A Review on Dye Reduction Mechanism using Nano Adsorbents in Waste Water

Kavithayeni V, Dr Geetha K, Akash Prabhu S

ABSTRACT:

Novel Nano adsorbents recently pay its attention in various applications, especially in pollution control division. The kinetic studies of different adsorbents for the treatment of wastewater discharged from Textile industries have been discussed here. The methods involved in the adsorbent synthesis, which is a major part on adsorption of numerous dyes used has also taken into consideration. This paper reviews about the unique and exuberant performances of three categories of adsorbents which could be a key to treat textile waste water in future aspect.

Keywords: Adsorption, Dyes, Nano adsorbents, Textile wastewater.

I. INTRODUCTION

Water pollution is a major crisis around the world, where one- third of the pollution is caused by the industrial effluents discharge from chemical and Food industries after the industrial revolution, which increases the surface water pollution more than 20 times [1], [2]. Moreover, 20% of people died every year, due to the consumption of unsafe drinking water [3]. Discharge of wastewater from textile industries, power plants, chemical industries contain the recalcitrant organic compounds and heavy metals1 which are toxic and non-bio gradable [4], [5]. Among various Industries, textile industries are noticeable, because they are consuming a large amount of water, energy, and chemicals [6]. Especially dyeing process in Textile industries use over 100,000 synthetic dyes in which 1-20% dyes are directly dispensed into the Textile wastewater, which causes 17-20% of water to get pollute [7] - [10]. Generally, synthetic dyes preferred are azo dyes, VAT dyes, Indigoid, Catinoic, phthalocyanine, Anthroquinone, Anionic, sulfur and Reactive dyes for dyeing and finishing processes [11], [12]. Comparing with other dyes Azo dyes are commercially viable which contains azo groups (N=N) with both the naphthalene and benzene rings [13].

Human health and Environmental related effects due to the presence of Benzidine in azo dyes are needed to be considered because of carcinogenic and mutagenic derivatives along the allergic reactions, skin degeneration, asthma, nausea, dysfunction of kidney, liver etc [5], [11], [14], [15]. Heavy metals present in wastewater such as Lead, Nickel, Cadmium etc., affects the agriculture products [16]. Aquatic systems also get contaminated and the effluents discharged in that blocked the sun light penetration to the organisms present [17]. Dye reduction technologies followed in a conventional way includes Membrane filtration, ozonation, coagulation, reverse osmosis, filtration, precipitation, enzymatic decomposition, active sludge bio chemical process etc facing the challenges owing to synthetic origin and complex structure present in dyes. Where these dyes are having the color discharging effect instead of removing toxic substances [15], [18], [17], [19], [20], [21]. Though Biological methods have the advantage of low cost, it takes long duration time to proceed [20]. Chemical methods and Filtration method exhibits heavy sludge to dispose with hazadorous contaminates [20], [21].

Low cost adsorption and Advanced Oxidation Process (AOP) attracted the recent researchers to work on the dye degradation application, because of its rapid dye removal rate [22], [23]. Adsorption is the way of decolorizing, separating, and detoxifying than the other conventional techniques followed with the ease of operation and high efficiency [24], [25]. Comparing with AOP, Adsorption method is widely used because of its efficacy of removal of toxic substances in a better way [26]. Recent researchers are focused on Nanoparticles based adsorbents owing to its distinctive surface strucuture, composition and functionalism with compact size and less concentration adsorption [27], [28]. Both Conventional and Non-conventional adsorbents are existed where conventional adsorbents includes wood, peat, coals, silica gel, bark, sawdust etc., and Non- conventional adsorbents holds clays, sagaun, cotton waste, hydro gels, bio sorbents [29] - [32].

Adsorption mechanisms and interactions are needed to be considered to understand the kinetics of dye reduction. Physisorption, Ion-exchange, Precipitation and Chemisorptions are the main mechanisms associated with the interactions like Surface adsorption, Electrostatic interaction, and VanderWaals interaction, chelation, Proton displacement and covalent binding etc., [33], [34]. Several isotherm models like Langmuir model, Elovic liquid film deposition model, Halsey model, Brener-Emmet Teller model and etc., are present to define the contact between the pollutants adsorbed by the adsorbent and in water [35]. The general characteristics possessed by the nano adsorbents should be of eco friendly basis, recyclable, excessive selectivity along adsorption capacity and ease of removal of dyes [36], [37].

