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Review

A Brief Review on the Regeneration of Spent Activated Carbon and Pollutant Migration Behavior

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Abstract: The regeneration of spent activated carbon saturated with adsorbates inherently involves pollutant migration, including volatile organic compounds (VOCs) and heavy metals. However, a comprehensive and systematic understanding of the varying pollutant migration behaviors across different regeneration techniques remains limited. This study reviews the regeneration of spent activated carbon and innovatively contrasts pollutant migration behavior for three widely used regeneration techniques: thermal, solvent, and electrochemical regeneration. A key contribution lies in highlighting the distinct pollutant migration profiles associated with traditional methods like incineration compared to emerging techniques such as microwave thermal regeneration and supercritical CO_2 solvent regeneration, emphasizing their implications for secondary pollution control. The current state of spent activated carbon regeneration technologies is critically evaluated in light of both engineering practices and research progress, and potential avenues for future development are proposed. This research provides valuable insights for the efficient regeneration of spent activated carbon and pollution control during the regeneration process.

Keywords: spent activated carbon; pollutants; migration and transformation; thermal regeneration; electrochemical regeneration

1. Introduction

Activated carbon is a processed carbonaceous material characterized by a porous structure that confers excellent adsorption properties [1]. Typically produced from abundant and high-carbon-content materials such as coal and biomass through chemical or physical activation processes, activated carbon is available in various forms, including granular, powdered, and honeycomb, each with widespread applications [2–5]. Possessing strong adsorption capacity, stable physicochemical properties, high strength, and resistance to high temperatures, acids, and bases, activated carbon is extensively used in diverse fields such as environmental protection and chemical engineering [6,7]. During the adsorption of pollutants, the adsorption capacity of activated carbon gradually decreases. Once it no longer meets the requirements, it becomes spent activated carbon and is managed as a solid waste. With the increasing annual consumption of activated carbon, the proper disposal of spent activated carbon has become a widespread concern. In 2023, the State Council

released the Action Plan for Continuous Improvement of Air Quality, highlighting the importance of spent activated carbon regeneration.

The conventional approach to managing spent activated carbon involves incineration [8]. However, this method produces considerable quantities of combustion emissions that necessitate further treatment, while simultaneously squandering the inherent value of the high-grade carbonaceous material. Spent activated carbon regeneration is an effective means of resource utilization, as it substantially restores the carbon's initial activity, enabling subsequent reuse. This process presents notable economic and environmental advantages over traditional disposal strategies. Currently, various effective regeneration techniques, such as thermal and solvent regeneration, have been developed [9–14]. However, the process of restoring the adsorption capacity of spent activated carbon through regeneration is also a process of desorbing pollutants into the environment. Because pollutants are merely transferred rather than effectively degraded, this undoubtedly increases the environmental and human health risks associated with spent activated carbon regeneration, hindering industry development. Therefore, the migration patterns of pollutants during spent activated carbon regeneration warrant attention.

The use of activated carbon for removing contaminants from water and air streams generates significant quantities of spent activated carbon. While regeneration offers a sustainable approach to managing this waste, the process can inadvertently transfer adsorbed pollutants from the carbon matrix into other environmental compartments, posing secondary pollution risks. This study critically reviews commonly employed spent activated carbon regeneration methods, encompassing thermal, solvent, and electrochemical approaches. Furthermore, it provides a detailed analysis of the fate and transport of various pollutants during regeneration, including volatile organic compounds, heavy metals, and emerging contaminants. The overarching goal is to offer valuable insights and guidance for optimizing regeneration processes to achieve greater efficiency, minimize environmental impacts, and safeguard public health.

2. Research Status of Spent Activated Carbon Regeneration

To assess the current research landscape, the Web of Science Core Collection database was searched using "Spent activated carbon regeneration" as the topic search term, covering the period from 2008 to 2024. This search yielded a total of 2003 publications. Figure 1 illustrates that the number of related studies grew consistently from 26 in 2008 to 251 in 2021, representing a substantial average annual growth rate of 19.1%. The surge in publications post-2018 may correlate with stricter environmental policies. While the number of publications has decreased since 2021, a considerable volume of articles continues to be published annually, demonstrating the continued widespread interest in spent activated carbon regeneration.

In terms of research content, the focus of related studies is diverse and complex. Keywords related to research objectives include "adsorption," "removal," and "kinetics." The studied systems encompass "waste water," "groundwater," and "drinking water." The activated carbon raw materials of interest are primarily "graphene oxide," "biomass," and "agricultural waste." Regarding widely studied

regeneration methods, thermal regeneration (44.3%), electrochemical regeneration (40.9%), and chemical regeneration (solvent regeneration, 14.8%) are the most prevalent. Additionally, microwave regeneration and biological regeneration techniques are commonly applied in practical engineering. Biological regeneration offers the advantages of a simple process, low construction and operating costs, and minimal carbon loss. However, it is characterized by long regeneration times and is generally unsuitable for activated carbon with high acid, alkali, or chlorine content [15].

