INTERNATIONAL JOURNAL OF ADVANCE RESEARCH, IDEAS AND INNOVATIONS IN TECHNOLOGY

ISSN: 2454-132X
Impact factor: 4.295
(Volume 5, Issue 5)
Available online at: www.ijariit.com

Efficiency of using functionalized mesoporous silica-based composites in organic textile dyes removal from aqueous media: Review

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ABSTRACT

Due to its enormous significance for human life, at the present time water treatment is the subject of greatest importance. Technological progress, which is an integral part of the modern world is also the cause of a number of environmental problems. One such problem is water pollution by organic dyes which are mainly a consequence of the development of the textile industry. The importance of this problem led to the development of many different methods and techniques. Due to a number of advantages (stability, low cost, efficiency, etc.), the use of compounds based on silica is a matter of great interest to scientists at the present time. This review discusses the applications of functionalized silica-based inorganic/organic copolymers in removal of the most common organic dyes for water and wastewater purification process.

Keywords— *Silica nanomaterials, Inorganic/organic copolymers, Organic dyes removal, Water treatment*

1. INTRODUCTION

The importance of water cannot be overestimated. Life on the Earth is impossible to imagine without water. Water is a prerequisite for the functioning of living organisms, factories, factories and industrial enterprises. We all need water, starting from the roots of the plant in the soil and ending with the astronauts at the space stations. The Earth's surface is about 70% composed of water, presented in the form of oceans, seas, rivers, lakes, natural rainfall and so on (Baker B., Omer A., Aldridge K., 2016). Water use is represented by a wide range of applications, the most important of which are freshwater ecosystems, agriculture, energy production, sanitation, rural development and so forth (World Water Council, 2014). Also water resources have a great potential in poverty reducing, food security, human health, sustainable energy sources, etc. (Balasubramanian A., 2015).

Due to its importance water is also called “life”. Water is not only a necessary part of human life but it is also part of human body. As known, the body of an adult human being is about 60% consists of water. Water in various amounts is distributed in our organs, for example, the water content in the human brain is 74 percent, skin – 64%, muscles – 79%, lungs – 83%, liver – 71%, skeleton – 31%, heart – 73% and kidneys – 79% (Mitchell H., Hamilton T., Steggerda F., Bean H., 1945). Thus, the harmful substances contained in polluted water, getting into human organs become the cause of many diseases (Hatami, 2013).

One of the most catastrophic consequences of the lack of clean water, as mentioned above, is the disease caused by the use of contaminated water resources. Thus, according to World Health Organization report the consumption and use of contaminated water is a source of a number of serious diseases, including diarrhoea, malnutrition, intestinal nematode infections, lymphatic filariasis, trachoma, schistosomiasis, malaria, drowning and other quantifiable diseases. So, according to this report, about 2200 million people worldwide suffer from these infections every year. Among them, 1.4 million child deaths every year are caused by diarrhoea; 860 000 child deaths annually are caused by malnutrition; one third of the population of the world is affected by intestinal nematode infections; every year in the world 25 million people become infected with lymphatic filariasis, which leads to a limitation of their capabilities; trachoma - 5 million people with visual impairment; schistosomiasis - 200 million cases of disease per year; drowning – 280 000 deaths per year (Prüss-Üstün A., Bos R., Gore F., Bartram J. , 2008). For comparison, according to the 1997 report provided by Commission on Sustainable Development (the United Nation) 2.3 billion people, each year were suffering from water-related diseases (Lundqvist J., Gleik P., 1997). However, despite the enormous importance of water in people's lives today 3 out of 10 people on our planet still do not have access to clean drinking water (UNESCO, 2019). Taking into account the enormous importance of water sources in human life and the sustainable functioning of society, access to water was recognized as one of the human rights in the framework of the right to adequate standard of living (Substantive Issues Arising in the Implementation of the International Covenant on Economic, Social and Cultural Rights, 2003). And nowadays, following the work done in this area, we can conclude that some success has been achieved, so according to the data presented in table 1, it can be noted that the number of people with limited access to clean water sources significantly decreased in 2015 compared to 1990.

