

Accepted Manuscript

Title: Sustainability of construction materials: electrolysytic technology as a tool for mortars production

Authors: C Magro, JM Paz-Garcia, LM Ottosen, EP Mateus, AB Ribeiro



PII: S0304-3894(18)30900-2
DOI: <https://doi.org/10.1016/j.jhazmat.2018.10.010>
Reference: HAZMAT 19830

To appear in: *Journal of Hazardous Materials*

Received date: 3-10-2017
Revised date: 22-9-2018
Accepted date: 3-10-2018

Please cite this article as: Magro C, Paz-Garcia J, Ottosen L, Mateus E, Ribeiro A, Sustainability of construction materials: electrolysytic technology as a tool for mortars production, *Journal of Hazardous Materials* (2018), <https://doi.org/10.1016/j.jhazmat.2018.10.010>

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

Sustainability of construction materials: electrodialytic technology as a tool for mortars production

C Magro^{1,2,*}, JM Paz-Garcia³, LM Ottosen², EP Mateus¹, AB Ribeiro^{1,*}

¹CENSE, Departamento de Ciências e Engenharia do Ambiente, Faculdade de Ciências e Tecnologia, Universidade Nova de Lisboa, Caparica 2829-516, Portugal

²Department of Civil Engineering, Technical University of Denmark, Lyngby, Denmark

³Department of Chemical Engineering, University of Malaga, Malaga, Spain

**corresponding author*

Highlights

- Electrodialytic technology (ED-T) removal rates: between 50-99.7%
- Mortars mix with the replacement of 50% and 100% of ED-T effluent showed similar initial setting times, workability, flexural and compressive strengths compared to the mortar reference
- ED-T can enhance the sustainability of the building industry

Abstract

The reduction of tap water consumption in all activity sectors, including the building industry, is crucial to the sustainability of water resources. Effluents from wastewater treatment plants have the potential to replace freshwater in the construction sector but they contain a critical mixture of impurities, which hampers their use in mortars production. In this work, the viability of using effluent as an alternative to potable water for the production of mortars, after electro-dialytic treatment, was assessed.

Electrodialytic technology (ED-T) is a proven technique for decontamination of porous and aqueous matrices.

ED-T experiments were conducted with 500 mL of effluent for 6, 12 and 24h, with a current intensity of 25 mA. The results showed that after ED-T 6h, the removal efficiencies of critical components were above 85% of their initial concentrations. Mortar properties such as setting time, workability, flexural strength,

compressive strength and morphology were obtained for 100% effluent and tap water/effluent mixtures (50:50) with and without ED-T pre-treatment. The mortars with the ED-T treated effluent showed similar initial setting times and workability, higher flexural and compressive strength compared to the mortars reference.

Keywords: electro dialytic technology, mortars production, secondary water resources, circular economy

1 Introduction

The world's freshwater resources are in short supply, as economic activities, population growth, climate change, and the lack of appropriated resources management have caused a significant increase of water use. The United Nations' Sustainable Development Goals stated in 2015 that: water scarcity affects more than 40% of the global population and is estimated to rise [1]. Currently, water scarcity affects an estimated 1.9 billion people and 2.1 billion people live with drinking water services not safely managed [2]. The critical point of water scarcity has led scientists to look for new and efficient ways to make the most out of nontraditional sources, including sea water, brackish water and wastewater. To match the demand of "zero-waste" system, it is essential to find solutions that minimize the consumption of freshwater.

Concrete is the second most consumed material in the world after water, being directly related to the use of freshwater [3]. Water use for the concrete industry represents 9% of all industry withdrawals and 1.7% of all global water consumption [4]. In a concrete mix, the water-cement ratio is around 0.45 to 0.60. In other words, more than 17 billion m³ of freshwater are used in the production of concrete per year [4]. Nowadays, freshwater is the only type of water that fulfils the industrial standards, for quality and safety demands. In fact, if polluted water was used, the quality of the concrete would be significantly lower, leading to structural failures that would represent 5% to 9.4% of the total costs in concrete production [5].

Various sources of non-freshwater have already been tested as alternatives to freshwater in concrete mixtures, such as (a) processed wastewater sludge [6], (b) polyvinyl acetate resin wastewater [7], (c) car washing wastewater [8], (d) untreated/treated textile effluent [9] and (e) domestic wastewater before chlorination [10]. Noruzman et al. [11] proved that the use of non-freshwater in the concrete mix can provide a proper mix design and acceptable standards regarding tolerable limits. However, due to the composition specifications and to different types of impurities that can exist in each type of water, it is difficult to draw a sound conclusion. Thus, some improvements in past studies need to be addressed in order to take water reuse into

consideration: (1) expected degree of human contact, (2) concentration of microbiological and chemicals and, (3) which pre-treatments are necessary [12].

The electro-dialytic technology (ED-T) consists on the application of a low level current density, either direct current or alternate current, of a few mA/cm and a low potential gradient of V/cm, between suitably located electrodes [13]. In the ED-T, ion-exchange membranes are used to separate the matrix to be treated from the electrode compartments. An anion-exchange membrane (AEM) is used at the anode end and a cation-exchange membrane (CEM) at the cathode end. This set-up allows selective separation of anions and cations from the matrix. Cationic species move towards the cathode through the CEM, that hinders the transport of anions and anions move towards the anode through the AEM, that only partially hinders the transport of protons, as it does not work as a perfect rectifier. The ED-T has proved to be efficient for the removal of organic or/and inorganic contaminants, such as heavy metals and salts [14], in decreasing organic matter and fats [15] and in the inactivation of microorganisms in fresh sewage sludge from wastewater treatment plant (WWTP) [16].

Therefore, the present study aims to investigate the use of effluent from an urban WWTP secondary treatment, after electro-dialytic process in mortars' production. The physical and functional mortar properties were performed and compared with the industry main standards.

2 Materials and Methods

2.1 Materials

Tap water was collected at the Technical University of Denmark, Lyngby, Denmark. The effluent was sampled after the secondary clarifier at Mølleåværet A/S WWTP in Lyngby, Denmark. The plant uses the BioDenitro method in the aeration tanks and no tertiary treatment [17]. The plant treats 8-12 million m³ of wastewater on a yearly basis, corresponding to a 115 000 population equivalent.

The cement used for mortar preparation was the Ordinary Portland Cement (OPC) and the sand was the CEN standard sand.

2.2 Tap water and effluent characterization

All experiments were carried out in triplicate.

The pH in the effluent and tap water were measured with a Radiometer pH-electrode and conductivity in a Radiometer Analytic.

Total concentrations of Ca, K, Mg, Ni, P, S, Si and Zn were determined by Inductively Coupled Plasma with Optical Emission Spectrometry (ICP-OES), and Cl^- , NO_3^- and SO_4^{2-} by Ion Chromatography (IC).

