

A Comprehensive Review of MOF Based Ceramic Membranes for Removal of Dye: A review

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Abstract

Clean water is necessary for whole human society and environment as well but this water is being wasted by industrial effluents like dyes, heavy metals etc. Due to high stability and anti-oxidation properties, ceramic membranes have been used in waste water treatment. Porous material has been used for good separation due to high selectivity by using processes like microfiltration and ultrafiltration. Some problems regarding ceramic membranes can be solved by coating MOFs on ceramic membrane. Moreover, metal organic frameworks are a combination of metals and organic ligands, can be used worldwide for waste water treatment, separation of gas and in field of energy. There is an issue regarding the stability of MOFs in water, researchers still working on enhancement of applications of MOFs in waste water treatment. It has been observed that by regulating the intracrystalline structure performance of MOFs can be improved. In this review, synthesis methods of MOFs based ceramic membranes like seeding method, layer by layer method and microwave thermal deposition method and applications in separation of heavy metals, separation of salts and minerals from saline water are discussed.

Keywords: MOFs based ceramic membranes, removal of dye, water treatment

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1. Introduction

Over the past ten years water consumption by industries is increasing day by day [1]. This large amount of waste water produced by industries effecting both the environment and human health [2]. One of most important waste water including oily waste water or water containing dyes produced by many industries like textile, oil and gas extraction, metal finishing, food and beverages. [3]. For the treatment of this water, a lot of techniques have been produced like flotation, coagulation, flocculation, adsorption and gravitational settling [4]. Moreover, use of these techniques is limited due to low efficiency, requires a large space for installation and use of large amount of chemical [5]. Due to increasing demand of organic dyes in textile, pharmaceutical, food and cosmetics, it has become the emerging power in many industries. Previous studies have suggested that organic dyes like even in very low amount can cause hazardous pollution because of high solubility, stability and high toxicity [6]. Aquatic system disturbs due to organic dyes present in water by retards the amount of light reaching deep in water and directly affect photosynthetic activity [7]. Organic dyes are not easily removed by using traditional methods like coagulation, air flotation, sand bed filtration, and sedimentation because of their intense color that are left

at the end [8-9]. The organic dye residues remain in aquatic environment for at least 50 years due to their ability to retards light and heat, high chemical stability, and low availability of oxygen to aquatic animals [10-11].

So, the main focus of researchers is to find a suitable way to remove these pollutants as well as their persistent use under extreme environmental conditions. One of the advanced techniques is membrane separation like ceramic membranes which has higher efficiency, high mechanical, thermal and chemical stability that make them appropriate for waste water treatment, in comparison to traditional methods. So, as compared to other polymer membrane, ceramic membranes have high porosity, high hydrophilicity, and narrow pore size [12-13]. During separation process, membrane fouling is observed that is actually contamination of membrane surface due to particles or dissolved substances. By constructing the fouling resistant membrane by modification of membrane, surface problem of membrane fouling can be overcome [14]. Now a days, ceramic membranes are widely used in microfiltration, ultra-filtration, reverse osmosis and gas separation processes. To enhance the application in separation process even in unfavorable conditions, metal organic framework-based ceramics membranes are used due to their high performance. Actually,

ceramic membranes act as a substrate and by applying one or more layer of MOFs on substrate increase its efficiency [15]. Metal organic framework has opened a new door for the formation of nano porous membrane on ceramic substrate. Metal organic frameworks are actually hybrid materials containing metal ions and organic linker with the complex pore structure.

Many types of MOFs are used to increase stability in waste water treatment but not all because of poor stability of certain metal organic frameworks. So, it can be produced in one or more dimensions, which make it appropriate component for formation of membrane. For the formation of MOFs membrane on ceramic substrate at solvothermal temperature, ceramic membranes act as support due to high structural stability in high temperature as well as solvent environment. This feature makes them differentiable than other organic membranes. So, to construct a stable MOFs based ceramic membranes the combined benefits of nano porous metal organic frameworks and ceramic membrane is of great importance. MOFs based ceramic membranes have many applications in both water treatment and gas separation but it works better in gas separation due to poor stability of some MOFs [16-17]. For the treatment of water, many water stabilized metal organic frameworks and membranes have been used. It has been observed that by regulating intracrystalline structure, performance of MOFs membranes can be improved. Dong et al. summarized the data about application of ceramic membranes that it can be used even in unfavorable conditions like contamination in waste water treatment, oil and water separation and saline water [18]. In this article, we will review existing studies on efficiency of different methods of MOFs based ceramic membranes, their way of working and potential application for dye removal.

