Combined Electrocoagulation and Adsorption Processes

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The electrocoagulation (EC) process is a possible alternative to conventional wastewater treatment methods. Characteristics of the process, such as its flexibility, easy operation, no need for additional chemicals, and its ability to deal with different contaminants, have been increasing the interest in its implementation. The EC process found application in the treatment of different contaminated waters, and several studies have shown the potential of this technology. Adsorption (AD) is another attractive way of treating wastewaters due to the potential of using low-cost and environmentally friendly adsorbents. Due to their high surface area and well-developed pore structure, activated carbons are the most used adsorbents in wastewater treatment systems. The high price of activated carbon limits its application. The combination of the EC and AD processes can be used to amplify the advantages that each process presents in treating wastewaters. As a first step, the EC process reduces the pollutant loading and the suspended solids concentration, which can benefit the AD process by delaying the adsorbent saturation and preventing clogging. Additionally, each adsorption/regeneration cycle could result in the adsorbent losing some of its capacity; as a result, delaying adsorbent saturation helps to increase its useful life.

Keywords: electrocoagulation ; adsorption ; economy

1. Applications of Combined Electrocoagulation and Adsorption Processes

Wastewater treatment processes combining electrocoagulation (EC) with adsorption (AD) have been used to remove target pollutants. **Table 1** summarizes processes combining EC and AD to treat different wastewaters.

Table 1. Combined electrocoagulation and adsorption on the treatment of different wastewaters (COD—chemical oxygen demand; BOD—biochemical oxygen demand; TDS—total dissolved solids; TSS—total suspended solids).

Wastewater	Adsorbent	Electrodes	Removal Efficiency	Reference
Industrial wastewater	**Ectodermis of Opuntia**	Al	**84% (COD) 78% (BOD) 97% (color) 98% (turbidity) 99% (fecal coliforms)**	[1]
Aqueous solution	**Granular activated carbon**	Zn	**99.88% (Pb(II))**	[2]
Textile wastewater	**Crude Tunisian clay**	Fe	**96.87% (color) 89.77% (COD) 84.46% (TSS)**	[3]
Beverage industry wastewater	**Activated carbon**	Al	**98.66% (COD) 92.15% (TSS) 90.12% (color)**	[4]
Semiconductor wastewater	**Activated carbon**	Al	**67.25% (fluoride)**	[5]
Produced water	**Coconut shell activated carbon**	Al/Fe	**98.39% (COD) 93.54% (TDS) 75.16% (ammonia) 97.56% (oil content) 92.5% (phenol)**	[6]
Nitrate-contaminated ground water	**Zeolite**	Al	**96% (nitrates)**	[7]
Automobile wastewater	**Activated carbon**	Al	**71.58% (COD) 77.91% (surfactant)**	[8]

Wastewater	Adsorbent	Electrodes	Removal Efficiency	Reference
Tanning wastewater	**Eggshell**	Al	**99% (Cr(VI))**	[9]
Anaerobic wastewater	**Granular activated carbon**	Al	**100% (COD)** **100% (BOD)** **96.5% (turbidity)** **97.5% (phosphorus)**	[10]
Paper mill effluent	**Granular activated carbon**	Al/Fe	**98.97% (COD)**	[11]
Model solution	**Red onion skin**	Al	**97% (Cr(VI))**	[12]
Cellulose and paper industry wastewater	**Granular activated carbon**	Al	**93% (humic acid)**	[13]
Dye solution	**Banana peel**	Al	**99% (methylene blue)**	[14]
Mine waters	**Rice straw activated carbon**	Al	**95.2% (sulphate)**	[15]
Dairy wastewater	**Granular activated carbon**	Al	**99.39% (turbidity)** **87.12 (COD)**	[16]

Several combinations of electrodes and adsorbents have been used in the combined EC and AD processes to treat different wastewaters. This characteristic of the process increases its versatility in dealing with specific pollutant removal problems.

Wang et al. [17] applied EC combined with peanut shell adsorption to remove malachite green from an aqueous solution. They found that with an adsorbent dosage of 5 g/L, the removal efficiency increased significantly, requiring lower current density and shorter operating time than the EC process alone. At optimal conditions, the removal efficiency of 98% was obtained in 5 min, corresponding to an increase of 23% relative to the removal obtained by the EC process in 60 min. Moreover, it was concluded that the combined process reduces 94% of the unit energy and electrode material demand compared to the EC process alone.