The regulatory parameters for effluent discharge in water bodies – namely biological oxygen demand (BOD_5), chemical oxygen demand (COD), NH_4^+ , NO_2 and N_{total} – were determined by Spectrophotometer DR2800 kits, and total suspended solids (TSS) by gravimetric analysis.

2.3 Electrodialytic experiments

Three different electrodialytic experiments ED-T were carried out, in triplicate, according to the conditions presented in Table 1.

The ED-T cell used (Figure 1) was a 3 compartment conventional one [18], made of plastic (RIAS, Acryl XT). The internal diameter was 8 cm and the central compartment length was 10 cm. The 2 electrode compartments were separated from the central one by ion exchange membranes, an AEM AR204SZRA, MKIII, Blank and a CEM CR67, MKIII, Blank, both from Ionics. The electrodes were platinum coated titanium wire ($\varnothing = 3$ mm) by Grønvold & Karnov A/S. A power supply (Hewlett Packard E3612A) was used, maintaining a constant current of 25 mA.

The effluent (500 mL) was placed on the central cell compartment. Both anolyte and catholyte compartments were set with 500 mL of 0.01 M NaNO_3 adjusted to pH 2 with HNO_3 , and recirculated by using “Pan World” magnetic pumps (Plastomec Magnet Pump model P05).

Control experiments for each treatment, without applied current, were also carried out. pH and conductivity in the electrolytes and in the central compartment were measured twice (T_0 and $T_{6,12,24\text{h}}$) per treatment.

At the end of the ED-T experiments, the effluent in the central compartment was analyzed by ICP-OES and IC (as in section 2.2). Membranes were soaked in 1M HNO_3 and the electrodes in 5M HNO_3 , overnight, being their aqueous phases collected and analyzed for Ca, K, Mg, Ni, P, S, Si and Zn by ICP-OES, as well as the electrolytes.

2.4 Mortars bars: Physical and functional tests

After ED-T, the treated effluent was used to prepare mortars according to EN 196-7 [19]. The mixture consisted of 225 mL water (tap water or effluent), 450 g of OPC and 1350 g of CEN standard sand, with 0.5 water/binder ratio of and a sand/cement ratio of 3%. Mortar samples were performed using tap water, ED-T pre-treated effluent and

mixtures of 50% each. The tested mortar samples consisted of horizontal prismatic specimens (160 x 40 x 40 mm) made in triplicates. The mortar bars were defined as:

- Reference (100% tap water)
- Raw (effluent without pre-treatment; 50% or 100%)
- ED-T (effluent with ED-T pre-treatment during 24h, 12h, 6h; 50% or 100%)

Mortars demolding was carried out after 24h and the samples were cured horizontally in a tap water bath for 7, 14 or 28 days at 20 - 21 °C.

The following tests were carried out on the mortar specimens in accordance with:

- Setting time (min) - Standard ASTM C 191 [20] performed by a Vicat Needle. The period of penetration tests was performed by allowing a 1 mm Vicat needle to settle into this paste. The Vicat initial setting time was the time elapsed between the initial contact of raw materials and the time when penetration was measured to be 25 mm or less. The Vicat final setting time was the end point to be the first penetration measure that does not mark the specimen surface with a complete circular impression.
- Workability - Standard EN-1015-3 [21] using a flow table test, where vertical difference between the top of the mold and the displaced original center of the top surface of the specimen was measured as the slump value.
- Flexural strength at 28-day - Standard EN 196-1 [22] with third point loading (two steel supporting rollers of $\varnothing 10.0 \pm 0.5$ mm and a third steel loading roller of the same diameter placed centrally between the other two) at a loading rate of 0.2 kN/s, performed by Instron 6022 10kN;
- Compressive strength at 7, 14 and 28 days - Standard EN 196-1C [22] by a Toni machine using a loading rate of 2.5 kN/s (t-test was performed to find statistical differences, 95% of confidence level)
- Morphology at 28 days with 100% of replacement by raw and ED-T effluent - performed by scanning electron microscope (SEM). No sample pre-treatment was performed. Acceleration voltage of the SEM was 20-25 kV with large field detector, in low vacuum with a magnification of 800x.

3 Results and Discussion

3.1 Characterization of tap water and effluent before and after electrolytic treatment

3.1.1 Initial characterization

Table 2 presents the characteristics of water samples under study: Tap water and effluent. The data shows that almost all raw effluent components were higher than those found in tap water. The exception was for Ca, Mg, Si, Zn average concentrations, nevertheless within the BS EN 1008 (2002) requirements [23].

Chemical impurities in water are active players in the reactions between water and cement and, being prone to interfere with the setting and hardening of cement and consequently with the development of strength and durability of concrete [24].

Comparing the chloride content in the raw effluent and in tap water (207.3 vs 63.0 mg/L, Table 2), and according to BS EN 1008 (2002) [23] requirements (to pre-stressed concrete or grout propose, chloride shall not exceed the level of 500 mg/L; Cl_2 residual ≤ 1 mg/L), both are under the standard, but not under NP 423 ≤ 50 mg/L [25]. The lower the chloride concentration the better, as a high Cl^- content will increase the potential for corrosion in reinforced concrete. Also, sulfate (65.7 vs 31.7 mg/L; BS EN 1008 ≤ 2000 mg/L; NP 413 ≤ 30 mg/L [26]) together with macro elements, can lead to the formation of complexes, increasing the cracking formation.

The value of COD, was higher in the raw effluent (comparing with the other experiments), and presented a large variability between samples (69% RSD). This parameter plays an important role in water and cement reaction, promoting a negative effect in the control of the cement paste of mortars (delays in setting time) [27]. Moreover, when handling effluents, their pathogen content is also relevant due to safety reasons and material acceptance for further uses.

Thereby, the ED-T proposed as a pre-treatment with the potential for the removal of unwanted parameters was studied.

3.1.2 ED-T experiments

The conductivity decreased with time, from 1.3 mS/cm to 0.1 $\mu\text{S}/\text{cm}$ in about 12h (Table 2), which is a consequence of the desalination of the effluent, leading to increase system resistivity (expressed by the cell voltage). In the ED-T experiments the final voltage achieved 35.9, 74.2 and 74.1 V, for respectively 6, 12 and 24h. In the case of the experiments with 12 and 24h, the voltage increased up to the limit of the power

supply and, thus, the current decreased below the established 25 mA, down to almost open-circuit conditions.

The final pH of the ED-T effluent was 4-5 (Table 2), inside the range of the standard BS EN 1008 (2002) \geq 4-8 [23].

3.1.3 Removal rates

The ED-T treatment reduced the concentration of all ions, elements and parameters in the effluent in all experiments (Table 2).

The data shows that the transport in the bench reactor is fast achieving, in a few hours, the necessary removal of ions to prevent the side effects on the mortar properties (Table 2). Anions removal at the end of the ED-T experiments ranged between 85-99.7%. The results showed (Table 2) that an optimal removal of the ions was achieved in approx. 6h, indicating that there is no need of expending more energy as in ED-T 12h or 24h. The anions removal is an important variable due to its weak interaction with cement, for example, on the mortars final compressive strength.

