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# Assessment of the energy needs for the arsenic remediation of drinking water by electrodialysis

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## ABSTRACT

In this work, three different arsenic-rich well waters from the territory of Viterbo (Italy) were processed via electrodialysis (ED) to remove arsenic. These waters having different chemico-physical parameters (i.e. pH, electric conductivity, main anion and cation, as well as arsenic, concentrations) were preliminarily submitted to limiting current tests to determine the electric current to be applied in constant-current batch desalination trials by using a laboratory-scale electrodialyzer. Arsenic removal was non-selective, its percentage removal ($\rho_{As}$) being about 71% of the desalination degree (DD) achieved (i.e. 80–85%) at the end of any batch desalination. The specific energy consumption ($w$) was found to be linearly related to DD in the range of 71–75 W h m$^{-3}$. Thus, contrary to generally accepted belief, arsenic removal by ED resulted to be by far less energy intensive than reverse osmosis, its specific energy consumption being almost of the same order of magnitude as that associated with the ED removal of nitrates from drinking water. Two empirical relationships ($\rho_{As}$ and $w$ against DD) were developed to assess firstly the DD associated with prefixed arsenic removal and then to estimate the corresponding specific electric energy consumption.

*Keywords:* Arsenic removal percentage; Drinking waters; Electrodialysis; Limiting current; Energy consumption

## 1. Introduction

Among the numerous pollutants in water streams identified as toxic and harmful to the environment and human health, arsenic is considered as a high priority one. It occurs naturally in rocks and soils, water, air, plants, and animals, being released into the environment by volcanic activity, erosion of rocks and minerals, and forest fires, as well as by anthropogenic activities related to the use of pesticides and fertilizers in agriculture, fossil fuel combustion, mining, smelting, etc.

Arsenic levels are generally higher in underground drinking water sources than in surface waters. Quite severe arsenic poisoning has been reported in some countries, such as Bangladesh, India, etc., where its content in water ranges from 300 to 4,000 μg L$^{-1}$ [1,2]. The presence of arsenic in drinking waters has been also reported in Taiwan, Chile, and Argentina [3], as well in the USA [4,5] and Europe [6,7].

The predominant dissolved arsenic species in ground water are the trivalent As(III) and pentavalent

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As(V) inorganic forms, the latter being more prevalent in surface water while the former in anaerobic ground waters [2]. Generally, inorganic forms are more toxic than organo-arsenic species, their toxicity varying as follows: arsenite > arsenate > mono-methylarsenate > dimethylarsenate [8]. Thus, arsenic is currently regarded as highly toxic and the World Health Organization recommended to lower the maximum contaminant level (MCL) of arsenic in drinking water to 10 μg $L^{-1}$ from an earlier value of 50 μg $L^{-1}$ [9]. Such a new MCL has been accepted by the United States Environmental Protection Agency (US-EPA) since 2002 [10], as well as by the European Community (EC) since 1998 [11]. Such a Directive was then extended to Italy by 2 February 2001 [12]. Such a new MCL has affected a significant number of water suppliers and users in many regions of the world, particularly in the United States [13] and Italy [6]. There is therefore a remarkable demand for developing efficient methods to remove arsenic from drinking waters [2,4,14,15]. Among the treatment methods developed so far, it is worth citing sorption and ion-exchange, precipitation, coagulation and flocculation, reverse osmosis (RO) and nanofiltration, electrodialysis (ED), biological processes, lime softening, etc. Most of these methods are effective to remove As(V), thus the arsenite form As(III) has to be oxidized to As(V) by adding ferric chloride, potassium permanganate, or chlorine. Lime softening and iron co-precipitation appear to be the most effective and inexpensive removal technology, the arsenic removal effectiveness ranging from 90 to 95% [4,14,16]. Also membrane separation methods, such as RO and ED, have been found to be very effective, their As removal efficiency being as high as 80–99% [4]; but both of them are regarded as costly owing to their high specific energy consumption [17]. Actually, very few studies have been carried out to assess the ED process for the specific removal of arsenic [14,18]. Among these, it is worth pointing out the application of the electrodialysis reversal (EDR) process (that is a conventional ED process with periodic reversal of the polarity of the electrodes to minimize membrane fouling) to treat city water containing a mixture of As(III) and As(V), and groundwater containing mostly As(III) at San Ysidro (New Mexico, USA) [19]. The removal percentage of As was as low as 28% for the latter, but as high as 81% for the former. From such results the ED process appeared to be inefficient in arsenite removal, even if the removal of fluoride, nitrate, and arsenate was greater than that obtained via RO.

The only industrial-scale plant applying the EDR process to remove As from drinking water is operating in the town of Magna (Utah, USA). Its daily capacity amounts to 22,728 $m^3$, the incoming arsenic being reduced by 82% to 2.2 μg $L^{-1}$ [20].

The aim of this work was to assess the removal degree and especially the specific energy consumed to remove arsenic from three typical well waters of the Cimino-Vico volcanic areas in Central Italy by the application of ED.

## 2. Materials and methods

### 2.1. Raw materials and chemicals

The As-rich water samples used in this study were collected from three different wells, namely Cetti, Ripa, and Tobia, located at Ponte di Cetti, Piscin di Polvere, and Tobia in the countryside of the town of Viterbo (Italy) (Fig. 1), and labeled as PC, PP, and TO, respectively. In order to remove stagnant water from the pipe of each well, a long-time pumping was carried out before collecting each water sample in 5-L PET bottles. Immediately after transportation to the laboratory, any sample was filtered through membrane filters (type WNC Cellulose Nitrate Membranes, 47 mm diameter, 0.2-μm pore size; Whatman, USA) and then immediately submitted to ED testing.

Several chemicals, such as sodium borohydride ($NaBH_4$), sodium hydroxide, and ascorbic acid (Sigma-Aldrich, Steinheim, Germany), potassium iodide and hydrochloric acid (Merck, Darmstadt, Germany), stock solutions containing 1 g $L^{-1}$ of $As_2O_3$ in diluted aqueous nitric acid (mono-element, calibration standard for inductively coupled plasma-optical emission spectroscopy, ICP-OES) from C.P.A. Ltd (Stara Zagora, Bulgaria), were used for sample pretreatment and total inorganic arsenic determination. All chemicals used were of analytical-reagent grade. De-ionized water (electric resistivity of 18 MΩ cm) was produced by a Milli-Q system (Millipore Corporation, USA).

The solutions used to fill the electrode and concentrating compartments and clean the ED stack were prepared by dissolving analytical-grade sodium chloride or sodium hydroxide into or mixing an analytical-grade HCl solution with de-ionized water.

