

REVIEW PAPER

Removal of Reactive Black 5 from Waste Waters by Adsorption: A Comprehensive Review

Mohammad Reza Jalali Sarvestani¹ and Zohreh Doroudi^{2*}

¹Young Researchers and Elite Club, Yadegar-e-Imam Khomeini (RAH) Shahr-e-Rey Branch, Islamic Azad University, Tehran, Iran

²Department of Chemistry, Yadegar-e-Imam Khomeini (RAH) Shahr-e-Rey Branch, Islamic Azad University, Tehran, Iran

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ABSTRACT

Reactive Black 5 is a toxic dye that has adverse effects on the environmental ecosystems and the health of human beings. Therefore, its removal is very important. Among the reported methods adsorption gathered huge attention in recent years because of its simplicity and low-cost. In this review paper, the removal of reactive black 5 by adsorption method from wastewaters was evaluated and all of the achievements from the past to the present were discussed in detail. The influence of important operational parameters on the adsorption efficiency of reactive black 5 such as pH, temperature, adsorbent dosage, and initial dye concentration was investigated. In addition, the reported adsorbents for reactive black 5 were divided into different groups based on their nature (like nanostructures, natural materials, by-products, and chitosan-based adsorbents) and their important characteristics, including adsorption capacity, removal percentage, initial dye concentration, repeatability, the synthesis cost, and optimized experimental parameters are compared with each other in detail. Moreover, important conclusions have been made from the surveyed literature and some suggestions are proposed for future works.

Keywords: Removal of dyes, Adsorption, Remazol Black B, Wastewater treatment, Azo dyes

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INTRODUCTION

Reactive Black 5 (RB5) is an economical azo dye widely utilized in different industries including the production of papers, textiles, colors, leathers, carpets, cosmetics, plastics, ink, shoes polish, electroplating, and mineral processing [1-5]. As a consequence, wastewaters of these industries are highly polluted with this teratogenic, mutagenic, and carcinogenic dye and the discharge of these effluents is the main way of RB5 release to the environment. Annually, more than 1000 tons of dyes are discharged by the effluents of various industries to natural waters and 45% of this amount belongs to the reactive dyes [6-12]. The chemical structure, IUPAC name, molecular formula, molecular weight, and maximum absorption wavelength (λ_{\max}) of RB5 are presented in Table 1.

* Corresponding Author Email: zohreh.doroudi@gmail.com

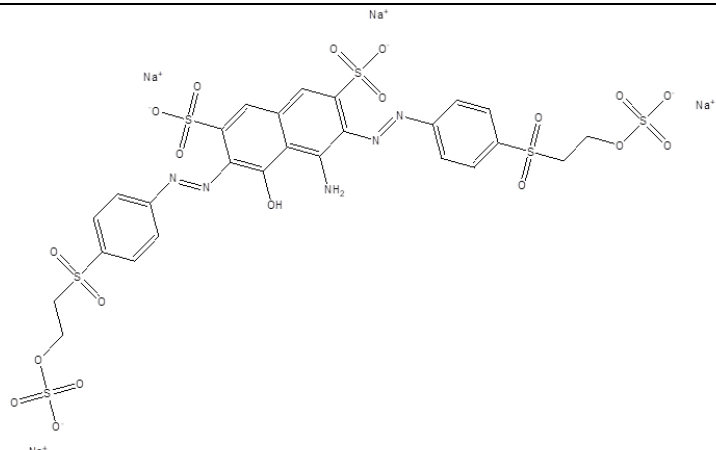

As can be seen, RB5 is a vinyl sulphone type reactive dye with azo chromophores in its structure. RB5 is also known as Remazol black B in the industry and its high popularity is due to a relatively low cost, low energy consumption, bright color, and high stability [13-18]. Reactive dyes are generally more stable than other kinds of dyes because they bind to desired materials through covalent bonds, which enhances the colorization efficiency prominently [19-21]. RB5 can obstruct light penetration in environmental waters even in trace amounts and consequently reduce the photosynthesis process in aqueous mediums leading to the impaired growth of aquatic plants, decreased oxygen concentrations, and finally the eutrophication phenomenon in the waters [22-25]. RB5 can not only affect the ecosystem but also cause serious problems for the health of human beings and other living organisms. Former reports