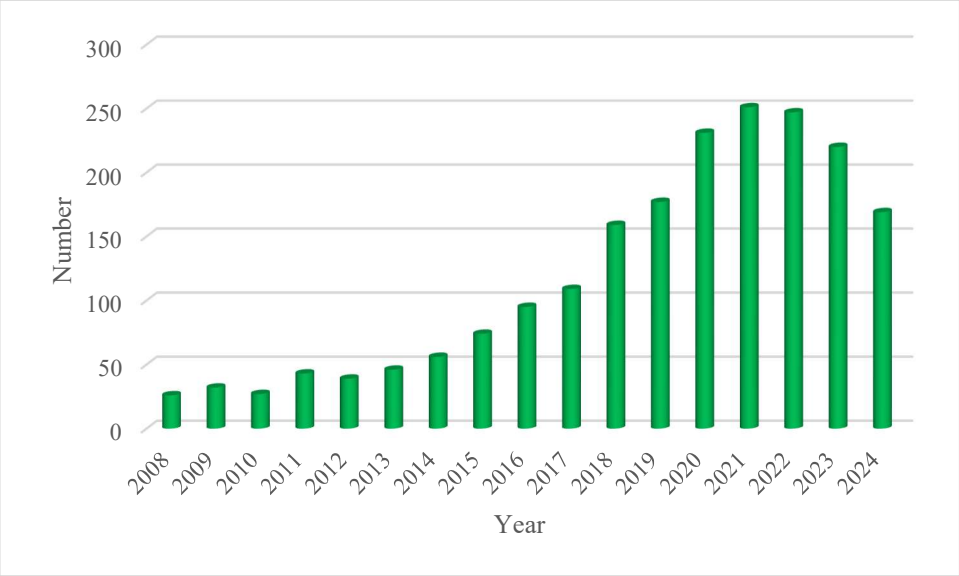

Figure 1. Number of publications from 2008 to 2024.

As illustrated in Figure 2, “regeneration” is strongly associated with keywords such as “removal” and “degradation,” underscoring the fundamental goal of restoring the activated carbon’s capacity to eliminate pollutants. This close interrelationship suggests that understanding the migration and transformation pathways of pollutants during regeneration is a central focus of the field. Further insights arise from examining the clustering of specific regeneration techniques. The proximity of “advanced oxidation processes”, “electrochemical regeneration,” and “thermal regeneration” reveals a growing trend of combining these methods to enhance regeneration efficiency and broaden applicability. Specifically, the link between “oxidation” and “electrochemical regeneration” reflects the increasing integration of these approaches to leverage their synergistic effects. Electrochemical methods can generate powerful oxidizing agents in situ, such as hydroxyl radicals or ozone, which then degrade adsorbed organic pollutants directly on the activated carbon surface. This combination enhances the oxidative power and avoids the need for external addition of chemical oxidants, offering improved control and potentially reduced costs. The dual focus on regeneration alongside the oxidative removal of pollutants reinforces the current trend toward integrated systems for both reclaiming the adsorbent and eliminating the adsorbed contaminants.

Within this context, dyes and other relatively simple pollutants exhibit the strongest associations, likely reflecting the emphasis on fundamental mechanistic

studies in related research. These studies often aim to elucidate the reaction pathways and limiting steps involved in pollutant degradation. In contrast, the red cluster encompasses a diverse array of pollutants, including heavy metals, ions, and methylene blue, suggesting a research focus on more complex pollution scenarios involving both organic and heavy metal contamination. These studies tend to investigate the distribution mechanisms, kinetics, and thermodynamics of diverse pollutants during regeneration processes relevant to real-world applications. Furthermore, a significant portion of the research explores the relationship between spent activated carbon regeneration and its source materials (blue cluster). Beyond conventional biochar, these studies consider raw materials such as agricultural waste, biomass, and even shells, and examine the impact of the type of raw material on the activated carbon's surface area, pore structure, and surface chemistry. This strong correlation underscores the importance of tailoring the activated carbon material to the specific regeneration process and target pollutants.

In summary, research in this area comprehensively addresses both the migration and transformation of pollutants during spent activated carbon regeneration and the regeneration process itself. However, the multifaceted nature of these studies means that the central research themes and their priority remain somewhat diffuse, and issues within the field, as well as future directions, require further clarification. A deeper understanding of pollutant migration patterns during spent activated carbon regeneration is crucial for promoting the long-term and sustainable development of the activated carbon industry.