The removal of the elements quantified by ICP analysis was in the range of 50-97.8%.

The parameters TSS and COD, that interfere with the cement paste final properties, were also reduced after ED-T (COD removal rates: ED-T 24h 44%; ED-T 12h 43%; ED-T 6h 45%) complying with the requirements of standard NP 1414 (COD \leq 500 mg/L) [27]. The effluent values of BOD₅ before and after ED-T experiments were in accordance with USEPA limit of BOD₅ \leq 30 mg/L [28]. Furthermore, Guedes et al. 2016 [29] reported the inactivation of biological activity in similar environmental matrices using ED-T, thus is expected the same phenomena in the present work.

The qualitative and quantitative analysis of ED-T treated effluent shows that its overall composition after 6h of treatment matches the standards and can supply a product with less risk components, such as corrosion, than the used conventional tap water.

Assuming a linear behavior of the voltage in time, for the ED-T 6h experiment at 25 mA, the treatment required approximately 4.05 kWh/m³ of pre-treated effluent. With an average energy price in Denmark of 0.30 € per kWh (in Europe is ~ 0.2 € per kWh), the estimated cost related to the energy consumption is 1.215 €/m³ of pre-treated effluent. The electricity energy cost can be significantly reduced by an optimization study on the duration of the ED-T pre-treatment based on the material's mechanical properties.

The results suggest that the operational time, and therefore the energy costs, could be reduced by using less concentrated electrolytes in the electrode compartments. The low pH found in the effluent at the end of the experiments suggests that protons are

entering the central compartment via permeation through the non-ideal AEM or by diffusion from the cathode compartment. Optimizing the electrolyte concentrations for the process will increase the selectivity and improve the energy efficiency.

Nevertheless, for optimization purposes, a variability in the effluent characteristics is expected to occur as they are influenced by daily factors such as weather conditions, influent composition or operational conditions at the biological reactor, and must be taken into account.

3.2 Mortars bars: Physical and functional quality parameters

3.2.1 Effects on cement paste - Setting time and workability

Setting time and workability are important properties of materials. Setting time is defined as a specified time required for concrete or mortar to change from liquid state (initial setting time) to a solid state (final setting time), where the surface becomes sufficiently rigid to withstand a definite amount of pressure. Workability can be divided into three different categories (low, medium and high), describing the ability to mix, place, consolidate and finish with minimal loss of homogeneity a mortar preparation. Workability is a property that directly impacts strength, quality, appearance, and the cost of labor operations. Setting time and workability are influenced by pH, dissolved organic matter, COD and salts. The aim of the tests was to understand if the moderately low pH after the ED-T would affect those two parameters.

Table 3 presents the initial and final setting time, workability category and slump values tests according to pH of the aqueous matrices used. Standards BS EN 1008 (2002) [23] and ASTM C94 [30] recommend that the water used for concrete production should have a pH \geq 4-8, since a lower pH can promote the dissolution of cement in the mortar mix. The final pH obtained in all effluents was in accordance to the values referred in the two standards (pH between 8-4).

Setting time can be divided into initial setting time and final setting time based on the degree of rigidity. Comparing the different mixtures, with reference tap water, there was a delay on the initial setting time of 20 min, when the mix was performed with 100% or 50% raw effluent and 10 min when performed with 50% ED-T treated effluent (Table 3). Using 100% ED-T treated effluent no measurable difference was observed comparing to the reference mortar. The final setting time increased when raw and ED-T effluent is incorporated, having a reference time difference of 50-130 min.

Delay in setting times is related to pH, temperature, humidity, sugars and dissolved organic matter (constituent of COD) presented on raw effluent. Sugars adsorb on the

surface of the cement grains and thus might delay the cement hydration. The obtained results are in line with other studies [31,32]. Dissolved salts also influence the setting time process [31]. The ammonia in the raw effluent has been reported to cause bleeding action (flow of water to the top of the concrete due to osmotic pressure) [33]. In the current study, applying ED-T as a pre-treatment decreased approximately 60% the NH_4^+ concentration. This will eliminate the possible bleeding action and turn the paste similar with the reference's setting time. According to BS EN 1008 the initial time shall not be less than 1h from the reference mortar (experiments achieved a range of difference 0-20min) and the final setting time should not exceed 12h (experiments between 4.8 and 7h).

Workability tests were conducted as described in section 2.4, where vertical difference between the top of the mold and the displaced original center on the specimen top surface was measured as the slump value. The use of effluent, either with or without ED-T treatment, increased the slump: 16-33 mm differences (Table 3). The cement paste produced with reference water was considered, according to EN 1015-3 [21], as a high workability mortar. The experiments using mixed effluents resulted in a high workability category cement paste as well [21]. High workability paste can be used in inaccessible locations, large flat areas, underwater applications and pumping concrete over long distances (BS EN 206) [34]. This similarity in the paste prepared with effluent, can present a positive enhancement in the mortars production, since it can be an alternative to the use of superplasticizer to stabilize and manipulate the paste [10,35,36].

3.2.2 Effects on mortar properties – Flexural strength, compressive strength and morphology

Flexural strength of concrete is a key mechanical property, which represents the ability of a beam or slab to resist failure in bending. The load, extension and flexural strength (also known as modulus of rupture, or bend strength, or transverse rupture strength) results were determined for three 40 x 40 x 160 mm prisms specimens. These specimens were casted and tested after 28 days of curing, as showed in Figure 2.

Comparing with the reference, flexural strength results showed ED-T 12h experiment effluent replacement as the highest strength (data in line with the similar morphology as Figure 4a and 4d). Consequently, the same experiment had the highest load, where the most similar to the reference were ED-T 6h and 24h with 50% of tap water replacement. Comparing the 100% of replacement (Figure 2b), the highest flexural stress was obtained after ED-T 6h treated effluent, followed by ED-T 12h. Regarding the extension - deflection of the material - for all the mortars made with untreated or

treated effluent, after the application of the load, the moment of failure occurred earlier comparing to the reference. In this way, the largest extension for the reference mortar indicates that the strengthening was more required in this experiment than in the ED-T or raw effluent experiments.

Figure 3 presents the compressive strength of mortars produced with tap water, 50% and 100% of raw effluent and treated effluents (ED-T 6, 12 and 24h) with 7, 14 and 28 days of curing age. The increase of material strength is related with mortar age. The higher the strength value, less deformable is the material.

The strength variations addressed between the reference (7, 14 and 28 days) and the other mortar experiments (7, 14 and 28 days) are within BS EN 1008 [23] (Figure 3), which refers that the average compressive strength shall be at least 90% of the average compressive strength of corresponding specimens prepared with tap water.

