



# Electrochemical regeneration of spent activated carbon from drinking water treatment plant at different scale reactors



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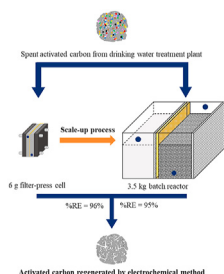
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## HIGHLIGHTS

- Electrochemical regeneration of spent ACs was studied at different reactor scales.
- A recovery of the surface area of 96% was achieved after 8 h of treatment.
- In the 3.5 kg electrochemical cell, a 95% of the AC surface area was recovered.
- Electrochemistry can provide a green and *in-situ* alternative to regenerate spent AC.

## GRAPHICAL ABSTRACT



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## ABSTRACT

The electrochemical regeneration of real spent activated carbons (AC) used in drinking water treatment plants was studied at different reactor scales. The electrochemical regeneration was carried out in a 6 g filter-press cell and a 3.5 kg batch reactor, allowing the scaling-up of the process between the two electrolytic reactors. The effect of the electrolyte, the divided/undivided compartment configuration and the current density were studied in the filter-press cell. The effect of the regeneration time and the influence of the regeneration time were studied in the scaled-up reactor. A current density of  $0.025 \text{ A cm}^{-2}$  was used and the electrodes were Pt/Ti as anode and Pt/Ti and stainless-steel as cathode. The ACs were characterized by  $\text{N}_2$  adsorption isotherms to analyse the recovery of porosity and TPD-MS to analyse the AC surface after the electrochemical treatment. In filter-press cell, a recovery of the surface area of 96% was achieved after 8 h of treatment, by introducing the AC in the cathodic compartment using  $0.05 \text{ M H}_2\text{SO}_4$  solution as electrolyte. In the 3.5 kg electrochemical reactor, 95% of the pristine AC surface area was recovered. Thus, electrochemical methods can provide a green alternative to the regeneration of spent AC.

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## 1. Introduction

Adsorption onto activated carbons (AC) is an established advanced technology to remove organic and inorganic pollutants in water treatment plants. This treatment is one of the most versatile, having the advantage of no by-products generation in drinking water treatment plants (DWTP) (Jaria et al., 2019). The global production of AC is annually increasing around 10% due to the

growing demand of waste water reutilization, the development of advanced drinking water treatment processes and the approval of a more restrictive environmental protection legislation (Jones et al., 2018), reaching a global consumption of 4.28 million tons in 2012 (Zanella et al., 2014). Nowadays, the industrial sector requires large amounts of AC to remove contaminants from gas and water.

AC plays a key role to reduce the organic matter in secondary and/or tertiary treatment of drinking water and wastewater. This is due to its capacity to adsorb those organic compounds which have not been eliminated by the natural biological degradation, as well as micropollutants and heavy metals at trace quantities (Wong et al., 2018). Also, during the tertiary treatment the AC adsorbs organic compounds, such as: pesticides, dyes, chlorinated solvents, phenols and hydroxylated derivatives, aromatic derivatives and other substances that introduce flavours and odours (Crini et al., 2019).

Nevertheless, there are several problems related to this technology that still need to be addressed, the main one being the manipulation and management of spent AC, that must be considered as highly contaminant waste, and can vary at regional-national level. Accordingly, the viability of AC use at industrial level relies on the regeneration and reactivation of spent AC. When the lifetime of the AC is reached, which occurs when the adsorption capacity of AC is partially depleted, the DWTP must replace it in order to maintain the water quality. Then, the spent AC must be either regenerated, what allows AC reutilization after the removal of adsorbed pollutants, or discarded as a hazardous contaminant, whose legislation is increasingly restrictive (McQuillan et al., 2018). Proper management of AC at this last stage is necessary to ensure the sustainability of the whole adsorption process.

Salvador et al. (2015a, 2015b) describe up to four different technologies for the regeneration of AC: thermal, chemical, microbiological and vacuum regeneration. Widespread use of the thermal method makes it the most developed treatment for the regeneration of AC. However, it has some drawbacks, such as the high energy consumption, low recovery yield due to undesired carbon gasification and high transportation costs. Through the process of thermal regeneration between 10 and 20% of AC is lost (Muñoz et al., 2007) and, this method is only economically feasible when AC regeneration is over 620 kg day<sup>-1</sup> (Jeswani et al., 2015).

Beyond this classification, many novel AC regeneration methods are being developed with a high regeneration efficiency such as electro-Fenton method (Bañuelos et al., 2013) and wet oxidation method (Ledesma et al., 2015). However, these methods are at the initial stage of development on a laboratory-scale and present numerous disadvantages for their application on a larger scale. For example, in the electro-Fenton method the potential distribution in the cathode packed bed or the stability and practical lifetime of the resin are relevant disadvantages. Other alternative technologies, such as electrochemical and supercritical regeneration (Salvador et al., 2015b), which have already been developed at lab-scale, show their potentialities to be used at larger scales, and microwave regeneration whose advantage to regenerate the spent adsorbents is that it is over two orders of magnitude faster than the conventional heating process but its drawback is that the technical and economic benefits of microwave regeneration have to be better defined compared to conventional heating (Durán-jiménez et al., 2019).

In electrochemical regeneration, the spent AC is placed between two electrodes and, when applying an electric field, desorption and, in the adequate conditions, degradation processes of the adsorbed contaminants are produced (McQuillan et al., 2018). This method has several advantages compared to other regeneration processes of AC: i) higher efficiency in terms of energy consumption due to the use of electron as a reagent, ii) it can be applied by a direct current

source, iii) it can be applied *in-situ*, iv) minimal adsorbent losses, v) less CO<sub>2</sub> emission and vi) its suitability for small and medium-sized water treatment equipment (Berenguer et al., 2010a; Sharif et al., 2017).

Advances in the recovery of the adsorption capacity of AC have focused on gram-scale studies carried out in two types of electrochemical reactors, a batch reactor (Ania and Béguin, 2008; Delpeux-Ouldriane et al., 2015; Zanella et al., 2017) and different designs in a column reactor (Weng and Hsu, 2008; Narbaitz and Karimi-Jashni, 2009; Narbaitz and McEwen, 2012). In these studies, AC regeneration efficiency greater than 75% has been obtained and 100% can be reached depending on the experimental conditions. However, the lab-scale used and the amount of regenerated AC in these studies are not enough to validate the process on an industrial-scale because only few studies deal with the feasibility of the process at larger scales. In this sense, Wang and Balasubramanian (2009) were able to obtain a regeneration efficiency close to 100% on a 6.3 kg reactor using spent AC with organic compounds. However, the development of up-scale process is very important from scientific and technological point of view.

