REVIEW ARTICLE

Dye removal from water and wastewater by nanosized metal oxides - modified activated carbon: a review on recent researches

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Abstract
The conventional water and wastewater treatment methods are unable to provide up-to-data organized standards for drinking water and discharging effluents into natural ecosystems. Therefore, developing advanced and cost-effective methods to achieve published standards for water and wastewater and population needs are nowadays necessity. The important parts of this article are providing literature information about dyes and their effects on the environment and human health, adsorption properties and mechanism, adsorbent characteristics, and recent information on various aspects of modified activated carbons with nanosized metal oxides (AC- NMOs) in the removal of dyes. This review also summarized the effect of main environmental and operational parameters such as adsorbent dosage, pH, initial dye concentration, contact time, and temperature on the dye adsorption using AC-NMOs. Furthermore, the applied isotherm and kinetic models have been discussed.

Keywords Nanoparticles · Activated carbon · Adsorbent · Dye · Wastewater · Water

Introduction

Synthetic dyes are widely applied in various industries such as leather, textiles, pharmaceutical, printing, food, paper and pulp, packaging, etc. [1, 2]. These dyes mainly include aromatic rings, azo groups with the ability to produce toxic amines [1]. The results of previous research works have confirmed the low biodegradability and high stability of discharged dyes into natural ecosystems [3]. Pirkarami & Olya, (2017) reported that about 5000 tons of dyeing materials are discharged into the environment each year [4]. About 1000 tons/year of dyes are discharged into the aquatic systems from different effluents [5]. Dyes have special properties such as high water solubility, high stability to light that can disturb the transmission of sunlight into the water, mutagenic, carcinogenic, and toxic effects [6]. Therefore, dyes removal from water and wastewater is a great worldwide issue for protecting the environment and human health [7].

Researchers have used different methods such as ultrafiltration membrane [8], ion exchange [9], electrocoagulation [10], advanced oxidation process [11], photocatalytic degradation [12], adsorption [13], coagulation and flocculation [14], and phytoremediation [15] to treat dye solutions. Among them, the adsorption technique has received tremendous attention because of its cost-effectiveness, lack of susceptibility to toxins, easy access with high performance, and reusability [16, 17]. Over the past decades, activated carbon (AC) has been well known as the most useful technique for the removal of dye from aqueous solutions [18, 19]. But the application of activated carbon is sometimes limited due to the expense of raw materials related to the AC synthesis [20]. Therefore, because of the economic limitations of commercial activated carbon, the development of new economical adsorbents with high-performance is necessary [21]. Recently, different types of lower-cost adsorbents have been developed and applied for eliminating dyes from polluted water such as banana trunk [22], tea waste [23], rice straw [24], oil palm fruit fiber [25], mango peels [26], corn stalks [27], coconut shell [28], grape wood [29], Jerusalem artichoke stalk [30], orange

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peels [31], *N. microphyllum* (AC-NM) [32], pineapple peels [33], rice husk [34], walnut shell [35].

The removal efficiency of dyes depends on the physico-chemical characteristics of an adsorbent such as well-developed internal pore construction, high specific surface area, distribution of pore size, pore-volume, surface charge (hydrophobic and hydrophilic character of the surface) and the presence of surface functional groups [36, 37]. Therefore, researchers have applied different methods to enhance AC quality by focusing on the aforementioned factor.

Recently, nanoparticles have received special attention in the environmental field, because of high surface-to-volume ratio, high mechanical strength, high ordered structure, and lower size [38]. The loaded nanoparticles on the AC can improve the adsorption capacity and make the possible recovery of powdered adsorbents when using magnetic nanomaterials, especially iron oxide for developing a magnetized activated carbon (MAC) by simple economic separation [39, 40].

Magnetic separation as a reliable fast and very convenient method has been performed in previous research works by using different iron oxide loaded on the AC such as $CoFe_2O_4$ [41], Fe_2O_3 [39], $Au-Fe_3O_4$ [42], $NiFe_2O_4$ [43], Fe_3O_4 [44]. The magnetized activated carbon show some shortcomings such as creating a polarity on the activated carbon surface, and decreasing surface area because of pores trapping by magnetite, which can decrease the adsorption capacity [20, 45, 46]. The nanosized metal oxides (NMOs) such as zinc oxide (ZnO) [47], copper oxide (CuO) [48], tin oxide (SnO_2) [49], lead oxide (PbO) [50], titanium oxide (TiO_2) [1], zinc hydroxide ($Zn(OH)_2$) [51], magnesium oxide (MgO) [52], manganese dioxide (MnO_2) [53], cuprous iodide–cupric oxide [54], and $Ce-TiO_2$ [55] have been loaded on the AC. They can synergically improve the operation feature, recovery, and efficiency of adsorption processes [40, 51].

Juang et al. (2018) confirmed that magnetic Fe_3O_4/activated carbon nanocomposites have prior properties such as the easy recovery of the synthesized adsorbent for removing methyl orange (MO) from aqueous solutions with an adsorption capacity of 324 mg/g [20]. Joshi et al. (2019) studied the efficient elimination of methylene blue (MB) and brilliant green (BG) using Fe_3O_4 nanoparticles loaded on activated carbon. The results displayed that the maximum adsorption ability of the nanosized metal oxides - modified activated carbon was 138 and 166.6 mg/g for MB and BG dyes, respectively [2]. Alipanahpour Dil et al. (2019) studied the efficiency of $Ce-TiO_2$-NPs-AC for the elimination of Basic Red 46 (BR 46) from aqueous solutions. The system showed more than 99% removal of BR 46, at the constant value of other factors such as 25 mg/l initial concentration of dye, pH 5, and adsorbent dosage of 0.02 g, with 4 min sonication. The isotherm and kinetic studies of the process revealed that the adsorption method was successfully correlated with pseudo-second-order while equilibrium data well described by the Langmuir

isotherm model with a high adsorption capacity of 58.61 mg/ g [55]. Prajapati et al. (2020) reported the MB removal using developed copper oxide nanoparticles loaded on the nanoporous activated carbon (CuO-AC). The results showed that the developed NMOs-AC highly removed MB (more than 95%) under conditions (contact time 180 min, pH 9, a dosage of 3 g/l, and temperature of 318 K). Furthermore, the results depicted that among the isotherm models (Langmuir, Freundlich, and Sips), the Sips model was best fitted [56].

