INTERFACE SCIENCE AND TECHNOLOGY

Advanced Low-Cost Separation Techniques in Interface Science

Edited by
George Z. Kyzas
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Advanced Low-Cost Separation Techniques in Interface Science

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Chapter 1

The impact of raw materials cost on the adsorption process

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1. Introduction

Recently, the "12 Principles of Green Chemistry" [1] have been more widely adopted and have been preferred over conventional methods. It has been used to a high extent for industrial separation and purification, ensuring that adsorbents are created in optimal conditions so that the optimal characteristics are expressed. The advantages of green synthesis include its cost-effectiveness, environment friendliness, scalability, and lack of requirement for extreme conditions like high pressure, energy or temperature, or the need to utilize toxic chemicals [2].

Organic dyes are a major type of water pollutants that may have severe environmental effects due to being highly toxic and may cause cancer [3,4]. Dyes are not easily degradable due to their stability and properties [5,6]. Multiple efforts have been made to address the issue of dye removal from wastewater systems, such as ion exchange, extraction of solvents, adsorption, use of filters, coagulation-flocculation, advanced oxidation and electrochemical methods [7−12]. Adsorption is one of the preferred methods because its operation mode is simpler and easier and it has little to none by-products compared with other methods [13]. As a result, the research on new and more effective materials has been getting more focus by scientists worldwide [13−16].

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A variety of methods have been utilized to address dye residues, including coagulation, irradiation, photocatalytic oxidation, and more. Adsorption is a cost-efficient and easy-to-apply method for the removal of existing dyes [17]. Physicochemical and biological processes are utilized for dye treatment in polluted wastewater [18]. The most common methods are coagulation-flocculation [19] and advanced oxidation process [20]. Nevertheless, such options are costly and have other operational issues, such as toxic by-products, lower removal effectiveness, and requirement for better matches to specific group of dyes [21].

The adsorption process is frequently used for the treatment of organic pollutants owing to its simplicity, performance, lack of sensitivity to toxic materials, and scalability [22]. Adsorption is considered to be better than alternative options for water reuse based on starting costs, clean design, and insensitivity to toxic substances [23,24]. Adsorption can be used either in single or combinational mode for partial or total wastewater cleaning. Recently, different, cheaper materials, such as biosorbents, industrial and agricultural waste, zeolite, silicon, and clays, have been tested as adsorbents at the third stage of waste treatment, thus replacing activated carbon (AC) [25–28].

2. Market trend for various adsorbents

Fig. 1.1 presents a comparison of the market trends and future projections for different adsorbents. There is a big increase in market size, with chitosan and activated carbon displaying the highest rise individually.

Activated carbon. The AC market might be split into different categories, including powdered, granular, and palletized, depending on the final product form. The high demand for powdered AC in applications such as decolorization and deodorization in pharmaceuticals is likely to enhance the market growth [29]. According to Statistics MRC, the global activated carbon market reached about $4.12 billion in 2017 and is anticipated to go beyond $14 billion by 2026 [30,31,32].

Chitosan. The global chitosan market report explores chitosan and its most common applications such as water treatment, cosmetics, food, and pharmaceuticals, among others. The most significant driver of the chitosan market is the abundance of the source material along with developing applications in various fields [33]. The size of the global chitosan market is expected to surpass $2500 million by 2022, compared with $1205 million in 2015 [34].

Graphene. Graphene is a carbon allotrope that contains graphite and diamond. The most common graphene types that form the market include graphene oxide; nanoplatelets; mono-, bi-, or few-layer graphene. The global graphene market is expected to achieve a noticeable growth of more than 811 million USD by 2023 [35–37].

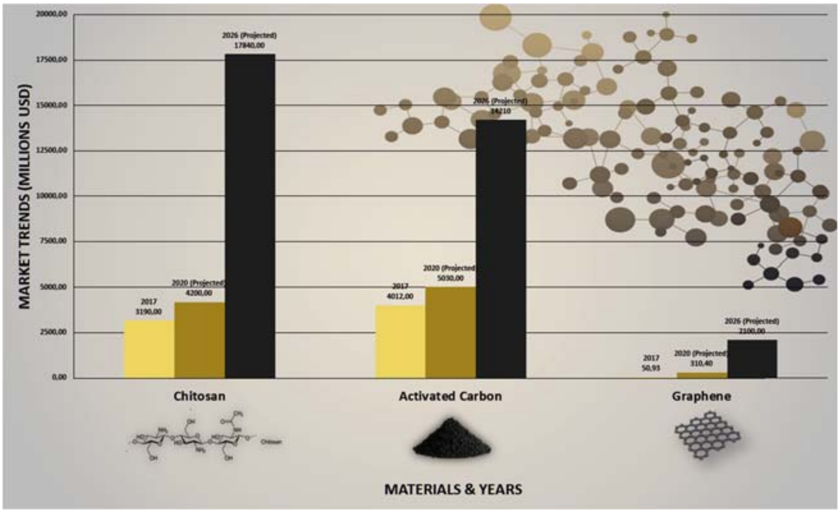

FIGURE 1.1 Trends and future projections for different adsorbents.

