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Supporting Information

Amorphous Fe/Mn Bimetal-Organic Frameworks: Outer and Inner Structural Design for Efficient Arsenic(III) Removal

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Supporting Figures

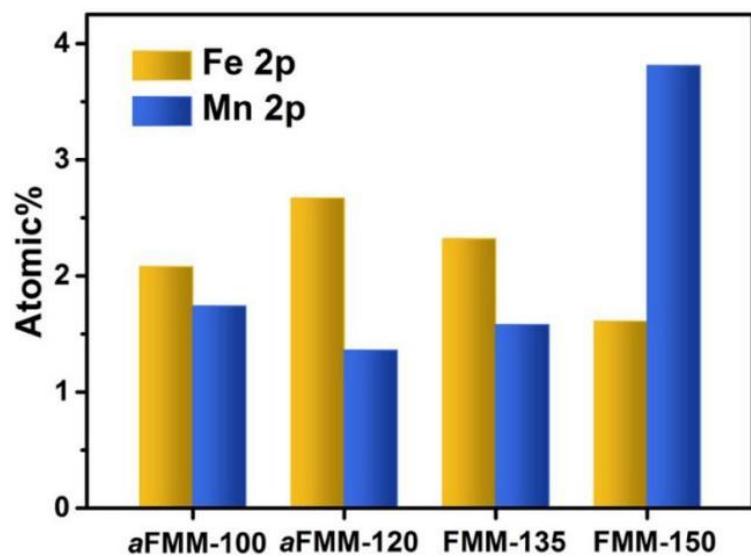


Figure S1. Distribution of the Fe and Mn contents of FMMs by using XPS in this work.

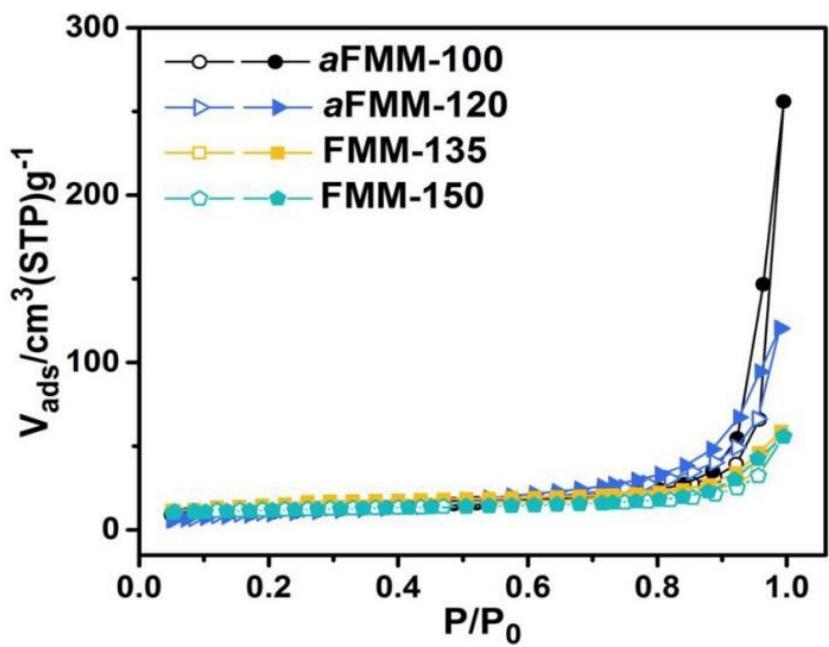


Figure S2. N_2 adsorption/desorption isotherms of aFMM-100, aFMM-120, FMM-135 and FMM-150 taken at 77 K.

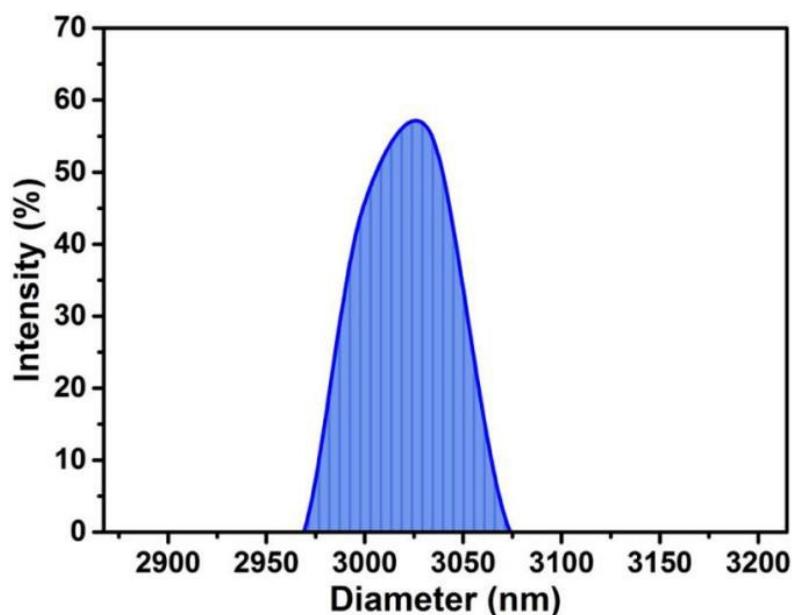


Figure S3. Size distribution of aFMM-120 in water determined by using DLS.

