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Regeneration and reusability of non-conventional low-cost adsorbents to remove dyes from wastewaters in multiple consecutive adsorption–desorption cycles: a review

Noureddine El Messaoudi¹  · Mohammed El Khomri¹ · Abdelaziz El Mouden¹ · Amal Bouich² · Amane Jada³ · Abdellah Lacherai¹ · Hafiz M. N. Iqbal⁴ · Sikandar I. Mulla⁵ · Vineet Kumar⁶ · Juliana Heloisa Pinê Américo-Pinheiro^{7,8}

Abstract

The global economy's growths due to industrialization, accompanied by urbanization high rate, have led to severe anthropogenic pollution impacting both the environment and human health. Regarding particularly the aquatic systems' chemical pollution, as resulting mainly from the release of liquid effluents containing dye molecules and organic pollutants, it is therefore necessary to use innovative processes for treating the polluted water. At first sight, adsorption seemed like an excellent process due to its simplicity and efficiency, but this technique simply moves the dye from an aqueous medium to the adsorbent surface, thus creating sludges which are new pollution forms. Hence, we need to treat this polluted adsorbent in order to recover the removed dye and to reuse once more the adsorbent for others dye removal adsorption cycles. This review focuses on regenerating low-cost adsorbents (LCAs), saturated with dyes, by using different desorbing agents and on exploring their potential to be reused. These low-cost materials have been classified into four classes based on their origin: agricultural solid wastes; industrial by-products and wastes; natural materials; and sea materials and biomasses. Different chemical agents such as acids, bases, salts, organic solvents, alcohols, and chelating reagents have been used as eluents to regenerate the dye-covered adsorbents. Therefore, in the present review citing 210 literature articles, it is demonstrated the excellent regeneration and high reusability of the LCAs suited for dye removal from environmental wastewater. In this review paper, we show that the agricultural solid wastes (39.56%) are the most regenerated and reused, in comparison to, sea materials and biomasses (22.31%), industrial by-products and wastes (18.71%), and natural materials (19.42%).

Keywords Dyes · Low-cost adsorbents · Desorbing agent · Regeneration · Reusability · Wastewater treatment

Abbreviations

AB25 Acid blue 25
AR73 Acid red 73
AR88 Acid red 88
AO7 Acid orange 7

AO10 Acid orange 10
AV49 Acid violet 49
BB Bromophenol blue
BF Basic fuchsine
BG Brilliant green

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BR	Basic red
BR2	Basic red 2
BR0	Basic red 9
CR	Congo red
CV	Crystal violet
DB	Drimaren blue
DB73	Disperse blue 73
DG	Drimaren gold
DR	Drimaren red
DY28	Direct yellow 28
EBT	Eriochrome black T
EY	Eosin yellow
IC	Indigo carmine
LCA	Low-cost adsorbent
MB	Methylene blue
MG	Malachite green
MO	Methyl orange
MV	Methyl violet
MV2B	Methyl violet 2B
NR	Natural red
RB5	Reactive black 5
RB19	Reactive blue 19
RB49	Reactive blue 49
RBBR	Remazol brilliant blue R
RE5	Red 5
ReB5	Remazol black 5
Rh6G	Rhodamine 6 g
RhB	Rhodamine B
RO122	Reactive orange 122
RY	Reactive yellow
RY86	Reactive yellow 86
RY145	Reactive yellow 145
RY176	Reactive yellow 176
PB	Pyronin B
SO	Safranin-O
Y6	Yellow 6

1 Introduction

Colored wastewater is caused by the presence of synthetic organic dyes [1]. It is regarded as a polluted water resources having many negative impacts on human health and aquatic life [2–8]. It is noteworthy to note that the ingestion of these dyes causes cancer, mutagen, teratogen, cardiovascular shock, vomiting, gastrointestinal pain, diarrhea, etc. (Fig. 1). Thereby, there is an urgent need to develop treatment technologies for the water solutions polluted by the dye. Scientific workers have made great efforts to investigate various removal techniques for dye, including biological flocculation, electrolytic route, Fenton oxidation, photocatalysis, membrane, and adsorption [9–16]. Among these methods, adsorption plays an important role because of its low cost,

simple operation, and high removal efficiency, making it a reliable removal technique [17–25]. Adsorption is a physico-chemical technique based on the mass transfer between two phases: liquid–solid, liquid–liquid, liquid–gas, and gas–solid [26, 27]. Adsorption efficiency (%) was calculated using Eq. 1 [28–30]:

$$\%Adsorption = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

In the above formula, C_0 (mg/L) and C_e (mg/L) are, respectively, the initial concentration and the final concentration at the equilibrium of the organic pollutant.

The dyes' adsorption performance from water onto LCAs depends on the properties of the adsorbent used, such as its surface area, its pore volume, and its pore distribution, which may be affected by the LCA chemical composition and the thermal conversion process conditions, as well as, the chemical activation [31–33]. Note that the dye-covered low-cost adsorbent (LCA) obtained from wastewater treatment may pose a significant threat when it is disposed directly into the environment, due to the possibility of the dye leaching [34, 35]. Therefore, a sustainable approach to mitigate the negative environmental impact resulting from such secondary pollutant is to regenerate the LCA [36, 37]. However, the regeneration and the reuse of adsorbents ensure that the entire remediation process is cost-effective [34, 38, 39]. The regeneration process basically shifts the adsorption equilibrium from adsorption to desorption [40–42]. A number of researchers regenerate and reuse the LCAs for eliminating dyes from wastewater for different reasons: these materials are abundantly available in large quantities, are environmentally friendly materials, have high regeneration percentages, are easy to regenerate, and are excellent in reusability for eliminating dyes [43–53].