3. Activated carbon efficiency

ACs stem from natural materials like wood and coal, which renders their production and regeneration process quite high in price [38,39]. Many scientists have thus researched potential alternate low-cost adsorbents production processes. AC has been suggested as an adsorbent because of its structure and large surface which make it a great candidate for the removal of organic pollutants (including hard biodegradable ones) from aquatic environment. It is considered as one of the most efficient adsorbents and is used widely due to its nontoxicity for purifying drinking water and treating wastewater [22]. More specifically, AC's efficiency in removing a large variety of dyes from wastewaters has rendered it an excellent option when compared with more expensive alternatives [40].

AC is commonly retrieved from substances with high carbon content. Its adsorption ability is affected by its porous structure and surface area, which can be adjusted, thus making AC suitable for a wide application range, including pollutant removal and catalyst support. AC is not cheap, and the better the quality, the higher its cost. All methods of spent carbon regeneration, however, are costly, are impractical, or result in further waste discharge [41], despite its efficiency as adsorbent [42]. Despite its multiple applications in cleaning processes, AC is still quite costly. Commercial AC is limited due to cost, lower performance, as well as efficiency after regeneration [43].

So far, natural adsorbents, such as chitin, biomass, and chitosan, as well as industrial and agricultural waste have been for the treatment. Bioadsorbents are considered more selective, less costly, and more effective than resins and commercially activated carbon [44]. Carbon-based materials are widely used in dye removal processes [13]. During the past few years, graphene-based materials have been tested in various areas, due to attributes that make them ideal candidates, such as physicochemical, optical, and other properties [45]. Recently, they have been successfully tried as alternative adsorption options [46,47]. Graphene is, however, costly and not too easy to produce; therefore significant efforts are dedicated toward finding cheaper yet still effective ways to take advantage of graphene and related materials. Graphene oxide (GO) is one such material created by graphite oxidation [48]. Graphene oxide has shown remarkable adsorption abilities compared with other carbon-based materials and thus has an important role in wastewater treatment. Furthermore, due to its comparatively high surface area (2630 m^2/g), it has received even more attention [49]. Graphene oxide may assist in removing cationic metals and dyes due to its negative charge density. Graphene oxide sheets are made of a single-layered graphite structure connected with oxygen atoms and encompass carboxyl, ketone, epoxy, and hydroxyl groups, which transmit negative charges when in liquid solutions, in a broad pH range.

4. Biosorbents efficiency (the case of chitosan)

Biopolymers such as chitosan and chitin have emerged as effective adsorption solutions [44]. Chitosan is useful as it possesses amino and hydroxyl groups, which may act as the active sites [50] and is also nontoxic, noncorrosive, and safe to manage [51]. The main advantage of chitosan is that it is able to bind dyes in low concentrations as opposed to metal salts and is also effective in cold water. Chitosan composites are cost-effective due to their easy preparation requirement of cheaper chemical reagents. The removal of even the small amounts of pollutants is very expensive, so traditional methods are not practical [3]. Currently, multiple adsorption methods based on chitosan composites are being researched as a substitute for the pricier conventional methods [52].

5. Agricultural waste efficiency

Agricultural solid waste such as leaves, fruits, seeds, and peels, as well as forest waste materials such as sawdust and bark, have been widely used as adsorbents for dye removal [53]. Their availability reaches extremely high quantities, and their physicochemical attributes make them useful as sorbents. Several agricultural solid wastes are effective in removing both cationic and anionic dyes; however, they require activation [54,55]. The most significant factor modifying the adsorption is the pH; a high pH allows for the adsorption of cationic dyes, whereas a low pH is more effective on anionic dyes· [54]. A review by Demirbas [15] provided a comprehensive list of agricultural waste and a variety of otherwise unwanted plant parts that provide a cheap and renewable supplementary source of AC. These materials have little to none economical value and would commonly be problematic to dispose, they are used in natural or modified form for dye removal. The most commonly used forms of agro-waste include pine wood, corn cob, nut shells, fruit peels, bamboo, bark [15]. Furthermore, they have desirable physicochemical attributes that render them highly efficient and easy to renew with or without being treated (they occasionally might need to be grounded, washed, and/or dried) which lowers the production and energy costs [56]. An example of agro-wastes with potential are banana and orange peels that can be used as adsorbents for the removal of copper, zinc, carbon dioxide, nickel, and lead cations from water. The residues of the peels can be treated and modified to become adsorbents due to their large surface areas, swelling capacity, strength, ease of use, and ability to adsorb hazardous pollutants.

