Efficient As(III) Removal by Novel MoS₂-Impregnated Fe-Oxide–Biochar Composites: Characterization and Mechanisms

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1. INTRODUCTION

Concentrations of heavy metal(loid)s are increasing in aquatic environments. Among these, arsenic is of significant concern worldwide owing to its high toxicity, bioaccumulation, and environmental persistence. Increasing As concentrations in water occur due to natural weathering; geochemical reactions; use of agricultural pesticides, wood preservatives, and desiccants; metal-ore smelting; leaching of man-made arsenic compounds; gold mining; volcanic eruptions; combustion of fossil fuels; and biological and numerous anthropogenic activities. The World Health Organization (WHO) has set 10.0 μg L⁻¹ of As as the maximum safe limit for drinking water.

In nature, As exists in two valence states: arsenite [As(III)] and arsenate [As(V)]. Arsenite is more toxic, highly mobile, and more difficult to eliminate from an aqueous solution than As(V), owing to its nonionic H₃AsO₃ species and its low affinity for many adsorbents. The high risk of environmental contamination means that more effective methods of eliminating As(III) must be developed urgently. Adsorption is the most efficient method for removing As(III) because of its high efficacy, low cost, and simple ease of implementation. Different materials have been developed for removing As from aqueous solutions. For example, metal (hydr)oxides (especially iron (hydr)oxides) and activated carbons can effectively remove As from aqueous systems. Although metal–organic frameworks such as nanospheres, graphene–oxide composites, and carbon nanotubes have shown effective adsorption capacities for the removal of As, such materials are relatively expensive and some even show cytotoxicity, which limits their applications.

The use of biochar (BC), produced through oxygen-free pyrolysis of organic and agricultural-waste materials, offers an innovative and effective approach to eliminating As from aquatic systems. Because of its negative charge, good physicochemical properties, high stability, and large surface area, BCs could be used as an active adsorbent for many toxic heavy metal(loid)s. It has been found that BCs impregnated with iron (hydr)oxide particles shows high sorption capacities for removing As from aqueous solutions. However, more
research is needed to develop innovative materials to improve the physicochemical characteristics of BC-based adsorbents and increase their sorption capacities.

In the past few years, molybdenum disulfide (MoS\textsubscript{2}), a two-dimensional dichalcogenide, has shown a wide range of mechanical, physicochemical, electrical, and catalytic applications\textsuperscript{16} owing to its momentous charge-transfer potential and active sulfur functional groups.\textsuperscript{17} However, few studies on the use of MoS\textsubscript{2} to remove toxic metal(loid)s from aquatic environments are available in the literature,\textsuperscript{18–20} although Zhang et al. explored the electrocatalytic properties of Fe\textsubscript{3}O\textsubscript{4}/MoS\textsubscript{2},\textsuperscript{17} which enhanced the removal of metal(loid)s. It is expected that MoS\textsubscript{2}-functionalized iron-based BCs could synergistically improve As(III)-removal efficiency via As-S interactions.\textsuperscript{18} In this way, a new class of adsorbents could be developed by combining MoS\textsubscript{2} with iron-based BCs for potential environmental remediation applications. To the best of our knowledge, the synthesis of MoS\textsubscript{2}-impregnated iron-based BCs and characterization of their synergistic effects on As sorption have yet to be accomplished.

With this in mind, we developed a strategy for increasing As(III)-sorption capacity by hydrothermally impregnating iron-based BCs with MoS\textsubscript{2}. The present study provides new insights into the removal of As(III) from aqueous systems and into the application of MoS\textsubscript{2}-impregnated iron-based BCs to environmental decontamination.

### 2. RESULTS AND DISCUSSION

The morphologies of the prepared adsorbents were explored by scanning electron microscopy (SEM). As shown in Figure 1, the morphologies of Fe\textsubscript{3}O\textsubscript{4}@BC600 and Fe\textsubscript{3}O\textsubscript{4}@BC800 were different from those of BC600 and BC800. Figure 1c,d clearly shows that Fe\textsubscript{3}O\textsubscript{4}@BC600 consisted of spherical-like nano-particles, while the morphology of Fe\textsubscript{3}O\textsubscript{4}@BC600 was different from that of Fe\textsubscript{3}O\textsubscript{4}@BC800. MS@BC800 showed flowerlike structures grown on the BC surface (Figure 1e). The morphologies of MSF@BC600 and MSF@BC800 were significantly different from those of Fe\textsubscript{3}O\textsubscript{4}@BC600 and Fe\textsubscript{3}O\textsubscript{4}@BC800, suggesting that MoS\textsubscript{2} had concentrated on the FeO\textsubscript{3}@BC600 and FeO\textsubscript{3}@BC800 surfaces (Figure 1f,g).

