



## Review

# A short review of activated carbon assisted electrosorption process: An overview, current stage and future prospects

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## ABSTRACT

Stepping into the new globalized and paradigm shifted era, a huge revolution has been undergone by the electrochemical industry. From a humble candidate of the superconductor resources, today electrosorption has demonstrated its wide variety of usefulness, almost in every part of the environmental conservation. With the renaissance of activated carbon (AC), there has been a steadily growing interest in this research field. The paper presents a state of art review of electrosorption technology, its background studies, fundamental chemistry and working principles. Moreover, recent development of the activated carbon assisted electrosorption process, its major challenges together with the future expectation are summarized and discussed. Conclusively, the expanding of electrosorption in the field of adsorption science represents a potentially viable and powerful tool, leading to the superior improvement of pollution control and environmental preservation.

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## 1. Introduction

Concern about environmental protection has increased over the years from a global viewpoint. Throughout the past several decades, an increasing trend in the extensive emission of various organic and inorganic pollutants into the ecosystems has been witnessed [1], underlying the exponential growth rate of population and social civilization, and development of industry and technology as its key drivers [2]. Accordingly, the developing research by the invention of a large variety of treatment technologies (precipitation, coagulation–flocculation, sedimentation, flotation, filtration, membrane processes, biological processes, chemical reactions, adsorption and ion exchange) with varying levels of success [3],

have been promulgated for the removal and recovery of pollutants from the contaminated water and waste streams [4].

Of major interest, adsorption process particularly by utilization of activated carbons is recognized as a prevailing technique which has become the focus of intense research and applied to almost every field of chemistry [5], mainly hinges on its simplicity of design, ease of operation, insensitivity to toxic substances and complete removal of pollutants even from dilute solutions [6]. Despite its prolific use in adsorption process, the biggest barrier of its application by the industries is the high cost of adsorbents presently available for commercial use and difficulties associated with regeneration [7]. This has stimulated a growing research interest in establishing a leading selective, reliable and durable alternative for the environmental conservation [8].

The adoption of environmental electrochemical approach, electrosorption which combines the enhancement of adsorption rate and capacity by polarization and electrochemical regeneration in

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situ [9], has received extensive stern consideration and supports, attributed to the dramatic advantages of both waste minimization and reduced processing costs [10]. In this respect, a 34% improvement of the benzene adsorption capacity on activated carbon was indicated by Plaisance et al. [11] in a study conducted in a 0.2 mol/L KCl solution. Meanwhile, Guo et al. [12] reported that a 60% enhancement of the chloroform adsorption capacity could be effectively achieved by the electrochemical polarization in his investigation. With the above aforementioned, this review attempts to summarize the principle, properties, progress and potential applications of electrosorption technology as remedial alternative to activated carbons in environmental preservation. The present work is aimed at providing a concise and up to date picture of the present status of electrosorption enhancing the adsorption rate and capacity. The challenges together with its future perspectives has been highlighted and outlined, to familiarize the reader with pertinent information regarding electrosorption technology.

## 2. Electrosorption technology

### 2.1. Definition and background studies

In general, electrosorption is defined as a current or polarization potential-induced adsorption phenomenon on the surface of the charged electrodes [13]. Specifically, when an external electrostatic field is imposed to the surface of the electrodes immersed in an aqueous electrolyte solution, charged ions are forced to move towards the oppositely charged electrodes, allowing the occurrence of charge separation across the interface [14], resulting in the formation of strong electrical double layers near the high conductivity and high surface area surfaces [15]. By the removal of the electrical field, the charge on the electrode is compensated for ionic countercharges at the electrolyte side of the interface, accomplished by the contact adsorption of ions [16], or by an accumulation of counter ions in the outer Helmholtz plane and the diffuse (Gouy–Chapman) layer (Fig. 1).