Several techniques are used in the regeneration of the LCAs for dye removal from wastewaters such as the chemical method, the biological method, the advance oxidative method, the thermal regeneration, the ultrasound regeneration, the microwave-assisted method, and the electrochemical regeneration [34, 54–56]. Table 1 represents the advantages and disadvantages of some adsorbent regeneration methods. Chemical regeneration is a simple technique in which acids, alkalis, and organic solvents having either oxidizing or solubilizing capacity are used to dissolve the adsorbates and to restore the adsorption ability of the LCA [36, 57, 58]. The chemical regeneration method has the main advantages of high regeneration efficiency, almost zero carbon loss, quick regeneration, and possible adsorbate recovery [34, 59]. It is useful, especially when the adsorbates form strong bonds with the adsorbent's surface [60, 61]. The regeneration capacity of any adsorbent depends on the solution pH and the rate of oxidation and degradation by

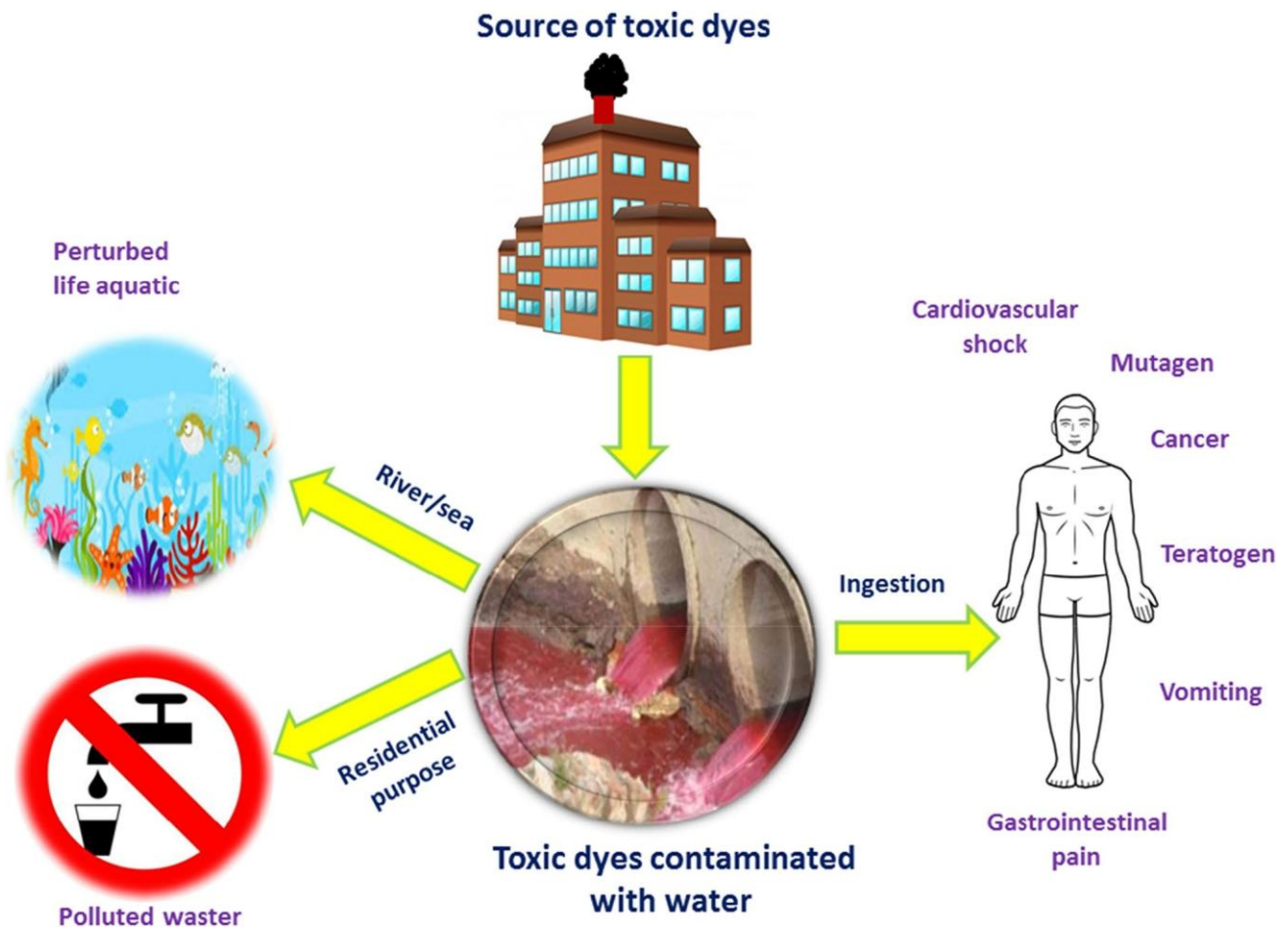


Fig. 1 Impacts of the toxic dyes on the environment, aquatic life, and human health

Table 1 Advantages and disadvantages of some adsorbent regeneration methods [81]

Regeneration method	Advantages	Disadvantages
Biological method	Efficient, less time-consuming, eco-friendly	Very slow, block the pores of adsorbent, only applicable for biodegradable pollutants
Thermal regeneration	Efficient	Highly expensive, degrade the pores of adsorbent, time-consuming, create air pollution
Advance oxidative method	Efficient	Highly expensive, degrade the pores of adsorbent, time-consuming
Microwave regeneration	Efficient, less time-consuming, eco-friendly	Highly expensive, degrade the pores of adsorbent, required further treatment
Ultrasound regeneration	Efficient, less time-consuming, eco-friendly	Highly expensive, degrade the pores of the adsorbent

complexion [59, 62]. The thermal regeneration under an inert atmosphere has been widely used due to its simplicity and versatility [63, 64]. During this process, exhausted adsorbents are exposed to high temperatures (500–800 °C) in an inert gas, requiring subsequent oxidation of released volatile compounds [65, 66]. Recently, the application of the microwave heating method in LCA regeneration has been employed by many researchers [67–70]. The improvement of LCA regeneration via the microwave method results from

its heating mechanism, where the energy supplied directly penetrated into the carbon bed and then it is transformed into heat inside the carbon matrix by dipole rotation and ionic conduction [71–73]. Electrochemical regeneration is a promising option, having characteristics which include environmental compatibility, energy efficiency, safety, selectivity, and cost-effectiveness [74–76]. Among all the regeneration techniques, no stimulating technique has been individually found to keep or to improve the adsorption efficiencies of all

adsorbents. Indeed, researchers are more inclined toward the regeneration and the reuse of adsorbents due to their production high cost, their stabilization, and their disposal [77]. It is cost-effective to regenerate LCA for their reuse during the pollutants' sorptive removal from wastewater. However, the used LCAs have known limitations in terms of their cost, their regeneration, and their reusability which should be addressed [78]. Hence, the capacity of an adsorbent to regenerate and its cost are two of the greatest challenges associated with the adsorption process [79].