Morad et al. (2012) studied the MB removal efficiency using impregnated Fe_3O_4 onto activated maize cob powder. The effects of main variables such as pH (3–8), initial concentration (100–250 mg/l), adsorbent dosage (0.2–1.0 g), and contact time were reported and the results demonstrated that among the aforementioned parameters, the MB adsorption was dependent to pH and the greater efficiency of MB adsorption was attained at pH 6. Moreover, the data for kinetics was well described by the pseudo-second-order kinetic model [57]. Nekouei et al. (2015) investigated the removal efficiency of acid blue 129 (AB 129) using copper oxide nanoparticle loaded on activated carbon (CuO-NP-AC) as a novel adsorbent. The results showed that the maximum removal efficiency (around 100%) occurred at pH 2.0, the contact time of 20–25 min, a dosage of 0.045 g, and 10 mg of AB 129. Also, the results of isotherm models investigated by the Langmuir, Freundlich, Tempkin, and Dubinin–Radushkevich models showed that among them, the Langmuir model ($R^2 = 0.9985$) provides a greater association with the experimental data [48].

To the best of our knowledge, there is no comprehensive review of the removal of dye using modified activated carbon with nanosized metal oxides. Therefore, this study has been developed to review the effects of dyes on the environment and human health, the activated carbon preparation and adsorption mechanism, the modification of activated carbon using nanosized metal oxides, the effect of environmental and operational parameters on the removal of dyes, and finally describing the isotherm and kinetics studies.

Dyes classification and characteristics

Dyes as the main class of pollutants based on solubility and chemical properties can be classified into three common groups of anionic (acidic, reactive, and direct), cationic (basic dyes), and nonionic (dispersed) [58]. The molecules of dyes include two main components (Table 1); auxochromes (NO_2, NO, N=N; as a functional group of atoms attached to the chromophore and assist the dye solubility) and chromophores (NH_2, NR_2, OH, NHR, Cl and COOH; responsible for producing color) [59, 60].

Table 1 The chemical structure of synthetic dyes

Chemical class	Dye	Molecular structure	Chemical formula	Mw (g/mol)	References
Cationic	Basic Yellow 28 (BY28)		$C_{21}H_{27}N_3O_5S$	433.52	[64]
	Methylene blue (MB)		$C_{16}H_{18}N_3SCl$	319.85	[66]
	Malachite green (MG)		$C_{23}H_{26}N_2Cl$	364.92	[67]
Anionic	Direct Blue (DB)		$C_{40}H_{23}N_7Na_4O_{13}S_4$	1029.87	[43]
	Acid blue 129 (AB 129)		$C_{23}H_{19}N_2NaO_5S$	458.46	[48]
	Acid Yellow 17 (AYD 17)		$C_{16}H_{10}Cl_2N_4Na_2O_7S_2$	551.29	[73]
Nonionic	Disperse Blue 56 (DB 56)		$C_{15}H_{13}BrN_2O_4$	365.18	[77]
	Disperse Red 354 (DR 354)		$C_{22}H_{24}ClN_5O_7$	505.723	[79]
	Disperse Orange 25 (DO)		$C_{17}H_{17}N_5O_2$	323.35	[80]

The molecular structure of synthetic dyes generally includes a complex aromatic, which cause more stability, toxicity, and non-biodegradability of them (Table 1) [61]. The coal-tar based hydrocarbons including naphthalene, benzene, xylene, toluene, etc. are sources of the aromatic molecular structure of synthetic dyes [62].

Dyes based on carrying change can be categorized into cationic and anionic, in which cationic and anionic dyes have a positive and negative charge on their molecules, respectively [63]. Among cationic dyes, the removal of basic yellow [64], rhodamine [65], methylene blue [66], and malachite green [67], basic blue 26 [68], Alizarin red S [69], Brilliant green [70] form water and wastewater have been intensively studied using adsorption process. Anionic dyes consist of different subclasses with many components that are different in a structure such as azoic, triphenylmethane, anthraquinone, xanthene, nitroso, azine dyes, and nitro [71, 72]. Previous studies have applied nanosized metal oxides - modified activated carbon for eliminating different anionic dyes such as acid blue (AB 129) [48], acid yellow [73], direct Blue (DB) [43], Direct Blue 71 (DB71) [74]. Azo dyes as the largest group of synthetic dyes are characterized by the presence of nitrogen-nitrogen bond (-N=N-) with aromatic structures that may link to sulfonic acid groups [75]. Nonionic dyes such as disperse yellow [76], disperse blue [77], disperse orange [78], disperse red [79], and disperse orange 25 (DO) [80] are a heterogeneous collection of several chemical types.

Environmental and health problems

As mentioned in the introduction section dyes release into the environment from industrial and domestic activities. Industries such as textiles, rubber, plastics, pharmaceuticals, paper, leather, food, cosmetics are the most important sources of discharging dyes into the natural ecosystem [56]. The textile industries as main sources of discharging of untreated effluents are responsible for damaging of environment and human health because of their characteristics including high chemical oxygen demand (COD), non-biodegradable organic compounds, complex structure (aromatics), stability, and toxicity [81, 82].

Among organic compounds, dyes based on their components and properties have a different effect on the plants and animals [83]. Dyes can cause aesthetic damage to the water bodies, and they can prevent the penetration of light into water that causes a reduction in the rate of photosynthesis because of a decrease in the level of dissolved oxygen furthermore, it can alter the main characteristics of wastewater such as chemical oxygen demand (COD) and pH [84].

The aquatic toxicity of dyes includes changes in primary productivity, protozoan colonization rate, and species composition of phytoplankton. Furthermore, dyes have different effects on the macro-invertebrate community systems and also the behavior of macro-invertebrate [85]. Among the dyes used in industries, the azo dyes are a large group that is resistant and stabile against degradation due to the double bond of nitrogen in which their complete removal by conventional treatment systems is not possible [1, 86, 87]. Azo dyes have effects on flora and fauna in the environment due to their structural properties and it could result in carcinogenic, and mutagenic [88, 89]. The physicochemical properties of soil can be affected by dyes disposal, furthermore, agricultural productivity and growth of plants decrease because of the germicide effect of dyes on the soil microorganisms [90, 91]. Azo dyes can have acute and chronic effects due to their toxicity and mutagenic properties, depending on the type and concentration of dyes in the products [92].

