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# Reusability of spent adsorbents for a circular materials economy in the sustainable chemical industry

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Circular adsorption systems, particularly the management of spent adsorbents, are reaching a pivotal stage in industrial adoption and large-scale implementation. Simultaneously, the production and scaling of spent adsorbents are increasingly aligning with commodity applications. However, the prevailing approach to spent adsorbents at the end of their lifecycle primarily focuses on disposal or recycling to mitigate secondary pollution. A more economically favourable alternative involves prioritizing efficient reprocessing and recycling over disposal. In this context, the review underscores the decisive role of cost management in both the synthesis and regeneration of adsorbents. The synthesis stage has a strategic and multifaceted impact on adsorption performance, with several parameters, either individually or jointly, exerting a direct influence on cost. Key economic determinants include preparation and modification expenses, process complexity, and overall yield, all of which are essential in assessing the feasibility of adsorbent technologies. The review also combines scientific and strategic perspectives by grouping adsorbents according to their synthesis and regeneration cost profiles. Materials such as graphene oxide, silica, carbon nanotubes, and MOF-based composites fall into the high-cost category due to their costly production and regeneration requirements. In contrast, agricultural waste-based adsorbents emerge as a cost-effective solution, offering low synthesis and regeneration costs. Although composites hold strong potential, their high cost remains a major obstacle to large-scale implementation. On top of that, we present strong economic incentives for adopting spent adsorbent reclamation over alternative pathways. The sustainable management of spent adsorbents—including recovery and regeneration processes—is reviewed through the lens of the circular economy.

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## Sustainability spotlight

In terms of sustainability, recent studies have focused on methods for regenerating adsorbents, such as direct desorption and converting spent adsorbents into new materials, with minimal treatment between uses to ensure a cost-effective and sustainable approach. Reusing spent adsorbents can be environmentally beneficial and help reduce overall costs, but the regeneration process often involves complex procedures that increase operational costs and energy consumption, limiting their sustainability. Cost, controllability, and scalability are significant challenges for the practical use of multifunctional adsorbents, especially since their synthesis can be complex, and they often exist at the nanoscale. The strategic role of synthesis in the adsorption evaluation equation is critical. In this review we present strong economic incentives for adopting spent adsorbent reclamation over alternative pathways. This works aligns with UN SDG 12 and SDG 13.

## 1. Introduction

Material circularity is widely regarded as a top priority within the research community,<sup>1</sup> driven by increasing demands to mitigate the environmental, climate, and energy impacts associated with adsorbent production and consumption. However, the persistent nature of used adsorbents prevents effective

closed-loop recycling, often leading to their disposal. This disposal process introduces secondary pollution from both the contaminants adsorbed and the chemicals used in treatment, with improper disposal further exacerbating environmental harm. To achieve a truly circular, net-zero materials economy, sustainable practices must be implemented at both the production and disposal stages of an adsorbent's lifecycle.<sup>2</sup> A key advantage of adsorbents lies in their versatility in synthesis. While some are used in their natural state, recent trends favor modified adsorbents with enhanced properties. Functionalization and the integration of adsorbents into composite materials

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improve their adsorption capabilities. In some cases, adsorbents serve as scaffolds for the development of advanced materials with tailored functional groups, following an adsorbent1@adsorbent2@adsorbent3 structure.

Adsorption efficiency is largely influenced by the dominant functional groups present on the surface and within the pores of an adsorbent.<sup>3</sup> The future may see the emergence of even more complex grafted structures, such as adsorbent1@adsorbent2@adsorbent3@adsorbent4 composites. However, while adsorption is traditionally considered a simple and cost-effective method, the increasing complexity and expense of chemical modifications raise an important question: can adsorption still be advocated as an affordable and straightforward approach in light of these advancements?

The cost of adsorbents is a critical factor that warrants careful consideration, as several compelling arguments highlight its significance. The high costs of recovery and regeneration processes can significantly affect the long-term viability of reusing spent adsorbents.<sup>4</sup> Given that the American Chemistry Council (ACC) introduced Economic Elements of Chemistry as a key resource for understanding the chemical industry's economic influence,<sup>5</sup> it is essential to recognize that cost is a fundamental aspect of any technology. However, these expenses can often be justified by performance. Additionally, the abundance, affordability, and diverse functional groups found in agro-based by-products have drawn scientific interest in their potential for pollutant removal from water.<sup>4</sup>

Spent adsorbents—the solid waste remaining after adsorbate has been recovered or regenerated from an adsorbent<sup>6</sup>—have gained significant attention in recent decades as they offer opportunities to advance a circular materials economy.<sup>7–9</sup> As demand for spent adsorbents grows, sustainability principles suggest that these materials should be recycled,<sup>10</sup> with reuse emerging as a viable strategy to address both disposal challenges and environmental concerns. Some spent adsorbents may even become key contributors to a future bio-based and circular economy. Given the limitations of current waste management systems,<sup>11</sup> exploring multiple reuse and recycling pathways for spent adsorbents is crucial. Ideally, these materials should be regenerated for reuse, minimizing disposal needs while conserving resources. Recycling can further extend their lifecycle by repurposing spent adsorbents or their components for diverse industrial applications. This perspective offers a comprehensive discussion on the essential role of spent adsorbents in advancing circular economy principles.

One of the key challenges in evaluating adsorbents is the limited number of review studies<sup>12–14</sup> that specifically analyze groups of adsorbents in terms of their adsorption, desorption, and regeneration properties under defined conditions from an economic perspective. However, none of these studies have systematically categorized adsorbents based on cost, distinguishing between high- and low-cost options. The high cost of certain adsorbents raises concerns among stakeholders regarding the development and adoption of new technologies, particularly given potential fluctuations in price and availability. Each available method has distinct characteristics in terms of balancing cost-effectiveness and performance while

maintaining scalability. The choice of adsorbent ultimately depends on whether high performance is prioritized or if cost reduction is the primary objective. To bridge this gap, this review aims to consolidate the extensive and fragmented literature, extracting key insights on the regeneration potential and economic viability of various adsorbents. The goal is to provide a comprehensive and comparative analysis of different adsorbent groups based on their economic factors, performance, and reusability. A major limitation in existing research is the lack of standardized methodologies for assessing the economic impact of spent adsorbents, which can lead to misleading conclusions and unsustainable solutions. Cost estimation is often overlooked, likely due to the absence of a unified framework for conducting such evaluations. Despite numerous studies focusing on the technical performance of spent adsorbents, this review represents the first comprehensive assessment of their economic aspects, offering a novel perspective on the subject.

This study aims to compare the cost–regeneration profiles of various adsorbent groups, identify the most promising options, and provide a comprehensive assessment for reliable evaluation. It updates existing literature with a comparative analysis of spent adsorbent reuse, focusing on MOFs, graphene, carbon nanotubes, activated carbons, clays, polymers, zeolites, alginate, lignin and chitosan-based materials. Key factors influencing regeneration, desorption efficiency, and post-regeneration performance are examined, with adsorption data presented in tables for easy comparison. Section 2 presents the adsorption evolution and classifies adsorbents based on economic factors, aiding researchers and industry professionals in selecting optimal materials. Moreover, it evaluates standalone and composite adsorbents towards enhanced adsorption performance and discusses regeneration methods and their merits and shortcomings. Section 3 highlights the determinants that governs synthesis and regeneration cost and reports on the economic returns by evaluating the regeneration potential of high-cost adsorbents and low-cost adsorbents. Section 4 analyzes regeneration studies and Section 5 discusses the feasibility of sequential reuse (recycling) of spent adsorbents, as a sustainable and cost-effective strategy. Section 6 underscores the importance of sustainability and life cycle assessment in the adsorbent agenda. Finally, Section 7 provides key conclusions based on the study's findings.

## 2. Adsorption technology for a circular materials economy

### 2.1. Adsorption evolution

A key factor in applying adsorption technology effectively is creating an integrated system that combines adsorption, desorption, regeneration, and contaminant recovery to ensure sustainability and efficiency.<sup>15</sup> Desorption is essentially the reverse of adsorption, where adsorbates are released from the adsorbent surface either by ion exchange with a higher-affinity ion or through chemical interactions with the eluent.<sup>16</sup> The desorption and regeneration of adsorbents play a vital role in determining the economic viability of water treatment



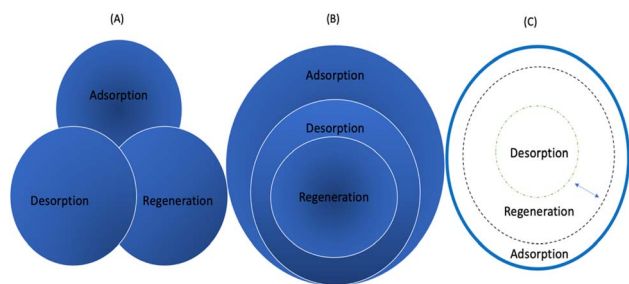


Fig. 1 Evolution of an adsorption system involves: (A) recognizing regeneration, adsorption and desorption as equally essential, (B) embedding regeneration as a key element within the desorption process, both fundamentally linked to adsorption, and (C) redefining regeneration as a resource-reuse strategy that enhances both environmental and economic sustainability while supporting long-term performance through multiple adsorption–desorption cycles.

technologies.<sup>2</sup> Fig. 1 illustrates how perspectives on adsorption systems within the circular economy have evolved through three key viewpoints.

## 2.2. Adsorbents market study confirms reusability to create a circular economy

The global adsorbents market was valued at USD 5470.1 million in 2023 and is expected to grow by 5.1% year-over-year, reaching USD 5751.1 million in 2024. With a projected CAGR of 5.8% (2024–2034), the market is estimated to reach USD 10 102 million by 2034, reflecting a 1.8-fold increase from current levels.<sup>17</sup> The rising demand is driven by global efforts to enhance water and air purification, ensuring clean water access and improved air quality. The industry is increasingly prioritizing recyclability and reusability, fostering a circular economy. Advancements in polymeric adsorptive materials are enhancing recyclability and multiple-use cycles, supporting sustainable operations with lower resource consumption through cradle-to-cradle material regeneration.

## 2.3. Beyond single use: the neglected aspect of adsorbent reuse and material reutilization after adsorption

In this vein, a recent study by Gkika *et al.* highlighted that, in the context of adsorption, regenerated adsorbents can serve as a strategic resource for reuse, contributing to both economic and environmental sustainability.<sup>18</sup> Regeneration and recycling are consistent with circular economy principles, which focus on reusing, remanufacturing, and recycling materials to preserve their value throughout their entire lifecycle. Collectively, these strategies significantly advance sustainable waste management.<sup>19</sup>

However, despite their potential, only a limited number of studies in the past four years have focused on reutilization of materials after adsorption. Arun V. Baskar *et al.* discussed sustainable spent adsorbent management by examining processes related to their recovery and regeneration for reuse within the framework of resource recovery and circular economy principles.<sup>9</sup> K. Mohanrasu *et al.* emphasized the

significance of reusing spent adsorbents for various applications.<sup>20</sup> Y. B. Nthwane and colleagues investigated their revalorization for blood fingerprint applications, demonstrating a dual-purpose use that connects environmental remediation with advancements in forensic science.<sup>21</sup> Anka Jevremović and collaborators explored the emerging field of reusing spent adsorbents in electrochemical devices.<sup>22</sup> Moreover, Muhammad Faheem and colleagues provided an in-depth review that combines environmentally friendly regeneration techniques for smart adsorbents with the sequential recycling of conventional spent adsorbents into high-value products.<sup>19</sup>

## 2.4. Bibliometric section

To understand the current trends and emerging directions in the reuse of spent adsorbents through a circular economy perspective, the literature was retrieved and analysed using the Scopus database. For the automated search strategy, Scopus was selected as the primary scientific database because of its broad coverage across diverse scientific disciplines and its availability of systematic search tools.<sup>23,24</sup> The final search query used was (“spent adsorbents” and “reuse” and “circular”). This query was applied to titles, abstracts, and keywords of publications dated from January 1, 2015, to October 31, 2025, with data retrieved on November 4, 2025. Inclusion criteria were based on metadata provided by Scopus. Eligible studies were (i) full research articles, review papers, conference papers, book chapters, and books, excluding short surveys, (ii) written in English, and (iii) published within the examined period (2015 to 2025). The search yielded a total of seven relevant records.

The reuse of spent adsorbents within a circular materials economy framework has not yet received extensive scientific attention over the past decade, which demonstrates that this field remains underdeveloped and requires further investigation. Nevertheless, several positive insights can be drawn from the bibliometric findings. The presence of multi-authored publications suggests ongoing collaborative research efforts. Furthermore, the topic is disseminated across multiple journals and spans diverse subject areas, including analytical chemistry, filtration and separation, pollution, management, monitoring, policy and law, general environmental science, general chemical engineering, waste management and disposal, and environmental chemistry. This distribution highlights the multidisciplinary nature of the field and the broad range of themes associated with the circular reuse of spent adsorbents.