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Table 1. Chemical structure, chromophore group, IUPAC name, molecular weight, molecular formula and maximum absorption wavelength of RB5

Chemical Structure	Chromophore	Molecular Weight
		991.82 (g/mol)
IUPAC Name	Molecular Formula	λ_{max}
tetrasodium;4-amino-5-hydroxy-3,6-bis[[4-(2-sulfonatooxyethyl)sulfonyl]phenyl]diazonyl]naphthalene-2,7-disulfonate	$C_{26}H_{21}N_3Na_4O_{13}S_6$	597 nm

showed that continuous and long-time exposure to RB5 could cause skin rashes, bladder cancer, chromosomal aberration, respiratory and kidney failure, blindness, shock, cardiovascular collapse, and asthma. Also, the presence of trace amounts of RB5 gives the water an unsightly appearance from aesthetic views [26-30]. In this respect, the removal of RB5 is of great importance.

To date, different techniques including chemical oxidation, electrochemical destruction, photocatalytic degradation, ozonization, coagulation/flocculation, aerobic and anaerobic biological treatment, membrane separation, and ionizing radiation degradation have been reported for the removal and decolorization of RB5 from wastewaters [31-33]. However, these methods have serious disadvantages that limit significantly their large-scale industrial applications. For example, coagulation and flocculation methods do not have enough removal efficiency due to the very high solubility of RB5. Biological treatment techniques are not also applicable because of the non-biodegradable essence of RB5. The chemical oxidation, electrochemical destruction, photocatalytic degradation, ozonization, membrane separation, and ionizing radiation degradation are very time-consuming, expensive, intricate and in some cases, in their procedures, several by-products

that are potential environmental contaminants themselves are produced [34-37]. However, adsorption is an ideal alternative for the above-mentioned techniques because of its simplicity, low-cost, versatility in the chemical and physical features of adsorbents, high efficiency, insensitivity towards hazardous materials, rapidness, flexibility and easy operability [38-45]. The goal of this review paper is to gather all scattered data about RB5 and its removal from aqueous mediums by adsorption techniques and to evaluate all developments in this area of research from the past to the present. In this respect, adsorbents reported for the removal of RB5 were categorized in different groups based on their quiddity. Moreover, their main characteristics including repeatability, the cost of synthesis, adsorption capacity, and so on were compared with each other, and experimental parameters playing a key role in the removal efficiency were also discussed comprehensively.

Parameters Influencing the Adsorption Efficiency of RB5

In the adsorption technique, several factors including pH, temperature, adsorbent dosage, and initial dye concentration can play a key role in increasing the removal percentage of RB5. To develop novel adsorbents with relatively higher