Historically, the pioneering work of electrosorption was firstly reported by Grahame, who conducting a potential-controlled adsorption study of n-octanol, a non-ionic surfactant in 1946 [17]. In the middle of the 1960s, the early concept regarding the retention of ions in the electric double layer has been introduced by Caudle et al. and published at the University of Oklahoma, describing the flow-through capacitors with porous activated carbon electrodes for desalination of sea water [18,19]. In the 1970s, similar investigations have been advanced by Johnson et al. [20,21], demonstrating the preferential adsorption of divalent ions onto the porous carbon electrodes, incorporated with the verification of theoretical

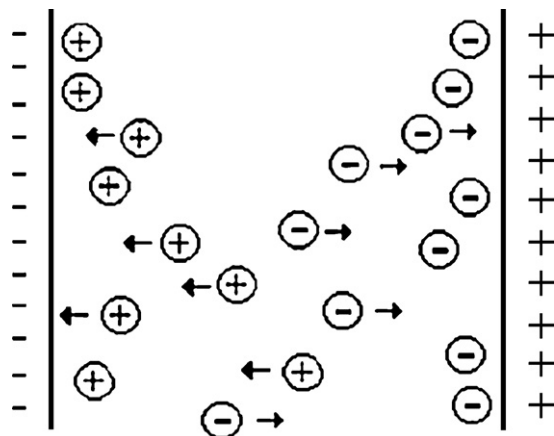


Fig. 1. Electrosorption deionization operating principle [10].

basis, parametric studies, and evaluation of a variety of candidate electrode materials.

More recently, in the 1980s, desalination work with porous activated carbon electrodes was initiated [22,23], utilizing a column with two separated vertical beds of activated carbon, one serving as the anode and the other serving as the cathode, for attaining of ion contaminants with concentration ratios as high as 1/100. Today, electrosorption chemistry has become a rapidly developing branch in numerous fields of research, industry and application, which finds diverse applications in the areas of catalysis, production of computer hardware, medical diagnostics, biological sensors, hydrogen storages and pollutants purification processes (phenol, non-ionic surfactant, benzene, aniline, bipyridyl and inorganic compounds) [8,24].

### 2.2. Electrosorption theoretical studies and its working principles

During the last few years, special attention was paid towards the theoretical and computational understanding of the statics and dynamics of the electrosorption processes [25]. In fact, as early as 1879, the first model of the electrode/electrolyte interface has been proposed by von Helmholtz and later presented as the Gouy–Chapman–Stern model in 1913 [26]. Theoretically, at any interface between an electronic conductor (a solid electrode or an immiscible liquid phase) and electrolyte solution, there exist some regions of which the two sides of the interface are carrying an excess of opposite electrical charges, while the whole interfacial system is electrically neutral, defined by the term of electrical double layer [27] (Fig. 2). Hereby, the electrical capacity of the interface,  $C_T$  ( $\mu\text{F m}^{-2}$ ) is expressed as the summation of two capacitors in series as

$$\frac{1}{C_T} = \frac{1}{C_{M-H}} + \frac{1}{C_{H-S}} \quad (1)$$

of which  $C_{M-H}$  ( $\mu\text{F m}^{-2}$ ) is denoted to the capacity of a compact double layer between the electrode surface,  $M$  and the plane of closest approach for the ions,  $H$  [known as the outer Helmholtz plane (OHP)], where  $C_{H-S}$  ( $\mu\text{F m}^{-2}$ ) is the capacity of the diffuse double layer spanning from plane  $H$  into the electrolyte solution. Meanwhile, the electrical potential distribution of the symmetric  $z:z$  electrolyte solution is described by the planar Poisson–Boltzmann

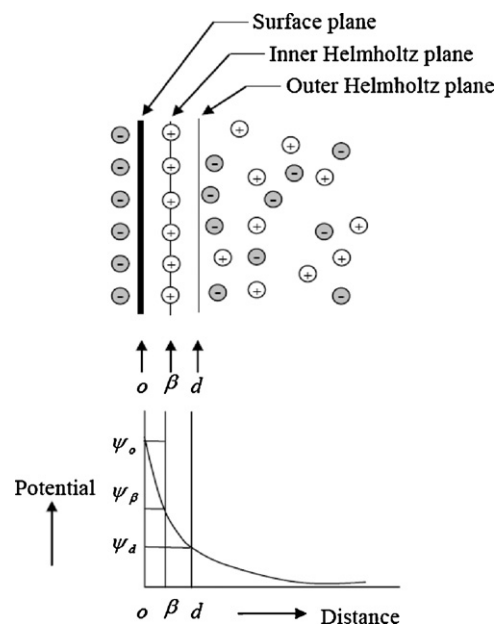


Fig. 2. The schematic diagram of the electrical double layer [28].