The main objective of this work is to study the electrochemical regeneration process at kilogram-scale and using spent AC provided by a municipal DWTP. The spent AC has been recovered from a filter located in the tertiary treatment system of the plant after a servicing time of three years, meaning that its adsorption capacity has been partially depleted while working in real conditions. It is important to highlight that, from the best of our knowledge, this is the first time that real spent AC used in DWTP has been electrochemically regenerated in kg-scale. In a first step, spent AC is subjected to electrochemical treatment using a filter-press cell at 6 g-scale, what allows to explore the most suitable regeneration conditions. The effects of the electrolyte, the electrodes composition and the treatment time, have been analysed to find the optimum conditions that maximize the porosity recovery. Afterwards, the process is scaled-up to a 3.5 kg-scale in an electrochemical batch reactor in order to optimize the design of the filter-press cell in terms of the amount of regenerated AC, total electrolyte volume and energy consumption.

## 2. Experimental

### 2.1. Materials and reagents

The AC used was a commercial AC (Filtrisorb TL-820, Chemviron Carbon, Belgium) in granular form (particle size between 0.9 and 2 mm). The spent AC was supplied by a municipal DWTP (Empresa Mixta Valenciana de Aguas S.A., Valencia, Spain). This real spent AC was used during 3 years for the retention of odours and organic compounds as geosmin and 2-methylisoborneol, in unknown concentration, improving the organoleptic properties of water. In addition, due to the hardness of the water in Valencia, spent AC has adsorbed a large amount of inorganic species such as magnesium, silicon, aluminium and calcium.

Sulphuric acid, H<sub>2</sub>SO<sub>4</sub>, from VWR International, sodium sulphate, Na<sub>2</sub>SO<sub>4</sub>, and sodium hydroxide, NaOH, both from Merck, being analytical grade reagents, were used for preparing the aqueous electrolyte solutions. Tap water was used for the preparation of all solutions for the electrochemical regeneration at g- and kg-scale.

### 2.2. Electrochemical regeneration of AC at 6 g-scale

Experiments at 6 g-scale were performed to determine the adequate range of operating conditions (i.e. current density, pH of electrolyte, reactor configuration and type of regeneration) that

allows regeneration of the spent AC at feasible time scale. The electrochemical filter-press cell used in this work has been previously described (Berenguer et al., 2009, 2010b). The lab-scale plant, in divided configuration, consists of two stainless steel tightening plates of dimensions 9 cm × 10 cm and 1 cm thick, a polyvinyl chloride (PVC) hydraulic distributor by compartment of dimensions 5 cm × 5 cm and 0.5 cm thick for electrolyte inlet and outlet, and two mesh-shaped electrodes with a nominal area of 20 cm<sup>2</sup> with 0.5 cm of separation between anode-cathode. In this divided compartment configuration, an ionic membrane, with dimensions of 5 cm × 5 cm, is added between the PVC hydraulic distributors. In addition to the lab-scale cell, several components are needed to fully install this reactor. It consists of a 1 L tank (two in the case of divided configuration) which acts as an electrolyte tank connected to the inlets and outlets of the filter-press electrochemical cell, a magnet pump (PMD-211, Sanso Electric, Japan) (two in the case of divided configuration) which recirculates the electrolyte between the tank and the filter-press cell, a direct current power supply (AL 924A, ELC, Spain) that operates in galvanostatic mode and, a multimeter to monitor the potential of the electrodes.

Electrochemical regeneration tests were carried out applying a current density of 0.025 and 0.075 A cm<sup>-2</sup> from 3 to 24 h to 6 g of dry spent AC placed either in the anodic or the cathodic compartment of the cell, connected through a cationic membrane (Ionac MC-3470, LANXESS Sybron Chemicals Inc., USA). Platinized titanium mesh (Inagasa, Spain) was used as anode while a stainless-steel mesh (Inagasa, Spain) served as cathode. These tests were carried out 200 mL per tank of three different electrolytes (0.05 M H<sub>2</sub>SO<sub>4</sub>, 0.05 M Na<sub>2</sub>SO<sub>4</sub> and 0.05 M NaOH aqueous solutions). The regenerated ACs were recovered from the cell and rinsed in 200 mL deionized water to remove the remaining electrolyte and ACs were dried at 70 °C overnight.

### 2.3. Scale-up study

Taking into account the classification of the scale of operations of electrochemical reactors by Walsh and Ponce de León (2018), we propose the electrochemical AC regeneration from laboratory-scale reactor to pilot-scale reactor. A 3.5 kg-scale electrochemical batch reactor has been designed in accordance to the operating conditions that showed the best recovery of porosity at 6 g of dry AC, and the scaled-up reactor keeps the basic configuration and elements from the lab-scale one. Table 1 compiles the design parameters used for the scale-up of the laboratory reactor. The following

considerations have been drawn during the scaling-up of the process:

- The maximum amount of AC in the batch reactor was set to  $W_{AC} = 3.5$  kg.
- The specific amount of AC ( $P_{AC}$ ) was fixed at 0.500 kg L<sup>-1</sup> of electrolyte.
- The compartment volume,  $V_C$ , was determined from the specific amount of AC ( $P_{AC}$ ):

$$V_C = \frac{W_{AC}}{P_{AC}} = 7 \text{ L} \quad (1)$$

- The bed porosity,  $\epsilon_B$ , was estimated from the carbon bed density ( $\rho_{AC}$ ) that has a value of 0.630 kg L<sup>-1</sup> and the water density ( $\rho_w$ ) that has a value of 1 kg L<sup>-1</sup>:

$$\epsilon_B = \frac{\frac{1}{\rho_{AC}} - \frac{1}{\rho_w}}{\frac{1}{\rho_{AC}}} = 0.37 \quad (2)$$

- The total electrolyte volume per unit of compartment volume ( $V_{\text{electrolyte}}/V_C$ ) was estimated from the carbon density in water ( $\rho_{AC(w)}$ ) which was experimentally determined being 1.130 kg L<sup>-1</sup>:

$$\frac{V_{\text{electrolyte}}}{V_C} = \frac{\frac{1}{P_{AC}} - \frac{1}{\rho_{AC(w)}}}{\frac{1}{P_{AC}}} = 0.56 \quad (3)$$

