Electrified Technologies for Physical Separation of Arsenic from Water

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Abstract

The presence of arsenic (As) in groundwater limited their use due to the high risk to human health. Electrified technologies (electrocoagulation, electrodialysis, and electrosorption) for physical As separation have been presented as a decentralized alternative. However, these technologies should overcome limitations and risks to be more feasible and competitive with current to arsenic (As) removal treatments. Herein, an electrified system that involves electrochemical sensors coupled with electrochemical treatment of As to ensure safe drinking water is presented as a feasible system. Electrified technologies are described in terms of their principles, limitations, and technology readiness level classification. Furthermore, the difference between the As concentration in the environment and the concentration used in electrified technologies is discussed.

1. Introduction

The United Nations (UN) defined sustainable development goals (SDG) as a blueprint for peace and prosperity. SDG number six aims to achieve universal coverage of essential drinking water by 2030 [1]. To reach this holistic objective, tapping into groundwater sources will be crucial. However, these vital hydric sources cannot be used raw and will require the implementation of decentralized treatments to enable their exploitation as off-grid alternatives. Contamination with arsenic (As) is a common trait of many water wells. It has been flagged as a significant health issue in many communities in South Asia (e.g., India, Bangladesh, etc.) [2–4] and South America (e.g., Mexico, Chile, and Argentina) [5–8]. Arsenic species are highly toxic and have demonstrated carcinogenicity. Thus, the World Health Organization (WHO) established a maximum contaminant level (MCL) of arsenic as low as 10 $\mu g\ L^{-1}$ [9].

As is a challenging pollutant that cannot be degraded or transformed into an innocuous species. Therefore, it can only be removed by physical separation methods. Off-grid technologies disconnected from the centralized water distribution grids, and sometimes the electrical grid, will be needed to decontaminate water sources by removing As. Processes such as adsorption, ion exchange, or chemical precipitation have been proven effective [10–15]. More recently, electrified approaches are gaining more relevance due to their modularity and high adaptability as scaled-down systems that are easy to operate by end-users [16,17]. Electrochemical technologies can provide the opportunity to implement electrochemical monitoring for data collection and analysis [18–22]. Coupled electrochemical removal systems with an electrochemical As sensor can provide quick information that is easy to understand by the user, such as red light (concentrations above MCL) and green light (concentrations below MCL), as depicted in Figure 1. This modular coupled system can activate treatment if required and ensure that treatment effectively removes arsenic levels below the MCL threshold. Several electrodes have proven electrochemical detection of arsenic at different concentrations levels above and below the MCL in natural water matrixes. Compared to colorimetric methods or induced coupled plasma mass spectroscopy (ICP-MS), electrochemical sensing is a reliable enough, cheaper, and faster way to make field decisions [23].

Herein, we describe opportunities for electrified arsenic removal systems, including their work principles, benefits, drawbacks, and technology readiness level (TRL).