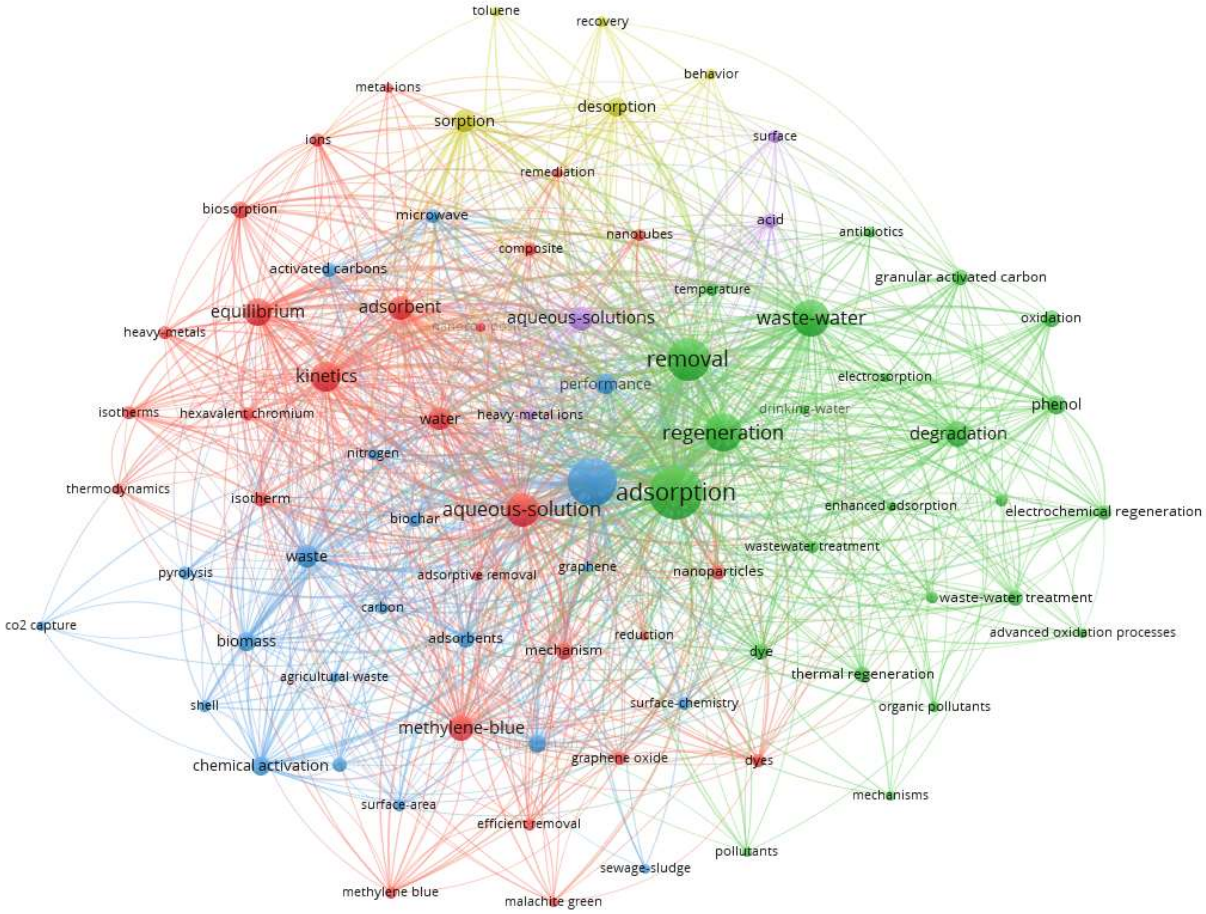

Figure 2. Visualization of keyword relationships in relevant research over the past five years based on VOSviewer analysis.

3. Pollutant Migration Behaviors during Spent Activated Carbon Regeneration

A comparison of thermal, solvent, and electrochemical regeneration, three common methods for regenerating spent activated carbon, is presented in Table 1, focusing on energy consumption, pollutant removal efficiency, and waste by-products. By comparison, thermal regeneration is characterized by high energy consumption and a medium level of pollutant removal efficiency. The main waste by-products from this process are flue gas (CO, NO_X, SO_X), ash, and volatilized organic compounds. Solvent regeneration consumes a moderate amount of energy but achieves lower pollutant removal, resulting in concentrated liquid waste (containing pollutants). Electrochemical regeneration boasts low energy consumption and high pollutant removal efficiency, but its waste stream primarily consists of electrolyte solution (potentially containing degraded pollutants) and electrode corrosion products.

Table 1. Comparison of spent activated carbon regeneration methods.

Regeneration Method	Energy Consumption	Pollutant Removal Efficiency	Waste By-products
Thermal regeneration	High	Medium	Flue gas (CO, NO_X, SO_X), Ash, Volatilized organics
Solvent regeneration	Medium	Low	Concentrated liquid waste (containing pollutants)
Electrochemical regeneration	Low	High	Electrolyte solution (potentially containing degraded pollutants), electrode corrosion products

3.1. Pollutant Migration Behaviors during Thermal Regeneration

Thermal regeneration is one of the earliest techniques applied for the regeneration of activated carbon saturated with organic pollutants [16–20]. Its advantages include short processing times, high efficiency, and broad applicability, leading to its widespread adoption. The thermal regeneration process for saturated activated carbon typically comprises three stages: (1) Drying stage: Heat is applied to evaporate moisture and some low-boiling-point organic compounds from the activated carbon. In engineering applications, this stage is sometimes supplemented with hot purging to facilitate the volatilization of low-boiling-point organics. (2) Carbonization stage: Within a temperature range of approximately 200–900 °C, large organic molecules are thermally decomposed into smaller organic molecules or inorganic substances such as CO_2 and CO. Organic pollutants that are difficult to desorb undergo carbonization and remain trapped within the pores of the activated carbon. (3) Activation stage: Under conditions of 800–1000 °C, with the presence of gases such as steam or O_2, the remaining carbonized pollutants react, promoting their decomposition or gasification. This process clears the occupied adsorption pores of the activated carbon, leading to the regeneration of its adsorption capacity. In engineering applications, this stage can also be supplemented with purging to promote the volatilization of thermal decomposition products and enhance the thermal regeneration effect.

However, despite its widespread application, thermal regeneration has limitations. The specific surface area and adsorption capacity of activated carbon are compromised to some extent after thermal regeneration. Furthermore, significant carbon loss, reduced mechanical strength, and high energy consumption hinder its further development. Therefore, it is imperative to develop more advanced and efficient thermal regeneration techniques. In recent years, microwave thermal regeneration has attracted research attention due to its advantages of selective and rapid heating [17,18,21–25]. In contrast to conventional heating, microwaves generate heat by causing polar molecules within the sample to undergo directional motion and friction within an alternating electric field [26]. Activated carbon, possessing a high dielectric constant, readily absorbs microwaves, enabling rapid heating under microwave irradiation [27]. Upon reaching a critical temperature, the adsorbate can then be desorbed, as shown in Figure 3a. By establishing a multiphase porous media model

that couples electromagnetic, heat transfer, and mass transfer (as shown in Figure 3b), researchers can study the characteristics of the microwave thermal regeneration process to optimize microwave reactors, potentially reducing energy consumption by 41% and significantly shortening the regeneration time by 86% [28]. In addition, a novel ultrafast method combining high temperature shock and ultrasonic pickling has demonstrated exceptional efficiency in regenerating spent activated carbon, achieving a 104% regeneration rate in just 150 s [29]. Schematic illustration of the ultrafast regeneration of spent activated carbon as shown in Figure 4. Therefore, traditional thermal regeneration methods are no longer sufficient to meet the growing demand, high-efficiency and low-energy thermal regeneration technologies will be an important future development direction.