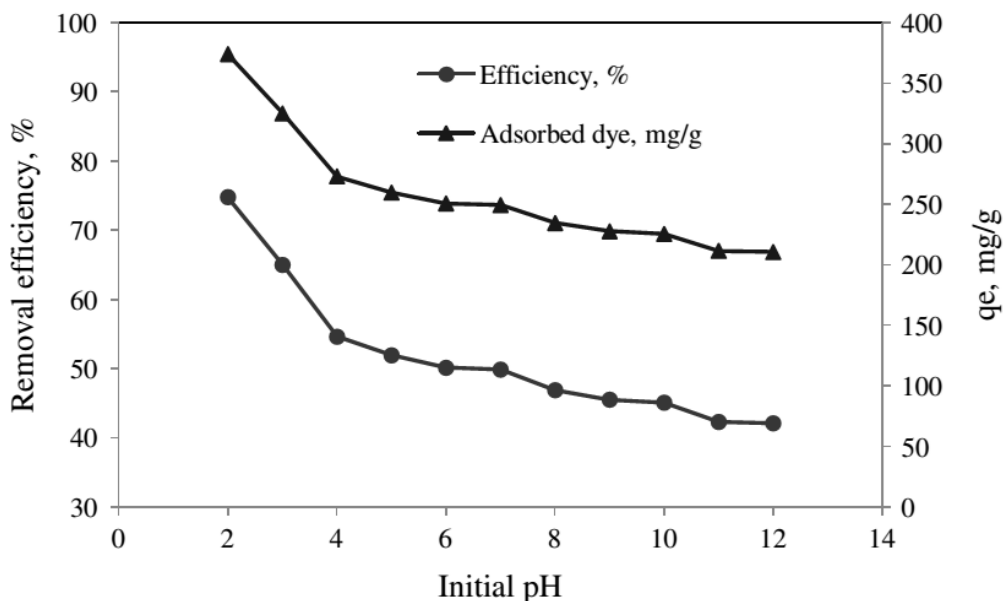


Fig. 1. Effect of initial pH on RB5 adsorption on to multiwalled carbon nanotube [24].

adsorption capacity for RB5 removal, it is helpful to look more precisely into the mechanisms by which experimental variables affected the removal efficiency of RB5. In this respect, the effects of important factors influencing the RB5 removal percentage will be discussed in the following.

The Influence of Solution pH

Owing to the fact that RB5 is an anionic dye and electrostatic interactions are one of the most effective factors in its adsorption process, it is critical to evaluate the effect of solution pH because it determines the surface charge of the utilized adsorbent. Numerous adsorbents have been reported for the removal of RB5, 70.46% of which showed the highest removal percentage in acidic solutions because the functional groups of the adsorbent surface usually become protonated in acidic conditions and, as a consequence, a positive charge covers the surface of adsorbent [11]. On the other hand, RB5 has anionic SO_4^- sites in its chemical structure. Hence, the electrostatic attractions are stronger in the acidic solutions and the adsorption efficiency will be higher at low pH values [15]. In the alkaline mediums, however, excess amounts of OH^- ions are present in the solution which can compete with RB5 to be adsorbed on the surface of the utilized adsorbent. In addition, the adsorbent surface does not have a positive charge anymore in alkaline solutions and consequently, electrostatic

forces become weaker [19]. As an example, the pH optimization diagram of RB5 removal by the multiwalled carbon nanotube (MWCNT) is presented in Fig. 1 [24]. As can be seen, the removal efficiency and the adsorption capacity decreased gradually by increasing the pH value from 2 to 12 by adding NaOH to the system. It should be noted that adsorption efficiency was still better in acidic mediums in most of the works where the removal of RB5 occurred in neutral pH values. Nonetheless, the authors decided to choose the natural pH of solutions (about 6-7) to make the work simpler and remove the step of pH adjustment of solutions in the process, with the consideration of no remarkable differences in removal percentages in neutral and acidic solutions.

The Influence of Temperature

Temperature plays a crucial role in the adsorption rate and dye equilibrium uptake. The effect of temperature on the adsorption efficiency depends on the thermodynamic properties of the system. In the case of endothermic systems, the removal percentage enhances significantly by increasing the temperature because the required energy or heat for the implementation of the process will be supplied from the environmental heat more conveniently at higher temperatures [22]. For exothermic systems, on the other hand, the adsorption process is more favorable at lower