This paper expanded its review on dye degradation ability of three different classes of nano based adsorbents along its fabrication methods and efficiency.

II. DIFFERENT CLASSES OF NANO ADSORBENTS

Wastewater treatment involves

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Kavithayeni V, Nanotechnology Division, Periyar Maniammai Institute of Science and Technology, Thanjavur, India.

Dr Geetha K, Nanotechnology Division, Periyar Maniammai Institute of Science and Technology, Thanjavur, India.

Akash Prabhu, Nanotechnology Division, Periyar Maniammai Institute of Science and Technology, Thanjavur, India.

the different classes of nano adsorbents which could contribute more in a way of dye removal. Carbon based, Metal Oxide based, Polymer based nano adsorbents are some of the prominent one to be applied for the efficient and rapid dye removal[37], [39].

A.Carbonaceous Adsorbents

Wastewater treatment techniques commonly includes carbon based adsorbents, for the sake of porous nature, structure stability, non-lethal with high toxic adsorption ability [40], [41], [42]. Carbon Nano Tubes (CNTs), Graphene and Activated carbon are the most preferential carbonaceous materials used in most of the processes of dye adsorption [43], [44].

a. Activated Carbon

Activated Carbon works through the Vander-waals force of attraction with the adsorbate that can exhibit the micro porous nature and elaborative surface area along the ion exchange characteristics for weak acidic ions, especially for adsorbing heavy metal ions [43], [45]. Granular Activated Carbon (GAC) and Power Activated Carbon (PAC) are the two basic types exist in AC, in which GAC is a favorable form of adsorbent for both the organic and inorganic pollutant removal [46], [47]. Advanced models of AC with ease of operation is also found out as AC fibers and pellets form [46]. The factors which determine the prominent AC are molecular size, Functional groups attached with the surface, Size distribution of particles, solubility and etc.,[48] [49]. The general term of precursor materials consumed for the fabrication of AC included wastes from agriculture, shells of coconut and derivatives of wood and coal mainly [50], [51].

Al-degs et al., (2001) reported that the Activated Carbon FS-400 has the high capacity of adsorbing anionic dyes especially methylene blue [52].Yasin Y et al., (2007) reports the Methylene blue dye removal at high percentage and attains the equilibrium stage after 180 minutes of treatment with activated carbon [53]. Kanawade et al., (2011) reported about the methylene blue dye removal followed by Isotherm model with three different concentrations of 5, 10, 20 ppm at 80 minutes of duration process at 27^{o} C. [54]. Pathania et al., (2017) reported that the Methylene Blue dye was removed in a maximum level through the activated carbon at the pH of 7.8, followed by Pseudo-second order kinetics and Intraparticle diffusion model [55].

b. Graphene Oxide

Graphene is basically a two-dimensional layer belongs to carbon family with excellent physical, chemical, mechanical, and electronic properties along high optical transparency and thermal conductivity [37], [56], [57]. Other than these properties Graphene oxide put forth the properties like compressive strength, large surface area along high pore volume [58]. The functional groups attached with Graphene oxide are carbonyl, hydroxyl and epoxy which highlights the material for various applications [59]. Due to its magnetic properties, it is possible to attain the liquid- solid separation [60]. Exfoliated GO and reduced GO pave its application in synthetic and natural organic pollutants removal from wastewater [37], [44]. Especially GO is a biodegradable adsorbent with outstanding adsorption capacities [58].

Ramesha et al., (2011) reported about the two types of GO where Exfoliated GO is applied for cationic dyes of 95% efficiency and reduced GO was applied on anionic dyes with 95% dye removal efficiency [44]. Apul et al., (2013) reported the GO capability of Natural Ogganic and synthetic Organic pollutants removal in a better way [61].Robati D et al., (2016) reported the dye (BR-12, MO) removal efficacy of Graphene Oxide for the contact of 100 minutes with pH of 3 [62].

c. Carbon Nanotube

CNTs are noticeable semiconductors because of distinctive structure with venerable chemical, opt electrical, physical and mechanical properties [37], [63]. The specific surface area, layered and hollow structure of CNTs marked it as a ideal adsorbent for the removal of both organic and inorganic pollutants like heavy metals, arsenic, lead, 1,2-dichlorobenezene, radionuclides and organic chemicals [64], [65], [66], [67], [68]. Hydroxyl and carboxyl groups attached with the CNTs exhibited its functionality in a better way [69]. CNT based adsorbents attracted with the organic molecules through the various interactions and forces where they are, hydrogen bonding, Vander Waals forces, hydrophobic interactions, non-covalent forces and mainly the presence of π-π bonding [70], [71].

