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Modifying MIL-100(Fe) through co-growing iron oxides to enhance its adsorption for selenite

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1. Introduction

Selenium (Se) is an essential micronutrient for animals and humans, and there is a very narrow range between deficient and toxic levels of selenium in mammals (Fernández-Martínze, 2009). High concentration of Se in water is the main cause of selenosis. Adsorption is an effective technology to deal with the Se-contaminated water. The most important issue of adsorption technology is improving the adsorption capacity and rate of adsorbent. Recently, metal-organic framework (MOF) has attracted wide attention due to its special structure of abundant Lewis and Brönsted acid sites and opening pore (Jiang, 2015), which make MOF an ideal adsorbent for the environmental remediation. However, the reported MOF can not exhibit its most potential on the adsorption for Se (Wang, 2019). Therefore, modification is an alternative strategy to improve the adsorption performance of MOF towards Se-contaminated water. This study explored the composite of MOF@iron oxides and investigated its adsorption performance for the selenite (Se(Ⅳ)).

2. Materials and Methods

The composites was fabricated through in-situ co-growing iron oxides with MIL-100(Fe), a kind of MOFs. Adsorption of Se(Ⅳ) on those composites were investigated based on the batch experiments of isothermal, kinetic and thermodynamic adsorption, as well as the influence of pH.

3. Results

3.1 Crystal structure

XRD patterns suggested that iron oxides in the composites prepared at 70 ℃ and 98 ℃ were akaganeite and hematite, respectively. The pore of MIL-100(Fe) was filled by the co-grown akaganeite, while the hematite prefered to co-exist with MIL-100(Fe).

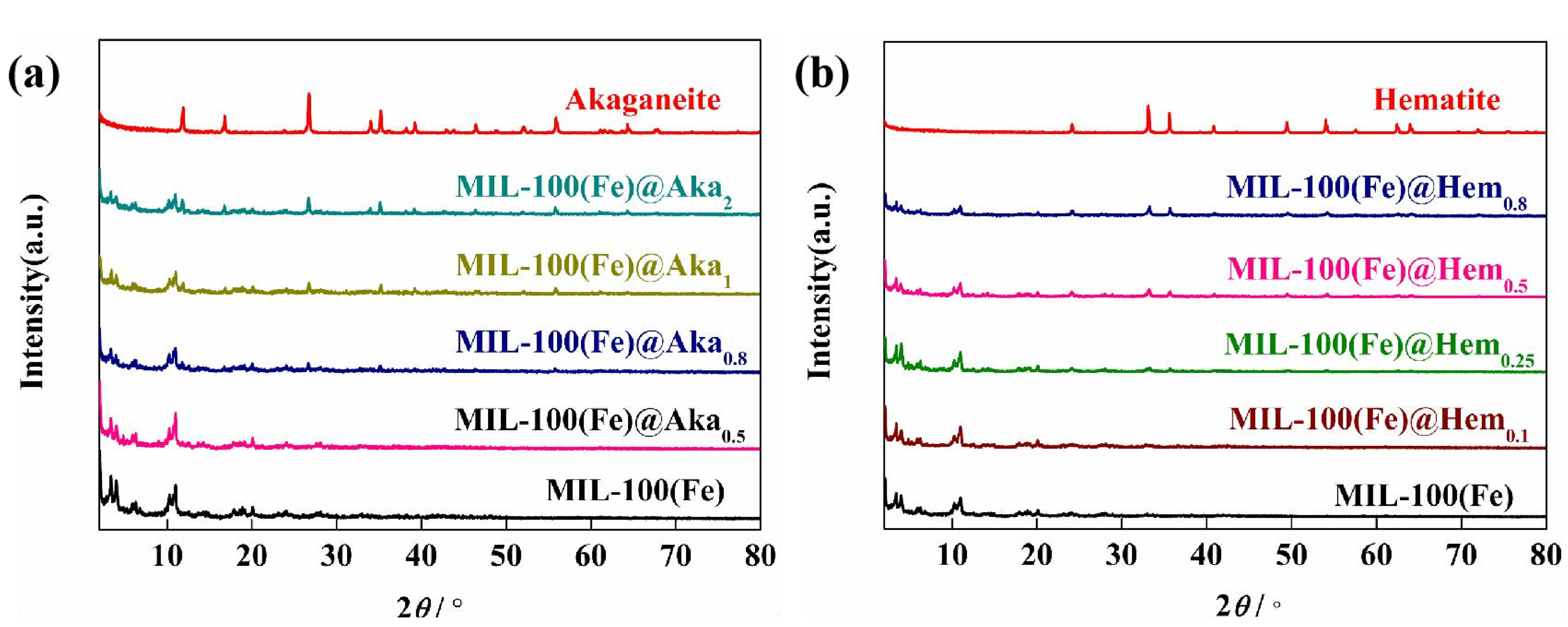

Fig.1 X-ray diffraction patterns of as-prepared materials

3.2 Component and pore structure

The component of those composites revealed by TG analysis and pore parameters listed in Table 1. Results indicated that the co-growing of iron oxides with MIL-100(Fe) in-situ decreased the surface area and increased the active sites of MIL-100(Fe).