- The linear velocity of the electrolyte along the carbon bed,  $v$ , used in the kg-scale reactor was matched with that of the laboratory-scale reactor, 0.25 cm s<sup>-1</sup> (Table 1). In consequence, mass transfer rates on the surface of the particle were expected to be similar in both reactors (i.e. value of Sherwood dimensionless number that governs the external mass transfer rate is similar).
- Residence time of the electrolyte was set to 20 s, what allowed to calculate the cross-section area (SEC) of the reactor:

$$\tau = \frac{V_C \cdot \epsilon_B}{v \cdot \text{SEC}} \cdot \text{SEC} = \frac{V_C \cdot \epsilon_B}{v \cdot \tau} = 518 \text{ cm}^2 \quad (4)$$

**Table 1**  
Dimensions and operational parameters.

Parameters from the electrochemical filter-press reactor at 6 g-scale	
Specific amount of AC	0.5 kg L <sup>-1</sup>
Linear velocity of electrolyte	0.25 cm s <sup>-1</sup>
Contact time of electrolyte	20 s
Electrode separation	0.5 cm
Specific volume of electrolyte	33.3 L kg <sup>-1</sup>
Electrode geometric surface area	20 cm <sup>2</sup>
Volume of electrolyte per tank	0.2 L
Parameters from the electrochemical batch reactor at 3.5 kg-scale	
Specific carbon loading	0.5 kg L <sup>-1</sup>
Linear velocity of electrolyte	0.25 cm s <sup>-1</sup>
Contact time of electrolyte	20 s
Electrode separation	10 cm
Specific volume of electrolyte	5 L kg <sup>-1</sup>
Height of the carbon bed	13.5 cm
Width of the carbon bed	50 cm
Cross-section area of the bed	518 cm <sup>2</sup>
Electrode geometric surface area	675 cm <sup>2</sup>
Volume of electrolyte per tank	17.5 L

- Once the area and the volume were determined, the rest of the compartment dimensions as height ( $h_{\text{reactor}} = V_C/\text{SEC} = 13.5 \text{ cm}$ ) and width ( $W_{\text{reactor}} = \text{SEC}/L_{\text{reactor}}$ , where  $L_{\text{reactor}}$  is the electrode separation) were calculated. Since the electrode separation from the lab-scale reactor was impractical, it had been set to 10 cm to allow a proper dimensioning of the reactor, rendering a reactor width of 50 cm.
- The demanded flow rate was  $Q = \text{SEC} \cdot v = 129.5 \text{ cm}^3 \text{ s}^{-1}$ , so that centrifugal pumps that can serve  $138.9 \text{ cm}^3 \text{ s}^{-1}$  were selected.
- The operating intensity current depends on the surface of the lateral size of the reactor, which corresponds to the surface area of the membrane (SME) acting as separator:

$$\text{SME} = h_{\text{reactor}} \cdot W_{\text{reactor}} = 675 \text{ cm}^2 \quad (5)$$

- The operating voltage relies on the reactor resistance, that could be estimated from the electrolyte resistance, membrane resistance and the electrode polarization at  $0.025 \text{ A cm}^{-2}$ . The contribution to the ohmic drop of the last two parameters were estimated from the laboratory-scale experiments ( $\Delta V = 2.5 \text{ V}$ ), whereas the electrolyte resistance was  $1.55 \Omega$ , calculated from the electrical conductivity of  $0.05 \text{ M H}_2\text{SO}_4$  ( $0.0034 \text{ S cm}^{-1}$  at  $25^\circ \text{C}$ ) and the reactor dimensions. In total, the expected voltage demanded to operate the reactor at  $0.025 \text{ A cm}^{-2}$  would be:

$$\Delta V = 2.5 + 1.55 \cdot 16.9 = 28.7 \text{ V} \quad (6)$$

This means that the regeneration experiments requested the power supply to work at very demanding conditions. At this point, two alternatives were proposed: i) performing the AC regeneration at a lower current density of  $0.010 \text{ A cm}^{-2}$  (Experiments labelled as EK-1, Table 2), ii) increasing electrolyte concentration to  $0.5 \text{ M H}_2\text{SO}_4$  (Experiments EK-2 to EK-4, Table 2) and current density of  $0.025 \text{ A cm}^{-2}$ , which has been checked as possible in the regeneration experiments at laboratory-scale. For the latter alternative, the conductivity of the electrolyte was increased 7.46 times due to the higher sulphuric acid concentration, resulting in a less demanding operating voltage of 6.8 V.

- The DC power supply was selected to deliver a maximum voltage of 30 V and a current of 20 A, in agreement with the estimated voltage and current requirements of the reactor.
- The specific volume of electrolyte employed at laboratory-scale was  $33.3 \text{ L kg}^{-1}$ . In order to increase the sustainability of the process, this ratio was decreased to  $5 \text{ L kg}^{-1}$ .

## 2.4. Electrochemical regeneration of AC at 3.5 kg-scale

The electrochemical batch reactor consisted in two compartments of  $50 \text{ cm} \times 10 \text{ cm} \times 15 \text{ cm}$  that were connected through a cationic membrane (Ionac MC-3470, LANXESS Sybron Chemicals

**Table 2**  
Experimental parameters for the electrochemical regeneration of spent AC in batch reactor.

Variable	EK-1	EK-2	EK-3 <sup>a</sup>	EK-4	Units
Electrolyte concentration	0.05	0.5	0.5	0.5	M
Cathode	Pt/Ti	Pt/Ti	Pt/Ti	stainless-steel	–
Anode	Pt/Ti	Pt/Ti	Pt/Ti	Pt/Ti	–
Current	16.9	16.9	16.9	16.9	A
Average voltage	30	6	6	6	V
Regeneration time	5, 8, 24	4	4	1, 2, 3, 4	h

<sup>a</sup> Undivided configuration.

Inc., USA). The reactor walls were made of 1 cm thick methacrylate, having inlet and outlet ports with a diameter of 2.5 cm for the flow of electrolyte. The junctions between the walls and the membrane were sealed with silicone. The outlet port was sealed with a filter to avoid that powdered carbon particles leave the reactor with the electrolyte. No fouling of the filter was observed at the end of the regeneration experiments. A scheme of the electrochemical batch reactor is shown in Fig. 1.