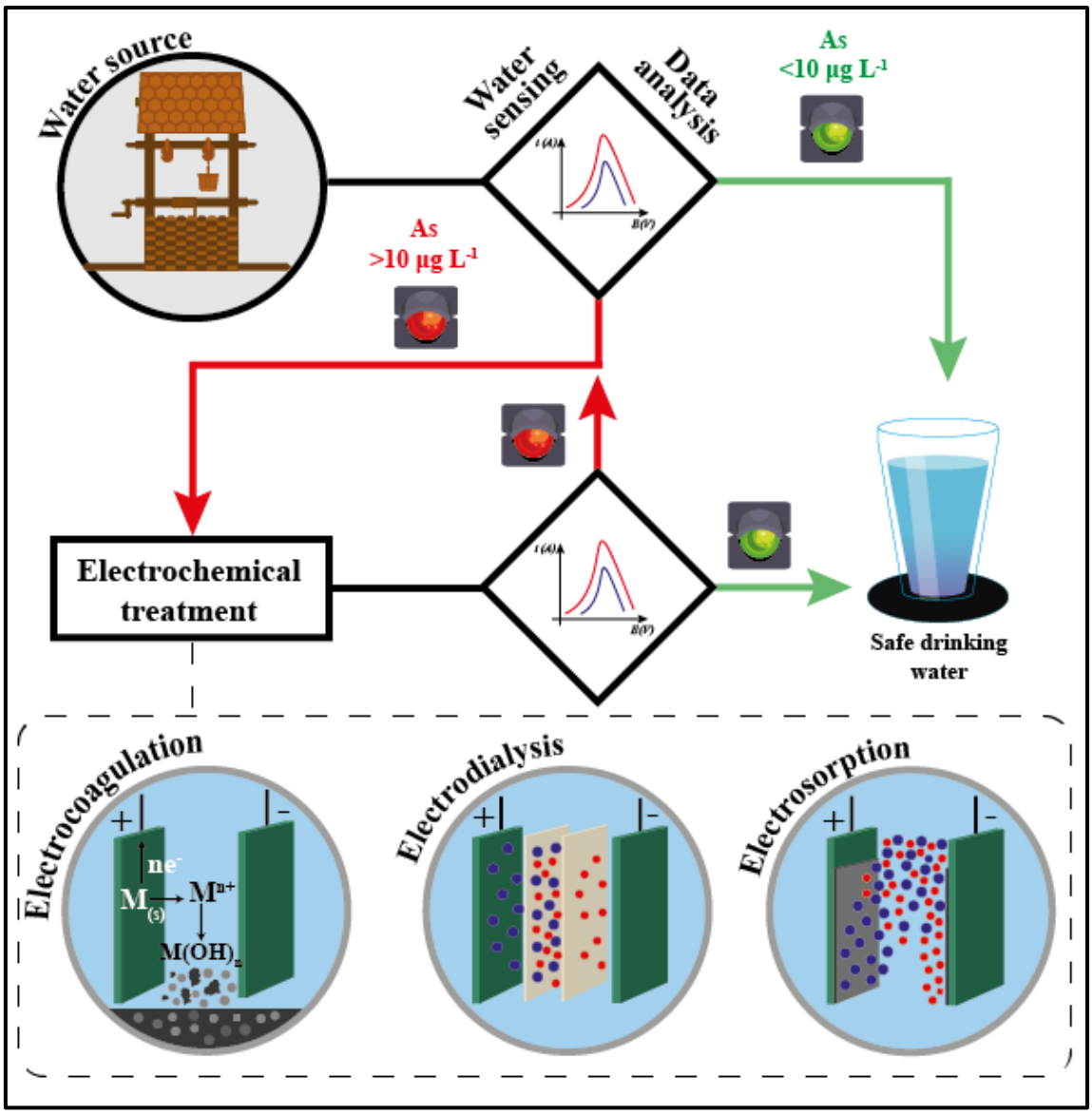

Figure 1. Schematic of a coupled electrochemical system using electrochemical sensors and data management to decide actions about water treatment to reach safe drinking water.

2. Electrified systems for arsenic removal

As depicted in Figure 1, among electrified methods for arsenic removal, the technologies considered to be competitive are electrocoagulation, electrodialysis, and electrosorption. This section will discuss these three electrified technologies in terms of their principles, benefits, and challenges.

2.1 Electrocoagulation

Electrocoagulation (EC) is based on the generation of coagulants in-situ by the electro-dissolution of sacrificial anodes. Generally, iron (Fe) and aluminum (Al) are used as anodes due to the yield of Fe(II), Fe(III), and Al(III) species from the oxidation of the bulk electrode. These soluble ionic species precipitate as hydroxides and poly hydroxides, which act as coagulants that remove undesired pollutants from the solution. The hydroxide anions are produced at the cathode from the water reduction reaction, which provides a self-buffering capacity to the EC treatment around pH values ranging between 6-8. The metal hydroxide coagulants can then remove pollutants in water by following different mechanisms, such as pollutant entrapment, charge neutralization, adsorption, and complexation [24,25]. Iron-based sacrificial electrodes are the preferred choice, given their outstanding removals (~ 99 %) compared to Al electrodes (~95 %) [26]. Taking a closer look at As speciation in the function of pH, it can be deduced that at pH 6-8, As(III) is present as H_3AsO_3, and As(V) is present as $H_2AsO_4^-$ and $HAsO_4^{2-}$. These As oxyanions have a high complexation affinity with iron oxides and iron hydroxides but not with aluminum hydroxides. Thus, the selective complexation of As species on iron coagula explains the higher removals, given the predominant role of adsorption removal mechanisms [27,28].