Desorption is the reverse reaction of adsorption; it is about removing the adsorbed molecule from the adsorbent surface by using different desorbing agents (Fig. 2). The percentage of dye desorption (%) is calculated by using Eq. 2 [80]:

$$\%Desorption = \frac{C_d}{C_a} \times 100 \quad (2)$$

where C_a (mg/L) and C_d (mg/L) are the concentrations of adsorbed and desorbed dye, respectively.

The present work reports on the regeneration and the reuse of non-conventional LCAs (Fig. 3) as prepared from agricultural solid wastes (sugarcane bagasse, wheat straw, nutshell, plant leaf, plant fiber, wood shaving, sawdust, oil palm wood, fruit waste, fruit peel, seed, tea waste, etc.), industrial by-products and wastes (fly ash, sludge ash, metal hydroxide sludge, bottom ash, red mud, etc.), natural materials (natural clay, zeolite, sepiolite, bentonite, soil,

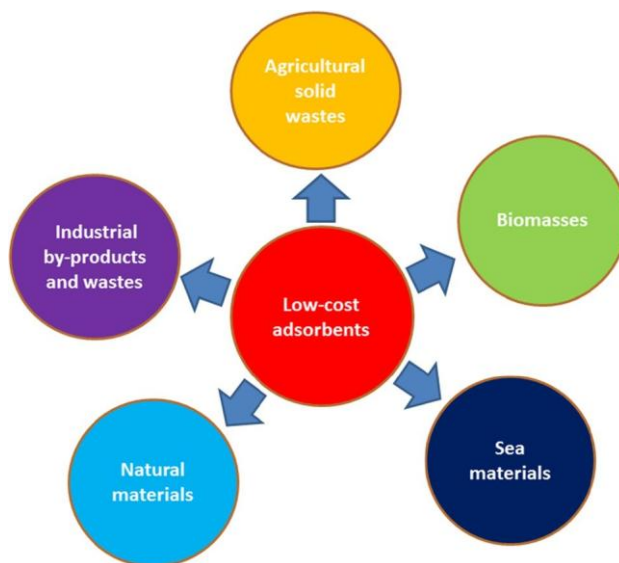
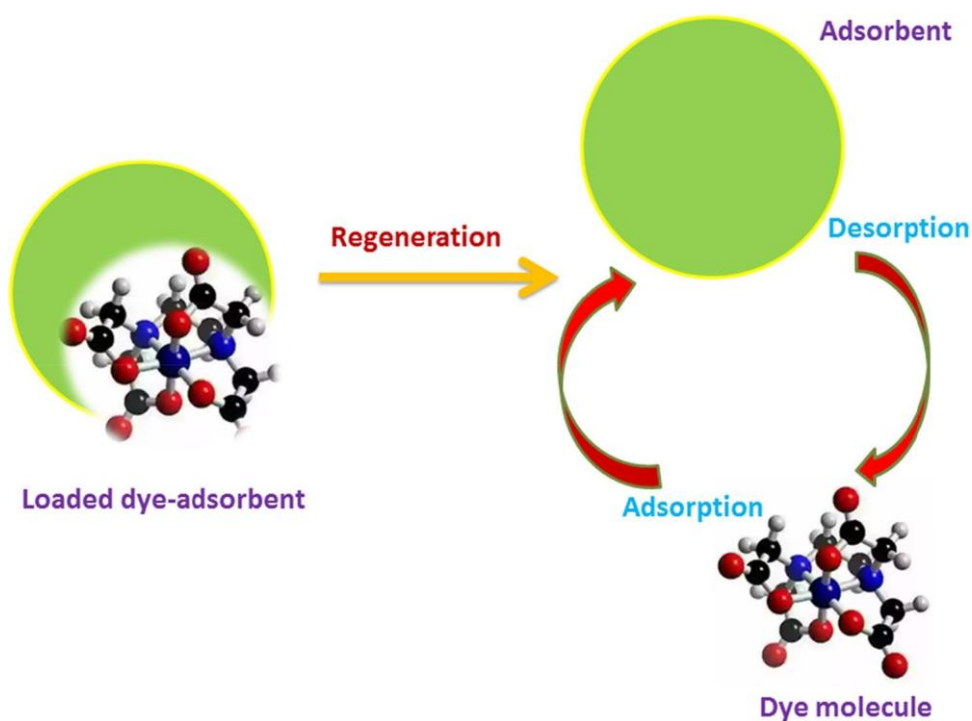


Fig. 3 Different types of non-conventional LCAs

montmorillonite, kaolin, halloysite, dolomite, siliceous materials, etc.), sea materials (alga, chitosan, and chitin, peat moss, seafood wastes, etc.), and biomasses (fungi, bacteria, yeast, etc.), to remove dyes from wastewaters in multiple adsorption–desorption cycles. Furthermore, different desorbing agents (acids, bases, salts, organic solvents, alcohols, water, and chelating reagents) for the regeneration of these materials are discussed.

Fig. 2 Dye adsorption and desorption from the dye-covered adsorbent



2 Regeneration and reuse of non-conventional LCAs

2.1 Agricultural solid wastes

Currently, available adsorbents are based on agricultural solid wastes, as the main, mostly non-renewable materials. Therefore, the development of renewable and recyclable agricultural solid adsorbents has contributed to the achievement of sustainable development. Many agricultural solid wastes have been developed, and they were used as renewable adsorbents with good adsorption properties [82–84]. However, relevant studies have shown that agricultural solid wastes have porous fibrous structures and contain various functional groups, like carboxyl and hydroxyl groups [85]. Regeneration of these material wastes for their reusability to eliminate dyes from wastewater is economically feasible with high adsorption efficiency mostly due to their physicochemical characteristics [86].

In this context, El Messaoudi et al. [80] have regenerated and reused the date stones and jujube shells for the Congo red (CR) removal in four biosorption–desorption cycles, using NaOH as the desorbing agent (Fig. 4); the biosorption efficiency was found to decrease from 94.91 to 73.24% for date stones and from 97.88 to 67.22% for jujube shells at a concentration of CR before biosorption of 100 mg/L and temperature of $50 < \text{°C}$ for 90 min of contact time. Similar results were reported by Al-Zoubi et al. [87] and Sharma et al. [88].