Platinized titanium mesh (geometrical electrode area of  $675 \text{ cm}^2$ , Inagasa, Spain) was used as anode and a cathode while a stainless-steel mesh (geometrical electrode area of  $675 \text{ cm}^2$ , Inagasa, Spain) served as cathode. The anode was fixed to the membrane side of the anodic compartment, while the cathode was emplaced at the outer wall of the cathodic compartment so that the separation between electrodes was set to 10 cm. The current was set to 16.9 A to maintain a current density of  $0.025 \text{ A cm}^{-2}$  and was supplied by a power source (Stelltrentrafo, Phywe, Germany) operating at galvanostatic mode.

Two independent circuits were constructed for servicing respectively the anodic and cathodic compartments with the electrolyte. These circuits consisted of a 25 L polystyrene tank that feeds the centrifugal pumps (P0101, Plastomec, Italy), followed by a ball valve and a flowmeter (FSIV040D, Aliaxis, Belgium) that allowed to keep the electrolyte continuously flowing from the electrolyte tank to the reactor compartment at a volumetric flow rate of  $480 \text{ L h}^{-1}$ . The outlet of the flowmeter was connected to the inlet port of the reactor, so that the electrolyte flows upwards through the compartments. The electrolyte leaves the reactor through the outlet ports, returning by gravity to the tank. The flow of electrolyte was through PVC pipes, with a diameter of 2.5 cm.

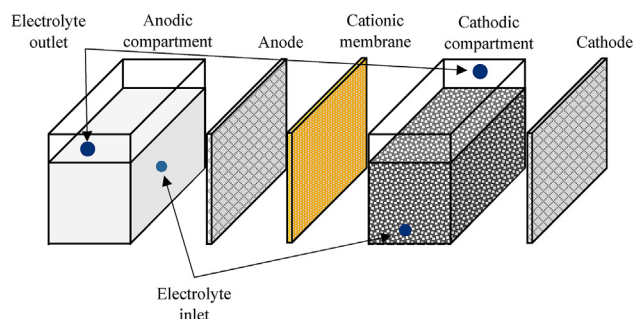
During the experimental study, 3.5 kg of dry AC was loaded in the cathodic compartment of the reactor, spent AC was in contact between cationic membrane and cathode material. The height of the resulting AC bed was 13.5 cm. The total electrolyte to AC ratio was set to 5:1, meaning that 17.5 L of  $\text{H}_2\text{SO}_4$  solution was always present into the cathodic and anodic compartment. Samples of regenerated AC were taken at each hour of treatment from different points of the batch, thus having a homogeneous sample of the process. After each test, ACs were rinsed with 200 mL of deionized water and dried at  $70^\circ \text{C}$  overnight. The experiments were repeated at least two times obtaining similar results.

Table 2 summarizes the different operating conditions (electrolyte concentration, effect of cathode, separation of compartments, voltage and time of regeneration) that have been tested for the AC regeneration at kg-scale.

## 2.5. Analysis of the regenerated AC

### 2.5.1. Physicochemical characterization of the AC

The porous texture of pristine, spent and regenerated samples



**Fig. 1.** Scheme of the 3.5 kg electrochemical batch reactor. The spent AC was placed in the cathodic compartment.

was determined by physical adsorption-desorption of N<sub>2</sub> at -196 °C using an automatic adsorption system (Autosorb-6, Quantachrome Corporation, USA). 100 mg of dry AC was out-gassed at 150 °C under vacuum for 8 h. Total volume of micropores ( $V_{DR}(N_2)$ ) was calculated applying the Dubinin-Radushkevich (DR) equation, the volume of mesopores ( $V_{meso}$ ) was calculated by subtraction of the adsorbed volume at relative pressures of 0.9 and 0.2 and the apparent specific surface area was determined by the Brunauer-Emmett-Teller (BET) equation (Lozano-Castelló et al., 2009). The pore size distribution (DFT-PSD) was calculated by applying the bidimensional nonlocal density functional theory method (2NL-DFT) to the N<sub>2</sub> adsorption isotherms using SAIEUS software (version 2.0, Micromeritics Instrument Corp. 2000–2015) (Jagiello and Olivier, 2012).

Temperature programmed desorption (TPD) experiments were performed with 10 mg of dry AC in a DSC-TGA equipment (SDT 2960 Simultaneous, TA Instruments, USA) coupled to a mass spectrometer (GSD 300 T3 Thermostat Balzers, Pfeiffer Vacuum, Germany) which was used to follow the  $m/z$  signals related to the decomposition of surface functional groups from the AC. The thermobalance was purged for 1 h under a helium flow rate of 100 mL min<sup>-1</sup> and then heated up to 900 °C (heating rate 20 °C min<sup>-1</sup>).

The surface morphology of ACs were studied by scanning electron microscope (SEM) (S-3000 N, Hitachi, Japan), which was coupled to a Rontec X-ray detector for energy dispersive X-ray (EDX) analysis.

### 2.5.2. Evaluation of the recovery of porosity

The percentage of recovery of porosity (%RE) was calculated as the ratio of regenerated to pristine BET surface area, according to the Eq. (7) and it is the parameter used for evaluation of the efficiency of the regeneration method:

$$\%RE = \frac{S_{BET} \text{ of regenerated AC}}{S_{BET} \text{ of pristine AC}} \cdot 100 \quad (7)$$

This parameter has only been calculated for samples whose BET surface is higher than that of the spent AC and with a %RE higher than 85% with respect to the pristine AC after the electrochemical treatment, thus considering them as regenerated and suitable for reuse.

## 2.6. Characterization of the electrolyte solution

The presence and degradation of different organic molecules that were desorbed from the spent AC were followed by conventional UV-VIS absorption spectroscopy (V-670 UV-VIS-NIR spectrometer, Jasco, Spain) over the wavelength range of 180–440 nm. In addition, various aliquots of the electrolyte were collected at different times and submitted to total organic carbon (TOC) determination by Total Organic Carbon Analyzer (TOC-V CSN, Shimadzu, Japan).

## 3. Results and discussion

### 3.1. Regeneration of AC at 6 g-scale

Table 3 summarizes the results obtained from the electrochemical filter-press regeneration experiments. BET surface area ( $S_{BET}$ ) of the regenerated sample and recovery of porosity are also reported and used as a benchmark to follow the achieved degree of adsorption capacity by the AC. The  $S_{BET}$  of the pristine AC sample decreases from 875 to 735 m<sup>2</sup> g<sup>-1</sup> for the spent AC, whereas the micropore volumes decreases from 0.36 to 0.29 cm<sup>3</sup> g<sup>-1</sup>.