2. Mechanism of dye removal by MOFs

Researchers have given importance to metal organic frameworks due to high surface area and high stability. Coordination bond is formed between metal ions like Cu, Zn, Co, Mg and inorganic linkers amines, carboxylates, azolates [19]. By using MOFs, Adsorption mechanism for removal of toxic dyes including anionic and cationic dye have been observed. MOFs used to remove Cationic dye as, rate of adsorption process depends on functional group of MOFs and molecular structure of dyes. Adsorption process carried out by many ways like Hydrogen bonding, ion exchange, Hydrophobic interactions, π - π interaction [20]. Positively charged MB^+ electrostatically adsorbed on negatively charged nitrates that contain Cu-MOF-1 and adsorption occurred in about 5mins but same type of MOF was used to remove Rhodamine B dye so, due to size restriction it was observed that adsorption process is very low as adsorption occurred in about 30mins. It was observed that size of Rhodamine B larger than MB^+ [21]. Electrostatic interaction is the main factor that controls the rate of adsorption. MOFs doped with metals increase the surface area and sites for adsorption so, as a result electrostatic interaction increases. Moreover, π - π stacking, hydrogen bonding, ion exchange mechanisms used to remove Anionic dyes as well.

Negatively charged dye adsorbed on positive surface of MOFs by electrostatic interactions. Previous studies showed that for the removal of AB80 by Al-MOF several mechanisms were observed like hydrogen bonding (bond formed between amine group of dye and oxide group

of linker MOF), hydrophobic interaction and electrostatic interaction (between oppositely charged material) [22]. Same type of mechanism was observed for the removal of Congo red dye using MIL-100Fe with additional pore filling (Figure 1) [23]. So, electrostatic interaction and hydrogen bonding combined together play an important role for removal of dye using MOFs. Metal organics frameworks are highly efficient for removal of dyes due to high surface area, good recyclability, high stability and performed well than other traditional adsorbents but lack thermal stability, mechanical strength and chemical resistance. So, MOFs based ceramic membranes are used that provides not only stability but also mechanical strength. ceramic materials like alumina, zirconia. are highly resistant to mechanical stress, high temperature and harsh environmental conditions. So, combination of MOFs and ceramic membranes responsible for high performance separations on industrial scale.

3. Comparison of different ceramic membranes

Many ceramic membranes like zirconia, alumina, silica and titania are used for waste water treatment [24]. Due to high hydrophilicity, mechanical strength, microstructure and chemical stability it's very difficult to choose distinct ceramic membrane [25]. Zirconia has high mechanical strength, chemical resistance and permeability is relatively low [26]. Alumina has high mechanical strength but less fouling resistance while silica has high chemical resistance but expensive one [27]. Besides all these characteristics, ceramic membranes have a problem of membrane fouling. membrane fouling is the deposition of suspended particles on the surface of membrane [28]. suspended particles may be organic (carbohydrates, proteins, oils and grease) or inorganic can accumulate in pores and surface of membrane. Some salts like calcium sulphates, calcium carbonate, silica in water deposit on surface of membrane and process of deposition is known as scaling [29]. So, by coating metal organic frameworks on ceramic membrane, problem of membrane fouling can overcome. This improves overall separation process and increase the anti-fouling abilities.

4. Synthesis methods of MOFs membrane on ceramic substrate

4.1. Situ growth method

Situ growth method is the simplest method for preparation of MOFs membranes on substrate. Two main stages are discussed, one is nucleation stage and other one is growth stage. In nucleation stage crystals randomly grow on substrate and adsorb to its surface. While in growth method, membrane layer formed by continuously growing the crystal around the nucleus and these crystals cross linking to each other and undergoes mis generation [30]. ZIF-8 membrane was synthesized by Jeong et al [31]. And recently UIO-66 membrane on aluminum oxide modified zirconium oxide substrate was also synthesized by situ growth method [32]. As far as industrial applications are concerned, main benefit of situ method is that it can be operated in simple conditions. Due to chemical properties of substrate MOFs crystals grow more efficiently in solution than on surface of substrate. it's very difficult to synthesize MOFs at normal conditions by situ growth method because many MOFs have low growth rate at different substrate surface. So, conditions for synthesis of metal organic frameworks are very strict that effects the quality of membrane [32] Before using situ growth method,

it's very important to modify the ceramic substrate to increase the growth ability of MOFs crystals. So, due to homogenous

nucleation all the MOFs cannot be synthesized by situ growth method.