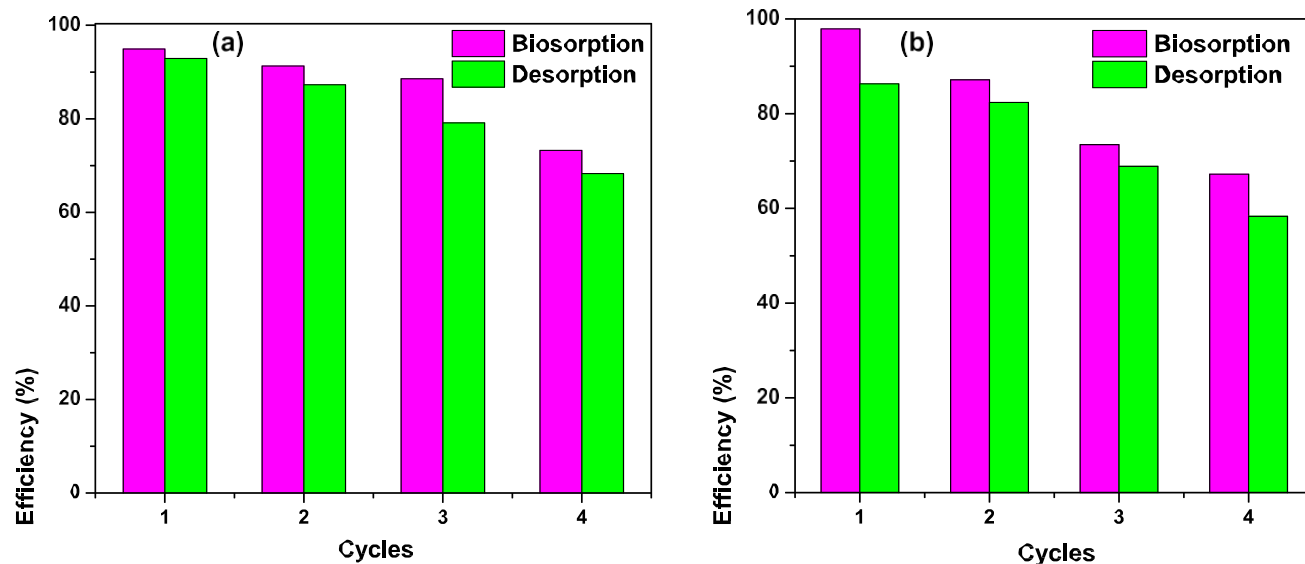


Fig. 4 Biosorption/desorption percentages cycles of CR from **a** date stones (*Phoenix dactylifera*) and **b** jujube shells (*Ziziphus lotus*) [80]

The reuse of treated *Pinus elliottii* sawdust to remove methylene blue (MB) four times was reported by Bortoluz et al. [89]. These authors have found that the removal percentage of MB ranged from 97 to 73%, between the first and the fourth cycle using 1 M HCl. A similar observation was reported by Mashkoo and Nasar [90] in the regeneration of magnetized *Tectona grandis* sawdust with 0.1 M HCl; the desorption capacity was 70.60% in the fourth cycle.

The main adsorbents for dye removal, which were prepared from agricultural solid wastes and were reported in the literature, are presented in Table 2. In general, the dye adsorption and desorption efficiencies, by using adsorbents originating from agricultural solid wastes, decrease after each cycle. Such a decrease was attributed to the decrease of the available adsorbent surface sites [91]. In addition, the NaOH and HCl aqueous solutions were found to be the best eluents for this regeneration.

2.2 Industrial by-products and solid wastes

Several industrial by-products have been proposed by several researchers for dye removal from wastewater [128, 129].

Dash et al. [129] studied the regeneration of modified coal fly ash to remove rhodamine 6G (Rh6G) and malachite green (MG) in 3 cycles of adsorption–desorption using NaOH ($pH = 8$) (Fig. 5). The Rh6G desorption efficiency decreased from 97.98 (first cycle) to 87.61% (third cycle), while MG desorption efficiency decreases from 97.38 (first cycle) to 88.31% (third cycle).

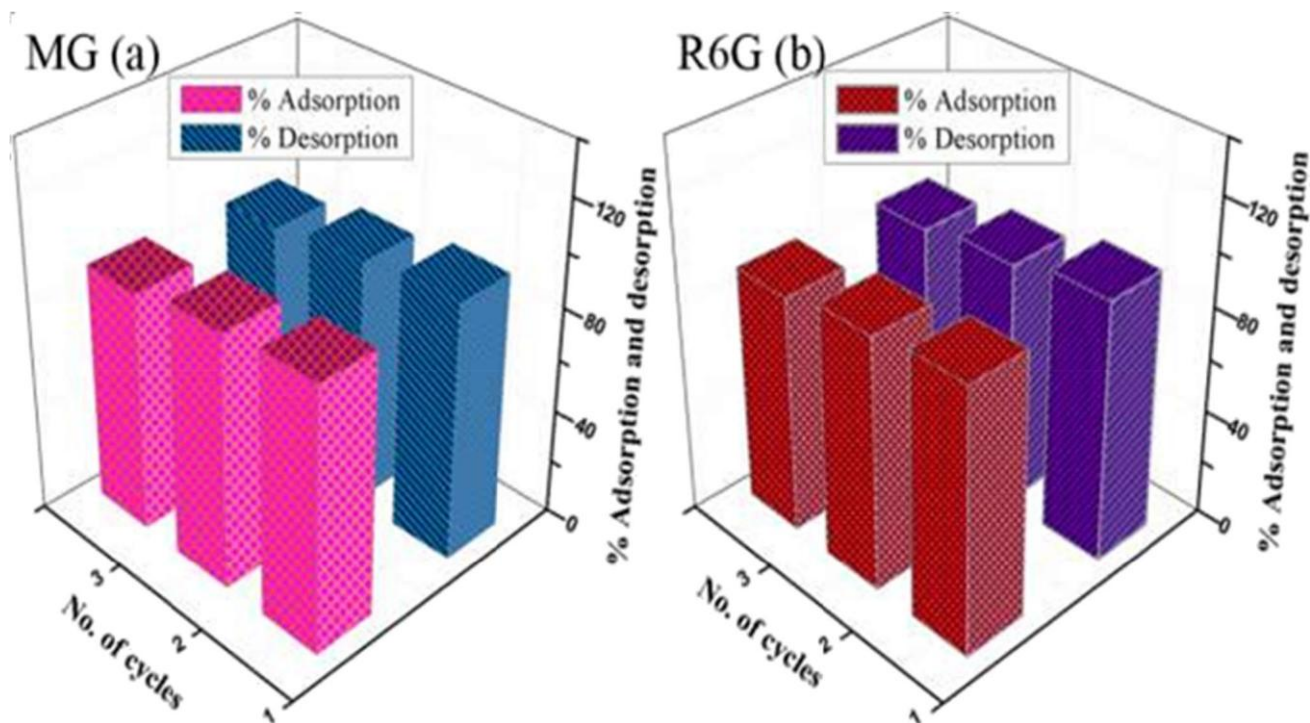
The regeneration and the reusability of the keratin protein (0.5 M wool) for rhodamine B (RhB) removal from