In the coming years, it will become clear whether this emerging upward trend in publication activity will persist.

## 2.5. Classification of adsorbents

A wide range of adsorbents has been studied for their effectiveness in removing different pollutants from water and wastewater.

Research on adsorbents has expanded rapidly in recent decades, becoming a major field of scientific interest. Adsorbents encompass a wide range of microstructures, adsorption capacities, and formats, including synthetic polymers, nano-materials, biomaterials, and waste-derived materials.<sup>25</sup> This



diversity makes it challenging to establish a consistent and comprehensive classification system, particularly given the variety of criteria and conditions that influence classification approaches. The significance of structured classification was highlighted by Gkika *et al.*,<sup>26</sup> who demonstrated that grouping adsorbents (i) enabled clear comparisons of cost–regeneration profiles, (ii) helped identify the most efficient option within each group, and (iii) provided detailed group-level information that enhanced the reliability of individual adsorbent evaluations. Multiple classification frameworks have been proposed. Leandro Pellenz categorized adsorbents as organic, inorganic, or hybrid materials, further distinguished by particle size (nano or micro) and membrane form.<sup>27</sup> Crini *et al.* proposed a simplified scheme dividing adsorbents into conventional and non-conventional categories.<sup>28</sup> Wai Siong Chai and collaborators further distinguished between conventional adsorbents and novel nanostructured materials.<sup>29</sup>

In addition to these structural or material-based categories, sustainable design principles must be considered to support long-term adsorbent performance. Circular economy strategies aim to develop sustainable models aligned with the UN Sustainable Development Goals, ensuring that adsorbent design provides both environmental and economic benefits. In this context, Steiger *et al.* classified adsorbents as single-component or composite systems,<sup>30</sup> while Faheem *et al.* proposed a classification based on spent and smart adsorbents. Smart adsorbents are engineered for integration into different systems, offering tunable properties, high pollutant retention, and reusability after regeneration. Their adaptive nature allows them to respond to variations in pH, temperature, ionic strength, magnetic fields, or light, extending their lifespan and reducing reliance on hazardous solvents.<sup>19</sup> Finally, adsorbents can also be grouped according to their cost profiles, ranging from high to low, enabling clearer economic evaluation alongside technical performance.<sup>13,26,31,32</sup> An ideal adsorbent is eco-friendly, low-cost, and highly efficient, with strong mechanical properties, high surface area, good selectivity, and reusability, making it suitable for industrial-scale use.<sup>33</sup> Table 1 presents various adsorbent's performance/cost ratio profiles classified according to material type.

## 2.6. Comparative evaluation of standalone and composite adsorbents: toward enhanced adsorption performance

Various types of adsorbents have demonstrated strong potential in removing harmful pollutants from wastewater. These materials differ in accessibility, cost-effectiveness, regenerative capacity, environmental impact, and the extent to which they can be derived from sustainable sources. Nanomaterials can be used either as standalone adsorbents or as essential building blocks in the development of composite adsorbents.<sup>61</sup> Numerous studies have explored the influence of both single-component and composite adsorbents on adsorption capacity.<sup>78</sup>

**2.6.1 Standalone adsorbents.** Standalone or single-component systems (such as zeolites, cellulose, and chitosan) consist of a single material, with or without chemical modification.<sup>30</sup> A large body of research indicates that specific surface

area and structural characteristics are key factors that give composite adsorbents a performance advantage over single-material systems. Consequently, there is growing interest in developing composite adsorbents with optimized structures and large surface areas using simple, scalable preparation methods.<sup>78</sup> Several standalone adsorbents show promise for water treatment. Biochar has gained attention as a sustainable and cost-effective option with strong environmental compatibility, especially for industrial wastewater in dye-intensive sectors.<sup>79</sup> Several scholars have also leveraged graphene oxide<sup>80,81</sup> and silica,<sup>82</sup> which have been effectively applied as single materials. However, certain natural adsorbents face performance limitations when used alone. Natural clays may exhibit low efficiency;<sup>83</sup> chitosan suffers from low adsorption capacity, poor thermal stability, and weak mechanical strength;<sup>84</sup> and nanocellulose is often not economical as a standalone option.<sup>85</sup> These limitations can lead to reduced adsorption efficiency in practical applications. When single-material sorbents face issues such as structural degradation or surface fouling, composite systems provide enhanced stability and durability by introducing complementary functionalities.<sup>86</sup>

**2.6.2 Composite adsorbents.** The integration of different materials through chemical modification or physical blending embodies the principle that “the whole is greater than the sum of its parts”. Synergistic and additive effects within composite structures often result in significantly improved adsorption performance compared to what would be expected from the individual components alone.<sup>30,87</sup> Composites, especially polymer-based systems, represent a more advanced alternative, offering improved mechanical stability, durability, and higher adsorption capacity compared to single adsorbents.<sup>88</sup> In these materials, one component typically serves as a filler or matrix, often derived from natural polymers (*e.g.*, plant fibers and biopolymers), while inorganic or organic materials act as binders to enhance pollutant removal.<sup>89</sup> For example, graphene-based composites combined with biopolymers such as chitosan, alginate, or cellulose demonstrate significantly higher adsorption capacities, largely due to an increased number of active sites for pollutant interactions. Graphene also improves the mechanical strength and reusability of the adsorbent, enabling multiple adsorption–desorption cycles. Similarly, carbon nanotubes contribute flexibility and strength, enhancing the material's durability without significant performance loss. Hybrid composites made from nanofibers and biopolymers offer increased adsorption rates, benefiting from the high surface area of the nanofibers. Metal oxide nanocomposites provide high surface reactivity, complementing the adsorption capabilities of biopolymers and further boosting performance.<sup>90</sup>

**2.6.3 The role of components in composite equation.** Nanocomposites are materials made of two or more components with different properties, where the matrix is the main phase. They feature small reinforcing nanomaterials with high surface area and aspect ratio, well dispersed in the matrix, and can appear as particles, sheets, or fibers. These properties provide significant advantages: using only small amounts of



Table 1 Adsorbent's performance/cost ratio profile

| Adsorbent                   | Economic elements   |   | Adsorption properties/<br>performance   |
|-----------------------------|---|---|---|
|                             | Regeneration cost   | Synthesis cost  |   |
| <b>Carbon-based</b>         |   |   |   |
| Commercial activated carbon | High cost and the difficulties involved in its regeneration <sup>34</sup> | The abundant availability of raw materials helps to lower production costs <sup>35</sup>                                | The outstanding performance can be ascribed to two key factors: (i) its high surface area stemming from a highly porous structure and (ii) the presence of numerous polar functional groups <sup>36</sup>   |
| CNTs                        | High regeneration cost <sup>37</sup>                                      | Complex preparation processes, typically increase preparation costs <sup>38</sup>                                       | High specific surface area and tubular structure. <sup>39</sup> However, CNT-supported catalysis faces challenges with catalyst regeneration. <sup>40</sup> Additionally, single-walled CNTs exhibit a higher adsorption capacity than multi-walled CNTs, as MWCNTs often experience purification issues that compromise their active sites <sup>41</sup> |
| Graphene oxide              | High regeneration cost <sup>42</sup>                                      | The high production cost significantly limits its use in practical water treatment <sup>43</sup>                        | GO exhibits excellent adsorption properties. <sup>44</sup> Both GO and reduced graphene oxide are increasingly used, thanks to hydroxyl, carboxyl, and epoxy groups, which play a crucial role in binding metal ions. <sup>45</sup> However, when used in their pristine form, GO and RGO tend to restack and agglomerate <sup>46</sup>                   |
| Carbon xerogels             | —   | Lengthy synthesis processes. Ongoing research aims to reduce synthesis time and decrease production costs <sup>47</sup> | The adsorption capacity and selectivity of xerogels can be improved by functionalizing their surfaces with groups such as carboxyl, hydroxyl, or amine. Their exceptional pollutant uptake and rapid adsorption rates are mainly due to their tunable porosity and large surface area <sup>48</sup>   |
| 3d printed adsorbents       | —   | Activated carbon monolithic adsorbents have low synthesis costs <sup>49</sup>   | 3D realize complex structures that are difficult for traditional molding methods <sup>50</sup>  |
| <b>Mineral-based</b>        |   |   |   |
| Clay-based adsorbents       | Poor recyclability and high regeneration costs <sup>40</sup>              | Low production cost <sup>51</sup>   | Clays exhibit high adsorption efficiency due to their net negative charge and large surface area. <sup>42</sup> However, they face challenges in regeneration through desorption and pH control <sup>52</sup>   |
| Zeolites                    | High regeneration cost <sup>53</sup>                                      | The preparation of synthetic zeolites is costly <sup>54</sup>   | Zeolites offer excellent ion exchange properties, a high surface area, and a hydrophilic character, making them effective for the removal of metals <sup>52</sup>   |
| Silica-based adsorbents     | Regeneration costs have hindered the widespread use <sup>55</sup>         | High manufacturing cost <sup>55</sup><br>Complicated synthesis high cost of reagents <sup>56</sup>                      | Mesoporous silica materials, characterized by their high specific surface area, well-defined pore size, and large pore volume, are used as supports in wastewater treatment. <sup>57</sup> High recoveries <sup>4</sup>   |



Table 1 (Contd.)

| Adsorbent  | Economic elements                    |   | Adsorption properties/<br>performance   |
|--|--------------------------------------|---|---|
|  | Regeneration cost                    | Synthesis cost  |   |
| <b>Nanomaterials</b>                             |                                      |   |   |
| Hydroxyapatite nanoparticles                     | —                                    | Large-scale production requires considerable amounts of chemicals, leading to high economic costs <sup>58</sup> | The adsorption efficiency of these nanoparticles is closely linked to their surface functional groups. These nanoparticles have shown remarkable effectiveness in removing heavy metals <sup>59</sup>   |
| <b>Metal organic frameworks (MOFs)</b>           |                                      |   |   |
| MOFs   | High regeneration cost <sup>60</sup> | Involve higher production costs due to their complex synthesis procedures <sup>61</sup>                         | Large surface areas, high scalability, and highly ordered porous structures. Their tunable physicochemical properties and adaptability allow them to outperform many conventional adsorbents. A key advantage of MOFs is their ability to maintain structural stability in challenging environments <sup>62</sup> |
| <b>Waste (agricultural/industry)-based</b>       |                                      |   |   |
| Agricultural waste-based adsorbent               | Low regeneration cost <sup>63</sup>  | Low synthesis cost <sup>63</sup>  | Low surface area but abundant functional groups. The removal efficiency is low <sup>4</sup>   |
| Adsorbents from stainless steel slag             | —                                    | High costs associated with their post-treatment processes <sup>64</sup>   | Limited specific surface area. <sup>65</sup> To enhance their performance, slag can be modified by introducing functional groups <sup>66</sup>  |
| <b>Polymer-based</b>                             |                                      |   |   |
| Porous organic polymers (POPs)                   | —                                    | High synthesis cost <sup>66</sup>   | Calixarene-based polymers create adsorbents with strong selectivity and high adsorption capacity toward targeted pollutants <sup>66</sup>   |
| Calixarene-based polymers                        | —                                    | Synthesis cost is low <sup>67</sup>   | After the template molecule is removed, memory regions are formed within the material, enabling it to selectively recognize and rebind the original template from complex mixtures, even under harsh physical and chemical conditions, while maintaining high stability <sup>68</sup>                             |
| Synthetic polymers                               | —                                    |   |   |
| Molecularly imprinted polymers (MIPs) adsorbents | —                                    |   |   |
| Biopolymers                                      | —                                    | Low synthesis cost <sup>69</sup>  | It has functional groups such as phenolic, aliphatic hydroxyl, and carboxylic groups that enable dye binding through ion exchange or complex formation <sup>70</sup>  |
| Lignin-based adsorbents                          | —                                    |   |   |
| Chitosan   | —                                    | Low synthesis cost <sup>71</sup>  | Chitosan's primary amine group facilitates strong electrostatic interactions between the amine groups and dye molecules, ensuring effective sorption. <sup>72</sup> However, chitosan has some drawbacks such as controlling its pore size <sup>52</sup>  |



Table 1 (Contd.)

| Adsorbent   | Economic elements |  | Adsorption properties/<br>performance  |
|---|-------------------|--|--|
|   | Regeneration cost | Synthesis cost                                 |  |
| <b>Composites</b><br>Metal oxide<br>composite<br>adsorbents | —                 | Very expensive synthesis cost <sup>73,74</sup> | Composite metal oxides exert synergistic effects of multiple metals <sup>75</sup><br>Graphene-metal oxide composites are widely favored for their well-controlled morphology, large specific surface area, versatile surface chemistry, strong adsorption capacity, abundant oxygen-containing functional groups, and notable catalytic activity <sup>76</sup> |
| MOF-biochar<br>composite<br>adsorbents                      | —                 | High costs <sup>77</sup>                       | The adsorption capacity of composites is typically twice that of standalone biochar, while MOFs retain their crystallinity even after multiple regeneration cycles, demonstrating the composites' durability and long-term potential for sustainable use <sup>77</sup>   |

lightweight nanofillers with size-dependent behavior can enhance the electrical, thermal, mechanical, chemical, optical, and magnetic performance of the composite material. A wide range of nanoparticles has been utilized in nanocomposite fabrication.<sup>91</sup>

