alumina and sulfate phases, changing the mineralogy of hydrated cement pastes [38]. The calcite contents in the samples were sufficient to form hemicarbonate [38], detected in E10, ET10, E25 and ET25 mortars at low/trace amounts.

Amorphous silica and silicates are key factors for pozzolanic reactivity and durability properties of mining residues, in presence of  $\text{Ca}(\text{OH})_2$  and water. The use of pozzolans may minimize mortars cost and embodied energy. The pozzolanic contribution can be categorized in physical/filler effect attributed by fine particles and in chemical/pozzolanic effect due to the pozzolanic reaction [39].

The pozzolanic reactivity of mining residues treated under different conditions was assessed through modified Chapelle test [40]. The total amount of fixed  $\text{Ca}(\text{OH})_2$  by siliceous or aluminosilicates amorphous substances from 1 g of raw mining residues was reported as 7.9 mg, from mining residues treated with only ED was 71.4 mg, and coupled with a thermal treatment was 547.6 mg, representing the last high pozzolanic potential [16]. Another study concerning the reuse of solid mining waste in cement-bricks showed that mining waste can achieved good mechanical performance and be considered a pozzolanic material [41].

Furthermore, minor crystalline compounds were identified, as amphibole, monosulfoaluminate (AFm) and ettringite (AFt). Amphibole is an inosilicate mineral that may contains iron (Fe) or magnesium (Mg). Fe was detected in Panasqueira mining residues (Table S2 in appendix A), which may justify the amphibole detection in E10, E25, E50, ET10 and ET25 formulations.

AFm and AFt result from the hydration reaction of calcium aluminates from cement clinker with gypsum. AFm is crucial in anion binding in cement, exchanging its original interlayer ions ( $\text{SO}_4^{2-}$  and  $\text{OH}^-$ ) with chlorides [42]. AFt can slowly dissolve and reform available voids or microcracks and can control stiffening properties. However, AFt may be formed after hardening in the presence of sulfate sources, which may cause materials deterioration due to its expansive behavior. This can lead to cracking of the hardened mortars and concrete, loss of dynamic modulus and, in reinforced concrete, debonding of reinforcing steel bars [43]. AFt was detected in trace proportions/low proportion in all samples, except in the global fraction of the Control mortar. AFm was only detected at small amounts in Control and E10 mortars, which can be justified by the differences in  $\text{Al}_2\text{O}_3/\text{SO}_3$  ratio on these samples.

From TGA/DTA curves (Fig. S1 in appendix A), the mass losses at specific temperature ranges were assessed, as well as  $\text{Ca}(\text{OH})_2$  and  $\text{CaCO}_3$  contents (Table 3).

Table 3

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Mass losses and calcium hydroxide and calcium carbonate contents from TGA/DTA mortars' tests.

Mortar	Mass loss per temperature range (%)		Mass loss of ignition (%)			$\text{Ca}(\text{OH})_2$ (%)	$\text{CaCO}_3$ (%)
	20–230 °C	230–390 °C	390–500 °C	500–900 °C	20–1000 °C		
Control	2.45	0.84	1.03	5.28	9.67	4.23	12.01
E10	2.58	0.77	0.96	4.56	9.00	3.95	10.37
E25	2.27	0.78	0.93	4.78	8.90	3.82	10.87
E50	1.26	0.47	0.44	4.31	6.63	1.81	9.80
ET10	3.64	0.87	1.03	4.19	9.82	4.22	9.53
ET25	2.66	0.72	0.86	3.49	7.76	3.54	7.94
ET50	1.56	0.43	0.32	3.37	5.71	1.32	7.66

The mass loss registered between 20 and 230 °C occurs due to the loss of adsorbed water and the hydrated compounds decomposition (e.g. ettringite). Regarding the range of 230–390 °C, mass losses are mainly from hydrated calcium silicate and calcium aluminate decomposition. The mass loss at 390–500 °C is due to calcium hydroxide decomposition. The losses reported in 500–900 °C interval are essentially caused by carbon dioxide decomposition [44]. Regarding all studied mortars, the higher mass loss occurred between 500 and 900 °C, indicating the presence of carbonates ( $\text{CaCO}_3$ ) and the calcite detected in XRD analysis (Table 2).



Control showed the higher content of  $\text{CaCO}_3$  (approximately 12%), and, as mining residues incorporation increased,  $\text{CaCO}_3$  decreased proportionally in both E and ET cases. The same trend was verified for  $\text{Ca(OH)}_2$  (Table 3). This may be justified by the pozzolanic reaction between portlandite from mortars with thermal treated and/or ED residues and their pozzolanic content. These mortars have lower portlandite contents due to the lower amount of cement, in comparison to Control. In addition, during the thermal treatment, different hydration levels and, consequentially, carbonation, may have contributed to slightly reduce the portlandite detected in ET, when compared to E mortars.