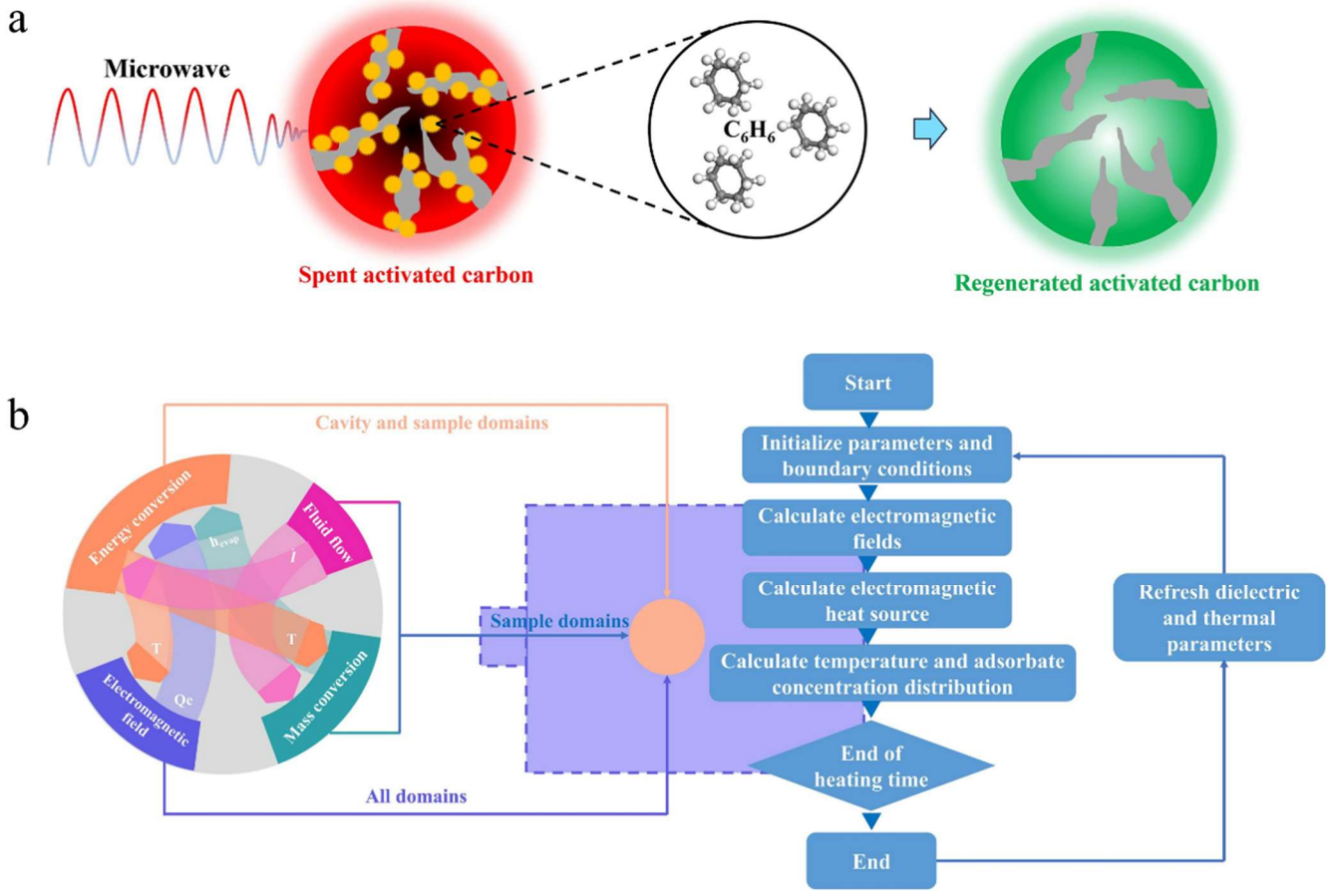

Figure 3. Mechanism of microwave thermal regeneration of spent activated carbon (**a**); schematic diagram of a model of a multiphase porous media coupled with electromagnetism, heat and mass transfer and flowchart of the overall calculations (**b**) [28].