Prolonged exposure to dyes during production, and consumption in industries, as well as polluted water cause health hazards and therefore, manage all dyes and chemicals, which are important to protect human health [46, 93]. Furthermore, dyes have carcinogenic, mutagenic, and teratogenic effects on humans [84]. In previous studies, different effects of cationic dyes such as methylene blue include increased heart rate, vomiting, gastritis, cyanosis, aesthetic, allergic problems tachycardia, irritation in the skin, gastrointestinal tract, mental disorders, and carcinogenic have been reported [2, 94, 95]. Furthermore, Malachite green (tri-phenyl methane dye) as a cationic dye have carcinogenic properties and it accelerates the generation of the liver tumor [49].

Dye removal

The treatment of colored effluents as an environmental and health problem has been of particular importance since their effects on natural ecosystems and human health have been discovered. Therefore, they have been upgrading with the development of standards as well as advancements in techniques and methods of analyzing [40, 96]. With the development of more effective methods, including biological treatment as an eco-friendly and cost-effective method, it was possible to remove many natural and synthetic dyes [97]. But, high concentrations of dyes, as well as complex compounds of dyes showed the inability of biological systems in their removal due to the harmful effects of dyes on some microorganisms [4]. To overcome this problem, researchers have proposed and implemented an advanced treatment that involves a variety of physical and chemical methods, which, of course, has led to heavy costs for the industries. Physicochemical methods such as coagulation [98], adsorption [99], and filtration [100] are used for wastewater containing high concentrations of dyes. Advanced oxidation processes involve ozonation, ultraviolet radiation, ozone/UV, photocatalysis UV/)TiO$_2$), Fenton oxidation is capable of dyes oxidation, but these methods require

a large number of chemical substances that make them uneconomical [101, 102]. The process of coagulation-flocculation is known as an economical, easy-to-use, simple and energy-saving alternative, but the coagulation process is not suitable for dye removal, moreover, a large amount of sludge is created, which may be turned into a pollutant and increase the cost of purification [103, 104]. Among the above-mentioned methods, the adsorption process due to superior advantages such as easy control and availability to raw materials, high efficiency, cost-effectiveness, simple recovery, design simplicity, ease of operation, and non-toxicity have been used widely to remove dyes from water and wastewater [105, 106].

Adsorption theory and mechanism

Adsorption is a technic for the separation of some of the components from the fluid phase on the surface of adsorbent [107]. The adsorbent has external and internal surfaces, which the internal surface due to the more effect on the adsorption process is more important than the external surface. At the first step of adsorption, pollutants are dispersed at the external surface of the adsorbent. In the second stage with sufficient time, an internal diffusion occurs that adsorbed molecules penetrate in the depth of adsorbent, that is, the transfer of contaminated mass to the interior surfaces. Then the process continues until a layer is made on the surface of the active pores, which is the end of the adsorption process. Adsorption is a reciprocating process that will continue until equilibrium [107–109]. One of the two main adsorption mechanisms, physical or chemical is used in the process of removing pollutants using adsorbents. In physical adsorption, target molecules bind to pores in the walls of high-level adsorbents by the forces of van der Waals, which have a low adsorption heat. In chemical adsorption, the target pollutant is placed under a chemical covalent reaction that attaches to certain sites in the adsorbent, and its heat is much more than adsorbing and is approximately equal to the heat of reaction [110].

Conventional activated carbon

Activated carbon is used as a highly effective ingredient in a variety of applications such as lithium batteries, electrocatalytic, and pollutants removal from aqueous solutions, biocatalysis, and electrochemical capacitors [111]. It has been estimated that the AC has a surface area of 1000 to 1500 m^2/g [112]. Generally, activated carbon has two types; granular activated carbon (GAC) and powdered activated carbon (PAC), which powdered AC is more effective than granular AC for reducing/removing contaminants [113, 114]. Activated carbon may be generated from any raw material with a high content of carbon, but commonly wood, coal,

and lignite have been used more among others. Furthermore, several studies have been undertaken to find alternatives materials for the production of activated carbon to reduce the generation cost such as agricultural and industrial wastes [29, 115]. Over the past decades, the interest in using lignocellulosic as a precursor of AC has been increased [115]. Moreover, researchers suggested two main methods for activation of ACs; physical and chemical methods. Physical activation can be developed in the existence of oxidizing gases, such as carbon dioxide or steam at a varied temperature (800–1000 °C). The chemical activation can be carried out by various chemicals such as KOH, NaOH, H_3PO_4, $ZnCl_2$, and other chemicals [116, 117]. Besides, it has mentioned that the structure of carbon compounds mainly includes functional groups, such as carboxyl, phenol, carbonyl, Quinone, and lactone, which are capable of contaminant adsorption. On the other hand nitrogen, sulfur, oxygen, and hydrogen are often found in the form of reactive groups or chemical atoms in the AC [118].

Various studies used the prepared activated carbon from agricultural wastes for the removal of dye (Table 2). According to data have been summarized in Table 2, pH as a main independent variable has been investigated in different ranges from 2 to 11. Some of the investigations depicted that high removal efficiency (more than 95%) of cationic dye can be achieved at neutral pH (6 to 7) [119–121], but others declared that lower pH (2–5) has a significant effect on the dye removal so that when pH increased more than acidic range, the removal efficiency of dye declined [122–125]. Furthermore, it should be noted that all studies have shown that at the initial concentration of dye, highly removal was observed on the other hand, by increasing dye concentration the removal efficiency of dye was decreased sharply [27, 29, 126]. Also, based on Table 2, the result of the studies by Balarak et al. (2015) [127], Kazemi et al. (2016) [128], Shahbazi et al. (2020) [29], and Daneshvar et al. (2014) [124] have verified that contact time had a positive effect on the dye removal so that the maximum removal efficiency obtained by increasing contact time to equilibrium time. In brief, according to Table 2, the applied AC as a promising and reliable adsorbent for the dyes elimination showed high efficiency of about 80–100% [122, 123].