### 2.2. Equipment

A laboratory-scale electrodialyzer model EUR2 (Eurodia Industrie SA, Wissous, France) was used. The ED stack was composed of 9 cation- (Neosepta CMX-Sb) and 8 anion- (Neosepta AMX-Sb) exchange membranes (Tokuyama Soda Co, Tokyo, Japan), forming a number of cell pairs ($N_{cell}$) equal to eight. The main characteristics of the ED stack and electro-membranes used were reported previously [21].

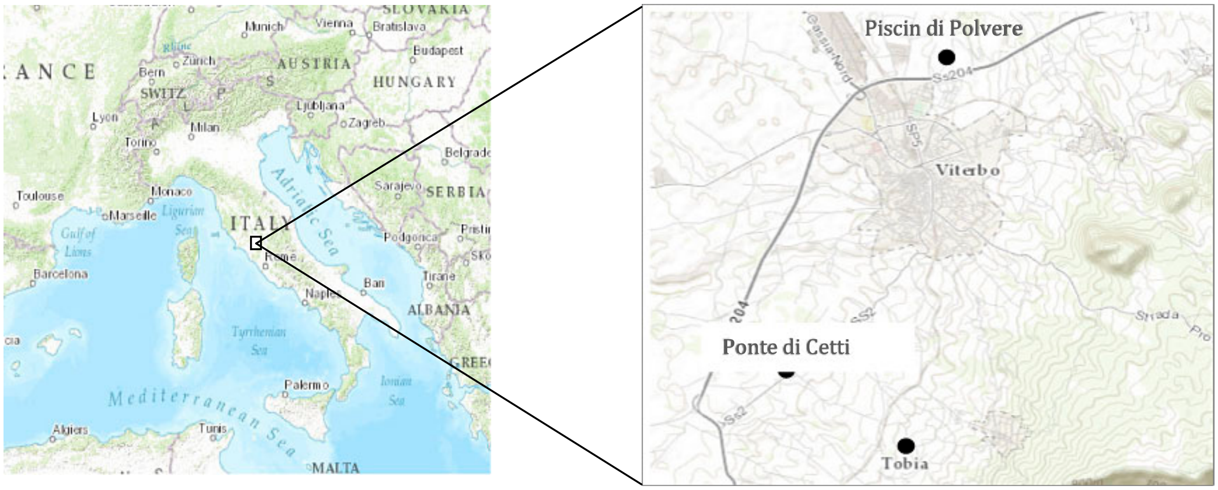

Fig. 1. Location map of the area of Viterbo (Italy), from where well water samples used in this work were collected.

The direct current (DC) generator Mod. N5767A (Agilent Technologies Inc., Santa Clara, CA, USA) could supply voltage (*E*) and current (*I*) in the ranges of 0–60 V and 0–25 A, respectively.

The dilute (D), concentrate (C), and electrode rinsing solution (ERS) were stocked into three 1.6-$dm^3$ PVC tanks equipped with coils for temperature control and re-circulated through the ED stack by means of 3 polypropylene centrifugal pumps. Both C and D tanks were equipped with a 2-m high Plexiglas tube provided with a millimeter scale to precisely assess volume variation in C and D tanks. The D, C, and ERS recirculation flow rates were manually regulated by means of ball valves and flow-meters (accuracy: 5% of the top scale).

The electric conductivity ($\kappa$) of the solutions flowing out of C, D, and ERS compartments was on-line measured as reported previously [21].

Before starting the experimental campaign, the ED plant was thoroughly cleaned-in-place by performing a series of re-circulation cycles of 20 min each with neutral (de-ionized water), acidic (aqueous HCl 2.5% w/w), and alkaline (aqueous 3 kg $m^{-3}$ NaOH) solutions. A series of re-circulation cycles with de-ionized water was performed between batch experiments. When the plant was not in use, the membrane stack was filled with 0.5 M aqueous NaCl.

### 2.3. Experimental procedure and operating conditions

All the experiments were operated in batch mode at 20°C by continuously recycling both D and C at about constant flow rates (148.2 L $h^{-1}$) and ERS at a nominal flow rate of 300 L $h^{-1}$. Electrode rinsing was carried out by recirculating an aqueous solution containing (4.53 ± 0.15) g $L^{-1}$ of NaCl for all experiments. In all experiments, hold-up volume of residual de-ionized water in C and D compartments after the rinsing cycles was estimated as equal to about (0.28 ± 0.10) and (0.24 ± 0.09) L, respectively.

#### 2.3.1. Current–voltage tests

A first series of trials (*I–E* tests) was performed to assess the experimental *I–E* curves for the three water samples under treatment, each one being diluted with de-ionized water to obtain an electric conductivity of ~9 mS $m^{-1}$, this value corresponding approximately to the final electric conductivity of the water samples submitted to ED desalination tests. Such *I–E* tests were carried out by flowing the aforementioned diluted samples through the C and D compartments of the ED stack. In this way, it was possible to assess the limiting current ($I_{\text{lim}}$) so as to perform any desalination test at $I < I_{\text{lim}}$. To this end, the hydraulic circuit of the ED laboratory-scale plant was modified by discharging the solutions undergoing concentration or dilution as flown out of the ED stack into tank D or C, respectively. Consequently, the electric conductivity ($\kappa$) of the solutions in the alternating compartments did not vary, thus resulting in about a constant overall resistance of the ED stack. After re-circulating any solution through the ED stack up to monitor a constant electric conductivity in C, D and ERS tanks, the DC generator was switched on to allow the voltage applied to the ED stack to be increased step by step from 3 to 40 V. After any step increase in the voltage applied, the system was allowed to reach the steady state for 3–5 s before recording the current applied to the ED stack.

### *2.3.2. Desalination tests*

Tank D was fed with (2.24 ± 0.09) L of any water sample, while tank C was fed with (2.28 ± 0.10) L of an aqueous solution containing (4.60 ± 0.15) g $L^{-1}$ of NaCl.

After re-circulating such solutions through the ED stack and corresponding reservoir up to achieve a constant electric conductivity in any tank, the DC generator was switched on and the electric current set to 0.1 A. The desalination test was stopped when the desalination rate of the water sample under treatment reached about 80–87%. A validation test was also carried out at a lower current (0.05 A) using the PP water sample. The duration of the tests at $I = 0.1$ A ranged from (8.4–13.0) min. During any desalination trial, five water samples (20-mL each) were withdrawn and analyzed as reported below.