Comparing 7 days cured specimens, with 100% of raw effluent and 50% of ED-T 12h treated effluent, with the reference mortar, a trend in increased mortar strength was observed, c.a. 5 and 11% of gain; $\sigma = 0.2$ and 1.4 , respectively). Still, this was only statistical relevant when the 50% of effluent treated in ED-T for 12h was used ($p < 0.05$). The same trend for increased strength was also observed for the specimens after 28 days of curing, ED-T 12h effluent either with 50% or 100% replacement and ED-T 6h effluent with 50% of replacement (55.9, 56.8, 58.6 MPa; $\sigma = 1.3, 2.1$ and 0.8 , respectively), although without statistical relevance at 95% confidence level. This highest “early” strength could be a result of changes in effluent chemical properties during ED treatment, that highly influences mortar physico-chemical parameters. This might be due to the decreased COD in the effluent after electrochemical treatment (Table 2), a parameter that is known to decrease material strength [37]. After the ED-T pre-treatment, a COD removal was achieved in the three ED-T experiments (43-45%; Table 2).

3.2.3 Morphology SEM

Scanning electron microscope was used to visualize the dimensional morphology of the prepared mortar samples to compare visual characteristics, such as formation of different crystals, void spaces and crack formation, in relation to the reference (using tap water Figure 4a). Figure 4 showed that the most significant difference was obtained when the tap water was replaced by 100% of raw effluent. It was also possible to observe the new formation of subhedral to anhedral crystals, more pronounced in Figures 4b) and 4e). The bed matrix became denser and with less void spaces compared to the reference. In the case of ED-T 6, 12 and 24h (Figures 4e, d and c), mortar specimens' structures between aggregate are structurally intact. This

observation corroborates the higher strength in compressive tests in ED-T specimens, as less interatomic spacing requires higher stresses for unwanted cracks to occur, being, therefore, a positive outcome [38]. In all mortar specimens' micro cracks effect were not observed.

4 Conclusion

Concerned with water scarcity around the world, the aim of the present study was to explore alternatives for freshwater use in the building industry. Herein, the feasibility of using ED-T as a pre-treatment in mortar preparation was investigated, to remove effluent contaminants such as Ca, Zn, Cl^- , SO_4^{2-} and COD that influence properties of the mortars' material. Based on the experimental conditions observed the following main conclusions are drawn:

1. The electrodiolytic pre-treatment achieved a high removal of critical elements and a decrease in the effluent discharged parameters that affect materials' quality and durability.
2. Workability that affect the mortar were moderate and tolerable. The workability increased in all the mortars compared to the reference. These results can avoid further use of superplasticizer. Compressive tests had an increase on resistance in specimens prepared with ED-T effluent, comparing with the reference and raw effluent. SEM showed changes in the morphology, where the replacement with raw effluent seems to show no space between pours, causing a possible problem. However, even if the void spaces are inferior with ED-T 12h and ED-T 6h, they are still tolerable and had a positive impact in flexural and compressive strength.
3. In some specimens, even if the raw effluent showed the potential to replace water in construction materials, the authors are concerned about the pathogens and other impurities that affect the material final quality, durability and safety. Hence, the ED-T can be a way to make effluents non-hazardous contributing for a large scale use.

Further studies are needed on the long-term durability performance and micro-biological activity of mortars mixed with urban effluent treated with ED-T, as well as on the process optimization.

Acknowledgments

C. Magro acknowledges Fundação para a Ciência e a Tecnologia for her PhD fellowship (SFRH/BD/114674/2016). Paz-Garcia acknowledges the financial support

from the “Plan Propio de Investigación y Transferencia de la Universidad de Málaga”, code: PPIT.UMA.B1.2017/20 and PPIT.UMA.B5.2018/17, and the “Ministerio de Educación, Cultura y Deporte - Subprograma Estatal de movilidad, y Plan Estatal de Investigación Científica y Técnica y de Investigación 2013-2016”, code: CAS17/00196. This work has received funding from the European Union’s Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No. 778045, and from FCT/MEC through grant UID/AMB/04085/2013.

ACCEPTED MANUSCRIPT

References

- [1] United Nations General Assembly, Transforming our world: The 2030 agenda for sustainable development, <https://Sustainabledevelopment.Un.Org/Content/Documents/7891Transforming%20Our%20World.Pdf>. Pdf. (2015) 1–5. doi:10.1007/s13398-014-0173-7.2.
- [2] WWAP (United Nations World Water Assessment Programme)/UN-Water., The United Nations World Water Development Report 2018: Nature-Based Solutions for Water, Paris, 2018.
- [3] M.-S. Low, Material Flow Analysis of Concrete in the United States, (2005), M.Sc. Thesis in Building Technology, Massachusetts Institute of Technology, MA, US, 189 pp..
- [4] S.A. Miller, A. Horvath, P.J.M. Monteiro, Impacts of booming concrete production on water resources worldwide, *Nat. Sustain.* 1 (2018) 69–76. doi:10.1038/s41893-017-0009-5.
- [5] P.E. Josephson, Y. Hammarlund, The causes and costs of defects in construction: A study of seven building projects, *Autom. Constr.* 8 (1999) 681–687. doi:10.1016/S0926-5805(98)00114-9.
- [6] C. Barrera-Díaz, G. Martínez-Barrera, O. Gencel, L.A. Bernal-Martínez, W. Brostow, Processed wastewater sludge for improvement of mechanical properties of concretes, *J. Hazard. Mater.* 192 (2011) 108–115. doi:10.1016/j.jhazmat.2011.04.103.
- [7] Z.Z. Ismail, E.A. Al-Hashmi, Assessing the recycling potential of industrial wastewater to replace fresh water in concrete mixes: Application of polyvinyl acetate resin wastewater, *J. Clean. Prod.* 19 (2011) 197–203. doi:10.1016/j.jclepro.2010.09.011.
- [8] K.S. Al-Jabri, A.H. Al-Saidy, R. Taha, A.J. Al-Kemyani, Effect of using wastewater on the properties of high strength concrete, *Procedia Eng.* 14 (2011) 370–376. doi:10.1016/j.proeng.2011.07.046.
- [9] S.B. M.Kanitha, P.Ramya, V.Revathi, Potential Utilisation of Untreated/Treated Textile Effluent in Concrete, *Int. J. Res. Eng. Technol.* 3 (2014) 518–522. doi:10.15623/ijret.2014.0319093.
- [10] G. Asadollahfardi, M. Delnavaz, V. Rashnoiee, N. Ghonabadi, Use of treated domestic wastewater before chlorination to produce and cure concrete, *Constr. Build. Mater.* 105 (2016) 253–261. doi:10.1016/j.conbuildmat.2015.12.039.
- [11] A.H. Noruzman, B. Muhammad, M. Ismail, Z. Abdul-Majid, Characteristics of treated effluents and their potential applications for producing concrete, *J. Environ. Manage.* 110 (2012) 27–32. doi:10.1016/j.jenvman.2012.05.019.
- [12] M. Silva, T.R. Naik, Sustainable Use of Resources – Recycling of Sewage Treatment Plant Water in Concrete, in: J. Zachar, P. Claisse, T.R. Naik, E. Ganjian (Eds.), *Second Int. Conf. Sustain. Constr. Mater. Technol.*, 2010. INC 1-10
- [13] Y.B. Acar, A.N. Alshwabkeh, Principles of electrokinetic remediation, *Environ. Sci. Technol.* 27 (1993) 2638–2647. doi:10.1021/es00049a002.
- [14] G.M. Kirkelund, C. Magro, P. Guedes, P.E. Jensen, A.B. Ribeiro, L.M. Ottosen, Electrodialytic removal of heavy metals and chloride from municipal solid waste