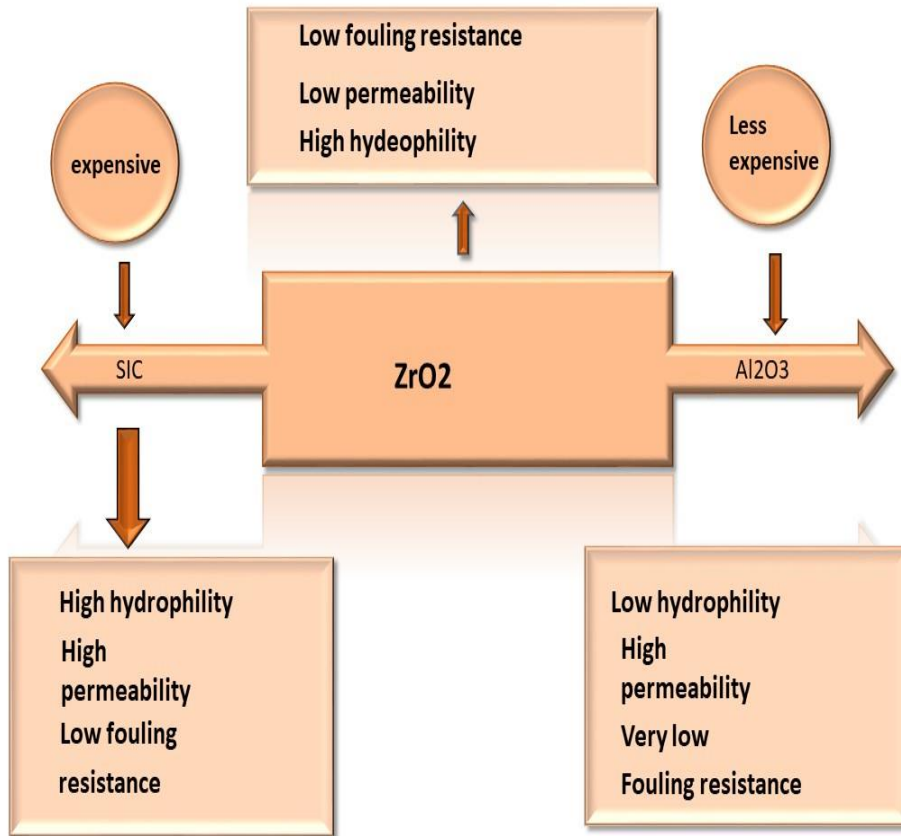


Figure 1. Synthesis methods of MOFs membrane on ceramic substrate

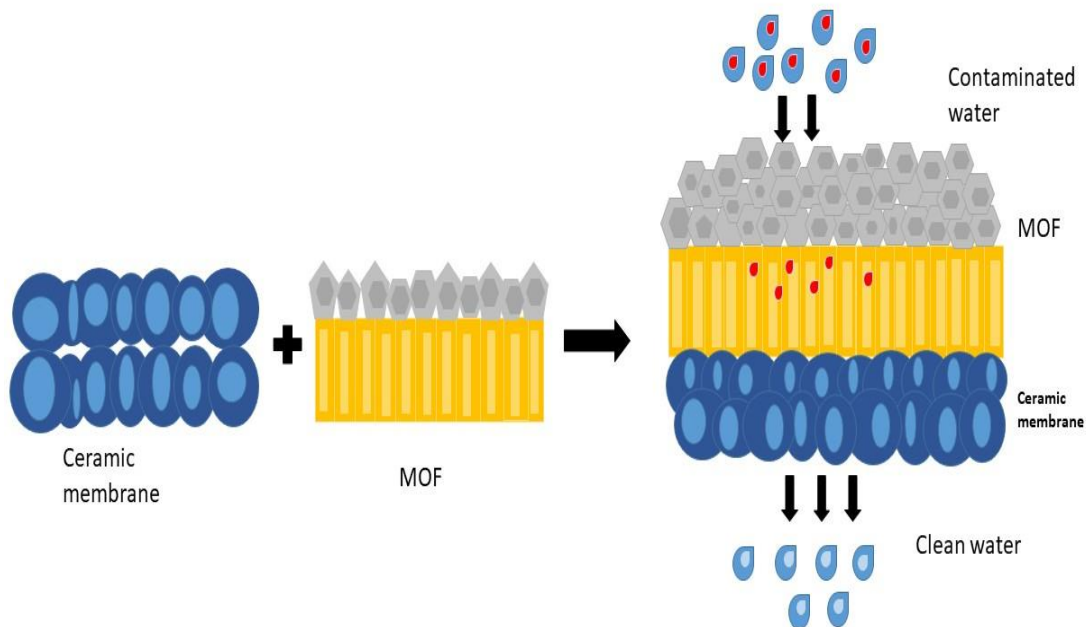


Figure 2. Separation of dye by MOFs based ceramic membranes**Table 1.** Comparative evaluation of synthesis methods of MOF membrane on substrate

Method	Pros	Cons	References
Microwave- induced thermal method	Requires less reaction time, high efficiency	Expensive equipment for heating is required	[33]
Electrochemical method	Capable of working successfully on industrial scale	Membrane fouling may occur	[34]
Layer by layer method	Requires low temperature	Multiple steps involve requires more time for reaction	[35]
Situ growth method	Easy and simplest method	Random growth of crystals	[36]
Seeding method	Crystal growth can easily be controlled, dense membrane easily obtained	Significant demand of seed quality	[37]

Table 2. Comparative evaluation of potential applications of different MOFs based ceramic membranes for waste water treatment

Method used	Ceramic membrane	MOFs	Applications	References
Situ solvothermal	Aluminum oxide	Uio-66	Removal of Humic acid	[38]
Seeding method	Aluminum oxide	ZIF-300	Removal of heavy metals and dye	[39]
Situ solvothermal	Aluminum oxide	MOF-303	Removal of salt and minerals from water	[40]
Situ solvothermal	PDA-Aluminum oxide	ZIF-8	Removal of salt and minerals from water	[41]
Situ Method	AAO	Uio-66NH2	Separation of ions	[42]

4.2. Layer by layer method

In this method, modified substrate dipped in both metal and organic linker solution Shekhah et al synthesized Zn(II)-BTC and Cu₂(BTC)₃, their preparation methods, structure and performance are also discussed by using this method [35]. By using this layer by layer method, DA-MOF and L2MOF also synthesized by Wiederrecht et al in 2013 [43]. One of the main benefits of this method is that it can be easily operated at room temperature, cycle number and membrane thickness can easily be controlled. So, by using this method Li et al synthesized the Zn-MOF membrane on ceramic substrate for the removal of dye from water [44].

