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Easy-separative MoS_2-glue sponges with high-efficient dye adsorption and excellent reusability for convenient water treatment

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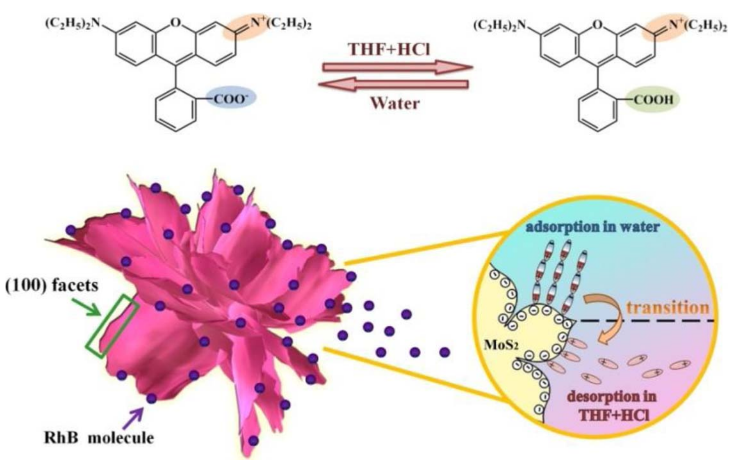
GRAPHICAL ABSTRACT



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ABSTRACT

This study demonstrated high-efficient dye adsorption for wastewater using MoS_2-glue sponge as an easy-separative and regenerative adsorbent. Flower-like molybdenum disulfide (MoS_2) nanopowders with rich polar edge sites were firstly synthesized by a facile one-pot hydrothermal method. Chemical and morphological features of MoS_2 nanoflowers were then examined by XRD, XPS, SEM and TEM. Afterwards, different concentrations of the MoS_2-glue sponges with easy separability were successfully fabricated by a convenient two-step dip-coating method. The adsorption performance was estimated by the removal of Rhodamine B (RhB) dyes. The results proved that the maximum adsorption capacity of MoS_2-glue sponge was 127.39 mg RhB/g product in 60 min. The RhB uptake behaviors can be well described by the Sips isotherm model and pseudo-second-order kinetic model, indicating a multilayer and chemical adsorption process. The adsorption mechanism proposed that the zwitterionic RhB molecules in aqueous solution would aggregate on the surface of negative charged MoS_2 in a head-to-tail way through electrostatic interaction. Furthermore, RhB dyes could desorb easily from MoS_2-glue sponges when changing the polarity of solvent and the reusability experiment showed that the MoS_2-

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3% glue sponge could be reused for 15 times with above 85.8% of the adsorption capacity retained. Thus, the novel MoS_2-glue sponge is considered as a competitive adsorbent for treatment of dyestuff waste water.

1. Introduction

The entire world is nowadays facing drinking water shortage due to the pollution in water and the unreasonable discharge of industrial contamination. Waste water with hazardous organic dyes (such as Rhodamine B (RhB), methylene blue (MB), methyl orange (MO), congo red and so on) from textiles, printing, pulp and paper industries poses a giant threat to human health and eco-environment, which may further leak in the ground water [1,2]. To remove organic dyes from wastewater and maintain the drinking water supply allow of no delay. Currently, several methods have been developed to deal with dye-polluted water, such as adsorption, photo-degradation, chemical oxidation, membrane filtration, biochemical method and so on [3–8]. Among all the proposed techniques, adsorption is considered as the most convenient, inexpensive and efficient approach that can remove organic dyes in a large scale. As is well known, an ideal adsorbent material should exhibit outstanding properties including large adsorption capacity, fast adsorption rate, easy separation from the suspension and recollection, simple desorption procedure and excellent reusability. As shown in Table 1, various functional materials have been used as adsorbents to remove organic dyes from wastewater. For example, commercial activated carbon, a traditional powder adsorbent, possessed a large adsorption capacity of 765.6 mg/g for MB dyes. However, it took a contact time over 1800 min, showing a time-consuming process [9]. Other adsorbents, like tungstate oxide nanourchins [10], hypercrosslinked polymer [11] and reduced graphene oxide-ferrite hybrids [12], needed a shorter adsorption time, but they had generally limited adsorption capacities only 20–80 mg/g toward RhB. Core-shell Fe_3O_4@ mesoporous carbon nanoparticles exhibited a fast adsorption rate for RhB dye within 30 min, as well as a satisfying adsorption capacity of 198.9 mg/g, but at the same time an insufficient reusability where it failed to reuse after five cycles [13]. Productions of these materials above, however, are high cost with complicated processing and difficult to scale up for practical use. Moreover, some environmentally unfriendly powder sorbents dispersed in water may cause secondary pollution and affect the cell proliferation to some extent due to the difficulty of sorbent recollection and material losses during each cycle [14,15]. The traditional methods such as centrifugation and filtration are tedious to separate adsorbents from the suspension completely. Thus, the development of cheaper, more eco-friendly, efficient and reusable adsorbents is a subject of intensive research.

