



## Review

## Emerging adsorptive removal of azo dye by metal–organic frameworks

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## HIGHLIGHTS

- Clean water demand is one of the most important worldwide issues.
- MOFs show great attention for the improvement of adsorption/separation of azo dyes.
- The effectiveness of MOFs adsorbents toward dyes adsorption are reviewed.
- The adsorption mechanism and the applied kinetic and thermodynamic models are discussed.

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## ABSTRACT

Adsorptive removal of toxic compounds using advanced porous materials is one of the most attractive approaches. In recent years, the metal-organic frameworks (MOFs), a subset of advanced porous nanostructured materials, due to their unique characteristics are showing great promise for better adsorption/separation of various water contaminants. Given the importance of azo dye removal, as an important class of pollutants, this paper aims to review and summarize the recently published research on the effectiveness of various MOFs adsorbents under different physico-chemical process parameters in dyes adsorption. The effect of pH, the adsorption mechanism and the applicability of various adsorption kinetic and thermodynamic models are briefly discussed. Most of the results observed showed that the adsorption kinetic and isotherm of azo dyes onto the MOFs mostly followed the pseudo-second order and Langmuir models respectively. Also, the optimum pH value for the removal of majority of azo dyes by MOFs was observed to be in the range of ~5–7.

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## 1. Introduction

Water quality has deteriorated globally and provision of clean water is one of the most important worldwide issues. Nowadays, various toxic organic and inorganic compounds have been detected at critical levels in waste water, ground and surface waters (Bhatnagar et al., 2015). Azo dyes are an important class of pollutants in the effluents of the textile, food, leather, paper, plastic and cosmetic industries (Rafatullah et al., 2010). Globally, it has been estimated that  $2.8 \times 10^5$  tons of textile dyes are discharged in corresponding industrial effluents each year (Jin et al., 2007). Most industrial dyes are toxic, carcinogenic and teratogenic and the release of this huge quantity of these dyes into the ecosystem is harmful (Yagub et al., 2014).

There is therefore an urgent need to develop robust, economically feasible and environmentally friendly processes to remove them from wastewater. In this regard, several physico-chemical treatment technologies have been reported to control/minimize water pollution due to azo dyes (Ayati et al., 2014a, 2014b; Bhatnagar et al., 2015; Särkkä et al., 2015; Singh et al., 2015). The adsorption process has received considerable attention because of its convenience, ease of operation and simplicity of design (Sivashankar et al., 2014; Yagub et al., 2014; Kyzas and Matis, 2015; Tan et al., 2015; Tanhaei et al., 2015, 2016).

Metal organic frameworks (MOFs) are a new class of crystalline organic–inorganic hybrid solids which have been applied in various adsorption processes for hazardous materials removal. Several earlier review papers have summarized the adsorption applications of MOFs in pollutants removal from water (Canivet et al., 2014) and gas phase (Li et al., 2011; Uzun and Keskin, 2014). In the recent literature, Khan et al. have reviewed the adsorptive removal of various hazardous compounds mainly from fuel and water by modified MOF materials (Khan et al., 2013), while discussing the possible interactions between the active adsorption sites of the MOFs and adsorbates. Ahmad and Jhung also reviewed the various methods and paths for the preparation of MOFs composites for those which have been applied to gas and liquid phase adsorptions. Moreover, they summarized the adsorptive desulfurization and denitrogenation using MOFs, in their very recent work (Ahmed and Jhung, 2016). Elsewhere the plausible mechanisms for selective adsorption of water pollutants by MOFs were investigated comprehensively (Khan et al., 2013; Hasan and Jhung, 2015) showing that MOFs are capable of strong electrostatic interactions with the guest molecules.

Due to the importance of azo dye elimination, it is necessary to critically review their adsorptive removal by promising MOFs structures. This review article concerns the effectiveness of various MOFs adsorbents under different physiochemical process parameters for the removal of dyes from aqueous solution. A compilation of relevant published research with respect to adsorption kinetics, isotherm models and adsorption mechanisms under various process conditions is presented along with key findings.