Table 2 Regeneration of adsorbents produced from agricultural solid wastes, for dye removal

Adsorbent	Dye	Ads. (%)	Des. (%)	N ^o of cycles	Desorbing Agent	Ref
<i>Clitoria fairchildiana</i>	Rh6G	90.27	21.78	2	C ₂ H ₅ OH	[92]
<i>Cornulaca monacantha</i> stem	CR	45.88	72.75	6	0.01 M NaOH	[88]
Date stones	CR	73.24	68.24	4	0.02 M NaOH	[80]
Jujube shells	CR	67.22	58.33	4	0.04 M NaOH	[80]
Olive pomace	DB73	100.00	67.00	5	CH ₃ COOH	[93]
Modified sugarcane bagasse	RB5	69.88	–	5	0.1 M NaOH	[94]
Modified sugarcane bagasse	AO10	93.6	–	5	0.1 M NaOH	[94]
Modified <i>Pyracantha coccinea</i>	MB	56.56	–	14	0.01 M HCl	[95]
Natural wheat straw	MB	57.20	–	3	0.1 M HCl	[96]
Modified wheat straw	MB	43.5	–	3	0.1 M HCl	[96]
<i>Salvinia minima</i>	MB	87.72	70.59	3	0.1 M HCl	[97]
Modified celery	MB	–	97.00	5	0.1 M HCl	[98]
Leaves of platanus	MB	68.00	–	3	Deionized water	[99]
Alkaline-treated orange tree sawdust	MB	29.49	–	5	0.5 M NaCl	[100]
Seeds of <i>Punica granatum L</i>	MB	79.00	–	2	CH ₃ COOH	[101]
Seeds of <i>Punica granatum L</i>	CV	85.00	–	2	CH ₃ COOH	[101]
Chemically treated olive pomace	CV	–	73.98	4	0.5 M HCl	[102]
Carboxylation kapok fiber	NR	74.20	–	10	0.1 M HCl	[103]
Carboxylation kapok fiber	MB	47.30	–	10	0.1 M HCl	[103]
Carboxylation kapok fiber	Rh6G	39.80	–	10	0.1 M HCl	[103]
Fatted jojoba residues	EBT	61.15	–	4	0.1 M NaOH	[87]
Fatted jojoba residues	CR	22.21	–	4	0.1 M NaOH	[87]
Walnut shell	MB	–	94.77	1	0.1 M HCl	[104]
Defatted jojoba residues	EBT	73.80	–	4	0.1 M NaOH	[87]
Defatted jojoba residues	CR	49.45	–	4	0.1 M NaOH	[87]
Modified sugarcane bagasse cellulose	MB	–	85.40	5	0.1 M HNO ₃	[105]
Modified sugarcane bagasse cellulose	CV	–	83.10	5	0.1 M HNO ₃	[105]
Alkali-treated orange tree sawdust	MB	–	93.00	1	0.5 NaCl	[106]
Treated <i>Pinus elliottii</i> sawdust	MB	73.00	–	4	1 M HCl	[89]
Jackfruit seed	MG	87.00	–	2	0.1 M NaOH	[107]
Magnetized <i>Tectona grandis</i> sawdust	MB	–	70.60	4	0.1 M HCl	[90]
Citric acid-modified sawdust	MB	–	90.00	3	0.01 M HCl	[108]
<i>Calotropis procera</i>	CR	32.00	–	6	1 M HCl	[109]
Modified- <i>Actinidia deliciosa</i> peels	CV	–	45.00	4	1 M CH ₃ COOH	[110]
Modified sugarcane bagasse	MB	–	78.30	5	TiO ₂ hydrosol (pH=2)	[111]
Mandarin peel	MB	86.12	–	3	1 M HCl	[112]
<i>Artocarpus odoratissimus</i> leaves	RhB	44.00	–	5	1 M HCl	[113]
<i>Terminalia ivorensis</i> seed waste	MB	–	86.37	1	0.1 M HCl	[114]
NaOH- <i>Artocarpus odoratissimus</i> leaves	MV2B	90.00	–	5	1 M NaOH	[115]
<i>Irvingia gabonensis</i> endocarp	RhB	–	81.82	1	CH ₃ COOH	[116]
Activated charcoal of <i>Thuja orientalis</i> leaves	RBRR	73.00	–	4	0.1 M HCl	[117]
Banana peel	CR	–	93.30	1	0.1 M NaOH	[118]
Acid modified-hazelnut shell	MB	90.00	–	5	0.5 M HCl	[119]
NaOH activated-betel nut husk	MB	74.00	–	5	NaCl/ C ₂ H ₅ OH	[120]
<i>Eragrostis plana</i> nees	CV	–	72.00	1	1 M CH ₃ COOH	[121]
NaOH treated- <i>Caesalpinia ferrea</i> fruits	MB	95	–	5	0.2 M HCl	[122]
Waste of <i>citrus sinesis</i>	RB19	–	35.97	1	NaOH	[123]
Waste of <i>citrus sinesis</i>	RB49	–	42.58	1	NaOH	[123]
H ₂ SO ₄ activated- <i>Gossypium hirsutum</i> seed	BR2	50.00	–	3	CH ₃ COOH	[124]

Table 2 (continued)

Adsorbent	Dye	Ads. (%)	Des. (%)	N ^o of cycles	Desorbing Agent	Ref
<i>Luffa cylindrical</i>	MB	–	6.43	1	HCl	[125]
Chemically treated <i>Lawsonia inermis</i> seeds	BG	–	39.00	5	1 M (CH ₂ COOH) ₂	[126]
Citrus limetta peel	MB	–	32.46	1	0.1 M HCl	[127]

**Fig. 5** Adsorption/desorption cycles of RhB and malachite green from modified coal fly ash with permission [129]

an aqueous solution were investigated by Tissera et al. (2020) [130]. The dye removal decreased from 100 to 94% in the fourth cycle using 0.5% NaOH ($pH = 2$) as eluent, by keeping all the parameters constant (concentration of *RhB* = 80 mg/L, loaded dye-adsorbent dosage = 0.8 g/L, contact time = 30 min), while the desorption capacity remains constant during three cycles. A similar result was reported by Thakur and Kaur [131] using paper industry sludge waste as a LCA and HCl as an eluent. The regeneration of adsorbents produced from the industrial by-products and wastes and reported in the literature is summarized in Table 3.

