

REVIEW

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Removal of dye molecules from aqueous solution by carbon nanotubes and carbon nanotube functional groups: critical review

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For the removal and separation of wastewater pollutants, one widely used technique is adsorption. Different materials have been used as adsorbents. Among several adsorbents, carbon nanotubes (CNTs) are emerging as potential adsorbents because of their large surface area, defined cylindrical hollow structure, high ratio aspect, easy modification and hydrophobic wall surfaces. In this review, the capability of adsorption of dyes on CNTs and functionalized CNTs has been compiled. This paper provides data about defects, adsorption sites on CNTs and adsorption process studies under the effect of such various operational parameters as solution pH, contact time, and temperature, and deals with the mechanisms involved in the adsorption of dyes onto CNTs. According to observations from the articles reviewed, functionalized carbon nanotubes have better sorption capacity than as-grown CNTs. For the removal of dyes from aqueous solution, carbon nanotubes (CNTs) have shown good potential and some of the functional groups improve the adsorption potential according to evidence from the literature. For example, the maximum adsorption amount (q_{\max}) of methylene blue dye on an MWCNT surface has been reported to be 100 mg g^{-1} while the maximum adsorption amount of methylene blue dye on functionalized MWCNT-SH surface was 166.7 mg g^{-1} . This indicates that functionalization is beneficial in increasing the amounts of absorption. However, the development of highly efficient, cost effective environmentally friendly CNT-based nano adsorbents for their commercial applications should be the focus of still more study.

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1. Introduction

Pollution of ground and running waters by industrial dyes is a threat to aquatic life and human beings and is a main environmental problem.^{1–4} Synthetic dye stuffs are extensively used as coloring agents in the paper, textile, gasoline, food, and pharmaceutical industries. The hydrosphere contaminated by the discharge of dyes into the environment is a notable source of pollution due to its recalcitrant nature, visibility even at very low concentrations, giving an undesirable color to the water, biological attack, and reducing sunlight penetration.^{5–10} The aromatic molecular structure of dyes presumably comes from coal tar-based hydrocarbons such as naphthalene, benzene, toluene, and xylene.^{11,12} Today, the yearly production of commercial dyes is $>7 \times 10^5$ tons per year, and to date, more than 100 000 dyes are known.^{12–16} Therefore, a significant area of applied and basic research deals with the removal of dye pollution from industrial wastewater.^{17,18}

To remove dyes from wastewater, some methods such as ozonation, flocculation, membrane separation, aerobic or anaerobic treatment, coagulation and adsorption are used by industry.¹⁹ Nevertheless, adsorption is one method which has been most widely used because it is inexpensive, and easy and impressive for dye removal from industrial wastewater. Various adsorbents are used in adsorption processes such as activated carbon,^{20–25} graphene nanosheets, multi-walled carbon nanotubes, cedar sawdust, crushed brick, magnetic chitosan/grapheme, and oxide composites.^{26–35} To remove dyes from aqueous solutions, activated carbon is one of the best adsorbents.^{36–40} Because of their large surface area and high porosity, activated carbons have high adsorption capacity. But the low regeneration and high cost of activated carbon restrict its use.⁴¹ Different studies have shown the adsorption ability of dyes on a large variety of non-conventional low-cost adsorbents, such as agricultural and industrial waste.^{41–53} Nonetheless, in very few cases, for non-conventional adsorbents and low-cost adsorbents, is the adsorption capacity higher than for activated carbons and these adsorbents showed mixed results. So, to develop more efficient adsorbents with a higher capacity for regeneration, the study of nanotubes is an ongoing process. One carbon allotrope is carbon nanotubes (CNTs, 1D) consisting of sp^2 -hybridized carbons,⁵⁴ graphene (2D),^{53,54} as well as