## 2. History and background of MOFs

Metal–Organic Frameworks are a crystalline subset of microporous materials that have attracted the attention of many scientists around the world in recent decades (Langmi et al., 2014; Kumar et al., 2015; Li et al., 2016a; Venna and Carreon, 2015). Due to the lack of an accepted standard definition during the development of this new family of hybrid materials (Li et al., 2012), they are also known as porous coordination polymers (PCPs) (Kitagawa et al., 2004), porous polymer networks (PPNs) (Lu et al., 2010b), microporous coordination polymers (Cyshosz et al., 2008), iso-reticular MOFs (Kim et al., 2002), metal peptide frameworks

(MPF) (Manton et al., 2008) and zeolite-like MOFs (Liu et al., 2006).

Although some scientists believe that “until the mid-1990s, there were basically two types of porous materials, namely inorganic and carbon-based materials” (Kitagawa et al., 2004), the first preparation of porous coordination polymers, which nowadays are called Metal–Organic Frameworks, was introduced by Tomic in the mid-1960s (Tomic, 1965). The main interest in organic–inorganic hybrid materials arose in the early 1990s and the body of research on such types of porous structures increased considerably in the following. In 1990, Hoskins and Robson (Hoskins and Robson, 1990) prepared a new class of porous coordination polymer named scaffolding-like materials which was used in an anion-exchange process.

Four years later, Fujita et al. reported a two-dimensional square network material for catalytic purposes (Fujita et al., 1994). The definition of the new concept of MOFs and the systematic surveys and studies was pioneered by Yaghi and co-workers between 1995 and 1999 (Yaghi et al., 1995; Li et al., 1999). In 1995, they reported the synthesis of a novel MOF designed to bind aromatic guest molecules selectively (Yaghi et al., 1995). In late 1999, they were able to synthesize the first robust and highly porous structure of MOF-5 (also known as IRMOF-1) and investigated its x-ray single crystal structure as well as its low-temperature and low-pressure gas sorption properties (Li et al., 1999). It should be noted that between these years, other papers related to porous polymers frameworks were published, such as the work reported by Kitagawa's group in 1997 (Kondo et al., 1997), in which they measured the gas adsorption properties of CH<sub>4</sub>, N<sub>2</sub> and O<sub>2</sub> on their as-synthesized porous structure (Kondo et al., 1997). They found that the synthesized porous structure was able to reversibly adsorb these gases within a pressure range of 1–36 atm without crystal framework deformation. Today numerous valuable and informative reviews have been presented in fields of the PCP and MOF, which summarize the fast growing related-research efforts. Some of the most comprehensive ones are those by Kitagawa (Kitagawa et al., 2004), Yaghi (Rowell and Yaghi, 2004) and Férey (Férey, 2008).

Generally, MOFs consist of two inorganic units of metal ions/clusters and organic building units, such as carboxylates or other organic anions (phosphonate, sulphonate, and heterocyclic compounds), which are linked via coordination bonds (Férey, 2008; Lu et al., 2014).

They have some exceptional and unique properties, such as simple synthesis, extremely high internal surface area, high thermal and mechanical stabilities, low density, great micro pore volumes, permanent porosities and flexible frameworks that make them different from other conventional porous solids (Aguilera-Sigalat and Bradshaw, 2016). The properties of MOFs have been discussed at length in the reported review papers in recent years (Kitagawa et al., 2004; Férey and Mellot-Draznieks, 2005; Kitagawa and Matsuda, 2007; Férey, 2008; Qiu and Zhu, 2009; Sumida et al., 2012; Furukawa et al., 2013). MOF materials are linked via coordination bonds by the combination of metal ions or metal clusters (inorganic unit) and bridging ligands or linkers (Organic unit) (Kuppler et al., 2009; Furukawa et al., 2014; McGuire and Forgan, 2015). This combination is achieved via self-assembly to form one, two or three-dimensional networks with very different topologies (i.e., crystal structures, pore sizes and surface areas) (Cook et al., 2013; Wu and Navrotsky, 2015). The properties of MOFs can be easily systematically controlled to achieve the desired targets or improved performance by tuning their structures and functionalities. For instance, altering the connectivity of the inorganic moiety and the character of the organic linker may affect the surface area, shape and pore size (Jhung et al., 2012; Lu et al., 2014) or changing the organic linker can lead to increasing of the surface area (Cavka et al., 2008). On the other hand, because of the diversity of metal

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