Yang et al. [18] studied the removal of several micropollutants from real municipal wastewater using graphene adsorption and simultaneous electrocoagulation/electrofiltration. They concluded that the main mechanisms involved in the combined process were carbon adsorption, size exclusion, electrostatic repulsion, EC, and electrofiltration. The removal efficiencies obtained with the combined process were the following: di-n-butyl phthalate 89%, di-(2-ethylhexyl) phthalate 85%, acetaminophen 99%, caffeine 94%, cefalexin 100%, and sulfamethoxazole 98%.

Narayanan and Ganesan [19] used batch EC with an Al–Fe electrode pair combined with adsorption using granular activated carbon to remove Cr(VI) from synthetic effluents. They identified that the combined effect of chemical precipitation, coprecipitation, sweep coagulation, and adsorption are the main mechanisms involved in the Cr(VI) removal. The results showed that adding adsorbent to the process provides a higher Cr(VI) removal rate, requiring lower current intensities and operating time than the conventional EC process.

Castañeda-Díaz et al. [20] studied the removal of the cationic dye malachite green and the anionic dye remazol yellow from aqueous solutions using EC followed by an AD process. The AD was performed in a packed column with carbonaceous material from industrial sewage sludge and commercial activated carbons as adsorbents. Batch and continuous set-ups were tested for the EC part of the process. The results showed that the continuous EC–AD process was more efficient than the EC alone.

Rubí-Juárez et al. [21] performed a comparative study of the COD and turbidity removal from car wash wastewater using an EC process followed by AD and simultaneous EC–AD. Removal of 78% for COD and 92% for turbidity was obtained after 60 min using aluminum electrodes. After AD with commercial granular activated carbon (GAC) using a dose of 22.5 g/L, the final removals were 99.97% and 98% for COD and turbidity, respectively. The simultaneous EC–AD process using aluminum electrodes and a GAC dosage of 22.5 g/L provided a final removal of 94% for COD and 98% for turbidity.

Nizeyimana et al. [22] studied the removal of copper and nickel ions from synthetic wastewater through AD with low-cost and sustainable activated green tea residue combined with EC with iron electrodes. The adsorbent was prepared without applying any activating agents. The optimized combined process presented a removal efficiency of almost 100% for both copper and nickel ions. Moreover, they concluded that the combined process is the cheapest due to lower adsorbent dosage and energy consumption than the single AD and EC processes.

Bazrafshan et al. [23] proposed a sequential chemical coagulation process with poly aluminum chloride (PAC), EC with aluminum electrodes, and AD with pistachio nutshell ash. The process was applied to the treatment of actual textile wastewater. The chemical coagulation step with a PAC dosage of 30 mg/L presented removals of 40% for COD, 34% for BOD5, and 44.5% for dye. After the EC step with an applied voltage of 60 V, the removals obtained were 93.1, 88.8, and 98.6% for COD, BOD5, and dye, respectively. The combined process, including the AD step, provided 98, 94.2, and 99.9% removals for COD, BOD5, and dye, respectively. The authors concluded that the combined process presents a superior performance compared with processes alone in textile wastewater treatment.

2. The Combined Process from the Perspective of Circular Economy

The circular economy (CE) concept appears as an alternative to the traditional linear economy approach based on the ”take-make-use-dispose” model, in which there is a continuous resource throughput, and the products are usually disposed of as waste at the end of their life cycle. The CE model aims for more sustainable use of raw materials and energy while reducing the volume of waste. In a linear economy model, economic growth is coupled with the negative consequences of resource depletion and environmental degradation. However, in the CE approach, the raw materials and products are kept in the economy for as long as possible, and the wastes are regarded as secondary raw materials that can be recycled and reused [24]. The European Commission recognized the importance of this issue and proposed an action plan to promote the implementation of the CE [25].

Reusing treated wastewater in applications such as agriculture, industrial processes, toilet flushing, and irrigation of parks and recreation grounds can be an essential element of the CE model [24][26]. Moreover, sludges produced from wastewater treatment can be a valuable source of nutrients [27]. From this perspective, wastewater can be regarded as a resource instead of a waste. The water reuse strategy is becoming increasingly important in the present context of water scarcity worldwide. However, when using conventional centralized wastewater treatment plants, the economic aspect can be a significant obstacle to the widespread implementation of water reuse projects [28]. A solution to this limitation can be the development of decentralized wastewater units able to be used far from the centralized wastewater treatment plants usually located near urban centers. To facilitate their application, these treatment units should be efficient, compact, easy to operate, and have low investment and operating costs.