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graphite (3D).⁵⁵ Since CNTs were discovered in 1991 by Iijima,⁵⁶ a variety of them have become available, such as single, double, few, and multi-walled carbon nanotubes.^{57–60} Carbon nanotubes (CNTs) have been intensively studied as a potential material to be used in a variety of applications based on their unusual chemical and physical properties.⁶¹ Compared with adsorbents such as clay and activated carbon, CNTs are more attractive because of their high selectivity, structural diversity and favorable physicochemical stability.⁶²

In this paper, the adsorption capacities of dyes on carbon nanotubes and functionalized carbon nanotubes are reviewed. The results of recent research are used to compare the usage of carbon nanotubes and functionalized carbon nanotubes as the basis of adsorbents for the removal of dyes. The effect of various parameters such as experimental conditions (*i.e.* solution pH, contact time, initial concentration, and temperature), dyes, and adsorbent characteristics on the adsorption capacity are discussed and presented. Also, in this paper thermodynamic, kinetic, and adsorption isotherm studies are reviewed.

2. Dye classification

There are many ways to classify dyes in terms of method of application, colour, and structure.⁶³ But the application classification is often favourable due to the extent and complexity of the nomenclature of colour from the system of chemical structure.⁶⁴ Table 1 presents the usual classes of dye classification according to the chemical structure of the dyes. Table 2 shows the classification according to the application of the dyes. Dyes are usually categorized according to the charge of the particle upon dissolution in aqueous solution⁶⁵ such as anionic (acid, reactive, and direct dyes), cationic (all basic dyes), and non-ionic (dispersed dyes).

Several industries such as the paper, dye stuff, plastic, printing, textile, carpet, cosmetics, and food, use dyes in their products to provide colour. These dyes are always left over in industry and the waste is usually discharged into the environment.^{66,67}

2.1. Dye toxicity

Dyes have great colour intensity and are easily visible even in very low concentrations.⁶⁸ Complex dyes are generally based on chromium, which is carcinogenic.⁶⁹ There are many aromatic structures in dye compounds that form stable complexes, which are resistant against heat, optical oxidizing agents, and biodegradation.⁷⁰ Because of the presence of toxic amines in the effluent, azo dyes are classified as toxic.^{71,72} Degradation-based dyes are highly sustainable and have higher longevity in effluents.⁷² In the environment, reactive dyes cause serious trouble because they are soluble in water and give a highly coloured effluent.⁷³ Also, having low biodegradability and being chemically stable, reactive dyes are likely to pass through conventional treatment plants, so their elimination is of high importance.⁷⁴ Dyes affect the photosynthetic activity of aquatic life as they decrease the penetration of light and are replete with metals, and aromatics, which may be toxic to some aquatic life.⁷⁵ Also,

dyes are carcinogenic, teratogenic, or mutagenic in different microbiological species of fish. Additionally, they can severely damage human beings through dysfunction of the reproductive system, kidneys, central nervous system, brain, and liver.⁷⁶

3. Dye adsorption process on CNTs

The active sites and defects of the CNT surface play an important role in dye adsorption on CNTs. CNTs consist of graphene or graphitic sheets, which have been rolled up into a cylindrical shape and exhibit a special sidewall curvature, and possess a π -conjugative structure with a highly hydrophobic surface. CNTs are called single-walled carbon nanotubes (SWCNTs) when they have one graphitic shell and multi-walled carbon nanotubes (MWCNTs) when they have several concentric graphitic shells. The main mechanisms of absorbing different dye compounds by CNTs depend on whether the dyes are cationic or anionic. In most cases, the adsorption prediction of dyes on carbon nanotubes may not be simple,⁷⁷ and different possible interactions between carbon nanotubes and dyes have been proposed. Hydrophobic, van der Waals forces, π - π stacking, hydrogen bonding, and electrostatic interactions might act simultaneously or individually.⁷⁸