The X-ray diffraction (XRD) spectra generated for Fe\textsubscript{3}O\textsubscript{4}@BC800, MS@BC800, and MSF@BC800 are shown in Figure 2a, while those of Fe\textsubscript{3}O\textsubscript{4}@BC600 and MSF@BC600 are presented in Figure S1a. Diffraction peaks at 31.5, 35.7, 43.8, 51.8, and 62.8° could be indexed to the (220), (311), (400), (422), and (440) planes of Fe\textsubscript{3}O\textsubscript{4} (JCPDS no. 19-0629), respectively, which is in line with previously published findings.\textsuperscript{21} Peaks at 49.3, 64.8, and 74.38° could be indexed to the (024), (300), and (220) planes of Fe\textsubscript{3}O\textsubscript{4} (JCPDS no. 33-0664), respectively. The diffraction peaks at 14.2, 33.0, 39.6, 47.6, and 58.0° in MS@BC800 indicate a hexagonal phase and could be indexed to the (002), (100), (103), (105), and (110) planes of MoS\textsubscript{2} (JCPDS no. 037-1492), respectively. The presence of the (002) plane strongly suggests that MoS\textsubscript{2} had been concentrated on the BC surface.\textsuperscript{22} Furthermore, the broad band at 20–30° in the XRD spectrum for MS@BC800 indicates that some of the BC remained amorphous. Comparing the XRD spectra for Fe\textsubscript{3}O\textsubscript{4}@BC800 and MS@BC800, four new weak peaks appeared at 37.0, 38.9, 53.3, and 56.2° in the spectrum for MS@BC800, while some Fe\textsubscript{3}O\textsubscript{4}@BC800-related peaks were lost or became weaker, suggesting that chemical reactions had occurred during composite preparation.\textsuperscript{23} Overall, the XRD results suggest that novel-adsorbent composites had been formed.

To examine the surface functionalities of the prepared adsorbents, Fourier transform infrared (FTIR) spectroscopy was used. FTIR spectra generated for BC800, Fe\textsubscript{3}O\textsubscript{4}@BC800, MS@BC800, and MSF@BC800 are presented in Figure 2b, while those for BC600, Fe\textsubscript{3}O\textsubscript{4}@BC600, and MSF@BC600 are presented in Figure S1b. The spectrum for BC800 showed bands around 1026 and 1586 cm\textsuperscript{-1}. After Fe modification, the spectrum for Fe\textsubscript{3}O\textsubscript{4}@BC800 showed new peaks at 564, 1393, 1465, 2918, and 3422 cm\textsuperscript{-1}. After MoS\textsubscript{2} modification, the spectrum for MS@BC800 showed new peaks around 471, 1112, 1401, 2923, and 3417 cm\textsuperscript{-1}. After Fe\textsubscript{3}O\textsubscript{4}@BC800 was modified with MoS\textsubscript{2}, the spectrum for MSF@BC800 presented new peaks at 468 and 1101 cm\textsuperscript{-1}, while the peak at 1465 cm\textsuperscript{-1} disappeared.

Broad peaks at 3426 cm\textsuperscript{-1} in the spectra of all the modified adsorbents were assigned to hydroxyl (–OH) group vibrations,\textsuperscript{25} while peaks at 2966–2853 and 1393–1402 cm\textsuperscript{-1} resulted from –CH\textsubscript{2} stretching vibrations.\textsuperscript{26,27} Peaks at around 1585 cm\textsuperscript{-1} corresponded to carboxyl (C=O) functional groups,\textsuperscript{28} and after adsorbent modification, spectra for all the modified adsorbents showed slightly shifted carboxyl peaks, indicating that chemical reactions occurred on the BC800 surface during modification.\textsuperscript{29} Sharp peaks at 1026–

![Figure 1](https://dx.doi.org/10.1021/acsomega.0c01268)

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1101 cm\(^{-1}\) mainly were attributed to C–O stretching vibrations and OH bending modes of carboxylic groups and alcohols. Peaks at 564–601 cm\(^{-1}\) were attributed to the Fe–O stretching vibration, while peaks at 468–471 cm\(^{-1}\) were attributed to S–Mo stretching. FTIR results showed that FeO\(_x\) and MoS\(_2\) both had concentrated on the BC surface, thereby intensifying the functional-group peaks and introducing new functional groups to the BC surface, which could potentially act as new active sites to improve sorption.

X-ray photoelectron spectroscopy (XPS) was conducted to further analyze the chemical composition of MSF@BC800 (Figure S2), and the spectrum generated for MSF@BC800 showed peaks such as S 2p, Mo 3d, O 1s, and Fe 2p (Figure S2). Figure S2b presents the Fe 2p XPS spectrum of MSF@
BC800, showing binding energy peaks attributed to Fe 2p3/2 at about 710.2 and 711.6 eV for Fe2+ and Fe3+ transitions, respectively.33 A peak corresponding to Fe 2p1/2 and a satellite peak characteristic of Fe2+ and Fe3+ appeared at 724.8 and 717.98 eV, respectively.