The components within composites can interact through covalent or noncovalent associations. Noncovalent associations arise through physical blending, while covalent associations involve chemical bonding between additive components. The resulting benefits of combining two or more components are multifaceted, including cost reduction and synergistic improvements in surface chemistry, textural properties, and electronic structure. Such enhancements often result in overall effects that surpass the sum of the individual components. Composite materials can be based on inorganic, organic–inorganic hybrid, natural biopolymer, or fully synthetic (block) copolymer systems, allowing for diverse advanced applications such as serving as adsorbents for sulfate removal. This approach provides an environmentally friendly route to modify low-cost adsorbents, improving their efficiency in selectively removing both cationic and anionic species.<sup>30</sup> Synergistic effects combined with variable compositions can significantly alter adsorption behavior and anion selectivity, as demonstrated by Hassan *et al.* Matrix effects in such materials are particularly important, as they influence chelation and the degree of cation incorporation. These properties depend on the chemical groups surrounding the metal cations, such as the relative proportions of COO<sup>−</sup> and NH<sub>2</sub> groups on the biopolymer backbone.<sup>92</sup> Despite their potential, comparative studies evaluating standalone materials (*e.g.*, alginate) against their corresponding composites remain limited.<sup>93</sup> These studies are essential to confirm and quantify synergistic

effects. By merging advantageous properties such as low synthesis cost, low regeneration cost, and high performance, innovative composites can be designed to enhance pollutant removal efficiency. Although adsorption capacity is often used to assess performance, it alone does not accurately represent the overall effectiveness of an adsorbent due to the complexity of influencing factors, such as adsorbent type and functional group characteristics.<sup>4</sup> Standalone adsorbents are often favored for their low material and sustainability costs,<sup>94</sup> whereas the economic profile of composites is more variable and strongly dependent on their components.<sup>61</sup> Both synthesis and regeneration costs directly shape the total production cost of an adsorbent,<sup>95</sup> but these costs can be balanced by enhanced performance. However experimental studies analyzing regeneration costs remain scarce.

### 2.7. Design of composite adsorbents: assessment of component contributions

When adsorbents become saturated, their pollutant removal efficiency decreases, leaving behind hazardous residues. Proper handling is crucial, as improper disposal can lead to secondary contamination. Beyond adsorption capacity, sustainability depends on regenerating these materials to restore performance and minimize waste. Regeneration removes retained contaminants over multiple cycles but often demands high energy or chemicals. An alternative is repurposing spent adsorbents for secondary uses such as catalysts, fertilizers, cement additives, secondary adsorbents, or biofuels, aligning with circular economy principles by turning waste into valuable products.<sup>22</sup>



Table 2 Comparison of regeneration methods

| Method                       | Description   | Merits  | Demerits   | Economic elements  |
|------------------------------|---|---|--|--|
| Chemical regeneration        | Chemical reagents are used as desorbing agents to remove pollutants from adsorbent surfaces and pores <sup>102–104</sup>  | The process often results in rapid regeneration efficiencies <sup>103</sup>   | Reagents can generate toxic by-products and cause degradation <sup>102,103</sup>   | Ensuring safe waste disposal is crucial for managing overall costs. <sup>105</sup> High cost of reagents limits the large-scale application <sup>105</sup> |
| Thermal regeneration         | Thermal regeneration involves exposing the adsorbent to high temperatures in an inert atmosphere to desorb or decompose pollutants <sup>106</sup>   | Efficiency loss due to attrition of the adsorbent occurs <sup>105</sup>   | High energy consumption that can create harmful by-products <sup>107</sup>   | High energy cost of operation, making thermal regeneration energy-intensive and expensive <sup>108</sup>   |
| Biological regeneration      | A synergistic approach combining biodegradation and adsorption, promoting desorption and biodegradation of organic pollutants <sup>108</sup>  | Can result in the complete regeneration of the adsorbent <sup>103</sup>   | Slow regeneration rates, selectivity towards adsorbents, microbial fouling, and adsorbent surface deterioration restrict its commercial use <sup>105</sup> | The process of biological regeneration is cost effective <sup>103</sup>  |
| Electrochemical regeneration | Divided into anodic, cathodic, and combined anodic-cathodic regeneration processes. Adsorbents are placed at the anode or the cathode. In a combined process, the material is positioned in the bulk electrolyte <sup>107</sup> | This method converts organic pollutants into non-toxic products and is simple to set up and operate <sup>103</sup>                    | Potential for oxidation of the adsorbent by $\cdot\text{OH}$ , <sup>109</sup> and it tends to have lower regeneration efficiencies <sup>103</sup>          | High operational costs make it a costly method <sup>103</sup>  |
| Ultrasound regeneration      | The ultrasonic regeneration process accelerates the regeneration of spent adsorbents, which generates potent, non-selective oxidants for organic pollutants in effluents <sup>105</sup>   | This method is clean, safe, and energy-saving, with minimal carbon loss, low equipment cost, and low water consumption <sup>110</sup> | Ultrasound can damage the adsorbent depending on the frequency and intensity <sup>105</sup>  | High economic efficiency <sup>110</sup>  |

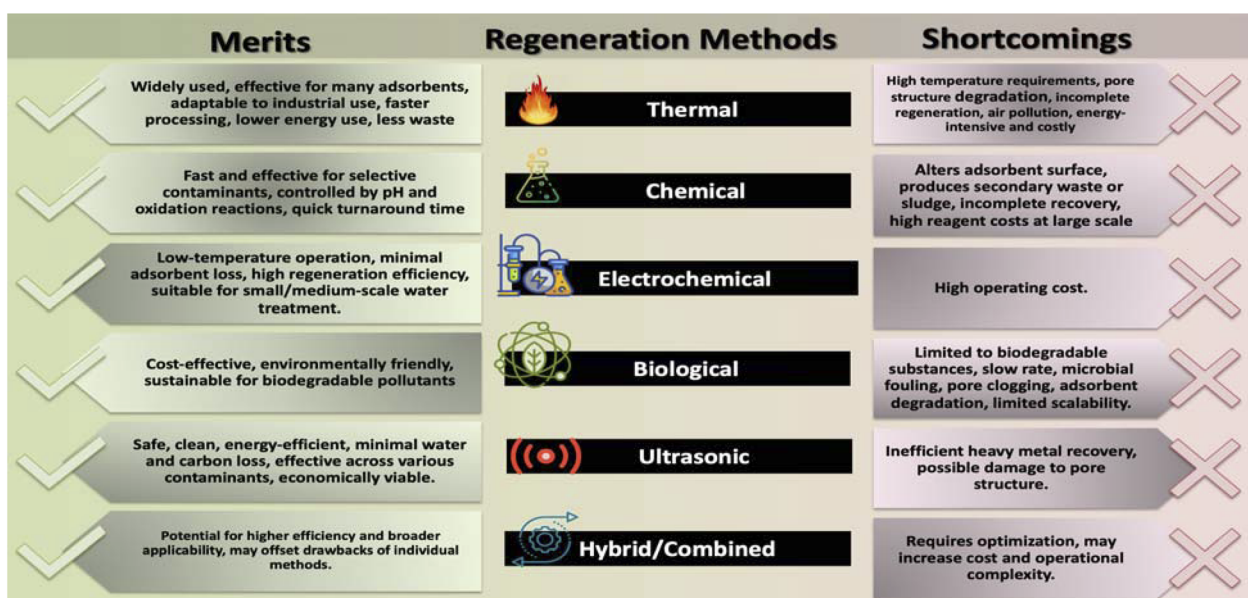


Fig. 2 Merits and shortcomings of regeneration methods.



Recycling is currently prioritized in circular economy efforts, but the main objective is to preserve a product's complexity and functionality for as long as possible rather than breaking it down into raw materials after each use. This has led to growing discussions on whether recycling should remain the dominant strategy. Increasing research focuses on alternative approaches such as repurposing, remanufacturing, refurbishing, and reusing.<sup>96</sup> It is also important to note that the release of volatile compounds during processing may pose a risk of secondary pollution.<sup>9</sup> Composite adsorbents integrate biopolymers with materials such as graphene, metal oxides, or carbon nanotubes to exploit the unique advantages of each component and generate synergistic effects.<sup>90</sup> When nanoparticles are combined with organic compounds or other metal oxides, the resulting composites often exhibit enhanced adsorption performance and improved stability under various environmental conditions.<sup>97</sup>

The primary objective is to leverage the unique strengths of each component to maximize performance. In a comprehensive review, Muhammad Faheem *et al.* emphasized the distinction between smart adsorbents, engineered for easy system integration, tunable properties, and effective pollutant retention, and spent adsorbents, which have completed their primary use. Integrating eco-friendly regeneration of smart adsorbents with sequential recycling of spent adsorbents into high-value products provides a sustainable way to reduce secondary pollution, restore functionality, and create valuable resources. This requires focused research to improve smart adsorbents' specificity, responsiveness, and reusability.<sup>19</sup>

Ahmed M. Omer *et al.* demonstrated that the Fe<sub>3</sub>O<sub>4</sub>/AP-coke/N-Cs magnetic composite adsorbent can be scaled from bench to industrial applications. Its advantages include the use of

abundant, low-cost components, simple processing without complex equipment or high energy consumption, excellent recyclability, rapid and easy separation from the medium, and strong adsorption performance. The hydrophobic nature of N-Cs enhances interactions with o-NP molecules, while AP-coke and Fe<sub>3</sub>O<sub>4</sub> improve adsorption capacity and removal efficiency.<sup>98</sup> Adelina-Gabriela Niculescu *et al.* further highlighted that incorporating magnetic components allows for straightforward separation of adsorbate-adsorbent complexes and easy regeneration. This greatly facilitates reuse in multiple decontamination cycles, enhancing both operational efficiency and sustainability.<sup>99</sup> Low-cost adsorbents often have limited reusability, as many natural materials degrade after a few cycles. For example, chitosan requires acidic or alkaline regeneration, which is costly and unsustainable. Current research focuses on low-impact regeneration methods such as solvent-free techniques, and composite formulations to enhance stability and extend reuse.

## 2.8. Regeneration methods

Spent adsorbents can be regenerated multiple times, though with reduced adsorption capacity.<sup>6</sup> The choice of regeneration method is crucial for improving desorption efficiency and depends on factors such as adsorbent type, contaminant nature, stability, toxicity,<sup>100</sup> and cost.<sup>101</sup> Various techniques (Table 2) are used to recover and regenerate adsorbents for industrial applications. Their respective merits and shortcomings are illustrated in Fig. 2.

No single regeneration method is universally effective for all adsorbents. The choice depends on the adsorbate and adsorbent's characteristics, such as toxicity, combustibility, and

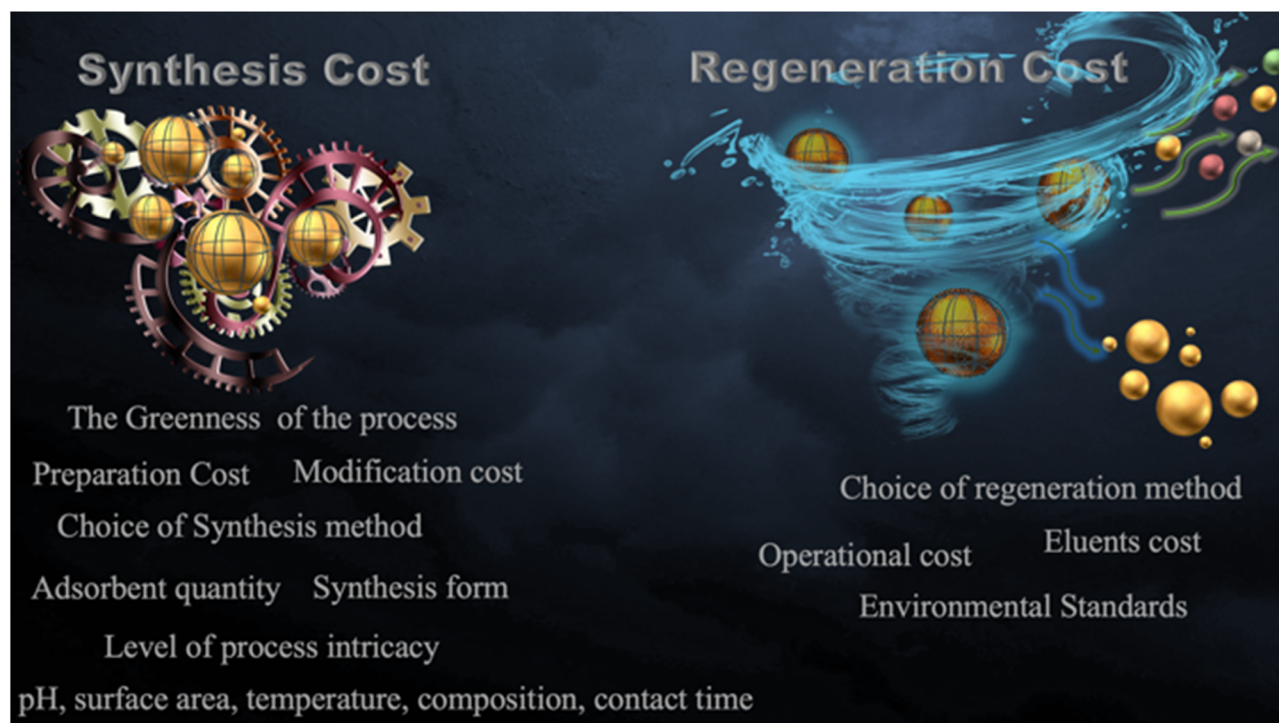


Fig. 3 Determinants governing synthesis and regeneration costs.