- incineration fly ash and air pollution control residue in suspension - Test of a new two compartment experimental cell, *Electrochim. Acta.* 181 (2015) 73–81. doi:10.1016/j.electacta.2015.03.192.
- [15] B. Ebbens, L.M. Ottosen, P.E. Jensen, Electrodialytic treatment of municipal wastewater and sludge for the removal of heavy metals and recovery of phosphorus, *Electrochim. Acta.* 181 (2015) 90–99. doi:10.1016/j.electacta.2015.04.097.
- [16] P. Guedes, C. Magro, N. Couto, A. Mosca, E.P. Mateus, A.B. Ribeiro, Potential of the electrodialytic process for emerging organic contaminants remediation and phosphorus separation from sewage sludge, *Electrochim. Acta.* 181 (2015) 109–117. doi:10.1016/j.electacta.2015.03.167.
- [17] Lyngby-Taarbæk Kommune - Økonomiudvalget, (2014) 1–90. <https://lyngbytaarbaek.plan.ramboll.dk/view.html?planid=08541641-0f8e-45b1-87fd-820222c7840e>.
- [18] A.B. Ribeiro, J.T. Mexia, A dynamic model for the electrokinetic removal of copper from a polluted soil, *J. Hazard. Mater.* 56 (1997) 257–271. doi:10.1016/S0304-3894(97)00060-5.
- [19] BS EN 196-7:2007 - Methods of testing cement. Methods of taking and preparing samples of cement, 2008, British Standards Institution.
- [20] ASTM C191-18, Standard Test Methods for Time of Setting of Hydraulic Cement by Vicat Needle, West Conshohocken, PA, 2018. doi:10.1520/C0191-18.
- [21] BS EN 1015-3:1999 - Methods of test for mortar for masonry Part 3: Determination of consistence of fresh mortar (by flow table), 1999, British Standards Institution.
- [22] BS EN-196-3:2016 - Methods of testing cement. Determination of setting times and soundness, 2016, British Standards Institution.
- [23] BS EN 1008:2002 - Mixing water for concrete. Specification for sampling, testing and assessing the suitability of water, including water recovered from processes in the concrete industry, as mixing water for concrete, 2002, British Standards Institution.
- [24] P. Tiwari, R. Chandak, R.K. Yadav, Effect of salt water on compressive strength of concrete, *Int. J. Eng. Res. Appl.*, 4 (2014) 38–42. ISSN 2248-9622, https://ijera.com/papers/Vol4_issue4/Version%205/H044053842.pdf.
- [25] NP 423:1966, Água. Determinação do teor de cloretos, 1966, Lisboa, IGPAI (in Portuguese).
- [26] NP 413:1966, Água. Determinação do teor de sulfatos, 1966, Lisboa, IGPAI (in Portuguese).
- [27] NP 1414:1977, Determinação do consumo químico de oxigénio de águas de amassadura e de águas em contacto com betões. Processo do dicromato de potássio, 1977, Lisboa, IGPAI (in Portuguese).
- [28] Environmental Protection Agency (EPA), Guidelines for water reuse technical issues in planning water reuse systems, Washington (DC), 2004.
- [29] P. Guedes, E.P. Mateus, J. Almeida, A.R. Ferreira, N. Couto, A.B. Ribeiro, Electrodialytic treatment of sewage sludge: Current intensity influence on phosphorus recovery and organic contaminants removal, *Chem. Eng. J.* 306

- (2016) 1058–1066. doi:10.1016/j.cej.2016.08.040.
- [30] ASTM C94/C94M-17a - Standard Specification for Ready-Mixed Concrete, West Conshohocken, PA, 2017. doi:10.1520/C0094_C0094M-17A.
- [31] O.S. Lee, M.R. Salim, M. Ismail, MD.I. Ali, Reusing treated effluent in concrete technology, *J. Teknol.* 34 (2001) 1–10. doi:10.11113/jt.v34.648.
- [32] I. Al-Ghusain, M.J. Terro, Use of treated wastewater for concrete mixing in Kuwait, *Kuwait J. Sci. Eng.* 30 (2003) 213–228.
- [33] A.M. Neville, *Properties of Concrete*, Pearson Education Limited, England, 2011. doi:10.4135/9781412975704.n88.
- [34] BS EN 206-1:2000 Concrete - Part 1: Specification, performance, production and conformity, 2001, British Standards Institution.
- [35] P.C. Aïtcin, Cements of yesterday and today - concrete of tomorrow, *Cem. Concr. Res.* 30 (2000) 1349–1359. doi:10.1016/S0008-8846(00)00365-3.
- [36] K.S. Al-Jabri, A.H. Al-Saidy, R. Taha, A.J. Al-Kemyani, Effect of using wastewater on the properties of high strength concrete, *Procedia Eng.* 14 (2011) 370–376. doi:10.1016/j.proeng.2011.07.046.
- [37] K. Sarkor, T.M. Miretu, B. Bhattacharjee, Curing of concrete with wastewater and curing compounds : Effect on strength and water absorption, (2014). *Indian Concr. J.* 88 (2014) 87-93.
- [38] B. Muhammad, M. Ismail, M.A.R. Bhutta, Z. Abdul-Majid, Influence of non-hydrocarbon substances on the compressive strength of natural rubber latex-modified concrete, *Constr. Build. Mater.* 27 (2012) 241–246. doi:10.1016/j.conbuildmat.2011.07.054.