Table 1 Component and pore parameters of as-prepared materials

Sample	BET surface area ($m^2 \cdot g^{-1}$)	Pore volume ($cm^3 \cdot g^{-1}$)	Theoretical pore volume ($cm^3 \cdot g^{-1}$)	Filling degree of pore (%)	Content of akaganeite or hematite (%)
MIL-100(Fe)	1490	1.156	1.156	0	0
Akaganeite	41	0.264	0.264	0	100
MIL-100(Fe)@$Aka_{0.5}$	1208	0.599	1.148	48	0.9
MIL-100(Fe)@$Aka_{0.8}$	1282	0.779	0.988	21	18.8
MIL-100(Fe)@Aka_1	1217	0.733	0.934	22	24.9
MIL-100(Fe)@Aka_2	1177	0.857	0.851	0	34.2
Hematite	4	0.160	0.160	0	100
MIL-100(Fe)@$Hem_{0.1}$	1536	0.991	1.094	10	6.2
MIL-100(Fe)@$Hem_{0.25}$	1422	0.991	0.998	1	15.9
MIL-100(Fe)@$Hem_{0.5}$	1180	0.752	0.909	17	24.8
MIL-100(Fe)@$Hem_{0.8}$	1013	0.722	0.722	0	43.7

3.3 Surface property

Zeta potential (Fig. 2) associated with TG analysis suggested that the main active site of MIL-100(Fe)@Aka was Lewis acid, while it was Brönsted acid for MIL-100(Fe)@Hem.

