Mini Review

Potential Application of Membrane Capacitive Deionization for Heavy Metal Removal from Water: A Mini-Review

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Heavy metal pollution of the aquatic environment has become a global concern in recent decades, and conventional methods for heavy metal removal, like coagulation, precipitation, and membrane filtration, have their limitations. In this mini-review, we discuss four aspects of heavy metal removal by membrane capacitive deionization (MCDI): i) the role of electrode materials; ii) role of ion-exchange membrane; iii) operating conditions; and iv) water chemistry. Based on this discussion, we determine MCDI to have good potential for heavy metal removal in water. Future MCDI studies should focus on synthesizing ion-selective electrode to promote the removal of specific ionic species; developing multifunctional electrodes for the simultaneous removal of different heavy metals; performing pilot-scale studies instead of bench-scale ones; applying MCDI to raw water or real-life water samples and controlling MCDI with different current modes.

1. INTRODUCTION

In recent decades, the problem of heavy metal pollution has gradually become a global concern [1–6]. In fact, heavy metals are highly toxic, carcinogenic, mutagenic, and difficult to degrade; moreover, they easily accumulate and their properties are long-lasting [7–10]. In the natural environment, the mobility and stability of heavy metals are very strong. Therefore, once heavy metals

are released in the natural environment, they are likely to pollute water and the soil, and they may enter the human body through a range of mechanisms, causing incalculable harm to human health [11–16].

The main methods for removing heavy metals from water include coagulation, precipitation, resin-based ion exchange, reverse osmosis, nanofiltration, electro-flocculation, and electrodialysis [17–21]. In detail, coagulation and sedimentation require large and complex processing facilities, and the efficiency of the removal of trace pollutants is poor through these approaches. Furthermore, the chemicals added to perform the coagulation procedure and the sludge thus generated are likely to cause secondary pollution. The disintegration of ion-exchange resins fragments will cause an increase of particles in water and cause secondary pollution. The filters used in reverse osmosis and nanofiltration need high pressures to remain operational, and implementation of electrocoagulation and electrodialysis involves high monetary investments, high energy consumption, and some operational obstacles [22,23]. Therefore, treatment methods that do not require the injection of chemicals are highly efficient, involve low energy consumption, and produce no secondary pollution that needs to be explored [24,25].

Based on electric field-driven separation, capacitive deionization operated under low voltage conditions by applying an external electric field affords the "electrical adsorption" of contaminants to the equipment's cathode and anode. This technique is especially suitable for the removal of low-concentration dissolved ions [26–28]. In particular, when an electric field is generated between parallel electrodes, ions present in the contaminated water employed as the electrolyte migrate to the pores of the electrode structure to form an electric double layer. Notably, when the polarity of the electric field is cut-off or reversed, the absorbed ions are released back into the water to regenerate the active material for the next cycle. To prevent the discharge of contaminants back into the feed solution during desorption, an ion exchange membrane can be placed between the electrodes to enhance the recovery efficiency and reduce energy consumption. This approach, dubbed membrane capacitive deionization (MCDI; see Figure 1), has been the focus of much research attention [29,30].
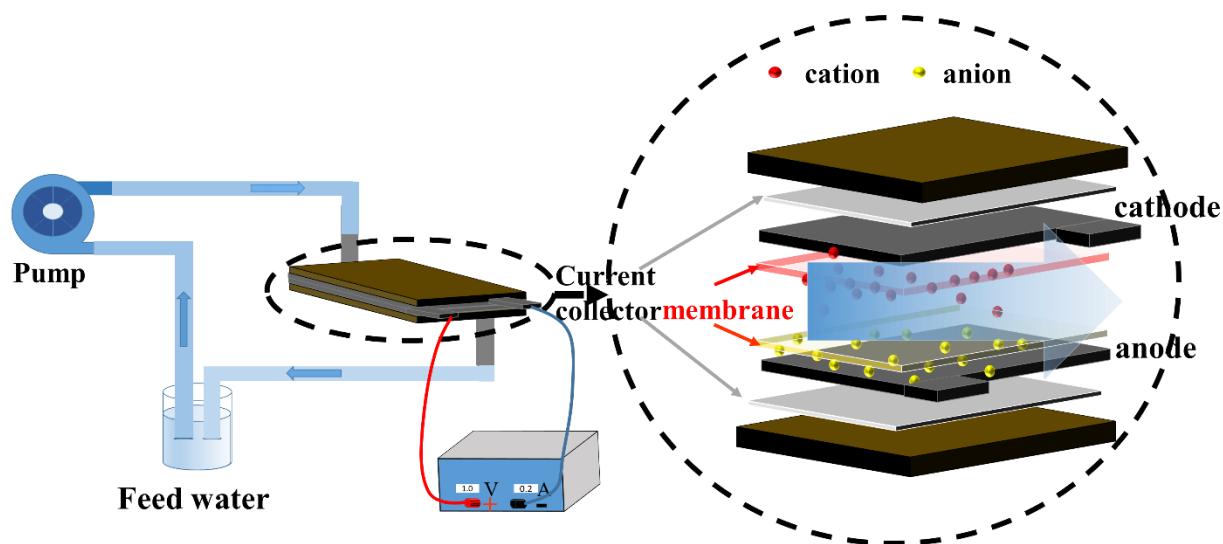