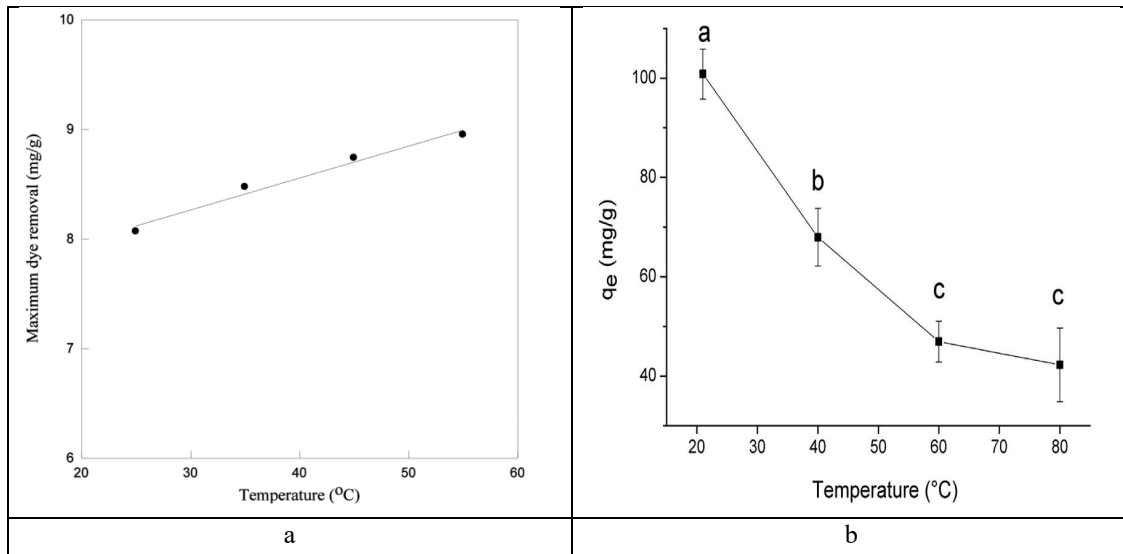


Fig. 2. The effect of temperature on the adsorption of RB5 by COP (a) [38] and HZN (b) [39].

temperatures because the heat resulting from the interaction of RB5 with the adsorbent is hardly conveyed to the environment at high temperatures [26]. As an example, the temperature optimization diagrams for RB5 removal by activated carbon made from orange peel (COP) and hollow zein nanoparticles (HZN) as endothermic and exothermic systems are presented in Figs. 2a and 2b respectively [38, 39]. As can be seen, the enhanced adsorption capacity of RB5 is observed on the surface of COP by increasing the temperature because the system is endothermic. In the case of HZN, however, the adsorption capacity declined remarkably by rising the temperature due to the exothermic nature of the system. It should be noted that adsorbents with high removal efficiencies at the room temperature and low impressment from temperature are more desirable because of the repeatability and simplicity of the work [30].

The Influence of Adsorbent Dosage

One of the factors that play a key role in the removal efficiency is the amount of utilized adsorbent. Generally, removal percentages enhanced tangibly by increasing the amount of adsorbent because more binding sites for the adsorption of RB5 are available at higher doses of adsorbent [31]. However, using large amounts of adsorbent is not preferable because of economic aspects. In this respect, developing adsorbents with higher binding sites, specific surface area,

porosity, and consequently better adsorption capacity is of great importance [41]. As an example, the optimization adsorbent dosage diagram for the removal of RB5 by chemically modified polyacrylamide/silica nanoporous composite (CMCA) is presented in Fig. 3 [21]. As it is clear, the removal percentage increased proportionally by increasing the amount of adsorbent in both 400 and 800 ppm solutions of RB5.

The Influence of Initial Dye Concentration

Reactive dyes are distinguishable in water at 1 ppm concentration by the naked eye while the RB5 concentrations in the effluents of industries are usually in the range of 10-100 ppm [11]. Therefore, a very important aspect is to evaluate the applicability of adsorbents utilized at different initial concentrations of RB5 under optimized conditions. Generally, the removal percentage was reported to decrease proportionally with increased amounts of adsorbed dye by increasing the initial dye concentration [15]. This phenomenon is usually observed because the adsorbent has finite binding sites on its surface for the RB5 adsorption and the binding sites will be occupied rapidly by the dye molecules with increasing the RB5 concentration. Hence there is no further place for interacting with other adsorbate molecules. Moreover, the selectivity of the adsorbent is also important because the matrix of industrial wastewaters is very complicated and is composed of various species. Thus, if the adsorbent