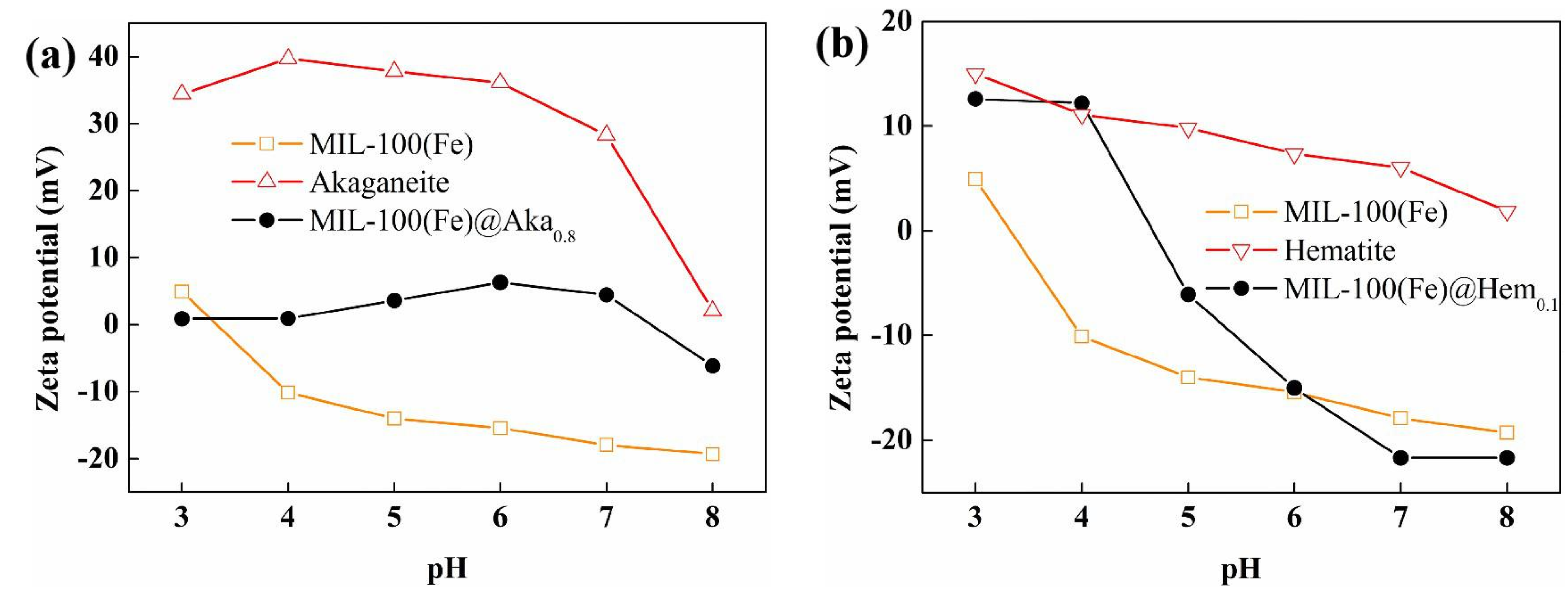

Fig. 2 Zeta potentials aganist pH for the as-prepared materials

3.4 Adsorption performance

There was a synergistic effect on the adsorption for Se(Ⅳ) by the composites of MIL-100(Fe)@Aka and MIL-100(Fe)@Hem (Fig. 3). The most synergistic effect on the adsorption for Se(Ⅳ) was obtained using MIL-100(Fe)@$Aka_{0.8}$ and MIL-100(Fe)@$Hem_{0.8}$ with an enhanced coefficient of 154% and 153%, respectively.

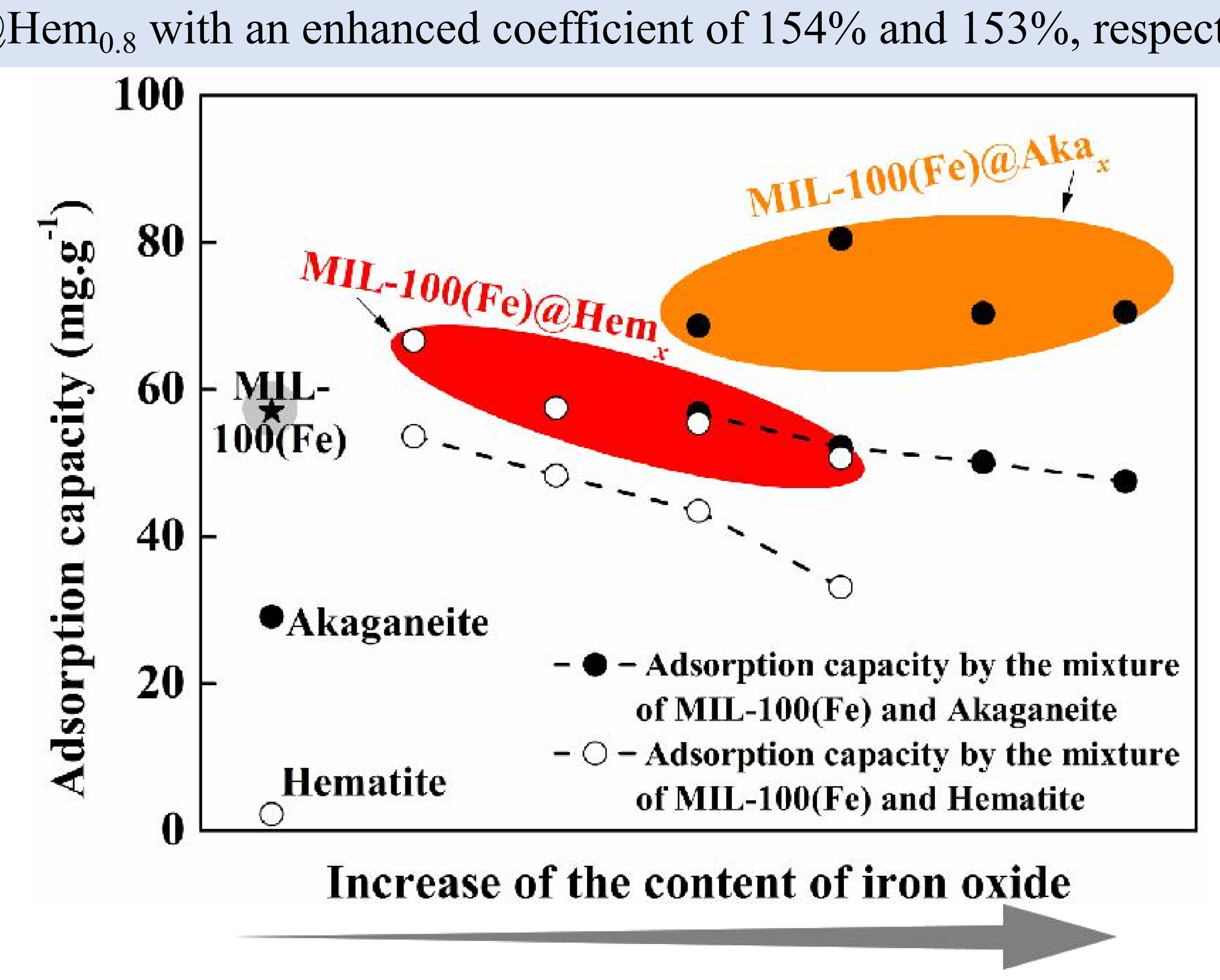

Fig. 3 Adsorption capacities of the as-prepared composites and the mixture of MIL-100(Fe) and akaganeite/hematite at the initial Se(Ⅳ) concentration of 200 mg/L

3.5 Isothermal adsorption

Fig. 4 showed the isothermal adsorption of Se(Ⅳ) and its fitting curves. MIL-100(Fe)@$Hem_{0.1}$ exhibited the maximum adsorption capacity of 301.93 mg/g fitted by Sips model among the reported adsorbents.