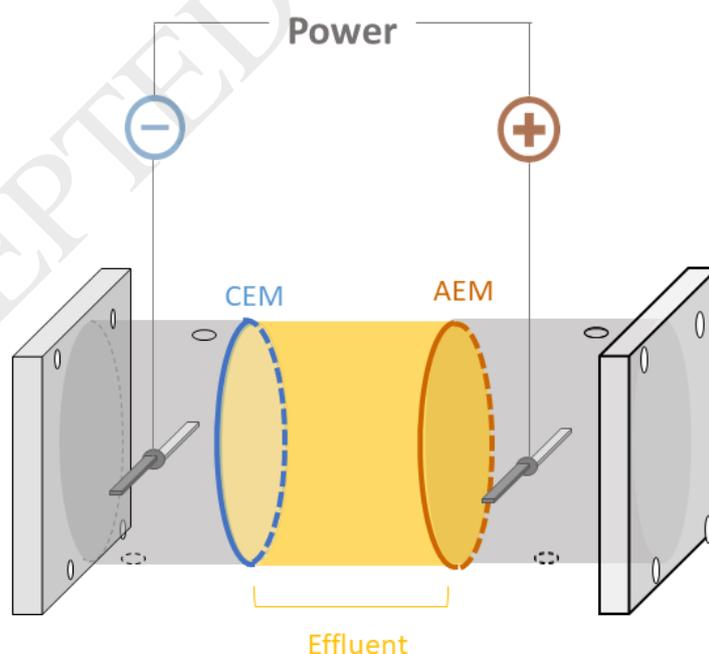
Figure captions

Figure 1 Electrodialytic 3 compartments cell with $\varnothing = 8$ cm, central compartment L = 10 cm and electrolyte compartment L = 5 cm. CEM – cation exchange membrane; AEM – anion exchange membrane

Figure 2 Load, vertical displacement and flexural stress values of mortar at 28 days of curing age for tap water (reference), (a) raw effluent and effluent after ED-T treatment with 50% of replacement and (b) raw effluent and effluent after ED-T treatment with 100% of replacement

Figure 3 Compressive strength of age 7, 14 and 28 days using: tap water (reference), raw effluent (50 and 100% replacement) and effluent with ED-T treatment (6, 12 and 24h - 50% and 100% replacement)

Figure 4 Images of SEM: (a) reference mortar, (b) 100% of replacement with raw effluent, (c) 100% of replacement with ED-T 24h effluent, (d) 100% of replacement with ED-T 12h effluent, (e) 100% of replacement with ED-T 6h effluent; magnification: 800x



(a)

Figure 1 Electrolytic 3 compartments cell with $\varnothing = 8$ cm, central compartment L = 10 cm and electrolyte compartment L = 5 cm. CEM – cation exchange membrane; AEM – anion exchange membrane

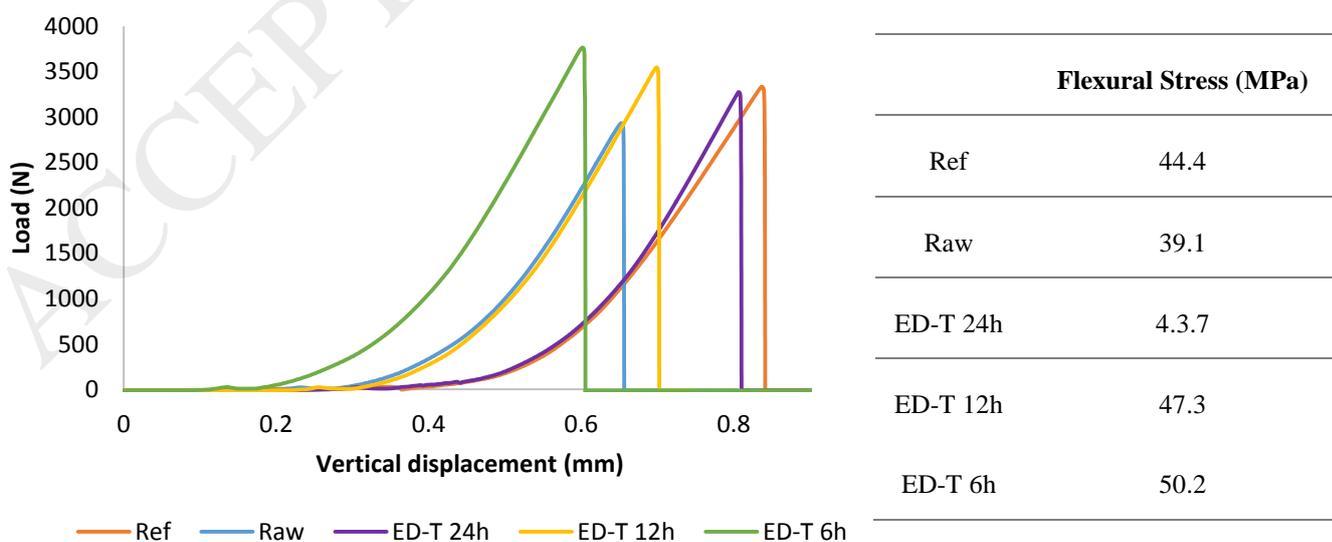
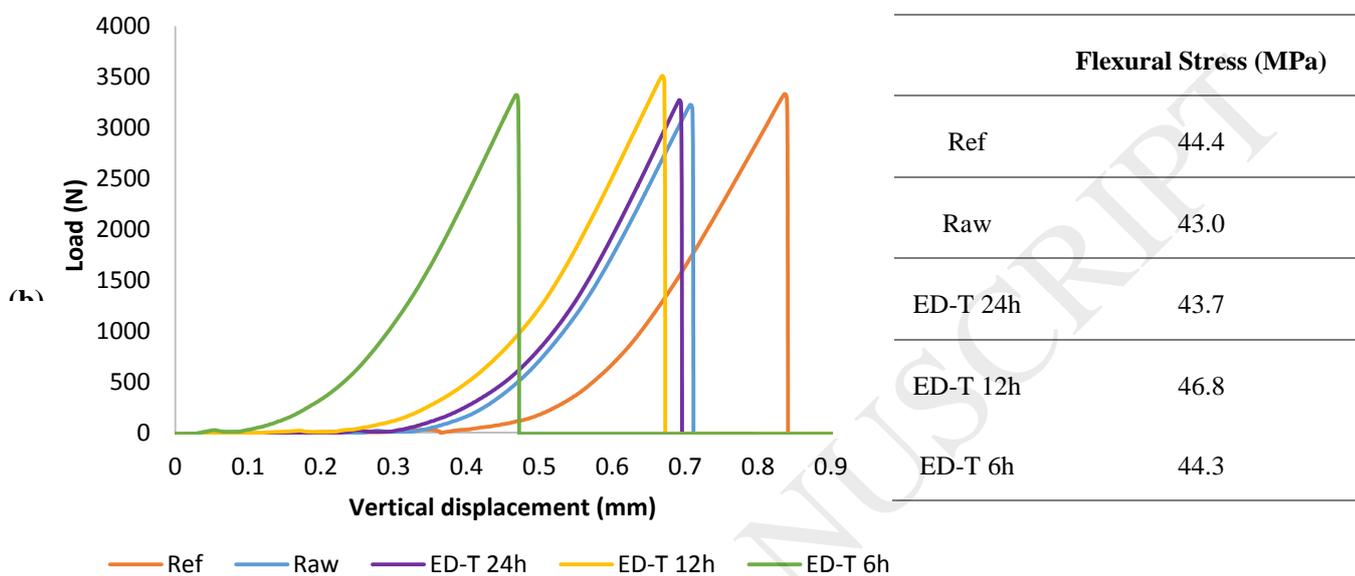


Figure 2 Load, vertical displacement and flexural stress values of mortar at 28 days of curing age for tap water (reference), (a) raw effluent and effluent after ED-T treatment with 50% of replacement and (b) raw effluent and effluent after ED-T treatment with 100% of replacement.