Table 1: Number of people without access to an improved water service (WHO, 2015)

Year	Population without access to improved drinking water services
1990	1.26 billion
1995	1.18 billion
2000	1.07 billion
2005	943.56 million
2010	806.39 million
2015	665.55 million

All this progress in the field of water resources was made possible largely due to efforts by the UN. Thus, the problem of water and sanitation has been recognized as one of the goals of sustainable development. According to the 2030 Agenda for Sustainable Development, by 2030, every inhabitant of our planet will have access to safe and affordable drinking water. (Transforming Our World: The 2030 Agenda for Sustainable Development, 2015).

With the development of industry, the growth of cities and the world's population the problem of pollution of water sources is becoming more and more relevant. The main sources of water pollution can be divided into two large groups: point sources – those that have a direct source of pollution and diffuse (non-point) sources – those that do not have an obvious source of contamination (Sharma S., Bhattacharya A., 2016). The main point sources include industrial and municipal wastewater; runoff from animal feedlots, waste disposal sites, oil fields, mines, and construction sites; etc. Examples of non-point sources may include runoff from agriculture, range, and pasture, from abandon mines and urban runoff; activities on land that produces pollutants, such as logging, wetland conversion, construction and development waterways; atmospheric deposition above the surface of the water and so on (Carpenter S., Caraco N., Correll D., Howarth R., Sharpley A., Smith V., 1998).

The most generalized is the separation of pollutants into inorganic and organic pollutants. Inorganic pollutants are represented mainly by heavy metals, nitrates, and phosphates from agriculture runoff and chemical waste. (Singh M., Gupta A.,, 2016). Metal pollution is a constant problem in many contaminated sites. Surface and groundwater can be contaminated by metals as a result of wastewater discharges or as a result of direct contact with soils, sediments, mining waste and garbage contaminated with metals (Evanko C., Dzombak D., 1997). Heavy metals pose a serious danger to human health, as they can cause disruption of the functioning of the organs of the human body. For instance, lead causes delays in physical or mental development in children, in adults causes impaired kidney function; mercury also causes impaired kidney function; uranium increases risk of cancer; barium leads to increasing blood pressure (Office of Water EPA, 2018).

Organic pollutants include insecticides and herbicides, volatile organic compounds, orgaohalogenides and other types of chemicals; bacteria from livestock and sewage; food industry waste; pathogens; etc. (Tsuchiya, Y., 2010). Thus, water contamination from organic pollutants in the aqueous environment comes mostly from artificial pollutants from human waste and industrial chemicals and also from natural products of aqueous microorganisms.

Residual dyes from various sources which will be discussed in this review are considered the most diverse organic pollutants that fall into water resources and into wastewater treatment systems. The textile industry is one of the fastest growing, oldest and most technologically complicated industries. The rapid growth rate of the textile industry is primarily due to an increase in the world's population and demand. So, textile mills and their wastewater increased proportionally, causing a serious pollution problem worldwide. And today, the textile industry accounts for 2/3 of the total dye market. This is recognized as the main cause of environmental contamination. The textile industry and its bumps are one of the main causes of natural water pollution with organic dyes around the world (Baban A., Yediler A., Ciliz N., 2010).

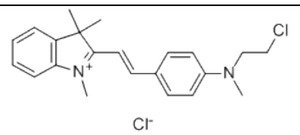
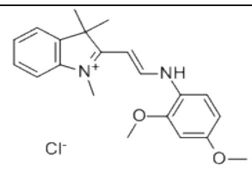
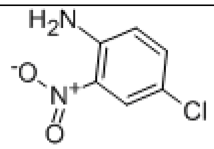
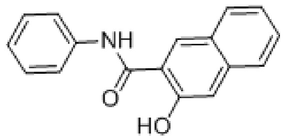
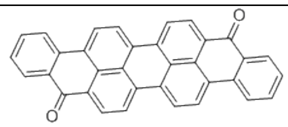
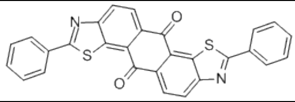
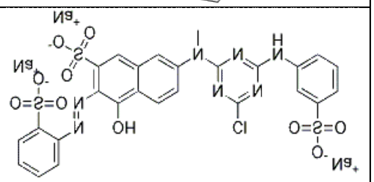
Textile dyes are recognized as synthetic chemical compounds that have an aromatic structure and are biodegradable because of their xenobiotic nature. They are mainly obtained from two sources: petroleum-based intermediates and coal tar. These dyes are sold in the form of pastes, liquid dispersions, powders or granules. The general chemistry of dyes as one of the basis for the classification assorts textile dyes into the following 12 groups: direct dyes, acid dyes, azoic dyes, Sulphur dyes, disperse dyes, basic dyes, reactive dyes, mordant (chrome) dyes, oxidation dyes, solvent dyes, optical dyes and vat dyes (Samchetshabam G., Ajmal H., Choudhury T., 2017). Chemical structure of some different organic dyes are represented in table 2 (Zollinger H., 2003)