adsorption type. Regeneration methods must be efficient, non-toxic, eco-friendly, cost-effective, easy to operate, and suitable for reusing spent adsorbents in water treatment.<sup>72</sup>

### 2.9. How regeneration pathways govern the structural stability of adsorbents

Retaining the structural stability of adsorbents is essential to ensure their integrity throughout regeneration and repeated reuse cycles.<sup>111</sup> An effective regeneration process should eliminate target and competing contaminants while preserving the adsorbent's structure and functional groups, allowing repeated use without replacement.<sup>112</sup> In practice, however, preserving this integrity remains a major challenge.<sup>25</sup> Thermal and chemical regeneration methods may result in substantial mass reduction (10–20%) and structural degradation, which diminish reusability and adsorption efficiency.<sup>103</sup> For example, prolonged thermal regeneration can deteriorate the microporous structure of AC, resulting in reduced adsorption capacity in subsequent cycles.<sup>113</sup> Recent studies have shown that thermal treatment can also modify spent adsorbents, creating new porous structures and surface chemical properties. These regenerated adsorbents often retain similar or slightly lower contaminant removal performance compared to their original form.<sup>9</sup> Chang *et al.* regenerated montmorillonite at 600 °C for 2 h after adsorption of the antidepressant amitriptyline. The regenerated material exhibited 71.7 mg g<sup>-1</sup> removal capacity, approximately 26% of the original, due to physico-chemical alterations.<sup>114</sup> Thus, achieving successful regeneration by thermal decomposition requires careful control of temperature and treatment atmosphere. Chemical regeneration poses similar risks. High acidity can deform the adsorbent's structure, reducing its adsorption and desorption efficiency.<sup>9</sup> Strong acidic or basic materials used for regenerating MOFs can damage their frameworks and cause secondary pollution.<sup>115</sup> For example, Kołodyńska *et al.* achieved 95% Cu desorption efficiency using 3.5 M HNO<sub>3</sub> as the eluent,<sup>116</sup> but such treatments should only be applied when the sorbent has sufficient mechanical strength.<sup>9</sup> Alternative methods offer more sustainable options. Tallat Munir *et al.* developed synthetic clinoptilolite (SCP) capable of removing multiple metals from aqueous solutions and retaining over 90% of its capacity after five regeneration cycles.<sup>117</sup> Supercritical fluid regeneration preserves structural integrity, while advanced oxidation processes enable efficient regeneration with minimal degradation, facilitating multiple reuse cycles and reducing operational costs.<sup>118</sup> Microwave-assisted regeneration has attracted interest for its shorter processing times and better structural preservation, although debates remain regarding its overall efficiency.<sup>119</sup> MOFs, in particular, can be regenerated through activation, which removes guest molecules from their porous frameworks without damaging the structure, resulting in “second-generation MOFs”.<sup>120</sup> Another promising route is direct conversion, where spent adsorbents are transformed into new composite materials with targeted functionalities through chemical interactions between the adsorbent and adsorbate.<sup>121</sup> The potential of spent adsorbents depends on their raw

materials and the specific pollutants they capture. They can be repurposed into various value-added products, including (i) construction materials, (ii) antimicrobial agents, (iii) catalysts, (iv) secondary adsorbents, and (v) fertilizers. Conventionally, hazardous spent adsorbents containing oxyanion-forming elements and heavy metals are stabilized with lime or cement before landfilling, a process that is both environmentally and economically burdensome. Incorporating spent adsorbents into ceramic materials offers a more sustainable and cost-effective strategy, reducing environmental impact while stabilizing hazardous elements.<sup>122</sup>

## 3. Determinants governing synthesis and regeneration cost: scientific and strategic perspectives

### 3.1. Synthesis cost

For a technology to be considered essential, it must provide both economic and environmental advantages,<sup>123</sup> and each selection of adsorbents is influenced by a complex interaction of scientific and strategic factors<sup>124</sup> (Fig. 3).

**3.1.1 Level of process intricacy.** Many currently used adsorbents face significant challenges due to their complex production processes.<sup>19</sup> Extended equilibrium times of 24–48 hours further limit their scalability and cost efficiency.<sup>125</sup> Nearly thirty years ago, Trost underscored the importance of atom economy,<sup>126</sup> and since then, advancements in catalytic methodologies have frequently improved chemical efficiency by minimizing the number of synthetic steps and optimizing overall reaction economy.<sup>127</sup> The choice of synthesis pathways has a decisive effect on both the experimental timeframe and total cost. This decision is guided by several factors, either individually or in combination, depending on the context. A major challenge lies in accurately estimating and prioritizing the real costs of these pathways, taking into account not only the number of synthetic steps and the cost of raw materials but also the structural organization of the synthesis route and the strategic timing of expensive reagents.<sup>128</sup>

**3.1.2 Cost implications of green synthesis approaches.** A key advantage of green synthesis is the abundant availability of bio-based materials, which can substantially reduce costs.<sup>129</sup> One of the most effective strategies to lower synthesis costs is substituting chemical agents with renewable, bio-based alternatives. Multiple studies have highlighted that plant-based synthesis can accelerate reaction kinetics and provide cost benefits.<sup>130</sup> Nevertheless, despite these strengths, bio-based synthesis still faces limitations. The production costs of these materials have not yet reached levels suitable for industrial-scale implementation, partly because excessive cost reductions can compromise adsorption performance. Furthermore, although resources such as lignocellulosic biomass are abundant, fully exploiting their potential remains challenging. To enhance their performance for various pollutants, bio-based materials require tailored surface modifications and optimized synthesis strategies, areas that demand further research and technological development.<sup>131</sup>



**3.1.3 Preparation cost.** Preparation costs are a key factor in evaluating the economic feasibility of adsorbents, as they have a direct impact on overall cost–performance results.<sup>132</sup> Moreover, purification processes often require substantial time and energy.<sup>125</sup> The cost of precursors or the final adsorbent is influenced by multiple parameters, making cost evaluations difficult to standardize.<sup>133</sup> A recent study highlighted that complex synthesis methods can hinder the broader adoption of adsorption technologies.<sup>134</sup> This has generated increasing interest in developing adsorbents with simplified synthesis procedures and lower preparation cost.<sup>135</sup> The availability and control of preparation conditions strongly affect cost estimates, with any variability leading to fluctuations in total cost.

**3.1.4 Modification cost.** Numerous studies have demonstrated the benefits of modification.<sup>64</sup> However, modifying natural adsorbents generally increases their overall cost. Additionally, the relationship between cost and adsorbent performance after modification remains underexplored in many studies.<sup>136</sup> Kyzas *et al.*<sup>137</sup> emphasized that modification costs should be incorporated into techno-economic assessments of adsorbents, suggesting that using washed agricultural waste directly can be more cost-effective than producing activated carbon from the same source. Although modified adsorbents generally exhibit higher efficiency than unmodified ones, their high modification costs and reliance on toxic additives limit their large-scale application. Future research should therefore focus on developing alternative modification techniques that are both cost-effective and environmentally friendly.<sup>52</sup> Many existing modification methods face challenges such as secondary pollution, high costs, and labour-intensive procedures. Developing sustainable modification strategies is essential to produce eco-friendly adsorbents with high adsorption capacity.<sup>138</sup> Modification cost is also influenced by the type of pollutant targeted for removal.<sup>36</sup> Among the simplest and most economical modification methods is acid treatment,<sup>139</sup> whereas metal modification tends to be more expensive and

unsuitable for large-scale use.<sup>140</sup> Physical and chemical modification techniques can substantially increase pore volume and introduce various functional groups, providing rapid and low-cost enhancement options.<sup>141</sup>

**3.1.5 Synthesis form.** Although nanocomposites and hybrid materials hold considerable promise for water treatment applications, their large-scale implementation can be expensive.<sup>90</sup> The labour-intensive nature of layer-by-layer assembly and the need for precise control during modification processes limit their scalability and cost efficiency in industrial settings.<sup>125</sup>

**3.1.6 Adsorbent quantity.** Adsorption capacity, which refers to the amount of contaminant an adsorbent can retain under defined conditions, is determined by multiple factors. These include the properties of the adsorbent the characteristics of the adsorbate, and environmental parameters (temperature, pH, and contact time). Effective contaminant removal typically demands large amounts of adsorbent, leading to higher operational costs and greater process complexity.<sup>61</sup>

**3.1.7 pH, surface area, temperature, composition, and contact time.** Larger surface areas enhance adsorption capacity and contaminant removal efficiency but often come with increased manufacturing complexity and higher production costs. Elevated temperatures can accelerate adsorption rates but may compromise polymer stability and raise energy expenses. Tailoring the adsorbent's composition to specific applications, including biodegradable alternatives, demands further research and leads to higher production costs. Longer contact times improve adsorption equilibrium but reduce throughput and elevate operational expenses.<sup>142</sup> Incorporating pH-responsive functionalities can increase material versatility, offering a more cost-effective solution.<sup>143</sup>

**3.1.8 Synthesis methods.** Several nanomaterial synthesis approaches, including solvothermal processes, are energy-intensive, time-consuming, and reliant on organic solvents, making them expensive and environmentally problematic for large-scale production. Although increasing reactant volumes can help reduce heating costs, microwave-assisted synthesis



Fig. 4 Synthesis cost/regeneration cost profile of various adsorbents.



provides better energy and time efficiency. Conversely, chemical vapor deposition requires significant power input, further driving up costs. Reducing its energy demand and simplifying processing steps is therefore essential to enable industrial-scale adoption. Electrospinning also typically involves prolonged high-temperature calcination, adding to the overall energy burden.<sup>144</sup> A major research priority remains the development of new synthesis methods that lower costs, particularly energy consumption, while maximizing nanomaterial yields.

### 3.2. Regeneration cost

Regeneration cost is a critical factor in the overall economics of adsorption processes and can account for more than 50% of the total operational expenses.<sup>145</sup>

**3.2.1 Operational cost.** Operational cost plays a key role in the overall regeneration process.<sup>146</sup> The choice of regeneration method and the number of regeneration cycles directly influence operational costs.<sup>103</sup> For instance, chemical regeneration costs can be reduced by optimizing temperature conditions; thermal regeneration costs can be lowered by substituting expensive gasifying agents and decreasing treatment temperatures; microwave-assisted regeneration can be made more efficient through heat optimization; microbial regeneration can be improved by optimizing the conditions for microorganism activity; and ultrasound-based regeneration can benefit from careful control of power consumption. In some cases, however, the operational cost of regeneration may exceed the initial cost of preparing the adsorbent. This can be mitigated through alternative disposal methods such as incineration or landfilling.<sup>147</sup> To reduce costs and waste, adsorbents are reused through multiple desorption cycles until their capacity is depleted.<sup>148</sup>

**3.2.2 Regeneration method.** Although regeneration is a crucial and integral step in adsorption processes, economic sustainability often limits its efficiency, underscoring the need for further research into cost-reduction strategies. While some regeneration methods offer high efficiency, they are frequently associated with substantial costs. No single technique provides a universal low-cost solution for regenerating all types of adsorbents. Thermal regeneration, currently the most widely used method, faces challenges due to its high energy consumption and costly equipment. Microwave-assisted regeneration has emerged as a promising alternative, but it can generate undesirable by-products such as HCl, CO<sub>2</sub>, and N<sub>2</sub> when applied to adsorbents containing chlorinated or nitrogenous compounds, requiring secondary treatment and increasing total costs. Chemical regeneration has been successfully applied on a laboratory scale for many adsorbates but typically involves significant capital investment. Electrochemical regeneration has shown promise, yet the cost of required accessories remains a major barrier to large-scale implementation.<sup>149</sup> Supercritical water regeneration offers short processing times that can lower costs, but its high-pressure requirements raise extraction costs, making it more suitable for small-scale applications.<sup>72</sup> Conversely, photosensitized oxidation, which relies on metal phthalocyanine activated by visible light rather than UV light (as required by photocatalysts such as TiO<sub>2</sub>), provides a more cost-effective alternative.<sup>149</sup>

**3.2.3 Eluent cost.** The cost of eluents varies.<sup>150</sup> Common desorption agents such as ethanol or NaOH can influence both the economic and scalability aspects of the regeneration process.<sup>125</sup>

**3.2.4 Environmental standards.** Furthermore, the treatment of wastewater generated during regeneration to comply with environmental standards adds another layer of cost to the process.<sup>150</sup>

### 3.3. Economic returns

The complexity of the synthesis or regeneration pathways is a decisive factor that shapes both costs and outcomes. Turning risk into opportunity becomes feasible only when the potential risks and returns are clearly identified. The success of circular economic models relies on the economic returns achieved through regeneration. Each analyzed profile reflects a specific degree of risk associated with regeneration costs. A high-synthesis cost can be offset with a low-regeneration cost. Fig. 4 presents the synthesis cost/regeneration cost profile of various adsorbents.