4.3. Seeding Method

In 1994, Lai et al synthesized zeolite molecular sieve membrane [31]. Two steps are discussed for the formation of MOFs membrane on substrate. In the first step, Nano-MOFs seed deposited on ceramic membrane at room temperature undergoes physical adhesion or weak chemical bonding, a loosely bound layer is formed. Other techniques like microwave assisted method also used to enhance the adhesion process. In second step, MOFs layer is formed by secondary growth method on seeded substrate [45]. Dong et al grown UIO-66 on Titanium oxide substrate by situ method and moreover, membrane thickness is reduced 1µm by secondary growth method [46]. This method has a lot of benefits as it maintains structure and thickness of membrane, produces nano size crystals, due to division of nucleation and growth period it also reduces crystal synthesis time, accurately regulates the crystal growth and produces high quality crystals. Production of high quality and nano size seeds as these seeds must be able to cover the surface of substrate is the main challenge in secondary growth method.

4.4. Microwave induced thermal deposition method

In this method, microwave heating is used rather than traditional solvothermal heating. Jeong et al synthesized MOF5 membrane by microwave assisted method (Table 1) [33]. Wei et al synthesized UIO-66 membrane by decreasing the synthesis time and prepared the MOF membrane in just one-hour High temperature is required for their synthesis. In this method, Crystals formed on thin plate near the surface of substrate and to enhance the crystal growth and nucleation process, thermal conductivity is most important factor. A lot of substrates like Pd, graphite has high thermal conductivity. Main benefits of this method it takes less time for reaction and also accelerate nucleation power for synthesis of MOFs (Figure 2).