Nanosized metal oxides - modified activated carbon

Currently, nanotechnology has been developed expressively and broadly has been applied individually and by integrating with other processes in the treatment of water and wastewater for the pollutant removal [54, 56]. Nanotechnology as a promising method can overcome some shortcomings of previous methods due to good characteristics such as metallic or semi-

Table 2 Comparison of dyes removal efficiency using various AC

Adsorbent	Adsorbate	Maximum capacity (mg/g)	Dosage (g/l)	pH	Time (min)	Initial con. (mg/l)	Removal (%)	References
Celery (Apium graveolens)	Methylene blue	476.19	1	6.5	120	100	99	[119]
Cerastoderma lamarcki shell	Acid black 1	–	7	2	60	50	100	[122]
Indian seed husk of Bengal gram	Congo red	41.66	6	5.85	180	50	92	[126]
Modified sphagnum peat moss	Malachite green	121.95	0.6	6.5	90	60	100	[120]
Garlic straw (GS)	Methylene blue	256.41	4	7	200	100	85	[121]
Cornstalk	Methylene blue	–	1.4	11	50	10	90	[27]
Cerastoderma lamarcki shell	Malachite green	35.84	1	5	60	20	–	[128]
Canola residues	Methylene blue	–	3.5	7	75	25	97.5	[127]
Ageratum conyzoides leaf powder (ACLP)	Methylene blue	192.4	0.06	3	20	20	>80	[123]
Roots of Eichhornia crassipes	Congo Red	1.580	1.0	–	90	104.45	96	[129]
Shrimp shell	Acid Blue 25	1093	0.1	2	–	70	–	[124]
Rice straw-based carbons	azocoumarin	156.34 at 60 °C	0.9	3	75	100	70	[125]
Sugarcane bagasse	Methyl red	–	0.4	6	180	50	100	[130]
Grape wood	Methylene blue	4.82	12.25	11	90	100	>98.5	[29]

metallic behavior [131], favorable thermal features [83], high mechanical strength [38], high surface area [38], interact with some organic molecules via non-covalent forces [83], high interparticle interactions with pollutant [56], a high number of vacant reactive surface sites [131], and by integration with them. Nowadays, preparing cost-effectiveness AC with high efficiency that does not have common carbon problems is a research priority [132, 133]. Therefore, separation of powdered carbon from the effluent, improvement of activated carbon quality, reusability, and the use of integrated methods are the most important up-to-date way that can minimize the weaknesses of common activated carbon. Among them, loading metallic nano-oxides on the AC make possible aforementioned improvements because of their short diffusion route and high surface area to volume ratio [39, 46, 69, 134]. In addition to the ability to form magnetite, metal oxides can improve the physicochemical properties of activated carbon. Furthermore, these compounds increase the ability to adsorb contaminants such as dyes, due to the high special surface area [40, 135]. The metal oxide nanoparticles such as iron oxide [136, 137], titanium oxide [138], zinc oxide [139], magnesium oxide [140], nickel oxide [141], manganese oxide [142], copper oxide [143] and zirconium oxide and its combination with iron are the most used nanoparticles [144, 145]. As mentioned, to receive the advantages of the AC and metal oxide nanoparticles and overcoming their weaknesses, integration of active carbon with nano metal oxides has been developed in various studies by loading zinc oxide (ZnO) [135], iron(III) oxide (Fe_2o_3) [39], lead oxide (PbO) [50], tin oxide (SnO_2) [49], and copper oxide (CuO) [48] on the activated carbon.

NMOs - AC characterization

The results of previous research works showed that the nanosized metal oxides loaded on activated carbon could significantly improve the physicochemical properties on nanomaterials and activated carbon and therefore their efficiency due to synergist effect on their ability to remove aqueous pollutants [55]. In some studies, NMOs- AC has been used to remove dyes, which their properties before and after dye removal have been investigated using various tests such as scanning electron microscope (SEM), energy dispersive X-ray (EDX), Fourier transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD), Brunauer–Emmett–Teller (BET), thermal gravimetric analysis (TGA), and transmission electron microscopy (TEM) [55, 69, 146]. The chemical and physical characteristics of different NMOs- AC has been presented in Table 3. It can be observed that the surface area of prepared adsorbents was ranged between 268 to 940 m²/g. Studies have confirmed that loading NMOs onto AC can improve the surface area and micropore volume of the adsorbent. Altıntıg et al. (2017) stated that the surface area of pristine AC was

Table 3 The NMOs-AC characteristics

NMOs- AC	BET (m²/g)	SEM/FESEM	FTIR	XRD	TEM	References
Magnetic loaded AC	940.13	The non-porous and random layer surface, which after loading iron oxide on the AC long and deep cracks occurred on the surface of the AC.	Stretching vibration of C=C bonds of the aromatic ring: 1610–1500 cm⁻¹, Alcohol groups (R–OH):1026 cm⁻¹, Stretching vibration of benzene ring: 1582, 1410, 1342 cm⁻¹	No-crystal structure	–	[147]
MnO₂ -AC	612.03	The sheet-like morphology of the MnO₂-NP-AC with a thickness of about 50–100 nm.	Mn–O vibrations modes: 586 cm⁻¹, O–Mn–O bond: 458 cm⁻¹	Amorphous structure	–	[131]
ZnO -AC	603	Randomly distributed NPs with an average diameter of 46 nm and there were holes and cave type openings on the surface of the adsorbent that accessible more surface area available for adsorption.	O–H stretching vibrations of water: 3434 cm⁻¹, Asymmetric and symmetric C–H bands of methyl and methylene groups: 2963 and 2853 cm⁻¹, Carbonyl C=O group: 1700 cm⁻¹, C=C band:1600 cm⁻¹, Metal–oxygen (M–O) stretching: 423 and 634 cm⁻¹	Hexagonal structure	–	[47]
MAC-α-Fe₂O₃	–	The presence of metal oxide (white aggregates) in the pores of tile-like structured AC was observed.	Peaks in the range 1100–1200, 1540–1570 and 2320–2370 cm⁻¹: carbon was not destroyed in composites during pyrolysis, $v_{Fe–O}$ of iron (II) gluconate: 585 cm⁻¹, $v_{Fe–O}$ of α-Fe₂O₃: 592 cm⁻¹	Crystallite structure of α-Fe₂O₃ in the carbon matrix	Iron oxide nanoparticles are embedded in the pores of a carbon matrix and also nanosize of embedded iron oxide nanoparticles is in the range 5–17 nm.	[73]
Fe₃O₄ -AC	–	Bare AC and Fe₃O₄/AC nanocomposites reveal the fiber-like shape.	N–H stretching vibration: 3437 cm⁻¹, C–H stretching vibrations: 2924 and 2854 cm⁻¹, N=N vibration: 1421 cm⁻¹, S=O vibration: 1122 cm⁻¹	AC: amorphous like structure Fe₃O₄-AC: pure Fe₃O₄ phase	Fe₃O₄-NPs are attached to AC with a size of 5–20 nm.	[20]
ZnO-AC	–	Agglomerated ZnO on the AC surface and ZnO-AC nanoparticles are distributed randomly over the AC surface. Furthermore, the hydrodynamic diameter of ZnO-AC ranged between 200 and 1000 nm.	ZnO stretching: 475 cm⁻¹, C–H bending vibrations: 950 cm⁻¹, C–OH stretching vibrations: 1210–1320 cm⁻¹, C=O stretching vibration: 1702 cm⁻¹, stretching vibrations of CH₂ bonds: 2332 and 2362 cm⁻¹, O-H stretching vibrations: 3300–3400 cm⁻¹	Hexagonal structure	–	[148]
Zn(OH)₂-AC	268	The surface morphology of the AC is homogeneous and relatively smooth and also the size of each Zn(OH)₂ nanoparticles is in the range of 20–40 nm.	–	The Zn(OH)₂ is orthorhombic structured	–	[51]
TiO₂–AC	667.82	Homogeneous dispersion of TiO₂-NPs onto the highly porous carbonaceous substrate is confirmed.	C-H vibrations in methyl and methylene groups: 2923 and 2854 cm⁻¹, respectively, OH groups and the stretching vibration of N-H groups:	–	–	[1]