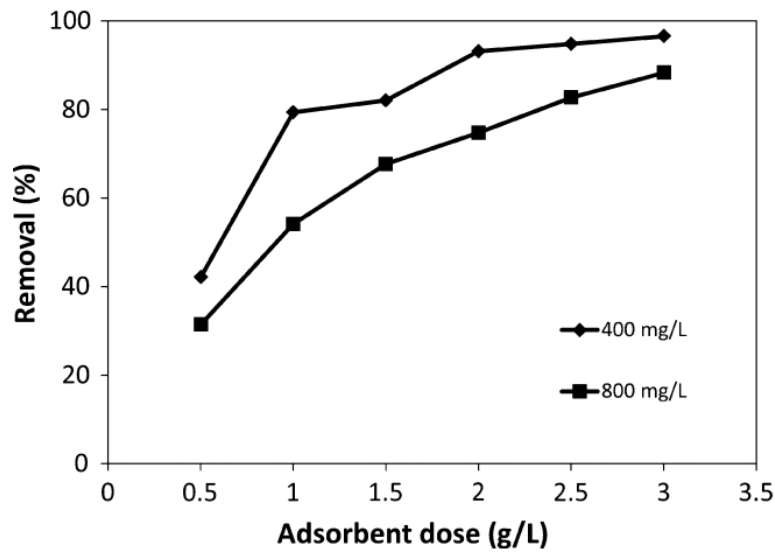


Fig. 3. The effect of adsorbent dosage on the removal of RB5 by CMCA [21].

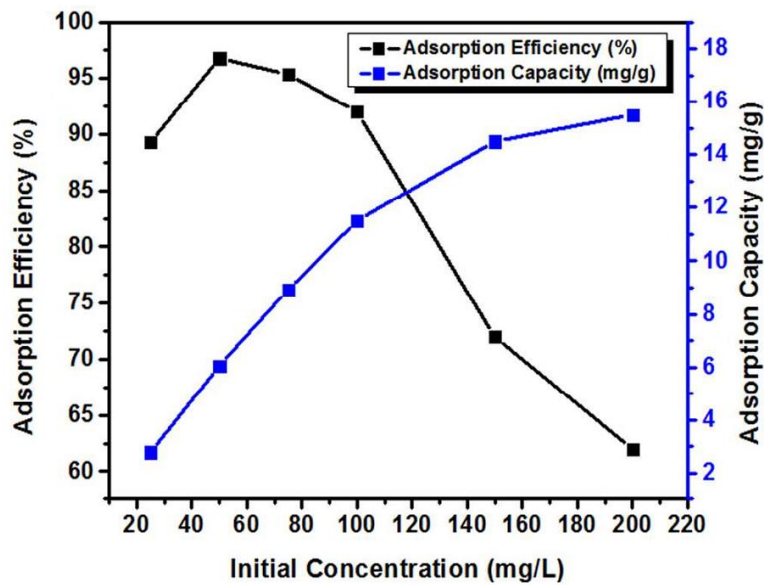


Fig. 4. The effect of initial concentration on the removal percentage and adsorption capacity of RB5 by electrospun zein nanofiber [22]

has no selectivity, other molecules can also occupy the binding sites and consequently, the adsorbent performance will be very poor in the effluents [19]. In this respect, adsorbents with higher adsorption capacity and selectivity have better performance in specimens with higher dye concentrations in the presence of other species [22]. As an example, the effect of initial dye concentration on the adsorption of RB5 by electrospun zein nanofibers is presented in Fig. 4. It indicates that increasing the initial

concentration of RB5 led to decreased removal percentage, but the adsorption capacity increased significantly [22].

Classification of Proposed Adsorbents for the Removal of RB5

To obtain further insights about the removal of RB5 by adsorption, reported adsorbents for RB5 were classified into three groups including natural and by-product adsorbents, nano-adsorbents, and