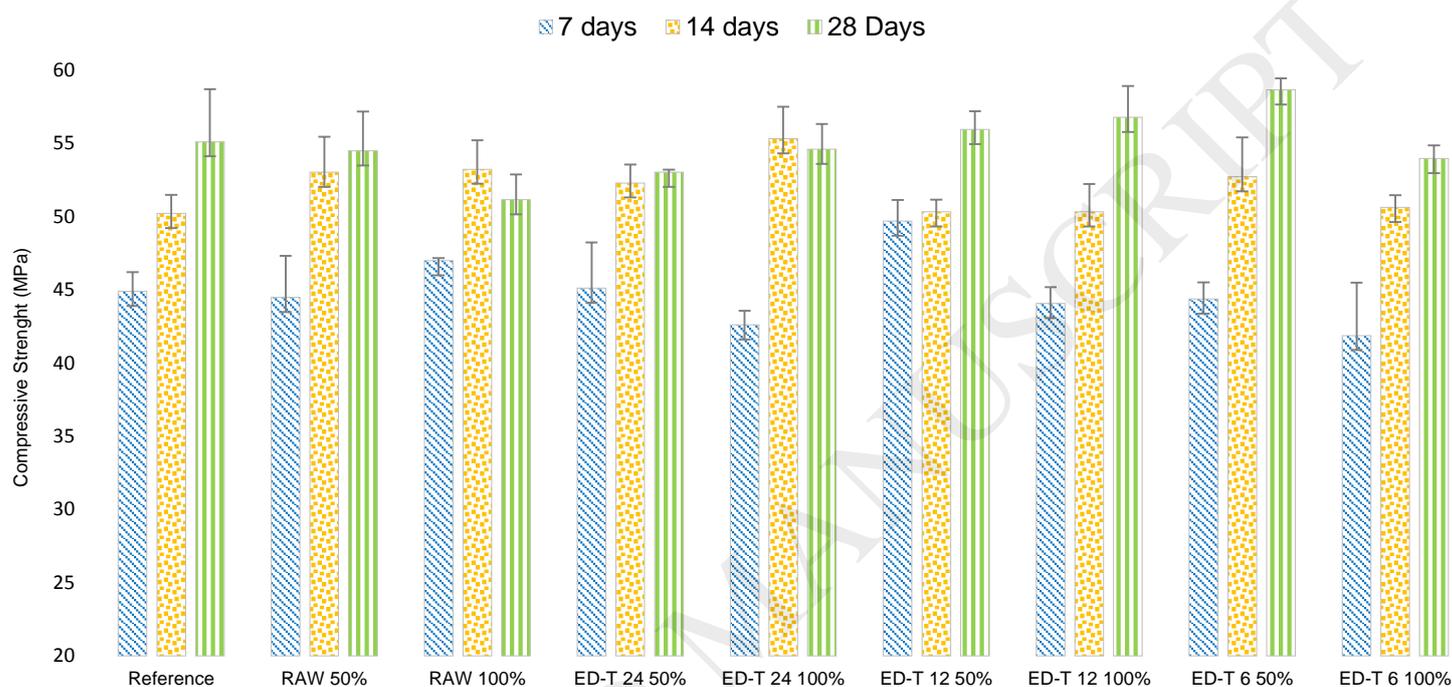
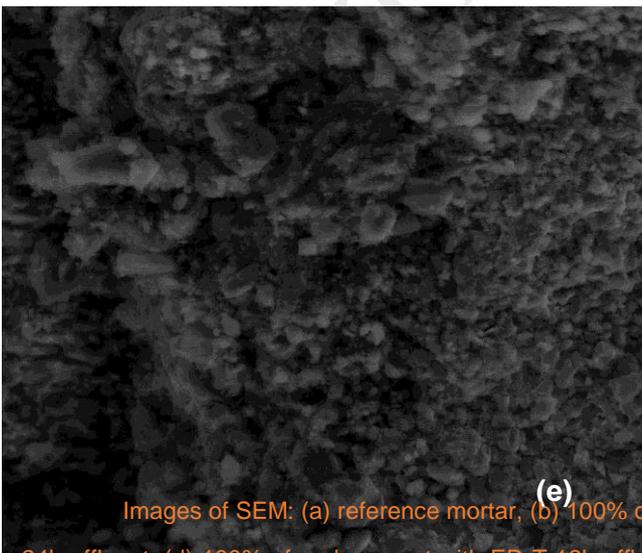
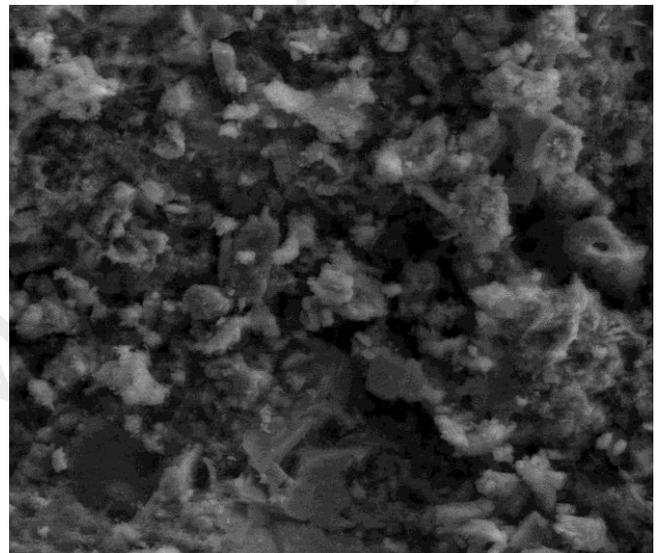
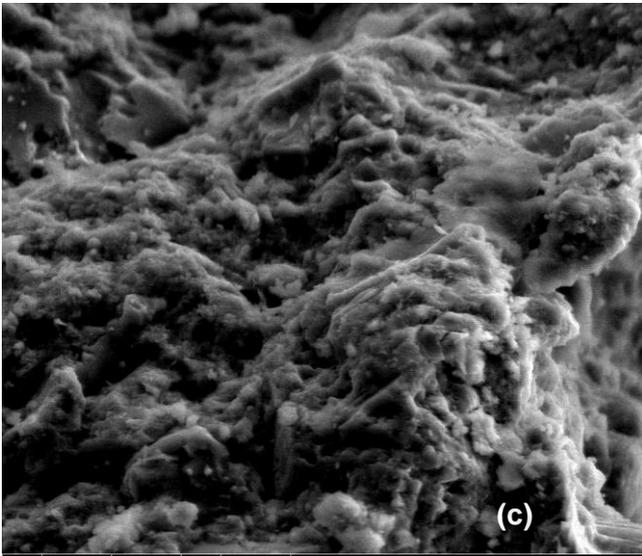
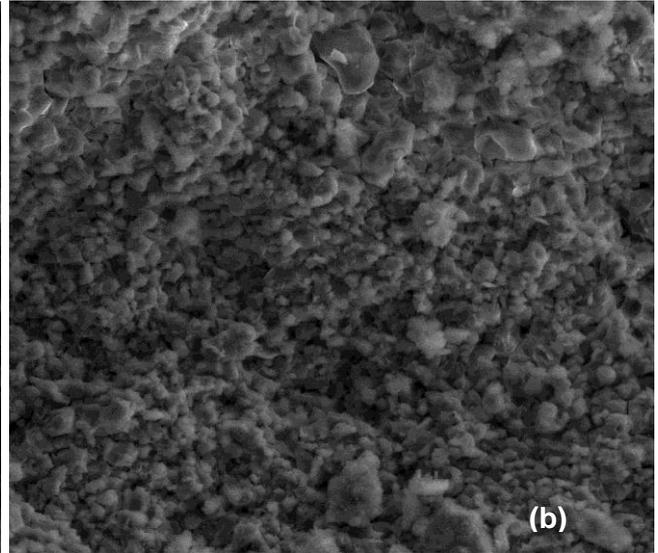
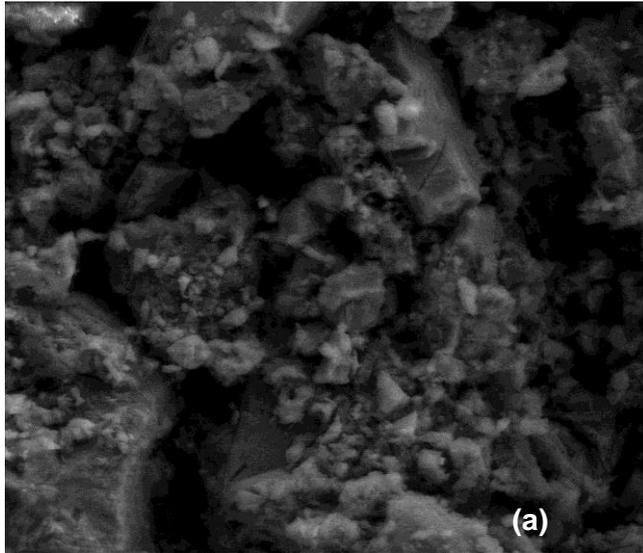


