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## **Dye Contaminated Waste Water Treatment through Metal-Organic Frameworks (MOFs) based Materials**

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**Table S1.** Comparison of important survey papers related to the MOF-based various water treatment techniques.

Sr. No.	Adsorbents	Research group	Main Focus	Main contribution	Ref
1	Ferric-MOFs	Johari <i>et al.</i>	Dye removal using Fe based MOFs	A comprehensive review of the MIL-100(Fe), used for removing different types of dyes in wastewater treatment.	<sup>1</sup>
2	MOFs	Lee <i>et al.</i>	Adsorptive removal of dyes	A thorough survey of different MOFs utilized in dye removal, especially, focusing on the factors affecting the adsorption process.	<sup>2</sup>
3	MOFs based membrane	Yoon <i>et al.</i>	Water purification	MOF-based FO/RO/NF/UF membrane filtration used in water purification, a brief survey of their utilisation in dye removal.	<sup>3</sup>
4	MOFs	Ayati <i>et al.</i>	Removal of azo dyes from wastewater	Primarily focusing on the azo dye infected water treatment via different MOFs along with a general overview of the effectiveness of various MOFs adsorbents under different physiochemical processes.	<sup>4</sup>
5	MOFs	Jhung <i>et al.</i>	Removal of hazardous material	A review on the separation of various hazardous compounds using different MOFs, includes dye removal.	<sup>5</sup>
6	MOF based membranes	Hu <i>et al.</i>	Water treatment	A survey on MOF-containing membranes in the potential environmental application, especially focusing on the removal of heavy metal ions, micropollutants, dyes, and seawater desalination.	<sup>6</sup>
7	MOFs based polymer mix matrix membrane	Nady <i>et al.</i>	Water purification	A full review on the different recently introduced MOFs-MMMs materials in water purification applications. A brief discussion of their application in dye removal.	<sup>7</sup>
8	MOFs	Bello <i>et al.</i>	Dye adsorption	A detailed review of the selected MOFs used for the Laboratory, industrial, environmental, and dye adsorbents.	<sup>8</sup>

9	Magnetic Nanoparticles@MOFs material	Zhu <i>et al.</i>	Sustainable Environment Adsorbents	An in-depth review on the removal of hazardous contaminants (organic pollutants, heavy metal ions, and dye) from the environment.	9
10	MOFs	Jhung <i>et al.</i>	Removal of hazardous material: possible adsorption Mechanism	MOFs-based adsorptive removal of hazardous material reviewed (includes dye) and also discuss interactions or mechanisms of dye adsorption.	10
11	Water stable MOFs	Duu-jong <i>et al.</i>	As adsorbents in aqueous solution	A mini-review on the efforts to develop water-stable MOFs as efficient adsorbents in aqueous solutions.	11
12	MOFs	Armentano <i>et al.</i>	Water remediation	A comprehensive survey, critically highlighting the latest developments achieved in the adsorptive removal of hazardous material (metal cations, inorganic acids, oxyanions, nuclear wastes, organic – pharmaceuticals and personal care products, artificial sweeteners, and feed additives, agricultural products, organic dyes, and industrial products).	12
13	Graphene oxide and MOFs composite	Huang <i>et al.</i>	Synthesis and application	A detailed review on the synthesis and applications of GO/MOF composites, especially focusing on the adsorption of various organic pollutants (includes dye).	13
14	MOFs	Zhou <i>et al.</i>	Removal of organic pollutants	A review on recent literature of the effectiveness of MOFs for the adsorption of selected organic pollutants (dyes, antibiotics and pesticides) from aqueous solution.	14
15	Magnetic MOFs (MMOFs)	Zeng <i>et al.</i>	Environmental monitoring and remediation	A detailed survey on the synthesis and classification of magnetic MOF composites, focusing on their physicochemical properties and remediation of environmental contaminants (includes dye).	15