CNTs, graphene, silica, and MOFs are linked to less favourable economic outcomes. These materials are positioned in the upper-right quadrant of the cost–return profile, representing high synthesis and high regeneration costs. In contrast, activated carbon and clays fall into the lower-right quadrant, characterized by low synthesis cost but high regeneration cost, which translates into lower economic returns. Agricultural-based adsorbents demonstrate the most advantageous performance, combining low synthesis and regeneration complexity with the highest economic returns. This aligns with recent technological advancements highlighting the potential to convert agricultural waste into high-value products, such as activated carbon and biochar, which can deliver returns three to five times higher than conventional applications.<sup>151</sup> The inherent complexity of synthesis and regeneration often constrains the potential for substantial economic gains. The central objective is to maximize profitability while minimizing risk through low-cost regeneration strategies. This approach provides a foundation for designing adsorbents that balance reduced risk—achieved through simpler, less chemical-intensive synthesis routes—with higher economic benefits from low regeneration costs. By integrating cost management with favorable performance outcomes, this strategy promotes informed decision-making. In the long term, optimizing synthetic and regeneration pathways can become a key driver of sustainable economic and environmental progress, mitigating the drawbacks of conventional, higher-risk methods. The cost of synthesis or regeneration methods can vary depending on the specific conditions used in the process. The sorption application and uptake efficiency are often influenced by the adsorbent's class and physicochemical properties.<sup>152</sup> When treating spent adsorbents, three options are available: (i) disposal, (ii) regeneration, and (iii) reuse. The latter two options have environmental and economic drawbacks, making regeneration the more preferable choice.<sup>153</sup> Moving forward, the use of low-cost regeneration methods in combination with joint regeneration



Table 3 High-cost spent adsorbents and their regeneration profile<sup>a</sup>

| Adsorbent                   | Adsorbate           | Adsorption operational conditions   | Isotherm and kinetic models | Performance                        | Eluent  | Regeneration efficiency (%)  | Ref. |
|-----------------------------|---------------------|---|-----------------------------|------------------------------------|---|--|------|
| PG/BCC                      | Imipramine          | Dosage: 10 mg PG/BCC, initial concentration: 250 ppm, temperature: 321 K, contact time: 34 min      | PSO, Langmuir               | 458.95 mg g <sup>-1</sup>          | Pure methanol                                 | 81.60% to 44.90% at the end of the 3rd cycle   | 156  |
| 3D-CTG                      | CV                  | Dosage: 10 mg, initial concentration: 70 mg L <sup>-1</sup> , temp: 30 °C, contact time: 45 min     | PSO, Langmuir               | 94.29%<br>583.6 mg g <sup>-1</sup> | 0.01 mol per L HCl                            | In the CV-CTG system values were maintained at around 71.7% at the end of the 4th cycle  | 157  |
| 3D-CTG                      | MG                  | Dosage: 12.5 mg, initial concentration: 40 mg L <sup>-1</sup> , temp: 40 °C, contact time: 31 min   | PSO, Langmuir               | 81.07%<br>344.8 mg g <sup>-1</sup> | 0.01 mol per L HCl                            | The MG-CTG system exhibited a significant decline in regeneration efficiency, with only 41.1% effectiveness observed after the 4th cycle | 157  |
| IA/CNT                      | MB                  | Dosage: 0.8–8 g L <sup>-1</sup> , initial concentration: 10–100 mg L <sup>-1</sup> , temp: 25–55 °C | PSO, Langmuir               | 32.78 mg g <sup>-1</sup>           | 0.1 M HCl solution and then treated with NaOH | The adsorbents were reused six times, with the percentage removal of IA-CNT adsorbent decreasing from around 83% to 74%                  | 158  |
| PANI/CNT                    | MB                  | Dosage: 0.8–8 g L <sup>-1</sup> , initial concentration: 10–100 mg L <sup>-1</sup> , temp: 25–55 °C | PSO, Langmuir               | 12.78 mg g <sup>-1</sup>           | 0.1 M HCl solution and then treated with NaOH | Adsorbents were reused six times   | 158  |
| MOF-5 and cellulose aerogel | Acid blue (anionic) | pH 7.8, T: 45 °C, duration: 180 min   | PSO, Langmuir               | 76.58%                             | Ethanol                                       | After three reuse cycles, the adsorption capacity of the MOF-5/cellulose aerogel composite declined by just 5%                           | 159  |

<sup>a</sup> 3D-CTG refers to three-dimensional cellulose/titanium/graphene oxide, MG stands for malachite green, CV represents crystal violet, PG/BCC is phosphorus-doped 3D graphene oxide with bentonite and carboxymethyl cellulose crosslinking, IA/CNT denotes itaconic acid carbon nanotubes, PANI/CNT is polyaniline carbon nanotubes, and MB refers to methylene blue.



Fig. 5 (a) Percentage removal as a function of the adsorption run, and (b) regeneration efficiency as a function of cycle number at varying methanol concentrations.<sup>156</sup>



Table 4 Low-cost spent adsorbents and their regeneration profile<sup>a</sup>

| Adsorbent            | Adsorbate        | Adsorption operational conditions  | Isotherm and kinetic models | Adsorption performance                       | Eluent  | Regeneration efficiency (%)   | Ref. |
|----------------------|------------------|--|-----------------------------|--|---|---|------|
| GA/CS-PEI-PVA (GCPP) | Cr(vi)           | Initial concentration ( $C_0$ ) of 400 ppm   | Langmuir, PSO               | 290.77 mg g <sup>-1</sup>                    | NaOH solution   | The results showed that the adsorption capacity remained notably high even after 5 cycles   | 164  |
| Ch-PDC               | Pt and Pd        | Initial concentration: 25 mg L <sup>-1</sup> to 1000 mg L <sup>-1</sup> , temperature: 20 ± 1 °C   | Langmuir, PSO               | 262.6 mg per g Pd(II), 119.5 mg per g Pt(IV) | Thiourea  | The efficiency of Pd(II) in the third cycle was 94.1%, while the efficiency for Pt(IV) was 97.7%  | 163  |
| Ch-BPDC              | Pt               | Initial concentration: 25 mg L <sup>-1</sup> to 1000 mg L <sup>-1</sup> , temperature: 20 ± 1 °C   | Langmuir, PSO               | 154.7 mg per g Pd(II), 98.3 mg per g Pt(IV)  | Thiourea  | The adsorption performance in the third cycle was 97.6% for Pt(IV)  | 163  |
| Ch-GA-HQC            | Pd               | Initial concentration: 25 mg L <sup>-1</sup> to 1000 mg L <sup>-1</sup> , temperature: 20 ± 1 °C   | Langmuir, PSO               | 340.3 mg per g Pd(II), 203.9 mg per g Pd(II) | Thiourea  | The adsorption efficiencies of Pd(II) in the third cycle was 99.6%  | 163  |
| PMKC                 | As(v)            | Dosage: 40 mg L <sup>-1</sup> , initial concentration: 100 mg L <sup>-1</sup> , temperature: 40 °C, contact time: 60 min                                     | Dubinin–Radushkevich PSO    | 337.22 mg g <sup>-1</sup>                    | Na <sub>2</sub> CO <sub>3</sub> (0.10 mol L <sup>-1</sup> ) | The adsorption efficiency of As(v) decreased to 85.10% after the 10th cycle   | 165  |
| PMKC                 | MG               | Dosage: 40 mg L <sup>-1</sup> , initial concentration: 100 mg L <sup>-1</sup> , temperature: 40 °C, contact time: 60 min                                     | Dubinin–Radushkevich, PSO   | 274.73 mg g <sup>-1</sup>                    | Na <sub>2</sub> CO <sub>3</sub> (0.10 mol L <sup>-1</sup> ) | The percentage adsorption of MG decreased to 81.00% after the 10th cycle  | 165  |
| CS/DS@ZIF-8          | Pb <sup>2+</sup> | Dosage: 10 mg, initial concentration: 50–800 mg L <sup>-1</sup> , contact time: 300 min, temperature: 25 °C  | Langmuir, PSO               | 340.94 mg g <sup>-1</sup>                    | Ethanol   | CS/DS@ZIF-8 maintains 81.3%   | 166  |
| CS/DS@ZIF-8          | Cu <sup>2+</sup> | Dosage: 10 mg, initial concentration: 50–800 mg L <sup>-1</sup> , contact time: 300 min, temperature: 25 °C  | Langmuir, PSO               | 308.27 mg g <sup>-1</sup>                    | Ethanol   | CS/DS@ZIF-8 maintains 72.9%   | 166  |
| FE/AS/CS             | Pb <sup>2+</sup> | Doses: 0.05–2 g L <sup>-1</sup> , initial concentrations: 10 and 50 mg L, contact time intervals: (5–150 min), temperature range: (313–353 K)                | Langmuir, PSO               | 305.5 mg g <sup>-1</sup>                     | 0.1 M of HNO <sub>3</sub>                                   | Pb(II) was recovered at rates of 97%, after the third cycle. No significant decline in the adsorption capacity for Pb(II) on the prepared composite was observed after the third consecutive adsorption/desorption cycles | 167  |
| FE/AS/CS             | Cu <sup>2+</sup> | Doses: 0.05–2 g L <sup>-1</sup> , initial concentrations: 10 and 50 mg L <sup>-1</sup> , contact time intervals: (5–150 min), temperature range: (313–353 K) | Langmuir, PSO               | 284.2 mg g <sup>-1</sup>                     | 0.1 M of HNO <sub>3</sub>                                   | Cu(II) was recovered at 95.6%, after the third cycle. The adsorption capacity for Pb(II) and Cu(II) on the prepared composite showed no noticeable decrease after the third consecutive adsorption/desorption cycles      | 167  |



Table 4 (Contd.)

| Adsorbent  | Adsorbate  | Adsorption operational conditions  | Isotherm and kinetic models        | Adsorption performance                                  | Eluent                               | Regeneration efficiency (%)  | Ref. |
|--|--|--|------------------------------------|---|--------------------------------------|--|------|
| ZnO/SA-NFs   | Pb <sup>2+</sup> and Cu <sup>2+</sup>                      | Initial concentration of TC, 500 mg L <sup>-1</sup> ; amount of adsorbent, 10 mg; volume, 5 mL; contact time, 120 min; temperature, 303 K; pH, 3.0 | Liu model, PSO                     | 369.6 mg g <sup>-1</sup> , 124.1 mg g <sup>-1</sup>     | <i>N,N</i> -Dimethyl formamide (DMF) | The adsorption capacity of TC decreases by less than 20% after five cycles   | 168  |
| Alg@MgS  | Pb <sup>2+</sup>   | pH of 4, dosage 20 mg, <i>t</i> : 60 min   | Freundlich, PSO                    | 84.7 mg g <sup>-1</sup>                                 | Simple acid washing technique        | The adsorption capacity decreased after 6 consecutive cycles   | 169  |
| CANRC  | Pb <sup>2+</sup> , Zn <sup>2+</sup> , and Cd <sup>2+</sup> | Dosage of 2.5 g L <sup>-1</sup> , pH = 5.0–6.0   | Langmuir                           | 247.99, 71.77, and 47.27 mg g <sup>-1</sup>             | 0.1 M NaNO <sub>3</sub>              | After 4 times of regeneration, the removal rates of Pb <sup>2+</sup> , Zn <sup>2+</sup> , and Cd <sup>2+</sup> remain above 96%, 15%, and 10%  | 170  |
| Fe <sub>3</sub> O <sub>4</sub> @TAC@SA                             | Diclofenac   | Dosage: 0.02 g/25 mL, initial concentration: 0.0002755 mol, adsorption period: 100 min, <i>T</i> : 50 °C, pH: 3                                    | Langmuir, PSO                      | 858 mg g <sup>-1</sup>                                  | NaOH (0.1 mol L <sup>-1</sup> )      | Highly efficient after three regenerative cycles   | 171  |
| CE/CSA   | Congo red (CR) and Cu <sup>2+</sup>                        | Dosage = 0.25 g L <sup>-1</sup> , <i>T</i> = 298 K, C <sub>(CR)</sub> or C <sub>(Cu<sup>2+</sup>)</sub> = 20 mg L <sup>-1</sup>                    | Thomas and Yoon–Nelson models, PSO | 380.23 mg g <sup>-1</sup> and 260.41 mg g <sup>-1</sup> | DMF and 0.2 M EDTA-2Na solution      | The initial removal rate of CEA, CSA, CE/CSA-1 and CE/CSA-2 for CR was 27.22%, 77.63%, 89.70% and 96.34%, respectively, and the removal rate decreased to 21.70%, 61.72%, 82.00% and 90.45% after six cycles, respectively | 172  |
| Cellulose–Sn(IV) (CSn) cellulose and stannic chloride biocomposite | As(III)  | Dosage: 400 mg initial concentration: 5 mg per L pH 7.0  | Freundlich, PSO                    | 16.64 mg g <sup>-1</sup>                                | 5% (w/v) NaCl                        | Over five adsorption–desorption cycles, As(III) removal decreased gradually from 95% to 78%, demonstrating the environmentally friendly performance of the CSn   | 173  |