The O 1s spectrum of MSF@BC800 (Figure S2c) was deconvoluted into four peaks assigned to various oxygen-containing compounds, suggesting the presence of inorganic oxygen (in iron oxides) and organic oxygen (bound in alkoxyl, carbonyl, and carboxyl groups).33 The peaks with binding energies of 530.4 and 531.5 eV probably originated from metal–O and metal–OH bonds, respectively, while binding energies of 532.9 and 532.2 eV represented C–O bonds in C=O and O–C≡O, respectively.34 The Mo 3d and S 2s spectra for MSF@BC800 (Figure S2d) were deconvoluted into three characteristic peaks at 227, 231.7, and 234.8 eV, which were attributed to S 2p3/2, Mo 3d5/2, and Mo 3d3/2, respectively.20 As presented in Figure S2e, the S 2p spectrum could be deconvoluted into three peaks at 167.5, 163, and 161.8 eV, which were attributed to S(VI), S(II) (S 2p3/2), and S 2p1/2, respectively.35 The spectra for MSF@BC600 was deconvoluted into Fe 2p, O 1s, Mo 3d, and S 2p peaks, as presented in Figure S3, and the MSF@BC600-associated binding energies fluctuated more than the MSF@BC800-associated ones.

The N2 adsorption and desorption isotherms generated for BC800, FeO@BC800, and MSF@BC800 are presented in Figure 2c. MSF@BC800 exhibited a type (IV) isotherm, indicating the presence of micropores and mesopores.36 The Barrett–Joyner–Halenda (BJH) pore size distributions of BC800, FeO@BC800, and MSF@BC800 are shown in Figure 2d–f. Isotherms for FeO@BC800 and MSF@BC800 showed major pore-size distributions and sharp peaks at 2–10 nm, indicating the presence of mesopores. The total pore volume (cm3 g−1), mean pore size (nm), and specific surface area (m2 g−1) were 0.236, 3.54, and 266.87 for FeO@BC800 and 0.151, 5.08, and 104.2 for MSF@BC800, respectively (Table S1). Clearly, the presence of Fe particles increased the adsorbent surface area. However, loading MoS2 particles on the BC surface blocked the micropores, thereby remarkably decreasing the specific surface area, suggesting that the specific surface area is not a key factor in As(III) adsorption. Furthermore, the N2 adsorption isotherms of BC800 and FeO@BC800 increased more rapidly than the MSF@BC800 isotherm near P/P0 = 1, indicating that BC800 and FeO@BC800 surfaces had more macropores than MSF@BC800 (Figure 2c).37

MSF@BC800 mass loss and thermal stability were determined by thermogravimetric analysis (TGA) in N2 at 25–800 °C (Figure 3). MSF@BC800 weight loss increased with increasing temperature from 30 to 800 °C, and the corresponding mass loss was calculated as 29.4%. Thermal decomposition of MSF@BC800 showed four stages: the first resulted in 5.6% weight loss owing to evaporation of physically adsorbed water; weight loss in the second stage (7.8%) was mainly ascribed to lattice-water loss; the weight decreased by 11.3% in the third stage, possibly owing to decomposition of some MSF@BC800 organic ligands;38 and finally the weight decreased by 4.7% in the fourth stage, possibly because of C–O bond breakage38 and FeOx and MoS2 loading.

2.1. Sorption Efficiency. Adsorption experiments were performed in batches. Industrial concentrations [5, 10, and 20 mg L−1 (ppm)] of As(III) were used for As(III) sorption efficiency and adsorbent affinity tests, which were performed after adsorbents had been in contact with As(III) for 5 h.

Herein, adsorbent affinities for As(III) are represented in terms of distribution coefficient Kd (defined in the Supplementary Information). The results demonstrated that MSF@BC800 displayed superior As(III) affinity and removal ability (Table 1). For initial As(III) concentrations of 5, 10, and 20 mg L−1, MSF@BC800 removed 99.98, >99.73, and ≥94.27% As(III) and showed Kd ≈ 106, 105, and 104 mL g−1, respectively. More importantly, when the initial As(III) concentration was 5 mg L−1, As(III) concentration decreased to 1 μg L−1, which is below the WHO safety limit established for drinking water.39

Typically, adsorbents are considered excellent if they show Kd ≥ 104 mL g−1.18 MSF@BC800 displayed good affinity for As(III) because Kd was 105–106 mL g−1. The results show that MoS2 and FeOx had certain synergistic effects and demonstrated that MSF@BC800 was an excellent filter for purifying As-contaminated water.

