Chapter

Adsorption Processes in the Removal of Organic Dyes from Wastewaters: Very Recent Developments

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Abstract

The problem of the treatment of contaminated wastewaters is of the upmost worldwide interest. This contamination occurs via the presence of inorganic or organic contaminants of different nature in relation with the industry they come from. In the case of organic dyes, their environmental impact, and thus, their toxicity come from the air (releasing of dust and particulate matter), solid (scrap of textile fabrics, sludges), though the great pollution, caused from dyes, comes from the discharge of untreated effluents into waters, contributing to increase the level of BOD and COD in these liquid streams; this discharge is normally accompanied by water coloration, which low the water quality, and caused a secondary issue in the wastewater treatment. Among separation technologies, adsorption processing is one of the most popular, due to its versatility, easiness of work, and possibility of scaling-up in the eve of the treatment of large wastewater volumes. Within a miriade of potential adsorbents for the removal of organic dyes, this work presented the most recent advances in the topic.

Keywords: organic dyes, adsorbents, wastewaters, water quality

1. Introduction

Nowadays, one of the most important environmental issues is related to water scarcity, water contamination and water quality. In many cases, water contamination is due to pollution by means of the presence of inorganic and/or organic compounds; particularly, the presence of organic dyes in waters, or in general, in effluents must be avoided since they are toxic (mutagenic, i.e. Azure B or Disperse Red 1, and carcinogenic, i.e. Basic Red 9 and Crystal Violet) against life and humans. Under the word "dye," is included substances which.

produced a color on materials of diverse nature, being the textile industry (54%) which produced the highest quantity of dyes-bearing effluents, followed by the dyeing industry (21%), paper and pulp industry (10%), tannery and paint industry (8%) and the own dye manufacturing industry (7%).

These compounds can be classified on structural bases, i.e. basic, acid, azo, disperse, anthraquinone or metal complex. The international standard of dye effluent discharge into the environment considered the next tolerable limits [1]:

i) biological oxygen demand: under 30 mg/L, ii) chemical oxygen demand: below 50 mg/L, iii). color: under 1 ppm; .iv) pH value in the 6–9 range, v) suspended solids: less than 20 mg/L, vi) temperature: below 42° C, and vii) toxic pollutants: completely avoid. However and despite all prohibitions, still today there is an important number of illegal dyes (i.e. Solvent Yellow 4, a member of the azolipophilic compounds) used in the textile industry. There are a number of useful technologies to eliminate these organic dyes from waters-effluents [2, 3], and among them, adsorption processes gained a paramount interest due to their manipulation easiness, scaling up, and the possibility of using a countless number of potential adsorbents.

The present work, and due to space constrains, reviewed the most recent results (January–April 2020) about the removal of organic dyes by adsorption processes.

2. Adsorbents and organic dyes: the adsorption results

Being a plant practically present around the world, cactus, and due to their chemical composition and biological and nutritional properties, find various applications, being one of them, their use as adsorbents for toxic metals and organic dyes [4]. Thus, different cactus parts: fruit seeds, peel, clodades, among others, had been investigated in the topic of the removal of organic dyes from waters. Often, these bioadsorbents were subject, before use, to some type of treatment, such as heat treatment, chemical treatment, sun-dehydration. Some results about the adsorption capacity of these adsorbents are given in **Table 1**.

These cactus-based adsorbents presented maximum adsorption capacities in close relation with those derived from other materials of different origins (**Table 2**).

Others bioadsorbents used in the removal of organic dyes (maximum capacity) are: leaves of *Lawsonia sp.* (malachite green (no data)) [5], *Platanus orientalis* (rhodamine B (557 mg/g), methyl orange (327 mg/g)) [6], *α-chitin* (methylene blue

Adsorbent	**Dyes (maximum capacity, mg/g)**
Cladodes of *Tacinga palmadora*	Crystal violet (229)
Fruit peels of *O. ficus indica*	Crystal violet (312). Metyhylene blue (416)
Natural cladodes of *O. ficus indica*	Methylene blue (3.5)
Palm cactus	Crystal violet (173-220)
O. ficus indica	Basic Blue 9 (35-278)

Table 1.
Use of cactus in dyes adsorption.

Adsorbent	**Dyes (maximum capacity, mg/g)**
Leaves of *Tacinga Palmadora*	Crystal violet (229)
Tarjuna sawdust waste	Crystal violet (46)
Rice husk	Crystal violet (293)
Cactus cladodes (activated carbon)	Methylene Blue (750)
Walnut shell	Methylene Blue (315)
Wood apple rind	Methylene Blue (40)

Table 2.
Crystal violet and methylene blue adsorption onto different adsorbents.

(95 mg/g)) [7], pods of *Clitoria fairchildiana* (rhodamine 66 (571 mg/g)) [8], and diatomite waste (methylene blue (25 mg/g), acid orange (35 mg/g)) [9].

Nanomaterial-based adsorbents are another type of materials that, due to their properties and adsorption capacities, have applications in the removal or organic dyes from waters. Including in these nanomaterials, carbon nanotubes (CNTs), grapheme sheets (GS), and metal oxides (MO) are found [10].