Figure 1. Membrane capacitive deionization setup for contaminant removal in flow-by mode. Reproduced from ref. [31] with permission from The Royal Society of Chemistry.

The use of MCDI can avoid disadvantages like the low efficiency of the coagulation and precipitation processes and the possibility that ion-exchange resins produce debris as a result of disintegration. In contrast to reverse osmosis, in MCDI high pressure is not required for contaminant separation and water purification; therefore, this method can effectively reduce membrane fouling, and its operation is associated with low unit treatment costs. MCDI has been applied for desalination, hard water softening, heavy metal and nutrient removal, and has the potential for resource and energy recovery [23,32,33]; its operation requires no chemicals and a low power investment; MCDI is also highly efficient and produces no secondary pollution, and it has shown good prospects for employment in heavy metal removal [34–36].

Although over the past few years MCDI has become a research hotspot in many fields [23,37], it remains in the exploratory stage as it relates to water treatment. Problems associated with electrode regeneration and performance degradation resulting from long-term use have hindered the application of MCDI to heavy metal remediation of water samples [38]. Therefore, studying the efficiency of MCDI may help solve the problem of this approach's long-term practical applications.

2. MAIN FACTORS AFFECTING MCDI EFFICIENCY

2.1. Role of electrode materials

The interest in electrode synthesis is surging among researchers in the fields of electrochemistry, chemical engineering, and environmental engineering. Since 2012, 536 studies on electrode synthesis have been published, with 54.5% of them focusing on the preparation of novel electrode materials to improve the adsorption capacity of the electrode [23,39,40].

The materials that make up the electrodes have a substantial influence on the electrodes' deactivation/regeneration performance [41]. Materials characterized by high specific surface area, multi-material composite, and high specific capacitance have good application potential [42–44]. Han [45] found that the surface area and microporosity of carbon cloth are important parameters; in particular, the higher the porosity, the higher efficiency of removing heavy metals; however, when the surface area is too large, the desorption time may be extended. Conventional adsorption treatment involves the use of arsenic-imprinted material that selectively removes As(V) [46]; if the imprinted material can be loaded onto the electrode, it allows the selective separation of contaminants as part of the MCDI operation. Liu *et al*. developed highly porous N-doped graphene nanosheets for the rapid removal of heavy metals from water by capacitive deionization; in particular, owing to the large surface area of the nanosheets, Pb^{2+} and Cd^{2+} ions could be simultaneously removed within 30 minutes [47]. Liu obtained porous carbon spheres *via* microwave-assisted synthesis that could be used for capacitive deionization and applied to ion removal [48]. Gaikwad removed Cr (VI) and the fluoride ions simultaneously. Hu prepared a manganese dioxide/carbon fiber electrode for the electrosorption removal of copper ions from water [49]. A summary of published studies on MCDI electrodes for heavy metal removal is presented in Table 1. The above evidence indicates that there will be a large market for multifunctional electrodes in the future [50].