Gong et al., (2009) reported the effect of cationic dyes (MB, NR, BCD) removal by Multi- Walled CNT (MWCNT) followed by Freundlich model [64]. Yao et al., (2010) presented the dye adsorption efficiency of MB as 55.34 mg/g and MV as 46.2 mg/gusing MWCNT [72]. Ai L et al., (2011) reported the dye removal efficacy of MWCNT with contact time, pH, initial dye concentration as important factors of 48.06 mg/g adsorption capacity [65]. Ghaedi et al., (2012) reported the better adsorption capacity of MWCNT of 108.97 mg/g for Methylene blue dye [73]. Vinod Gupta et al., (2013) reported the maximum adsorption of cationic dyes based on electrostatic and π-π bond interaction [74].

B. Metal Oxide Based Adsorbents

Generally Metal Oxide (MO) adsorbents has size quantum effect with high capacity and selectivity [75]. Nano sized MO adsorbents have high specific surface energy and affinity due to its reduction in size from micro level [76].Metal Oxide Nanoparticles (MONPs) engrossed the recent researchers, due to its unique structure with edge surface sites and high density along physical and chemical properties [77]. It exhibits pore volume in varying sizes, Low solubility, low cost, and strong mechanical structure [37], [78] [79]. MO adsorbents are chemically stable and resist against the organic chemicals, alkalis, oxidants and acids [80]. The presence of oxides in MO possess ligand sorption in a better way due to the formation of acid-base interactions [81]. Owing to its fast adsorption rate, it can be applied in wastewater treatment [79]. Secondary pollution formation is also no possible in MO adsorbents because of the minimal environmental impact [37].

Gao et al., (2008) reported about the MgO as an outstanding adsorbent for the removal of heavy metal ions and organic pollutants [82]. Feng et al., (2012)reported the arsenic metal ions removal from wastewater using Fe_3O_4 [83]. Hua et al., (2012) reported about the fast kinetics, high preferable adsorption of Nano MO adsorbents for the improved dye

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removal process [76].Piao Xu et al., (2012), reported the non-invasive Iron oxide application on wastewater with adsorption rate of 104.2 mg/g [84]. Anitha et al., (2015) reported about the Copper Oxide (CuO) adsorbent on dye removal of 55% at the pH level of 5.2 [85]. Acelas et al., (2015) reported the phosphate removal of Zirconium oxide with 83% and Copperoxide adsorbent with 86% [86]. Bincy Rose et al., (2018), reported the Malachite green dye removal by $CuFe_2O_4$ of 200 mg/g with 85% efficiency [87].

C. Polymer Based Adsorbents

In recent years organic polymers with tailor made cross link attracted the researchers in adsorption process, due to their reversible process and variable properties such as vast surface area, mechanical rigidity, high porosity, high adsorption capacity and regeneration of adsorbent at mild condition without decrease in sorption capacities [88-91]. Polymers have different classes like copolymers, linear free chains, membranes, permanently and physically cross-linked hydrogels and amphiphillic block [89, 92, 93]. The network structure of polymer has been achieved by interpretrating the polymer networks (IPNs), which increases the adsorption rate of polymer material [89]. The interaction between polymer and dyes are listed as hydrogen bonding, hydrophobic, electrostatic and ion exchange interactions [89,94]. Most common natural polymer used for adsorption are starch, chitosan, alginate, cellulose and cyclodextrin [89].

Susan et al., (2011) reported the Porous poly(vinyl alcohol) gels with the adsorption efficiency of 35% against congo red dye [95]. OMER et al., (2011) reported the adsorption efficiency of Poly(acrylamide-co-sodium methacrylate)/poly (ethylene glycol) semi-IPN on janus blue B as 87% [96]. Niyaz et al., (2012) study and reported the adsorption capacity of poly (amidoamine-co-acrylic acid) copolymer against direct red 31, direct red 80 and acid blue 25. The maximum dye adsorption rate was 3400 mg/g, 3448 mg/g and 3500 mg/g for direct red 31, direct red 80 and acid blue 25 respectively [97]. Omer et al., (2013) reported adsorption capacity on poly(acrylamide-co-sodium 4-styrenesulfonate) against the janus blue B as 67% [98]. Jianwei et al., (2014) study and reported the adsorption capacity and efficiency of polydopamine microspheres about 98.62 mg/g and 98.62% against methylene blue [99].