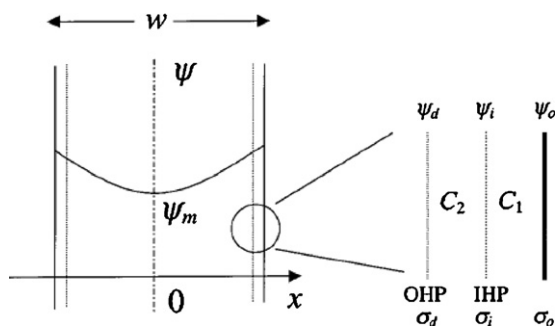


Fig. 3. Theoretical distribution of electrical potential on the Helmholtz plane [29].

(PB) equation [29] as

$$\frac{d^2\psi}{dx^2} = \frac{2zeN_0}{\varepsilon} \sinh\left(\frac{ze\psi}{kT}\right) \quad (2)$$

where  $\psi$ ,  $z$ ,  $e$ ,  $N_0$ ,  $x$ ,  $\varepsilon$ ,  $k$  and  $T$  are individually representing the electrical potential (V), valence of the ions, electrical charge of the electron, total number of the ions in the bulk solution, distance variable (m), position-dependent dielectric constant, Boltzmann constant ( $VK^{-1}$ ) and absolute temperature (K). In the perspective, the inner interface is subdivided into two separated layers, consisting of the inner Helmholtz plane (IMP) and outer Helmholtz (diffuse-layer) plane (OHP), of which its width is half of the diameter of the hydrated ions (Fig. 3) [30], following of the boundary conditions:

$$\frac{d\psi}{dx} = 0 \quad \text{and} \quad \psi = \psi_m \quad \text{at} \quad x = 0 \quad (3)$$

$$\psi = \psi_d \quad \text{at} \quad x = \pm \frac{w-a}{2} \quad (4)$$

$$\psi = \psi_o \quad \text{at} \quad x = \pm \frac{w}{2} \quad (5)$$

where  $\psi_m$ ,  $\psi_d$ ,  $\psi_o$ ,  $w$  and  $a$  are individually referred to the electrical potential at the midplane (V), the diffuse-layer potential (V), surface potential (V), pore width ( $\text{\AA}$ ) and the diameter of a fully hydrated counter ion (usually  $4.25 \text{\AA}$ ) [31]. In most cases, the relationship of the diffuse-layer potential is given by [32,33]:

$$\psi_d = \psi_o - \frac{\sigma_0}{C_1} \quad (6)$$

$$= \frac{V}{2} - \phi_{ecm} - \frac{\sigma_0}{C_1} \quad (7)$$

Table 1

Previous researches of activated carbon assisted electrosorption processes.

Adsorbent/precursor	Adsorbate	Maximum adsorption capacity (mg/g)	Reference
Activated carbon fibers (commercial)	Acid Orange 7 dye	644.59	[9]
Polyacrylonitrile resin	Phenol	225.86	[13]
Activated carbon fibers (commercial)	Naphthalenesulfonic acid	320.00	[16]
	Benzyl alcohol	210.00	
	Naphthoic acid	200.00	
Activated carbon cloth (commercial)	Bentazone	30.47	[25]
Spectracarb 2225 (commercial)	Chromium (VI)	7.28	[43]
Activated carbon fibers (commercial)	Phenoxide ions	207.04	[46]
	<i>p</i> -Nitrophenol	407.59	
	Sodium dodecylbenzene sulfonate	766.66	
Activated carbon fibers (commercial)	Uranium	5.02	[55]
Polyacrylonitrile resin	Aniline	315.71	[56]
Polyacrylonitrile resin	<i>m</i> -Cresol	367.68	[57]
NFEN 12915 (commercial)	Metribuzin pesticide	210.00	[58]
Spectracarb 2225 (commercial)	Nitrate	1.12	[59]
	Nitrite	0.83	
Coconut charcoal	Phenol	188.00	[60]
Spectracarb 225 (commercial)	Ethyl xanthate	1120.55	[61]
	Thiocyanate	823.25	