2.2. Sorption Kinetics. With the exception of MSF@BC800, the sorbents showed fast sorption during the first hour and then slowed between the first and second hours owing to particle diffusion into pores, before gradually reaching equilibrium within 3.5 h (Figure 4a). Meanwhile, MSF@BC800 required longer (about 4 h) to reach equilibrium. Therefore, the maximum contact time for the rest of the experiments was fixed at 5 h.

Pseudo-first- and second-order parameters for fitting As(III) sorptions are listed in Table S2. Pseudo-first-order coefficients (R2 = 0.933–0.993) were lower than pseudo-second-order coefficients (R2 = 0.988–0.997) for all adsorbents, demonstrating that the pseudo-second-order model was more suitable for As(III) adsorption. Thus, overall adsorption might be governed by electron sharing or exchange between the sorbent and sorbate, which may indicate that chemisorption was the rate-limiting step.40,41

Intraparticle diffusion for As(III) uptake is plotted as a function of t0.5 in Figure 4b, and the associated parameters for MSF@BC600 and MSF@BC800 are presented in Table S2. Intraparticle diffusion showed multilinearity during adsorption, indicating that adsorption was affected by at least two steps for MSF@BC600, and three steps for MSF@BC800. The results showed that some other mechanisms alongside pore diffusion were involved in determining the rate-limiting step, because the straight line did not pass through the origin (i.e., C ≠ 0).
The plot shown in Figure 4b initially increases sharply, indicating electrostatic attraction at the external surface. This was followed by a second stage wherein intraparticle diffusion (i.e., particle diffusion from the external surface into macro-pores and mesopores) began, resulting in chemisorption characteristics and plateau formation extending to equilibrium, representing micropore diffusion.37,42 MSF@BC800 displayed dramatically higher sorption capacity than the other adsorbents, which could be due to the synergistic effects of FeO\textsubscript{x} and MoS\textsubscript{2} on the MSF@BC800 surface, as both FeO\textsubscript{x} and MoS\textsubscript{2} have excellent charge-transfer capacities.17 Furthermore, the dramatically higher sorption capacity might be due to the presence of plentiful FeO\textsubscript{x} nanoparticles growing in situ on the MoS\textsubscript{2}-impregnated MSF@BC800 surface.22 The S content decreased after adsorption, as shown in Table S1, suggesting that S helps to generate more Fe−OH sites for As sorption by activating Fe atoms.43

### 2.3. Adsorption Isotherms

The maximum sorption capacities were measured using sorption isotherms. Sorption saturation curves were plotted for each adsorbent as a function of As(III) adsorption, as shown in Figure 5a; ascending curve portions indicate effective electrostatic interactions between As(III) and adsorbents, while relatively flat regions at high \( C_e \) show adsorption-site saturation.44 MSF@BC800 exhibited high As(III) adsorption and removal and a high distribution coefficient \( K_d \) for an initial As(III) concentration of 10 mg L\textsuperscript{−1} (Table 2).

Fitting parameters for Langmuir and Freundlich models are given in Table 2. Results showed that the Langmuir model (\( R^2 = 0.982−0.994 \)) was the best fit for the experimental data obtained for BC600, BC800, FeO\textsubscript{x}@BC600, and FeO\textsubscript{x}@BC800, indicating monolayer adsorption on adsorbents showing uniform sorption energies. On the other hand, MoS\textsubscript{2}-modified adsorbents (MS@BC800, MSF@BC600, and

### Table 1. Removal % and Affinities of Different Adsorbents toward As(III)\textsuperscript{a}

| adsorbents   | \( C_0 \) (mg g\textsuperscript{−1}) | \( C_f \) (mg g\textsuperscript{−1}) | removal % | \( Q_m \) (mg g\textsuperscript{−1}) | \( K_d \) (mL g\textsuperscript{−1}) |
|--------------|---------------------------------------|--------------------------------------|------------|------------------------------------|---------------------------------|
| BC600        | 4.68                                  | 3.9651                               | 15.28      | 0.72                               | 180                             |
|              | 8.52                                  | 7.58428                              | 10.98      | 0.94                               | 123                             |
|              | 17.94                                 | 16.4237                              | 8.45       | 1.52                               | 92                              |
| BC800        | 4.68                                  | 3.51                                 | 25.08      | 0.44                               | 334                             |
|              | 8.52                                  | 7.23                                 | 15.16      | 0.91                               | 178                             |
|              | 17.94                                 | 16.47                                | 8.16       | 1.36                               | 89                              |
| FeO\textsubscript{x}@BC600 | 4.39                                  | 2.67                                 | 39.08      | 1.71                               | 641                             |
|              | 9.75                                  | 7.12                                 | 26.96      | 2.63                               | 369                             |
|              | 18.53                                 | 14.33                                | 22.65      | 4.19                               | 292                             |
| FeO\textsubscript{x}@BC800 | 4.88                                  | 2.69                                 | 44.98      | 2.19                               | 817                             |
|              | 8.93                                  | 5.62                                 | 37.07      | 3.31                               | 589                             |
|              | 19.37                                 | 14.26                                | 26.38      | 5.11                               | 358                             |
| MS@BC800     | 4.72                                  | 3.64                                 | 22.83      | 1.08                               | 295                             |
|              | 9.07                                  | 7.45                                 | 17.86      | 1.62                               | 217                             |
|              | 18.72                                 | 16.61                                | 11.28      | 2.11                               | 127                             |
| MSF@BC600    | 4.72                                  | 0.36                                 | 92.27      | 4.36                               | 1.1 \times 10\textsuperscript{4} |
|              | 9.11                                  | 1.17                                 | 87.17      | 7.94                               | 6.8 \times 10\textsuperscript{3} |
|              | 18.23                                 | 5.39                                 | 70.58      | 12.83                              | 2.3 \times 10\textsuperscript{3} |
| MSF@BC800    | 4.68                                  | 0.001                                | 99.98      | 4.68                               | 4.6 \times 10\textsuperscript{3} |
|              | 8.52                                  | 0.03                                 | 99.73      | 8.49                               | 2.8 \times 10\textsuperscript{3} |
|              | 17.94                                 | 1.026                                | 94.27      | 16.91                              | 1.6 \times 10\textsuperscript{4} |