Carbon nanotubes presented a sp^2 allotropic carbon of graphite structure in cylindrical or tube shaped sheets. Based on the number of these sheets presented in the adsorbent, CNTs can be found as single-walled carbon nanotubes (SWCNTs) or multi-walled carbon nanotubes (MWCNTs). Typically, SWCNTs presented a diameter in the 0.4–10 nm range, whereas MWCNTs have a diameter in the 10–100 nm range and spacing between sheets in the 0.34–0.38 nm range.

The adsorptive effectiveness of these carbon nanotubes can be improved by functionalizing them or modifying some of their characteristics: specific area, charge density, porosity, and hydrophilicity. These modifications can be done by acid/oxidant treatment, combination with metals/MO, and grafting special functional groups, such as polymer and surfactants.

These carbon materials presented four characteristics adsorption sites in their surfaces. Thus, the adsorption process occurred at i) the external, and/or ii) internal surface of the nanotubes, iii) the interstitial pathways between individual nanotubes sheet, and iv) the external groove sites. In the case of MWCNTs, the space between the sheets can also be used to adsorb organic dyes.

Organic dyes uptake onto these nanotubes responded very often to the Langmuir and Freundlich isotherm models, and the adsorption kinetics is best fitted to the pseudo-second-order kinetic model.

Graphene is formed by a single layer of sp^2 allotropic carbon atoms arranged in a two-dimensional hexagonal honeycomb lattice structure.

Similarly to carbon nanotubes, single-layer (SLG) or multiple-layer graphene (MLG) materials can be yielded in a 2D structure from a graphite-based material. Other derived materials, such as graphene oxide (GO) and reduced graphene oxide (RGO), with enhanced adsorptive characteristics, can be produced by chemical oxidation of graphite and reduction of grapheme oxide, respectively. These two last materials, presented better adsorption characteristics than the above grapheme materials.

A graphene/wastepaper composite [11], had been used in the removal of methylene blue and Congo red from waters, with maximum capacities of 58 and 90 mg/g, respectively. Nanoribbons of graphene were used in the adsorption of methylene blue and orange II dyes [12], in this case the maximum capacities, presented for the adsorbent, were of 280 and 265 mg/g, respectively. Others graphene-based materials had been recently used in the adsorption of crystal violet (69 mg/g) [13], rhodamine B (963 mg/g) [14], etc.

It was found that for selected organic dyes, graphene-based adsorbent had an average 2–5 times higher dye adsorption capacity than carbon nanotubes and metal oxides.

MOs adsorbents applied for the treatment of organic dyes-bearing waters included, iron oxide (Fe_3O_4), zinc oxide (ZnO), titanium dioxide (TiO_2), magnesium oxide (MgO), alumina oxide (Al_2O_3), and zirconium oxide (ZrO_2). Among them, iron oxide nanoparticles presented good properties i.e. high specific surface area, to adsorb organic dyes, and they are magnetic. This characteristic facilitated the dispersion of the nanoparticles in the aqueous solution, and their removal from it, when an external magnetic field is applied [15].

Other investigations described the use of nanohybrids of Cu_xO/Fe_2O_3/MoC as materials used in the adsorption of reactive red 195A and reactive yellow

84 (maximum capacities 435 and 278 mg/g, respectively) [16], also the use of Cr-doped ZnO in the adsorption of methyl orange (19 mg/g) and methylene blue (41 mg/g), the adsorption of methyl orange (833 mg/g) by a magnetic composite [17], and Fe_3O_4/PPy composites (eosin Y, methyl orange and brilliant green: 212, 149 and 264 mg/g, respectively) [18]. It was described in the literature [19], the usefulness of Ag_2O as adsorbent of Congo red (181 mg/g), acid orange 7 (125 mg/g) and amido black 10B (83 mg/g), however this investigation, as many others, did not give any information about the desorption step.

The list of nanoparticles or nanomaterials used to remove organic dyes from waters seemed not to end [20, 21], considering that a series of materials such as biomass, clay minerals, different wastes, etc., when modified with magnetic nanoparticles enhanced their respective adsorption capacity towards organic dyes, because they increased their surface area and porosity and with the addition of the magnetic nanoparticles, they adopt a new property, as is the magnetic character, which improve their separation from the treated water. Moreover, by the addition of adequate functional groups to these nanoadsorbents, basically on their surface, they further improve their respective capacities on the treatment of waters contaminated with organic dyes. Not being exhaustive, **Table 3** summarized some of the results encountered in this field.

Polyaniline, polypyrrole and other conducting polymers, had been also used in the removal of organic dyes from waters. In fact, these polymers reacted with organic dyes due to the similarity of the conjugated molecular structures, of both types of compounds, which enhanced the reactivity between them. The use of these organic dyes as templates in the conducting polymer synthesis may affect both the conductivity and morphology control of the end product, specially in the case of polypyrrole.

It was described [22], how conducting polymers and organic dyes reacted:

i. π-π interaction between the aromatic rings,

ii. electrostatic ionic interactions,

iii. hydrogen bonding, and.

iv. hydrophobic interactions.