Table 3 (continued)

NMOs-AC	BET (m²/g)	SEM/FESEM	FTIR	XRD	TEM	References
AC/γ-Fe₂O₃	347.8	Maghemite nanoparticles deposited on the surface of AC are uniform with the spherical shape.	3200 and 3400 cm⁻¹, C=O groups: 1734 cm⁻¹ C=C band: 1634 cm⁻¹ γ-Fe2O3 nanoparticles existing in the activated carbon: 453, 581, 630, 797 and 889 cm⁻¹	AC is amorphous and maghemite NPs are well covered on the AC surface	–	[69]

870.356 m^2/g but after modifying with iron oxide nanomaterials (Fe-AC), the surface area boosted to 940 m^2/g [147]. Based on Table 3, the high surface area of synthesized NMOs- AC exhibited that these adsorbents are so preferable and environmentally friendly for dye removal. According to XRD results, it can be concluded that some research studies showed that the structure of prepared AC is amorphous and NMOs successfully covered AC [20, 69, 131], while the results of Ghaedi et al. (2014) [47] and Saini et al. (2017) [148] have verified that the prepared nanoparticles had the hexagonal structure. Furthermore, it is better to note that based on Table 3, different functional groups were observed onto the NMOs-AC surface. In brief, the main functional groups can be summarized as follows: a) a sharp peak at a range of 3200–3434 cm^{-1} is associated with the hydroxyl group (O-H), b) the peaks occurred at 2923 and 2854 cm^{-1} may be due to asymmetric and symmetric C–H bands of methyl and methylene groups, respectively, c) a broad absorption at 3400–3437 cm^{-1} was ascribed to N-H groups, and d) the wide peak a peak at 1700, 1702, and 1734 cm^{-1} corresponds to carbonyl groups (C = O), and f) C = C band was detected at 1600 cm^{-1} and 1634 cm^{-1} [1, 20, 47, 148]. Moreover, according to TEM results, it can be determined that NMOs are usefully embedded into AC with a size range of 5–20 nm [20, 73].

Nano-adsorbent performance

The current review tries to explain the effects of process parameters such as contact time, initial concentration of pollutant, pH, and adsorbent dosage on the efficiency of NMOs-AC for the removal of dyes from aqueous solutions [38]. Identifying and optimizing the effects of the aforementioned parameters reduce the cost and increase the applicability of process in the elimination of dyes from water and wastewater [149].

Effect of adsorbent dosage

The effects of adsorbent dosage as a key process parameter can be investigated by adding different amount of adsorbents into solution. In addition, the amount of adsorbent is a major element influencing the adsorbent capacity with a certain volume of adsorbent under the operating conditions [150]. In general, the removal efficiency of dye increase by increasing adsorbent dosage due to a direct increase in the number of sorption sites that are available sites at the surface of adsorbent [6]. By increasing the adsorbent dosage, the ratio of dye molecule to the unoccupied site is very low, which could results in higher movement and distribution of dye molecules to the surface of adsorbent with requiring less energy to adsorb. Therefore, dye adsorption takes place faster, while it has

Table 4 NMOs-AC: performance and efficiency

Adsorbent	Adsorbate	Adsorption capacity (mg/g)	Adsorbent dosage (g/l)	pH	Initial con. (mg/l)	Contact time (min)	Temp (K)	Removal(%)	References
Magnetic AC/α-Fe$_2$O$_3$	Acid Yellow 17	–	1	5	60	30	–	100	[73]
CuO-AC	Acid blue 129	65.36	0.9	2	10 20	25	333.15	100	[48]
Magnetic cellulose/Fe$_3$O$_4$/AC	Congo red	66.09	3	4	20	–		99.8	[40]
Magnetic-AC	Methylene Blue	357.11	0.5	4	50	30	–	>80	[134]
ZnO-AC	Malachite green	322.58	0.15	7	20	15	–	100	[47]
SnO$_2$-AC	Malachite green	142.87	0.3	5	15	–	–	95	[49]
Zn(OH)$_2$-AC	Sunset yellow	83–114	0.46	3	30	5	–	97	[51]
PbO-AC	Methyl orange	333.33	0.4	2	15	30	–	98	[50]
ZnO-AC	Brilliant green	142.9	0.6	5	15	–	–	>97	[70]
ZnO-AC	Rhodamine B	128.2	0.6	7	50	100	343	>99	[148]
ZnO nanorods-AC	Crystal violet	113.64	0.5	7	24	5	–	99.82	[165]

mentioned that the adsorption capacity will decrease as the adsorbent dosage increases [57, 94]. Further increase of adsorbent dosage cannot significantly increase the process efficiency [94, 151].