The engineering design of adaptable EC reactors has been a fruitful line of research. Moving from simple batch tests to larger treatment systems elucidates some final user challenges associated with the sacrificial character of the electrodes, which turns them into consumables that should be “easily” replaced (10.1016/j.watres.2021.117595). Recent attractive designs consider the transition from bulk two-dimensional plate electrodes towards cartridges containing balls and pellets as three-dimensional electrodes [29–31]. These appear as an intelligent replenishable alternative that will facilitate the user using scrap metals and recycling other waste (e.g., soda cans, etc.). However, sustained performance studies that explore issues such as electrode passivation risks or continuous flow treatment at meaningful water flow rates should still be considered to advance EC technology.

The use of EC proves extremely efficient on As abatement but presents a significant challenge for decentralized treatment units that it is common to those of conventional treatment processes such as coagulation or adsorption: the disposal of an As-laden solid waste (i.e., sludge). Safe

handling of hazardous solids and the actions to manage this waste is pervasive since incorrect disposal of the waste will result in undesired leaching of As back to water sources. This is a sanitation challenge with no trivial solution, and it needs to be addressed in the literature that focuses mainly on the removal of As from water. More attention should be devoted to the whole process, including sludge removal and management after treatment [32,33].

2.2 Electrodialysis

Electrodialysis (ED) is a membrane separation process in which anions and cations in solution are separated under electrical potential as a driving force (10.1016/j.chemosphere.2020.126363). Positive charges migrate toward the negatively charged electrode, and negative charges migrate toward the positive charge electrode. Selective separation is allowed by different specific ionic exchange membranes (anionic and cationic membranes). Therefore, the separation of contaminants depends on their concentration and the specific properties of the membrane [34,35]. Two types of effluents are obtained from the treatment: the treated water and an As concentrated effluent. Treated water should be tested to ensure that pollutant concentration is below the MCL. ED has demonstrated a high As removal performance of 50 to 99% in single-pass treatment [36,37]. However, most studies evaluated treatment at concentrations several orders of magnitude higher than those found in the environment (see the whisker plot of Figure 2). Concentration gradient may impact ionic transport and, in some instances, overestimate removal capabilities. Therefore, further assessment at realistic concentrations would be recommended to ensure sufficient removal capacities are achieved.

The use of ED seems an emerging alternative that, despite the complexity of the ion-selective membrane arrangement, can achieve effective As removal. However, the main challenge remains the disposal of concentrated effluent. Releasing concentrated effluents may backfire by causing undesired environmental impacts associated with increasing As concentrations. Different alternatives have been proposed to manage concentrated effluents, such as deep well injection, evaporations ponds, and conventional crystallizers to protect environmental health [38,39]. The management of concentrated effluent still needs to be solved by researchers and engineers. The disposal of the concentrated effluent significantly impacts the techno-economics of the process

representing up to 30% of the total treatment cost [40].

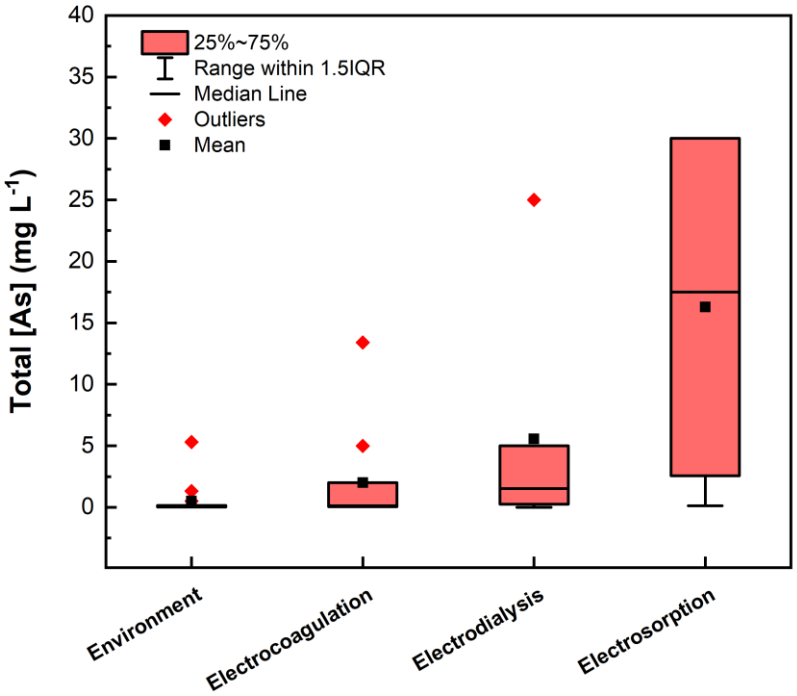

Figure 2. Whisker plot of different concentrations reported in environment [41–43], electrocoagulation [44–51], electrodialysis [36,37,52–54], and electrosorption [55–57] studies.