The mass losses from 20 to 500 °C were higher for ET10 (5.54%) > Control (4.32%) > E10 (4.31%) > ET25 (4.24%) > E25 (3.98%) > ET50 (2.31%) > E50 (2.17%), which may be caused by the presence of lower contents of hydrated compounds in sequence.

### 3.3 Physico-mechanical analysis

The loose bulk density of the materials used in mortars is presented in Table 1. Mining residues loose bulk density decreased from 1003  $\text{kg/m}^3$  (ED treatment only) to 970  $\text{kg/m}^3$  (ED and thermal treatment). This may be justified by the extraction of inorganic compounds during the ED process and minerals transformation and/or water losses during the thermal treatment. The replacement of the cement binder (1000  $\text{kg/m}^3$ ) by treated mining residues (970  $\text{kg/m}^3$ ) corresponds to almost the same weight. Thus, a similar filler effect is expected.

Hardened mortar results for dry bulk density, thermal conductivity, compressive and flexural strength and dynamic modulus of elasticity after 28 days of cure are presented in Table 4.

**Table 4**

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Hardened mortars characterisation: thermal conductivity, bulk density, dynamic modulus of elasticity, compressive and flexural strength.

Experiment	Thermal conductivity [W/(m.K)]	Bulk density ( $\text{kg/m}^3$ )	Dynamic modulus of elasticity (MPa)	Compressive strength (MPa)	Flexural strength (MPa)
Control	1.5	1975.56 ± 16.78	7208.66 ± 40.72	18.23 ± 0.76	4.68 ± 0.08
E10	1.4	1971.83 ± 12.35	4147.33 ± 78.78	11.91 ± 1.87	4.18 ± 0.34
E25	1.2	1997.86 ± 28.95	3513.00 ± 186.54	7.53 ± 1.46	3.63 ± 0.30
E50	1.3	2007.24 ± 28.33	1508.67 ± 33.50	4.66 ± 0.52	1.34 ± 0.81
ET10	1.7	2003.56 ± 30.82	8948.33 ± 38.50	10.29 ± 1.76	4.52 ± 0.25
ET25	1.7	1949.07 ± 5.19	3495.67 ± 229.24	5.82 ± 1.02	3.94 ± 0.13
ET50	1.2	1866.68 ± 40.90	1260.33 ± 121.72	3.85 ± 0.27	2.25 ± 0.01

When treated mining residues were introduced, dry bulk density slightly decreased on E10 (1971.8  $\text{kg/m}^3$ ) and ET25 (1949.1  $\text{kg/m}^3$ ), comparing to Control (1975.6  $\text{kg/m}^3$ ). This could be justified by the loose bulk density differences between the materials used, namely residues and cement (Table 1).

The lowest thermal conductivity was 1.2 W/(m.K) for E25 and ET50 samples, meaning that treated mining residues incorporation can improve the properties of mortars. The morphology of the matrix could be more relevant for thermal conductivity, comparing with chemical composition of the components. In addition, the pore system promoted by mortars' aggregates can have more impact in thermal conductivity than properties as total porosity and specific gravity [45].

The thermal conductivity of soil-cement bricks using mining waste also demonstrated a direct relation on the construction product density, once the increase in brick density provided a material with lower heat dissipation characteristics [41].

Mortars with low thermal conductivities have advantages in thermal comfort [46], although it depends on the mortar application. For renders and plasters applied with thicknesses of around 2 cm the differences are not significant. Nevertheless, for bedding mortars, namely when joints are thick, these differentiations may cause important changes.

The results obtained for flexural and compressive strengths, as well as dynamic modulus elasticity (DME), are also presented in Table 4. Mechanical properties of mortars decreased proportionally when cement was replaced by mining residues. As expected, besides both ED and thermal treatment, mining residues are still lower strength materials compared to cement.

The DME increased 19% in ET10, when compared to Control. However, when higher contents of mining residues treated with both ED and thermal treatment were incorporated in mortars the DME decreased from around 7209 MPa (Control) to 3496 MPa (ET25) and 1260 MPa (ET50). In E mortars, DME decreased sequentially: E10 (4147 MPa) > E25 (3513 MPa) > E50 (1509 MPa). Low DME values mean that mortars are more able to absorb deformations from the substrate, which may decrease vulnerability to crack, particularly important on rendering and plastering mortars [47].

The decrease on flexural and compressive strengths is proportional to the increase of mining residues content (Table 4). This trend, due to the lower proportion of cement, may have promoted a retarding effect on cement hydration for both E and ET formulations.

