

Supplementary Materials

S1. As(III) removal by different materials

Ye^[1] et al. used chemical modification to obtain functionalized CNTs and NMCNTs-PANI. The results showed that the maximum adsorption capacity was 14.80 mg/g at the experimental conditions of As(III) initial concentration of 10 mg/L, 25°C and t=35 min. Additionally, in this work, As(III) was mainly removed from the aqueous solution by adsorption without being changed into other less toxic compounds. Sherlala^[2] et al. synthesized CMGO nanocomposite for arsenic adsorption with high specific surface area (152.38 m²/g) and excellent saturation magnetization (49.30 emu/g). And the highest adsorption capacity (45 mg/g) and removal efficiency (61%) were obtained at pH 7.3. In comparison to this study, the maximum removal could reach 333.56 mg/g at 25°C and pH=5.0, which represented a significant increase over both economically and operationally.

Table S1 Comparison of As(III) removal performance of nZVI/MXene@CNTs and other materials

Materials	Experimental conditions	q _e (mg/g)	References
NMCNTs-PANI	C ₀ =10mg/L, 25°C, t=35min	14.80	[1]
CMGO	60 °C, pH=10	63.31	[2]
nZVI/MXene@CNTs	25 °C, pH=5	333.56	

S2. Adsorption kinetics, isotherms and thermodynamics

S2.1. Adsorption kinetics

In a 150 mL conical flask, 0.02 g of nZVI/MXene@CNTs and a 50 mL 200 mg/L As(III) solution were added, and the reaction was carried out at pH=5.0, 25 °C and 150

rpm. The As(III) content was measured at each time point, respectively. The adsorption process and mechanism of As(III) by nZVI/MXene@CNTs were determined by analytical fitting of the obtained data using pseudo-first-order and pseudo-second-order kinetic models(eq. S1 and S2).

$$\ln (q_e - q_t) = \ln q_e - k_1 t \quad (S1)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (S2)$$

Where, k_1 and k_2 are the pseudo-first-order and pseudo-second-order kinetic models adsorption equilibrium constants, respectively; q_t and q_e (mg/g) are the adsorption capacities of nZVI/MXene@CNTs on As(III) at a specific and equilibrium reaction time; t is the reaction time.

S2.2. Adsorption isotherms

In a 150 mL conical flask, 0.02 g of nZVI/MXene@CNTs and the 50 mL different initial concentration of As(III) solution were added, and the reaction was carried out at pH=5.0, 25 °C and 150 rpm. The content of As(III) was measured after 120 min of reaction. The adsorption process and mechanism of As(III) by nZVI/MXene@CNTs were further demonstrated using the fitted data via Langmuir(eq. S3) and Freundlich isothermal adsorption models(eq. S4)

$$q_e = \frac{q_m C_e K_L}{1 + K_L C_e} \quad (S3)$$

$$q_e = K_F C_e^{\frac{1}{n}} \quad (S4)$$

Where, K_L and K_F are the Langmuir and Freundlich adsorption constants, respectively; C_e (mg/L) is the concentration of As(III) at equilibrium; q_e (mg/g) is the adsorption amount corresponding to C_e ; and q_m (mg/g) is the theoretical maximum adsorption amount obtained from the two adsorption isotherm models.

S2.3. Adsorption thermodynamics

The three thermodynamic parameters, Gibbs free energy change (G^0), entropy change (S^0), and enthalpy change (H^0), are determined from the fitting and analysis of

the adsorption isotherm, and then k_d and the thermodynamic parameters are calculated (Eq. S5, S6).

$$\Delta G^0 = -RT \ln k_d \quad (S5)$$

$$\ln k_d = \frac{\Delta S^0}{R} - \frac{\Delta H^0}{RT} \quad (S6)$$

Where, ΔG^0 (KJ/mol) is the Gibbs free energy change; R (8.314 J/(mol·K)) is the thermodynamic model constant; T (K) is the absolute temperature; k_d (L/g) is the thermodynamic equilibrium constant

Table S2 The fitting parameters of pseudo-first-order and pseudo-second-order kinetic models for As(III) adsorption by nZVI/MXene@CNTs

Temperature (°C)	Pseudo-first-order kinetic model			Pseudo-second-order kinetic model		
	q_{cal} (mg/g)	k_1 (min ⁻¹)	R^2	q_{cal} (mg/g)	K (g/(mg·min))	R^2
15	317.48	0.022	0.9876	403.58	5.39×10^{-5}	0.9880
25	336.27	0.030	0.9922	408.19	7.93×10^{-5}	0.9942
35	342.93	0.035	0.9828	405.96	1.01×10^{-4}	0.9893

Table S3 The fitting parameters of Langmuir and Freundlich isotherm models for As(III) adsorption by nZVI/MXene@CNTs

Temperature (°C)	Langmuir isotherm models			Freundlich isotherm models		
	q_m (mg/g)	b(L/mg)	R^2	k_f (mg/g)	n	R^2
15	309.19	0.37	0.9857	127.19	5.26	0.9319
25	336.64	0.41	0.9771	146.38	5.47	0.9267
35	338.06	1.08	0.9756	173.18	6.33	0.9200

Table S4 The fitting parameters of thermodynamic model for As(III) adsorption by

nZVI/MXene@CNTs

The As(III) initial concentration(mg/L)	ΔH^0 (KJ/mol)	ΔS^0 (J/(mol·K))	ΔG^0 (kJ/mol)		
			15°C	25°C	35°C
25	94.31	367.65	-11.63	-15.30	-18.98
50	65.66	262.14	-9.88	-12.50	-15.12
75	38.77	164.53	-8.64	-10.28	-11.93
100	32.20	139.82	-8.09	-9.49	-10.89
150	25.92	106.99	-4.91	-5.98	-7.05
200	13.26	57.44	-3.29	-3.87	-4.44
300	8.47	33.92	-1.30	-1.64	-1.98

References

1. Ye, X.; Ren, G. Adsorption of As(III) from aqueous solution by amidated/oxidized carbon nanotube-polyaniline. *Chin. J. Environme. Eng.* **2019**, *13*, 2798 – 2807.
2. Sherlala, A.I.A.; Raman, A.A.A.; Bello, M.M.; Buthiyappan, A. Adsorption of arsenic using chitosan magnetic graphene oxide nanocomposite. *J. Environ. Manag.* **2019**, *246*, 547–556.