Adsorbent	**Organic dye (removal efficiency, mg/g)**
Magnetic baker's yeast biomass	methyl violet (61)
Modified multi-walled carbon nanotubes	alizarin yellow R (45)
Magnetic peach gum bead	methylene blue (232)
Magnetic polyacrylamide micropheres	methylene blue (1990)
Malachite@clay nanocomposite	Congo red (238)
ZnO nanorods loaded activated carbon	brilliant green (58)
Sorel's cement nanoparticles	methyl orange (21)
Alkaline treated timber sawdust	methylene blue (694)
Ultrathin $MoSe_2$ nanosheets	rhodamine B (133)
Cellulose nanocrystal-reinforced keratin	reactive black 5 (1201)

Table 3.
Adsorbents and adsorbed organic dyes.

Some dyes used in the preparation of polyaniline are: methyl orange and green GS, whereas in the case of polypyrrole, the list included both mentioned above and Congo red, Thymol blue, cresol red and rhodamine B among others.

In the conducting role, polyaniline and polypyrrole are polycations, thus, it is expected that they normally reacted with anionic dyes, however, experimentally it was found that both cationic and anionic dyes reacted with these conducting polymers; it seemed that electrostatic ionic interactions are not the most important factor to explain this reactivity.

As it is mentioned above, these conducting polymers had been used, alone or in the composite forms, in the removal of numerous organic dyes from waters, and basically their success is due to that they have a relative low production cost.

In the case of polyaniline, the list of investigations related to the removal of organic dyes from waters included: Congo red, eosin Y, rose bengal, indigo carmine, etc. Polyaniline composites are classified along the non-conducting component, and included composites containing (in various forms): aluminum, bismuth, carbon, iron, silicium, natural polymers (cellulose), synthetic polymers, etc.

Pristine polypyrrole had been investigated in the removal of methyl orange, methylene blue, etc. Polypyrrole composites again contained in various forms: aluminum, carbon, titanium, zinc, etc.

Table 4 presented a comprehensive (but not exhaustive) list of organic dyes adsorbed by polyaniline and polypyrrole composites.

The use of polyaniline-related materials, such as aniline oligomers and copolymers, polyaniline chemically modified, etc., had a further interest in the removal of organic dyes from waters. The use of polypyrrole-related materials in this environmental role has a significant minor development.

Due to their environmentally friendly and availability, polymer derivatives based on polysaccharides are also of interest in the removal of organic dyes from contaminated waters. Thus, a variety of modified polysaccharides, i.e. chitosan, starch, dextran, cellulose, have been investigated as adsorbents in this role; however, and despite some of pullulan characteristics such as: high solubility and flexibility of the backbone when compared with other polysaccharides, they are not amply used in waters purification [27].

	Composite	Organic dye (maximum capacity, mg/g)	Ref.
Polyaniline	TiO_2	Methyl orange (62)	[23]
	TiO_2	Methylene blue (83) Congo red (20) Crystal violet (50) Rhodamine 6G (57)	[24]
	SiO_2	Methylene blue (55) Congo red (33) Crystal violet (25) Rhodamine 6G (2)	[24]
	resin	Methylene blue (40)[a] Methylene blue (28)[b]	[25]
	PSMA	Methyl orange (148)	[26]
Polypyrrole	Fe_3O_4	Eosin Y (712) Methyl orange (149) Brilliant green (264)	[18]

[a] *Mixed solutions of Cr(VI) and methylene blue.*
[b] *Solutions containing only methylene blue.*

Table 4.
Organic dyes adsorbed onto polyaniline and polypyrrole composites.

Pullulan, having a chemical formula $(C_6H_{10}O_5)_n$, is a linear, non-ionicpolysaccharide consisting of maltotrioseunits:α-(1 → 6)-linked(1 → 4)-α-d-triglucosides. The known pullulan derivatives are: i) soluble ionic pullulan derivatives: this type of compounds can be synthesized by chemical modification of the polysaccharide. It can include i.i) various content and length of grafted chains, and i.ii) various content of tertiary amine groups, ii) pullulan microspheres: they can be formed by suspension cross-linking of the previously grafted pullulan with cationic moieties (P-g-pAPTAC); iii) nonionic thermosensitive pullulan copolymer: it was prepared by graft-polymerization of p(N-isopropylacrylamide) onto the pullulan. Here, cerium(IV) was used as initiator. The resulted thermosensitive material has the (P-g-pNIPAAm) acronysm; iv) nonionic pullulan-graft-polyacrylamide hydrogel: this pullulan was synthesized by free radical polymerization in presence of a cross-linking agent and calcium carbonate.

Pullulan derivatives showed high removal efficiency of organic dyes contaminants, i.e. P-g-APTAC microspheres were used in the adsorption of azocarmine B (maximum capacity: 114 mg/g), acid orange 7B (65 mg/g) and methyl orange (55 mg/g); pullulan-graft-polycrylamide hydrogel was used in the adsorption of methylene blue and reactive blue with maximum capacities (70° C) of 399 and 356 mg/g, respectively.