Comparing to Control, flexural performance decreased 11, 22 and 71% in E10, E25 and E50, respectively. Lower flexural decreases were observed in ET mortars, where 3, 16 and 52% decays were registered for ET10, ET25 and ET50, respectively. Also, for the same formulations, a decrease of 35–74% (E) and 44–79% (ET) on compressive strength was verified, showing that the loss in compression was more accentuated. This decrease can be related to the low pozzolanic activity of mining residues.

Calcined clays were studied as a supplementary cementitious material [48]. The flexural strength could be improved with calcined materials, even at a high cement substitution. However, the decrease in compressive strength was also verified, probably due to the dilution effect. The activity of calcined materials depends on the content and composition of the reactive silica, fineness and calcination. Inadequate grinding may reduce mechanical properties although a progress on anti-cracking properties of plastering mortars could be achieved [48]. The introduction of magnesia, for instance, as an additive in construction products may lead to a greater compressive strength and minimize the chances of micro-crack occurrence due to a delayed hydration [49].

DME, flexural and compressive strengths are correlated in 67% (DME vs. Fstr), 57% (Cstr vs. Fstr) and 55% (DME vs. Cstr). All these properties decreased when cement was replaced by mining residues in the formulations. However, some properties assume more relevance for certain mortars application. For renders and plasters, resistance to crack and capacity of deformability are important characteristics. The DME/flexural strength ratio is an indicator, where lower values mean more appropriate mechanical performance. Thus, mortars showed better behaviors in sequence: ET50 > ET25 > E25 > E10 > E50 > Control > ET10.

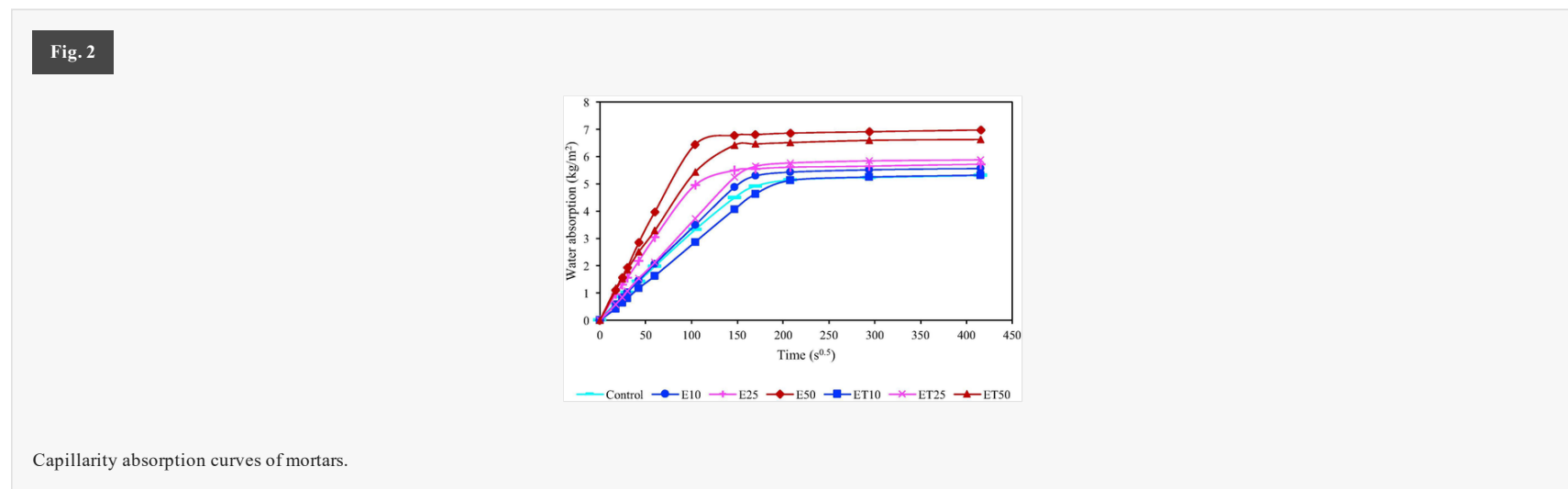
Based on EN 998-1 [50] and EN 998-2 [51], E and ET mortars can be classified considering compressive strength data at 28 days and their application (Table S3 in appendix A).

For rendering or plastering, E and ET mortars are classified as CS III or CS IV (the latter as the Control). Regarding masonry bedding mortars, E and ET formulations are categorized as M5 and M10, while the Control classification is M15. Nevertheless, several applications of mortars require relatively low compressive strength, meaning that upper classes are not often needed. Therefore, the mortars studied showed mechanical properties compatible with conventional concrete and contemporary fired brick

masonry walls. For traditional vernacular walls applications, E and ET mortars are still exceedingly stiff. Other studies corroborated the suitability of using mining waste to partially replace cement in concrete (up to 10%) and reported the considerable decrease in compressive resistance [52].