Remarkably, we found that some of the studies just described did not employ the MCDI technique in real-life scenarios; we thus suggest that future studies should not focus only on the electrochemical analysis of the electrode properties, but they should also include investigations whereby the synthesized electrode is utilized to achieve desalination, heavy metal removal, or water softening [51–53]. Existing research reports are mostly bench-scale, and exploring the optimization mechanism of the efficiency of different types of electrodes in pilot-scale tests and on-site operations still needs more comprehensive exploration [54,55]. These new electrodes should be considered to be applied in real-life operations [56–59].

Most electrodes are not selective in ion removal. However, in many water pollution scenarios, not all ions present in solution need to be removed. In such a situation, some electrodes have the potential to selectively remove cations or anions; for example, Lee developed a carbon electrode that could selectively remove arsenic [60]. Selective electrodes will be of great importance, as their use will allow operational costs to be cut by avoiding the unnecessary removal of ions that are not harmful [61–63]. Therefore, the manufacture of ion-selective electrodes should be emphasized in future studies.

Table 1. Published studies on membrane capacitive deionization electrodes used for heavy metal removal

Heavy metal species	Electrodes material	Removal rate (%)	Ref.
Arsenic	Carbon electrode as the anode and cathode	80	[64]
Arsenic	Porous activated carbon electrode	99.0	[26]
Chromium (VI)	MOFs MIL-53(Fe) as the anode, activated carbons as the cathode	77.2	[65]
Chromium (VI)	Tea waste biomass activated carbon as the anode and cathode	88.5	[66]
Lead	Activated carbon electrode as the anode and cathode	65	[67]
Lead	Activated carbon cloth as the anode and cathode	81	[68]
Lead	N-doped graphene nanosheets as the anode and cathode	99	[47]
Cadmium	Activated carbon cloth as the anode and cathode	42	[68]
Copper	Mg/Fe layered double hydroxide loaded with Magnetic(Fe_3O_4) carbon spheres as the anode and cathode	40	[69]
Iron	Graphene and resol (RGO–RF) nanocomposite as the anode and cathode	43.8	[70]

2.2. Governing operating factors affecting electrode efficiency

In addition to electrode materials, a lot of studies have focused on pollutant adsorption characteristics and kinetics at the electrode as part of MCDI operations [71]. The mechanism of ion desorption during electrode regeneration is often overlooked, yet understanding the mechanisms of desorption and separation are crucially important to solving practical problems.

Voltage, current density, flowrate, and charge-discharge control modes are the key control factors for electrode deactivation/regeneration [72–74]. When the water flow rate in the inlet is low, the effectiveness of pollutant removal is high; however, a low value for the flowrate will also mean long hydraulic retention time, so finding the appropriate treatment flowrate for different water qualities is necessary. Conventional electrodes are regenerated *via* power cut-off, but this approach has two drawbacks: (1) the desorption rate is slow, so electrode regeneration takes a long time; (2) the desorption efficiency is low, so the pollutant removal efficiency gradually decreases as the running time is extended. If we can learn from the studies on pulse regulation that have been published in recent years in the field of electrochemistry to adjust the pulse duty cycle, the pulse mode (alternative pulsed current shown in Figure 2) is expected to control the multi-step electron transfer process and achieve the rapid and efficient desorption of contaminating pollutants [75]. The directional conversion separation is helpful for the long-term operational efficiency of MCDI, and it increases energy recovery.
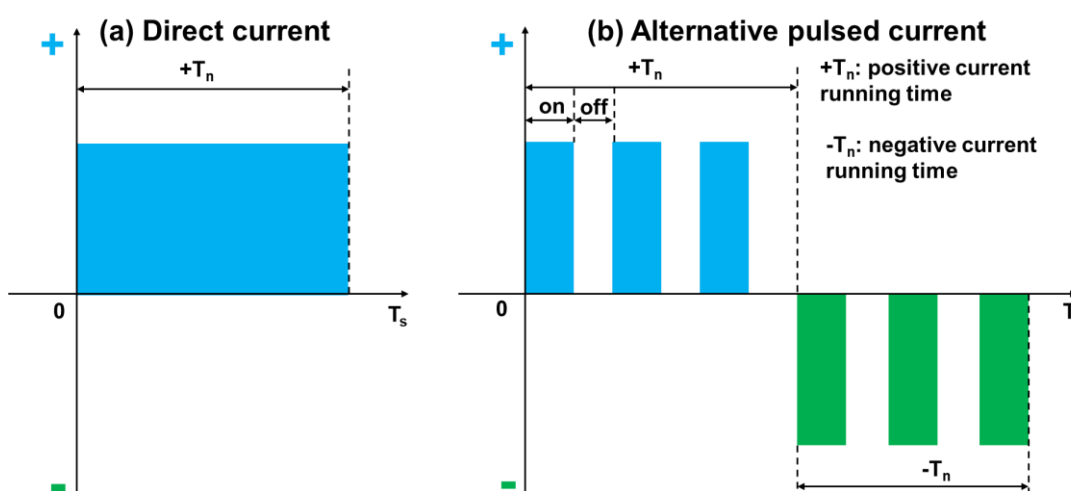