\textsuperscript{a}m = 0.02 g, \( V = 20 \) mL.

**Figure 4.** (a) Adsorption kinetics of unmodified and modified adsorbents and (b) intraparticle diffusion of MSF@BC600 and MSF@BC800.
MSF@BC800) followed the Freundlich model ($R^2 = 0.961 - 0.988$) rather than the Langmuir one ($R^2 = 0.947 - 0.984$), signifying heterogeneously adsorbent surfaces or pores and multilayer coverage, indicating chemisorption properties.45

The As(III) adsorption capacities and affinities of MoS2-impregnated iron-based BCs (MSF@BC600 and MSF@BC800) were remarkably higher than those of MS@BC800, FeOx@BC600, and FeOx@BC800 (Table 2), which might be related to additional functional groups inducing the formation of As-MoS2 complexes and to synergistic effects of MoS2 and FeOx on the MSF@BC600 and MSF@BC800 surfaces, both of which showed high charge-transfer capacities.17,22

Figure 5. (a) Adsorption isotherms and (b) Temkin models of As(III) on unmodified and modified adsorbents.

Table 2. As(III) Adsorption Parameters of Different Adsorbents

| adsorbents   | removal % | $K_d$ (mL g$^{-1}$) | $Q_{max}$ (mg g$^{-1}$) | $K_L$ (L mg$^{-1}$) | $R^2$ | $K_f$ (mL$^3$ g$^{-1}$)$^{1/n}$ | $R^2$ | $b_T$ (J mol$^{-1}$) | $A_T$ (L mg$^{-1}$) | $B_T$ | $R^2$ |
|--------------|-----------|----------------------|--------------------------|--------------------|-------|-------------------------------|-------|---------------------|---------------------|-------|-------|
| BC600        | 10.98     | 123                  | 2.6734                   | 0.0883             | 0.936 | 0.4425                        | 0.409 | 0.977               | 5352                | 1.894 | 0.463 | 0.932 |
| BC800        | 15.16     | 178                  | 2.6772                   | 0.0631             | 0.985 | 0.3048                        | 0.484 | 0.974               | 5243                | 1.260 | 0.472 | 0.955 |
| FeOx@BC600   | 26.96     | 369                  | 6.9229                   | 0.1049             | 0.980 | 1.4805                        | 0.354 | 0.973               | 2407                | 4.194 | 1.029 | 0.921 |
| FeOx@BC800   | 37.07     | 589                  | 7.6794                   | 0.1424             | 0.967 | 1.8196                        | 0.336 | 0.981               | 2312                | 7.884 | 1.071 | 0.926 |
| MS@BC600     | 17.86     | 217                  | 5.7551                   | 0.0395             | 0.977 | 0.4783                        | 0.532 | 0.991               | 2675                | 1.083 | 0.926 | 0.929 |
| MS@BC800     | 87.17     | $6.8 \times 10^3$    | 17.299                   | 0.7888             | 0.985 | 6.6481                        | 0.268 | 0.947               | 1032                | 31.64 | 2.400 | 0.983 |
| MSF@BC600    | 97.23     | $3.5 \times 10^4$    | 28.434                   | 1.6464             | 0.968 | 13.568                        | 0.215 | 0.973               | 772.6               | 250.02 | 2.375 | 0.871 |
| MSF@BC800    | 97.23     | $3.5 \times 10^4$    | 28.434                   | 1.6464             | 0.968 | 13.568                        | 0.215 | 0.973               | 772.6               | 250.02 | 2.375 | 0.871 |

$^a$As(III) concentration, 10 mg L$^{-1}$. pH = 6, and temperature = 25 °C.