## *2.4. Analytical methods*

Water samples, as such or submitted to ED testing, were filtered through 0.45-μm filters (Millipore, USA), split into two aliquots, and collected in clean Nalgene® narrow-mouth bottles. Each bottle was previously soaked with 10% hydrochloric acid for 48 h, rinsed sequentially with distilled water, and then with deionized-distilled water two or three times. The first aliquot was used to measure the electric conductivity ($\kappa$), pH, and concentrations of major anions (i.e. $HCO_3^-$, $SO_4^{2-}$, $NO_3^-$, $Cl^-$, and $F^-$) and cations (i.e. $Na^+$, $K^+$, $Ca^{2+}$, and $Mg^{2+}$). The second one was acidified by adding 1 μL of a 12% (w/w) aqueous HCl solution per mL and stored at 4˚C in the dark for later determination of total inorganic arsenic.

The aforementioned major anions and cations were determined by ion chromatography using a conductivity detector. More specifically, a Hamilton PRP-X 100 column (4 mm × 100 mm; Hamilton, USA), fed with an aqueous solution containing 4 mM p-hydroxybenzoic acid and 2.5% (w/w) methanol at pH 8.4 at a flow rate of 1.0 mL $min^{-1}$ as eluent, was used to measure anion concentrations (chemical suppression was used for all anions, except for $HCO_3^-$); whereas an Alltech® Universal Cation HR column (4.6 mm × 100 mm; Alltech Associates Inc., USA), fed with aqueous 1.5 mM $H_2SO_4$ at a flow rate of 1 mL $min^{-1}$ as eluent, was used to determine cation ones. The coefficient of variation of replicated measurements varied between 0.0 and 15.5% with a mean of 5.1%.

The electric conductivity ($\kappa$) and pH of any water sample was measured by means of a WTW conductometer (model Inolab Cond Level 1, WTW, Germany) and an ORION pH-meter (model 420A+; Orion, USA), respectively.

Total inorganic As concentration was assessed by means of an ICP-OES spectrometer (model Optima™ 8000 DV, Perkin Elmer, USA) coupled to a commercial hydride generation (HG) system (model FIAS, Perkin Elmer, USA) [22,23]. The measurements were carried out at three wavelengths (i.e. 193.696, 188.979, and 197.197 nm) presenting the lowest interference and highest analytical signal-to-background ratio. All samples were read in duplicate and averaged, the coefficient of variation varying between 0 and 4.3% (mean 1.5%). The wavelength of 193.696 nm was the one chosen to estimate the As concentration, the other two lengths (197.197, 188.979) being used to confirm its accuracy, as well as absence of potential interference.

Prior to HG, samples and standards were pre-reduced in a polypropylene vessel by mixing 5 mL of any sample or standard with 5 mL of a reducing solution (prepared by dissolving 5 g of pure grade ascorbic acid and 5 g of KI in 100 mL of de-ionized water), and 5 mL of aqueous 10% (w/w) HCl, and letting the resulting mixture stand for 60 min. The reducing agent for HG was freshly prepared every day by dissolving 2 g $L^{-1}$ of $NaBH_4$ in an aqueous solution containing 0.5 g $L^{-1}$ of NaOH.

Calibration was carried out by using several aqueous solutions at diverse As concentrations, these being prepared by spiking appropriate amounts of the As standard into a volumetric flask and diluting to required volume with de-ionized water. The calibration straight lines were accepted when the coefficient of correlation ($r$) was >0.999.

As(III) and As(V) concentrations were determined by estimating the total inorganic As concentration as reported above, while As(III) concentrations were determined according to the method by Anthemidis et al. [24] and Howard [25], that required all samples to be acidified with hydrochloric acid to a final concentration of 2.3 mol $L^{-1}$ prior to undergoing HG so as to avoid the aforementioned pre-reduction step.

## 3. Results and discussion

### *3.1. Characterization of well water samples used*

Table 1 shows the chemico-physical analyses of the three well water samples used in this work. Their As concentrations varied from 41 to 49 μg $L^{-1}$, that is within the range of (1.6–195) μg $L^{-1}$ previously assessed in a survey of 65 well water samples collected in the same Cimino-Vico volcanic area by Angelone et al. [6]. Their pH ranged from 7.4 to 8.5. Only As(V) was present in the water samples tested, as assessed

Table 1
Main physico-chemical characteristics of the three water samples (TO, PP, and PC) used in this work: mean and standard deviation of concentration of major anions ($HCO_3^-$, $SO_4^-$, $NO_3^-$, $Cl^-$, $F^-$) and cations ($Na^+$, $K^+$, $Ca^{++}$, $Mg^{++}$), electric conductivity ($\kappa$) and pH at 20°C and total inorganic arsenic concentration [As]. All measurements were replicated three times

| Sample | pH (–) | $[F^-]$ (mg $L^{-1}$) | $[Cl^-]$ (mg $L^{-1}$) | $[NO_3^-]$ (mg $L^{-1}$) | $[SO_4^-]$ (mg $L^{-1}$) | $[HCO_3^-]$ (mg $L^{-1}$) | $[Na^+]$ (mg $L^{-1}$) | $[K^+]$ (mg $L^{-1}$) | $[Mg^{2+}]$ (mg $L^{-1}$) | $[Ca^{2+}]$ (mg $L^{-1}$) | $\kappa$ (mS $m^{-1}$) | [As] (μg $L^{-1}$) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| TO | 7.7 ± 0.3 | 1.6 ± 0.1 | 15.5 ± 0.7 | 7.4 ± 0.6 | 8.6 ± 0.5 | 89.0 ± 6.3 | 14.3 ± 1.4 | 18.4 ± 1.0 | 5.5 ± 0.5 | 14.7 ± 0.1 | 22.4 ± 0.2 | 30.9 ± 0.3 |
| PP | 8.5 ± 0.1 | 1.4 ± 0.1 | 22.6 ± 1.9 | 20.9 ± 1.6 | 21.4 ± 1.6 | 143.2 ± 8.6 | 35.2 ± 0.5 | 7.7 ± 0.4 | 8.0 ± 1.2 | 29.2 ± 2.8 | 35.8 ± 0.6 | 49.1 ± 0.9 |
| PC | 7.9 ± 0.1 | 1.4 ± 0.0 | 15.4 ± 0.5 | 15.5 ± 0.9 | 20.6 ± 0.3 | 138.3 ± 10.1 | 41.3 ± 0.1 | 13.1 ± 0.2 | 5.8 ± 0.1 | 17.1 ± 1.2 | 26.9 ± 0.5 | 39.9 ± 4.1 |

by using the method reported by Anthemidis et al. [24] and Howard [25]. Thus, by referring to the redox potential-pH diagram plotted by Angeloni et al. [6], the dissolved arsenic species in the samples under study fell within the arsenate domain ($H_2AsO_4^-$ and $HAsO_4^{-2}$), with a preponderance of such species.