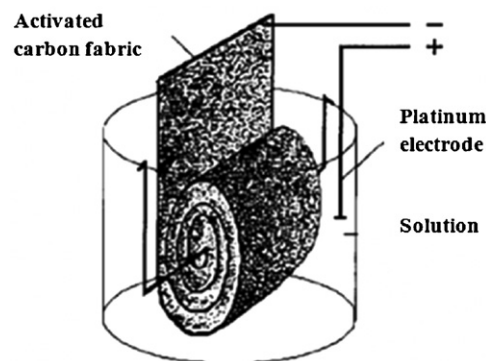


Fig. 4. The basic experimental setup of the electrosorption process [62].

of which the difference between  $\psi_d$  and  $V/2$  can be viewed as the potential difference across the electrical double layer, implying a driving force in the ions adsorption mechanism. Accordingly, the surface charge density for a single symmetric electrolyte solution is derived as [34]:

$$\sigma_0 = \sqrt{4\varepsilon RTI} \left[ \cosh\left(\frac{e\psi_d}{kT}\right) - \cosh\left(\frac{e\psi_m}{kT}\right) \right] \quad (8)$$

where  $\phi_{ecm}$ ,  $C_1$ ,  $\sigma_0$ ,  $R$  and  $I$  are corresponding the potential at the electrical capillary maximum (V), inner-layer capacitance ( $\mu\text{F m}^{-2}$ ), surface charge density ( $\text{C m}^{-1}$ ) gas constant ( $\text{L at m mol}^{-1} \text{K}^{-1}$ ) and ionic strength ( $\text{mol m}^{-3}$ ), respectively.

### 3. Activated carbon assisted electrosorption

Within recent decades, intensive wide spread contamination of atmosphere and surface water related to extensive industrial operations has inspired a great global attention for many environmentalists [35]. Undoubtedly, activated carbon, an adsorbent with its large porous surface area, controllable pore structure, thermo-stability and low acid/base reactivity [36], is gaining huge momentum and popularity, owing to its competent ability in wastewater treatment processes [37] and as air purifier in cleaning of atmosphere contaminants [38]. Along with the notable trend of its growing investigation interest and research efforts, a wide variety of publications covering the theory of adsorption science and its manifestations measurements have currently been proliferated [39–41].

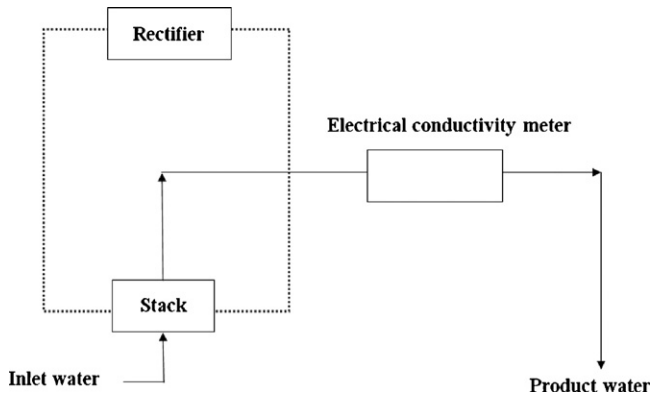


Fig. 5. Schematic flow diagram of the electrosorption desalination equipment [47].