Based on the general structure textile dyes are also classified into cationic, nonionic and anionic dyes. The main anionic dyes are acidic, reactive and direct dyes, and the most problematic among them are acidic, brightly colored and water-soluble dyes (it is not possible to remove these dyes using conventional processing systems). The main nonionic dyes are not ionized in the aqueous environment dispersed dyes. The main cationic dyes are reactive, azo basic and disperse dyes, etc. (Robinson T., McMullan G., Marchant R., Nigam P. , 2001).

Many different techniques and methods have been developed to remove organic dyes from water. Textile organic dyes should be separated and removed from water especially from industrial wastewater by viable and efficient treatment at a wastewater treatment plant or in situ, following 2 various treatment concepts, such as: separation of organic contaminant from water or partial/complete mineralization/decomposition of organic pollutants (Anjaneyulu Y., Sreedhara Ch., Suman R., 2005).

In order to remove organic dyes from wastewater a lot of methods were applied (Robinson, McMullan, Marchant R., Nigam P., 2001), among them: chemical treatment such as ozonation (Shriram B., Kanmani S. , 2014), oxidation with NaOCl (Pizzolato T., Carissimi E., Machado E., 2002), electrokinetic coagulation (El-Hosiny F., Abdel-Khalek M., Selim K., Osama I. , 2016), ionic exchange and Fenton process; physical methods include irradiation, microfiltration, membrane process, nanofiltration, reverse osmosis, ultrafiltration, and adsorption; some biological treatment methods are also applied, among them aerobic and anaerobic processes, Single cell (Fungal, Algal and Bacterial) etc. (Karthik V., Saravanan K., Bharathi P., Dharanya V., Meiaraj C., 2014).

Table 1: Chemical structure of different types of dyes

Class	Name	Synonyms	Molecular formula	Structure
CATIONIC DYES	Astrazon Pink FG	Basic Red 13	$C_{22}H_{26}Cl_2N_2$	
	Basic Yellow 11	Yellow 3GL	$C_{21}H_{25}ClN_2O_2$	
AZOIC DYES	Sanyo Fast Red Salt	4-Chloro-2-nitroaniline	$C_6H_5ClN_2O_2$	
	Naphthol AS	Orange Base GC	$C_{17}H_{13}NO_2$	
VAT DYES	Vat Violet 10	Benzadone Violet B	$C_{34}H_{16}O_2$	
	Vat Yellow 2	Indanthrene Yellow	$C_{28}H_{14}N_2O_2S_2$	
REACTIVE DYES	Reactive Orange 5	Concion Orange	$C_{26}H_{17}ClN_7Na_3O_{10}S_3$	

Biological purification is known as the most economically efficient alternative of physicochemical methods of treatment. However, the use of biological treatment is often limited due to technical limitations. Biological methods of treatment require a large land area and are limited by the daily variations and toxicity of certain chemicals and the rigidity of the design and operation (Crini G., 2006). The most important drawback of chemical techniques is the making of sludge what is the result of the using of chemicals and in addition the high cost of the removal of sludge. There is also the possibility of a secondary pollution problem due to overuse of chemicals (Huang C., Cang K., Ou H., Chiang Y., Wang W., 2011).

The most common treatment used in water pollutant removal is adsorption. Adsorption is recognized worldwide as the most promising wastewater treatment technique. This method is characterized by low cost, ease of use, the possibility of repeated use of the adsorbent, biostability, etc. (Kaykhaii M., Sasani M., Marghzari S., 2018). Although most adsorbents are inexpensive and readily available their application in treatment processes was limited due to the problems caused by their utilization, regeneration, poor mechanical stability, the low removal efficiency of a wide range of dyes and sludge production (Nguyen T., Juang R., 2013). The most commonly used adsorbent is activated carbon. However, for a number of reasons, recently activated carbon in the field of wastewater treatment has been replaced by various polymers and composite materials. One such material is silica-based inorganic/organic copolymers. This review will be discussed using functionalized mesoporous silica materials in dye removal from wastewater.