The major anion present was bicarbonate ($HCO_3^-$), generally followed by sulfate ($SO_4^{2-}$) and chloride ($Cl^-$), whereas the major cation was sodium ($Na^+$), followed by calcium ($Ca^{2+}$) and potassium ($K^+$) in agreement with the composition of natural ground waters [26]. By accounting for their molar concentrations and valences, it was possible to calculate the sum of positive and negative charges and assess that their differences differed by (0.01–0.8) %, thus establishing the accuracy of the analytical methods used.

Since the Piper trilinear diagram [27] is highly used to classify water samples, it was possible to construct the Piper diagram shown in Fig. 2 by using JMP software release 11 (SAS Institute, Cary, NC). In particular, the ionic composition of several water samples from the Cimino-Vico volcanic area was extracted from Angeloni et al. [6] and compared to that of the water samples examined here. The apexes of the cation plot (Fig. 2(a)) are calcium, magnesium, and sodium plus potassium cations, while those of the anion plot (Fig. 2(b)) are hydrogen carbonate, sulfate, and chloride anions. It can be noted that the ionic composition of well waters tested here is quite similar to that of those assayed by Angelone et al. [6]. Thus, well waters under testing are to be regarded as characteristics of ground waters of the Viterbo area.

The electrical conductance ($\kappa$) of natural waters mostly depends on their anionic and cationic contents and varies in the range of 10–500 mS $m^{-1}$ [26]. Owing to the fact the such waters are dilute solutions, Fig. 3 shows a linear relationship between $\kappa$ and the total ionic concentration (TIC), that was expressed in mg $L^{-1}$:

$$\kappa = a\mathrm{TIC} \quad (r^2 = 0.989) \qquad (1)$$

where $a$ is an empirical proportionality coefficient, equal to $(0.115 \pm 0.009)$ mS $m^{-1}$ L $mg^{-1}$, and thus in line with that characterizing numerous natural waters [26].

Conductivity measurements are used regularly in many industrial and environmental applications as a fast, inexpensive, and reliable way of assessing the ionic content in a solution [28]. Thus, the time course of $\kappa$ was monitored in the following ED desalination tests to estimate the reduction in TIC. Moreover, owing to the small $Ca^{2+}$ and $Mg^{2+}$ concentrations in the water samples used (Table 1), electro-membrane fouling was regarded as negligible and no hardness removal pretreatment was carried out.

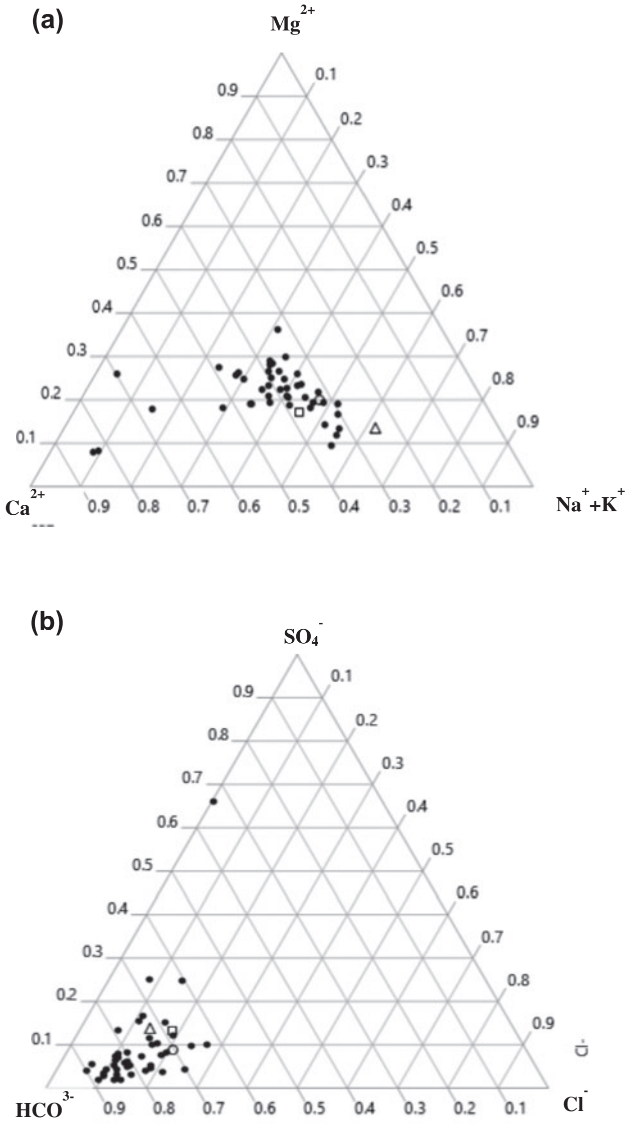

Fig. 2. Piper diagram comparing the ionic composition of the three waters investigated in this work (○, TO; □, PP; △, PC) to that of several well water samples (closed symbols) from the same geographical area [6]. Compositions on the triangular plot for cations (a) and anions (b) are expressed as equivalent fractions.

### 3.2. Limiting current experiments

The so-called limiting current density is the first value at which current density is diffusion limited

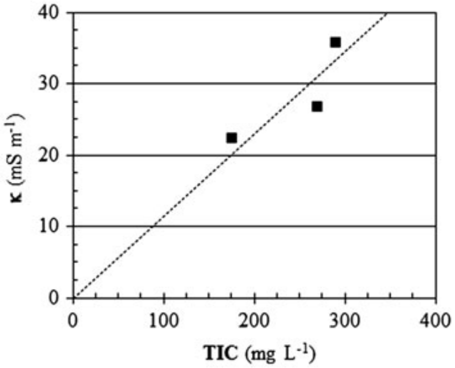

Fig. 3. Effect of the TIC on the electrical conductance ($\kappa$) of well waters used in this work.

[29], that is the value at which the electrolyte concentration at any membrane surface falls to zero. It can be determined by measuring the current ($I$) flowing through the ED stack as the voltage $E$ applied is progressively increased and the same solution is re-circulating through both the ED compartments. Generally, in the first region (the so called *ohmic region*) the $I$–$E$ relationship is linear. As $E$ increases, the solute concentration at the membrane surface reduces. As soon as it falls to zero, the first deviation in the linear trend of the $E$ vs. $I$ plot is observed. Any further increase in $E$ gives rise to a smaller increase in $I$, thus leading to the second region (sometimes called *plateau region*) [30]. By continuing to increase $E$, $I$ tends to increase again, thus allowing the presence of the third over-limiting region to be revealed. In conventional ED stacks, as the limiting current is exceeded, the apparent resistance of the cell rises sharply, the pH of the dilute falls, while that of the concentrate increases, the Coulomb efficiency falls [30].