5. Applications

5.1. Removal of salts from sea water

Bivalent and monovalent ions can easily remove by MOFs base ceramic membranes. By situ growth method, Liu et al synthesized MOF Uio-66 membrane on Al₂O₃ substrate and ion removal rates of Ca, Mg, Al was 85.6%, 97.0%, 98.9% based on mechanism of molecular sieving. By solvothermal method, Cong et al synthesized 303 membrane on Al₂O₃ ceramic substrate, ion removal rate of MgCl₂, Na₂SO₄ were 94.5% and 97.0% based on electrostatic exclusion method [40]. The main problem is to remove monovalent ions from water as its difficult due to pressure driven filtration so MOFs based ceramic membranes efficiently removes monovalent ions. ZIF-8 MOFs on Al₂O₃ ceramic membranes was synthesized by Zhu et al through solvothermal method and it was experimentally proved that it removes salts and minerals from sea water and moreover, showed high stability in sea water. About 99.8% ions was removed by ZIF-8 membrane and water fluxes rate were 8, 1, 10.7, 13.6L per square meter per hour at 50, 70, 100-degree Celsius temperature [41]. Therefore, MOFs based ceramic membranes have high selectivity for multivalent ions but ion

removal rate relatively low for monovalent ions because of dehydration effect when molecules of water pass through nano size MOFs. Overall, effectively separation of salts by MOFs based ceramic membranes was observed (Table 2).

5.2. Separation of heavy metals

Industrial waste water contains heavy metals like lead, copper, nickel and zinc. These heavy metals effecting the both environment and human health. ZIF material have been used to remove heavy metals. It was observed by Wanqin Jin et al that due to high stability of ZIF-300 MOFs can be used to remove heavy metals when ZIF adsorbed on Aluminum oxide substrate and removal rate for copper sulphate was found to be 99.21% [39]. For the characterization of microstructure of membrane PAS (positron annihilation spectroscopy) was used. Growth of crystals depends on temperature, as temperature increases the growth of crystals also increases. So, it was observed that by increasing temperature from 80-120 pore size of membrane reduces from 3.5 to 3.11 angstrom. Size exclusion method was used to increase the removal rate of dyes and heavy metals. 92% lead was removed by ZIF membrane and it was observed by Mugahed Amran et al. So, studies showed that MOFs used to remove heavy metals and salt from waste water [47].

5.3. Separation of pollutants

Pollutants like dyes, fluorides and micropollutants present in environment also cause water pollution. Other traditional methods like size exclusion method, adsorption method is more convenient and efficient for removal of pollutants. Jinhui liv et al observed that Zr-MOF adsorbed on aluminum oxide substrate used for removal of fluoride ions and also studied that how concentration and flow rate effect the efficiency of removal of fluorides ions by dynamic filtration [48]. Characterization was done by FTIR and XPS. To increase the removal efficiency of pollutants, MOFs material was added. Xiao hu et al studied that Al MOF adsorbed on ceramic membrane have ion removal rate as high as 99.2% for 100mg Rhodamine B solution and this method also avoid the problem of preparing organic layer on inorganic substrate [49]. The connection between polyamide layer and ceramic substrate is increased by MOFs. Moreover, the nanofiltration performance of membrane also increased by using MOFs. ZIF-MOF adsorbed on composite membrane for removal of dye and separation of salts through combined methods situ and solvothermal was observed by Naixin Wang et al. It was studied that composite membrane have 18.9% salt removal rate and 98.2% drug dye removal rate [50]. so, in conclusion MOFs adsorbed on ceramic membrane used to remove pollutants and dyes from waste water.

6. Conclusion and Future outcomes

Ceramic membranes got much importance in separation of proteins and organic waste water treatment. Some problems regarding the ceramic membranes like membrane fouling could be solved by using nano materials as a separation layer. This review gives a detail explanation of synthesis methods and applications MOFs based ceramic membranes. Most stable MOFs like MOF-303 and Uio-66 much efficient in removing the salts and minerals from saline water. ZIF-67 on alumina membrane used efficiently for dye removal and heavy metal like Lead. Mechanism of separation is totally based on dehydration effect. Adsorption method is

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widely used for dye and heavy metal removal. In spite of high performance of MOFs based ceramic membranes it is important to solve the problem like low permeability and low anti-oxidant capability. For treatment of hypersaline water, high rejection, acid base ability all these show good performance but MOFs based ceramic membranes are developing now to be used for hypersaline treatment of water. Water long term stability and weak anti-oxidant problem need to be solved. For the formation of next level MOFs based ceramic it is important to understand the relationship between the physiochemical properties of MOFs pore structure and its separation. So, MOFs based ceramics used for hypersaline water treatment and applications such as ion separation and organic separation. Moreover, two important challenges that is economic and environmental friendliness for development of MOFs based ceramic membrane in near future should be given high priority.

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