<sup>a</sup> Ch-PDC refers to 1,10-phenanthroline-2,9-dicarbaldehyde cross-linked chitosan, Ch-BPDC to [2,2-bipyridine]-5,5-dicarbaldehyde cross-linked chitosan, and Ch-GA-HQC to glutaraldehyde cross-linked chitosan grafted with 8-hydroxyquinoline-2-carbaldehyde. Pt denotes platinum, Pd represents palladium, and PMKC corresponds to *Pterocarpus mildbraedii* integrated into mesoporous kaolin clay. As(V) represents arsenic ions, MG stands for malachite green, and CS/DS@ZIF-8 indicates chitosan microspheres doped with silica and zeolite imidazolate frameworks. FE/AS/CS refers to a Fuller's earth/aluminum silicate/chitosan composite. ZnO/SA-NFs designate alginate-based nanofibers loaded with ZnO nanoparticles. Alg@MgS refers to alginate microbeads encapsulating magnesium sulfide nanoparticles, and CANRC denotes a calcium alginate-nZVI-biochar composite. Fe<sub>3</sub>O<sub>4</sub>@TAC@SA stands for sodium alginate, magnetite, and activated carbon derived from tea waste polymer. CE/CSA represents cellulose extracted from waste reed (CE) and chitosan (CS), forming a three-dimensional (3D) hierarchical porous structure.

processes will likely become a key trend to improve regeneration efficiency and reduce costs.

## 4. Regeneration studies

Regeneration is the process of quickly recycling or recovering spent adsorbents using methods that are both technically and economically viable.<sup>72</sup> Since cost is a critical factor in the development of adsorbents, the regeneration process plays a crucial

role in effective pollution control. Researchers are prioritizing adsorbent regeneration and reuse because of the significant costs associated with production, stabilization, disposal, and preparation.<sup>103</sup> Regeneration studies assess adsorbent reusability and economic viability.<sup>154</sup> However, challenges include: (i) instability of many adsorbents, (ii) difficulty in desorbing ions or molecules, and (iii) the need for harmful eluents, which raise safety concerns. It is crucial to prioritize ease of regeneration and develop new reuse methods when designing adsorbents.<sup>19</sup>





Fig. 6 (a) The reduction ratio of Cr ions was compared for CS and GCPP at different concentrations. (b) The reduction ratio of Cr ions by GCPP was analyzed at various solution pH levels. (c) Adsorption capacity and (d) reduction ratio of Cr ions by GCPP were evaluated over five adsorption–reduction cycles.<sup>164</sup>

#### 4.1. Regeneration studies of high-cost spent adsorbents

In adsorption-based processes, adsorbent cost is a key factor, and current efforts are increasingly directed toward evaluating advanced, often higher-cost, materials.<sup>155</sup> A summary of high-cost spent adsorbents and their regeneration profiles is provided in Table 3. Wan Ting Tee *et al.* developed a phosphorus-doped 3D graphene oxide composite (PG/BCC) for efficient imipramine removal from wastewater. Batch experiments and central composite design (CCD) optimization resulted in a maximum adsorption capacity of  $458.95 \text{ mg g}^{-1}$ . Characterization confirmed imipramine incorporation. Fig. 5(a) shows removal percentages for different methanol concentrations, while Fig. 5(b) demonstrates the effect of eluent concentration and cycle number on regeneration efficiency.<sup>156</sup>

The regeneration of PG/BCC using methanol was highly feasible, supporting its potential as a sustainable graphene-based adsorbent for imipramine removal from pharmaceutical wastewater.<sup>156</sup> The group created an eco-friendly 3D-CTG adsorbent for removing CV and MG dyes, with adsorption fitting the Langmuir isotherm and following pseudo-second-order kinetics. Process optimization resulted in 94.29% CV removal. The 3D-CTG adsorbent showed strong reusability and efficient regeneration over four cycles.<sup>157</sup>

The study used itaconic acid- and polyaniline-modified CNTs to remove methylene blue dye, examining the effects of pH, dosage, concentration, and temperature. Characterization confirmed Langmuir isotherm behavior with maximum capacities of  $32.78 \text{ mg per g (IA/CNT)}$  and  $12.78 \text{ mg per g (PANI/CNT)}$ .<sup>158</sup>

Shiri M. and colleagues developed an innovative composite material, MOF-5/cellulose aerogel, using the Pampas plant as a natural source for cellulose aerogel production. Their study examined the adsorption efficiency and structural flexibility of this composite for removing organic dyes. Reusability tests showed that the material retained stable performance, with only a slight 5% decrease in adsorption capacity after multiple cycles. This consistent absorption rate highlights the MOF-5/cellulose aerogel as an environmentally friendly and reusable adsorbent for dye removal applications.<sup>159</sup>

#### 4.2. Regeneration studies of low-cost spent adsorbents

The high costs of traditional adsorbents have prompted researchers to explore more affordable alternatives. Biopolymers, known for their non-toxicity, availability, and cost-effectiveness, have gained attention for wastewater treatment. Among these, chitosan, natural zeolites, clays, and soil constituents are noted for their affordability and widespread availability.<sup>160</sup> Clays, in particular, are a promising alternative due to their natural abundance and being up to 20 times cheaper than activated carbon.<sup>161</sup> While nanomaterials are also potential adsorbents, they face challenges such as limited selectivity, structural issues, agglomeration, and difficulties in separation.<sup>162</sup> Silica, with its versatile surface chemistry and high porosity, is effective in pollutant removal but faces challenges such as heterogeneous pore structure and poor stability. To address these issues, new adsorbents featuring triple or double grafting composites are being developed. Table 4 summarizes various studies on low-cost adsorbents, including process conditions, capacities, and regeneration efficiencies.



Mincke S. *et al.* developed three green chitosan derivatives for Pd(II) and Pt(IV) adsorption. The Langmuir isotherm provided the best fit, with optimal performance at pH 3. Ch-GA-HQC showed the highest capacities. Kinetic studies indicated pseudo-second-order chemisorption with external and intra-particle diffusion. The materials were easily regenerated with over 95% recovery, and functionalization enhanced capacity, acid stability, and reusability, providing strong environmental advantages.<sup>163</sup>

Ling Peng *et al.* developed a novel chitosan-based adsorbent, GA/CS-PEI-PVA (GCPP), incorporating polyethyleneimine (PEI), polyvinyl alcohol (PVA), and 3,4,5-trihydroxybenzoic acid (GA) to create crosslinked networks for targeted adsorption. The GCPP showed improved thermal stability, mechanical strength, and a larger specific surface area. It achieved a Cr ion adsorption capacity of 290.77 mg g<sup>-1</sup> and reduced Cr(VI) to Cr(III) by 83.20%. The adsorption mechanism involved complexation and electrostatic attraction, with phenolic hydroxyl groups playing a key role in the reduction. This work advances the development of efficient adsorbents for Cr ion removal from wastewater.<sup>164</sup> The study also examined the effect of pH on reduction efficiency (Fig. 6(a) and (b)), revealing a variable reduction ratio linked to Cr ion speciation. GCPP's stability and recyclability were evaluated through cyclic tests, showing consistent adsorption capacity after five cycles, with regeneration through NaOH solution and deionized water washes (Fig. 6(a), (c) and (d)).<sup>164</sup>

Titus Chinedu Egbosiuba *et al.* developed a biogenic ultrasonic method to modify kaolin clay with *Pterocarpus mildraedii* (PMKC) for removing As(V) and MG. The flake-like PMKC achieved adsorption capacities of 337.22 mg g<sup>-1</sup> for As(V) and 274.73 mg g<sup>-1</sup> for MG under optimal conditions. The process followed Dubinin–Radushkevich and pseudo-first-order models, and the material remained stable and reusable for up to ten cycles, showing strong potential for pollutant removal.<sup>165</sup>

J. Li *et al.* developed a bifunctional composite microsphere adsorbent, CS/DS@ZIF-8, by combining chitosan microspheres with silica and ZIF-8. The material exhibited enhanced crystallinity, surface area, porosity, thermal stability, and active sites. Pb<sup>2+</sup> and Cu<sup>2+</sup> adsorption followed the Langmuir isotherm and pseudo-second-order kinetics, with capacities of 340.94 mg g<sup>-1</sup> and 308.27 mg g<sup>-1</sup>. It retained 81.3% and 72.9% efficiency after five cycles and showed strong antibacterial activity, highlighting its promise for wastewater treatment.<sup>166</sup>

Heba Kandil *et al.* developed a Fuller's Earth–Aluminum Silicate–Chitosan (FE/AS/CS) hybrid composite for Pb<sup>2+</sup> and Cu<sup>2+</sup> removal. It achieved maximum removal rates of 98.5% and 97%, with adsorption following the Langmuir model, indicating chemisorption. The composite maintained high efficiency after three adsorption–desorption cycles, confirming good reusability.<sup>167</sup>

Kouhua Zhang *et al.* created ZnO/SA-NFs, alginate-based nanofibers with ZnO nanoparticles, using electrospinning. The porous fibers (surface area 5.443 m<sup>2</sup> g<sup>-1</sup>, pore size 19 nm) showed adsorption capacities of 248.6, 244.5, and 388.6 mg g<sup>-1</sup> for tetracycline, oxytetracycline, and doxycycline. Adsorption followed the pseudo-second-order and Liu models, with a spontaneous, endothermic process driven by ZnO–alginate

interactions. The nanofibers performed well in real water samples, showing strong potential for tetracycline removal.<sup>168</sup>

Mehdi Esmaeili Bidhendi and co-workers employed alginate-caged magnesium sulfide (MgS) nanoparticles in microbead form to remove Pb<sup>2+</sup> ions from water. An optimal removal efficiency of 91% was achieved at pH 4, with an adsorbent dosage of 20 mg and a contact time of 60 min. The adsorption kinetics followed the pseudo-second-order model more closely than the pseudo-first-order model, supported by high *R*<sup>2</sup> values and further confirmed by the Elovich model (*R*<sup>2</sup> = 0.964). Equilibrium data fitted the Freundlich isotherm better than the Langmuir model, indicating heterogeneous surface adsorption.<sup>169</sup>

Ruohan Zhao *et al.* synthesized a calcium alginate–nZVI–biochar composite (CANRC) and applied it for the simultaneous removal of Pb<sup>2+</sup>, Zn<sup>2+</sup>, and Cd<sup>2+</sup> from water. Adsorption mechanisms were investigated using various models and site energy analyses. CANRC prepared at 300 °C with a 5 wt% Fe loading exhibited the highest adsorption capacities under conditions of 2.5 g L<sup>-1</sup> dosage and pH 5.0–6.0. The adsorption behavior was best described by the Langmuir isotherm, suggesting monolayer adsorption. Maximum adsorption capacities reached 247.99 mg g<sup>-1</sup> for Pb<sup>2+</sup>, 71.77 mg g<sup>-1</sup> for Zn<sup>2+</sup>, and 47.27 mg g<sup>-1</sup> for Cd<sup>2+</sup>.<sup>170</sup>

Salhah D. Al-Qahtani and co-workers employed an Fe<sub>3</sub>O<sub>4</sub>@TAC@SA polymer to remove diclofenac sodium from water, achieving an impressive adsorption capacity of 858 mg g<sup>-1</sup>. The adsorption mechanism involved a combination of ion exchange, π–π interactions, electrostatic pore filling, and hydrogen bonding. Notably, the adsorbent maintained high removal efficiency after three regeneration cycles, demonstrating its strong reusability.<sup>171</sup>