Table 4 shows the previously described studies on the effect of NMOs-AC dosage on dye removal efficiency. M. Ghaedi et al. (2016) investigated the effect of varied PbO-NP-AC (0.005–0.02 g) as a simple adsorbent for the elimination of methyl orange (MO) from the aqueous phase. Based on achieved results, it was observed that rising the adsorbent dosage from 0.005 g to 0.02 g makes it possible to enhance the adsorption efficiency from 63.5% to 99.5% over equilibrium time (30 min) [50]. Nekouei et al. (2015) synthesized the CuO-NP-AC as a novel, simple, and efficient adsorbent for the removal of acid blue 129 (AB 129) from aqueous solutions. It was concluded that the elimination efficiency of AB 129 by CuO-NP-AC improved sharply from 40% to nearly 100% as the adsorbent dosage boosted from 0.01 to 0.045 g, then got a nearly constant value, as well as adsorption capacity, reduced by increasing adsorbent dosage [48]. Ghaedi.et al. (2013) examined the effects of dosage alteration (0.005, 0.01, 0.015, 0.02 g) at room temperature on the malachite green (MG) removal by ZnO-NP-AC, and it was observed that, MG removal percentage decline with a rise in initial dye concentration. Moreover, the adsorption capacity was decreased from 322.58 to 76.9 mg/g at 0.005 to 0.02 g of ZnO-NP-AC [47]. Another study by Morad et al. (2012) studied the adsorption process of cationic dye (methylene blue (MB)) from environmental solutions onto Fe$_3$O$_4$ impregnated onto activated maize cob powder. They found that when the adsorbent dosage increased from 0.2 to 1.0 g/100 ml, the maximum efficiency removal of 99.89% was achieved [57].

Effect of pH

The acidity and basicity of dye solutions can be determined by pH measurement as one of the main and crucial factors in the adsorption process [40]. Therefore, the study of the initial pH of the dye solutions due to the effect on the rate of adsorption is necessary to be performed in all adsorption processes [152]. The alteration of solution pH can directly affect the efficiency of adsorption processes because of the surface protonation of materials, changing of the adsorbent surface properties, and charge of dye [69, 153]. The acidic conditions (low pH value) can cause protonation, in which the functional groups of the adsorbent led to the creation of positive charge and the presence of the powerful attractive forces between the adsorbent and dyes resulting in increasing anionic dyes adsorption and decreasing cationic dye adsorption [84, 154–156]. The basicity of dye solutions (high pH value) causes negative charges on the adsorbent surface, which increases the efficiency of cationic dye adsorption and decrease anionic dye adsorption

[155]. In many research works to a better understanding of the adsorption mechanism the point of zero charges (pHpzc) or the isoelectric point (pH_{IEP}) has been investigated [157, 158]. Their results revealed that pHpzc can significantly indicate the kind of active centers and the capability surface charge of the adsorbent for adsorbed dyes, which are the electrokinetic properties of an adsorbent surface [159]. Moreover, the pzc can be interpreted by the pH value, which pH > pHpzc is ideal for cationic dyes because of negative charge of surface, and pH < pHpzc is suitable for adsorption of anionic dyes due to existence of positive charge on the surface [56, 131, 160, 161].

Agarwal et al. (2016) studied the effects of pH alteration on the removal of eosin B (EB) and sunset yellow (SY) by copper oxide nanoparticle loaded on activated carbon (Cu_2 O-NP-AC), and the results displayed that by increasing pH at the range of 2 to 10, the removal of SY and EB decreased [162]. Nekouei et al. (2015) examined the effect of solution pH on the removal of acid blue 129 using copper oxide nanoparticle-modified activated carbon as adsorbent, and they revealed that the maximum adsorption efficiency (around 100%) achieved at lower acidic pH of 2 [48]. Jethave et al. (2018) investigated the rhodamine b (RB) removal using AC-Fe-Pb nanoparticle (FePbO) and also pH ranged from 2 to 9. The results revealed that the maximum removal efficiency of RhB occurred at pH 7 [65]. Ranjithkumar et al. (2014) used magnetic-activated carbon/α-Fe_2O_3 nanocomposite as an adsorbent to remove acid yellow 17 (AYD 17) from water. The results depicted that with increasing pH from 1 to 14, the removal of AYD 17 reduced gradually for AC and AC/α -Fe_2O_3 and high removal efficiency of AYD 17 was obtained at acidic pH [133]. On the contrary, a study by Ghaedi et al. (2013) was synthesized ZnO-NP-AC as a green adsorbent. The results showed that higher adsorption achieved at the neutral pH, and a further decrease of pH cause a net positive charge on the adsorbent, which inhibits more removal of cationic dyes such as brilliant green (BG) [70].

Initial dye concentration

The effects of the initial concentration of dye are considered as the most important factor in the adsorption process so that the efficiency of the adsorption system highly depends on dye concentration in the solution [163]. Various studies demonstrated that by increasing the concentration of dyes, the adsorption efficiency is reduced, because of the saturation of available binding sites on the surface of adsorbent [163]. The efficiency of the system directly depends on initial dye concentration and adsorbent dosage, which at high dye concentration and available unoccupied sites an increase in the loading capacity of the dye occur because of high mass transfer driving force for adsorption at a high unsaturated adsorbent's surface [159, 164].

The methylene blue (MB) adsorption as a cationic dye using magnetic loaded activated carbon was studied by Altıntıg et al. (2017) at varied numerical parameters such as initial temperature (298–318 K), pH (3–9), adsorbent dosage (0.05–0.2 g/100 ml), MB concentration (50–250 mg/l) and contact time (5–60 min). The results showed that as MB concentration increased from 50 to 250 mg/l at a constant adsorbent dosage of 0.05 g/100 ml, the efficiency of the system diminished from 100% to approximately 73% [147]. Livan et al. (2017) evaluated the $NiFe_2O_4$ magnetic nanoparticle loaded onto activated carbon for the adsorption of two anionic dyes (Direct Blue 78 and Direct Red 31). They reported that at the contact time of 8 h, pH 2.0, mixing speed of 300 rpm and at temperatures of 30–50 °C by increasing dyes concentration, adsorption capacity raised and maximum adsorption capacity (q_{max}) for DR31 and DB78 were 99.67 and 209.13 mg/g, respectively [43]. Dil et al. (2015) showed that the adsorption of crystal violet (CV) using zinc oxide nanorods loaded on activated carbon (ZnO nanorods-AC) is highly depended on the CV concentration so that the efficiency of the system was declined by increasing CV concentration from 8 to 24 mg/l and also maximum removal was 99.82 (Table 4) [165]. A similar study by Fayazi et al. (2015) showed that initial Alizarin red S concentration had a negative effect on the removal efficiency so that by increasing the from 50 to150 mg/l, the removal percentage of dye decreased, furthermore, the high maximum adsorption capacity was observed at 108.69 mg/g [69].