Figure 6. (a) Effect of pH on As(III) adsorption by MSF@BC800 and (b) pH changes after adsorption.
Table 3. Thermodynamic Parameters for the Adsorption of As(III) by MSF@BC800 at Different Temperatures

| T (K) | Qe (mg g⁻¹) | lnKc | ΔG° (kJ mol⁻¹) | ΔH° (kJ mol⁻¹) | ΔS° (kJ mol⁻¹ K⁻¹) |
|-------|-------------|------|---------------|---------------|-------------------|
| 288   | 124.99      | 7.874 | -27.413       | 18.965        | 0.1611            |
| 298   | 129.64      | 8.219 | -29.044       |               |                   |
| 308   | 132.62      | 8.667 | -30.6334      |               |                   |

Figure 7. (a) Zeta potential of MSF@BC800 and (b) FTIR of MSF@BC800 before and after As(III) adsorption.

2.5. Effect of Solution pH on As(III) Sorption. Generally, pH is the most important variable that impacts adsorption, affecting adsorbent characteristics such as surface charge and As speciation. The influence of pH across the 3.0–8.0 range on As(III) sorption on MSF@BC800 is shown in Figure 6a. As(III) sorption decreased with increasing pH, demonstrating that As(III) sorption on MSF@BC800 was pH-dependent and confirming that electrostatic interactions played a decisive role in adsorption. Similar pH effects previously have been reported for other adsorbents. Decreasing As(III) adsorption at higher pH values might be due to the predominance of negatively charged sites on the MSF@BC800 surface causing a repulsion effect. Furthermore, the zeta potential was studied at different pH values to further elucidate the effect of pH on As(III) sorption. The point of zero charge, pH_PZC of MSF@BC800 was 4.83 (Figure 7a), meaning that MSF@BC800 was negatively charged when pH > 4.88. The negative surface charge was due to deprotonation of O-, Fe- and S-containing functional groups with increasing pH, which repelled As(III). When solution pH < pH_PZC, the MSF@BC800 surface was positively charged owing to protonation by H₃O⁺, hence enabling reaction via electrostatic attraction. This could be responsible for the increasing As(III) sorption capacity of MSF@BC800 with decreasing pH, as more positively charged sites would enable electrostatic attraction between As anions and the adsorbent surface as well as complexation between H₂AsO₄⁻ and MSF@BC800 hydroxyl groups, leading to improved adsorption in the low pH range. Furthermore, we measured the final solution pH for different initial pH values. For initial pH values of 3.0 and 4.0, the final pH increased after sorption (Figure 6b), suggesting that –OH groups were released into the solution by arsenate or arsenite through ligand exchange during sorption. The final pH was lower than the initial value when the initial pH ≥ 5.0, probably owing to the release of H₂O⁺ during sorption after formation of Fe–...
O–As complexes. Similar changes in pH have previously been reported for other adsorbents.\textsuperscript{52} However, Chen et al. found that for pH < 9, As(III) was mainly removed as $\text{H}_2\text{AsO}_3^-$ while $\text{H}_3\text{AsO}_3$ formed.\textsuperscript{53} As(III) removal directly involves As(III) sorption, oxidation of As(III) to As(V), and subsequent As(V) sorption. Therefore, oxidation and electrostatic interactions both contributed to As(III) sorption.

**2.6. Effect of Ionic Strength on As(III) Sorption.** To investigate the effect of ionic strength on As(III) adsorption using MSF@BC800, NaNO\textsubscript{3} solutions containing various nitrate concentrations (0.001–0.1 M) similar to those usually found in natural surface waters and groundwater were used. Figure S4b clearly shows that As(III)-removal capacity increased remarkably as NaNO\textsubscript{3} concentration was increased from 0.001 to 0.01 M.\textsuperscript{54} The current system did not show competitive adsorption for As(III) on MSF@BC800, which is consistent with the inner-sphere surface-coordination complexation (i.e., strong bonding) mechanism previously reported.\textsuperscript{8,55} As(III) adsorption slightly decreased as NaNO\textsubscript{3} concentration was increased further from 0.01 to 0.1 M, indicating outer-
sphere surface-coordination complexation.  

Therefore, the results suggest that although outer- and inner-sphere mechanisms coexisted, inner-sphere surface-coordination complexation was the dominant mechanism.

### 2.7. Effect of HA Concentration on As(III) Sorption.

Being one of the major components of the natural organic matter, HA affects heavy-metal adsorption.  

We investigated the impact of HA concentrations of 0–30 mg L⁻¹ on As(III) adsorption using MSF@BC800 and found that HA concentration had a considerable effect on As(III) sorption (Figure S4c).  