Yanyang Liu *et al.* synthesized a multifunctional biomass-based aerogel (CE/CSA) composed of cellulose (extracted from waste reed) and chitosan. The aerogel exhibited a 3D hierarchical porous structure with a low density of 0.062 g cm<sup>-3</sup>. The maximum adsorption capacities of CE/CSA-1 for Congo red (CR) and Cu<sup>2+</sup> were 380.23 mg g<sup>-1</sup> and 260.41 mg g<sup>-1</sup>, respectively, in a binary system, representing a 49.05% and 28.64% increase compared to single-component adsorption. This enhancement was attributed to a synergistic bridging effect: preloaded CR introduced new adsorption sites (–NH<sub>2</sub>/–SO<sub>3</sub><sup>-</sup>) for Cu<sup>2+</sup>, while preadsorbed Cu<sup>2+</sup> facilitated CR binding. Fixed-bed column tests showed a CR adsorption capacity of 241 mg g<sup>-1</sup>, with breakthrough behavior fitting well to the Thomas and Yoon–Nelson models.<sup>172</sup>

Anita Shekhawat *et al.* developed a cellulose–Sn(IV) (CSn) biocomposite using microwave-assisted synthesis. It achieved an adsorption capacity of 16.64 mg g<sup>-1</sup> for As(III) at pH 7. Regeneration with 5% NaCl showed a gradual efficiency drop from 95% to 78% over five cycles, confirming the material's green and reusable properties.<sup>173</sup>

Juan Díaz *et al.* successfully synthesized a novel AL-based biocomposite, P(CLAPTA-AL), *via* radical polymerization and thoroughly characterized it for dye adsorption applications. The material demonstrated exceptional adsorption capacity for ARS dye, surpassing the performance of many conventional



Table 5 Primary use and reuse of spent adsorbents reported in the literature<sup>a</sup>

| Adsorbent                      | Primary adsorption conditions   | Removal efficiency/<br>adsorption capacity<br>(mg g <sup>-1</sup> ) | Spent<br>adsorbent                                   | Reuse   | Ref. |
|--------------------------------|---|---|--|---|------|
| PANI-RBTW                      | Cr(vi), PSO, Freundlich model   | 293.72  | PANI-RBTW/<br>Cr(vi)                                 | Photocatalyst for the degradation of tetracycline                                   | 178  |
| RWBO                           | Ni(II), PSO, Temkin model   | 99.75%  | RWBO-Ni(II)  | Photocatalyst for the degradation of ciprofloxacin                                  | 177  |
| N-CNPs/<br>ZnONP               | Cu <sup>2+</sup> PSO, Langmuir model                                    | 285.71  | Cu <sup>2+</sup> -N-CNPs/<br>ZnONP                   | Latent fingerprint detection  | 179  |
| CNS/<br>ZrO <sub>2</sub> NPs   | Zn <sup>2+</sup> hydrothermal method, temkin<br>model, Langmuir model   | 606.06  | Zn <sup>2+</sup> -CNS/<br>ZrO <sub>2</sub> NPs       | Latent fingerprint detection  | 180  |
| RWBO                           | Cd(II), PSO, Langmuir model   | 90.63%  | RBTW/Cd(II)  | Photocatalyst for the degradation of<br>sulfamethoxazole                            | 181  |
| CFA/GO/<br>WO <sub>3</sub> NRs | Hydrothermal method, PSO,<br>Langmuir model                             | 41.51   | CFA/GO/<br>WO <sub>3</sub> NRs +<br>Pb <sup>2+</sup> | Photocatalyst for the degradation of<br>acetaminophen                               | 182  |
| MC/TiO <sub>2</sub><br>NPs     | Pb <sup>2+</sup> , hydrothermal method, PSO,<br>Langmuir model          | 168.919   | Pb <sup>2+</sup> -MC/TiO <sub>2</sub><br>NP          | Latent blood fingerprint detection  | 203  |
| MnO <sub>2</sub> -CFA          | Pb <sup>2+</sup> , hydrothermal method, Elovich,<br>Langmuir model      | 141   | Pb <sup>2+</sup> -MnO <sub>2</sub> -<br>CFA          | Latent fingerprint detection  | 203  |
| CFA/C<br>HNCPs                 | Cd <sup>2+</sup> with hydrothermal method, PSO, 77 °C<br>Langmuir model |   | CFA/C-<br>Cd <sup>2+</sup> HNCPs                     | Photocatalyst for the degradation of MB   | 184  |
| PPy@L-<br>Cyst                 | Hg <sup>2+</sup> radical polymerization, PSO,<br>Langmuir model         | 2042.7  | PPy@L-Cyst/<br>Hg(II)                                | Catalyst in a reaction with phenylacetylene to<br>furnish acetophenone of 52% yield | 185  |

<sup>a</sup> CFA/C HNCPs – coal fly ash/carbon hybrid nanocomposite, CFA/C-Cd<sub>2</sub>+HNCPs – coal fly ash/carbon cadmium hybrid nanocomposite, Cd<sup>2+</sup> – cadmium ions, MB – methylene blue, MC/TiO<sub>2</sub> NPs – mesoporous carbon/titanium dioxide nanoparticles, Pb<sup>2+</sup>-MC/TiO<sub>2</sub> NP – lead mesoporous carbon/titanium dioxide nanoparticle, MnO<sub>2</sub>-CFA – manganese oxide-coated fly ash, PPy@L-Cyst – polypyrrole with L-cysteine, PANI-RBTW – polyaniline-decorated rooibos tea waste, RBTW – rooibos tea waste, N-CNPs/ZnONP – zinc oxide nanoparticle nanocomposite, CNS/ZrO<sub>2</sub>NPs – carbon nanosheets coated on zirconium oxide nanoplates, and CFA/GO/WO<sub>3</sub>NRs – graphene oxide-tungsten oxide nanorod nanocomposite.

adsorbents. Optimal adsorption conditions, established using a Box–Behnken design, included a pH of 12.0, a temperature of 20 °C, a contact time of 120 minutes, and a composite-to-ARS mass ratio of 10. The biocomposite maintained an adsorption efficiency of approximately 99% up to the fourth cycle and 81.1% after the seventh, indicating strong reusability. Its high capacity, ease of synthesis, environmental compatibility, and durability make P(CIAPTA-AL) a promising candidate for large-scale water treatment applications.<sup>174</sup>

Chao Wang and co-workers developed a bio-based hydrogel (LN-NH-SA) using aminated lignin and sodium alginate, which was evaluated for the removal of methyl orange (MO) and methylene blue (MB). The LN-NH-SA@3 hydrogel exhibited a maximum MB adsorption capacity of 388.81 mg g<sup>-1</sup>, demonstrating excellent performance as a bio-based adsorbent.<sup>175</sup>

## 5. Sequential recycling and reuse of spent adsorbents

Desorption and adsorbent recycling are critical factors in designing the desorption process, which involves recovering bound ions and regenerating the spent sorbent material.<sup>176</sup> In terms of sustainability, recent studies have focused on methods for regenerating adsorbents, such as direct desorption and converting spent adsorbents into new materials, with minimal treatment between uses to ensure a cost-effective and sustainable approach. The application of spent heavy metal adsorbents has expanded to areas such as photocatalytic degradation of

pharmaceuticals and latent fingerprint detection. Table 5 outlines the primary uses and reuses of spent adsorbents as reported in the literature.

Recent studies have explored the potential of rooibos tea waste (RTW) as an effective adsorbent for various pollutants. Opeoluwa I. Adeiga *et al.* developed a composite adsorbent by combining RTW with a binary oxide (Fe<sub>2</sub>O<sub>3</sub>-SnO<sub>2</sub>) for removing Ni(II) ions, achieving 99.75% removal efficiency. The adsorption process was endothermic and spontaneous.<sup>177</sup> In another study, the same team used polyaniline-decorated RTW (PANI-RBTW) to remove hexavalent chromium (Cr(vi)) and reuse the composite as a photocatalyst for tetracycline removal. The PANI-RBTW composite achieved 100% removal of Cr(vi) under optimal conditions. The composite also effectively degraded 10 mg per L tetracycline, achieving 80.4% degradation and 70% mineralization in 150 minutes. The PANI-RBTW composite proves to be an effective adsorbent for toxic metal ions and a viable photocatalyst for organic pollutant remediation.<sup>178</sup>

Opeoluwa I. Adeiga developed a low-cost rooibos tea waste (RBTW) adsorbent for Cd(II) removal and subsequent photocatalytic degradation of sulfamethoxazole. RBTW showed an adsorption capacity of 7.13 mg g<sup>-1</sup> and 90.63% removal at 45 °C and pH 7, following the Langmuir isotherm and pseudo-second-order kinetics. The Cd-loaded adsorbent degraded 69% of sulfamethoxazole with 53% mineralization under visible light. The endothermic, spontaneous process demonstrates RBTW's dual effectiveness for heavy metal removal and organic pollutant degradation in wastewater treatment.<sup>181</sup>



Fouda-Mbanga B. G. *et al.* developed a CNS/ZrO<sub>2</sub>NP nanocomposite for Zn<sup>2+</sup> removal. It achieved a maximum adsorption capacity of 606.06 mg g<sup>-1</sup> at pH 8 and 20 mg dosage. The process was exothermic and spontaneous. The Zn<sup>2+</sup>-loaded material was successfully reused for latent fingerprint detection, showing high selectivity and sensitivity, and reducing secondary pollution risk.<sup>180</sup>

The same research group developed a N-CNPs/ZnONP nanocomposite using pineapple leaves and zinc oxide nanoparticles for copper ion removal from water. The nanocomposite exhibited outstanding copper uptake efficiency, achieving 99.67% at the optimal pH and 99.78% at the correct dosage. The nanocomposite was also effectively used for latent fingerprint detection under normal light, proving its potential as a recyclable labeling agent for forensic applications.<sup>179</sup>

Emmanuel Christopher Umejuru *et al.* developed CFA/C HNCs from coal fly ash *via* hydrothermal synthesis for Cd<sup>2+</sup> removal. The material had a maximum adsorption capacity of 77 mg g<sup>-1</sup>, following the Langmuir isotherm and pseudo-second-order kinetics. The Cd<sup>2+</sup>-loaded adsorbent achieved 97.41% methylene blue degradation through photocatalysis, showing strong potential for combined heavy metal removal and pollutant degradation in environmental remediation.<sup>184</sup>

The same group modified coal fly ash with a graphene oxide-tungsten oxide nanorod composite (CFA/GO/WO<sub>3</sub>NRs) for Pb<sup>2+</sup> removal. The material showed an adsorption capacity of 41.51 mg g<sup>-1</sup>, following the Langmuir isotherm and pseudo-second-order kinetics. The Pb<sup>2+</sup>-loaded adsorbent was reused for acetaminophen photodegradation, achieving 93% degradation. This approach highlights the potential of reusing spent adsorbents for photocatalysis, minimizing secondary waste.<sup>182</sup>

Yvonne Boitumelo Nthwane and colleagues developed a composite for Pb<sup>2+</sup> removal and reusing the Pb<sup>2+</sup>-loaded spent adsorbent in blood fingerprint detection. The nanocomposite showed a high adsorption capacity of 168.92 mg g<sup>-1</sup> for Pb<sup>2+</sup> removal at pH 4, achieving a 98% removal rate. The adsorption process was exothermic and spontaneous. In fingerprint detection, the composite improved blood fingerprint clarity, highlighting its potential for use on nonporous surfaces while minimizing secondary pollution.<sup>203</sup>

M. W. Mofulatsi and colleagues synthesized manganese oxide-coated fly ash (MnO<sub>2</sub>-CFA), which exhibited a threefold increase in surface area compared to raw fly ash. Adsorption experiments showed a maximum capacity of 141 mg g<sup>-1</sup>. The adsorption process was endothermic and spontaneous, displaying high selectivity for Pb<sup>2+</sup> over other metal ions. The adsorbent removed 83.33% of Pb<sup>2+</sup> from a spiked water sample. Additionally, the spent adsorbent proved effective in latent fingerprint detection, yielding clearer images than MnO<sub>2</sub>-CFA, with clarity lasting up to 8 days, showcasing its potential as a labeling agent.<sup>183</sup>

Niladri Ballav *et al.* developed a polypyrrole-L-cysteine (PPy@L-Cyst) composite that serves as a highly efficient adsorbent for Hg<sup>2+</sup> removal, with an impressive adsorption capacity of 2042.7 mg g<sup>-1</sup> at pH 5.5. The adsorption process was well-represented by the pseudo-second-order rate equation and Langmuir isotherm model, with electrostatic interactions between the adsorbent and Hg<sup>2+</sup> ions being the dominant

mechanism. The composite's strong binding affinity was attributed to its electron-rich functional groups. Furthermore, the Hg<sup>2+</sup>-loaded spent adsorbent was successfully utilized as a catalyst for the conversion of phenylacetylene to acetophenone, yielding 52%. The PPy@L-Cyst composite shows great potential for both Hg<sup>2+</sup> removal and catalytic applications in environmental remediation.<sup>183</sup>