The properties of metal–organic frameworks (MOFs), made of them interesting materials for the removal of organic dyes from waters. Some of these properties are: thermal stability, high surface area and porosity, nanosized cavities, etc. The metal centers of these materials provided additional coordination locations aimed to fixing organic dyes, whereas one step ahead in the practical use of these MOFs is provided them with magnetic properties *via* the incorporation of magnetic materials within the framework. Some examples of the use of these materials are given in **Table 5**.

Similarly to the number of materials used to adsorb organic dyes, the number of these chemical compounds investigated to be adsorbed, seemed to be countless. Besides all the mentioned along this work, below, it is summarized further investigations and results (maximum capacities, mg/g):

i. methylene blue: modified Cs-ZnS (502) [33], Cr-doped ZnO nanorods (41) [34], attapulgite derivative (115) [35], mesoporous Zr-based polymer (60) [36], diatomite waste (32) [37], iron-carbon nanosheets (185) [38], graphene oxide derivative (1370) [39], zeolite/CeO_2 nanocomposite (2.5) [40], pyridine derivative (175) [41], cellulose nanocomposite (2067) [42], cellulose/carbon aerogel (1179) [43], MgO modified biochar (475) [44], MoS_2/WO_3 (228) [45],

ii. methyl orange: Cr-doped ZnO nanorods (16) [34], TOCN/CGG hydrogel (134) [46],

Adsorbent	Organic dye (maximum adsorption, mg/g)	Reference
Cd-MOF	Congo red (192)	[28]
Zn-MOF	methylene blue (116)	[29]
HSO_3-MOF	methylene blue(833)	[30]
LDH-MOF	orange II (1173)	[31]
aramid nanofibrils-MOF	methyl violet (114)	[32]

Table 5.
MOFs and adsorption of organic dyes.

iii. Congo red: iron-carbon nanosheets (532) [38], $Ba_5Si_8O_{21}$ microspheres (1239) [47], cellulose/carbon aerogel (585) [43], MIL-100(Fe) (1791) [48],

iv. bromophenol blue: *Hermetia illucens* larvae (571) [49],

v. acid chrome blue K: MIL-100(Fe) (926) [50],

vi. crystal violet: attapulgite derivative (69) [37], vii) thioflavin T: TOCN/CGG hydrogel (430) [46],

vii. methyl blue: nitrogen-doped carbon derivative (1054) [50],

viii. eriochrome black T: carbon derivative (166) [51],

ix. rhodamine B: WS_2/WO_3 (237) [52],

x. reactive yellow: diatomite waste (33) [37],

xi. indigo carmine: N and S organic framework (547) [53],

xii. direct red 31: nanoporous composites (526) [54],

xiii. acid blue 92: nanoporous composites (556) [54].

3. Conclusions

During the first quarter of 2020 year, an important number of dye adsorption procedures have been proposed in a series of published papers, all of them, claiming successful dye removal. However, in the opinion of the authors of the present review some criticism to the papers must be accounted for:

i. comparison of the maximum dye capacity presented different numbers, see **Table 6**. Thus, the successfulness of some of these adsorbents can be questioned if compared with the figure presented by other materials,

ii. near 52% of the published papers included some data about the desorption step, that is, the other 48% of the published papers are uncompleted in terms of the overall adsorption–desorption process. Authors must aware that as important is the adsorption step than the desorption one, and in all the cases in which the latter is included, the authors do not give any comment about what to do with the now dye-bearing desorption solution. In the manuscripts, organic dyes simply go to one solution (the feed one) to another solution (the desorption one) (except in the case of degradation occurs),

iii. all the reviewed manuscripts lacked the investigation of a sometimes key variables on adsorption processes and in batch mode, as the stirring speed and how the phases mixed can be. This is because, with the correct stirring speed, the.

iv. thickness of the aqueous film layer reached a minimum and the adsorption reached a maximum. Together with the above, the form in which the phases are mixed also ensured the best contact, and thus the best solute transfer, between the phases involved in the process.

Organic dye	Adsorbent	Maximum capacity (mg/g)	Ref.
Methylene blue	functionalized-organic polymer	2740	[55]
	cellulose hydrogel	756	[56]
	modified grapheme oxide	257	[57]
	functionalized lignosulfate	63	[58]
Methyl orange	NiAlTi	1250	[59]
	MIL-101-NH_2	462	[60]
	polyaniline composite	148	[26]
Crystal violet	fly ash	433	[61]
	polyaniline-metal oxides	50	[24]
Congo red	MIL-100(Fe)	1791	[31]
	cellulose/carbon aerogels	585	[43]
	polyaniline-metal oxides	33	[24]
Rhodamine B	Ni-graphene composite	963	[14]
	MoS_2 nanoflowers	365	[62]
	polyaniline-metal oxides	20	[24]

Table 6.
Difference in organic dyes adsorption capacity of various adsorbents.

It is needed to mention here, that in Refs. [63, 64] a heavy scientific fault was detected: the authors of the manuscripts investigated, besides the adsorption of organic dyes, the adsorption of Cr^{6+}, written as such, when this element in the VI oxidation state, never exists as a cation in aqueous solutions. This fault also is responsibility of the corresponding reviewers and of the Editors.