As(III) adsorption was enhanced by 10 mg L⁻¹ of HA compared to the control (0 mg L⁻¹ of HA), possibly by providing some adsorption sites via HA-induced changes in solution pH. However, higher HA concentrations (≥ 20 mg L⁻¹) decreased As(III) adsorption, probably because numerous MSF@BC800 surface-active sites were occupied by HA.

### 2.8. Adsorption Mechanisms.

Adsorption occurred via multiple mechanisms, including electrostatic interactions, surface complexation, and chemisorption, all of which acted as As(III)-removal mechanisms. To further elucidate the adsorption mechanism, FTIR and XPS measurements were conducted after As(III) adsorption on MSF@BC800 (Figures 7b and 8).  

Comparison of FTIR spectra generated for MSF@BC800 before and after sorption revealed that MSF@BC800-As(III) showed remarkable changes (Figure 7b). Peaks attributed to hydroxyl and C−H groups shifted slightly after adsorption, indicating that hydroxyl and C−H groups had participated in the adsorption of As(III). Peaks relating to other functional groups also shifted, including those of C=O, −CH, −OH and C−O groups, which were at 1585, 1402, and 1026–1101 cm⁻¹, respectively, after adsorption, implying possible surface-complexation mechanisms consistent with chemisorption. Furthermore, the intensities of the −CH, −OH, and C=O peaks increased, indicating that additional −CH, −OH, and C=O groups were introduced after As(III) adsorption; thus, while a portion of the −OH, −CH, and C=O groups were used to form As complexes, arsenic introduced more groups in MSF@BC800−As(III). The strong affinities of hydroxyl, −CH, and C=O groups for As(III) mean that they actively and significantly contributed to As adsorption. A new band, absent in the MSF@BC800 spectrum, appeared at 1462 cm⁻¹ after adsorption, which might be attributed to noncomplexed As−O bending and stretching vibration modes of MSF@BC800−As(III). A new, distinct, broad peak assigned to As-O stretching vibrations also appeared at 879 cm⁻¹ in the MSF@BC800−As(III) spectrum. A shifted, weaker Fe−O peak attributed to Fe−O−As vibrations appeared after adsorption, suggesting strong formation of inner-sphere complexes on MSF@BC800−As(III) surfaces. Markedly intensified and broadened Mo−S peaks shifted to 490 cm⁻¹ after adsorption, indicating strong interactions between As and Mo−S groups. Thus, MSF@BC800-surface functional groups such as C=O, −CH, −OH, Fe−O, and Mo−S showed strong affinities for As(III), and therefore played an important role in As(III) sorption by surface complexation and group exchange. Our results are similar to the findings of Liu et al., who reported that the intensification of peaks in As-loaded Fe−Mn−Ce oxide-modified BC composites was attributable to As(III)-surface-functional-group complexations. In summary, electrostatic interactions and complexations were important mechanisms for As(III) adsorption on MSF@BC800, showing that surface-functional groups were the dominant property controlling As(III) adsorption.

The interaction mechanism was further characterized by XPS spectroscopy. High-resolution 3d XPS spectra are presented in Figure 8b. MSF@BC800−As(III) presented distinct peaks at about 44.7 and 43.9 eV, corresponding to As(V) and (III), respectively. As(V) and (III) comprised 41.6 and 58.4% of total As, respectively, indicating that As(III) had been partially oxidized to As(V) upon adsorption, with As(III) being initially adsorbed through electrostatic interactions and then partially oxidized. Finally, oxidized As(V) was retained by the adsorbent.

To further elucidate the adsorption mechanism, structural activation by functional groups and corresponding changes in Fe, S, O, and Mo XPS peak intensities and binding energies were studied, with structural activation being found to increase adsorption capacity. Figure 8c shows that after As (III) adsorption, Fe 2p peaks shifted to higher binding energies, indicating inner-sphere Fe−O−As complex formation. Lin et al. previously reported that FeOx played an important role in As(III) adsorption.

In the O 1s spectrum (Figure 8d), the area ratios of the C=O and O−C=O peaks increased from 17.8 to 21.5% and from 23.6 to 26.2%, respectively, indicating that bonds between ligand carboxylates and Fe−O nodes might be partly broken during adsorption, resulting in the formation of free carboxylate groups. After adsorption, peaks at 530.4 and 531.5 eV (assigned to Fe−O and Fe−OH, respectively) weakened. Area ratios for O in metal−O and metal−OH nodes increased from 17.4 to 10.7% and from 41.2 to 28.8%, respectively, suggesting that the metal−O and metal−OH structures were involved in sorption. Furthermore, after adsorption, a new peak attributed to O−As/Fe−O−As appeared at 530.8 eV, suggesting coordination between O−As/Fe−O−As and adsorbed arsenic species and implying O−As bond formation during As adsorption on MSF@BC800.