## 6. The importance of sustainability and life cycle assessment in the adsorbent agenda

Despite their demonstrated advantages, adsorbent technologies remain an evolving research field, with increasing emphasis on improving sustainability profiles.<sup>186</sup> The overall viability of an adsorbent can only be achieved when three fundamental criteria are simultaneously satisfied, namely (i) high removal performance, (ii) economic feasibility, and (iii) environmental sustainability. A robust sustainability profile must be supported across the entire life cycle, including synthesis and regeneration stages, through simplified procedures that adhere to green chemistry principles while maintaining low costs.<sup>187</sup>

In this context, several studies have shown that the adoption of green synthesis routes and solvent recovery strategies can substantially reduce environmental impacts.<sup>188</sup> Accordingly, adsorbent synthesis pathways have undergone considerable evolution over recent years, enabling enhanced control over material properties and performance.<sup>189</sup> In parallel, regeneration strategies that extend adsorbent lifetime and improve sustainability<sup>20</sup> must be environmentally benign, cost effective, and non-toxic. Such approaches include the use of mild eluents, electrochemical regeneration, and biological methods.<sup>190</sup> Environmental sustainability is therefore a critical consideration in the design and application of adsorbent materials. However, significant challenges remain. Certain adsorbents may undergo degradation under prolonged exposure to harsh environmental conditions.<sup>142</sup> Moreover, the management of spent adsorbents and their net contribution to environmental burdens across the life cycle remain insufficiently explored. In particular, regeneration processes for bio adsorbents require further investigation. Although a gradual decline in adsorption efficiency following successive regeneration cycles is expected, this phenomenon raises concerns regarding long term applicability. This efficiency loss must be explicitly considered when employing regenerated bio adsorbents, and sustainable, cost effective strategies are required to restore or retain performance.<sup>191</sup> The combined environmental and economic burdens associated with adsorbent synthesis and regeneration underscore the need for comprehensive sustainability evaluations to ensure long term feasibility. In this regard, life cycle assessment (LCA) represents a critical quantitative tool for evaluating environmental, economic, and social aspects across the full life cycle, including carbon footprint and a broad range of environmental benefits or trade-offs,<sup>192</sup> as well as cost efficiency from raw material extraction to end-of-life management.<sup>193</sup> LCA is also widely applied to provide early stage assessments of emerging



technologies, enabling redesign and optimization of products and processes.<sup>194</sup> The LCA methodology is standardized under ISO 14040:2006, which defines four main phases, namely (i) goal and scope definition, (ii) life cycle inventory analysis, (iii) life cycle impact assessment, and (iv) interpretation.<sup>192</sup> Application of this framework has enabled the identification of critical improvement points in production systems, such as reducing energy and chemical consumption.<sup>194,195</sup>

To date, LCA studies addressing the environmental impacts associated with end-of-life treatment options for adsorbents remain limited. Furthermore, environmental assessments of nanoadsorbent synthesis are scarce within the current literature.<sup>196</sup> Many studies do not provide comparative analyses capable of substantiating sustainability claims and often fail to conduct full cradle-to-grave evaluations that account for energy intensive and chemically demanding synthesis steps. Consequently, recent efforts increasingly emphasize the development of sustainable synthesis and regeneration strategies.<sup>188</sup> Indeed, comprehensive life cycle analyses are strongly warranted, particularly given that improvements in sustainability may require trade-offs in performance or durability when compared with conventional approaches.

For example, Korhonen *et al.* identified kaolin calcination to metakaolin, sodium hydroxide consumption during synthesis, energy use, and wastewater generation as the primary contributors to climate impacts. The global warming potential was estimated at 2.01 kg CO<sub>2</sub>eq per kg of adsorbent, a value comparable to those reported for conventional adsorbents such as activated carbon.<sup>197</sup>

Similarly, Ahmed I. Osman and co-workers conducted an LCA to evaluate the environmental impacts associated with the production of composite adsorbent materials. For one functional unit, defined as 1 kg of pomace leaves used as feedstock, abiotic depletion of fossil fuels and global warming potential were quantified as 7.17 MJ and 0.63 kg CO<sub>2</sub>eq, respectively, for the production of magnetic char composite materials. The resulting magnetic char composite was applied for crystal violet dye removal under various operational conditions. Kinetic and isotherm analyses indicated that adsorption followed pseudo second order and Langmuir models, respectively, with a maximum adsorption capacity of 256.41 mg g<sup>-1</sup>. Furthermore, recyclability of 92.4 percent was achieved after five adsorption-desorption cycles. These findings highlight the potential for sustainable and cost effective magnetic sorbent production, particularly from combined biomass and plastic waste streams.<sup>198</sup>

Gopa Nandikes and colleagues provided comprehensive insights through LCA using both mass-based and adsorption capacity-based functional units to assess the sustainability of pine bark derived adsorbents. In addition to conventional midpoint indicators, cumulative energy demand and endpoint impacts were evaluated. The study benchmarked different physical and chemical activation strategies against alternative adsorbents and employed a prospective scale up LCA framework to explore industrial optimization of activated carbon production. End-of-life scenarios were also assessed to determine the potential for emission mitigation through alternative disposal strategies. By integrating experimental data with LCA

modeling, this work offers a systematic and quantitative pathway toward sustainable adsorbent development.<sup>199</sup>

Maria Nelly Garcia Gonzalez and co-workers conducted an LCA of silicate titanate nanotube chitosan beads used for cadmium removal from wastewater. Environmental impacts associated with nanomaterial synthesis, adsorbent production, use, and recycling were evaluated. The synthesis stage emerged as the dominant hotspot due to high electricity consumption, indicating that energy efficiency improvements are essential during scale up. Although granular activated carbon exhibited the lowest environmental impacts, the results emphasized the need to prioritize optimization of both energy and chemical use in emerging adsorbent technologies.<sup>200</sup>

Kavya Bisaria *et al.* performed a laboratory scale LCA comparing magnetic stirrer and ultrasonicator synthesis routes. The assessment considered the synthesis of 1 kg of nanofibrous composite and treatment of 1000 L of arsenic contaminated water, from an initial concentration of 50 mg L<sup>-1</sup> to World Health Organization acceptable limits. Environmental impacts associated with material handling and adsorbent recycling were included. Electricity consumption and chemical usage, particularly nickel and liquor ammonia, were identified as dominant contributors to global warming, human toxicity, freshwater ecotoxicity, and marine ecotoxicity. The manufacture of the nanomaterial was the most energy intensive step, highlighting the importance of reducing electricity demand during scale up. Comparisons with granular activated carbon revealed lower environmental impacts relative to layered double hydroxides.<sup>194</sup>

Despite these advances, conventional adsorption studies continue to prioritize kinetic performance and adsorption capacity, often neglecting cradle-to-grave environmental impacts.<sup>201</sup> Mohanrasu *et al.* emphasized that LCA enables the identification of cost drivers and environmental hotspots across raw material extraction, production, use, and disposal stages, thereby supporting more sustainable adsorbent design.<sup>20</sup>

Ivan Kozyatnyk and colleagues compared the environmental impacts of end-of-life management options for activated carbon, biochar, and hydrochar used in wastewater treatment. Incineration, regeneration, and landfilling were evaluated. Heavy metal emissions during production were identified as major contributors to carcinogenic and freshwater ecotoxicity impacts. Regeneration and the use of higher capacity materials were shown to reduce overall environmental burdens, while heat recovery during incineration resulted in net negative impacts across several categories. Recirculation of hydrothermal carbonization process water reduced freshwater ecotoxicity and eutrophication impacts.<sup>202</sup>

Similarly, an LCA of Ni-Fe layered double hydroxide-chitosan adsorbents for arsenic removal identified electricity consumption during nanomaterial synthesis as the primary environmental hotspot. Chemical usage, particularly nickel and liquor ammonia, contributed significantly to toxicity related impacts. Repeated regeneration cycles increased environmental burdens due to additional energy requirements for drying. Sensitivity analysis demonstrated substantially lower carbon dioxide emissions when renewable electricity sources were employed compared to fossil fuel dominated electricity mixes.<sup>194</sup>



Overall, from an LCA point of view, it is evident that although adsorbents have significant potential to realize a better environmental performance, problems persist. Comprehensive cradle-to-grave LCAs are urgently needed to verify the true environmental advantages of adsorbents compared to established alternatives.<sup>190</sup>

## 7. Conclusions

This review underscores the central importance of cost management in both the synthesis and regeneration of adsorbents. The synthesis process plays a strategic and multifaceted role in determining adsorption performance, with multiple parameters, individually or in combination, directly influencing cost. Key factors such as preparation and modification costs, process complexity, yield, surface area, temperature, composition, and contact time are critical in evaluating the economic feasibility of adsorbents. Choosing appropriate synthesis and regeneration pathways represents a major cost challenge that requires further innovation. In addition, the use of specific additives during fabrication can further increase overall expenses. Addressing these economic factors is essential to ensure the commercial viability of emerging adsorbent technologies. This review also integrates both scientific and strategic considerations by categorizing adsorbents based on their cost profiles for synthesis and regeneration. Materials such as graphene oxide, silica, carbon nanotubes, and MOF-based composites are classified as high-cost adsorbents due to their expensive production and regeneration processes. Future research should focus on lowering these costs by employing simpler, greener, and more scalable methods, thereby expanding their versatility and application potential. In contrast, activated carbon (AC) and clays benefit from low synthesis costs and ease of preparation but are hindered by high regeneration expenses. Agricultural waste-based adsorbents stand out as an optimal low-cost option, offering both low synthesis and low regeneration costs. However, the regeneration and reutilization potential of low-cost composites still requires further investigation.

Compared to standalone adsorbents, composites offer dual functionality, combining multiple active components and exploiting synergistic interactions to enhance performance. Given the current momentum from academia and industry, the diverse types and synthesis strategies of composite adsorbents are expected to drive a surge of research in the coming decades, particularly in regeneration, recyclability, scalability, and stability. However, despite their potential, cost remains a significant barrier to their widespread adoption. Repurposing spent adsorbents is gaining traction, offering tangible economic and industrial benefits. The reuse of adsorbents is a key priority in the chemical and manufacturing industries to minimize both environmental impacts and operational costs. Spent adsorbents can make a substantial contribution to a circular economy, promoting resource conservation and reducing waste.

The sustainable management of spent adsorbents is therefore a crucial environmental engineering challenge. Emerging field practices and sequential application strategies offer promising pathways to enhance performance, lower costs, and improve long-term sustainability compared to traditional single-use

systems. By consolidating recent advances and identifying current gaps, this review provides a comprehensive roadmap for future research. It highlights the urgent need to address synthesis and regeneration costs as a key driver for achieving sustainable development goals, enabling broader commercial adoption and advancing the field of adsorption technologies.

A range of technologies have been utilized to remove pollutants from water and wastewater, with adsorption being the most commonly employed method due to its simplicity and cost-effectiveness. The choice of adsorbent is typically based on either high adsorption capacities for various pollutants or cost-effectiveness. Among natural adsorbents, activated carbon is considered the most effective for pollutant removal, though its high regeneration cost limits its use. Agricultural waste-based adsorbents are an example of an optimal representative in different categories, due to the fact that they are included in the low synthesis cost–low regeneration cost range, while all other alternatives lie in a high regeneration range. Reusing spent adsorbents can be environmentally beneficial and help reduce overall costs, but the regeneration process often involves complex procedures that increase operational costs and energy consumption, limiting their sustainability. Cost, controllability, and scalability are significant challenges for the practical use of multifunctional adsorbents, especially since their synthesis can be complex, and they often exist at the nanoscale. The strategic role of synthesis in the adsorption evaluation equation is critical. The results reveal that a complex interplay of several parameters either on their own or in combination that can be influential. The underlying reasons for the selection of regeneration or synthetic pathways are complex and involve both scientific and strategic components. Synthesis methods present a significant cost challenge, requiring further innovation. Additionally, some additives used during the fabrication process can be expensive. However, the cost of regeneration itself has not been extensively studied. Economic considerations, such as the expenses related to regeneration and synthesis, need to be addressed to ensure that adsorbent advancements are commercially viable. While low-cost adsorbents may offer lower performance compared to high-cost alternatives, their availability and affordability can compensate for these limitations. These low-cost adsorbents are promising for pollutant removal and recovery from wastewater, especially when combined with their recyclability. Looking ahead, we emphasize three key points for the future of spent adsorbents: (1) aligning spent adsorbents with regeneration and repurposing principles within a circular materials economy, (2) ensuring that spent adsorbents' reuse remains relevant to application-specific needs, and (3) advancing research on adsorbent synthesis to reduce production costs and close the lifecycle loop.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

All data generated or analysed in this study are included in this article. Any additional data related to this paper may be



requested from the corresponding author: kyza-s@chem.duth.gr; degkika@chem.duth.gr.

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