Figure 2. Potential current mode for membrane capacitive deionization control. Reproduced from ref. [76] with permission from Elsevier.

From the perspective of the mechanism of the reactor, the types of water flow (flow-through mode *versus* flow-by mode) will affect MCDI efficiency during operation *via* the effect on mass transfer and reaction rate. Peng [77] found that flows into the anode-cathode effluent system performance will be better than the cathode–anode effluent system.
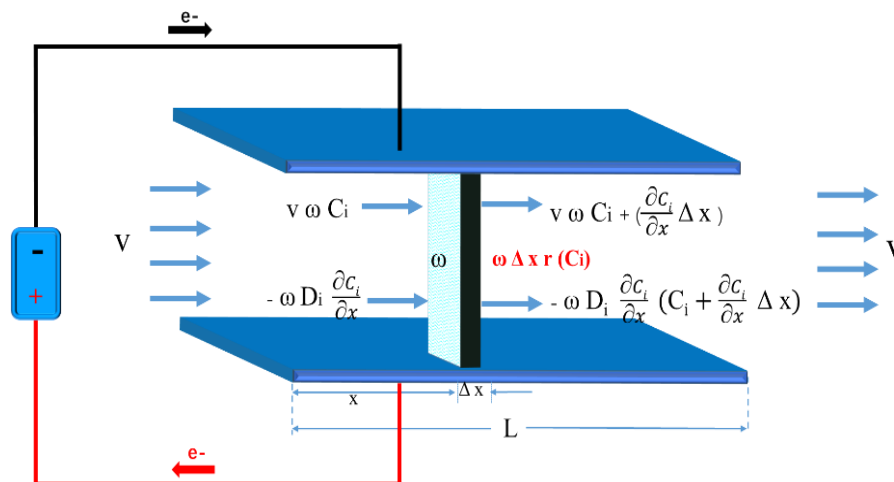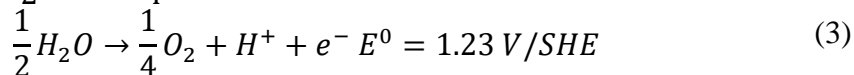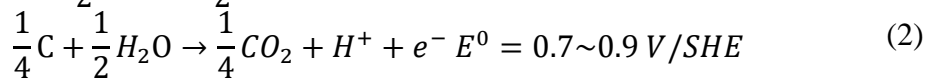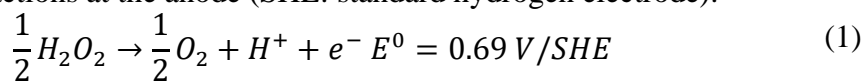
Figure 3. Dispersion model for the mass transfer of pollutants passing through the membrane capacitive deionization system.
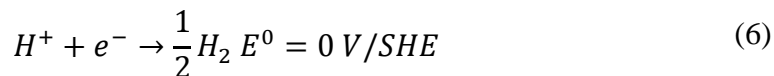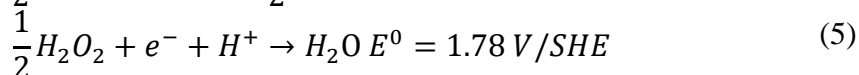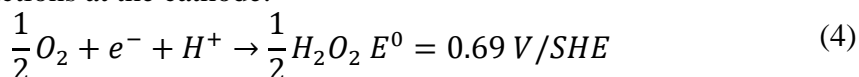
The dispersion model in water quality engineering has not been used in the study of electrode deactivation/regeneration kinetics. If the dispersion model can be applied to the simulation of the evolution of kinetic characteristics (Figure 3), the time required to predict electrode deactivation/regeneration can be shortened; on the other hand, benefits are associated with optimizing the hydraulic retention time and implementing MCDI reactor temporal control in industrial applications.