3.1. Effective factors in dye adsorption on CNTs

Many factors are involved in dye adsorption by carbon nanotubes, such as initial concentration of dye, solution pH, temperature, and dosage of carbon nanotubes. To remove dyes in order to optimize industrial-scale development processes, an in-depth study of these parameters will be of great help. So, these factors are discussed here.^{79–81}

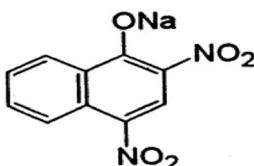
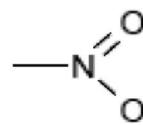
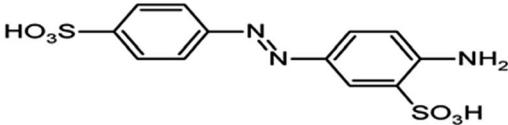
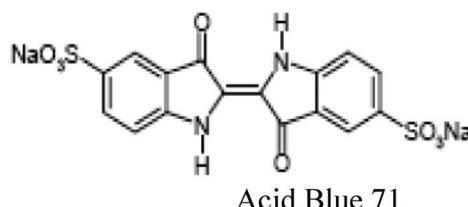
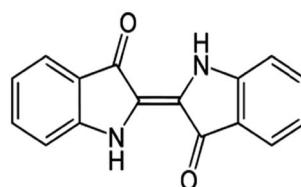
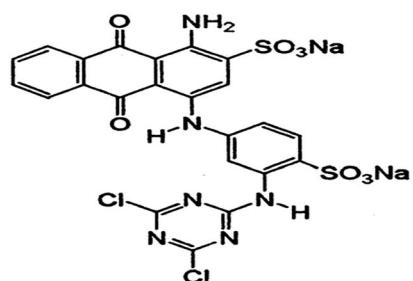
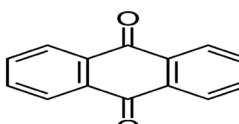
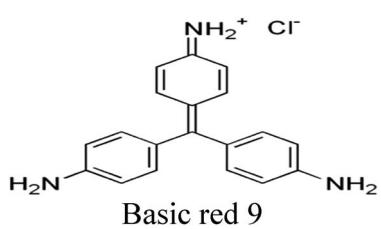
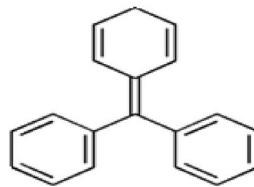
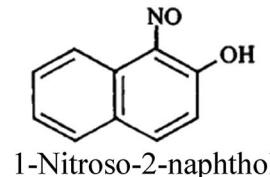
3.1.1. Initial concentration of dye. The initial concentration of dye has a great effect on the removal of the dye. The initial dye concentration effect relies on the immediate relationship between the sites available for binding on the surface of the adsorbent and the concentration of the dye. As the initial concentration of the dye increases, the efficiency of dye removal decreases according to the saturation of adsorption sites on the surface of the adsorbent.⁸² At a low concentration of dye, there will be unoccupied binding sites on the surface of the adsorbent and when the initial concentration of dye increases, there will be insufficient sites for the dye molecules to adsorb: therefore, the efficiency of dye removal decreases.⁸³

3.1.2. The effect of solution pH. Many articles have investigated the effect of solution acidity on the removal of the dye by CNTs from aqueous solution, and in several studies on dye adsorption it was reported that the acidity effect is one of the most important factors affecting dye removal through carbon nanotubes. Therefore, the solution pH is an effective parameter in dye removal. For example, Seow and Lim in a review article expressed that whereas for adsorption of cationic dye a high value of solution pH was preferred, a low value of solution pH is more suitable for the adsorption of anionic dye.⁸⁴