2. MESOPOROUS SILICA

According to the IUPAC definition, mesoporous materials include materials with a pore size of 2-50 nm (IUPAC, 1972). For the first time, mesoporous silica material was synthesized in 1990 by Yanagisawa and co-workers in Japan (Yanagisawa T., Schimizu T., Kuroda K., Kato Bull C. , 1990). A new group of materials was investigated in 1992 by Mobile Oil Company's researchers. The new family of mesoporous materials including lamellar MCM-50, cubic MCM-48, and hexagonal structured MCM-41 was called M41S (Kresge C., Leonowicz M., Roth W., Vartuli J., Beck J. , 1992). Since that different types of mesoporous structured silica-based materials were synthesized, among them SBA – Santa Barbara Amorphous family of mesoporous silica, OMS – Ordered Mesoporous Silica, MSU – Michigan State University Silica, HMS – Hollow Mesoporous Silica, MCF – Meso Cellular Form Silica, FSM – Folded Sheet Mesoporous Silica, PMO – periodic mesoporous organosilica, etc.

Table 3: Different silica-based mesoporous materials characterization

Mesoporous silica type	Pore diameter, nm	BET surface area, m^2/g	Reference
MCM-41	30,0	980.0	(Chen C-Y., Li H-X., Davis M., 1993)
MCM-48	29,4	1055,8	(Chen F., Huang L., Li Q., 1997)
SBA-15	5,1	1000,0	(Zhao D. et al, 1998)
MSU	4,24	813,9	(Kharbouche L. et al, 2019)
HMS	3-4	426,0	(Sun Q. et al, 2003)
MCF	3,2	995,0	(Lukens W., Yang P., Stucky G., 2001)
FSM	3,8	600,0	(Inagaki S., 2004)
PMO	7,5	484,0	(Vidal C. et al, 2011)

Due to their high surface area, porous materials have been widely used in adsorbents, catalysts and carrier material. The Figure 1 shows data on the number of studies on request of mesoporous silica from 1990 to 2018 (SciFinder, 2019). Mesoporous materials are divided into 2 groups: silicon-based materials (pure and modified mesoporous silicates); non-siliceous materials (including transition metal oxides, non-metallic oxides, and metal sulfides). The main most important characteristics of mesoporous materials include large surface area what makes possible their application in different fields of chemical industry, variable pore size (2-30nm), thermal and hydrothermal stability gained in process of modification, ordered porous structure and so on (Chang-Sik Ha., Sung S., 2019). In order to synthesize materials with mesoporous structure generally applied following techniques: sol-gel method, microwave assisted method, template assisted method, chemical etching method. To establish the porosity and pore size, the gas adsorption method is mainly used (Sotomayor F., Cychosz K., Thommes M., 2018).

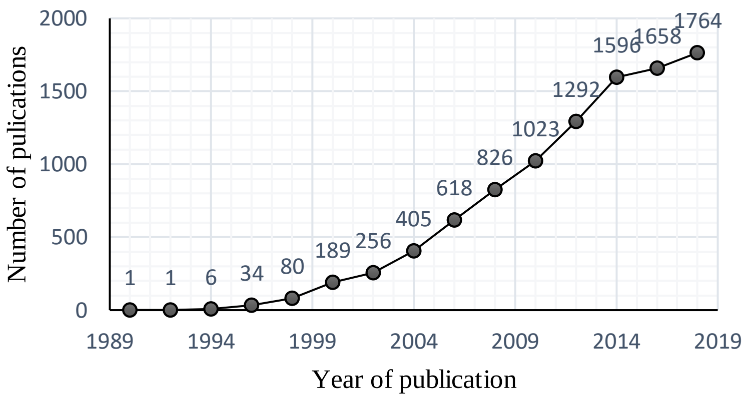


Fig. 1: Number of publications per year - key word 'Mesoporous silica' (SciFinder, 2019)

Mesoporous silica-based nanoparticles can by synthesized in different ways, with multiple pore size, dimensions, with different morphology and structure, using various silica source, surfactants or polymers, under various process controlling conditions like concentration, pH, temperature, etc. (Wu S., Mou C., Lin H., 2013) As for now, in order to synthesize mesoporous silica-based materials different methods were investigated. All of these approaches can be allocated into 4 techniques (Chang-Sik Ha., Sung S., 2019):

- Sol-gel method
- Template assisted method
- The liquid crystal template approach
- Microwave assisted method
- Chemical etching method.