Contact time

The adsorption time as a performance parameter is important because of the economic and technical points of wives [166]. Therefore optimizing contact time lead to developing an appropriate method for treating the pollutants [167], and also it can be effective to estimate the cost of water and wastewater treatment [56]. Studies have shown that in the adsorption process when the contact time rises, the active sites on the adsorbent surface are saturated which could result in a decrease in the adsorption rate. The adsorption does not take place with time when all sites occupy [168].

Altıntıg et al. (2017) employed magnetic loaded activated carbon (MAC) as a novel adsorbent for removing cationic dye (methylene blue). They investigated the effect of contact time (5–60 min) at a different dosage of MAC (0.05, 0.1, and 0.2 g/100 ml) and the results demonstrated that methylene blue adsorption was increased when contact time increased to 20 min but furthermore has not altered the adsorption of methylene blue [134]. Nekouei et al. (2015) reported the effect of varied contact time (1 to 30 min) on the acid blue 129 adsorption using activated

Table 5 Kinetic and Isotherms models

Models		Equations models	Constant parameters	References
Kinetic	PFO	$\frac{dq_t}{dt} = k1\,(q_e - q_t)$ $\log(q_e - q_t) = \log q_e - \frac{k1}{2.303}t$	q_t is the amount of adsorbed at any time (mg/g) q_e is the amount of adsorbed at equilibrium (mg/g) K_1 is the adsorption rate constant K_2 is the adsorption rate constant	[179]
	PSO	$\frac{1}{q_e - q_t} = \frac{1}{q_e} + k_2 * t$	k_i is the intraparticle diffusion rate constant	[180]
	IPD	$q_t = k_i * t^{\frac{1}{2}} + c_i$	α is the initial sorption rate (mg/ (g min))	[181]
	Elovich	$q_t = \frac{1}{\beta}\ln(\alpha\beta) + \frac{1}{\beta}\ln t$	β is the desorption constant (g/mg). The plot qt versus ln t having slope $1/\beta$ and intercept $[(1/\beta)\ln(\alpha\beta)]$	[182]
Isotherms	Halsey	$\ln q_e = [\frac{1}{n}\ln K] - \frac{1}{n}\ln C_e$	K and n are the Halsey constants Ce is a concentration in the aqueous phase (mg/L)	[184]
	Langmuir	$q_e = \frac{q_0 * b * c_e}{1 + b * c_e}$ $\frac{c_e}{q_e} = \frac{1}{K_L * q_m} + \frac{c_e}{q_m}$	b is Langmuir equilibrium constant (L g adsorbent^{-1}) q_0 is maximum monolayer adsorption (mg g adsorbent^{-1}) k_L is the Langmuir constant (L/mg) which is calculated by plot Ce versus Ce/q_e	[171, 185]
	Freundlich	$1/n\ Q = k_f * (ce)$ $\ln q_e = \ln k_f + \frac{1}{n}\ln c_e$	K_F and n are Freundlich coefficients	[172, 186]
	Temkin	$q_e = B\ln A * C_e$ $q_e = B\ln A + B\ln C_e$	B = is Temkin constant b (J mol^{-1}) is the adsorption heat A is fixed Temkin isotherm	[173, 178, 187]
	D-R	$\ln Q_e = \ln Q_{D-R} - \beta\varepsilon^2$ $\varepsilon = RT\ln(1 + \frac{1}{C_e})$	R (3314 J mol^{-1} K^{-1}) is a constant global gas in which T (K) is absolute Q_{D-R} the theoretical saturation capacity of the adsorbent b is a constant related to the mean free energy of adsorption per mole of the adsorbate ε is a logarithmic function of concentration called Polanyi potential R and T are gas constant and temperature in the Kelvin scale	[144]
	Sips	$q_e = \frac{Q_{max}\,K_s\,C_e^{1/n}}{(1 + K_s\,C_e^{1/n})}$	Q_{max}, Ks and1/n, which can be evaluated by fitting the experimental data	[174]

Table 6 Isotherm and Kinetics parameters

Adsorbent	Adsorbate	Isotherm constants									Kinetic constants									References
		Langmuir			Freundlich			Temkin			PFO			PSO			IPD			
		q_m (mg/g)	K_L or b or Ka	R^2	K_F	1/n	R^2	B1	K_T	R^2	k_1 (min^{-1})	qe, cal	R^2	k_2	qe, cal	R^2	kid	C	R^2	
ZnO -AC	bromophenol red	200	5	0.99	79.61	0.16	0.97	15.1	279.53	0.93	0.20	27.7	0.96	17.9	166.7	0.99	3.67	121.9	0.94	[183]
ZnO nanorods -AC	crystal violet	113.6	5.870	0.99	0.29	0.3	0.96	15.68	224.1	0.95	1.17	129.7	0.81	0.004	88.50	0.98	29.5	1.15	0.99	[165]
ZnO -AC	Rhodamine B	128.2	6.50	0.99	88.02	0.13	0.73	–	–	–	0.058	76.37	0.87	0.002	120.5	0.99	1.49	110.2	0.96	[148]
Ce-doped TiO$_2$-AC	Basic Red 46	58.61	9.57	0.99	4.12	0.36	0.92	5.85	2618.6	0.88	2.18	7.04	0.826	5.83	53.91	0.99	6.52	24.52	0.98	[55]
CuO-NP-AC	acid blue 129	65.36	1.14	0.99	31.35	0.24	0.97	7.97	108.92	0.97	0.18	5.14	0.86	0.01	11.52	0.99	0.71	7.83	0.94	[48]
Fe$_3$O$_4$/Au-AC	Rhodamine123	71.46	0.284	0.99	3.35	0.57	0.99	15.65	2.59	0.97	0.42	3.29	0.99	0.25	21.83	0.99	1.53	17.65	0.98	[188]
ZnO nanorods -AC	Eosin Y	61.72	3.05	0.99	5.28	0.2	0.88	10.57	42.53	0.93	0.72	14.34	0.83	0.00002	26.24	0.99	2.29	18.44	0.94	[149]
CuI-CuO-AC	malachite green	136.67	1.15	0.99	73.22	0.21	0.96	39.94	129.45	0.97	0.239	31.26	0.80	0.016	81.97	0.99	5.07	59.56	0.94	[54]

carbon loaded onto copper oxide nanoparticles (AC-PbO-NP). The results confirmed that maximum adsorbent efficiency of dye was obtained by increasing contact time [48].