16	MOFs	Kai-lv <i>et al.</i>	Removal of common aromatic pollutants	A comprehensive survey, especially related to the removal and preconcentration of aromatic pollutants (organic pollutants, pharmaceutical by-products, and dyes) using MOFs are provided in wastewater treatment.	16
17	MOFs/carbon based material	Feng <i>et al.</i>	Environmental remediation	A mini-review on recent advances of MOFs-Carbon based composites in environmental remediation.	17
19	MOFs	Rizwan <i>et al.</i>	Removal of environmental contaminations	A review article on the application of MOFs toward how to remove the toxic agents (organic, inorganic, dye and heavy metal pollutant) from water.	18
21	MOFs	Khan <i>et al.</i>	Dye removal mechanism	An in-depth review on the removal of the dye from wastewater using different MOFs, focuses on adsorption mechanisms or interactions	19
22	MOFs	Horcajada <i>et al.</i>	Removal of organic contaminations	A review article on the elimination of anthropogenic pollutant (Pharmaceuticals and Personal Care Products, Herbicides and Pesticides, and organic dyes) from waste water using MOFs.	20
23	Al-MOFs	Samokhvalov <i>et al.</i>	Sorption in solutions	A review Article provides an analysis of the published reports on adsorption of various organic and <u>inorganic compounds</u> (including dye) on <u>microporous</u> and <u>mesoporous</u> Al-MOFs in the liquid phase.	21
25	MOFs + MOF gels+ magnetic MOFs	Kurkuri <i>et al.</i>	Water remediation and separation of oils from water	A detailed review article on the MOF-based material like aerogels/hydrogels, MOF-derived carbons (MDCs), hydrophobic MOFs, and magnetic framework composites (MFCs) to remediate water from contaminants (includes dye) and for the separation of oils from water.	22
26	MOFs	Suresh <i>et al.</i>	Adsorptive dye removal	A review article on the adsorptive removal of hazardous dye molecules from wastewater utilizing MOFs.	23
27	MOFs	Man-Au <i>et al.</i>	Dye removal	A mini review on the removal of the dye moieties from waste water by MOFs.	24

28	MOFs+ Magnetic MOFs + MOF based membrane + MOF based gels		Dye removal using MOF based technologies	An extensive survey on MOF-based technologies includes magnetic MOFs, MOF-based membranes, and hydrogel/aerogels, which are used in the removal of hazardous cationic and anionic dyes from dye wastewater.	Current review article
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**Table S2.** Advantages and disadvantages of different dye removal methods.<sup>25-47</sup>

Methods		Advantages	Disadvantages
<b>Biological</b>	Adsorption by microbial biomass, both alive and dead	Several microbial species have a strong affinity for certain colors. Good option with a low cost.	The majority of hazardous dyes are incompatible.
	Dye bioremediation by Anaerobic biomass	Decolonization of water-soluble and azo dyes is easy.	Hydrogen sulphide and methane are created during the anaerobic process.
	White-rot fungi in dye degradation	To degrade dye, white-rot fungi produce enzymes.	The production of enzymes is not a reliable process.
<b>Chemical</b>	Photochemical	There will be no sludge produced, and COD will be reduced significantly.	Byproducts are formed and Light penetration limitation.
	Sonolysis	Chemical additives are not used, and so no additional sludge is produced.	It needs a large amount of dissolved gas (O <sub>2</sub> ). At this stage of reactor development, it is not economically viable.
	Fenton's reagent	Fenton's reagent is a chemically highly effective treatment method.	Produce sludge.
	Electrolysis	Decolonization of soluble and insoluble dyes. No sludge buildup and no chemical consumption.	High flow rates result in a direct reduction in dye removal. The high cost of electricity is a barrier.
	Oxidation	Very easy to use.	To start the process, need an oxidizing agent. Disperse dyes are not compatible. This method can be related with secondary contamination.
	Sodium hypochlorite (NaOCl)	Enables and enhances azo-bond cleavage.	Further chlorine pollution. This method can be related to secondary contamination.
	Wet air oxidation	This well-established method is especially well-suited to effluents that are both too dilute for incineration and too hazardous for incineration.	The use of high pressure and temperature is connected with high cost.
	Advanced oxidation Processes (AOPs)	Create a higher number of extremely reactive free radicals that outperform traditional oxidants in terms of decolonization.	Complete oxidation may not be achieved due to unwanted toxic byproducts. The presence of radical scavengers reduces the efficacy of various pH-dependent activities.
	Ozonation	Ozone can be used in a gaseous form, and it does not affect the volume of wastewater or sludge.	The half-life is relatively short (20 min)
<b>Physical methods</b>	Ion exchange	The adsorbent can be renewed without losing its effectiveness, and dye recovery is theoretically achievable.	Dye-specific ion exchange resins are expensive to regenerate.