3. MESOPOROUS SILICA FUNCTIONALIZATION

As mentioned before, silica-based nanocomposites are used in several fields of processes such as catalysis, adsorption, material carrying, etc. The functionalization of mesoporous silica-based nanomaterials is applied in order to improve physico-chemical properties the material. Also should be mentioned the fact that silica-based materials' surface covered by silanol groups makes possible the adjustable functionalization of silicas pore surface. As of this writing mesoporous silica surface functionalization is very intensively explored and described in literature (Stein, A., Melde, B., Schroden R. , 2000). In order to functionalize the surface of mesoporous silica with organic functional groups two main approaches have been investigated: grafting and co-condensation.

The grafting method belongs to post-synthesis methods of modification of mesoporous silica materials. This method consists of attaching functional groups to a previously prepared surface of inorganic component. In most cases, the grafting process occurs after removal of the surfactant. Silanol groups presented in high concentration on the surface of mesoporous silica acts in role of grafting points for organic modification which is usually carried out by process of sylation (Stein, A., Melde, B., Schroden R. , 2000). Another approach to functionalization of mesoporous silica surface is co-condensation which belongs to sol-gel chemistry. Co-

condensation is able to give uniformly distributed organic groups over the entire inner surface of the pores and can provide easy control of mesoporous silica particles' morphology (Yang H., Coombs N., Ozin G. , 1997).

Table 2: Applications of different types of mesoporous silica materials

Mesoporous silica type	Modified group	Applications	Reference
MCM-41	-	Catalysis, sorption, filter membranes, optics, etc.	(Kresge C., Leonowicz M., Roth W., Vartuli J., Beck J. , 1992)
MCM-48	-	Catalysis, sorption, drug delivery, etc.	(Xu J., Luan Z., He H., Zhou W., Kevan L., 1998)
MCM-41	$-NH_2$ (amino group)	CO_2 sorption	(Aghdas H., Habibollah Y., Zahra M., 2009)
MCM-41	Iminodiacetamide	Nd^{3+}, Ce^{3+}, Lu^{3+}, Gd^{3+}, and Eu^{3+}, sorption	(Glen, E., Wilaiwan C., Ryan D., 2011)
MCM-41	Ammonium	Anionic dyes adsorption	(Qin Q., Ma J., Liu K. , 2008)
MCM-41	Magnetic Aminopropyl group	U removal	(Li D. et al, 2016)
FSM	-	Adsorption of naphthoic acids	(Tozuka Y., Yokohama C., Higashi K., Moribe K., Yamamoto K., 2009)
FSM	Alanine racemase	Adsorption	(Nara T. et al, 2010)
MCF	Amphiphile	Cu(II) and Zn(II) ions removal	(Sharifpour E., Haddadi H., Ghaedi M., Dashtian K., Asfaram A., 2018)
MCF	Amino acid-functionalized ionic liquids	CO_2 capture	(Liu Sh-H., Sie W-H., 2016)
SBA-15	-	Catalysis, sorption, drug delivery, etc.	(Zhao D. et al, 1998)
SBA-15	Sulfonic acid	Catalysis, adsorption	(Shen J., Herman R., Klier K., 2002)
SBA-15	1-furoyl thiourea	Hg(II) adsorption	(Mureseanu M., Reiss A., Cioatera N., Trandafir I., Hulea V., 2010)
MSN	Amino groups	Methylene Blue adsorption	(Chueachota R. et al, 2018)
PMO	-	Adsorption of polycyclic aromatic hydrocarbons	(Vidal C. et al, 2011)
PMO	Bipyridine	Catalysis	(Yamaguchi Sh. et al, 2019)
MSU-1	Vanadium oxide	Catalysis	(Guosong S. et al, 2016)
MSU-1	-	Extraction of pesticides	(Kharbouche L. et al, 2019)

Table 3: Efficiency of using mesoporous silica-based materials comparing with different types of adsorbents