In practical terms, the real bottlenecks in the usefulness of these adsorbents are:

i. lack of information about the desorption step,

ii. evident loss of adsorption capacity under continuous adsorption–desorption cycles,

iii. adsorbent cost and possibility of production at large scale to scale-up the adsorption–desorption process,

iv. environmental friendship of the own adsorbent,

v. lack of information on the use of the purification of real waters bearing organic dyes.

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References

[1] Katheresan V, Kansedo J, Lau SY. Efficiency of various recent wastewater dye removal methods: A review. Journal of Environmental Chemical Engineering. 2018;6:4676-4697. DOI: 10.1016/j.jece.2018.06.060

[2] Ismail M, Akthar K, Khan, MI, Kamal T, Khan A, Asiri AM, Seo J, Khan SB. Pollution, toxicity and carcinogenicity of organic dyes and their catalytic bio-remediation. Current Pharmaceutical Design. 2019; 25:3653-3671. DOI: 10.2174/1381612825666191021142026

[3] Lellis B, Favaro-Polonio CZ, Pamphile JA, Polonio JC. Effect of textile dyes on health and the environment and bioremediation potential of living organisms. Biotechnology Research & Innovation. 2019. 3:275-290. DOI: 10.1016/j.biori.2019.09.001

[4] Amari A, Alalwan B, Eldirderi MM, Mnif W, Ben Rebah F, Cactus material-based adsorbents for the removal of heavy metals and dyes: a review. Materials Research Express. 2020;7:012002. DOI: 10.1088/2053-1591/ab5f32

[5] Sharmila S, Dinesh M, Kowsalya E, Kamalambigeswari R, Rebecca LJ. Biosorption of dye using Lawsonia sp. as adsorbent. Research Journal of Pharmacy and Technology. 2020;13:1651-1654. DOI: 10.5958/0974-360X.2020.00299.1

[6] Jiang X, Sun P, Xu L, Xue Y, Zhang H, Zhu W. Platanus orientalis leaves based hierarchical porous carbon microspheres as high efficiency adsorbents for organic dyes removal. Chinese Journal of Chemical Engineering. 2020;28:254-265. DOI: 10.1016/j.cjche.2019.03.030

[7] Abou-Zeid RE, Salama A, Al-Ahmed ZA, Awwad NS, Youssef MA. Carboxylated cellulose nanofibers as a novel efficient adsorbent for water purification. Cellulose Chemistry and Technology. 2020;54:237-245. DOI: 10.35812/CELLULOSECHEMTECHNOL.2020.54.25

[8] da Silva AMB, Serrão NO, de Gusmão Celestino G, Takeno ML, Antunes NTB, Iglauer S, Manzato L, de Freitas FA, Maia, PJS. Removal of rhodamine 6G from synthetic effluents using *Clitoria fairchildiana* pods as low-cost biosorbent. Environmental Science and Pollution Research. 2020;27:2868-2880. DOI: 10-1007/s11356-019-07114-6

[9] Liu N, Wu Y, Sha H. Magnesium oxide modified diatomite waste as an efficient adsorbent for organic dye removal: adsorption performance and mechanism studies. Separation Science and Technology. 2020;55:234-246. DOI: 10.1080/01496395.2019.1577456

[10] Awad AM, Jalab R, Benamor A, Nasser MS, Ba-Abbad MM, El-Naas M, Mohammad AW. Adsorption of organic pollutants by nanomaterial-based adsorbents: An overview. Journal of Molecular Liquids. 2020;301:112335. DOI: 10.1016/j.molliq.2019.112335

[11] Feng C, Ren P, Li Z, Tan W, Zhang H, Jin Y, Ren F. Graphene/waste-newspaper cellulose composite aerogels with selective adsorption of organic dyes: Preparation, characterization, and adsorption mechanism. New Journal of Chemistry. 2020;44:2256-2267. DOI: 10.1039/c9nj05346h

[12] Gao Y, Liang X, Han S, Wu L, Zhang G, Qin C, Bao S, Wang Q, Qi L, Xiao L. High-efficiency adsorption for both cationic and anionic dyes using graphene nanoribbons formed by atomic-hydrogen induced single-walled carbon nanotube carpets. Carbon Letters. 2020;30:123-132. DOI: 10.1007/s42823-019-00089-x

[13] Czepa W, Pakulski D, Witomska S, Patroniak V, Ciesielski A, Samorì P. Graphene oxide-mesoporous SiO2 hybrid composite for fast and efficient removal of organic cationic contaminants. Carbon. 2020;158:193-201. DOI: 10.1016/j.carbon.2019.11.091

[14] Tao X, Wang S, Li Z. Ultrasound-assisted bottom-up synthesis of Ni-graphene hybrid composites and their excellent rhodamine B removal properties. Journal of Environmental Management. 2020;255:109834. DOI: 10.1016/j.jenvman.2019.109834

[15] Luyen NT, Linh HX, Huy TQ. Preparation of Rice Husk Biochar-Based Magnetic Nanocomposite for Effective Removal of Crystal Violet. Journal of Electronic Materials. 2020;49:1142-1149. DOI: 10.1007/s11664-019-07798-z