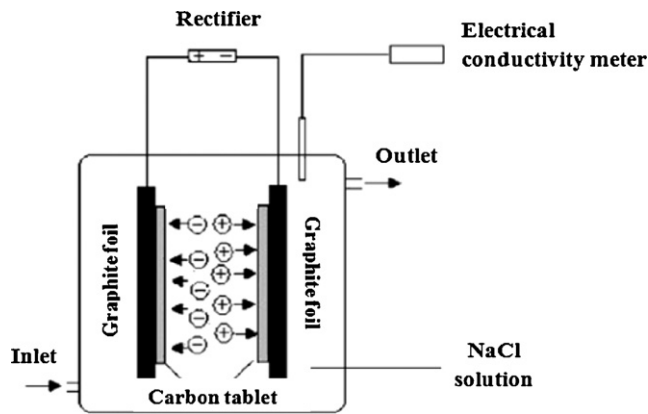


Fig. 6. The schematic of the electrosorption unit cell [10].

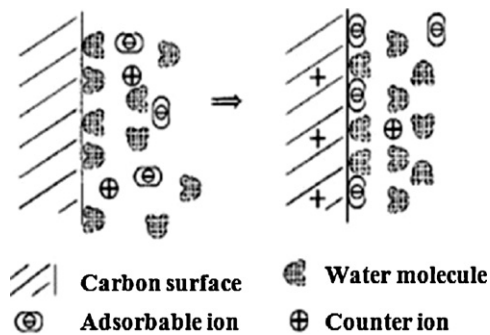


Fig. 7. The structure of the activated carbon surface/aqueous electrolyte interface [16].

Explicitly, despite various exploitations and extensive studies have been undertaken, such exertions are always handicapped by the constraints of its poor economic feasibility, limited suitability for large scale application [42,43] and difficulties associated with regeneration (adsorption capacity lost by attrition, burn-off and washout in thermal regeneration, large investment cost requirement for chemical regeneration and slow and low applicability for bio-regeneration) [44]. Moreover, in some cases, when an exhausted carbon reached its saturation limit, it was usually taken to a landfill or incinerator to be discarded [45], without establishing the real actions and standard procedure, contributing to another form of pollution phenomenon—secondary pollutants [46].

In view with the above matter, the departure of the concept of electrosorption, an attractive combination of the high adsorption capacity and the electrical conductivity of graphitic carbons, operating in the presence of an applied low direct current voltages