### 3.4 Capillary absorption and drying capacity

The average capillarity curves of mortars are presented in Fig. 2. Through this data is possible to address the initial absorption rate and the total water absorbed from samples by the determination of the capillary coefficient and the asymptotic value, respectively.



Considering capillary absorption of mortars, the capillary curves showed that E10, E25, E50, ET25 and ET50 behaved negatively in comparison to Control. In Table 5 is possible to observe an increase in the initial rate (capillarity coefficient) and in the total absorbed water (capillarity saturation value), in a proportional way to the residue increase. E50 [ $0.067 \text{ kg}/(\text{m}^2 \cdot \text{s}^{0.5})$ ] and ET50 [ $0.059 \text{ kg}/(\text{m}^2 \cdot \text{s}^{0.5})$ ] represented the worst cases of capillarity coefficient, once these formulations absorbed water at a faster ratio and in higher contents (E50 =  $6.976 \text{ kg}/\text{m}^2$  and ET50 =  $6.626 \text{ kg}/\text{m}^2$ ). ET10 showed an improvement in the initial absorption rate in relation to Control, where a similar asymptotic value (approximately  $5.3 \text{ kg}/\text{m}^2$ ) was achieved at a slower rate [Control =  $0.033 \text{ kg}/(\text{m}^2 \cdot \text{s}^{0.5})$ ; ET10 =  $0.028 \text{ kg}/(\text{m}^2 \cdot \text{s}^{0.5})$ ]. This behavior may affect abrasion and acid attack resistance and improve mortars durability [10]. The outcomes of another study suggest that molybdenum mine tailings up to 10% can be used to produce cement mortar with improved durability levels [53].

**Table 5**

*i* The table layout displayed in this section is not how it will appear in the final version. The representation below is solely purposed for providing corrections to the table. To preview the actual presentation of the table, please view the Proof.

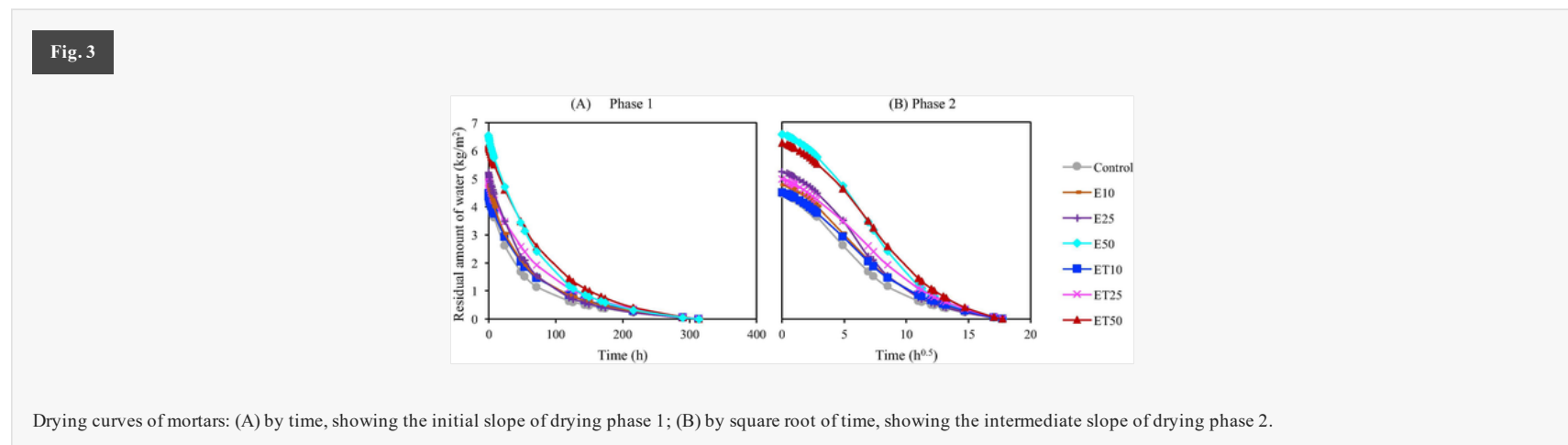
Capillarity absorption coefficient, capillarity saturation value, drying rates of phases 1 and 2 and drying index after 314 h, of mortars.