The results obtained from electrochemical regeneration experiments revealed that low pH use leads to higher porosity recovery (Table 3). This has been previously studied and may be due to the fact that acidic conditions limit oxygen formation in the anode and improve cathode efficiency (Wang et al., 2008), a decrease in the stability of the H<sub>2</sub>O<sub>2</sub> under basic conditions, and the formation of bicarbonate ions, that may act as radical scavengers (Sowmiya et al., 2016). For these reasons, Karabacakoglu and Savlak (2014) recommends working with an electrochemical regeneration cell with the lowest possible pH value. In this specific spent AC, the pH of the electrolyte is more relevant than the anodic or cathodic regeneration probably due to the important role of inorganic species adsorbed on AC.

The maximum  $S_{BET}$  was attained for AC electrochemically treated in the cathodic compartment (Table 3). This finding was in agreement with previous experiments about the electrochemical regeneration of spent AC in the literature, where the treatment on the cathodic compartment is more effective than the anodic one (Berenguer et al., 2010b; McQuillan et al., 2018). Then, when the regeneration is performed at the cathode, the AC is subjected to a negative potential which builds up a negative charge on the carbon material. It seems that the specific adsorbed molecules in this spent AC are more easily desorbed from the AC and removed away by repulsive interactions. In Table 3 it can be seen how an increase in current density from 0.025 to 0.075 A cm<sup>-2</sup> did not provide an improvement in %RE. Although an increase in current density could promote a better %RE (Zhou and Lei, 2006), the formation of gaseous bubbles may increase electrical resistance being detrimental for regeneration (Martínez-Huitle et al., 2015).

The effect of the treatment time on porosity recovery was analysed by using the most efficient electrochemical filter-press cell configuration (0.05 M H<sub>2</sub>SO<sub>4</sub> as electrolyte, current density of 0.025 A cm<sup>-2</sup>, divided cell and AC located in the cathodic compartment), showing a maximum regeneration after 24 h,  $S_{BET}$  of 850 m<sup>2</sup> g<sup>-1</sup>, %RE of 97% (Table 3). At regeneration times between 1 and 4 h, the desorption of the adsorbed pollutants in the most accessible porosity seems to be easily produced. When the regeneration time increases, the increase in porosity recovery is not so important probably due to the sluggish diffusion of the pollutants through the narrowest micropores and the adsorption of some oxidation products and the readsorption of desorbed organic compounds present in the electrolyte which is recirculated. However, most of the porosity recovery is attained after 4 h. These experimental conditions were considered optimal and were used in the electrochemical regeneration tests at the 3.5 kg-scale.

### 3.2. Electrochemical regeneration of AC at 3.5 kg-scale

Table 4 shows the porous texture and the %RE of the AC after the different regeneration experiments. As can be seen, the porous texture of the spent AC can be recovered with a %RE higher than 95% in the large-scale electrochemical batch reactor. Thus, the surface area of the electrochemically regenerated samples was 97% for EK-1 compared to the pristine sample, 99% for EK-2, 89% for EK-3 and 95% for EK-4. The effect of regeneration time will be discussed in 3.4 section and, the differences between EK-2 and EK-3 will be discussed in 3.5 section.

The N<sub>2</sub>-sorption isotherms obtained for the experiments carried out in the 3.5 kg batch reactor are shown in Fig. 2. The isotherm for the spent AC has a similar shape as that of the pristine AC but a lower adsorption capacity due to porosity being partially occupied by the adsorbed molecules. Interestingly, the electrochemically regenerated AC have higher adsorption capacity but the isotherms for the regenerated AC have a wider knee and higher slope at relative pressures higher than 0.2. This indicates that the

**Table 3**  
Electrochemical regeneration of spent AC at filter-press reactor using an electrolyte concentration of 0.05 M in all the cases.

Current density A cm <sup>-2</sup>	Electrolyte	AC compartment	Time h	Average Voltage V	S <sub>BET</sub> m <sup>2</sup> g <sup>-1</sup>	%RE
0.025	NaOH	Anodic	5	7	660	–
0.025	H <sub>2</sub> SO <sub>4</sub>	Anodic	5	6	790	90
0.025	Na <sub>2</sub> SO <sub>4</sub>	Anodic	5	8	730	–
0.025	NaOH	Cathodic	5	7	710	–
0.025	H <sub>2</sub> SO <sub>4</sub>	Cathodic	5	6	820	94
0.025	Na <sub>2</sub> SO <sub>4</sub>	Cathodic	5	8	720	–
0.025	H <sub>2</sub> SO <sub>4</sub>	Undivided	5	5	750	86
0.025	H <sub>2</sub> SO <sub>4</sub>	Cathodic	1	6	740	85
0.025	H <sub>2</sub> SO <sub>4</sub>	Cathodic	2	6	780	89
0.025	H <sub>2</sub> SO <sub>4</sub>	Cathodic	4	6	820	94
0.025	H <sub>2</sub> SO <sub>4</sub>	Cathodic	8	6	840	96
0.025	H <sub>2</sub> SO <sub>4</sub>	Cathodic	24	6	850	97
0.075	H <sub>2</sub> SO <sub>4</sub>	Cathodic	3	6.5	770	88

**Table 4**  
Characterization of the textural properties of pristine, spent and regenerated AC on a kg-scale using N<sub>2</sub> adsorption isotherms at –196 °C and recovery of porosity.

Sample	S <sub>BET</sub> (m <sup>2</sup> g <sup>-1</sup> )	V <sub>meso</sub> (cm <sup>3</sup> g <sup>-1</sup> )	V <sub>DR</sub> (N <sub>2</sub> ) (cm <sup>3</sup> g <sup>-1</sup> )	%RE
Pristine	875	0.05	0.36	
Spent	735	0.04	0.29	
EK-1-5 h	840	0.09	0.32	96
EK-1-8 h	835	0.08	0.32	95
EK-1-24 h	850	0.09	0.32	97
EK-2-4 h	865	0.09	0.33	99
EK-3-4 h <sup>a</sup>	780	0.07	0.30	89
EK-4-1 h	775	0.09	0.31	89
EK-4-2 h	805	0.08	0.31	92
EK-4-3 h	815	0.08	0.31	93
EK-4-4 h	830	0.10	0.31	95

<sup>a</sup> Undivided configuration.

regeneration produces some widening of the porosity of the AC. This agrees with the observed increase in mesopore volume after regeneration and the lower volume of micropores obtained after the regeneration compared to the pristine AC (Table 4). This widening of porosity suggests that some carbon gasification as consequence of the surface oxidation process may occur (see discussion on surface chemistry).