3.1.3. Temperature. One important factor affecting the dye adsorption on carbon nanotubes is temperature. Adsorption is



Table 1 Dye classification based on the chemical structure the dye comes from⁶³

Class	Example	Chromophores
Nitro dyes		
Azo dyes		$-\text{N}=\text{N}-$
Indigoid dyes		
Anthraquinone dyes		
Triarylmethane dyes		
Nitroso dyes		$-\text{N}=\text{O}$

a process which can be endothermic or exothermic for different adsorbents and dyes. If increasing temperature decreases the removal efficiency of the dye, dye adsorption on carbon nanotubes will be endothermic, and if increasing temperature decreases the removal efficiency of the dye, dye adsorption on carbon nanotubes will be exothermic.⁸⁵ For example, Kuo *et al.* examined some dye adsorption on carbon nanotubes and found that the removal of Direct Yellow 86 (DY86) dye by a carbon

nanotubes surface is endothermic,⁸⁶ as they observed that the adsorption capacity decreased slightly with an increase in temperature. Konicki *et al.* studied adsorption of Acid Red 88 (AR88) dye on a magnetic multi-walled carbon nanotubes–Fe₃C nanocomposite and found that removal of Acid Red 88 dye is exothermic.⁸⁷ Also, Yao *et al.* studied methylene blue adsorption on carbon nanotubes and found that removal of methylene blue dye was endothermic.⁸⁸



Table 2 Dye classification according to their chemical nature (adopted from⁶⁴)

Class	Method of application	Substrate	Chemical types
Direct	Used from slightly alkaline or neutral baths containing additional electrolyte.	Rayon, nylon, leather, paper, and cotton.	Azo, oxazine, phthalocyanine, stilbene.
Basic	Used from acidic dye baths.	Paper, inks, polyacrylonitrile, polyester, and treated nylon.	Azo, diazahemicyanine, cyanine, xanthene, acridine, triaryl methane, oxazine, azine diphenylmethane, and anthraquinone.
Acid	Commonly from neutral to acidic bath.	Silk, wool, inks, nylon, paper, and leather.	Xanthene, triphenylmethane, anthraquinone, nitro, and azo (including nitroso, premetallized).
Dispersed	Often applied by lower fine aqueous dispersions temperature or high pressure/temperature methods carrier; dye may be padded on thermo and fixed cloth.	Acrylic polyester, polyamide, plastics, and acetate.	Azo, benzodifuranone, nitro, styryl, and anthraquinone.
Reactive	Reactive site on dye reacts with functional group on fibre to bind dye covalently under influence of pH and heat.	Nylon, cotton, wool, and silk.	Azo, phthalocyanine, anthraquinone, basic, and formazan, oxazine.
Sulfur	Aromatic vatted substrate with reoxidized sodium sulfide and to form insoluble sulfur-containing products on fibre.	Cotton and rayon.	Structures indeterminate.

3.2. Isotherm of dye adsorption on CNTs

To study the dye adsorption isotherm on CNTs, it is necessary to measure the relationship between the equilibrium pressure, the amounts adsorbed, n_a , and P at constant temperature.⁸⁸

$$n_a = F(P)_T \quad (1)$$

3.2.1. Chemical and physical adsorption. The process of dye adsorption is normally considered to be a physical process in which van der Waals power in the adsorption process is usually dominant.^{89–94} The adsorption of physical liquid on solid surfaces occurs in an adsorption process during electron

Table 3 A comparison of the adsorption capacities of various adsorbents for some cationic dyes