Mortar	$C_{ac} [\text{kg}/(\text{m}^2 \cdot \text{s}^{0.5})]$	$C_s (\text{kg}/\text{m}^2)$	$DR_1 [\text{kg}/(\text{m}^2 \cdot \text{h})]$	$DR_2 [\text{kg}/(\text{m}^2 \cdot \text{h}^{0.5})]$	Drying index
Control	$0.033 \pm 0.001$	$5.305 \pm 0.593$	$0.204 \pm 0.003$	$0.456 \pm 0.002$	$0.216 \pm 0.009$
E10	$0.034 \pm 0.000$	$5.564 \pm 0.041$	$0.206 \pm 0.011$	$0.444 \pm 0.006$	$0.257 \pm 0.000$
E25	$0.051 \pm 0.000$	$5.717 \pm 0.742$	$0.200 \pm 0.019$	$0.507 \pm 0.016$	$0.228 \pm 0.000$
E50	$0.067 \pm 0.005$	$6.976 \pm 0.198$	$0.221 \pm 0.007$	$0.539 \pm 0.004$	$0.284 \pm 0.001$
ET10	$0.028 \pm 0.006$	$5.320 \pm 0.129$	$0.200 \pm 0.012$	$0.405 \pm 0.005$	$0.275 \pm 0.001$
ET25	$0.036 \pm 0.003$	$5.882 \pm 0.106$	$0.213 \pm 0.010$	$0.390 \pm 0.006$	$0.323 \pm 0.001$
ET50	$0.059 \pm 0.006$	$6.626 \pm 0.271$	$0.206 \pm 0.008$	$0.474 \pm 0.003$	$0.350 \pm 0.011$

Notation:  $C_{ac}$  – Capillarity absorption coefficient;  $C_s$  – Capillarity saturation value;  $DR_1$  – Drying rate (phase 1);  $DR_2$  – Drying rate (phase 2).

The samples internal cure may have influenced water absorption by mortars, once mining residues particles may had retained more water compared to conventional cement [54]. The short term cement hydration may have been hindered, changing the porous structure [34]. Both mortars microstructure and strength can be affected by mining residues incorporation because of the lower structuring of a stiff hydrated skeleton that could resist to an internal pressure promoted by capillary stress from self-desiccation [55].

The presence of water can damage mortars due to salts solubilization or harmful substances entry. A high drying capacity improves mortars durability and avoid fungi/algae growth [56]. Through the drying curves of mortars (Fig. 3) as function of time (drying phase 1 by a linear segment) and square root of time (drying phase 2 by a linear segment), drying rates were determined for phases 1 ( $DR_1$ ) and 2 ( $DR_2$ ), as shown in Table 5. The moisture movement during drying is often described by a diffusion process that depends on the moisture content. The moisture diffusivity is related to water in phase 1 and to vapor in phase 2 [57].



The  $DR_1$  was approximately  $0.2 \text{ kg}/(\text{m}^2 \cdot \text{h})$  for all tested mortars, although the lowest absolute value was observed for E25 and ET10, and the highest for E50. Regarding  $DR_2$ , the highest rate was observed for mortars with 50% of mining residues incorporation, meaning an increase in the evaporation rate due to improved diffusion and permeability properties.

The presence of soluble salts during the drying process may potentiate their crystallization in the sample surface causing staining [58]. In the samples studied, no salts crystallization was observed during drying. Thus, treated mining residues did not affect mortars drying phases.