Table 2. The reported natural and by products adsorbents for RB5

Adsorbent name	Optimum pH	Optimum contact time (min)	optimum adsorbent dosage (g/L)	Optimum temperature (°C)	Removal %	Initial dye concentration (mg/L)	Maximum Adsorption Capacity (mg/g)	Ref.
Chitin	4	600	1.0	25	NM	24.8	92	1
Cotton stalk	1	60	1.0	25	54.9	25	35.7	2
Cotton hull	1	60	1.0	25	79.6	25	50.9	
Potato peel waste	3	120	20.0	25	85.5	50	NM	3
Rhizopus arrhizus	2	120	1.0	35	94.0	800	500.7	4
Green alga <i>Chlorella vulgaris</i>	2	120	1.0	35	92.0	800	555.6	5
Live activated sludge	2	120	1.0	25	82.3	500	134.8	
Dried activated sludge	2	120	1.0	25	78.0	500	126.5	
Autoclaved activated sludge	2	120	1.0	25	75.4	500	123.1	6
H ₂ SO ₄ -treated activated sludge	2	120	1.0	25	64.4	500	118.2	
NaOH-treated activated sludge	2	120	1.0	25	69.0	500	113.2	
Canola stalk	3	120	10.0	25	93.6	200	32.8	7
Brazilian pine-fruit shell in natural form	2.0	720	2.5	25	55.6	100	74.6	
Activated carbon prepared from Brazilian pine-fruit shell	2.0	240	2.5	25	94.3	100	446.2	8
High lime fly ash	7	60	15.0	20	87.3	100	7.184	9
Sugar beet pulp	1	180	1.0	25	88.5	20	80.0	10
Banana peel powder (bpp)	3	180	0.6	25	NM	300	49.2	11
Natural clinoptilolite modified with apolaccase	6	60	20.0	25	NM	0.001	1.79	12
<i>Aspergillus flavus</i>	4.5	5760	1.4	30	91.0	1000	233	13
<i>Corynebacterium glutamicum</i>	1	720	2.5	35	94.1	500	419	14
Treated residue from the aluminum industry	4	5	1.0	25	NM	10.0	0.98	15
Surfactant-modified zeolite	7	30	8.0	30	NM	150	3.84	16
Zeolite imidazole framework-8 (ZIF-8)	9.2	250	0.4	45	NM	50	133.76	17
hexadecyl trimethyl ammonium bromide modified clinoptilolite	NM	30	3.0	NM	NM	50	NM	18

chitosan-based adsorbents. Then, their initial dye concentrations, removal (%), maximum adsorption capacity and optimized operational conditions,

such as optimum levels of pH, temperature, adsorbent dosage and contact time were tabulated and the results were compared with each other.

Natural and By-product Adsorbents

The natural and by-product adsorbents developed for the removal of RB5 are presented in Table 2. As shown clearly in Table 2, 18 adsorbents are classified in this group encompassing 51.1% of the total reported adsorbents. Natural and product adsorbents are abundant, economical, and even have no economic value in many cases [1-3]. Moreover, these types of adsorbents usually need not need any processing and organic solvents or other toxic chemicals are not used in their preparation processes [7, 8]. Furthermore, huge amounts of household and industrial garbage have become an environmental problem all over the world in recent years. Because managing and disposing of massive volumes of wastes is not easy, one of the appropriate solutions for this problem is to reuse the wastages in the utile fields such as water treatment and adsorption of environmental contaminants [10-12]. The aforementioned reasons show why natural and by-product adsorbents are widely investigated for the removal of RB5.

However, these adsorbents are not very stable in harsh conditions such as highly acidic or alkaline solutions and their reproducibility is relatively poor in most cases [4-6]. In addition, the reusability of these adsorbents is not very good due to the poor stability of natural adsorbents in harsh conditions. Besides, many common eluent solutions cannot be used for the desorption of adsorbate and consequently, the desorption process will not be effective. Lack of selectivity is the next matter that dissuades the use of natural and by-product adsorbents for the removal of environmental pollutants [15]. An ideal adsorbent should have an equal removal percentage for the adsorbate in both laboratory-made solutions and real wastewater specimens; however, this phenomenon is rarely observed as the industrial effluents contain numerous coexisting species, which are also present in the medium at high concentrations [17]. However, the stability and selectivity of this type of adsorbent can be improved by performing minor modifications or pretreatment steps [18]. As it is obvious from Table 2, in this group of adsorbents *C. vulgaris* (a green alga) has the maximum adsorption capacity and its initial dye concentration is relatively high. It, therefore, seems that *C. vulgaris* is the best adsorbent in this classification.