Fig. 4(a) shows the $I$-vs.-$E$ curves for the three waters under study. Although these tests were carried out by using a conventional ED stack composed of alternating anionic and cationic electro-membranes, all curves exhibited the typical pattern observed with ED stacks composed of a single electro-membrane type in conjunction with aqueous solutions of sodium salts of strong [31] or weak monoprotic [32–34], diprotic [35], and triprotic [36] acids. In particular, for the TO sample, the typical three-region pattern of the $I$–$E$ relationship was observed (Fig. 4(a)). According to Rapp and Pfromm [37], the limiting current ($I_{\mathrm{lim}}$) was roughly determined as the minimum value of the ($E/I$) vs. ($1/I$) curve (Fig. 4(b)). By operating under constant superficial velocity, for the water samples under study the limiting current ranged from 0.11 to 0.14 A. Thus, any desalination test was carried out at a constant current of 0.1 A or lower.

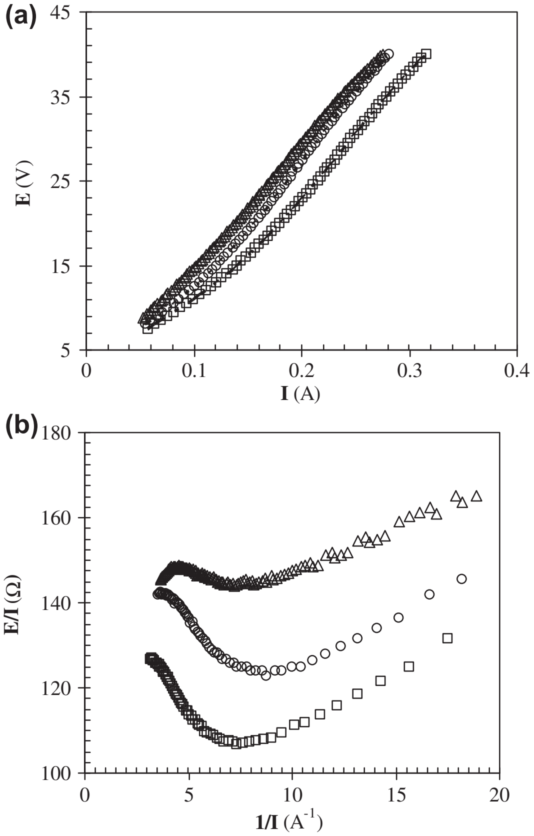

Fig. 4. Voltage–current ($E$–$I$) tests for the three waters under study (PC: □; PP: ○; TO: △) as carried out at 20°C and constant volumetric flow rate (150 L h$^{-1}$) using the membrane pack described in the text: (a) voltage applied to the ED stack ($E$) vs. the electric current ($I$); (b) apparent electrical resistance ($E/I$) vs. the reciprocal of the electric current ($I$). The original waters were diluted with deionized water so as to set their corresponding final values of the electric conductivity to 9.07 (PP), 10.74 (PC), and 9.59 (TO) mS m$^{-1}$, respectively.

### 3.3. Desalination tests

The three water specimens were treated in the lab-scale ED plant by setting the temperature and electric current at 20°C and 0.1 A, respectively. The time course of the electric conductivity in C ($\kappa_C$) and D ($\kappa_D$) compartments, voltage applied to the electrodes ($E$), and solution volume ($V_D$) in the D tank, as well as the concentrations of total arsenic [As], and sodium [Na$^+$], and chloride [Cl$^-$] ions, was monitored, as shown in Fig. 5.

As soon as the D, C, and electrode compartments of the ED plant had been charged with the water sample, brine, and ERS, respectively, any liquid was re-circulated through the ED stack up to monitor constant electric conductivity and temperature in the

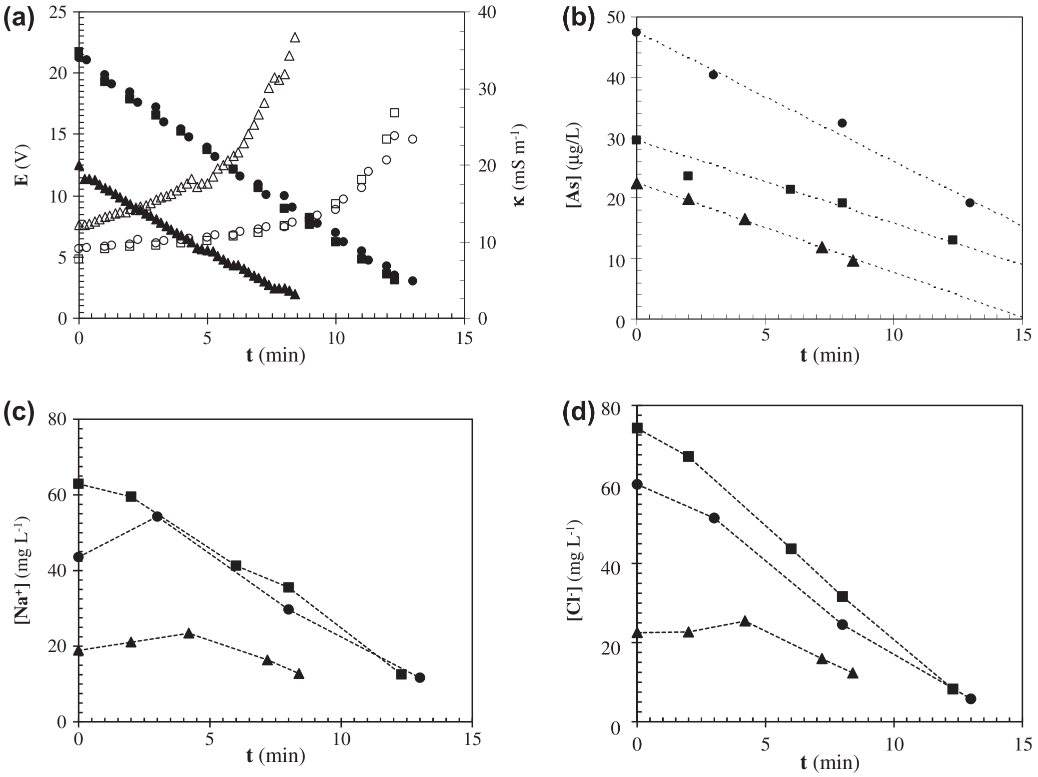

Fig. 5. Main results of the ED desalting tests carried out at $I = 0.1$ A and 20℃ for the three water samples under study (PC: ■, □; PP: ●,○; TO: ▲, △): time course of (a) the electric conductivity of the water sample under treatment ($\kappa$: closed symbols) and electric voltage applied to the ED stack ($E$: open symbols) vs. time ($t$); (b) total inorganic arsenic concentration ([As]: closed symbols); (c) sodium ion concentration ([Na$^+$]: closed symbols); (d) chloride ion concentration ([Cl$^-$]: closed symbols). The broken lines shows the trend of the experimental data.