III. SYNTHESIS ROUTE: A MAIN PART OF ABSORPTION

Adsorption Efficiency of different adsorbents can be achieved from the composition and structure, which mainly depends on the fabrication route of adsorbents and the particle size distribution [87]. Some of the researches have been proved that synthesis route pave its influence on adsorbent efficiency for dye reduction mechanism. Rafatullah et al., (2010) mentioned in his work that outstanding performance of activated carbon can be get by raw materials used to synthesis and activation temperature [29]. Kannan et al., (2001) reported the dye removal rate of 472.10 mg/g using Straw as a raw material in the fabrication of Activated carbon [100]. K. Legrouri et al., (2005) reported his work on activated carbon prepared with molasses and sulphuric acid with the dye adsorption rate of 435mg/g [101]. Ramesha et al., (2011) reported that there are two various physical and chemical techniques employed for the GO based adsorbents which was not suitable for the dye degradation applications. Exfoliated method of synthesizing GO is most preferable for that because it gives a hydrophilic effect which increases the negative charge density of functional groups attached with GO, results in the the maximum dye adsorption rate of 95% for cationic dyes [44]. Yujin et al., (2011) reported about the CNT adsorbent synthesis via Chemical Vapor Deposition through the cracking by acetylene is an efficient one [102].

Patil et al., (2002) mentioned in his work about the Solution Combustion Method, an efficient synthesis route for general MO based adsorbents which could be applicable for dye reduction due to its precised reaction time and yield [103]. Abdel et al., (2015) reported about the aqueous solution synthesis method of MO CO_3O_4/ SiO_2 with the adsorption rate of 53.87 mg/g [104]. Moussavi et al., (2009) presented in his work about the sol-gel method for MgO with the efficacy of 166.7 mg/g [105]. Maria Visa et al., (2009) reported about the TiO_2 adsorbent synthesis via Laser pyrolysis with the efficiency of 85.39mg/g [106]. Vesna et al., (2013), reported in his work about the Polymerization types like Free-radical, Atom Transfer Radical Polymer (ATRP) are the most prominent techniques for fabricating Cross linked polymers and graft polymers with high adsorption rate on dyes [107].

I. Table.1 represents the various synthesis routes for nano adsorbent applied on dye removal process

Dye Adsorbed	Material	Fabricated Method	Precursor	Adsorption Capacity (mg/g)	Adsorption Efficiency (%)	Reference
Methylene Blue	Activated Carbon	Thermal Reduction	Molasses	435	-	[101]
	KOH treated Activated Carbon	Thermal Combustion	Coconut shell	45.9	-	[53]
	Activated Carbon	-	-	533		[52]
Cationic Dye	Graphene Oxide	Chemical Exfoliation	Graphite	-	95	[44]
		Chemical Reduction			50	
Anionic Dye		Chemical Exfoliation			Negligible	
		Chemical Reduction			95	
Methylene Blue	Magnetic MWCNTs nanocomposite	Chemical Co-Precipitation	MWCNT and Iron Oxide (Fe_3O_4)	15.74	99.16	[64]
Natural Red				20.33	98.33	
Brilliant Cresyl Blue				23.55	98.8	
Methylene Blue	MWCNTs	CVD	Acetylene	53.74		[72]
Methyl Violet				46.2		
Methylene Blue	Magnetite loaded MWCNTs	Ultrasonification	FeCl3·6H2O, sodium acetate (NaAc), ethylene glycol (EG)	48.06		[65]
Methyl Red	Oxidized MWCNTs	Oxidation	MWCNT	108.7		[73]
Malachite Green	$CuFe_2O_4$	Solution Combustion	Copper nitrate Cu(NO3)2·3H2O, ferric nitrate Fe(NO3)3·9H2O, glycine (CH2NH2COOH)	200	85	[87]
Methylene Blue	Co_3O_4/SiO_2	Aqueous Solution Method	Cobalt(II) choride,sodium silicate, poly ethulene glycol, sodium hydroxide	53.87		[104]
Reactive Blue 19	MgO	Sol-Gel Technique	Magnesium Chloride dehydrateand and sodium Hydroxid	166.7		[105]
Reactive Red 198				123.5		
Methyl Orange	TiO_2	Laser Pyrolysis	Titanium Chloride	85.39		[106]
Janus Blue B	Poly(acrylamide-co-sodium methacrylate)/poly(ethylene glycol) semi-IPN	Polymerization	Acrylamide (AAm), sodium acrylate(SA), poly (ethyleneglycol) (PEG, poly(ethylene glycol) dimethacrylate (PEGDMA)		87	[96]
Direct red 31	Poly (amidoamine-co-acrylic acid)	Two Step Polymerization	Maleic anhydride, ethylenediamine, toluene, potassium persulfate, benzoyl peroxide	3400	-	[90]
Direct red 80				3448		
Acid blue 25				3500		