2.3 Natural materials

Natural materials such as clays which are hydrous aluminosilicate minerals are used in the treatment of textile effluents, due to their abundance in large quantities in most countries in the world [143, 144]. Other natural materials such as Zeolites which are aluminosilicates are also used for the treatment of inorganic and organic pollutants [145]. Finally,

siliceous materials are the most LCAs used for eliminating anionic dyes from aqueous solutions [146].

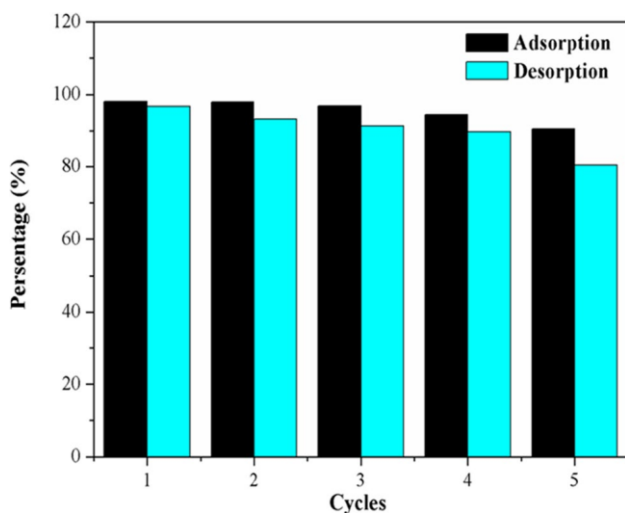
Bentahar et al. [147] tested the ability and reusability of Moroccan natural clay for removal of MB from aqueous solution in five cycles of adsorption–desorption using 0.3 M HNO₃ (Fig. 6). It found that adsorption efficiency was 90.57% after five cycles at an initial concentration of 100 mg/L of MB and 1 g/L of dye-adsorbent dosage. Similar results were reported by Ma et al. [148] using hydrolyzed polyacrylamide-modified diatomite and by Momina et al. [40] using bentonite coating (*Paintosorp*TM). Table 4 presents the regeneration of different adsorbents used to remove dyes from wastewater, as prepared from natural materials.

2.4 Sea materials and biomasses

Sea materials are economically attractive, abundant, and environmentally friendly. Chitosan and chitin which are natural polysaccharides extracted from crustaceans are the main sea materials employed for eliminating dyes from

Table 3 Regeneration of adsorbents produced from industrial by-products and solid wastes, for dye removal

Adsorbent	Dye	Ads. (%)	Des. (%)	N ^o of cycles	Desorbing agent	Ref
Hydroxyapatite-93	CR	64.23	–	6	0.1 M HCl	[132]
Modified coal fly ash	MG	–	88.31	3	NaOH (pH=8)	[129]
Modified coal fly ash	Rh6G	–	87.61	3	NaOH (pH=8)	[129]
Paper industry waste sludge	RhB	–	99.00	1	HCl	[131]
Cellulose	MB	98.20	–	4	0.1 M HCl	[133]
Dried waste sludge	MB	–	25.8	1	0.1 M HCl	[134]
Dried waste sludge	CV	–	30.8	1	0.1 M HCl	[134]
Keratin protein (0.5 M wool)	RhB	94.00	97.00	4	0.5% NaOH (pH=2)	[130]
Dried waste sludge	BF	–	69.3	1	0.1 M HNO ₃	[134]
Red mud modified with sodium dodecyl sulphate	SO	–	93.20	1	0.1 M HCl	[135]
Bottom ash	AO7	55.00	–	5	NaOH (pH=12)	[136]
Oiled soya	AO7	65.00	–	5	NaOH (pH=12)	[136]
Bottom ash	EY	54.00	–	4	NaOH	[137]
Lignite-based graphite	MO	84.00	66.00	5	0.5 M NaCl	[138]
Sludge-derived biochar	MB	60.00	–	3	C ₂ H ₅ OH/CH ₃ COOH	[139]
Aluminum lamination waste	DB	79.41	–	5	Distilled water	[140]
Aluminum lamination waste	DR	69.68	–	5	Distilled water	[140]
Aluminum lamination waste	DG	79.56	–	5	Distilled water	[140]
Industrial waste-transformed hydroxyapatite	CR	64.20	–	6	0.1 M HCl	[132]
Waste of rapeseed press cake	MG	–	94.5	1	0.1 M NaOH	[141]
Casuarina seed-biochar	MB	79.90	–	5	0.1 M HCl	[142]

**Fig. 6** Adsorption/desorption cycles of MB from natural clay [147]

wastewater [51, 166, 167]. The biomasses (dead and living) are used as efficient adsorbents in the treatment of wastewaters [168, 169]. The studies reported in the literature showed that sea materials and biomasses are efficient adsorbents for the removal of dyes from environmental wastewater [43, 170, 171]. Table 5 illustrates the efficiencies conducted in studies for the regeneration of adsorbents as prepared from different natural materials and biomasses.

Kooh et al. [172] recovered the methyl violet 2B (MV2B) from loaded dye-*Azolla pinata* in five cycles of adsorption–desorption using three desorbing agents: 0.1 M NaOH, 0.1 M MNO₃, and distilled water (Fig. 7). The MV2B adsorption capacity in the last cycle was 92.00%, 51.20%, and 47.10% using 0.1 M NaOH, 0.1 M NNO₃, and distilled water, respectively. A similar observation was reported by Kua et al. [171] using *Ipomoea aquatic*.

The regeneration and recyclability of aerobic granular sludge for the crystal violet (CV) in four adsorption–desorption cycles using acid TiO₂ hydrosol were investigated by Huang et al. [173]. The removal of CV was 70.00% in the fourth cycle at optimal conditions. Similar results were obtained by Elwakeel et al. [174] using sepia shells.