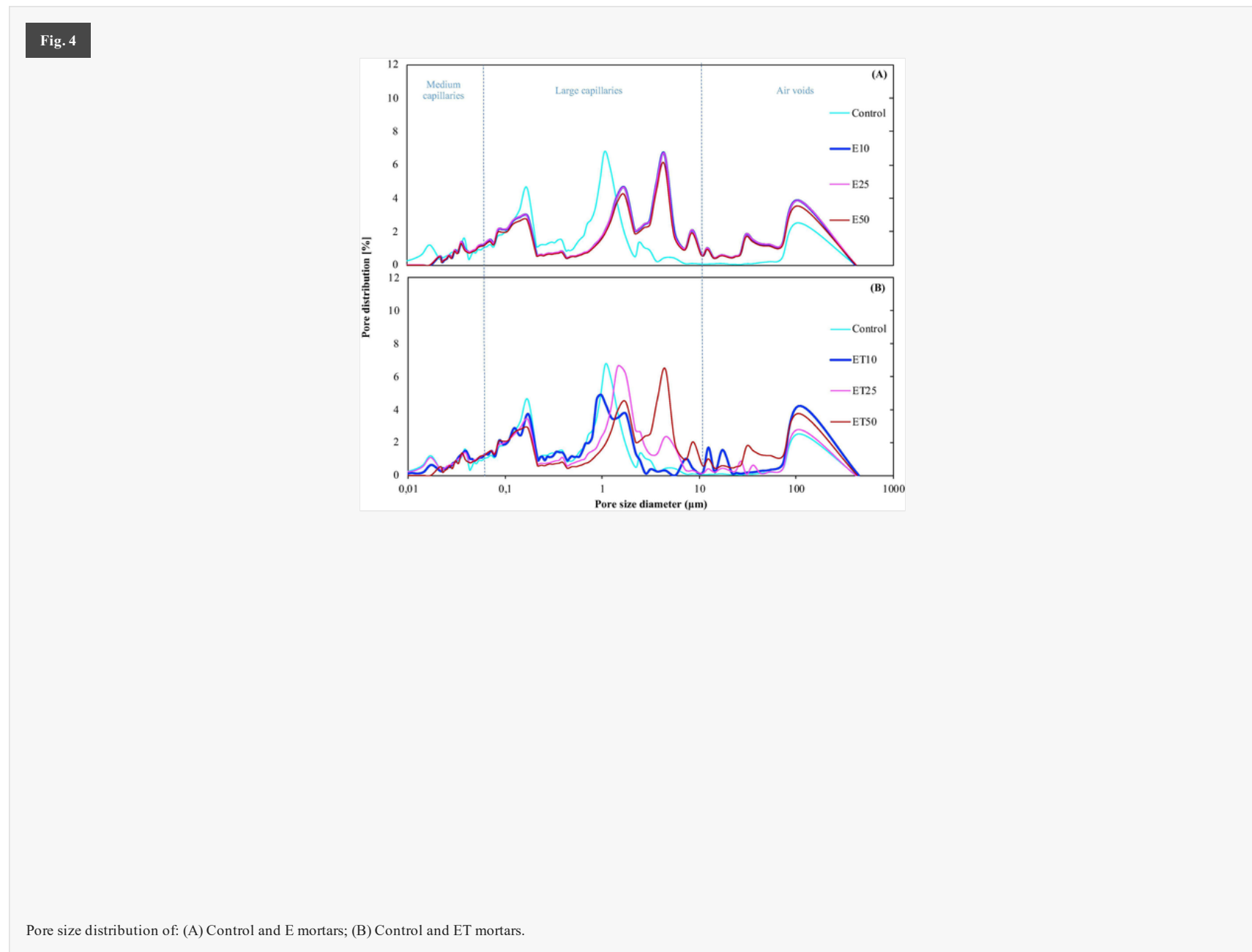
The drying index of mortars was also determined (Table 5). The lower drying index was found for Control (0.216), followed by E25 (0.228), E10 (0.257), ET10 (0.275), E50 (0.284), ET25 (0.323) and ET50 (0.350). Mortars with mining residues demonstrated a harder capacity to completely dry, proportionally to the residue content in ET

formulations.

During the drying process, the behaviour of mortars can be related to their water absorption, since transport mechanisms of water occur [56]. A faster transport velocity of water promotes a higher water absorption on the evaporation during the first drying phase.

### 3.5 Pore size distribution

The analysis of the porosimetry was carried out to compare Control, E and ET mortars. The open porosity was lower in sequence E50 (25.3%) > E10 (23.9%) > ET50 (23.7%) > ET25 (23.4%) > E25 (22.2%) > Control (22.0%) > ET10 (20.3%). Nevertheless, the samples revealed similar porosities, meaning that cement replacement by mining residues did not affected expressively the open porosity of mortars. The pore size distribution of Control, E and ET mortars is presented on Fig. 4.



Considering the range of large mesopores (medium capillaries), ET25 and Control mortars are dominant. For pore sizes lower than 0.05  $\mu\text{m}$ , the thermal treatment seems to induce differences in porosimetry, particularly in ET10 and ET25 mortars, that are closer to the Control sample porosimetry. On macropores (large capillary pores), Control has a prevalence until approximately 2  $\mu\text{m}$ . This could be justified by the clinker phase once the Control mortar has the highest content of cement [59]. The water retention by E and ET mortars during short term hydration may have been higher than cement [60].

After 2  $\mu\text{m}$  of pore size diameter, different trends were observed between E and ET formulations. In E mortars, pore size distribution behaved similarly and independently of the proportion of mining residues applied, while in ET mortars the distribution oscillated as function of the mining residues proportion. The peaks observed were: Control = 1.09  $\mu\text{m}/6.78\%$ ; E10 = 4.47  $\mu\text{m}/6.65\%$ ; E25 = 4.51  $\mu\text{m}/6.62\%$ ; E50 = 4.46  $\mu\text{m}/6.05\%$ ; ET10 = 1.45  $\mu\text{m}/3.50\%$ ; ET25 = 1.76  $\mu\text{m}/6.12\%$  and ET50 = 4.48  $\mu\text{m}/6.41\%$ . Hydrated elements formation, associated to hydration and carbonation reactions at early stages, could have been affected by lower cement contents [59].

Regarding the air voids interval, while for E mortars the influence of the cement replacement percentage is insignificant in porosimetry, for ET mortars it differs with the cement replacement ratio. ET50 mortar remains almost unchanged whereas the E25 case showed porosimetry similar to the Control mortar.