corresponding reservoir. After that, the DC generator was switched on. It was noted that the original ion concentrations of the water samples examined (Table 1) were different from those in the diluting compartment. This was in all probability due to the water sample dilution (this depending on the liquid hold-up of the D compartment), and the diffusion of selected ions across the membranes (owing to their different concentrations in C, D, and ERS). Also, the contact of the water sample with the electro-membranes resulted in unpredictable ion exchanges.

The voltage applied to the electrodes ($E$) increased with time ($t$) from an initial value of 5.6–7.6 V to 14.5–23.0 V (Fig. 5(a)) in consequence of the linear decrease in the electric conductivity ($\kappa_D$) in the diluting compartment. Similarly, a linear decrease in the arsenic concentration [As] was noted (Fig. 5(b)), this implying an implicit relationship between [As] and $\kappa$. On the contrary, in the PC and TO samples the concentration of sodium ions in the dilute tended to increase during the first 3–5 min (Fig. 5(c)), their electro-migration being overwhelmed by the diffusion of Na$^+$ from the concentrating compartment to the diluting one. Also the transport of chloride ions during the same time interval appeared to be affected by diffusion, especially in the TO water sample (Fig. 5(d)).

From the data collected, it was possible to estimate the reduction in the dilute electric conductivity or desalination degree (DD), electric energy consumed ($W$), specific energy consumed ($w$), net amount of total arsenic transferred from D to C tank ($\Delta m_{\mathrm{As}}$), and percentage arsenic removal ($\rho_{\mathrm{As}}$) as follows:

$$\mathrm{DD} = \frac{\kappa_0 - \kappa}{\kappa_0} \tag{2}$$

$$W = \int_0^t EI\,dt' \tag{3}$$

$$w = \frac{W}{V_{\mathrm{D}}} \tag{4}$$

$$\Delta m_{\mathrm{As}} = V_{\mathrm{D0}}[\mathrm{As}]_{\mathrm{D0}} - V_{\mathrm{D}}[\mathrm{As}]_{\mathrm{D}} \tag{5}$$

$$\rho_{\text{As}} = \Delta m_{\text{As}} / \left( V_{\text{D0}} [\text{As}]_{\text{D0}} \right) \quad (6)$$

where $[\text{As}]_\text{D}$ and $[\text{As}]_{\text{D0}}$ are the instantaneous and initial weight concentrations of arsenic in the diluting compartment, while $V_\text{D}$ and $V_{\text{D0}}$ are the instantaneous and initial volumes in compartment D.

Table 2 summarizes all the input and output variables ($t$, $\kappa_\text{D}$, $\kappa_\text{C}$, DD, $\text{pH}_\text{D}$, $V_\text{D}$, $W$, $w$, [As], $\rho_{\text{As}}$, $[\text{Cl}^-]$, and $[\text{Na}^+]$) and shows that all water samples reduced their initial electric conductivity by about 80% in less than 13 min. Similarly, the arsenic removal ($\rho_{\text{As}}$), overall electric energy consumed ($W$), and specific energy consumption ($w$) varied from about 56 to 60%, 0.157 to 0.170 W h, and 71.6 to 75.0 W h $\text{m}^{-3}$, respectively.

By operating at $I = 0.05$ A with the PP water sample, it was possible to almost achieve the aforementioned values of $\rho_{\text{As}}$ and DD in a longer processing time (35 min).

Fig. 6 relates the percentage removal of arsenic ($\rho_{\text{As}}$) to the reduction in the dilute electric conductivity (DD) for the three water samples under testing. It can be noted that all data fall below the identity function ($\rho_{\text{As}} = \text{DD}$) and, despite a certain dispersion degree, they were fitted by using the least squares method as:

$$\rho_{\text{As}} = (0.71 \pm 0.02)\text{DD} \quad \left( r^2 = 0.986 \right) \quad (7)$$

Thus, the arsenic removal by ED is not a selective process, being smaller than that of the other electrolytes present in the water samples tested. In fact, it is about 71% of the DD achieved. For instance, the ED treatment of the PC water sample yielded a reduction in the electric conductivity of 80.3%, while the percentage removal of As was just 57.8%. Such results appeared to be independent of the electric current used (Table 2).

Fig. 7 compares $w$ to DD for the water samples examined. Even in this case, the least squares method yielded the following:

$$w = (0.81 \pm 0.02)\text{DD} \quad \left( r^2 = 0.985 \right) \quad (8)$$

By setting $I = 0.1$ A, the energy consumed per unit water volume ($w$) tended to increase about linearly with DD. Actually, the energy consumed is proportional to $I^2$ times the electric resistance of the ED stack. The latter, in turn, is controlled by the electric resistance of the dilute (D), that is the water sample undergoing desalination. According to the second Ohm's law, the electric resistance of D is inversely proportional to $\kappa_\text{D}$. As long as the variation in $\kappa_\text{D}$ was small, the voltage applied to the ED stack appeared to increase linearly with time (Fig. 4(a)) and the energy consumption resulted to vary about linearly with DD, as shown in Fig. 6. This even held true for the specific energy consumed at a lower electric current ($I = 0.05$ A) for the PP water sample (see open symbols in Fig. 7).

For the TO sample assayed, the arsenic content was reduced from 22.5 to 9.7 μg $\text{L}^{-1}$ (that is, slightly below the aforementioned MCL for As) by consuming 73.8 W h $\text{m}^{-3}$ (Table 2). Actually, such a value accounts for the contributions of the thermodynamic potential and overpotential of electrodes ($E_{\text{el}}I$) and ohmic resistance of the electrode rinsing solution ($R_{\text{ERS}}I^2$). These contributions are negligible in the industrial-scale ED stacks, that may be composed of 500–1,000 cells. In contrast, in a laboratory-scale ED stack the ohmic resistance of the electrode rinsing solution ($R_{\text{ERS}}$) may represent more than 50% of the overall resistance of the ED stack [21], thus making the above values overestimated.