3 Comparison of regeneration of LCAs

The used adsorbents are generally of no use and could be disposed of as waste. But at the same time, its disposal can cause environmental pollution as most of them could be toxic due to adsorbed hazardous compounds/molecules [190, 191]. Apart from their disposal, the desorption of adsorbed pollutants and regeneration of the employed LCAs are other ways to use the material in an environment-friendly way [37]. A successful regeneration stage needs a relevant selection of eluents for the desorption of the loaded LCAs which

Table 4 Regeneration of adsorbents produced from natural materials, for dye removal

Adsorbent	Dye	Ads. (%)	Des. (%)	N ^o of cycles	Desorbing agent	Ref
Bentonite	MB	63.00	52.00	7	HCl	[149]
Moroccan natural phosphate	CR	–	48.30	1	0.1 M HCl	[150]
Moroccan natural phosphate	Rh6G	–	34.90	1	0.1 M HCl	[150]
Modified halloysite	MG	61.00	–	6	CH ₃ OH	[151]
Fe-zeolitic tuff	RE5	96.30	–	3	Fenton's reagent	[145]
Fe-zeolitic tuff	Y6	80.50	–	2	Fenton's reagent	[145]
bentonite coating (<i>Paintosorp</i> TM)	MB	92.60	–	3	C ₂ H ₅ OH	[40]
Kaolinite	RhB	86.00	–	5	Fenton's reagent	[152]
Calcareous soil	MB	–	94.00	1	H ₂ O/C ₂ H ₅ OH	[153]
Natural clay	MB	90.57	–	5	0.3 M HNO ₃	[147]
Kaolin/CuFe ₂ O ₄	MB	93.00	–	7	0.1 M HCl	[154]
Kaolin/CuFe ₂ O ₄	MV	92.00	–	7	0.1 M HCl	[154]
Surfactant-laterite soil	CV	82.00	–	4	0.1 M HCl	[155]
Surfactant-laterite soil	CV	85.00	–	4	1 M NaOH	[155]
Vermiculite	PB	–	41.00	1	CH ₃ OH	[156]
Modified montmorillonite clay	MB	–	99.95	1	1 M HCl/alcohol	[157]
Hydrolyzed polyacrylamide modified diatomite	MB	80.50	–	4	0.1 M HCl	[148]
Hydrolyzed polyacrylamide modified diatomite	RY	78.40	–	4	0.1 M HCl	[148]
Surfactant modified-zeolite	RY176	–	90.60	1	NaOH (<i>pH</i> = 12)	[158]
heat-treated and acid-leached halloysites	MG	61.00	–	6	CH ₃ OH	[151]
Romanian soil	CR	–	93.89	1	70% C ₂ H ₅ OH	[159]
Raw Algerian kaolin	MB	23.00	–	4	Distilled water	[160]
Carbon coated monolith	MO	14.00	–	6	1 M NaOH	[161]
Sepiolite	BR	–	9.40	1	HCl (<i>pH</i> = 2)	[162]
Spent sepiolite	BR9	–	36.00	1	Benzyl alcohol	[163]
Aquatic phosphate	DY28	–	13.6	1	Distilled water (<i>pH</i> = 12.6)	[164]
Calcined alunite	AR88	70.00	–	4	0.025 M NaOH	[165]

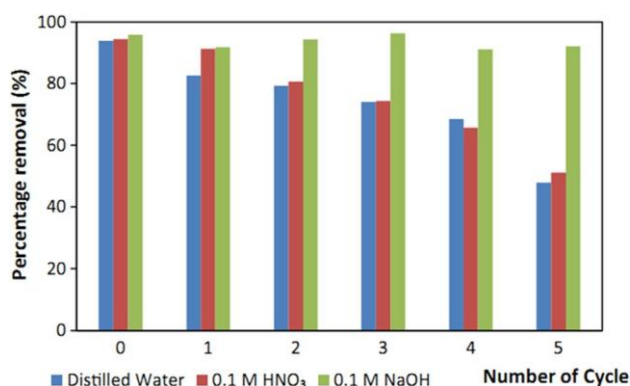
in turn depends upon the mechanism and type of dye remediation [34, 192]. This review reported the regeneration and recyclability of more than 150 non-conventional LCAs to remove dyes from environmental wastewaters in multiple consecutive adsorption–desorption cycles (Fig. 8). According to this study, we note that the agricultural solid wastes (39.56%) are the most regenerated and reused to remove dyes from an aqueous solution than sea materials and biomasses (22.3%), industrial by-products and wastes (18.71%), and natural materials (19.42%), due to their abundance in nature with large quantities. Also, these agricultural solid wastes are excellent adsorbents to remove dyes from wastewaters compared with other non-conventional LCAs, which may be due to their physicochemical properties sorption and the variety of functional groups sorption on their surface adsorbents. Finally, we notice that the bases and acids were the best eluents for anionic dyes and cationic dyes, respectively, for all non-conventional LCAs.

The disposal of LCAs is a major challenge, and it may cause an environmental threat if not managed properly

[193]. Some of the commonly used approaches for the disposal of spent adsorbents are incineration, landfilling, and stabilization/solidification [194]. In general, disposal in an open environment is not suggested due to the associated environmental risks [195]. Various factors should be considered in selecting the appropriate disposal method such as the amount of dye adsorbed, pH, and leaching tendency of dyes from the spent adsorbent [196, 197]. The toxicity characteristic leaching procedure may be employed to determine the extent of leaching [198, 199]. The cost and safety issues should also be considered while selecting a disposal method. For evaluating the real environmental impact of LCAs, risk assessments, environmental impact assessments, and life-cycle analyses should be performed [34]. In addition, the potential for recovery of dyes from LCAs and employing the adsorbents in other applications is a fascinating and suitable approach that needs to be explored. Conversion of LCAs into value-added products will be a good step towards a circular economy and sustainability [200].