Effect of temperature

Temperature is one of the critical variables for identifying the thermal character of the adsorption system [147]. Furthermore, the role of temperature can determine the adsorption mechanism as endothermic or exothermic [48]. Previous studies showed that high temperature also enhances the diffusion of adsorbate ions into the pores of the intra-particle [148]. As temperature rises, pores grow and new active sites are created due to bond fractures on the adsorbent surface, which can increase the adsorption efficiency [169].

Saini et al. (2017) investigated the effect of different temperatures (283–343 K) when other factors were kept constant. The results exhibited that as temperature increased, the removal efficiency of the Rhodamine B (RB) onto ZnO-AC increased from 92.0% to 99.5% [148]. A similar study by Mondal et al. (2020) demonstrated that increasing temperature from 293 to 318 K, at a varied initial concentration of MB (50 to 500 mg/l) had a positive effect on the MB removal efficiency [56]. A study by Nekouei et al. (2014) confirmed that with increasing temperature from 280 to 333.15 K the removal efficiency of the process increased sharply when parameters such as pH, dye concentration, adsorbent dosage, and contact times were 2, 10–20 mg/l, 0.9 g/l, 20–25 min, respectively (Table 4) [48].

Isotherm and kinetic studies

The interaction between adsorbate and adsorbent is indicated by the adsorption isotherms and also the parameters derived from various models provide details on the adsorption process [147]. An adsorption isotherm is an important tool for assessing adsorbent distribution over solid/liquid boundaries, as well as for the estimation of adsorption capacity [153]. Therefore, the data from isotherm studies can be used to design an appropriate adsorption system [170]. Several isotherms have been developed to describe equilibrium relations such as Langmuir [171], Freundlich [172], Dubinin-Radushkevich (D-R) [144], Temkin [173], and Sips [174]. Previous works showed that the Langmuir isotherm model is generally used to express the single-layer adsorption process [175], but Freundlich adsorption isotherm as an empirical model suggests that the adsorbent surface has heterogeneous structure [176]. Dubinin–Radushkevich isotherm as an empirical model is used for the isotherms analysis with a high degree of regularity

[177]. The Tempkin isotherm supposes that the heat of adsorption of all molecules instead of logarithmically diminishes linearly, moreover, the Temkin isotherm equation has the indirect effects of interaction between adsorbent molecules based on this, it assumes that the heat adsorption of all molecules in the layer can be linearly reduced by surface coating. This isotherm has predictive power in a wide range of concentrations [173, 178]. Moreover, the Temkin isotherm indicates that the decline in the adsorption heat of all molecules takes place with a linear pattern proving that the binding energy is homogeneous [169]. The Sips concept is an alternative empirical approach with the characteristics of the Freundlich and Langmuir isotherms [174]. The equations of the above models are summarized in Table 5.

Studies of the kinetic parameters as a valuable method are necessary to estimate the adsorption rate and offer the required information for the modeling and development of adsorption processes [40]. To better understand the behavior of the adsorption process, various kinetic models are developed but the main kinetic models which are used for the removal of the dye using NMOs-AC are Pseudo-first-order (PFO) [179], Pseudo-second-order (PSO) [180], Intraparticle diffusion (IPD) [181], and Elovich [182]. The pseudo-first-order equation is usually employed for estimating the initial stage of the adsorption mechanism [126]. Pseudo-second-order suggests that chemisorption is a rate-limiting step, and dye sorption is strongly related to the active ion functional group on the adsorbent surface [56], while the Elovich theory models are widely examined for chemisorption [160] and also intra-particle diffusion can be deemed as a widest method for the detection of the pathway involved in the adsorption system [126]. The equations of the kinetic models are summarized in Table 5.

Herein, experimental data, adsorption kinetics, correlation coefficients, and their constants for NMOs-AC as a dye adsorbent were compared in Table 6. According to the table, it was found that among the different Isotherms (Langmuir, Freundlich, and Tempkin), the experimental data for prepared NMOs-AC was best fitted to the Langmuir isotherm, which completely describes the adsorption is monolayer [149, 165, 183]. Nekouei et al. (2014) used major isotherm models (Langmuir, Freundlich, Dubinin–Radushkevich, and Tempkin) for evaluating acid blue 129 (AB 129) adsorption, and the results showed that Langmuir model provides a great link between experimental results, and also the maximum capacity of AB 129 was 65.36 mg/g [48]. Furthermore, studies have reported that the pseudo-first-order (PSO) kinetic model due to the high correlation coefficients ($R^2 \geq$ more than 99) is the most reliable model for describing the adsorption behavior of NMOs-AC [149, 165, 183]. Dil et al. (2019) applied four kinetic models (PFO, PSO, IPD, and Elovich). The results exhibited that the efficiency of the Basic Red 46 adsorption onto Ce-TiO2-NPs-

AC was expected by the PSO model ($R^2 \geq 99$) [55]. Also, the result of a study by Nekouei et al. (2015) depicted that the adsorption kinetic equilibrium data were best fitted by the PSO model with correlation coefficients R^2 more than 99 [54].

Conclusion

The development of dyes removal methods is a need of recent days, due to the inability of conventional methods and discharging of the high volume of toxic dyes into water bodies. Among developed methods, many researchers moved on the adsorption using metal oxide nanomaterials loaded on activated carbon as a promising method to overcome the previous shortcomings of adsorption by AC. This technique because of many advantages such as the recovery of magnetized powdered activated carbon, synergically improves the operation feature of integrated adsorbent, high effective dye removal and economic point of view has been developed. This paper highlighting the most important aspects of adsorption using AC- NMOs such as their physical and chemical characteristics, effects of main factors on the process, and isotherms and kinetics parameters.

Compliance with ethical standards

Competing interests The authors declare that they have no competing interests.

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