[16] Bastami TR, Khaknahad S, Malekshahi M. Sonochemical versus reverse-precipitation synthesis of CuxO/Fe2O3/MoC nano-hybrid: removal of reactive dyes and evaluation of smartphone for colorimetric detection of organic dyes in water media. Environmental Science and Pollution Research. 2020; 27:9364-9381. DOI: 10.1007/s11356-019-07368-0

[17] Natarajan S, Anitha V, Gajula GP, Thiagarajan V. Synthesis and Characterization of Magnetic Superadsorbent Fe3O4-PEG-Mg-Al-LDH Nanocomposites for Ultrahigh Removal of Organic Dyes. ACS Omega. 2020;5:3181-3193. DOI: 10.1021/acsomega.9b03153

[18] Zhang M, Yu Z, Yu H. Adsorption of Eosin Y, methyl orange and brilliant green from aqueous solution using ferroferric oxide/polypyrrole magnetic composite. Polymer Bulletin. 2020;77:1049-1066. DOI: 10.1007/s00289-019-02792-1

[19] Wang Y, Bi N, Zhang H, Tian W, Zhang T, Wu P, Jiang W. Visible-light-driven photocatalysis-assisted adsorption of azo dyes using Ag2O. Colloids and Surfaces A: Physicochemical and Engineering Aspects. 2020;585:124105. DOI: 10.1016/j.colsurfa.2019.124105

[20] Tara N, Siddiqui SI, Rathi G, Chaudhry SA, Inamuddin, Asiri AM. Nano-engineered adsorbent for the removal of dyes from water: A review. Current Analytical Chemistry. 2020;16:14-40. DOI: 10.2174/1573411015666190117124344

[21] Lu F, Astruc D. Nanocatalysts and other nanomaterials for water remediation from organic pollutants. Coordination Chemistry Reviews. 2020;408:213180. DOI: 10.1016/j.ccr.2020.213180

[22] Stejskal J. Interaction of conducting polymers, polyaniline and polypyrrole, with organic dyes: polymer morphology control, dye adsorption and photocatalytic decomposition. Chemical Papers. 2020;74:1-54. DOI: 10.1007/s11696-019-00982-9

[23] Cionti C, Della Pinal C, Meroni D, Falletta E, Ardizzone S. Photocatalytic and oxidative synthetic pathways for highly efficient PANI- TiO_2 nanocomposites as organic and inorganic pollutant sorbents. Nanomaterials. 2020;10:441. DOI:10.3390/nano10030441

[24] Maruthapandi M, Eswaran L, Luong JHT, Gedanken A. Sonochemical preparation of polyaniline@TiO2 and polyaniline@SiO2 for the removal of anionic and cationic dyes. Ultrasonics Sonochemistry. 2020;62:104864. DOI: 10.1016/j.ultsonch.2019.104864

[25] Ding J, Pan Y, Li L, Liu H, Zhang Q, Gao G, Pan B. Synergetic adsorption and electrochemical classified recycling of Cr(VI) and dyes in synthetic dyeing wastewater. Chemical Engineering Journal. 2020;384:123232. DOI: 10.1016/j.cej.2019.123232

[26] Liu Y, Song L, Du L, Gao P, Liang N, Wu S, Minami T, Zang L, Yu C, Xu X. Preparation of polyaniline/emulsion microsphere composite for efficient adsorption of organic dyes. Polymers. 2020;12:167. DOI: 10.3390/polym12010167

[27] Ghimici L, Constantin M. A review of the use of pullulan derivatives in wastewater purification. Reactive and Functional Polymers. 2020;149:104510. DOI: 10.1016/j.reactfunctpolym.2020.104510

[28] Ghomshehzadeh SG, Nobakht V, Pourreza N, Mercandelli P, Carlucci L. A new pillared Cd-organic framework as adsorbent of organic dyes and as precursor of CdO nanoparticles. Polyhedron. 2020;176:114265. DOI: 10.1016/j.poly.2019.114265

[29] Hong YS, Sun SL, Sun Q, Gao EQ, Ye M. Tuning adsorption capacity through ligand pre-modification in functionalized Zn-MOF analogues. Materials Chemistry and Physics. 2020;243:122601. DOI: 10.1016/j.matchemphys.2019.122601

[30] Saghian M, Dehghanpour S, Sharbatdaran M. Unique and efficient adsorbents for highly selective and reverse adsorption and separation of dyes: Via the introduction of SO3H functional groups into a metal-organic framework. RSC Advances. 2020;10:9369-9377. DOI: 10.1039/c9ra10840h

[31] Soltani R, Marjani A, Shirazian S. A hierarchical LDH/MOF nanocomposite: Single, simultaneous and consecutive adsorption of a reactive dye and Cr(VI). Dalton Transactions. 2020;49:5323-5335. DOI: 10.1039/d0dt00680g

[32] Zong L, Yang Y, Yang H, Wu X. Shapeable aerogels of metal-organic-frameworks supported by aramid nanofibrils for efficient adsorption and interception. ACS Applied Materials and Interfaces. 2020;12:7295-7301. DOI: 10.1021/acsami.9b22466