Adsorbent	Dye	q (mg g ⁻¹)	Time	Temperature (K)	pH	Ref.
MWCNTs	Malachite green	142.85	80 (min)	298	7	98
CNT/polyaniline composites	Malachite green	13.95	10 (min)	293	7	99
MWCNT-TiO ₂	Malachite green	—	240 (min)	298	5	100
SWCNT	Malachite green	4.9285	15 (min)	300	7	101
SWCNT-NH ₂	Malachite green	6.1340	15 (min)	300	7	101
SWCNT-COOH	Malachite green	19.841	15 (min)	300	7	101
MWCNT	Malachite green	55.25	80 (min)	298	8	102
MWCNT	Malachite green	41.15	80 (min)	318	8	102
MWCNT-COOH	Malachite green	11.63	10 (min)	328	9	44
MWCNT-SH	Methylene blue	166.7	60 (min)	298	6	82
MWCNT	Methylene blue	100	60 (min)	298	6	82
Carbon nanotubes	Methylene blue	35.4	45 (min)	273	7	88
Carbon nanotubes	Methylene blue	46.2	45 (min)	298	7	88
Carbon nanotubes	Methylene blue	64.7	45 (min)	333	7	88
Carbon nanotubes	Methylene blue	103.62	120 (min)	290	7	93
Carbon nanotubes	Methylene blue	109.31	120 (min)	300	7	93
Carbon nanotubes	Methylene blue	119.70	120 (min)	310	7	93
Carbon nanotubes	Methylene blue	188.68	5 (hour)	298	6	13
Composite of graphene–CNT	Methylene blue	65.79	30 (min)	283	7	104
SWCNT	Basic red 46	38.35	100 (min)	298	9	105
SWCNT	Basic red 46	33.12	100 (min)	308	9	105
SWCNT-COOH	Basic red 46	49.45	100 (min)	298	9	105
SWCNT-COOH	Basic red 46	45.33	100 (min)	308	9	105
MWCNT	Maxilon blue	187.69	120 (min)	303	10	106



exchange between the molecules of dyes into the solution phase and the carbon nanotubes surface, and chemical bond formation is not necessary.^{93–95} Unlike physical adsorption, for the molecular forces during the process of adsorption, an electron exchange reaction means that a chemical adsorption occurs between the dye molecules of the solution and the solid surface of the carbon nanotubes.^{94,95}

3.2.2. Monolayers and multilayers. For open surfaces, adsorption consists of a layer-by-layer loading process, where the first layer is filled as $\theta = n_a/N_m = 1$, where θ is the recovery of the surface, and N_m is the capacity of the monolayer. According to the result, it is understood that when $\theta = n_a/N_m < 1$, we have monolayer adsorption, and when $\theta = n_a/N_m > 1$, the adsorption is multilayer.

To optimize the adsorption system design for the dye adsorption process, it is important to establish the most appropriate correlation for the curves of equilibrium. Various isotherm equations have been used to describe the dye adsorption equilibrium on the carbon nanotubes surface. Some of these isotherms are Langmuir and Freundlich. The Langmuir isotherm model is one of the most widely used. It is observed that the Langmuir isotherm model can be linearized in at least four different types. The Langmuir isotherm model can be expressed as:^{96,97,103}

$$\text{Type (I): } \frac{C_e}{q_e} = \frac{1}{KQ_m} + \frac{C_e}{Q_m} \quad (2)$$

$$\text{Type (II): } \frac{1}{q_e} = \frac{1}{Q_m} + \frac{1}{KQ_m C_e} \quad (3)$$

$$\text{Type (III): } q_e = Q_m - \frac{q_e}{KC_e} \quad (4)$$

$$\text{Type (IV): } \frac{q_e}{C_e} = KQ_m - Kq_e \quad (5)$$

where Q_m (mg g⁻¹) and K (L mg⁻¹) are Langmuir constants corresponding to dye removal capacity and adsorption energy.

3.2.3. Carbon nanotubes properties. The discovery of carbon nanotubes was very important. Because of their seamless graphite structure, and being nearly cylindrical, they have attracted much attention. Their thermal, mechanical, electrical, and physical properties make them ideal for work on SWNT and MWNT. Because of their high efficiency, the process of production of carbon nanotubes has progressed rapidly. In dye adsorption on carbon nanotubes, these properties have been taken into account because the surface area to volume ratio in CNTs is greater than in other common adsorbents. So, the chemical reaction with dye molecules for CNTs is more effective than for other common adsorbents which leads to the high contact surface of the effective collisions between the molecules of the dyes and the surface of the carbon nanotubes which in turn leads to an improvement in the efficiency of the adsorbent.^{124–128} Tables 3 and 4.