The results in Fig. 2 and Fig. SM-2 and Table 4, show that there are some differences in the regeneration of the AC depending on the concentration of electrolyte and the separation of compartments. Thus, a lower concentration of electrolyte (samples EK-1) produces an adequate recovery of porosity but the reached voltage is higher than the 25 V limit established by the regulations, even at the low applied current due to the higher resistance of the electrolyte and needing higher regeneration time compared to the experiments done at higher electrolyte concentration (Fig. 2a compared to Fig. 2c). Fig. 2b and Table 4 show that a configuration without a division of compartments produced less recovery of the porosity of the AC. Analysing the EK-2 and EK-4 experiments (Table 4), in the 4th h of treatment, an effect on the cathode material on regeneration efficiency was observed. Pt/Ti cathode produced a higher %RE than stainless steel because the Pt/Ti allowed to desorb better inorganic compounds that favoured the subsequent desorption of organic compounds. In summary, the most interesting configuration to achieve the better regeneration of the AC at the lowest regeneration time, is an electrochemical reactor with separation of compartments and with a high concentration of electrolyte.

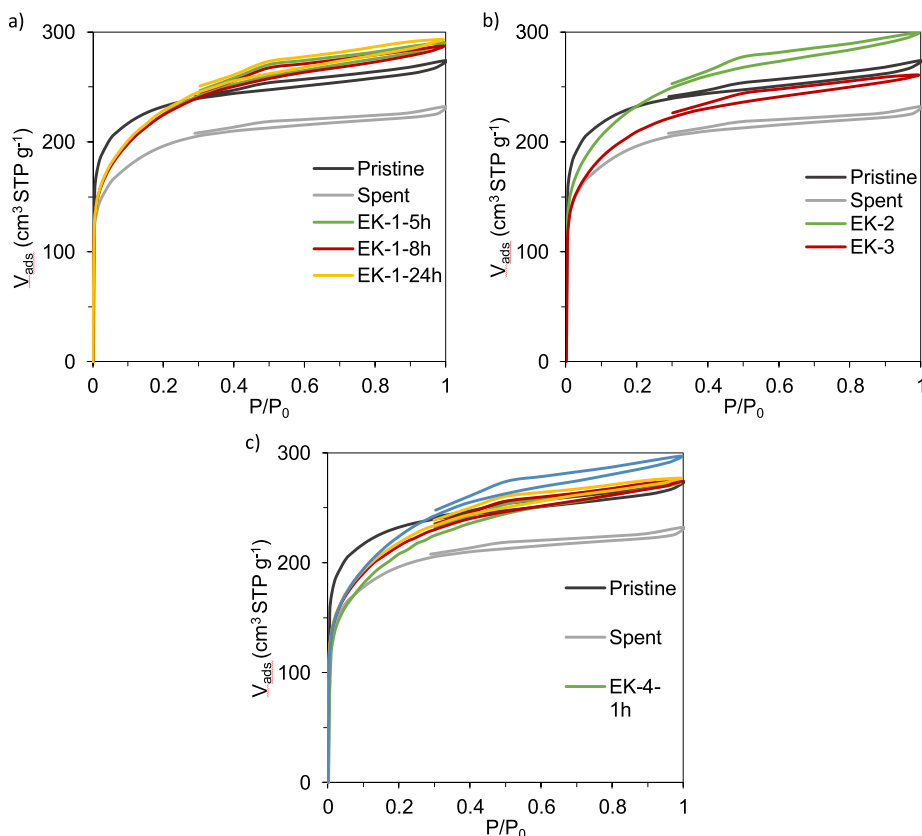
These results can be compared in the literature with the research carried out by Wang and Balasubramanian (2009) where

6.3 kg of spent AC was regenerated by an electrochemical process using a batch reactor with separation of compartments and a different geometry, achieving a regeneration efficiency of 100%. However, deionized water was used in the electrolyte, and the spent AC was retrieved from an industrial filter used in the manufacture of 4,4'-diamino stilbene-2,2'-disulphonic acid meaning that it is saturated with specific organic compounds. Thus, our study validates the process of electrochemical regeneration of AC from drinking and wastewater treatment plants at industrial-scale.

Fig. SM-2 includes the DFT-PSD for the different materials. The results are also in agreement with the changes observed in the isotherms and in the porous texture mentioned before. In Fig. SM-2a, due to the low electrolyte concentration and the long-time of regeneration, a widening of its microporosity is observed in all cases. However, in Fig. SM-2b we can see that the use of a divided or undivided electrochemical reactor did not affect the pore size distribution.

Fig. 3 shows the TPD profiles for CO and CO<sub>2</sub> for the pristine, spent and electrochemically regenerated AC for different experiments in 3.5 kg electrochemical reactor. The evolution of CO indicates the decomposition of surface anhydrides (400–600 °C), phenols (600–700 °C) and carbonyls or quinones (800–900 °C). The evolution of CO<sub>2</sub> indicates the decomposition of surface carboxylic acids (200–500 °C), anhydrides (400–600 °C) and lactones (600–800 °C) (Figueiredo et al., 1999). The results show that there are differences in the surface chemistry of the regenerated AC with respect to the spent and the pristine ones. An increase in CO and CO<sub>2</sub> TPD profiles obtained in the regenerated samples can be observed in relation to the pristine and the amount of functional groups created increases with the regeneration time. This increase in the amount of oxygen functionalities, and particularly carboxylic/anhydride and lactone/phenol groups, is in agreement with the observed carbon oxidation under cathodic polarization (Gineys et al., 2017). Interestingly, although the electrochemical oxidation of the carbon surface produces an increase in oxygen groups in the whole range of thermal stability measured with the TPD, it seems to promote the formation of carbonyl groups with a higher selectivity (see CO peaks for regenerated samples at around 850 °C in Fig. 3).

The observed oxidation can be related to the oxidative action of the hydrogen peroxide formed during the electrochemical oxygen reduction (Delpeux-Ouldriane et al., 2015), which takes part during the electrochemical regeneration tests at the cathode because the electrolyte is not deoxygenated and it is well-known that the oxygen can be reduced in the cathode forming hydrogen peroxide. In particular, Cheng and Teng (2003) proposes that cathodic oxidation may be due to electrogenerated peroxide oxidizing species during the reduction of oxygen dissolved in the electrolyte. Berenguer



**Fig. 2.**  $N_2$  adsorption isotherms at  $-196\text{ }^\circ\text{C}$  for samples obtained from the experiments carried out in electrochemical regeneration of spent AC in a 3.5 kg batch reactor. The data for pristine and spent AC are also included for comparison purposes: a) EK-1, b) EK-2 and EK-3, c) EK-4.

et al. (2009) showed that an electrochemical regeneration treatment using  $H_2SO_4$  as an electrolyte produces the oxidation of AC, due to the presence of oxygen in the solution.