IV. COMPARION VIEW OF ADSORPTION

Among the three different carbon based adsorbents, Sadegh et al., (2017) study correlates the carbon based adsorbents and resulted that Graphene and Graphene Oxide adsorbents are well suited for the adsorption mechanism than activated carbon and CNTs in most cases through the various comparison results [37].

II. Table

S.No	Type of Adsorbent	Dye/heavy metals/acids	Adsorption Capacity (mg/g)	Reference
1.	Activated Carbon	Reactive Red	10.00	[108]
		Methylene Blue	41.8	[53]
2.	Carbon Nano Tube	Nitrofurazone	59.9	[109]
		Methylene Blue	48.06	[63]
3.	Graphene (reduced GON)	Chlorpyrifos	1200	[110]
	Graphene (GO)	Methylene blue	1995.6	[71]

Table. 2 represents the adsorption rate of three carbon based adsorbents in different cases of dye removal. From the results, it is clear that the Graphene based adsorbents show the maximum adsorption capacity than activated carbon and CNTs.

Comparing with the Graphene/GO based adsorbents, Metal Oxide based adsorbents have relatively low adsorption capability [111-113]. By Table. 2, it can be concluded that MO based adsorbents are better absorbents when compare with CNTs and Activated carbon.

III. Table

S.No	Type of Adsorbent	Dye/Heavy metals/acids	Adsorption capacity (mg/g)	Reference
1.	TiO_2	Cd	16.69	[111]
	Iron Oxide	PhA	104.2	[84]
	MgO	Reactive blue 19	166.7	[105]
2.	Graphene	Cd	106.3	[112]
		Malathion	800	[110]
		Cationic red X	217.39	[113]

Table. 3 represents the different classes of MO adsorption efficiency with Graphene based adsorbents in a detailed manner. It is clear from the Table. 3 that Graphene has a reliable adsorption capacity than MO based adsorbents.

Polymer based adsorbentspossess maximum adsorption capacity than Activated carbon and CNTs but relatively matches the adsorption rate of Metal Oxide based adsorbents. While comparing the Graphene and Graphene based adsorbents, Polymer based adsorbents are low in adsorption capability can be clearly shown in the table 4.

Table. 4

S. No	Type of adsorbent	Dye/heavy metals/acids	Adsorption Capacity (mg/g)	Reference
1.	Acrylic Ester polymer	phenol	100	[114]
	Macroporous polystyrene	Phenol/salicyclic acid	82	[115]
	Aminated polystyrene	Aromatic acids	200	[116]
2.	GO	Methylene Blue	714	[117]
	rGO	Endosulfan	1100	[110]
	GO	Pb	842	[118]

These above mentioned tables clearly explains the Graphene based adsorbents are the most prominent dye removal agent than other carbon and metal based adsorbents.

V.CONCLUSION

Waste water treatment, a needed one in recent days due to the increase in insufficient water Dye degradation process involves numerous techniques which does the color removal but not the toxicity level of the dyes used in textiles. Many researchers moved onto Photo degradation and Adsorption techniques due to the effective dye removal kinetics. Low cost, conventional, Non-conventional adsorbents are the basic types of adsorbents involved in adsorption phantasm. This paper emphases about the three major categories of carbon based, MO based, Graphene based adsorbents which have different adsorption rate while comparing with each other. Synthesis is also an important

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parameter that could enhance the dye removal efficacy of adsorbents are also discussed here. From the results obtained on the basis of past and present year researches, the adsorption rate of Graphene based adsorbents are in the high level than carbon and metal oxide based adsorbents. Graphene and Graphene Oxide adsorbents are not only possesses best adsorption results but also reduces the sludge formation and secondary pollution during dye removal process.

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