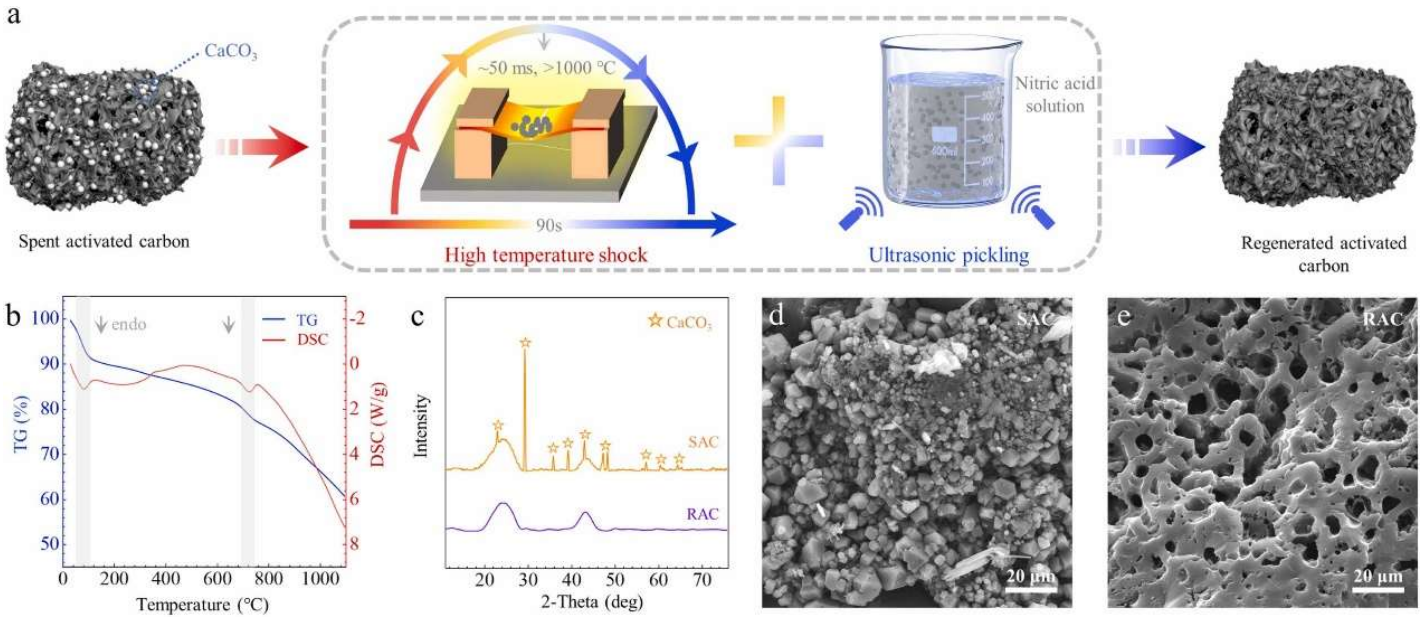

Figure 4. Schematic illustration of the ultrafast regeneration of spent activated carbon (**a**), TG-DSC curves of spent activated carbon (**b**), XRD patterns of spent activated carbon and regenerated activated carbon (**c**); SEM images of spent activated carbon (**d**) and regenerated activated carbon (**e**) [29].

Regarding pollutant migration, as illustrated in Figure 5, the adsorption force on organic pollutants, primarily volatile organic compounds (VOCs), decreases significantly upon heating. Some VOCs become free and migrate to the surface of the activated carbon or directly volatilize. When assisted by a hot gas purge process, the organic pollutants are not only heated but also have increased contact with the gas phase, which promotes their migration into the gas phase. Subsequently, as the temperature further increases, large VOC molecules decompose into smaller organic molecules or inorganic substances such as CO_2. Concurrently, some large VOC molecules are difficult to desorb and remain within the pores of the activated carbon as fixed carbon, leading to a decrease in the adsorption capacity and lifespan of the activated carbon. Furthermore, adsorbed VOCs can sometimes undergo coupling reactions; for example, phenol can lose a proton and transform into a phenoxy group, hindering its desorption. Finally, the VOCs and their decomposition products on the activated carbon surface come into contact with activating agents and undergo oxidation, further converting into non-polluting gases, mainly CO_2 and H_2O [30]. In summary, during the thermal regeneration process, the majority of pollutants decompose and convert into non-polluting substances, migrating into the environmental phase. However, it is worth noting that some VOCs or small organic molecules only migrate and escape from the surface of the activated carbon without complete decomposition, posing a continued environmental pollution risk.

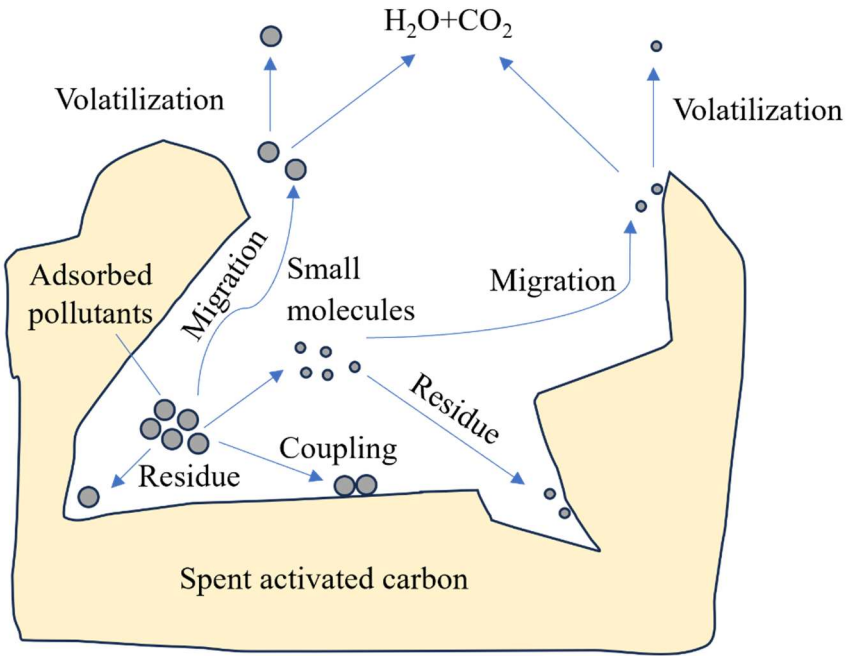

Figure 5. Schematic illustration of pollutant migration pathways during the spent activated carbon thermal regeneration process.

Although the migration and transformation mechanisms of simple pollutants like phenol are becoming clearer, the actual behavior of pollutants is much more complex. If we could clarify the specific relationship between thermal regeneration process conditions and pollutant migration patterns, calculate detailed relational expressions between the two, and summarize and develop theoretical systems based on engineering experience, it would greatly promote the development of spent activated carbon thermal regeneration processes and improve the efficiency ratio (regeneration efficiency per energy input).

3.2. Pollutant Migration Behaviors in Solvent Regeneration

Solvent regeneration is another widely used technique for regenerating spent activated carbon, offering advantages such as a simple process flow, minimal activated carbon loss after regeneration, and the potential for extracting pollutants for resource recovery [31–34]. Based on the reaction mechanism, solvent regeneration can be divided into reactive regeneration and extraction regeneration, or inorganic solvent regeneration and organic solvent regeneration. Reactive regeneration (inorganic solvent regeneration) utilizes inorganic solutions such as acids and bases to chemically react with pollutants adsorbed in the pores of spent activated carbon, transforming them into heavy metal ions or other substances readily soluble in the inorganic solution. This opens the adsorption pores of the activated carbon, restoring its adsorption capacity. Extraction regeneration (organic solvent regeneration) uses solutions with a high extraction capacity for the pollutants adsorbed onto the spent activated carbon (primarily organic pollutants), enhancing the desorption of these adsorbed pollutants from the pores of the spent activated carbon and promoting their migration into the liquid phase, thereby restoring the activated carbon's adsorption capacity. A novel modified supercritical CO_2 ($scCO_2$) extraction method has been developed for the

regeneration of spent activated carbon. By incorporating organic solvents and acid modifiers, this procedure achieved >99% perfluorooctanoic acid (PFOA) desorption after a 60 min treatment in a continuous flow reactor [35]. Figure 6 illustrates potential PFOA desorption pathways. Route I is facilitated by the high dielectric permittivity of $scCO_2$, which enhances proton availability from PFOA dissociation. Route II involves the interaction of CO_2 molecules with water retained in the spent activated carbon pores, leading to bicarbonate ion formation. Route III is predicated on disrupting the electrostatic interactions between spent activated carbon and per- and polyfluoroalkyl substance (PFAS).