	Filtration	Most dyes can be removed.	Generates a considerable amount of sludge. Membrane replacement is expensive.
	Floatation	High removal efficiency.	The high cost of electricity is a barrier.
	Coagulation	Removing dispersion, sulphur, and vat dyes in a cost-effective manner.	Method is pH-dependent; generates a considerable amount of sludge.
	Adsorption	Good at removing a wide range of dyes, including azo, reactive, and acid dyes; especially useful at eliminating basic dyes.	Regeneration is costly and results in an adsorbent loss.
<b>Hybrid methods</b>	Solar/Fenton	Highly effective on orange-II dye.	Produce sludge require sunlight for processing, and very costly.
	UV/Fenton	Effective on disperse dyes.	Requires UV radiation on a very large scale and produce sludge

**Table S3.** Dye adsorption through MOFs and MOF composite materials in last decades.

MOFs or MOFs composite	Dye name	Adsorption efficiency/Removal capacity	Year	Ref.
MIL-53	MO	57.9 mg/g	2010	48
MIL-101	MO	114 mg/g		
ED-MIL-101	MO	160 mg/g		
MOF-235	MB	252 mg/g	2011	49
	MO	501 mg/g		
MIL-101(Cr)	Xylenol orange	322–326 mg/g	2012-2014	50
ZIF-67	AB40	55 mg/g		51
Cu-MOF	CR	828.50 mg/g		52
ZJU-24-0.89	MB	902 mg/g		53
HKUST-1	MB	83.6 to 94.4 mg/g		54
Cu <sub>3</sub> (BTC) <sub>2</sub> (H <sub>2</sub> O) <sub>3</sub>	MB	95%		55
HKUST-1@ABS	MB	98.3%		56
HKUST-1@γ-Fe <sub>2</sub> O <sub>3</sub> /C	MB	370.2 mg/g		57
MIL-101(Cr)@PW <sub>11</sub> V	MB	98%	2015	58
	RhB	60 %		
	MO	19%		
MIL-101(Al)-NH <sub>2</sub>	MB	762 mg/g		59
H <sub>6</sub> P <sub>2</sub> W <sub>18</sub> O <sub>62</sub> @Cu <sub>3</sub> (BTC) <sub>2</sub>	MB	298.34 mg/g	2015	60
MIL-100(Fe)	MB	1105 mg/g		61
MIL-125(Ti)	RhB	59.92 mg/g		62
UiO-66	MB	90.59 %	2015	63
	RhB	77.47 %		
	Neutral red	74.84 %		
	MO	38.24 %		
	Acid chrome blue K	31.61 %		
UiO-66-NH <sub>2</sub>	MB	97.27 %	2015	63
	RhB	82.69 %		
	Neutral red	75.85 %		
	MO	28.80 %		
	Acid chrome blue	22.53 %		

	K		
Fe <sub>3</sub> O <sub>4</sub> @MIL-100(Fe)	Methyl red	625 mg/g	64
MIL-53(Al)-NH <sub>2</sub>	MG	38.09 mg/g	65
	MB	45.97 mg/g	
HKUST-1@Fe <sub>3</sub> O <sub>4</sub>	MB	245 mg/g	66
MIL-68(In)	CR	1204 mg/g	67
TMU-1	MB	100 %	68
	MO		
ZIF-8	CR	1250 mg/g	69
Fe <sub>3</sub> O <sub>4</sub> @MIL-100(Fe)	MB	73.8 mg/g	70
	RhB	28.36 mg/g	
MIL-68(Al)	MB	1666.67 mg/g	71
	RhB	1111.11 mg/g	
H <sub>6</sub> P <sub>2</sub> W <sub>18</sub> O <sub>62</sub> /MOF-5	MB	97 %	72
	MO	10%	
	RhB	10%	
MIL-68(In)-NH <sub>2</sub>	RhB	50 %	73
In-MOF@GO	RhB	267 mg/g	73
Fe <sub>3</sub> O <sub>4</sub> /MIL-101	MO	90 %	74
	Xylenol orange		
	Fluorescein sodium		
Fe <sub>3</sub> O <sub>4</sub> /MIL-101(Cr)	Acid red 1	97.9 %	75
	Orange G	97.7 %	
FJI-C2	MB	1323 mg/g	76
HKUST-1@GO@Fe <sub>3</sub> O <sub>4</sub>	MB	1604 mg/g	77
MIL-101(Al)-NH <sub>2</sub>	ICM	135 mg/g	78
	MG	274.4 mg/g	
MIL-101(Fe)@H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	MB	473.7 mg/g	79
	RhB	96 %	
ZJU-71	MB	9.52 mg/g	73
MIL-101(Cr)@GO	AM	111.01 mg/g	80
	CM	77.61 mg/g	
	SY	81.36 mg/g	
HLJU-2	CV	44.55 mg/g	81
	RhB	4789 mg/g	