After adsorption, three peaks obviously changed in the Mo 3d spectrum (Figure 8e). Peak binding energies are shown in Table S4. The Mo 3d₃/₂ and Mo 3d₅/₂ peak-area ratios slightly decreased from 26.4 to 22.0% and from 50.7 to 45.3%, respectively, while the Mo 3d₃/₂ peak shifted from 231.7 to 231.6 eV, thereby elucidating the mechanism for As(III) adsorption on MSF@BC800. Moreover, the S 2s peak shifted from 227.0 to 227.3 eV and the corresponding peak area increased from 22.9 to 32.7%, indicating the role of S groups in sorption. The S(VI) peak position and area ratio shifted from 167.5 to 168.2 eV and decreased from 55.6 to 33.6%, respectively. The S 2p₁/₂ peak-area ratio increased from 13.9 to 40.6% (Figure 8f). The S 2p₁/₂ peak position and area ratio shifted from 161.8 to 161.6 eV and decreased from 30.5 to 25.8%, respectively, revealing that S groups assisted in the adsorption of As(III) on MSF@BC800 through strong interactions between S groups and As(III).

In summary, the results showed that FeOx and MoS₂ synergistically affected As(III) removal by MSF@BC800 and that the mechanism of As(III) removal was governed by electrostatic attraction and inner-sphere complexations.

### 3. CONCLUSIONS

Novel MoS₂-impregnated iron-based BC (MSF@BC) adsorbents were hydrothermally synthesized, and FTIR, XPS, and XRD analyses were used to confirm surface modifications. MSF@BC800 displayed the best As adsorption among all the
prepared adsorbents. The synergistic effects of FeOx and MoS2 on MSF@BC800 enhanced the As(III)-removal efficiency and resulted in superior As(III) affinity over a wide range of As(III) concentrations in aqueous solutions compared to FeOx@BC800 or MS@BC800. Equilibrium and adsorption data followed the Freundlich and pseudo-second-order models, respectively. ΔH° > 0 indicated that sorption was endothermic. Therefore, MSF@BC800 is an excellent candidate as an eco-friendly material for effective As(III) adsorption and environmental decontamination.

4. MATERIALS AND METHODS

4.1. Materials. Analytical-grade chemicals including iron nitrate (Fe(NO3)3·9H2O), sodium hydroxide (NaOH), hexammonium heptamolybdate, thiourea, and nitric acid (HNO3) were purchased from Jiuxinyaozheng Co., Ltd., Beijing (China). Deionized water (18.25 MΩ·cm−1) was prepared by a Millipore Milli-Q water purification system.

4.2. BC Preparation. Corn straw obtained from the Tianjin suburbs was used to prepare BCs. Corn straw powder (100-mesh) was transferred to a muffle furnace and pyrolyzed in nitrogen at 600 and 800 °C for 2 h. The resulting BCs are hereafter named “BC600” and “BC800,” respectively.

4.3. Synthesis of Fe-Modified BCs. To prepare Fe-modified BCs, 5 g of corn straw powder was added to a 500 mL beaker containing 100 mL of 0.06 M aqueous ferric nitrate. The mixture was stirred for 1 h, sonicated for 2 h, and evaporated until dry in a constant-temperature (90 °C) water bath. The residue was ground, passed through a 0.154 mm mesh sieve, and pyrolyzed in nitrogen in a muffle furnace operating at 600 and 800 °C for 2 h. Fe-modified BCs are hereafter named “FeOx@BC600” and “FeOx@BC800,” respectively.

4.4. Synthesis of MoS2-Modified Fe-Based BCs. To prepare MoS2-modified adsorbents, 10 mmol hexaammonium heptamolybdate and 0.5 mol thiourea were added to a 250 mL beaker containing 80 mL of Milli-Q deionized water. To homogenize the solution, it was stirred for 0.5 h on a magnetic stirrer and then 1 g of either BC800, FeOx@BC600, or FeOx@BC800 was added to the homogeneous solution, which was continuously stirred for 2 h and then sonicated for 1 h. The mixture was then sealed in a Teflon-lined autoclave and heated in an oven for 10 h at 200 °C. After that the collected samples were rinsed repeatedly with a significant excess of deionized water. Before collection, samples were freeze-dried under vacuum. Final samples are hereafter named “MS@BC800,” “MSF@BC600,” and “MS@BC800,” respectively, where “MS” represents molybdenum disulfide.

All the prepared sorbents were ground, passed through a 0.154 mm sieve, and repeatedly rinsed with significant excesses of deionized distilled water. Detailed descriptions of instruments, characterization methods, and As(III)-adsorption experimental procedures are presented in the Supporting Information.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.0c01268.

Characterization of prepared composites before and after adsorption and detailed procedures of adsorption experiments for As(III) removal (PDF)

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Notes
The authors declare no competing financial interest.

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