Table 4 A comparison of the adsorption capacities of various adsorbents for some anionic dyes

Adsorbent	Dye	q (mg g ⁻¹)	Time	Temperature (K)	pH	Ref.
MWCNT-COOH	Methyl orange	5.580	25 (min)	298	2	32
Functionalized CNTs with 3-aminopropyltriethoxysilane	Methyl orange	42.85	30 (min)	298	6.5	107
MWCNT	Methyl orange	51.74	2 (hour)	298	7	108
MWCNT	Methyl orange	52.43	2 (hour)	318	7	108
CNTs	Methyl orange	46	60 (min)	298	7	109
Alkali-activated MWCNT	Methyl orange	149	60 (min)	298	7	109
MWCNT	Methyl orange	66.09	120 (min)	297	6.5	110
MWCNT	Methyl orange	60.94	120 (min)	312	6.5	110
Composites of GO-MWCNT	Congo red	66.67	350 (min)	303	5	111
Chitosan hydrogel beads impregnated with CNT	Congo red	450.4	360 (min)	303	5	2
CNT/Mg(Al)O nanocomposites	Congo red	1250	75 (min)	298	7	112
f-MWNTs	Congo red	148	60 (min)	298	6.5	1
MWNTs	Congo red	148	—	—	7	113
MWCNT	Congo red	231.8	60 (min)	298	11	114
MWCNT	Reactive blue 4	502.5	4 (hour)	298	2	115
SWCNT	Reactive blue 4	567.7	4 (hour)	298	2	115
MWCNT	Reactive blue 4	442.0	4 (hour)	298	2	116
SWCNT	Reactive blue 4	487.6	4 (hour)	298	2	116
MWCNTs	Reactive black 5	1082.07	60 (min)	298	2	117
MWCNT	Reactive red M-2BE	312.3	1 (hour)	298	2	118
CNTs	Procion red MX-5B	42.92	24 (hour)	281	6.5	119
MWCNT-Fe ₃ C	Direct red 23	172.4	21 (min)	303	7	120
MWCNT	Direct blue 53	409.4	3 (hour)	298	2	121
MWCNTs	Acid red 18	166.67	120 (min)	298	5	122
MWCNT	Acid blue 161	1000.0	60 (min)	298	3	123



4. Conclusion

In recent years, many papers have been published on dye adsorption. In this review, the adsorption potential of carbon nanotubes and functionalized carbon nanotubes in the removal of dye molecules from wastewater has been highlighted. This paper presented dye classification according to their chemical nature and based on the dyes' chemical structure, and discussed dye toxicity. It also presented the effect of different chemical and physical conditions of adsorption on the adsorption capacity of dyes on carbon nanotubes and functionalized carbon nanotubes, such as initial concentration of dye, solution pH, and temperature. The pH of the solution is one of the main parameters in the adsorption of cationic dyes where a high value of solution pH is more suitable, whereas for anionic dye adsorption, a low value of solution pH is preferred. For the effect of initial dye concentration, because of increased interactions between dye molecules and adsorbent surface, the efficiency of dye removal will increase with an increase in the initial dye concentration, but with a further increase in initial dye, the concentration efficiency of dye removal will decrease due to saturation of the adsorption sites on the surface of the adsorbent. Also, for temperature, if the adsorption process is endothermic, as the temperature increases, the capacity for dye adsorption will increase. If the adsorption process is exothermic, with increasing temperature, the capacity for dye adsorption will decrease. Also, the toxicity of the dyes used in industry should not be neglected while they are removed through the adsorption process. So, it can be concluded that all these factors should be taken into account while the adsorption capacity of various adsorbents is evaluated from an economic point of view.

Conflicts of interest

There are no conflicts of interest.

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