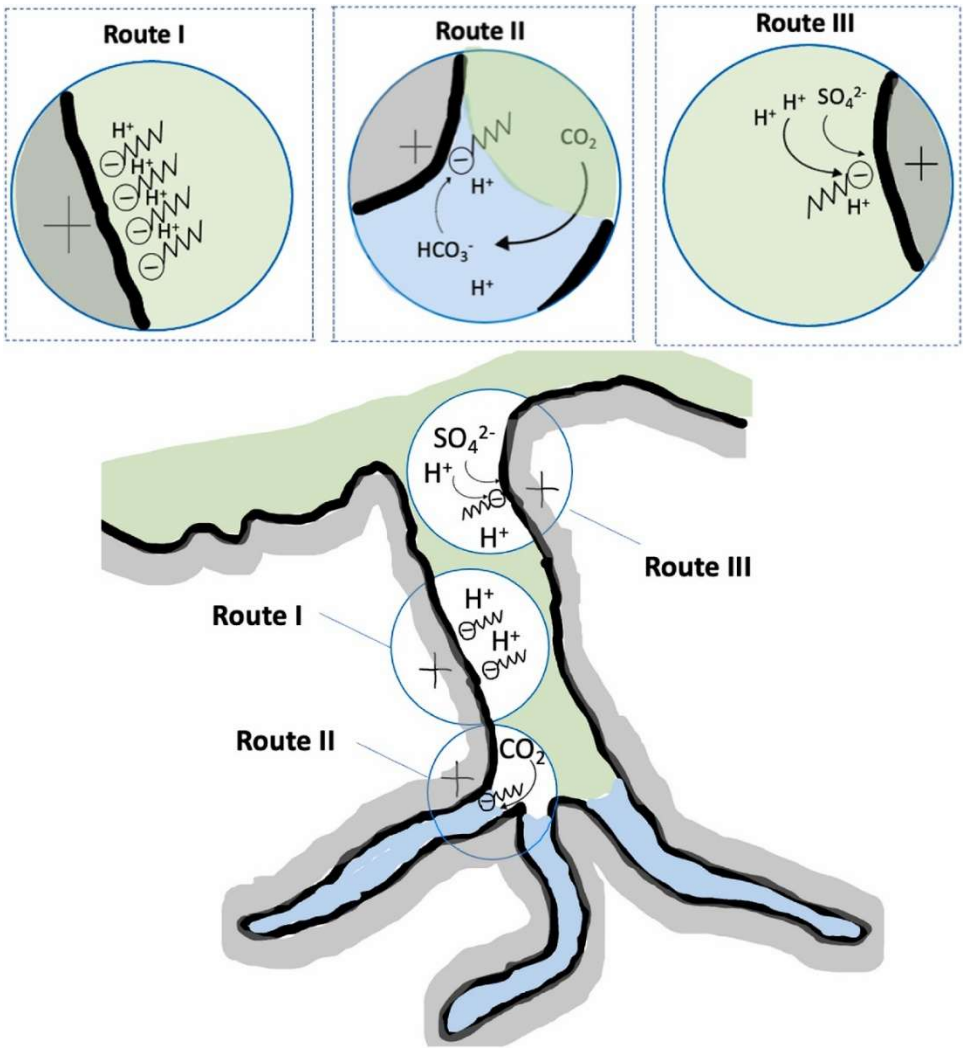

Figure 6. PFOA desorption routes from spent activated carbon, blue-porous water, and green-$scCO_2$. Route I-in pure $scCO_2$, high dielectric permittivity and subsequent proton association lead to partial PFOA protonation; Route II-in pure $scCO_2$, bicarbonate ions formed from CO_2 interaction with retained water compete with PFOA for adsorption sites; Route III-in $scCO_2/MeOH/H_2SO_4$ (i) competition of the sulfate ion for spent activated carbon active site and (ii) PFOA protonation [35].

However, solvent regeneration methods also have certain drawbacks. Studies have shown that residual air inside the pores of the activated carbon hinders the contact between the regeneration solution and the spent activated carbon, reducing the material reaction and migration between the two phases. On the other hand, when

using highly corrosive inorganic regeneration solutions such as strong acids and bases, it can also cause the micropores inside the activated carbon to shrink or collapse, reducing the specific surface area and permanently decreasing the adsorption capacity and efficiency of the activated carbon. In summary, existing solvent regeneration methods also have significant limitations and cannot meet current application demands. Exploring more superior activated carbon regeneration technologies is necessary.

The solvent regeneration process exhibits good regeneration effectiveness for activated carbon adsorbed with both organic pollutants like VOCs and inorganic pollutants, such as heavy metals. During the regeneration process, heavy metal pollutants react with the solution (e.g., acidic solution), transforming into metal ions and migrating into the liquid phase of the solution. For organic pollutants like VOCs, they can also react with the solution or dissolve into the solution phase, thereby achieving the migration of pollutants from the activated carbon surface to the solution phase.