### 3.3. SEM study

Fig. SM-3 shows the SEM images of different AC samples. The pristine AC sample is shown in Fig. SM-3a, while Fig. SM-3b depicts the spent AC sample. By comparison between them, it can be observed that the spent AC contains many white particles that can be related to inorganic matter precipitated during the water treatment (see **Table SM-1**) that can also contribute to the decrease in the adsorption capacity of this sample compared to pristine AC.

The Fig. SM-3c shows the electrochemically regenerated AC for EK-1-24 h test in 3.5 kg batch reactor. It can be observed that the treatment has cleaned the AC surface, leading to a similar surface than the pristine AC. **Table SM-1** shows that the presence of magnesium and calcium in the spent sample is not found in the original and regenerated samples. It can be concluded that this treatment of regeneration of AC does not produce any substantial changes in the morphology of the material, although the adsorption capacity is not completely recovered, due to the presence of some organic and inorganic compounds in the AC surface that cannot be easily removed (Weng and Hsu, 2008).

### 3.4. Effect of regeneration time

The process of desorption of the contaminants present in the ACs is directly related to the regeneration time during the electrochemical treatment (**Tables 3 and 4**). From these results, it can be

deduced that 4–5 h of treatment produces the highest regeneration of the spent AC, while longer times does not result in a significant improvement in the recovery of porosity, but it can produce a larger energy consumption. Moreover, high regeneration times can facilitate the re-adsorption desorbed compounds being detrimental for the adsorption capacity. Similar trends with treatment time are obtained by other authors (Narbaitz and Karimi-Jashni, 2009; Wang and Balasubramanian, 2009; Salvador et al., 2015a; McQuillan et al., 2018). In most of these works, an increase in porosity occurs during the regeneration of the AC due to greater desorption and more oxidation reactions at short times, until an equilibrium state is reached where the reaction is stabilized, and no further regeneration is achieved. The incomplete recovery of porosity of the AC may also be due to the adsorption of the by-products produced in the oxidation of the desorbed compounds, preventing a 100% regeneration (Berenguer et al., 2010b).

### 3.5. Effect of the separation of compartments

To investigate the effect of compartment separation on the electrochemical regeneration of AC, experiments carried out on a g-scale (**Table 3**) and the EK-2 and EK-3 experiments (**Table 4**) are analysed. The cationic exchange membrane is used to separate both compartments and to maintain the pH in both compartments. In addition, it also avoids non-desired reactions in the counter electrode. Therefore, this type of reactor configuration is especially useful when it comes to maintaining high concentrations of oxidizing agents as it is concluded in different studies (Narbaitz and McEwen, 2012; McQuillan et al., 2018). However, several investigations propose the use of reactors without separation of

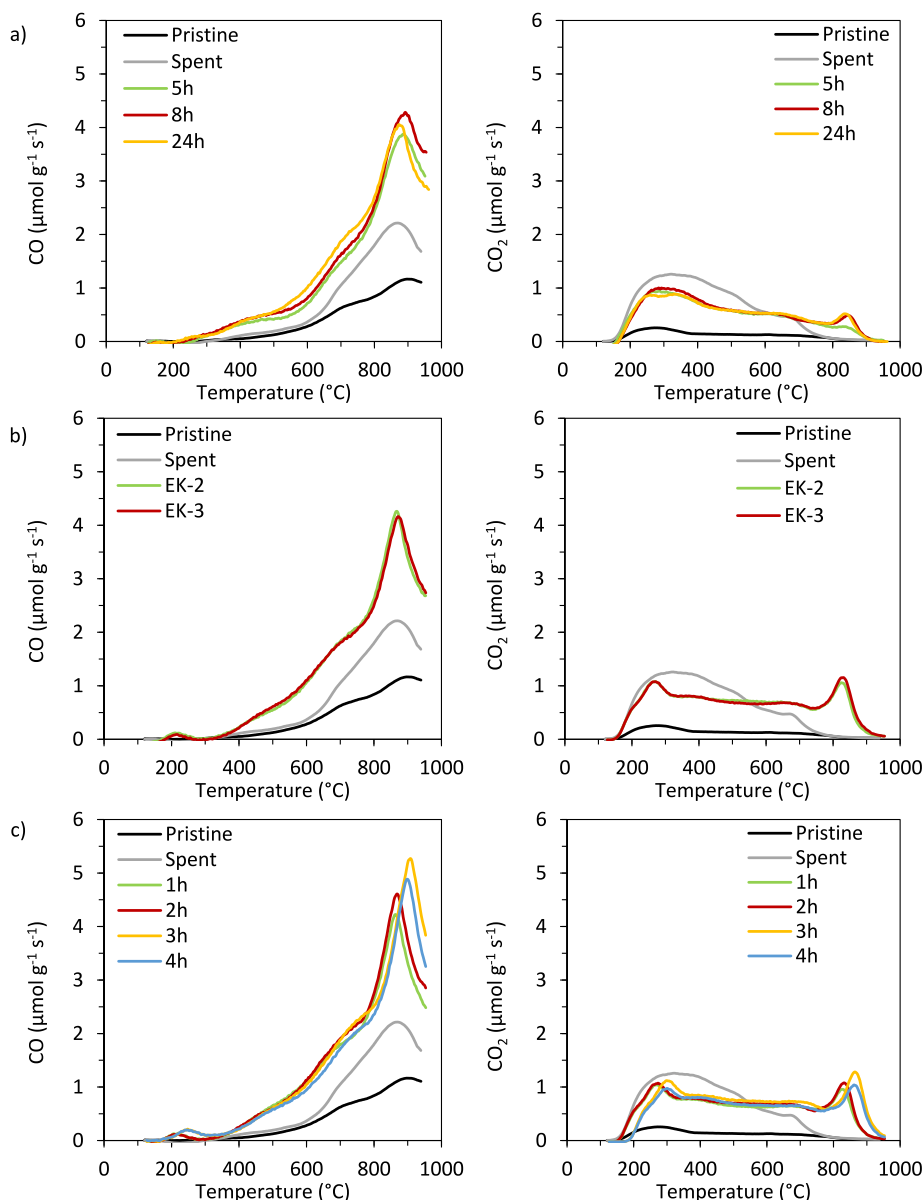


Fig. 3. TPD-MS analysis of pristine and regenerated AC after electrochemical treatment from a) EK-1, b) EK-2 and EK-3 and c) EK-4.

compartments since this configuration is simpler saving cost and time. However, in this way molecules desorbed at the cathode could migrate and be oxidized at the anode and as the conditions of the electrochemical regeneration is recirculating, these oxidized products can adsorb on the AC (Berenguer et al., 2010b; Wang et al., 2013). In addition, the cost of ion exchange membranes and the elimination of gases produced by the electrochemical regeneration treatment of AC must be considered (Narbaiz and Karimi-Jashni, 2012).