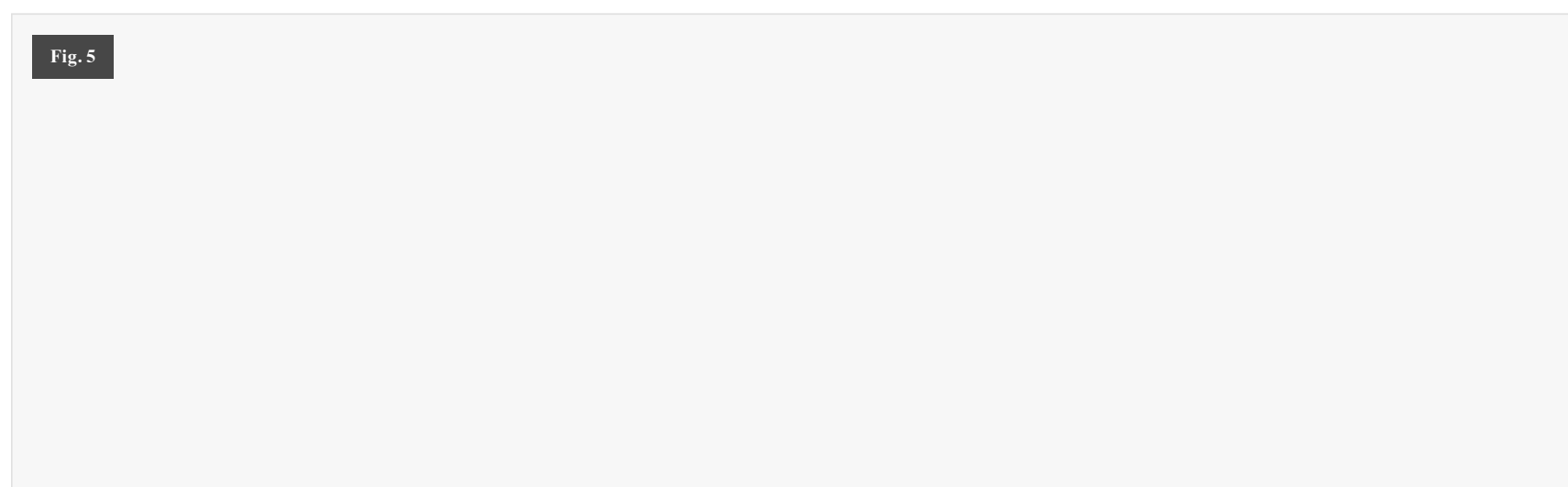
The trend observed for capillary water absorption may be related to the mortars porosimetry increase with the residue content increase, considering the interval 1–6  $\mu\text{m}$ . The high porosimetry of E50 and ET50 justified the high capillary water absorption observed.

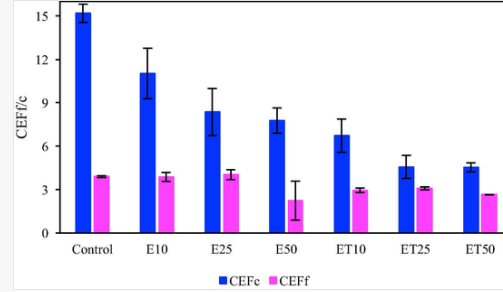
Samples with mining residues showed a rising trend in the evaporation rate. Their higher capillary, total porosity and pore diameter, in comparison to Control, justify this increase. Additionally, low drying index are related to a high large capillary porosity of samples [59].

Strength and durability may be influenced by samples compactness and air/liquid losses [61]. Generally, the increase in volume and diameter pore size of the coarse porosity promoted a decrease on mechanical performance, more pronounced in compressive strength case (Table 4).

### 3.6 Cost-efficiency analysis

Fig. 5 presents the cost effectiveness factor (CEF) of the mortars determined based on Eq. (1), considering flexural and compressive strength data (Table 4).





Cost effectiveness factor (CEF) of cement-based mortars incorporating mining residues as cement replacement. Notation: Cost ( $\text{€}/\text{m}^3$ ): Control – 120; E10 – 108; E25 – 90 ; E50 – 60 ; ET10 – 153 ; ET25 – 128 and ET50 – 85. CEF<sub>c</sub>-Cost effectiveness factor determined with compressive strength data; CEF<sub>f</sub>-Cost effectiveness factor determined with flexural strength data.

Although including a residue in the formulation, the cost assumed for ET mortars is only lower than Control for ET50 (Fig. 5 notation). That occurred due to the energy cost determined to prepare ET mining residues. The results could differ depending on the efficiency of the thermal treatment, emphasizing the influence that residues treatment may have in the overall analysis.