2.3 Electrosorption

Electrosorption (ES) are separation processes in which charged pollutants are adsorbed on the surface of porous electrodes under applied potential given electrostatic forces. Capacitive deionization (CDI) is the most studied electrosorption process for environmental remediation. The CDI process operates intermittently by charge-discharge cycles. Charge species are removed from the aqueous phase by forming an electrical double layer (EDL) during the charge step. Meanwhile, the electrode surface regenerates during the discharge step, releasing the charged species adsorbed as part of their EDL. Despite the most studied process being related to water desalination, the selective removal of contaminants has also been reported [58,59]. The main challenge of this process is the need for electrode specificity and capacity of adsorption/desorption during the charge/discharge steps, respectively.

In the case of As removal by CDI, the rational design of modified activated carbon or other high surface area is required to improve the attachment of As species. It is important to remark

that electrostatic forces are the main driver for adsorption; consequently, it has been observed higher removals for negatively charged As(V) species (usually found as $H_2AsO_4^-$ and $HAsO_4^{2-}$) than neutral As(III) (usually seen as H_3AsO_3) [55,60,61]. Some interesting approaches aim to oxidize As(III) at the applied potential to enhance electrosorption. However, other electrode design strategies may aim to include other interfaces prone to form complexes with As(III) (e.g., MnO_2, Fe/FeO/Fe_2O_3, TiO_2). Understanding how these modifications affect the charge/discharge cycles should be clarified further. The main challenge of CDI systems is associated with the As concentrated effluents released during the discharge cycle (similar to that of ED). Managing water effluents in the intermittent system (safe water / As rich water) may also become an engineering challenge that may result in water recovery losses to ensure further protection for the final user.

3. Assessing technology readiness level of electrified technologies for arsenic removal

Research and engineering development guide technology advance to solve challenges and cover specific needs. When considering the environmental and health issues associated with As pollution in terms of safe and drinking water supply, we have to critically assess the maturity of technologies to evaluate how close they are to be commercially implemented. We can classify the status of technology based on “Technology Readiness Levels” (TRL) divided into nine stages, as summarized in Figure 3a. In the research stage (TRL 1, 2, 3), fundamental and proof of concept studies are considered to evaluate the possible application of the approach. Development (TRL 4, 5, 6) and deployment (TRL 7, 8, 9) stages require the intervention of external factors such as the government, industries, and start-ups [62]. The successful advancement to stages closer to commercialization require techno-economic analysis (TEA) to validate the cost competitiveness of proposed processes [63,64]. Conducting more TEA for electrified processes appears as a need to understand further essential aspects to design effective business plans and identify niche applications such as removing As from water sources. In this framework, studies should be able to provide engineering figures of merit to benchmark them with current alternatives for the same application.

Reverse osmosis (RO) is an example of membrane-based technology that transitioned to TRL9 with several commercial options available. Meanwhile, electrified treatments such as

electrodialysis and electrosorption are classified between TRL 4-6. In this framework, electrocoagulation has already been scaled into competitive reactor designs and is transitioning into deployment stages (TRL 5-7) with some existing examples of early implementation. However, research efforts are still needed to clarify fundamental questions that arise within the translation to higher TRLs, feeding a closed loop that advances our understanding of these technologies. Figure 3b illustrates an increasing trend in electrocoagulation articles published during the last eight years to face As pollution challenges, while less attention is paid to other electrified processes. Overall, these trends and the technology stages suggest that electrocoagulation may emerge as the most competitive electrified option.

