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Hydrothermal synthesis of NH_2-UiO-66 and its application for adsorptive removal of dye

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ABSTRACT

In this research paper we report hydrothermal synthesis of NH_2-UiO-66, a metal organic framework (MOF) with zirconium as metal and amino terephthalic acid as a linker. The synthesized MOF was characterized by XRD, FTIR, SEM and BET surface area. As a potential application in water treatment, an adsorptive removal of safranin dye was studied using the synthesized material. The effect of initial concentration and pH of the dye solution was studied on the dye adsorption capacity of the material. An optimum set of conditions resulting into maximum dye adsorption was found out. The maximum adsorption capacity of the MOF was observed to be 390 mg/g at neutral pH of the solution and at room temperature. The experimental data was fitted with Langmuir, Freundlich and Temkin adsorption isotherm models. The kinetics of adsorption was studied using pseudo first order and pseudo second order model. The dye adsorption mechanism was also attempted.

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1. Introduction

Metal organic frameworks (MOFs) are a class of advanced porous, crystalline materials synthesized from organic linkers and transition metal ions (or clusters) [1]. These materials are receiving increasing scientific attention for variety of applications owing to their ease of synthesis, high surface area, high porosity, high thermal and mechanical stability, tunable structural and functional properties, etc. [1,2].

The MOFs have been extensively investigated as an adsorbent for the treatment of polluted water [3,4]. However, most of these materials are unstable in water and subject to degradation through ligands displacement or hydrolysis [4]. Among the reported MOFs, UiO-66 and NH_2-UiO-66 (UiO = University of Oslo) exhibit high stability in water and have excellent adsorption properties [4,5] because of which these are receiving considerable attention for adsorptive removal of water pollutants [4,6–8].

Residual synthetic dyes, in the effluent of textile, leather, paper, printing, cosmetics, petroleum, plastic, food, paint, rubber, and pharmaceutical industries are the major contributor to water pollution [9,10]. Many of these chemical pollutants are identified as mutagenic, carcinogenic and if consumed in excess quantity can cause severe damage to human health [11]. In addition, the water contaminated with these chemicals can be a potential threat to aquatic life [12]. Therefore, majority of the research in the field of water treatment is focused on removal of dye pollutants. Several conventional and advanced oxidation techniques are reported for the removal of these pollutants [13–16]. Among the available techniques, adsorption is an economical yet efficient method for the removal of variety of synthetic dyes [6,17]. In this context, a considerable amount of literature is available on variety of adsorbents including activated carbon, diatomaceous earth, resins, zeolites, carbon nanotubes, etc. In general, a good adsorbent should possesses high surface area, high porosity, ease of synthesis, less cost, high thermal and mechanical properties, regeneration ability and high adsorption capacity towards a wide range of pollutants within a short contact time [6,18]. Since UiO-66, NH_2-UiO-66 and few other MOFs satisfy most of the above requirements; these are considered to be potential alternative adsorbents for the treatment of dye polluted water.

Chen and coworker studied the adsorptive removal of various anionic and cationic dyes using UiO-66 and NH_2-UiO-66 MOF [6]. They observed superior adsorption of cationic dyes on to the NH_2-UiO-66 compared to UiO-66. A microwave-assisted synthesis

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of UiO-66 and its application for the adsorptive removal of acid chrome blue K is reported by Li and coworkers [8]. He and coworker reported adsorptive removal of Rhodamine B on UiO-66 [19]. They observed increase in the adsorption capacity with temperature and the maximum dye adsorption capacity was observed to be about 75 mg/g at 323 K. They also studied the regeneration of the spent adsorbent and demonstrated reuse of the material for six cycles of sorption/desorption. In an another study, an adsorptive behavior of UiO-66 for the removal of acid orange 7 (AO7) dye was investigated and a maximum adsorption capacity of about 358 mg/g was observed at 318 K [20]. Recently, Embaby et al. studied adsorption of a series of anionic (Alizarin Red S, Eosin, Fuchsin Acid and Methyl Orange) and cationic (Neutral Red, Fuchsin Basic, Methylene Blue and Safranine T) dyes onto the UiO-66 and reported a record maximum adsorption capacity of 400 mg/g for adsorption of Alizarin Red S dye [21]. In literature, most of the studies are performed with UiO-66 however, application of NH_2-UiO-66 for the adsorptive removal of dyes is least investigated.