6. Economics of adsorption

Cost is a significant factor when comparing sorbent substances. The adsorbent cost is influenced by a variety factors, such as availability, type (natural, agricultural, industrial, synthesized, by-products, etc.), conditions, recycle, stability, production country (in terms of development stage [57,58]. Due to the continual changes in adsorption, the scientific community has concentrated its efforts on developing a cheap, simple, and eco-friendly process for the removal of dyes. Traditional synthesis processes have many limitations due to their environmental risks and high costs [17]. Therefore efforts are concentrated toward establishing less expensive, more effective, and scalable separation processes as required by the industry. The feasibility of using low-cost agriculture-based waste materials has been widely studied, and biosorption technology has been found to be a sufficient alternative for the removal of heavy metals from wastewater [59]. Agricultural wastes are cheaper and often environmentally friendly due to their chemical attributes, abundancy, renewability, low cost, and efficiency for both organic and inorganic waste treatment [60].

7. Materials and methods

The information was collected from a variety of sources, primarily via in-person interviews with the managers of two Greek chemistry laboratories in Greece. The managers were chosen due to their expertise in the field of adsorption. The focal point of the interviews was the identification and analysis of the characteristics of “popular” adsorbents.

The advantage of personal interviews lies in the ability to observe the interviewee's expressions and feelings as well as listen to their responses unobtrusively. The chosen approach consisted of unstructured interviews since they are more flexible for all participants, as proposed by Gubrium and Holstein [61]. Furthermore, there is also the chance to further discuss specific "elite" issues of importance that might be brought up during interviews [62,63]. The term "elite" refers to experts on specific subjects [64]. The basic issue in such cases is to reach a balance between proper reporting of research outcomes and the potential risk of disclosing interviewees' identities, as opposed to hiding their data for less exposure but higher risk of results being disputed [65]. The confidentiality is necessary to protect the interviewees. Anonymity permits participants to express more freely [66]. For this specific research, the respondents are male professors. The interview was detailed, taking up more than 40 min. Based on the responses, we determined two important parameters affecting costs. These consist of the raw material and the adsorption energy costs.

Recipe selection: In order to further explore the adsorption phase and identify the attributes shaping the studied adsorbents, we performed a literature search to find the corresponding recipes. The most cited ones were chosen for each adsorbent, so that the cost parameters could be calculated.

Raw material price: The first cost factor to be estimated is the raw material cost. These costs refer to the necessity for the adsorption process resources. Chesbrough [67] proposes a new way of cooperation of various stakeholders in the value chain in order to gain new knowledge and technologies. The analysis included the compilation of market prices for the materials based on information of various vendors worldwide.

Energy cost: The second cost factor consists of energy costs. This refers to the energy utilized for the various stages of adsorption. Electricity costs are estimated based on the energy price in Greece for 2019 (0.194 Euro/kWh). These prices are provided by the Public Power Corporation S.A., Hellas [68]. The cost in Euros has been calculated as the energy cost in kWh multiplied by the price of energy in Greece as Euros per kWh.

Maintenance cost and labor cost: These costs are not included because corresponding data are not available for the adsorption phase.

8. Results and discussion

The effects of different parameters including solution pH, biosorbent's dosage, initial dye concentration, and contact time were studied. Table 1.1 attempts to provide an up-to-date list on adsorption properties or recycled and reused waste by-products.

A combination of multiple adsorbents (Table 1.1) has been found to yield promising results; however, this comes with the disadvantage of increased costs. Therefore, in spite of their effectiveness and suitability, certain financial considerations might limit the use of costly adsorbents.

TABLE 1.1 Synthesis recipes of the adsorption of dyes using different adsorbent (dosage: 1 g of adsorbent per 1 L of adsorbate solution).

Adsorbent	T(°C)	pH	Contact time (min)	Removal (%)	Pollutant	Ref
Graphene oxide (GO)	65	3	180	55%	Dye	[69]
GO-Chm	65	3	180	81%	Dye	[69]
Magnetic activated carbon	65	10	300	68%	Dye	[70]
CD-MIPs	25	2	225	62%	Dye	[71]
CHI-MIPs	25	2	225	62%	Dye	[71]
Chitosan biosorbent	65	12	120	97%	Metal	[72]
Chitosan biosorbent	65	4	120	85%	Metal	[72]
Coffee biosorbent	25	5	180	UCR: 70%; TCR: 77%	Metal	[73]
Coffee biosorbent	25	5	180	UCR: 60%; TCR: 62%	Metal	[73]
GO/CS	65	6	150	93%	Metal	[74]
GO/mCS	65	6	150	96%	Metal	[74]
GO	65	3	200	53%	Drug	[75]
CSA	65	3	200	68%	Drug	[75]
GO/CSA	65	3	200	93%	Drug	[75]
Activated carbon	65	3		B0: 50%; B1: 80%; B3: 82%; B4: 83%; B5: 65%	Drug	[76]
Cs	65	10		50%	Drug	[77]
CsNCB	65	10		78%	Drug	[77]
CsSLF	65	10		82%	Drug	[77]
Gho	65	2	180	PRO: 68%; ATL: 93%	Drug	[78]