Voltage and current values are also important factors affecting MCDI heavy metal removal effectiveness [78]; although results from studies have indicated the maximum voltage for the effective operation to be ~1.6 V, Tang was able to achieve a satisfactory removal rate in the context of desalination at a potential of 2.4 V [79]. The occurrence of faradaic reactions is also linked to the voltage value [29,79–82]. Chen found that increasing the voltage promotes faradaic reactions, increases ion removal efficiency, and reduces the system's energy consumption [83].

Oxidation reactions at the anode (SHE: standard hydrogen electrode):

$$\frac{1}{2}H_2O_2 \rightarrow \frac{1}{2}O_2 + H^+ + e^- \quad E^0 = 0.69 \, V/SHE \tag{1}$$

$$\frac{1}{4}C + \frac{1}{2}H_2O \rightarrow \frac{1}{4}CO_2 + H^+ + e^- \quad E^0 = 0.7{\sim}0.9 \, V/SHE \tag{2}$$

$$\frac{1}{2}H_2O \rightarrow \frac{1}{4}O_2 + H^+ + e^- \quad E^0 = 1.23 \, V/SHE \tag{3}$$

Reduction reactions at the cathode:

$$\frac{1}{2}O_2 + e^- + H^+ \rightarrow \frac{1}{2}H_2O_2 \quad E^0 = 0.69 \, V/SHE \tag{4}$$

$$\frac{1}{2}H_2O_2 + e^- + H^+ \rightarrow H_2O \quad E^0 = 1.78 \, V/SHE \tag{5}$$

$$H^+ + e^- \rightarrow \frac{1}{2}H_2 \quad E^0 = 0 \, V/SHE \tag{6}$$

A novel technique quite similar to MCDI but dubbed shock electrodialysis, which utilizes over-limiting current, is a quite promising method for heavy metal removal from aqueous phases that has been proposed by researchers from the Massachusetts Institute of Technology; results obtained by these scientists indicate that, if properly applied, over-limiting current can enhance heavy metal removal

efficiency. In the case whereby the energy demand and removal efficiency are well balanced [83–87], MCDI application to real-life scenarios may be accelerated [88–92].

2.3. Role of the ion-exchange membrane in MCDI

The existence of an ion-exchange membrane prolongs an MCDI electrode's activity [93]. Notably, the identity of the ion-exchange membrane influences the composition of the filtrate passing through the said membrane, which in turn affects removal efficiency [94–96]. Studies on heavy metal removal processes have focused mainly on the performance and membrane fouling specificity of conventional membrane materials [97]; many problems remain thus unsettled, such as (1), how to select suitable ion-exchange membranes for water samples of different quality; (2), the mass qualitative mechanism during the membrane filtration process; and (3), how the electrode characteristics change during the removal process. The issues just enumerated point also to key obstacles that must be cleared to solve practical problems and afford the industrial-scale utilization of MCDI.

The performance of the ion-exchange membrane will affect the electrochemical characteristics of the filtered water, which in turn indirectly affects the electrode deactivation/regeneration process. Published studies generally focus on the ions of different metallic elements, whereas more comprehensive studies are needed to determine the behavior of different ionic species of the same metallic element (e.g., As[III] *versus* As[V]) [98]. Dong developed an ion-exchange membrane affording the selective removal of lead ions [67]; the development of metal-selective membranes is expected to allow the selective removal of arsenic ions with a specific valence number [99–101]. In the scenario whereby selective ion-exchange membranes will be utilized that separate the ions of different metallic elements or different ionic forms of the same metallic element, the load on the electrode will be relieved. In the context of ion-exchange membrane materials characterized by high anti-pollution performance, a reduction in the extent of abrasion occurring during backwash will prolong the service life of the membrane [102].