In this work, it is found that an improvement of the electrochemical regeneration of the AC occurs when the reactor has a separation of compartments.

### 3.6. Characterization of the electrolyte solution

The analysis of the electrolyte solutions for EK-1 was carried out using UV-VIS spectroscopy (Fig. SM-4). The spectrum for the initial electrolyte is also included for comparison purposes. The spectra

show bands within the following ranges: 190–200, 240–260 and 305–310 nm. The band at 190–200 nm could be assigned to the presence of aromatic species (benzene derivatives), while bands of lower intensity at 240–260 and 305–310 nm can be associated to oxidized aromatic rings (phenols, benzoquinones, etc.). The analysis of the different aliquots of electrolyte samples points out that desorption of organic compounds occurs at short times. The amount of these products does not increase with time, in agreement with the slight variation in the  $S_{\text{BET}}$  after 5 h of treatment. However, for EK-1-24 h the peaks were more intense, especially in the 280–340 nm region. Therefore, the pollutants have just been moved from the AC into the electrolyte solution and re-adsorption of these compounds was not appreciated. The presence of such compounds in the electrolyte has been reported by different authors (Ania and Béguin, 2008; Delpoux-Ouldriane et al., 2015), and Berenguer et al. (2010a) attributed these peaks to the possible degradation in the cathodic compartment of organic compounds through an indirect oxidation process with electrogenerated



hydrogen peroxide. Those hydrogen peroxide or oxygen-containing radicals also react with the carbon material, creating oxygen groups on the surface, Fig. 3 (Maass et al., 2008; Berenguer et al., 2009; Liu et al., 2017).

The TOC results of EK-1 revealed an increase of the total organic carbon concentration as the time of regeneration of the AC increases (3.5 mg L<sup>-1</sup> for EK-1-5 h, 3.9 mg L<sup>-1</sup> for EK-1-8 h and 6.8 mg L<sup>-1</sup> for EK-1-24 h). Similar trend was reported by Weng and Hsu (2008). It must be noticed a clear correlation between TOC values and the UV-VIS spectra from EK-1 samples. As shown in Fig. SM-4, the samples EK-1-5 h and EK-1-8 h presented a very similar spectra and a difference of 0.4 mg L<sup>-1</sup> in the TOC; for the sample EK-1-24 h the UV-VIS peaks showed larger intensities and, consequently, the result in the TOC was also higher.

### 3.7. Energy consumption of the electrochemical regeneration

The energy consumption per unit mass of the regenerated AC, EC (kWh kg<sup>-1</sup>), was calculated according to the Eq. (8):

$$EC \text{ (kWh / kg)} = \frac{(U \cdot I \cdot t) \cdot 10^{-3}}{w} \quad (8)$$

where U is the average potential of the electrochemical cell (V), I is the applied current (A), t the regeneration time (h) and, w the mass of the AC (kg).

Although the filter-press cell did not have optimized design parameters, in the best regeneration conditions, the EC is 2 kWh kg<sup>-1</sup> in the filter-press reactor at the g-scale and 0.11 kWh kg<sup>-1</sup> for the batch reactor at the kg-scale as can be seen in Table SM-2. This difference between both prototypes, at the laboratory-scale and pilot-scale reactor, shows that the scaling-up process has improved the design parameters with the objective of obtaining a more optimized electrochemical reactor for the regeneration of spent AC. These results, in 3.5 kg batch reactor, have a similar trend to those presented by Karabacakoglu and Savlak (2014). Compared with the EC of our prototype with respect to thermal regeneration, the most established technology for this process, like that carried out by different authors (Bayer et al., 2005; Muñoz et al., 2007) a reduction of EC of 79% is obtained with the batch reactor compared to the thermal regeneration. With regard to reducing energy consumption by electrochemical regeneration of AC compared to production of new AC, a 94% decrease can be achieved, obtaining the same data as published by Weng and Hsu (2008). These results show that the electrochemical regeneration of real spent AC can be applied as an alternative process to thermal regeneration methods from the point of view of energy consumption.

## 4. Conclusions

This study shows that electrochemical regeneration of AC at 6 g-scale has been achieved using a commercial AC and a filter-press electrochemical reactor designed in previous works. The results show a S<sub>BET</sub> recovery up to 97%, providing the most suitable conditions for the scaling-up. Afterwards, a batch reactor for the electrochemical regeneration of AC at 3.5 kg-scale has been designed and used spent AC from DWTP samples at different operating conditions, obtaining a S<sub>BET</sub> regeneration efficiency up to 99% in the optimum case. The effect of the time of the regeneration treatment has been studied and the results indicate an increase of the regeneration efficiency during the four first hours. From that time, the regeneration efficiency only increases marginally, which can be related to the equilibrium between the adsorption/desorption processes being finally reached. A study conducted about the effect of the separation of compartments concludes that the better

results have been obtained when a cationic exchange membrane is used to divide the compartments, locating the AC in the cathodic compartment. To sum up, this work demonstrates for the first time the feasibility of electrochemical regeneration of spent AC used in DWTPs at kg-scale. However, after the electrochemical treatment there was a partial oxidation of AC that should be avoided, being the subject of further investigations. Moreover, electrochemical recovery of porosity of spent AC can be considered a green chemistry technology cleaner, cheaper and more sustainable than conventional methods like thermal regeneration.

### CRedit author statement

Borja Ferrández-Gómez: Conceptualization, Methodology, Investigation, Writing - original draft. Ramiro Ruiz-Rosas: Conceptualization, Methodology, Investigation, Writing - original draft. Samuel Beaumont: Methodology, Investigation. Diego Cazorla-Amorós: Conceptualization, Methodology, Supervision, Writing - review & editing, Funding acquisition. Emilia Morallón: Conceptualization, Methodology, Project administration, Supervision, Writing - review & editing, Funding acquisition.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2020.128399>.

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