[33] Ali, H. Ternary system from mesoporous CdS–ZnS modified with polyaniline for removal of cationic and anionic dyes. Research on Chemical Intermediates. 2020;46:571-592. DOI: 10.1007/s11164-019-03968-0

[34] Chen J, Xiong Y, Duan M, Li X, Li J, Fang S, Qin S, Zhang R. Insight into the Synergistic Effect of Adsorption-Photocatalysis for the Removal of Organic Dye Pollutants by Cr-Doped ZnO. Langmuir. 2020;36:520-533. DOI: 10.1021/acs.langmuir.9b02879

[35] Cui M, Mu P, Shen Y, Zhu G, Luo L, Li J. Three-dimensional attapulgite with sandwich-like architecture used for multifunctional water remediation. Separation and Purification Technology. 2020;235:116210. DOI: 10.1016/j.seppur.2019.116210

[36] Liang C, Ren J, El Hankari S, Huo J. Aqueous Synthesis of a Mesoporous Zr-Based Coordination Polymer for Removal of Organic Dyes. ACS Omega. 2020;5:603-609. DOI: 10.1021/acsomega.9b03192

[37] Ma T, Wu Y, Liu N, Wu Y. Hydrolyzed polyacrylamide modified diatomite waste as a novel adsorbent for organic dye removal: Adsorption performance and mechanism studies. Polyhedron. 2020;175:114227. DOI: 10.1016/j.poly.2019.114227

[38] Manippady SR, Singh A, Basavaraja BM, Samal AK, Srivastava S, Saxena M. Iron-Carbon Hybrid Magnetic Nanosheets for Adsorption-Removal of Organic Dyes and 4-Nitrophenol from Aqueous Solution. ACS Applied Nano Materials. 2020;3:1571-1582. DOI: 10.1021/acsanm.9b02348

[39] Neskoromnaya EA, Burakov AE, Melezhik AV, Babkin AV, Burakova IV,

Kurnosov DA, Tkachev AG. Synthesis and evaluation of adsorption properties of reduced grapheme oxide hydro- and aerogels modified by iron oxide nanoparticles. Inorganic Materials: Applied Research. 2020;11:467-475. DOI: 10.1134/S2075113320020264

[40] Nyankson E, Adjasoo J, Efavi JK, Yaya A, Manu G, Kingsford A, Abrokwah RY. Synthesis and kinetic adsorption characteristics of Zeolite/CeO2 nanocomposite. Scientific African. 2020;7:e00257. DOI: 10.1016/j.sciaf.2019.e00257

[41] Rajendran N, Samuel J, Amin MO, Al-Hetlani E, Makhseed S. Carbazole-tagged pyridinic microporous network polymer for CO2 storage and organic dye removal from aqueous solution. Environmental Research. 2020;182:109001. DOI: 10.1016/j.envres.2019.109001

[42] Wang G, Zhang J, Lin S, Xiao H, Yang Q, Chen S, Yan B, Gu Y. Environmentally friendly nanocomposites based on cellulose nanocrystals and polydopamine for rapid removal of organic dyes in aqueous solution. Cellulose. 2020;27:2085-2097DOI: 10.1007/s10570-019-02944-6

[43] Yu Z, Hu C, Dichiara AB, Jiang W, Gu J. Cellulose nanofibril/carbon nanomaterial hybrid aerogels for adsorption removal of cationic and anionic organic dyes. Nanomaterials. 2020;10:169. DOI: 10.3390/nano10010169

[44] Zheng Y, Wan Y, Chen J, Chen H, Gao B. MgO modified biochar produced through ball milling: A dual-functional adsorbent for removal of different contaminants. Chemosphere. 2020;243:125344: DOI: 10.1016/j.chemosphere.2019.125344

[45] Zheng Y, Wang J, Wang Y, Zhou H, Pu Z, Yang Q, Huang W. The combination of MoS2/WO3 and its adsorption properties of methylene blue at low temperatures. Molecules. 2020;25:2. DOI: 10.3390/molecules25010002

[46] Dai L, Cheng T, Xi X, Nie S, Ke H, Liu Y, Tong S, Chen Z. A versatile TOCN/CGG self-assembling hydrogel for integrated wastewater treatment. Cellulose. 2020;27:915-925. DOI: 10.1007/s10570-019-02834-x

[47] Xu L, Sun P, Jiang X, Chen J, Wang J, Zhang H, Zhu W. Hierarchical quasi waxberry-like Ba5Si8O21 microspheres: Facile green rotating hydrothermal synthesis, formation mechanism and high adsorption performance for Congo red. Chemical Engineering Journal. 2020;384:123387. DOI: 10.1016/j.cej.2019.123387

[48] Guo XZ, Han SS, Yang JM, Wang XM, Chen SS, Quan S. Effect of Synergistic Interplay between Surface Charge, Crystalline Defects, and Pore Volume of MIL-100(Fe) on Adsorption of Aqueous Organic Dyes. Industrial and Engineering Chemistry Research. 2020;59:2113-2122. DOI: 10.1021/acs.iecr.9b05715