UiO-66	Safranin T	97 %		78
Ni-MOFs	CR	2046 mg/g		82
TMU-7 (Cd)	CR	97 mg/g		83
$\text{Fe}_3\text{O}_4@\text{MIL}-100(\text{Fe})$	MB	221 mg/g	84	
	RhB	93.5 %		
UiO-66-P	MB	91.1 mg/g		85
UiO-66	MB	24.5 mg/g	86	
	Acid Chrome Blue K	228.6 mg/g		
	MO	172.5 mg/g		
	CR	493.1 mg/g		
MIL-53(Fe)/PAN	MB	70 %		87
HKUST-1@ $\text{Fe}_3\text{O}_4$ -1	MB	118 mg/g		88
HKUST-1@UiO-66	MB	526 mg/g		89
LIFM-WZ-3	CV	713.5 mg/g	2017	90
In-MOF	MB	724.64 mg/g		91
JLU-Liu39	MB	308 mg/g		92
	RhB	16 mg/g		
PCN-222(Zr)	MB	1239 mg/g	93	
	MO	1022 mg/g		
ZIF-8@GO	MG	3300 mg/g		94
HPU-5	RhB	01415 mg/g	95	
	MO	0.185 mg/g		
HPU-6	RhB	0.34 mg/g	96	
	MO	0.42 mg/g		
PCN-124-Cu	CBB	78.7 mg/g		96
<i>N</i> -doped $\text{TiO}_2$ nanoparticles caged in MIL-100 (Fe)	MB	99.1 %	91	
	RhB	93.5 %		
Co-MOFs	CR	4885.20 mg/g	2018	97
UiO-66	MB	69.8 mg/g		98
	MO	83.7 mg/g		
Zn-MOF	MB	326 mg/g		99
	RhB	3.75 mg/g		
	CV	90.77 %		
$\text{Fe}_3\text{O}_4@\text{SiO}_2@\text{UiO}-66-\text{NH}_2$	MB	128 mg/g		100

	MO	130 mg/g		
MgFe <sub>2</sub> O <sub>4</sub> @MOF	RhB	219.78 mg/g	101	
	RhBG	306.75 mg/g		
MOF-199	B41	1257 mg/g	102	
POM@UiO-66	RhB	222.6 mg/g	103	
	MG	190.6 mg/g		
	Orange G	40 mg/g		
MOF-5@Ag <sub>2</sub> O	AO	260.70 mg/g	104	
Fe-MOFs	CR	90.67 %	105	
	RhB	70.90 %		
	Orange II	88.21 %		
Fe <sub>3</sub> O <sub>4</sub> @MIL-100 (Fe)	AO10	100 %	106	
NH <sub>2</sub> -MIL-88B (Fe)	MB	100 %	107	
MIL-125(Ti)-NH <sub>2</sub>	B46	1257 mg/g	102	
	MB	862 mg/g		
Cd-MOF	CV	221 mg/g	108	
Zn-MOF	CV	7.355 mg/g	99	
	RhB	2.977 mg/g		
	MB	6.394 mg/g		
LIFM-WZ-3	MB	983 mg/g	108	
MIL-53(Al)-NH <sub>2</sub> @Fe <sub>3</sub> O <sub>4</sub> @CA, CA: citric acid	MB	325.62 mg/g	109	
	MG	329.61 mg/g		
MIL-100(Fe)	MB	96 %	105	
	RhB	99%		
	CBB	100%		
MIL-100 (Al)	MB	97%	105	
	RhB	34 %		
	CBB	100 %		
MIL-101(Cr)-NH <sub>2</sub> @Fe <sub>3</sub> O <sub>4</sub>	MB	370.3 mg/g	107	
MIL-101(Cr)@P <sub>2</sub> W <sub>18</sub> O <sub>62</sub> @Fe <sub>3</sub> O <sub>4</sub>	MB	200 mg/g	110	
	RhB	164 mg/g		
	MO	60%		
UiO-66@Ce	MB	145.3 mg/g	111	
	ACK	245.8 mg/g		
	MO	639.6 mg/g		

