### Accepted Manuscript

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PII: S1385-8947(17)31598-X

DOI: http://dx.doi.org/10.1016/j.cej.2017.09.099

Reference: CEJ 17684

To appear in: Chemical Engineering Journal

Received Date: 14 July 2017

Revised Date: 15 September 2017 Accepted Date: 15 September 2017



Please cite this article as: Q. Yang, S. Ren, Q. Zhao, R. Lu, H. Cheng, Z. Chen, H. Zheng, Selective separation of methyl orange from water using magnetic ZIF-67 composites, *Chemical Engineering Journal* (2017), doi: http://dx.doi.org/10.1016/j.cej.2017.09.099

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## **ACCEPTED MANUSCRIPT**

# Selective separation of methyl orange from water using magnetic ZIF-67 composites

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#### **ABSTRACT:**

Efficacious and convenient removal of organic dye contaminants from wastewater is a challenge for public health and ecosystem protection. Here we fabricate a novel type of Fe<sub>3</sub>O<sub>4</sub>@MOFs (Metal-organic frameworks) magnetic porous composite materials. ZIF-67 (Zeolitic imidazolate framework-67) nano-crystals as an attractive subfamily of MOF was selected to fabricate Fe<sub>3</sub>O<sub>4</sub>-PSS@ZIF-67 composites (defined as MZIF-67). MZIF-67 composites are core-shell structure, for which the aggregation core of Fe<sub>3</sub>O<sub>4</sub> nanoparticles is coated with petal-like ZIF-67 crystals, in which Co<sup>2+</sup> firstly combines with  $SO_3^{2-}$  provided by PSS (poly (styrenesulfonate, sodium salt)) to form nucleation. MZIF-67 composites perform well on methyl orange (MO) adsorption, which could be attributed to the highly porosity and the nature of Lewis base of coordinated Co<sup>2+</sup> centrals. The results show that the equilibrium adsorption capacity for MO is up to 738 mg·g<sup>-1</sup> (when pH =8.0, contact time is 7 h, adsorbent dose is 5 mg and initial MO concentration is 400 mg·L<sup>-1</sup>). In addition, MZIF-67 composites could selectively separate MO from the mixture solution of MO and MB (methylene blue). The removal rate of MO is up to 92%. The concentration ratio of MO/MB is 0.04. And the separation efficiency is up to 96%. The results suggest MZIF-67 composites could be a good candidate for

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