[49] de Souza PR, do Carmo Ribeiro TM, Lôbo AP, Tokumoto MS, de Jesus RM, Lôbo I.P. Removal of bromophenol blue anionic dye from water using a modified exuviae of *Hermetia illucens* larvae as biosorbent. Environmental Monitoring and Assessment. 2020;192:197. DOI: 10.1007/s10661-020-8110-z

[50] He Y, Fu X, Wu H, Zhu T, Li S, Na B, Peng C. Highly efficient removal of methyl blue from aqueous solution using a novel nitrogen-doped porous magnetic carbon, Desalination and Water Treatment. 2020;173:409-419. DOI: 10.5004/dwt.2020.24658

[51] Khan A, Wei D, Khuda F, Ma R, Ismail M, Ai Y. Comparative adsorption capabilities of rubbish

tissue paper–derived carbon-doped MgO and CaCO3 for EBT and U(VI), studied by batch, spectroscopy and DFT calculations. Environmental Science and Pollution Research. 2020;27:13114-13130. DOI: 10.1007/s11356-020-07796-3

[52] Li G, Wang Y, Bi J, Huang X, Mao Y, Luo L, Hao H. Partial oxidation strategy to synthesize WS2/WO3 heterostructure with enhanced adsorption performance for organic dyes: Synthesis, modelling, and mechanism. Nanomaterials. 2020;10:278. DOI: 10.3390/nano10020278

[53] Pan X, Qin X, Zhang Q, Ge Y, Ke H, Cheng, G. N- and S-rich covalent organic framework for highly efficient removal of indigo carmine and reversible iodine capture. Microporous and Mesoporous Materials. 2020;296:109990. DOI: 10.1016/j.micromeso.2019.109990

[54] Hasanzadeh M, Simchi A, Shahriyari Far H. Nanoporous composites of activated carbon-metal organic frameworks for organic dye adsorption: Synthesis, adsorption mechanism and kinetics studies. Journal of Industrial and Engineering Chemistry. 2020;81:405-414. DOI: 10.1016/j.jiec.2019.09.031

[55] Yan KY, Chen JY, Li XY, Wang Q, Kuang GC. Carboxylic Acid Enriched Porous Organic Polymer as a Platform for Highly Efficient Removal of Methylene Blue from Aqueous Solution. Macromolecular Chemistry and Physics. 2020;221:1900553. DOI: 10.1002/macp.201900553

[56] Li Y, Hou X, Pan Y, Wang L, Xiao H. Redox-responsive carboxymethyl cellulose hydrogel for adsorption and controlled release of dye. European Polymer Journal. 2020;123:109447. DOI: 10.1016/j.eurpolymj.2019.109447

[57] Wang Z, Gao M, Li X, Ning J, Zhou Z, Li G. Efficient adsorption of methylene blue from aqueous solution by graphene oxide modified persimmon tannins. Materials Science and Engineering C. 2020;108:110196. DOI: 10.1016/j.msec.2019.110196

[58] Abboud M, Sahlabji T, Haija MA, El-Zahhar AA, Bondock S, Ismail I, Keshk SMAS. Synthesis and characterization of lignosulfonate/amino-functionalized SBA-15 nanocomposites for the adsorption of methylene blue from wastewater. New Journal of Chemistry. 2020; 44:2291-2302. DOI: 10.1039/d0nj00076k

[59] Rathee G, Singh N, Chandr, R. Simultaneous Elimination of Dyes and Antibiotic with a Hydrothermally Generated NiAlTi Layered Double Hydroxide Adsorbent. ACS Omega. 2020;5:2368-2377. DOI: 10.1021/acsomega.9b03785

[60] Zhang W, Zhang RZ, Yin Y, Yang JM. Superior selective adsorption of anionic organic dyes by MIL-101 analogs: Regulation of adsorption driving forces by free amino groups in pore channels. Journal of Molecular Liquids. 2020;302:112616. DOI: 10.1016/j.molliq.2020.112616

[61] Sočo E, Papciak D, Michel MM. Novel application of mineral by-products obtained from the combustion of bituminous coal-fly ash in chemical engineering. Minerals. 2020;10:66. DOI: 10.3390/min10010066

[62] Xiao X, Wang Y, Cui B, Zhang X, Zhang D, Xu X. Preparation of MoS2 nanoflowers with rich active sites as an efficient adsorbent for aqueous organic dyes. New Journal of Chemistry. 2020;44:4558-4567. DOI: 10.1039/d0nj00129e

[63] Chen X, Cui J, Xu X, Sun B, Zhang L, Dong W, Chen C, Sun D. Bacterial cellulose/attapulgite magnetic composites as an efficient adsorbent for heavy metal ions and dye

treatment. Carbohydrate Polymers. 2020;229:115512. DOI: 10.1016/j.carbpol.2019.115512

[64] Li D, Tian X, Wang Z, Guan Z, Li X, Qiao H, Ke, H, Luo L, Wei, Q. Multifunctional adsorbent based on metal-organic framework modified bacterial cellulose/chitosan composite aerogel for high efficient removal of heavy metal ion and organic pollutant. Chemical Engineering Journal. 2020;383:123127. DOI: 10.1016/j.cej.2019.123127