Dye		Adsorbent										
		Mesoporous silica-based material							**Other types**			
Class	name	Type	Modification	Pore size, nm	S_{BET}, m^2/g	A, mg/g	R, %	Reference	Type	R, %	A, mg/g	Reference
Basic dye	Methylene Blue (MB)	MCM-41	-COOH	2,55	757	113	-	(Ho K., et al, 2003)	C_{act}	-	66	(Jawad A., et al, 2018)
		MSM	-COOH	2,1	213	102	-	(Fu X., et al, 2006)	Hydrogel	>99	-	(Wang W., et al, 2019)
		SBA-15	-	10,7	669	280	-	(Dong Y. et al, 2010)	Graphene	-	357	(Liu X., et al, 2019)
		CFA-MS	-	4,9	497	317	-	(Yuan N., et al, 2019)	Biochars	-	114	(Orfanos A., et al, 2016)
		HMS	Cu_2O	3,18	201	-	99,8	(Wang J., et al, 2018)	chitosan	99,4	199	(Guo J., et al, 2018)
Acidic dyes	Acidic Blue	MCM-41	$-NH_2$	2,57	774	256	-	(Ho K., et al, 2003)	chitin	85,2	177	(Gyeong K., et al, 2019)
		MPS	$-NH_2$	-	313	-	78,0	(Mahmoodi N., et al, 2011)	Rambutan	87,1	36	(Sivarama Krishna L., et al, 2018)
		SBA-15	$-NH_2$	4,1	166	1429	-	(Mirzaie M., et al, 2017)	zeolite	-	112	(Krishna L., et al, 2019)
		SBA-15	$(C_3H_4O_2)_n$	2,8	159	909	-	(Merat G., et al, 2018)	diatomite	72,8	-	(Badii K., et al, 2010)
		SBA-15	$-NH_2$	4,8	224	-	77,7	(Akbartabar I., et al, 2017)	CeF_3	99,9	-	(Jahedi F., et al, 2018)
		HMS	CDs	3,9	416	-	80,0	(Asouhidou D., et al, 2009)	sawdust	-	8	(Jahan Ara N., et al, 2013)
	RR	MCM-41	$-NH_2$	1,8	215	-	99,1	(Santos S, et al, 2013)	C_{act}	39,0	-	

Reactive dyes	RY	SBA-15	Al	6,6	1020	-	83,0	(Ahmed K., et al, 2016)	MM	-	8,62	(Gomez-Trevino D., et al, 2013)
	Yellow	MSM	$-NH_2$	2,3	516	351	-	(Moscofian A., et al, 2013)	chitosan	-	885,0	(Wu F., et al, 2001)
	Red					388			C_{act}	-	209,0	(Degs Y., et al, 2008)
	Blue					253			cellulose	-	14,4	(Xie K., Zhao W., He X., 2011)

5. MESOPOROUS SILICA IN DYE REMOVAL

As mentioned above, first silica-based mesoporous material was investigated in 1990, since then it has been widely used as an adsorbent in wastewater purification (Paul M., Pal N., Bhaumik A. , 2012). Table-4 presents a comparison of the effectiveness of the use of materials based on silica and other types of adsorbents in the removal of the same waste from the textile industry. The adsorption process of organic dyes on silica-based mesoporous nanoparticles is carried out mainly in 3 stages:

- Film diffusion - dye transfer to the outer surface of the adsorbent (only a small amount of pollutant can be absorbed on this surface);
- Particle diffusion – dye transfer within the pores of the silica pores;
- Dye adsorption on the inner surface of the mesoporous silica (Dinesh M., Sinhg P., Singh G., Kumar K., 2011).

6. CONCLUSION

In this work applications and efficiency of using mesoporous silica-based inorganic/organic materials in wastewater purification processes are reviewed. Due to their low cost, easiness of the procedure, chemical stability and the fact that silica is available from different natural sources, using mesoporous silica-based materials in adsorption processes in water purification are possible to become efficient method along with such traditional techniques as filtration, mechanical separation, flocculation, chemical treatment, coagulation, etc. Today, thanks to a number of exceptional advantages such as very high ability to remove organic components, being a cost effective product and reusability, activated carbon, the most common adsorbent in water treatment processes. However, due to the fact that the pores of activated carbon are very small, its use to remove polyatomic large molecules like organic pollutants or dyes seems ineffective. While the ability to modify the porosity of silica-based materials and the possibility of functionalization the pore surface with various functional groups, mesoporous silica-based materials can become very effective adsorbents for the selective removal of organic contaminants. As studies show, modification of the surface of mesoporous silica-based materials, in particular by amino and polymer groups, leads to the high efficiency of this material as an adsorbent for the removal of organic dyes during water treatment. However, despite all the advantages of this material, to date, scientists are faced with the task of transferring laboratory research and progress to an industrial scale.

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