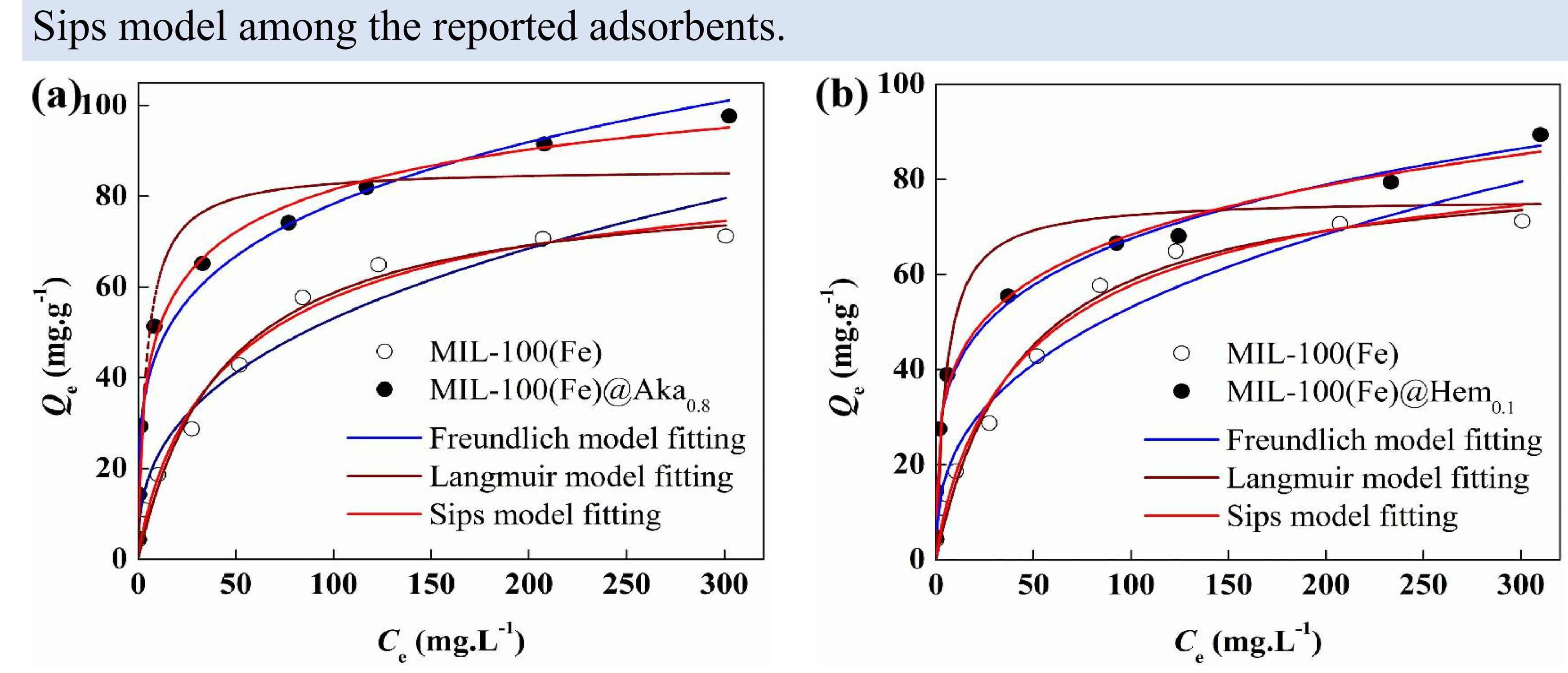

Fig. 4 Isothermal adsorption and fitting curves for Se(Ⅳ) by the as-prepared materials

4. Conclusion

Composites of MIL-100(Fe) was fabricated through co-growing iron oxides in-situ. The main active sites of MIL-100(Fe)@Aka and MIL-100(Fe)@Hem were Lewis acid and Brönsted acid, respectively. Both MIL-100(Fe)@Aka and MIL-100(Fe)@Hem exhibited synergistic effect on the adsorption for Se(Ⅳ). MIL-100(Fe)@$Hem_{0.8}$ possessed a very high adsorption capacity for Se(Ⅳ).

5. Acknowledgment

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6. References

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