The synthesis of MOFs is mainly reported by hydrothermal and microwave methods [6,8,19]. The hydrothermal method, under elevated temperature and pressure conditions, ensures complete solubilization of the organic molecules used as linker. These experimental conditions also help to overcome the activation energy required for intermediate metastable phases of synthesis and therefore, the synthesized material becomes highly crystalline [22,23]. The hydrothermally synthesized of UiO-66 family MOFs has been well reported in the literature. However, its application for dye adsorption is least investigated. For example, Shen and coworkers reported application of hydrothermally synthesis of NH_2-UiO-66 for photocatalytic applications [24]. Luu and coworkers studied the application of NH_2-UiO-66 for adsorption of CO_2 and CH_4 [25]. The only study available in the literature on application of hydrothermally synthesized NH_2-UiO-66 for dye adsorption is reported by Chen and coworker [6]. In the above work, they reported adsorptive removal of three cationic dyes (Methylene Blue, Rhodamine B and Neutral Red) and two anionic dyes (Methyl Orange and Acid Chrome Blue K) using UiO-66 and NH_2-UiO-66. They observed preferential adsorption for cationic dye onto the MOFs. However, in the above work, the experiments were carried out at 20 ppm and the equilibrium dye adsorption capacity is reported as 96 mg/g for adsorption of methylene blue dye. The authors (Chen and coworkers) have not studied the effect of initial dye concentration or pH and there was no attempt of optimizing the experimental conditions.

In the present study, we report hydrothermal synthesis of NH_2-UiO-66 and its application for the removal of cationic Safranin dye. The effect of initial concentration and pH of the dye solution was studied on the dye adsorption capacity of the material. The experimental data was fitted with Langmuir, Freundlich and Temkin adsorption isotherm models. The dye adsorption mechanism was also attempted.

2. Experimental

2.1. Chemicals and materials

All the chemicals used in the experiments were of analytical grade and were used without any further purification. The chemicals used were Zirconium chloride (S.D. Fine Chemicals Limited, Mumbai), 2-amino terephthalic acid (Sigma Aldrich), N,N-dimethyl formamide (DMF) (S.D. Fine Chemicals Limited, Mumbai) and methanol (S.D. Fine Chemicals Limited, Mumbai). deionized (DI) water was used to prepare all the solutions.

2.2. Synthesis of NH_2-UiO-66

In a typical synthesis of NH_2-UiO-66, 2.17 g of 2-amino terephthalic acid and 3.8 g of Zirconium chloride was added to 36 mL of DMF. The mixture was stirred for 30 min. It was then transferred to a stainless steel teflon-lined autoclave and maintained at 120 °C for 24 h. The solid thus obtained was washed first with DMF and then repeatedly with methanol. Finally, the as-obtained solids was dried at room temperature and further in an oven kept at 150 °C for 4 h.

2.3. Characterization of the synthesized material

The synthesized material, NH_2-UiO-66, was characterized by XRD, FTIR, SEM, Zeta potential analysis and BET surface area. A powder X-ray diffraction pattern was recorded on a Bruker D8 Advance X-ray diffractometer (Bruker, USA), with Cu Kα radiation in a range of 10–90° at a scanning speed of 0.2 s/0.02°. The crystallite size was calculated using Scherrer's calculator. An FTIR spectrum was recorded on a Spectrum Two™ spectrometer (Perkin Elmer, USA) in the range of 4000 cm^{-1} to 500 cm^{-1}. The surface morphology of the as-synthesized material was studied by a scanning electron micrograph taken on a JEOL JSM-6380 SEM instrument (JEOL, Japan). Zeta potential analysis of the synthesized materials was performed using a Zetasizer instrument (Malvern Instruments, UK). The BET surface area was measured by nitrogen adsorption-desorption isotherm using a PMI BET sorptometer (BET-201AEL-2OSEL, USA).

2.4. Batch adsorption experiments

Batch adsorption studies were performed using the synthesized material for the removal of Safranin dye from aqueous solution. Initially, a stock solution (1000 mL) of 1000 ppm of Safranin dye in DI water was prepared. A required concentration of the solution was prepared by diluting the stock solution. The observed error in the initial dye concentration of the solution was ±2 ppm. To this solution, a pre-weighed amount of adsorbent was added and the suspension was stirred on a magnetic stirrer at room temperature conditions. For analysis, samples were withdrawn for the solution at a regular interval of time. The samples were centrifuged in a Optima™ MAX-XP centrifuge, (Beckman Coulter, USA) in order to remove the adsorbent material and absorbance of the supernatant solution was noted using a UV–Vis spectrophotometer (Carry 50 UV–Vis, USA). The concentration of the unknown solution was then determined with the help of a pre-plotted calibration curve between the concentration of known solutions and their corresponding absorbance values. The experiments were continued until a constant concentration of the dye was observed at least in 5 successive readings. This constant concentration was attributed to equilibrium concentration.

The amount of dye adsorbed on the adsorbent at different time, t, is represented by q_t and calculated by:

$$q_t = (C_0 - C_t) * \frac{V}{W} \quad (1)$$

where C_o is the initial concentration (in mg/L) of dye, C_t is the concentration (in mg/L) of dye at time, t, V is the volume (in L) of the solution and W is the amount (in g) of adsorbent.

Further, the amount of adsorption at equilibrium, q_e (mg/g), was calculated by:

$$q_e = (C_0 - C_e) * \frac{V}{W} \quad (2)$$

where C_e is the equilibrium concentration (in mg/L) of dye. All other quantities on right side of the above equation have same meaning as that in Eq. (1).

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