Nano-adsorbents

In the recent decade, nanotechnology has

gained huge attention all over the world in the fields of water treatment and removal of environmental contaminants [19-21]. Because nanomaterials have special traits that make them a potential adsorbent, including high surface/area ratio, great surface reactivity, superb porosity, excellent thermal and structural stability, supreme specific surface area, high resistance to corrosion and ability to be modified by different agents to improve the selectivity and removal efficiency [25, 27]. Furthermore, the reusability of nano-adsorbents is usually better than that of natural ones because of their high structural stability even in harsh conditions [24].

Despite natural and by-product adsorbents the synthesis of nanostructures is costly and time-consuming and requires large amounts of different chemicals. Also, the separation of nano-adsorbents from water after the treatment process is difficult because of their very small size, and their uncontrolled release to the environment causes a lot of problems for the health of human beings and the environment [27-29]. For this reason, centrifugation and filtration techniques are commonly employed for the separation of nano-adsorbents from water, but these methods are expensive, tedious, and longsome [32]. Nevertheless, magnetizing the adsorbents can make the separation process more rapid and convenient because magnetic particles can easily be separated from an aqueous medium by applying an external magnetic field [26].

In this respect, the performance of 19 magnetic and non-magnetic nano-adsorbents was investigated for the removal of RB5 (42.2% of total reported adsorbents). The names of reported nano-adsorbents with their %removal, maximum adsorption capacity, initial dye concentration, and optimum experimental conditions are tabulated in Table 3. Accordingly, among the synthesized nano-adsorbents MWCNT has the highest adsorption capacity (1082.07 mg/g), with a relatively high initial dye concentration (200 mg/l) and fast adsorption kinetic. It seems that MWCNT is the most appropriate adsorbent for the removal of RB5 in this classification.

Chitosan-based adsorbents

Chitosan is a biocompatible, economic, eco-friendly, and biodegradable polymer that can be synthesized by deacetylation of chitin [34]. Chitin is the second most plentiful polymer in nature

Table 3. The developed nano-adsorbents for RB5

Adsorbent name	Optimum pH	Optimum contact time (min)	optimum adsorbent dosage (g/L)	Optimum temperature (°C)	Removal %	Initial dye concentration (mg/L)	Maximum Adsorption Capacity (mg/g)	Ref.
Chitin/graphene oxide composite	4	600	1.0	25	NM	NM	70	1
Polyaniline nanofibers	6	1440	1.0	25	NM	200	312.5	19
Poly 2-dimethylaminoethyl methacrylate-bentonite nanocomposites	11.3	500	0.1	40	NM	200	833.3	20
Polyacrylamide/silica nanoporous composite	2	60	3.0	25	97.0	200	454.5	21
Electrospun hollow Zein nanofiber	6	20	8.0	25	97.0	100	18.1	22
Graphite oxide/chitosan composite	2	1080	1.0	25	86.0	250	277	23
Graphite oxide	2	NM	1.0	25	54.0	250	205	
Multi-walled carbon nanotubes	2	60	1.0	25	98.0	200	1082.07	24
Magnetic Sodium Alginate Beads	7	240	180	25	92.3	150	NM	25
Magnetic iron oxide nanoparticles	2	10	6.25	45	90.0	50	18	26
Nanoscale zerovalent iron	5	120	5.0	25	100.0	100	NM	27
Mesoporous magnetic carbon composite	4	46.3	0.8	38.7	84.0	102	175.4	28
Magnetic graphene oxide synthesized (Fe ₃ O ₄ -GO) by impregnation method	3	1440	1.0	25	NM	500	164	29
Magnetic graphene oxide (Fe ₃ O ₄ -GO) synthesized by co-precipitation method	3	1440	1.0	25	NM	500	188	
Mesoporous Al ₂ O ₃ @TiO ₂ Nanocomposites	2-9	360	1.0	20	92.0	300	434.78	30
Magnetic graphene oxide (NiFe ₂ O ₄ -GO) nanocomposite	3	60	0.5	25	99.0	10	NM	31
Iron doped zeolite imidazole framework 8 (Fe-ZIF-8)	9.8	250	0.4	45	NM	40	193.56	17
Magnetic multiwalled carbon nanotube-Fe ₃ O ₄ nanocomposite	5.3	300	0.4	25	NM	30	0.3	32
multiwalled carbon nanotube-Fe ₃ O ₄ -κ-carrageenan-nanocomposite	6.4	300	0.4	25	NM	30	0.22	