By referring to the current treatment technologies for As removal, their specific electric energy consumption ranges from as low as 16–30 W h $\text{m}^{-3}$ in the case of As adsorption on iron ores or other materials coated with iron oxide, as well as titanium dioxide-loaded resins, to as high as 500 W h $\text{m}^{-3}$ in the case of RO [16]. Such a specific energy consumption is even less than that (810–1,090 W h $\text{m}^{-3)}$ measured in the case of brackish groundwater desalting by RO [38]. Thus, arsenic removal by ED resulted to be definitively less energy intensive than that by RO. In fact, with water salinity up to 5 g $\text{L}^{-1}$ ED is generally regarded as the most economic desalination process [39,40], while at total dissolved solid contents higher than 12 kg $\text{m}^{-3}$ RO is more profitable than conventional ED units as far as power consumption is accounted for [41]. Moreover, the specific energy consumption and overall operating costs to remove nitrate from drinking water were of the order of 89 W h and 0.66 € per $\text{m}^3$ of treated water, respectively [42].

Desalinated water costs are very sensitive to feedwater composition, as well as the plant type and capacity. For instance, the overall costs (capital, energy, and maintenance) of desalinating brackish water (2–5 g $\text{L}^{-1}$ of total dissolved solids) range from 0.36 to 0.25$ $\text{m}^{-3}$ by RO and from 0.35 to 0.19$ $\text{m}^{-3}$ by ED at plant capacities of about 4,000 and 100,000 $\text{m}^3\ \text{d}^{-1}$, respectively [43]. Thus, similar to brackish water desalting, As removal by ED also seems to be a method more economic than RO.

Table 2

Overall performance of the ED desalting tests carried out in this work at different process times (*t*) and constant electric currents (*I*) with the main characteristics (electric conductivity, $\kappa_D$ and $\kappa_C$, in tanks C and D; percentage reduction in the dilute electric conductivity, DDS; pH, $pH_D$, and volume, $V_D$, of desalted water; energy consumed, *W*; specific energy consumed, *w*; total arsenic concentration, [As]; percentage arsenic removal, $\rho_{As}$; sodium, $[Na^+]$ and chloride $[Cl^-]$ concentrations) of the water samples under study

| Sample identification | *I* (A) | *t* (min) | $\kappa_D$ (mS $m^{-1}$) | $\kappa_C$ (S $m^{-1}$) | DDS (%) | $pH_D$ (–) | $V_D$ (L) | *W* (W h) | *w* (W h $m^{-3}$) | [As] (μg $L^{-1}$) | $\rho_{As}$ (%) | $[Cl^-]$ (mg $L^{-1}$) | $[Na^+]$ (mg $L^{-1}$) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| TO | 0.1 | 0 | 19.95 | 0.71 | 0.0 | 7.3 | 2.28 | 0.000 | 0 | 22.5 | 0.0 | 22.5 | 18.9 |
| | | 2 | 14.88 | 0.72 | 25.4 | 7.5 | 2.28 | 0.027 | 12 | 19.8 | 12.0 | 22.7 | 21.1 |
| | | 4.2 | 10.44 | 0.72 | 47.7 | 6.9 | 2.28 | 0.063 | 27.7 | 16.5 | 26.8 | 25.4 | 23.4 |
| | | 7.2 | 4.82 | 0.72 | 75.8 | 6.6 | 2.28 | 0.128 | 56.3 | 11.8 | 47.4 | 15.9 | 16.4 |
| | | 8.4 | 3.10 | 0.73 | 84.5 | 6.3 | 2.28 | 0.168 | 73.8 | 9.7 | 56.9 | 12.3 | 13 |
| PP | 0.05 | 0 | 33.5 | 0.69 | 0.0 | 7.77 | 2.33 | 0.000 | 0 | 40.7 | 0.0 | 60.0 | 43.6 |
| | | 6.3 | 28.0 | 0.69 | 16.4 | 7.88 | 2.30 | 0.024 | 10.2 | 36.2 | 11.8 | 51.4 | 65.0 |
| | | 17 | 20.6 | 0.70 | 38.5 | 7.81 | 2.28 | 0.068 | 30.0 | 29.9 | 27.9 | 47.2 | 47.9 |
| | | 24 | 15.3 | 0.71 | 54.3 | 7.66 | 2.22 | 0.099 | 44.9 | 25.8 | 39.6 | 40.8 | 40.9 |
| | | 35 | 6.8 | 0.72 | 79.8 | 7.46 | 2.22 | 0.156 | 71.0 | 16.9 | 60.8 | 28.8 | 31.8 |
| | 0.1 | 0 | 34.0 | 0.720 | 0.0 | 8.40 | 2.33 | 0 | 0.000 | 47.4 | 0.0 | 60.0 | 43.6 |
| | | 3 | 27.5 | 0.721 | 19.1 | 8.34 | 2.33 | 0.03 | 0.013 | 40.3 | 15.1 | 51.47 | 54.3 |
| | | 8 | 15.9 | 0.729 | 53.2 | 8.2 | 2.33 | 0.09 | 0.037 | 32.3 | 31.9 | 24.51 | 29.7 |
| | | 13 | 4.9 | 0.738 | 85.7 | 8.32 | 2.33 | 0.17 | 0.075 | 19.2 | 59.5 | 5.77 | 11.7 |
| PC | 0.1 | 0 | 33.0 | 0.732 | 0.0 | 7.5 | 2.22 | 0.000 | 0 | 29.5 | 0.0 | 74.2 | 62.9 |
| | | 2 | 28.7 | 0.736 | 13.0 | 7.5 | 2.22 | 0.018 | 7.9 | 23.6 | 19.9 | 67.0 | 59.5 |
| | | 6 | 20.2 | 0.750 | 38.8 | 7.4 | 2.19 | 0.059 | 25.9 | 21.3 | 28.7 | 43.7 | 41.3 |
| | | 8 | 16.4 | 0.738 | 50.5 | 7.5 | 2.16 | 0.083 | 36.8 | 19.2 | 37.0 | 31.6 | 35.6 |
| | | 12.3 | 6.5 | 0.763 | 80.3 | 6.2 | 2.12 | 0.157 | 71.6 | 13.0 | 57.8 | 8.2 | 12.5 |

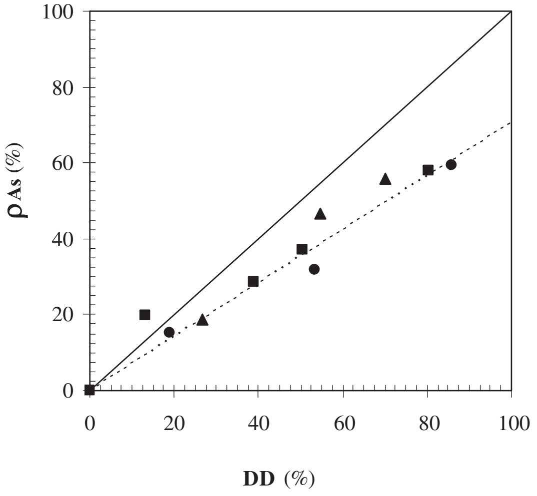

Fig. 6. Percentage removal of arsenic ($\rho_{As}$) against the reduction in the dilute electric conductivity (DD) for the three water samples under testing (PC: ■; PP: ●; TO: ▲). The continuous line refers to the identity function ($\rho_{As}$ = DD), while the broken one was calculated by using Eq. (7).