Molybdenum disulfide (MoS_2), an emerging graphene-like nanomaterial, is recently gaining significant interest due to its fascinating sandwiched S-Mo-S lamellar layers structure [16]. Furthermore, at the edges of lamellar layers along the in-plane direction, MoS_2 possesses strong polar Mo-S covalent bonds, which may attract polar species and lead to a tendency of adsorption [17–19]. He et al. [20] synthesized layered porous MoS_2 nanosheets and used them to remove RhB dyes from aqueous solution. The sample exhibited fast adsorption for dye removal and could reach a maximum adsorption capacity of 163 mg/g at 420 min for RhB, which can further reveal the feasibility of MoS_2 sorbent for dye adsorption. Recently, Han et al. [21] demonstrated that the prepared flower-like MoS_2 nanostructure had negative surface charge, and it showed more prominent adsorption of cationic dyes (eg. RhB, MB) compared with anodic dyes (eg. MO). What's more, the RhB-adsorbed MoS_2 can be easily regenerated through washing with alkaline solution, showing a promising water treatment application. For collection purpose, magnetic materials, such as Ni and Fe_3O_4, have been introduced into MoS_2 nanopowders to overcome the disadvantage of nonmagnetic materials in separation. Ni/MoS_2 and Fe_3O_4/MoS_2 nanocomposites with superior magnetic sensitivity were successfully prepared and they could be separated from the suspension easily by applying an external magnetic field, although their adsorption capacities toward RhB were limited (15.7 and 22.0 mg/g respectively) [22,23]. Therefore, it is of great significance to further enhance the adsorption capacity and adsorption rate of MoS_2 adsorbent toward organic dyes, explore the adsorption and desorption mechanism, and more importantly, improve the separation method and reusability.

Herein, MoS_2 nanoflowers were first synthesized via a facile one-pot hydrothermal method, where high concentration of the starting materials and excessive sulfur source to the stoichiometric ratio were employed. Hydrothermal reaction with high concentration and disproportionality of the precursors are more favorable to increase the defect richness of MoS_2 nanoflowers (such as edge dislocation and petal curve), which may lead to an increase in the exposed active edge sites [17,24,25]. Afterward, we fabricated an easy-separative, highly efficient and recyclable MoS_2-glue sponge adsorbent via a convenient and economic two-step dip-coating method, which can be mass-produced and remove organic dyes in a large scale. Commercial melamine-formaldehyde sponge (denoted MF sponge) with a three-dimensional interconnected macroporous structure was used as a framework for MoS_2 nanoflowers coating and 901 building glue solution was chosen as the adhesive to reinforce the adhesion of MoS_2 nanoflowers to MF sponges frame. Subsequently, the adsorption isotherms and kinetic studies of RhB, a representative cationic dye pollutant, adsorption onto MoS_2-glue sponges were investigated in detail. Furthermore, excellent cycle performance can be obtained through the polarity-triggered reversible adsorption/desorption procedure [24]. As for separation process, the sponge adsorbent can be taken out from dye solution simply just using a pair of tweezers instead of any tedious centrifugation or filtration process. The results demonstrate that the novel MoS_2-glue sponge can be used as a desirable sorbent for the primary or secondary wastewater treatment of the dyestuff waste water remediation in the future, which would greatly increase the available domestic water supply [26].

Table 1
Previously reported adsorption properties of various functional adsorbents.

Adsorbents	Dyes	Adsorption capacity (mg/g)	Adsorption time (min)	Cycle times	References
Commercial activated carbon	MB	765.6	1800	3	[9]
Tungstate oxide nanourchins	RhB	81.78	720	–	[10]
Hypercrosslinked polymer	RhB	57	600	–	[11]
Reduced graphene oxide-ferrite hybrids	RhB	23	400	–	[12]
Core-shell Fe_3O_4@mesoporous carbon	RhB	198.9	30	5	[13]
MoS_2 nanosheets	RhB	163.0	420	–	[20]
Flower-like MoS_2 nanostructure	RhB	55	180	–	[21]
Ni/MoS_2 nanocomposites	RhB	15.7	35	–	[22]
Fe_3O_4/MoS_2 nanocomposites	RhB	22	30	–	[23]