ESF@ZIF-8	MG	127.1 mg/g	112
	RhB	19 mg/g	
ESF@ZIF-67	MG	840.2 mg/g	113
MIL-125-NH <sub>2</sub> @Fe <sub>3</sub> O <sub>4</sub>	MG	457 mg/g	111
UiO-66-0.75-(COOH) <sub>2</sub>	RhB	94 mg/g	114
Al-SA MOF	Acid Black 1	199.1 mg/g	115
UiO-66	ARS	400 mg/g	116
IPM-MOF-201	ARS	50%	117
	MO	50%	
UiO-66@CNT, CNT: carbon nanotubes7	AY17	89.39 mg/g	118
	AO7	80.63 mg/g	
MIL-100(Fe)@GO	MB	1231 mg/g	119
	MO	1189 mg/g	
Cd-MOF	MO	93%	120
Zn-MOF	MO	100%	
Ni-MOF	CR	276.7 mg/g	121
Zn/Co ZIF	MB	92%	122
BUT-29	MB	1119 mg/g	123
SCNU-ZI-Cl	MO	285 mg/g	
	Acid orange A	180 mg/g	
	CR	585 mg/g	
	MB	262 mg/g	
Cd-ZIF	MG	395.87 mg/g	124
CoOF	Acid red 18	44.26 mg/g	125
UiO-66	Acid blue 92	73 %	126
	Direct red 2B	76 %	
	Maxilon blue M2G	46.6 %	
BUT-29	CV	832 mg/g	122
MOF-235	CR	1250 mg/g	127
	Lemon yellow	250 mg/g	
Mn-MOF	CV	938 mg/g	128
ZIF-67	Acid orange 7	738 mg/g	129
	CR	3899.8 mg/g	
NENU@0.03GO	B46	243 mg/g	131
PCN-222(Fe)	BG	854 mg/g	132

2019

	CV	812 mg/g		
	AB80	85%		
	AR	417 mg/g		
MIL-68(Al)@PVDF, PVDF: polyvinylidene fluoride	MB	74.35 $\mu\text{g cm}^{-2}$		133
MIL-140X, (X = F, -2COOH, -2NO <sub>2</sub> )	MB	100%		134
MIL-140X, (X = B, -2COOH, -2NO <sub>2</sub> , C, E, F, -SO <sub>3</sub> H)	NB	100 %		
SCNU-Z1-Cl	MB	262 mg/g		135
	RhB	130 mg/g		
	AO7	180 mg/g		
	MO	285 mg/g		
MIL-101(Fe)@dopa@Fe <sub>3</sub> O <sub>4</sub>	MG	833.33 mg/g		136
UiO-66@wood	RhB	690 mg/g		137
MIL-101(Cr)	AB92	185 mg/g		138
	Red 80	227 mg/g		
MIL-88A(Fe)@Fe <sub>3</sub> O <sub>4</sub>	BPB	141.5 mg/g		139
UiO-66-NH <sub>2</sub> @Tb-CP	CBB	397.3 mg/g		140
Fe-BDC	MB	94.74%		141
TFMOF(Zr)	CR	252.25 mg/g		142
Ce(III)-doped UiO-67	CR	799.6 mg/g		143
	MB	398.9 mg/g		
	MO	401.2 gm/g		
In-TATAB	CR	299 mg/g		144
	Acid chrome blue K	343 mg/g		
	Acid red 26	259 mg/g		
	Direct black 38	242 mg/g		
	Orange II	217 mg/g		
UiO-66/PGP	CR	99.6 %		145
	MB	100 %		
	MO	94.8 %		
	RhB	95.5 %		
Printed MOF/CA-GE	MB	99.8 %		143
GO-Cu-MOF	MB	262 mg/g		146
CuBDC	MB	41.01 mg/g		123

2020

MOF-NiZn	MB	398.9 mg/g		147
ABim-Zn-MOF	MB	174.64 mg/g		148
	Chicago sky blue	144.26 mg/g		
ZIF-8	MG	1000 mg/g		149
AC@MIL-101(Cr)	Direct Red 31	99.4 %		150
	Acid Blue 92	92.9 %		
MOF-2(Cd)	MB	99.90 %		151
[Ni <sub>2</sub> F <sub>2</sub> (4,4' bipy) <sub>2</sub> (H <sub>2</sub> O) <sub>2</sub> ](VO <sub>3</sub> ) <sub>2</sub> ·8H <sub>2</sub> O	CR	242.1 mg/g		152
MOF-5/COF (M <sub>5</sub> C)	AO	17.95 mg/g		153
	RhB	16.18 mg/g		
PFC-24-Zr	MO	12 mg/g		154
HKUST (Cu <sub>3</sub> (BTC) <sub>2</sub> )	MB	833.33 mg/g		155
{[Cu(bipy) <sub>1.5</sub> (H <sub>2</sub> - pdm)}·2NO <sub>3</sub> ·H <sub>2</sub> O] <sub>n</sub> (Cu-MOF-1)	MB	98.23 %		156
MOF-5@GO	RhB	151.62 mg/g	2021	157
PCN-224	MB	1015.7 mg/g		158
MIP-202 Zr-MOF	MB	36.071 mg/g		159
	Direct red 81	19.012 mg/g		
{[Ag(3-(4H-1,2,4-triazol-4-yl)-5-(trifluoromethyl)pyridine)](BF <sub>4</sub> ) <sub>n</sub>	MB	409		160
	CR	264		

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