after cellulose and can be extracted from prawns, insects, and crustacean shells. Due to the presence

of both hydroxyl and amine functional groups in the chemical structure of chitosan, it is an effective

Table 4. The reported chitosan based adsorbents for RB5

Adsorbent name	Optimum pH	Optimum contact time (min)	optimum adsorbent dosage (g/L)	Optimum temperature (°C)	Removal %	Initial dye concentration (mg/L)	Maximum Adsorption Capacity (mg/g)	Ref.
Pristine chitosan with %85 deacetylation degree	6.7	20	10.0	30	NM	6.7	NM	34
RB5 molecular imprinted chitosan crosslinked by epichlorohydrin	3	7200	1.0	30	90.0	0.006	2941	35
Chitosan beads	6	182.5	25.0	25	96.2	60	2.91	36
Pristine chitosan with %75 deacetylation degree	4	66	0.25	NM	85.0	30	NM	37
Chitosan granulates crosslinked by glutaraldehyde	2	1080	1.0	25	75.0	250	224	23

adsorbent for both anionic and cationic species [35]. However, the mechanical and structural stability of chitosan is really poor in harsh acidic conditions. To improving its stability, it is usually used in crosslinked forms or composites combined with other materials [36, 37]. The reusability and stability of crosslinked chitosans are better than the pristine ones. As can be seen from Table 4, five chitosan-based adsorbents have been reported for the removal of RB5 with different deacetylation degrees and various crosslinkers. Among this list RB5 molecular imprinted chitosan crosslinked by epichlorohydrin has the highest adsorption capacity. This adsorbent not only has the highest adsorption capacity in this group but also has the highest capacity among all the reported adsorbents (also considering the two previously discussed classifications). Although this adsorbent seems to be very effective and selective, it is not usable in industrial large scales because it has a very slow adsorption kinetic rate, and its optimum contact time is 7200 minutes (120 hours). As can be expected spending 120 hours for removal of dyes from the effluents of a factory does not seem logical.

Future perspectives

Although RB5 molecular imprinted chitosan has the highest adsorption capacity among the evaluated adsorbents, its adsorption kinetic rate is too slow and hence it is not applicable in the industrial scales. The second adsorbent was the MWCNT that showed a very high adsorption capacity with a relatively fast kinetic rate (1082.07

mg/g). Therefore, modification of this carbon-based nanostructure with various functional groups seems to be a reasonable way for developing a new adsorbent with higher adsorption capacity and also better selectivity, stability, and adsorption kinetic rate for removal of RB5.

CONCLUSIONS

In this review paper, scattered information about the removal of RB5 by the adsorption method from waste-waters in different papers was gathered and surveyed precisely. The reported adsorbents were divided into three groups of natural and by-product adsorbents, nanostructures, and chitosan-based adsorbents, and their characteristics were compared with each other. The natural and by-products adsorbents were economic and environmentally friendly but with poor stability in harsh conditions, reusability and selectivity. The nanostructures had better stability, selectivity, repeatability, and more efficiency but their synthesis was more costly and time-consuming than natural and by-products adsorbents. Chitosan-based adsorbents were biocompatible, low-priced, and effective but their adsorption rates were too slow. Generally, among the reported adsorbents the MWCNT seems to be the most appropriate candidate for removal of RB5 due to its very high maximum adsorption and relatively fast adsorption rate. Therefore, modification of this nanostructure with different functional groups or synthesizing novel composites using this material may be an appropriate way for designing new more effective adsorbents for RB5 removal.

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CONFLICT OF INTEREST

The authors do not have any conflicts of interest to disclose.

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