Table 5 Regeneration of adsorbents produced from sea materials and biomasses, for dye removal

Adsorbent	Dye	Ads.(%)	Des. (%)	N° cycles	Desorbing agent	Ref
Brown macroalga	MB	48.16	46.83	5	1 M HCl/butanol	[43]
H ₂ O ₂ treated-chitosan	AR73	–	82.30	5	0.05 M NaHCO ₃	[175]
Alga <i>Nostoc comminutum</i>	ReB5	–	84.00	10	Deionized water	[176]
Sepia shells	CV	87.22	96.91	4	0.5 M HCl	[174]
Ultrasonic surface-modified chitin	MB	–	60.00	1	0.2 M HNO ₃	[177]
Sepia shells	CR	89.55	97.67	4	0.5 M NaOH	[174]
H ₂ O ₂ -treated <i>Trapa bispinosa</i> 's fruit	RO122	–	20.66	3	0.01 M NaOH	[178]
H ₂ O ₂ -treated <i>Trapa bispinosa</i> 's fruit beads	RO122	–	18.36	3	0.01 M NaOH	[178]
Sepia shells	MB	86.55	94.07	4	0.5 M HCl	[170]
Sepia shells	RB5	88.85	95.89	4	0.5 M NaOH	[170]
Polyethylenimine-chitin	AB25	–	95.70	2	0.01 M NaOH	[179]
<i>Trametes pubescens</i>	CR	–	92.67	3	0.1 M NaOH	[180]
Fungal biomass	RY86	58.00	–	10	50 mmol Na ₂ O ₃	[181]
Composite fungal biomass	RY86	63.00	–	10	50 mmol Na ₂ O ₃	[181]
Citric acid-modified <i>Chlorella vulgaris</i>	RY145	80.00	75.00	5	0.1 M NaOH	[182]
Aerobic granular sludge	MO	50.00	–	4	Acid TiO ₂ hydrosol	[173]
Aerobic granular sludge	CV	70.00	–	4	Acid TiO ₂ hydrosol	[173]
<i>Ipomoea aquatic</i>	MV2B	56.81	–	6	0.1 M HCl	[171]
<i>Ipomoea aquatic</i>	MV2B	63.68	–	6	0.1 M NaOH	[171]
<i>Ipomoea aquatic</i>	MV2B	9.00	–	6	Distilled water	[171]
<i>Aspergillus oryzae</i> cells	RhB	74.00	–	2	0.1 M NaOH	[183]
<i>Aspergillus fumigatus</i>	AV49	82.82	–	5	0.1 M NaOH	[184]
<i>Trametes pubescens</i>	CR	92.67	–	3	0.1 M NaOH	[180]
<i>Azolla pinnata</i>	MV2B	92.00	–	5	0.1 M NaOH	[172]
<i>Azolla pinnata</i>	MV2B	51.20	–	5	0.1 M NNO ₃	[172]
<i>Azolla pinnata</i>	MV2B	47.10	–	5	Distilled water	[172]
Chitosan	IC	93.75	–	3	0.1 M NaOH	[185]
burned root of <i>Eichhornia crassipes</i>	CR	–	87.00	1	5 M NaOH	[186]
<i>Rhizopus stolonifer</i>	BB	–	90.00	5	0.1 M NaOH	[187]
Peat	RhB	22.00	–	5	0.1 M HNO ₃	[188]
<i>Bacillus subtilis</i> KK01	CR	–	86.24	1	5 M CH ₃ COOH	[189]
<i>Oedogonium subplagiostomum</i> AP1	MO	–	83.00	1	0.1 M NaOH	[169]

**Fig. 7** Reuse of *Azolla pinnata* in five cycles for MV2B removal with permission [172]

4 Life cycle of the LCAs and disposal mechanism

Eco-design is defined as the integration of environmental aspects into product development, with the aim of reducing the environmental impact of products throughout their life cycle [201]. Most life cycles of LCAs have focused on their application as agricultural soil modifiers and as co-products of energy generation. Only a few studies have evaluated the comparative life cycle impacts of using alternative adsorbents for wastewater treatment [202, 203].

Dye contaminants/pollutants could be ionic and non-ionic in nature and therefore involves complex adsorption mechanisms, depending upon the type of remediating dyes [204]. Numerous mechanisms are responsible for the removal of various dyes from aqueous solutions using LCAs, mainly

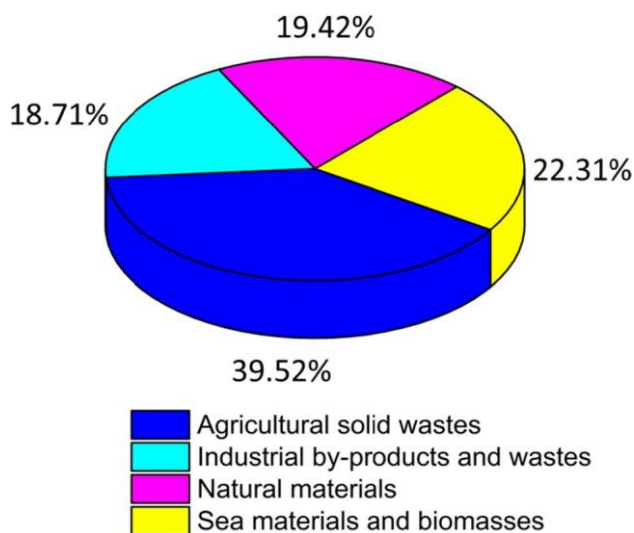


Fig. 8 Percentage of different LCAs regenerated and reused for eliminating dyes from wastewaters

guided by electrostatic, hydrophobic, and π - π interactions, van der Waals forces, H-bonding, acid–base reactions, and ion exchange [205–207].

5 Conclusions

The regeneration and the reuse of low-cost materials have been examined, which give useful insights into the possibility of adsorbent recovery and its operational behavior. Furthermore, in the present work, we have shown a general loss in the removal rate, due, on the one hand, to the irreversible binding of the dye to the adsorbent active sites, occurring during the regeneration phase and on the other hand, to the adsorbent protonated functional groups. In this context, the acids, the bases, the salts, the organic solvents, the alcohols, and the chelating reagents were used as desorbing agents for dye removal. Thus, it was concluded that an increase in the adsorbent regeneration cycles decreases both the adsorption and the desorption efficiencies. In addition, the increase in the desorbing eluent concentration was found to enhance the desorption efficiency. However, high eluent concentrations may affect the adsorbent structure and decrease the desorption efficiency. Furthermore, the adsorbent regeneration was easy for all non-conventional LCAs in multiple consecutive dye adsorption–desorption cycles. Basic and acidic aqueous solutions made with, respectively, NaOH and HCl were the most eluents used to recover dyes from low-cost materials. Finally, the results obtained in the present review paper reveal that the regenerated low-cost materials could be reused, and they have potential applications, such as recyclable adsorbents for wastewater treatment.

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