For compressive strength ratios, the Control mortar presented the highest CEF<sub>c</sub> ( $15.2 \pm 0.6$ ). The increase in the replacement level of mining residues resulted in a decrease of the cost effectiveness factor for E and ET mortars. Thus, for applications that need to comply high compressive resistances, e.g. masonry bedding mortars with high strength masonry units, Control is the most feasible option, followed by E10 ( $11.0 \pm 1.7$ ), E25 ( $8.4 \pm 1.6$ ), E50 ( $7.8 \pm 0.9$ ), ET10 ( $6.7 \pm 1.2$ ), ET25 ( $4.6 \pm 0.8$ ) and ET50 ( $4.5 \pm 0.3$ ). However, lower strength products are frequently used for bedding mortars and conventional masonry units.

Considering applications where flexural strength assumes more relevance, e.g. plastering and rendering mortars, E25 ( $4.0 \pm 0.4$ ) presented the highest CEF<sub>f</sub>, in comparison to Control ( $3.9 \pm 0.1$ ). The increase in the replacement level of mining residues resulted in an increase in the cost effectiveness factor of mortars, although the flexural strength decrease verified in E50 (Table 4) did not corroborate this trend. Most probably, an optimal cement replacement percentage between 25 and 50% would optimize flexural strength.

In an environmental perspective, the mechanical performance of ET mortars was not considerably improved when compared to E formulations, except for ET50 case. Although a decrease in compressive strength was registered for ET50 in comparison to E50 mortar (from  $4.7$  to  $3.9 \text{ kg}/\text{mm}^2$ ), a substantial increase on flexural strength was obtained (from  $1.3$  to  $2.2 \text{ kg}/\text{mm}^2$ ). The high dispersion registered by ET50 mortar and the superior energetic needs, that will promote equally higher CO<sub>2</sub> emissions release and cost, may not reward the thermal treatment. Further tests should be conducted to determine the technical and environmental feasibility at an industrial scale, for both ED and thermal treatments.

## 4 Conclusions

The reuse of mining residues in cementitious mortars contributes to reduce impacts in quarries, from where the calcareous marl is extracted, to minimize CO<sub>2</sub> impacts from cement manufacturing and to sustain a balanced ecosystem at mining sites. In this work, mining residues from Panasqueira mine were treated with an electro-dialytic (ED) technology, aiming the removal and recovery of elements, alone or followed by a thermal treatment to enhance pozzolanic reactivity and durability. The effect of incorporating treated mining residues on technical characteristics of mortars, by replacing cement in 10, 25 and 50% (wt%), was addressed:

- The mineralogical composition of mortars produced with both thermal and/or ED treated mining residues validated the compatibility of this material with cement mortars and the removal of elements.
- As expected, mining residues mortars are lower strength products compared to cement mortars. The increase in the replacement level of cement by mining residues treated with ED and thermal processes decreased both flexural (3 to 52%) and compressive strength (44 to 79%), comparing to the Control mortar.
- Once ED and thermally treated mining residues have demonstrated pozzolanic reactivity, the water curing optimization may be needed to enhance the pozzolanic reaction in mortars. In future studies, the influence of a humid curing should be addressed.
- Since a large majority of mortars applications require relatively low compressive strength, the studied mortars showed properties compatible with conventional concrete and contemporary fired brick masonry walls, and mining residues feasibility as cement replacement.
- An improvement in the initial absorption rate was only achieved when 10% of mining residues ED and thermally treated were incorporated in mortars, comparing to the reference, once a similar asymptotic value was obtained at a slower rate.
- The cost effectiveness analysis also validated the feasibility of treated mining residues reuse in cementitious mortars, namely considering applications where flexural strength is key factor (e.g. renders and plasters). However, further studies should be conducted to assess the technical and environmental feasibility of using ED mortars at an industrial scale and to quantify the optimized impacts of the thermal treatment.

## CRedit authorship contribution statement

**J. Almeida:** Conceptualization, Methodology, Writing - original draft, Visualization, Writing - review & editing, Software, Investigation. **P. Faria:** Conceptualization, Validation, Supervision, Writing - review & editing, Funding acquisition, Resources. **A.B. Ribeiro:** Validation, Supervision, Writing - review & editing, Funding acquisition, Resources. **A. Santos Silva:** Methodology, Validation, Supervision, Resources, Writing - review & editing.

## Declaration of competing interest

The authors have no affiliation with any organization with a direct or indirect financial interest in the subject matter discussed in the manuscript.

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## Appendix A Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.conbuildmat.2021.122483>.

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## Highlights

- Mining residues were submitted to an electrodialytic process alone or coupled with a thermal treatment.
  - Treated mining residues replaced cement from 10 to 50 wt% in mortars.
  - Flexural strength of mortars decreased from 3 to 52%, showing potential for renders.
  - Mortars' cost-efficiency with only electrodialytic treatment could be upgraded.
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## Appendix A Supplementary material

The following are the Supplementary data to this article:

[Multimedia Component 1](#)

**Supplementary data 1**

## Queries and Answers

Q1

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