Mass transfer during membrane filtration will directly affect the separation efficiency of heavy metal-contaminated water. The mass transfer afforded by ion-exchange membranes is affected by the membrane's interface structure and by the charge of the electrolyte, which indirectly affects the electrode performance. Pollution of an MCDI anion exchange membrane will produce a large internal resistance, which will negatively affect the electrochemical system's electricity generation performance. Tang found that the oxidative degradation of anion exchange membranes may result in maintenance cost increases [80]. The addition of cation exchange membranes alone can still prevent faradaic reactions taking place at the cathode. Additional investigations on the necessity of anion exchange membranes are needed [28,103]. In the process of ion permeation through the membrane, the Donnan effect and the concentration polarization will also affect the efficiency of pollutant separation and removal [94,104,105]. In the removal of pollutants by MCDI, the influence that the mechanism of the change of membrane flux has on the adsorption of pollutants onto the electrode remains to be explored. If heavy metal removal rate can be maintained constant under conditions of increasing water flux, the amount of generated clean water will increase.

2.4. Influence of water chemical composition and characteristics on MCDI performance

The chemical composition and characteristics of water are the core factors affecting pollutant removal efficiency. Published research on the removal of heavy metals by MCDI indicates that a single ion or component may have an impact on electrode deactivation/regeneration [106–109]. Fan showed that when NaCl concentration increases, anode-adsorbed chloride ions hinder electrode regeneration and decrease arsenic removal rate [22]. In addition to inorganic compounds, heavy metal complexation by dissolved organic matter widely present in drinking water sources will affect the extent of heavy metal electrochemical adsorption [110,111]. Taking humic acid in dissolved organic matter as an example, it is worth noting that humic acid can act both as an electron donor and an electron acceptor. As a result, the presence of electrode-adsorbed humic acid is likely to accelerate the electron transfer. The influence that humic acid protonation and the electron shuttle effect have on electrode inactivation, as a result of heavy metal complexation by the said acid, has not been reported.

Lin studied the heavy metal adsorption/desorption behavior of electrodes in the presence of both inorganic and organic substances using synthesized water [106]. In most published studies, laboratory water distribution has been utilized; however, water and wastewater usually comprise multiple coexisting components [68,112], so raw water samples containing different kinds of ions, macromolecules, and colloids, as well as hydration particles of different sizes, should be used to study the effect of complexation, chelation, adsorption, etc. In other words, the use of raw water samples in MCDI studies produces results of higher practical significance in terms of pinpointing the mechanism of electrode deactivation/regeneration.

Studying the effect that water chemistry has on electrode deactivation/regeneration will help further reveal the water quality conditions applicable to MCDI technology. Different electrical characteristics of the contaminant adsorbed onto the electrode will help increase contaminant separation efficiency [51]. Results of studies conducted by Wang [80,113] indicate that although usually high-valence ions are preferentially removed, under certain conditions monovalent ions like Na^+ will be removed before divalent ions like Mg^{2+}. This finding is closely related to the directional control of the separation and removal of ions of different valence. However, its cause and mechanism have not yet been explained. He [80,114] found that the amount of dissolved oxygen affects the selective removal of ions and H_2O_2 generation potential during electrode regeneration. The total hardness of groundwater in parts of the world is relatively high, and the main components of total hardness (Ca^{2+}, Mg^{2+}, etc.) can affect electrical transmitting. The faradaic reaction increases the H^+ electrical influence adsorption selectivity and pH changes [115,116]. In an oxidizing atmosphere, at pH values below 6.9, $HAsO_4^{2-}$ is the main species by contrast, in a reducing environment and at pH values below 9.2, $H_3AO_3^0$ is the main species [22]. To explore the pH of electrodes deactivation/reproducing mechanism and the impact of different valence and forms arsenic closely related separation removed is important.

3. CONCLUSIONS

In summary, the study of MCDI efficiency for water purification will help promote this technique from theoretical to practical applications; indeed, this mini-review shows that after years of study, MCDI has the potential to be employed in the removal of heavy metals from water/wastewater samples.

Future studies should focus on the synthesis of ion-selective electrodes affording the removal of ions with specific valence values; development of multifunctional electrodes for the simultaneous removal of different kinds of heavy metals; performance of pilot-scale studies instead of bench-scale ones; application of MCDI to raw water or real-life water samples; and apply the different types of current on the MCDI electrode.

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