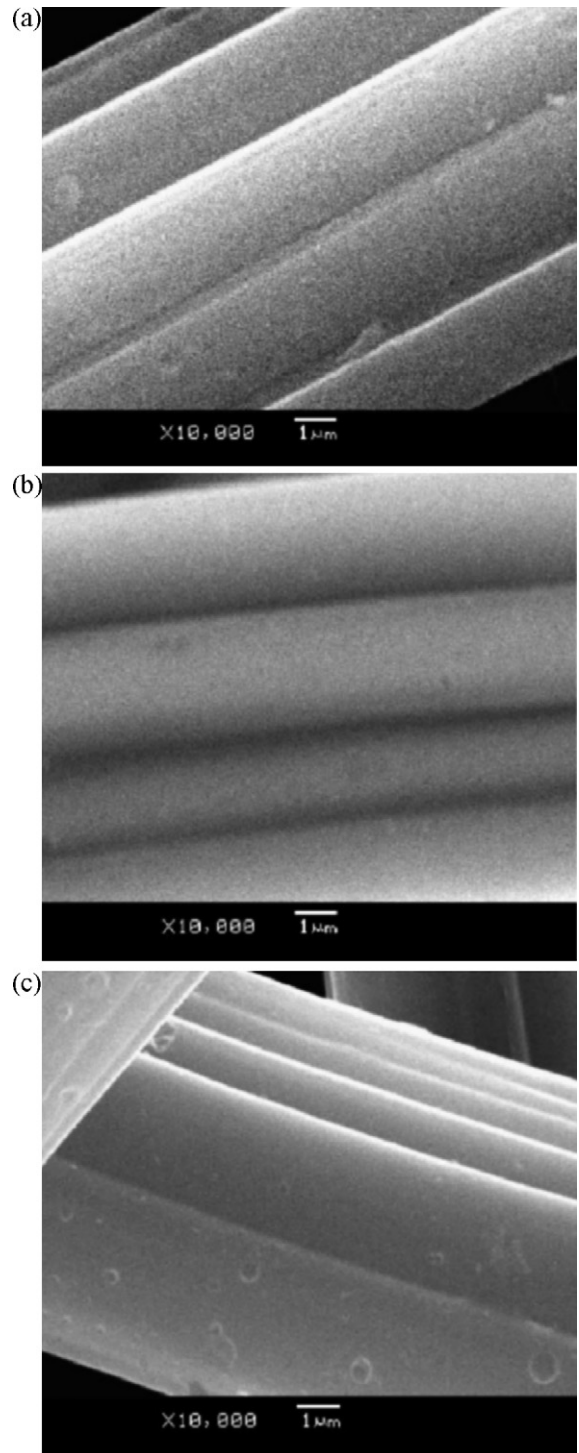
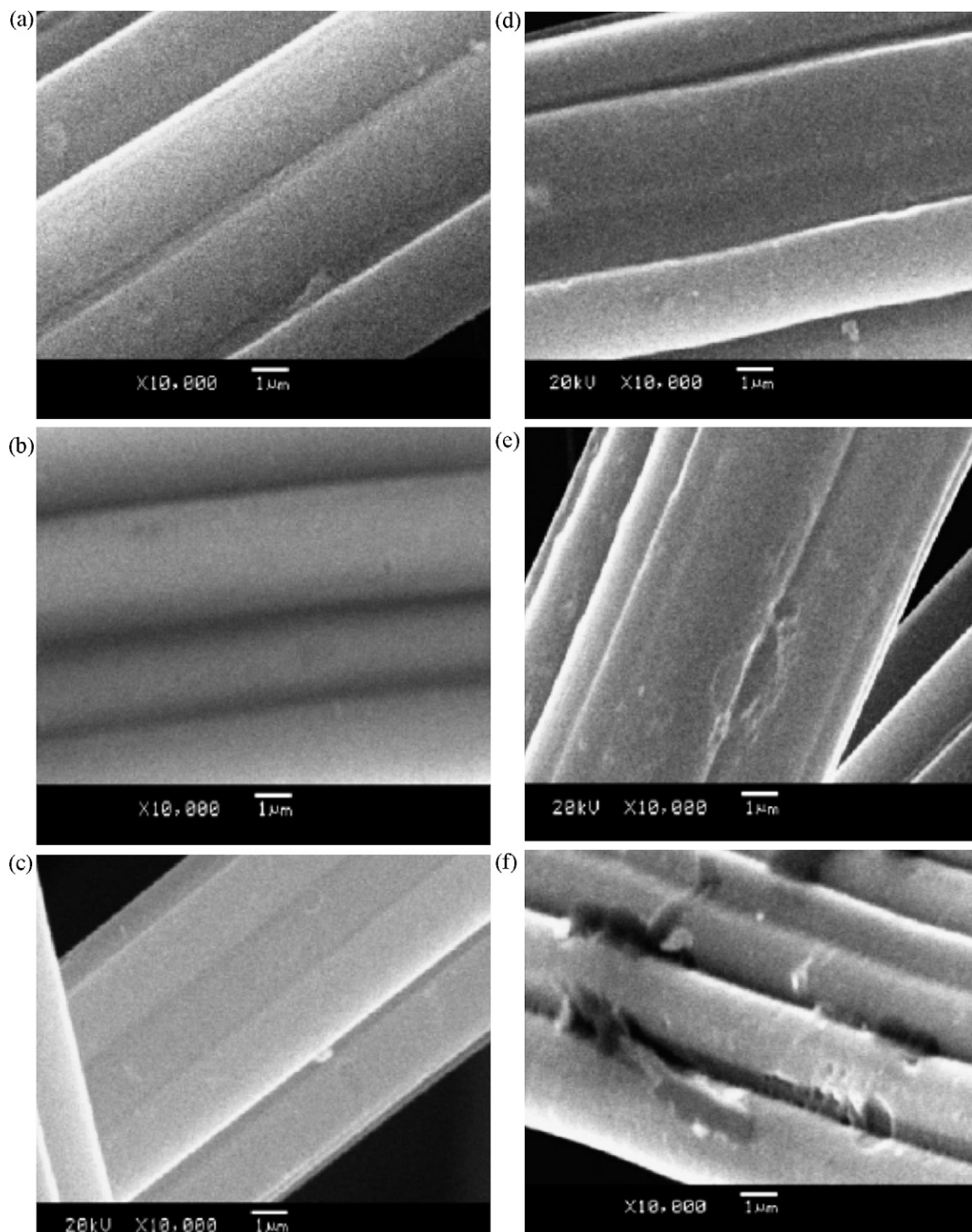