Using waste materials to produce added-value products is also a way to move several manufacturing processes toward a zero-waste circular economy [24]. In this context, the use of wastes as raw materials to produce low-cost adsorbents fits well in the CE model. From the perspective of wastewater treatment, using these kinds of adsorbent materials reduces the costs of the process, facilitating their implementation. This aspect is crucial in developing decentralized wastewater treatment systems outside urban centers.

Despite the potential of EC and AD processes in wastewater treatment, their wastes can represent a significant challenge. Therefore, implementing strategies for valorizing these wastes is fundamental for those processes’ sustainability and economic viability. In recent years, there has been an increasing interest in strategies to valorize the EC process wastes [29]. The use of the sludge resulting from EC process operation to produce fertilizers [30], pigments [31], construction materials [32], adsorbents [33], and catalysts [34] are possible ways to valorize this by-product. In addition to these applications, EC sludge has also been considered an alternative energy source. The energy can be obtained from the sludge by combustion, anaerobic digestion, pyrolysis, and gasification [35]. Hydrogen generated at the cathodes by water electrolysis is another potentially valuable by-product of the EC process. The hydrogen recovered during the EC process can be used as an alternative energy source or a reactant for industrial processes [36].

After an adsorption-based wastewater treatment, the saturated adsorbents are traditionally disposed to landfill or regenerated and reused. Hazardous compounds in the adsorbents make their disposal in landfills potentially detrimental to the environment. However, the regeneration of the saturated adsorbents may represent a large part of the treatment process costs. Moreover, the regeneration process can cause secondary pollution due to the addition of regeneration agents and the concentration of contaminants [37]. Alternative strategies for the valorization of spent adsorbents have been proposed, including their application as fertilizers [38] and catalysts [39], and in the production of cementitious materials [40] and energy production [41].

The combination of EC and AD for the treatment of wastewaters has the characteristics to be part of a CE model. First, this combined process has the potential to be used for decentralized wastewater treatment facilitating the implementation of water reuse projects and, consequently, more sustainable use of water resources. Another characteristic of the process is the possibility of using low-cost adsorbents produced from waste. Finally, the wastes produced during the wastewater treatment with the combined process can be valorized into useful materials. In addition to the obvious economic advantage, using this kind of materials also reduces waste from several manufacturing activities. **Figure 1** shows a schematic representation of the combined process application with low-cost materials for wastewater treatment, the different routes for valorizing its wastes, and the possible applications of the treated water.

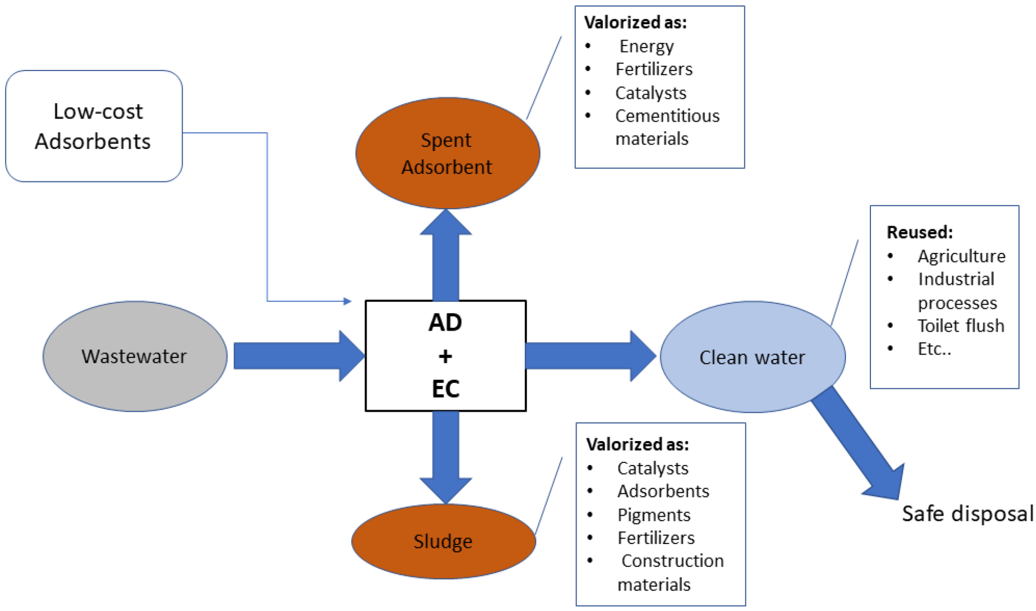

Figure 1. The combined process from the perspective of a circular economy.

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