As concerning the waste management requirements, they are similar to those of RO and ion-exchange processing. Nevertheless, the burden of wastewater disposal in ED systems should be insignificant for the high water recovery. Generally, in nitrate removal by ED units the nitrate level in the concentrate is about 10 times greater than that in the incoming water [42]. For the water samples submitted to ED here (Table 1), the concentrating stream to be disposed of would contain less than 0.5 mg $L^{-1}$ of As. According to the Italian regulation [44], the residues from arsenic remediation of drinking water are to be coded as CER 19 09 00 without asterisk, this meaning that they are non-hazardous wastewaters unless the pollutant concentration is ≥3% (w/v) [45]. Therefore, the concentrate from an ED unit can be disposed of either in water basins and sewers or at landfills provided that its arsenic content is lower than 0.5 or 0.05 mg $L^{-1}$, respectively (see Tables 3 and 5 of Annex V—Section III of the aforementioned Legislative Decree [44]).

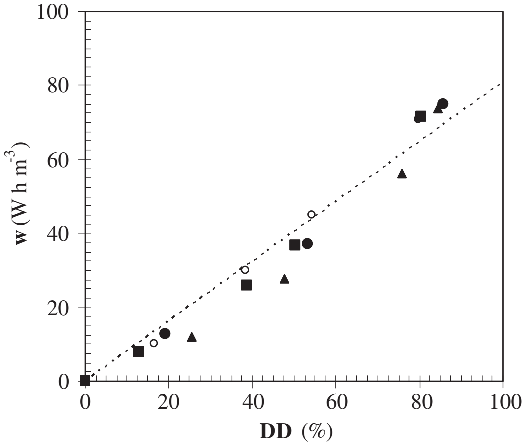

Fig. 7. Specific energy consumed ($w$) against the DD for the three water samples submitted to ED desalting at 20˚C and $I$ = 0.1 A (PC: ■; PP: ●; TO: ▲). The open symbols (○) refer to ED desalting of the PP water sample at $I$ = 0.05 A. The broken line was calculated by using Eq. (8).

In the circumstances, the concentrate represents a loss of potentially useful water, its quantity depending on the original arsenic concentration in feed water. Provided that the latter varies from 20 to 50 μg $L^{-1}$, the theoretical water wasted is expected to vary from 2 to 8% of the feed water volume.

Finally, at the end of their life cycle the electromembranes might be disposed of as non-hazardous solid wastes on condition that the elution test performed with distilled water led to eluates containing less than 2.5 mg $L^{-1}$ of As [46].

In conclusion, the experimental procedure described in this work allowed two empirical relationships of $\rho_{As}$ and $w$ against DD to be developed. These might be used to assess roughly the technical feasibility of the ED removal of As from the drinking water of concern. Once estimated the As removal needed to assure a final As level lower than MCL, Eqs. (7) and (8) enable the corresponding DD and specific electric energy consumption ($w$) to be in sequence estimated.

## 4. Conclusions

Batch desalting of a few well waters of the Cimino-Vico volcanic areas in Central Italy by ED was found to be technically feasible, the arsenate form As (V) being the most prevailing one. Thus, no oxidizing agent (i.e. ferric chloride) had to be injected into feed waters. For well waters tested, arsenic removal by ED was not a selective process, being about 71% of the DD achieved. Moreover, the specific energy consumption ($w$) was found to be linearly related to the DD. Contrary to the generally accepted belief, As removal by ED resulted to be by far less energy intensive than that by RO, its specific energy consumption (i.e. 71–75 W h $m^{-3}$) being of the same order of magnitude as that associated with the ED removal of nitrates from drinking water.

## Nomenclature

[As] — weight concentration of arsenic ($\mu$g L$^{-1}$)
$a$ — empirical coefficient, as defined by Eq. (1) (mS m$^{-1}$ L mg$^{-1}$)
C — concentrate
[Cl$^{-}$] — weight concentration of chloride ions (mg L$^{-1}$)
D — dilute
DD — desalination degree, as defined by Eq. (2)
$E$ — voltage applied to the ED electrodes (V)
ED — electrodialysis
EDR — electrodialysis reversal
$E_{\mathrm{el}}$ — thermodynamic potential and overpotential of electrodes (V)
EPA — environmental protection agency
ERS — electrode rinsing solution
$I$ — electric current (A)
$I_{\mathrm{lim}}$ — limiting current (A)
MCL — maximum contaminant level
[Na$^{+}$] — weight concentration of sodium ions (mg L$^{-1}$)
PC — As-rich water samples collected at *Ponte di Cetti* well (Viterbo, Italy)
PET — polyethylene-terephthalate
PP — As-rich water samples collected at *Piscin di Polvere* well (Viterbo, Italy)
$r^2$ — coefficient of determination
$R_{\mathrm{ERS}}$ — ohmic resistance of the electrode rinsing solution ($\Omega$)
RO — reverse osmosis
$t$ — process time (s or min)
$t'$ — dummy variable of integration (s or h)
TIC — total ionic concentration (mg L$^{-1}$)
TO — As-rich water samples collected at *Tobia* well (Viterbo, Italy)
$V_{\mathrm{k}}$ — volume solution in the generic $k$-th tank (L or m$^3$)
$W$ — electric energy consumed, as defined by Eq. (3) (W h)
$w$ — specific electric energy consumed per unit volume of water treated, as defined by Eq. (4) (W h m$^{-3}$)

*Greek symbols*

$\Delta m_{\mathrm{As}}$ — net variation in the mass of arsenic in tank D (g)
$\kappa$ — electric conductivity (S m$^{-1}$)
$\rho_{\mathrm{A}}$ — percentage removal of arsenic, as defined by Eq. (6)

*Subscripts*

0 — initial
C — referred to the concentrate
D — referred to the dilute
ERS — referred to the electrode rinsing solution

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