Fig. 8. Scanning electron micrographs of activated carbon fibers: (a) initial surface; (b) after adsorbing aniline; (c) after adsorbing aniline at the polarization of 600 mV [57].

[47] without high-pressure pumps, thermal heaters, membranes, distillation columns or chemicals [48,49], has recently receiving great supports and recognition worldwide as a minimally polluting, energy saving and potentially cost-effective purification technology [50]. In contrast to adsorption of activated carbon, earlier studies exhibited its promising role in enhancing the good extents of impurities removal and possibility of accelerating the adsorption rate and capacity by manipulating the interfacial potential of the conductive electrodes [51,52]. Increasingly, it provides a convenient means of



**Fig. 9.** Scanning electron micrograph (SEM) images of activated carbon fibers: (a) initial surface, (b) to (f) after polarizing at 200, 400, 600, 800 and 1000 mV, respectively [58].

controlling the adsorption and desorption processes, accomplishing a direction for in situ restoration of adsorbents into the purge stream through the reversed potential, enabling the capability of regeneration of the exhausted activated carbon [53,54].

Table 1 summarizes previous researches of activated carbon assisted electrosorption processes, where Figs. 4–6 signify schematic diagrams of the electrosorption setup. In this aspect, the double-layer adsorption theories state that the extent of adsorption capacities are governed primarily by the electrostatic attraction force between the ions and the electrode, affecting by the

ion solution concentration and applied voltage (the double-layer capacitance, available interfacial area and the applied potential), [63,64] and pseudocapacity due to Faradaic reactions, which depends strongly on the chemical characteristics of the solute and functional groups on the electrode surface (the orientation of the molecule at the interface and the type of bonding between the adsorbate with the adsorbents) [40,65].

In the presence of organic substances, the mechanism is presumed to be induced via  $\pi$ - $\pi$  dispersive interactions with the aromatic rings approaching to the carbon surface and electrostatic

attraction or repulsion force [25,42] (Fig. 7), of which the following expressions indicate the ion exchange reactions between the cations and the surface acidic groups [56]:

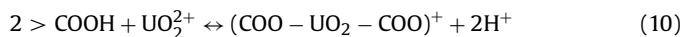
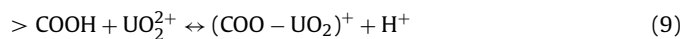


Fig. 8 shows scanning electron micrograph (SEM) images for activated carbon fibers in the electrosorption of aniline, an aromatic amine. The micrographs display a gradual increase in the smoothness of its surface from Fig. 8(a)–(c), indicating different degrees of adsorption [57]. Depending on the nature of the adsorbent, SEM images for activated carbon fibers at various polarized potentials are illustrated in Fig. 9. A comparison of the surfaces exhibited no evident difference while the polarized potential is less than 600 mV, and with the enhancement of 800 mV, pits formation appear to be occurred on the surfaces, attributed to the development of active oxidizing free radicals [58].

#### 4. Current stage and future prospects

The world is currently facing the worst environmental crisis in its entire history. For the past two decades, the enthusiasm of huge waste production and environmental preservation has been one of the most challenging topics which has focused greatest public concern and critical considerations towards the recovery of contamination resources. In line with the growing anxiety on the environment-friendly technologies and achieving the status of green industries policy, a supportive engineering measure, electrosorption, which incorporating electrochemical innovation in the adsorption science has lately released extended interest all around the globe [66]. During its operations, the adsorption capacity is associated closely with the accessibility, stability, permeability [67], surface properties (surface area, pore microstructure, and pore size distribution [68]) as well as the production cost of the electrodes, of which an appropriate electrodes selection is set to play a key role in accessing an electrosorption system [69].