However, the pollutants do not stabilize or decompose to eliminate pollution once they enter the solution phase. Instead, they form concentrated waste liquids with high pollutant concentrations. The treatment of concentrated waste liquids generated during spent activated carbon regeneration is a critical aspect of ensuring the overall sustainability of activated carbon technology. Membrane separation processes and advanced oxidation processes offer promising solutions for managing these complex waste streams, and integrated approaches hold the greatest potential for achieving effective and cost-efficient treatment. Future research should focus on optimizing these treatment strategies and developing novel technologies for minimizing the generation of concentrated waste liquid.

3.3. Pollutant Migration Behaviors in Electrochemical Regeneration

Electrochemical regeneration is a promising emerging technology for regenerating spent activated carbon [36–41]. This method involves placing the spent activated carbon in a specific liquid, and under the influence of an electric current, redox reactions, acid-base neutralization, and other processes alter the adsorption-desorption equilibrium of the pollutants. This separates the harmful substances attached to the activated carbon, thus achieving regeneration [42,43]. Studies on the regeneration mechanism of spent activated carbon suggest that applying an electric current across an exhausted bed of spent activated carbon leads to two main regenerative processes [44]. First, enhanced desorption commences from the spent activated carbon surface, as illustrated in Figure 7a–c, resulting in an adsorbent free of contaminant species. Second, electrochemical reactions occurring at the electrodes and polarized spent activated carbon particles stimulate the degradation of contaminant species, completely removing them from the system, as shown in Figure 7d. An ideal electrochemical regenerative process would promote both mechanisms, thereby avoiding further treatment of the desorbed compounds.

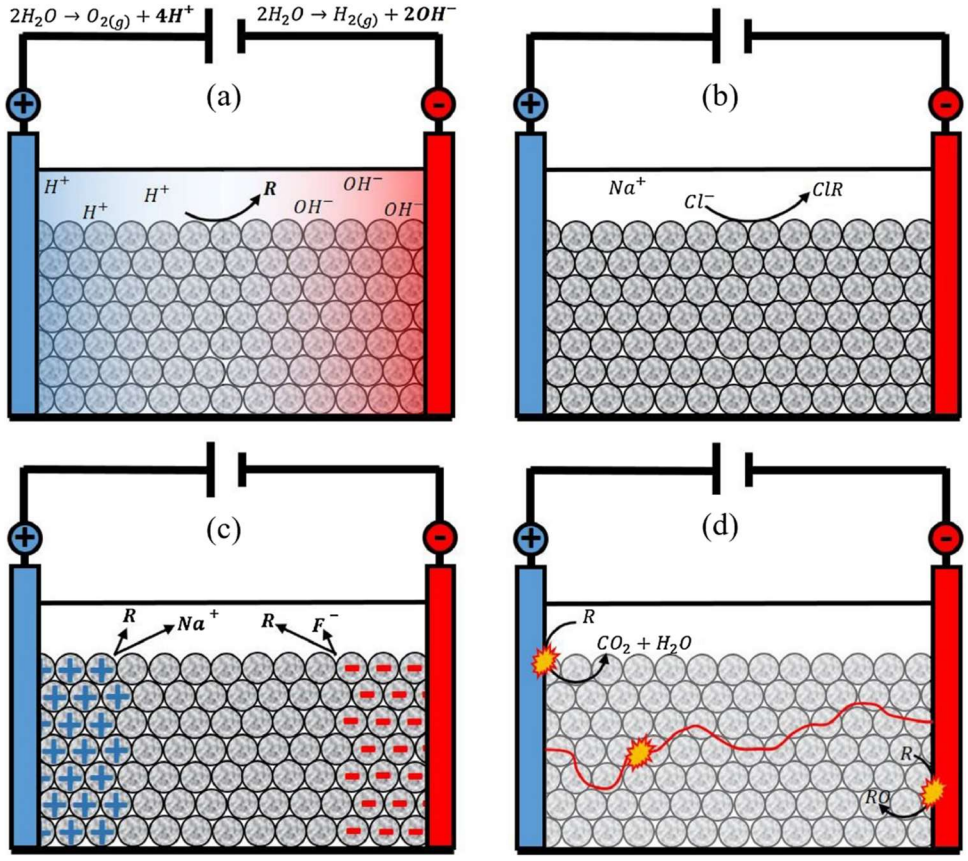

Figure 7. Regenerative mechanisms involved in the electrochemical regeneration of spent activated carbon: enhanced desorption due to changes in local pH (**a**); enhanced desorption due to local changes in salinity concentration, or adsorbate reacting with ionized species to produce a less readily adsorbed compound (**b**); electro desorption whereby species are repelled from the charged spent activated carbon surface (**c**); oxidation/degradation reactions occurring at either the electrodes or polarized spent activated carbon (**d**) particles [44].

As early as the 1990s, a simple current of 50 mA could restore more than 90% of the adsorption capacity of spent activated carbon saturated with phenol [45]. After decades of development, the degradation rate and effectiveness have been further improved. For example, when using sodium chloride solution to electrochemically regenerate activated carbon fiber felt loaded with potassium dichromate, the regeneration rate can reach up to 100% [46]. This indicates that electrochemical regeneration of spent activated carbon has the advantage of high regeneration efficiency.

The electrochemical regeneration process is influenced by various factors. Studies have shown that the magnitude of the current affects the change in Coulombic interactions between the pollutants and spent activated carbon. A certain current can enhance the oxidation efficiency of the pollutants, but a stronger current may also lead to side reactions such as hydrogen evolution. The regeneration efficiency differs between the electrolyte and the electrode surface; in related studies [47], the regeneration efficiency on the electrode surface was approximately 20% higher. The material properties of the cathode and anode also affect the electrochemical regeneration process. For example, inert electrodes such as lead dioxide have higher physical and chemical stability, making them less prone to side reactions [48]. In

addition, the properties of the electrolyte, the type of pollutants, and other characteristics also affect the effectiveness of electrochemical regeneration technology.