Figure 3 Compressive strength of age 7, 14 and 28 days using: tap water (reference), raw effluent (50 and 100% replacement) and effluent with ED-T treatment (6, 12 and 24h - 50% and 100% replacement). Each column represents an average (n=3), and standard deviations are provided.



Images of SEM: (a) reference mortar, (b) 100% of replacement with raw effluent, (c) 100% of replacement with ED-T 24h effluent, (d) 100% of replacement with ED-T 12h effluent, (e) 100% of replacement with ED-T 6h effluent; magnification: 800x

Table Legends

Table 1 Electrodialytic experimental conditions

Experiment	Code	Effluent treated (mL)	Duration (h)	Current intensity (mA)
1	ED-T 6h		6	
2	ED-T 12h	500	12	25
3	ED-T 24h		24	

ACCEPTED MANUSCRIPT

Table 2 Characterization of the tap water and effluent before and after 24, 12 and 6h of ED-T

	Tap Water	Effluent			
		Raw	ED-T 24h	ED-T 12h	ED-T 6h
pH	7.7 ± 0.1	7.9 ± 0.3	4.0 ± 0.3	4.0 ± 0.5	5.0 ± 0.3
Conductivity (µs/cm)	755.5 ± 23.3	1304.5 ± 109.6	0.1 ± 0.02	0.1 ± 0.01	0.2 ± 0.01
Parameters (mg/L)					
TSS	–	< 4	<i>n.d.</i>	<i>n.d.</i>	<i>n.d.</i>
BOD ₅	–	3.55 ± 0.05	3.1 ± 0.0	5.1 ± 0.4	4.1 ± 0.4
COD	–	403.5 ± 279.3	226	230 ± 4.2	221 ± 8.5
N _{total}	–	7	2	6	1
NO ₂	–	0.11 ± 0.00	0.02 ± 0.00	0.02 ± 0.00	0.02 ± 0.00
NH ₄ ⁺	–	0.16 ± 0.01	0.08 ± 0.01	0.07 ± 0.01	0.09 ± 0.01
P _{total}	–	0.63 ± 0.42	<i>n.d.</i>	<i>n.d.</i>	<i>n.d.</i>
Elements (mg/L)					
Ca	105.3 ± 12.1	99.7 ± 13.3	0.3 ± 0.0	0.3 ± 0.1	5.9 ± 3.6
K	4.9 ± 0.7	24.2 ± 0.7	0.5 ± 0.2	0.3 ± 0.2	1.2 ± 0.4
Mg	20.3 ± 0.7	15.7 ± 0.9	0.05 ± 0.01	0.04 ± 0.01	1.2 ± 0.7
Na	35.6 ± 4.2	118.7 ± 0.7	0.5 ± 0.1	1.5 ± 1.8	10.2 ± 8.3
S	11.1 ± 3.3	20.6 ± 1.9	0.2 ± 0.1	0.2 ± 0.1	3.0 ± 1.1
Ni	0.005 ± 0.000	0.06 ± 0.02	0.004 ± 0.005	0.003 ± 0.004	0.004 ± 0.004
Si	9.3 ± 0.7	8.4 ± 0.4	5.8 ± 0.6	6.6 ± 0.2	7.5 ± 0.6
Zn	0.8 ± 0.6	0.05 ± 0.02	± 0.01	0.01 ± 0.01	0.02 ± 0.02
Anions (mg/L)					
Cl ⁻	63.0 ± 2.2	207.3 ± 3.9	2.0 ± 0.2	2.1 ± 0.8	23.1 ± 10.5
NO ₃ ⁻	2.0 ± 2.8	13.5 ± 3.2	8.3 ± 3.8	9.1 ± 15.7	11.9 ± 19.4
SO ₄ ²⁻	31.7 ± 2.2	65.7 ± 2.3	0.4 ± 0.1	0.2 ± 0.3	9.9 ± 3.9

*n.d. – not detected

Table 3 Setting times and slump values using: tap water (reference), raw effluent (50 and 100% replacement) and effluent with ED-T treatment (50% and 100% replacement)

	Mix	Initial Setting time (min)	Final Setting time (min)	Workability	Slump (mm)
pH 7.7	Reference (tap water)	140	290	High 100 – 150 mm	101 ± 2
pH 7 - 8	Raw 50%	160	340		117 ± 2
	Raw 100%	160	420		129 ± 3
pH 4 - 5	ED-T 50%	150	400		131 ± 2
	ED-T 100%	140	420		134 ± 9