In the conventional approach, carbonaceous materials including the graphite granules or chips, glassy carbon grains, crushed graphite, screens of graphite and reticulated vitreous carbon (RVC) has represented an attractive family of electrodes [70], owing of its ease of processability, relatively low cost and environmental friendliness [71]. Nevertheless, a number of recent researches reported that several intrinsic problems have been encountered with the conventional activated carbon electrodes, characterized by a low conductivity, irregular pore structures, large potential and pressure drop (in thick electrodes and packed beds), high electrical or mass transfer resistance, binder degradation (susceptibility to both chemical attack and radiation-induced degradation) and channeling, thereby resulting in lower process efficiency and overall capacity [50].

In light with the above drawbacks, lately, the potentiality of the carbon aerogel, a monolithic, high-surface-area area ( $\sim 400\text{--}1000 \text{ m}^2/\text{g}$ ), low electrical resistivity ( $\leq 40 \text{ m}\Omega \text{ cm}$ ) and controllable pore size distribution ( $\leq 50 \text{ nm}$ ) porous material was noted having excellent properties in electrosorption processes, effectively removing a wide range ions of sodium, chloride, chromium, ammonium, and perchlorate from aqueous solutions [72]. In the Lawrence Livermore National Laboratory (LLNL) of Berkley, CA, it has been extensively utilized in a laboratory-scale non-membrane electrosorption process, capacitive deionization technology (CDT) for continuous treatment of the ionic impurities rich saline solutions [64].

Another novel nanoscale fiber, carbon nanotube (CNT) with its large surface area, high mechanical strength, remarkable electrical conductivities, superb chemical stability and light mass, has

currently illustrated tremendous viability for the application as electrode material [73,74]. Posing the highly  $\pi$ -conjugative and hydrophobic sidewalls consisting of  $\text{sp}^2$  carbons and open ends bearing oxygen-containing moieties, carbon nanotube has indicated its favorably function as a support for organic and inorganic electrocatalysts [75]. Although there have been some successful lab scale industrial applications, generally the industry is still facing various challenges, the availability of economically viable technology, sophisticated and sustainable resources management, and proper market strategies under competitive markets. Increasingly, several separated studies have recorded the deflections of high resistivity and low mechanical strength of carbon aerogels, and the restrictions of low production yield and high modification cost associated with carbon nanotubes [46].

Amidst these shortcomings, Dai et al. [76] has proposed a new electrochemical double layer electrode fabricating process combining active carbon and multi-walled carbon nanotubes for the brackish water desalination treatment. In the present state, the electrosorption module which encompassing primary of the electrodes, spacers, and current collectors [47], has not yet emerged from the laboratory, small pilot scale and demonstration units to become a valid and mature commercial technology system [15]. Although it is still in its infancy, the development and implementation of dedicated researches and additional insights is still the single and most important factor which determines the successfulness in this area. Ultimately, full co-operation and integration between different parties with compatible technology investigations is a key motivation for the race to the end line.

#### 5. Conclusion

To date, rapidly changing technologies, population growth, industrial products and practices are increasing worldwide, driving towards the overwhelming environment pollutions, ecological imbalance, upsetting of biological processes and threatening of the public health. The past ten years has seen a developing interest in the preparation of activated carbons in water and wastewater treatment processes. Lately, the limited success of adsorbents in field applications has raised apprehensions over the use of electrosorption as a measure to the environmental pollution control. The evolution has turned from an interesting alternative approach into a powerful standard technique by offering a numbers of advantages: minimally polluting, energy saving, cost-effective, capability of regeneration, enhancing the good extents of impurities removal, and possibility of accelerating the adsorption rate and capacity. Despite various drawbacks and challenges has been identified and clarified, a widespread and great progress of in this area can be expected in the future.

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