Regarding pollutant migration behaviors, in the aforementioned electrochemical process, the pollutants adsorbed by the spent activated carbon are subjected to Coulombic forces, and their adsorption-desorption equilibrium is disrupted. This causes them to migrate from the solid phase of the spent activated carbon to the liquid phase of the desorption solution [49]. However, in traditional electrochemical regeneration processes, this pollutant migration process is not accompanied by pollutant transformation, and the hazardous nature of the pollutants remains. Therefore, combined electrochemical and advanced oxidation technologies have emerged, leading to the development of integrated technologies such as electro-Fenton regeneration and electro-activated persulfate regeneration. Zhan et al. used electrochemical technology to regenerate spent activated carbon adsorbed with p-nitrophenol, achieving 98% regeneration efficiency and effectively mineralizing the intermediate products [50]. The electro-permanganate system presents a promising activated carbon regeneration strategy, characterized by a 93.11% reduction in energy consumption compared to electrochemical regeneration and a high regeneration efficiency [51]. Ding et al. used electro-activated persulfate oxidation technology to regenerate spent activated carbon adsorbed with phenol, achieving complete mineralization of 81.9% of the pollutants [52]. Liu et al. elucidated the mechanism of electro (E)-activated carbon fiber (ACF)-peroxydisulfate (PDS), as shown in Figure 8. The E-ACF-PDS process achieves remarkable carbamazepine removal from aqueous solution through the synergistic mechanisms of pollutant adsorption and in situ generation of active radicals on the ACF cathode. The electron-rich environment at the cathode minimizes oxidative damage to the ACF by sulfate radicals, hydroxyl radicals, and PDS, while simultaneously promoting continuous sulfate radical production, independent of the electron-donating ability of the activated carbon itself [53].

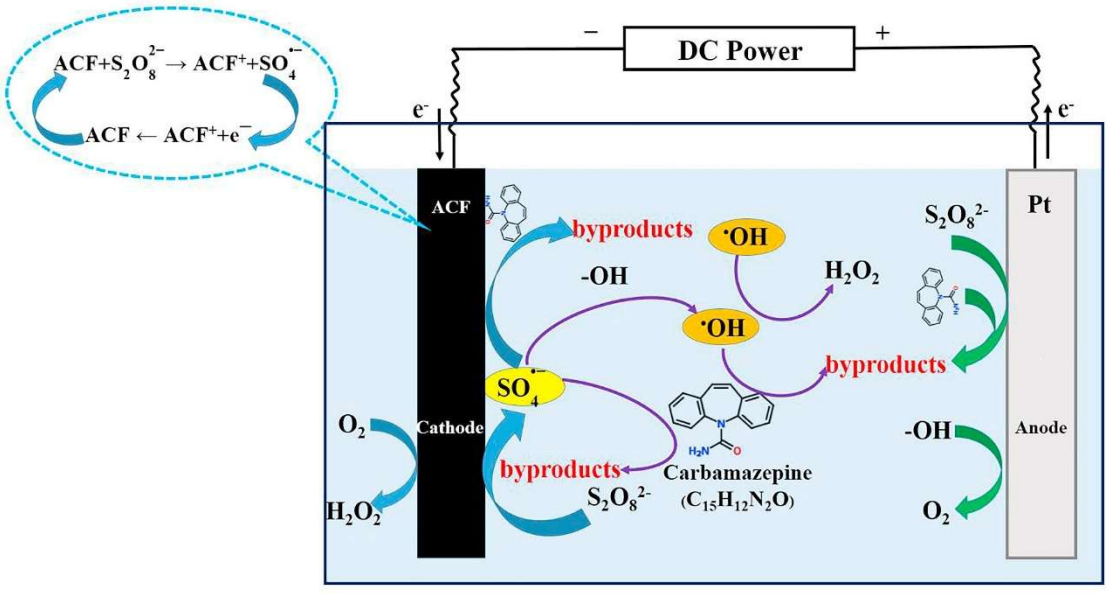

Figure 8. Proposed mechanism in E-ACF-PDS process [53].

In comparison, electrochemical technologies combined with advanced oxidation processes are relatively expensive. Although they achieve spent activated carbon regeneration, the large number of free radicals generated are not fully utilized, resulting in a low efficiency ratio. Further leveraging the role of advanced oxidation technologies is key to improving the efficiency ratio. On this basis, if the various wastewaters generated in co-treatment engineering are also considered for synergistic treatment, their application will be even more widespread.

4. Future Development and Prospects

Improving the efficiency ratio is one of the most important development directions for spent activated carbon regeneration. In thermal regeneration, it is necessary to clarify the specific relationship between thermal regeneration process conditions and pollutant migration patterns, and to summarize and develop theoretical systems based on engineering experience to improve the efficiency ratio. In solvent regeneration, the treatment and utilization of concentrated waste liquid byproducts is key to improving the efficiency ratio. Electrochemical regeneration is often combined with technologies such as advanced oxidation processes. Simultaneously regenerating spent activated carbon while considering the synergistic treatment of self-produced wastewater from the engineering process may be an important area of research in the future.

The complexity of spent activated carbon regeneration processes, involving numerous interacting parameters, presents a significant challenge for traditional optimization methods. These methods are often time-consuming, resource-intensive, and may fail to identify the global optimum operating conditions. Artificial intelligence, particularly machine learning algorithms, offers a powerful alternative for optimizing these complex processes. The application of artificial intelligence to optimize regeneration parameters will be a crucial area of investigation in the field.

The effectiveness of many regeneration methods, particularly chemical and thermal processes, can be significantly enhanced by the incorporation of catalysts. Conventional catalysts, however, may suffer from limitations such as high cost, limited activity, and environmental toxicity. Emerging classes of catalysts, such as single-atom catalysts, offer significant potential for overcoming these limitations and achieving more efficient and sustainable pollutant degradation during spent activated carbon regeneration. Therefore, the exploration and development of novel catalytic materials, especially single-atom catalysts, for enhanced pollutant degradation during spent activated carbon regeneration is an important future development.

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