Supporting information

Removal of arsenic from smelting wastewater using Fe_3O_4 as an situ Fe source: the effect of pre-dissolution and the evolution process of scorodite

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Number of pages: S1

Number of figures: 4

S1. Temperature dependence

The reaction temperature affects the scorodite synthesis. Combined with previous studies, we took the temperature from 23°C to 90 °C. According to Fig. S1a-c, with the increase of reaction temperature, the residual As concentration in the solution gradually decreases, the As removal rate gradually increases, and the toxic leaching gradually decreases. When the reaction temperature was 90 °C, the As removal rate reached up to 99.1%. At the same time, scorodite was generated when the reaction temperature was ≥80°C (Fig. S1d). At the same time, according to the research of Cai et al.[1], when the temperature was >90°C, the As removal rate will not change greatly. Therefore, finally, we choose the reaction temperature of 90°C as the best experimental condition.

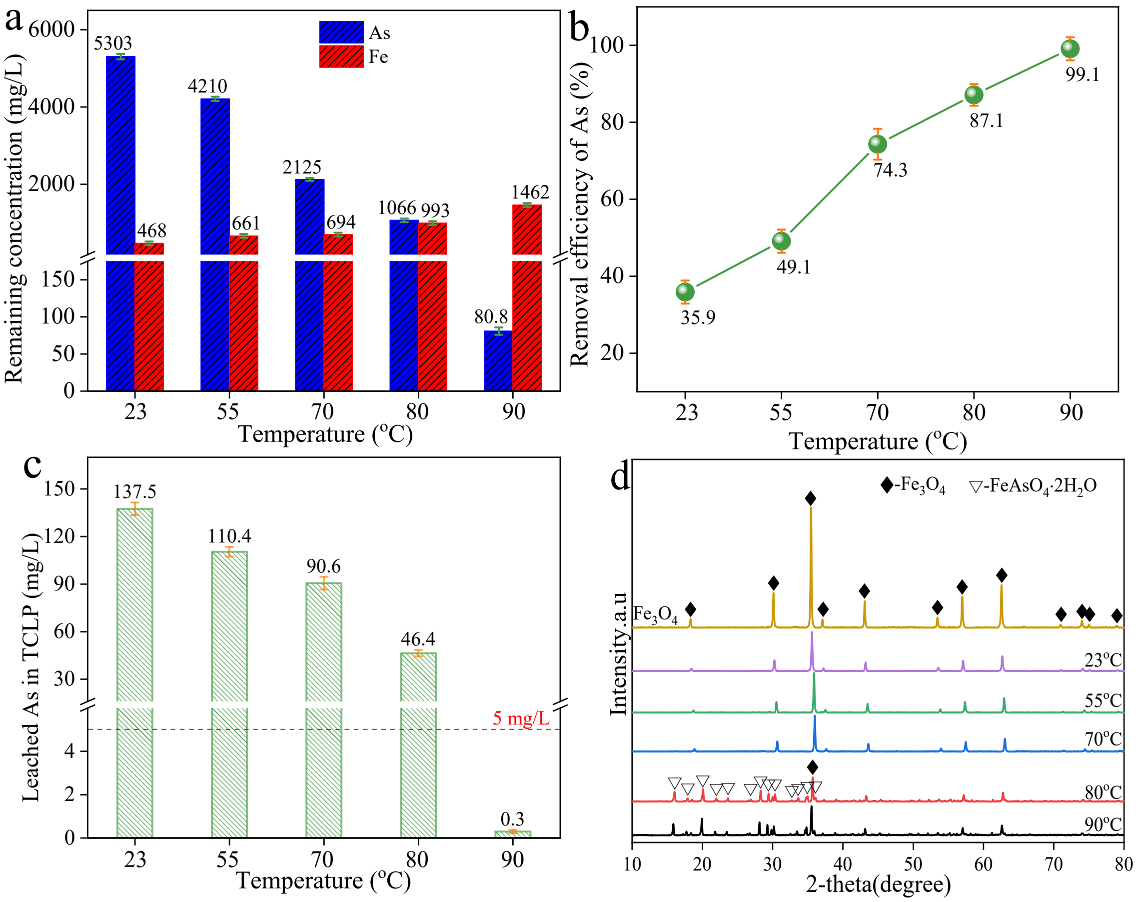

Fig. S1. (a) As and Fe concentrations in the residual solution, and (b) removal rate of

As, (c) TCLP (the dashed red line is the national standard), (d) XRD patterns of the precipitates at different temperatures (Experimental conditions: predissolution time=2 h; Fe/As molar rate=3:1; reaction time=24 h; pH values=2.5).

Our proposed process was compared with others reported in literature, as shown in Table S1. Generally, high temperature was required for the scorodite synthesis. In the reaction of liquid iron source, it was difficult to control a low Fe and As supersaturation to produce high-crystallization scorodite. Compared with the liquid iron source process, our process can overcome these shortcomings. The release of Fe ion from solid iron source could react with As(V) in the form of scorodite. Meanwhile, the Fe_3O_4 could neutralize H^+ in the wastewater, providing a stable pH for the crystallization of scorodite. Additionally, the As removal rate of Fe_3O_4 was higher and less toxic leaching than that of limonite.

Table S1 Comparison in terms of operational parameters between this study and previously available studies.

Mothods	wastewater composition	Iron sources	Reaction condition	As removal efficiency	Residual As concentration	TCLP (mg/L)
Liquid iron source[2]	pH=0.7 CAs=25000 mg/L	$Fe(NO_3)_3$	pH=0.73 Fe/As=1 T=160 °C Time=24 h	92	2000 mg/L	＜5
Solid iron source[3]	pH=0.9 C_{As}=10300	Limonite	pH=1.5 Fe/As=4	96.2	389 mg/L	2.3

	mg/L		T=90 ^oC Time=12 h			
Our proposed process	pH=0.8 C_{As}=10300 mg/L	Fe_3O_4	pH=2.5 Fe/As=3 T=90 ^oC Time=24 h	99.1	80.8 mg/L	0.3

References

[1] G. Cai, X. Zhu, K. Li, X. Qi, Y. Wei, H. Wang, F. Hao, Self-enhanced and efficient removal of arsenic from waste acid using magnetite as an in situ iron donator, Water Res, 157 (2019) 269-280.

[2] G.P. Demopoulos, D.J. Droppert, G.V. Weert, Precipitation of crystalline scorodite (FeAsO4 · 2H2O) from chloride solutions, Hydrometallurgy, 38 (1995) 245-261.

[3] X. Li, G. Cai, Y. Li, X. Zhu, X. Qi, X. Zhang, B. Shu, K. Li, Y. Wei, H. Wang, Limonite as a source of solid iron in the crystallization of scorodite aiming at arsenic removal from smelting wastewater, Journal of Cleaner Production, 278 (2021) 123552.