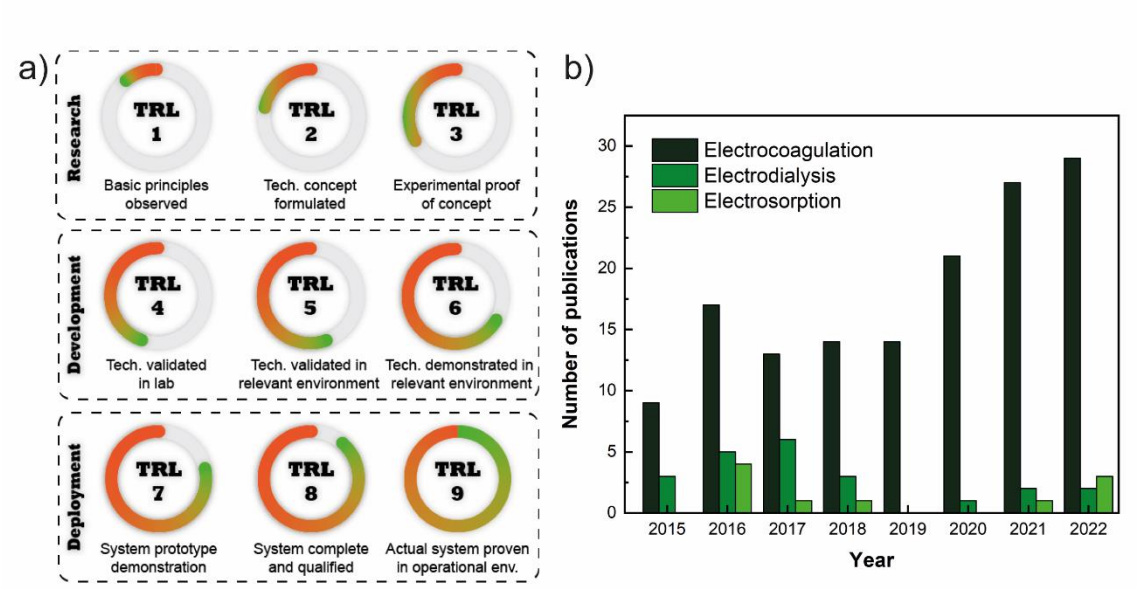

Figure 3. (a) TRL classification is divided into three groups of maturity: research, development, and deployment. (b) The number of publications related to arsenic removal using electrocoagulation, electrodialysis, and electrosorption. Results are obtained from the search of the following keywords in Scopus: “arsenic” AND “removal” AND “electrocoagulation, electrodialysis, electrosorption”.

3. Outlook and perspective

Electrified technologies rapidly emerge as translatable technologies to ensure clean and safe water access. All these electrified processes achieved more than 98% of removal from the initial arsenic content. The modularity and easy implementation of decentralized technology is the most

promising and competitive asset. Electrified technologies present unique modular capabilities and easy adaptability to many case scenarios. Research provides an understanding of system-level performance and increasing removal capabilities by advancing electrode and reactor design for electrocoagulation, electrodialysis, and electrosorption. Our critical assessment of technology competitiveness should consider the wide range of concentrations of arsenic reported. Environmental arsenic content in impacted regions presents concentrations ranging between 15 – 160 μg L^{-1} of As. Those As concentrations can rapidly bump up in certain areas affected by industrial and anthropogenic activities, reaching values from 0.5 to 5.0 mg L^{-1} (or 500 – 5000 μg L^{-1}) [41–43]. However, in many instances, such huge concentrations (even 500 times higher than the MCL) are still several orders of magnitude below concentrations used in lab tests (see Figure 2). Future research efforts should emulate real concentrations reported in order to accurately assess technical capabilities and minimize hype generated from overestimations. Benchmarking removal and associated costs at different levels will be essential to decide on technology implementation in each case. Using figures of merit and TEA will become key tools to guide engineering decisions for each case. Electrocoagulation is the technology that has been assessed at close to relevant concentrations, which is a need for higher TRL stages. Nevertheless, early-stage implementation of parametrized conditions closer to environmentally relevant situations will smooth and accelerate TRL transitions. In the case of arsenic removal technologies, careful attention will have to be paid to the resulting solid waste or concentrated effluents to avoid undesired unplanned environmental consequences.

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*10.1016/j.chemosphere.2020.126363 This review provides a thorough description of the principles and applications of electrified technologies for physical separations.