ATL, atenolol; *CD-MIPs*, β-Cyclodextrin MIPs; *CHI-MIPs*, chitosan MIPs; *CSA*, chitosan grafted with poly(acrylic acid); *CsNCB*, chitosan grafted with N-(2-carboxybenzyl groups); *CsSLF*, chitosan grafted with sulfonic groups; *PRO*, propranolol; *TCR*, treated coffee residues; *UCR*, untreated coffee residues.

In general, a potential low-cost dye adsorbent should show (1) effectiveness in removing a wide range of dyes; (2) high ability and rate of adsorption; (3) differentiation in selectivity based on concentration; and (4) tolerance of a variety of wastewater attributes. Low-cost adsorbents possess certain physical and chemical characteristics such as pore size, surface area, and physical strength, as well as a range of advantages and disadvantages that affect their efficiency in treating wastewater [44].

Raw material costs are considered "hard" costs—therefore any savings in these will allow for better profits directly. The high prices of raw materials makes stakeholders nervous about the development and exploitation of new technologies and how those can be affected by fluctuations in price and availability [30].

The results from Table 1.2 reveals that only the raw material cost actually matters for the adsorption phase. The reduced consumption of raw materials could result in lowered costs and other benefits, including an improved laboratory image and working conditions.

This work presents an initial attempt to optimize the dye removal during the adsorption stage; however, an effective methodology addressing the

TABLE 1.2 Raw materials and energy costs in selected recipes.

Material produced	Raw materials	Raw materials cost per 1 g of final product (€)	Energy cost (€)	Recipe cost (€)
Polymer/graphene oxide—synthesis of magnetic nanoparticles	$KMnO_4$ (>99.0%); graphite flakes; H_2SO_4 (95%–98%); H_2O_2 (30 wt%)	5.75	5.02	10.77
Polymer/graphene oxide—synthesis of magnetic chitosan (Chm)	$FeCl_2$ $4H_2O$ (p.a. >99.0%); chitosan (high molecular weight); $FeCl_3$ 6 H_2O (reagent grade, 97%); glutaraldehyde (50 wt% in water; acetic acid solution (>99%) $KMnO_4$ (>99.0%); graphite flakes; H_2SO_4 (95%–98%); H_2O_2 (30 wt%)	2.4	3.35	5.75
Polymer—synthesis of sulfonate-grafted	Dichloroacetic acid (>99%); formamide	3.22	3.89	7.11

TABLE 1.2 Raw materials and energy costs in selected recipes.—cont'd

Material produced	Raw materials	Raw materials cost per 1 g of final product (€)	Energy cost (€)	Recipe cost (€)
chitosan adsorbent (CsSLF)	(>99.5%); chitosan (high molecular weight); glutaraldehyde (50 wt% in water); acetic acid solution (>99%)			
Polymer—synthesis of N-(2-carboxybenzyl)-grafted chitosan adsorbent (CsNCB)	Chitosan (high molecular weight); glutaraldehyde (50 wt% in water); acetic acid solution (>99%); 2-carboxybenzaldehyde (97%); sodium borohydride (>96%)	11.39	0.84	12.23
Polymer—synthesis of cross-linking chitosan adsorbents	Chitosan (high molecular weight); glutaraldehyde (50 wt% in water); acetic acid solution (>99%): 1L	2.09	4,29	6.38
Graphene oxide—synthesis of adsorbents according to a modified Hummers method	$KMnO_4$ (>99.0%); graphite flakes; H_2SO_4 (95%–98%); H_2O_2 (30 wt%)	5.57	2.47	8.04

removal of all kinds of dyes in an inexpensive manner has been found. The outcomes suggest that there are some cheaper adsorbents that allow for higher dye removal rate, as shown in Table 1.1. Such low-cost adsorbents arose as an alternative option, and they support practical solutions to toxicity-related issues.

9. Conclusions

This chapter has presented a broad range of different adsorbents that can be utilized as low-cost adsorbents. The usage of such cheap adsorbents is highly advisable due to not just their low cost but also availability, renewability, and effectiveness. The advantage of low raw material cost should be further explored in terms of regeneration, design, and confinement of the waste material for improved effectiveness and recovery rates. The outcomes of this work could help in establishing a well-founded, environmentally friendly, simple, and cost-effective process for green synthesis especially with the constant developments in its application fields.

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