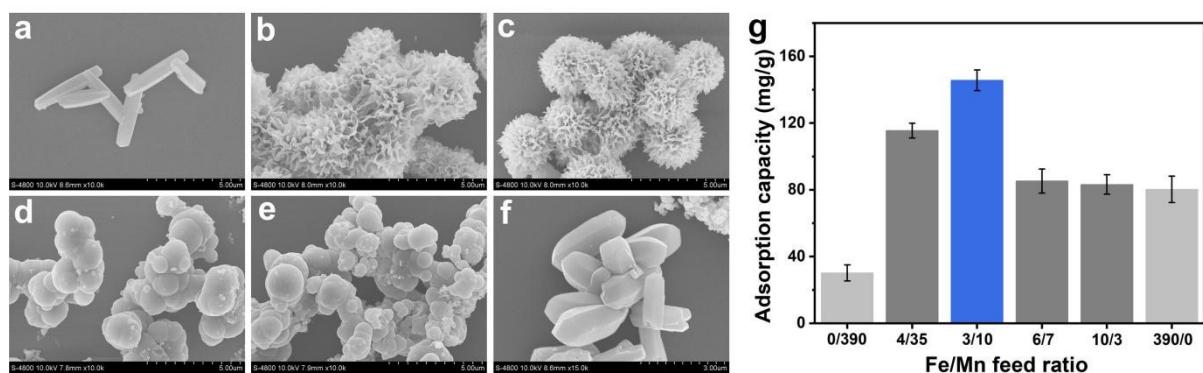


Figure S4. a-f) The SEM images of MOF-74s with increased Fe/Mn feed radios from 0/390 to 390/0. **g)** The adsorption performance of MOF-74s toward As(III) with different Fe/Mn feed radios.

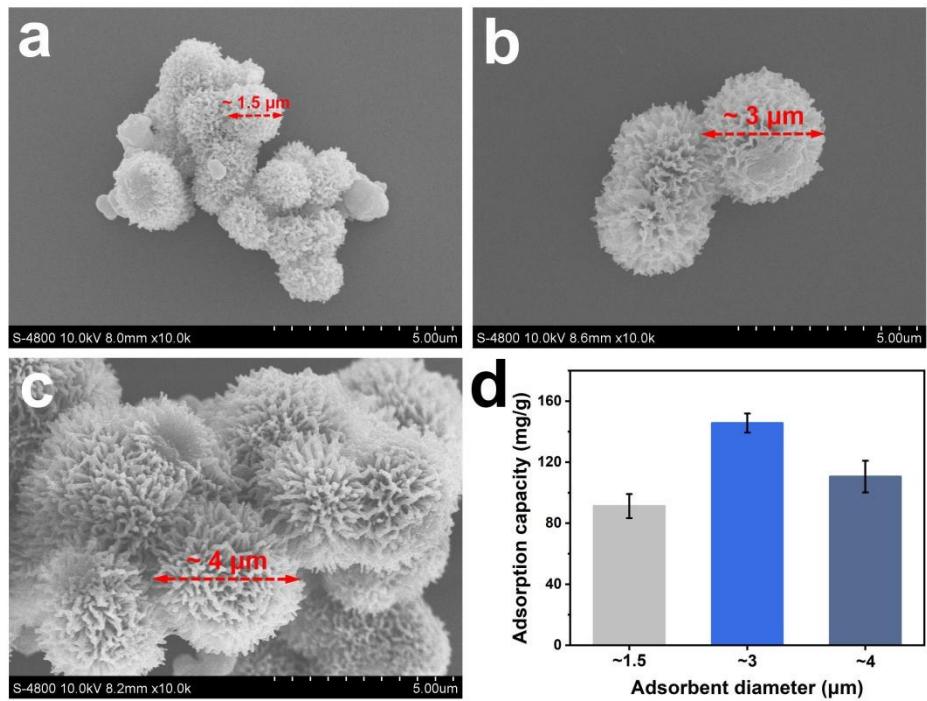


Figure S5.a-c) SEM images of aFMM-120s with different sizes of $\sim 1.5 \mu\text{m}$, $\sim 3 \mu\text{m}$ and $\sim 4 \mu\text{m}$ and **d)** the corresponding adsorption capacities toward arsenic species.

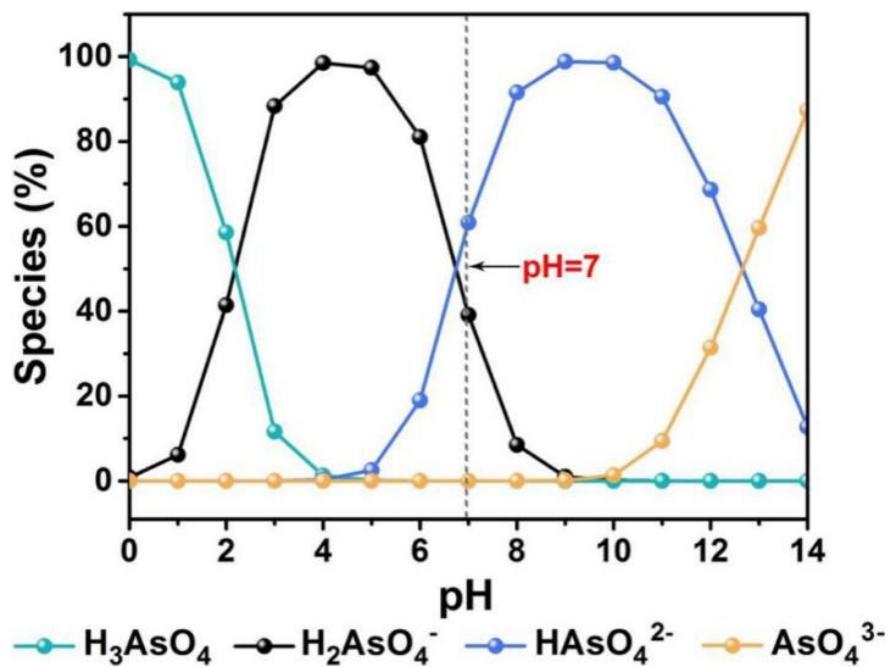


Figure S6. Different species of As(V) with changing pHs (0-14) obtained by Visual MINTEQ software programme.

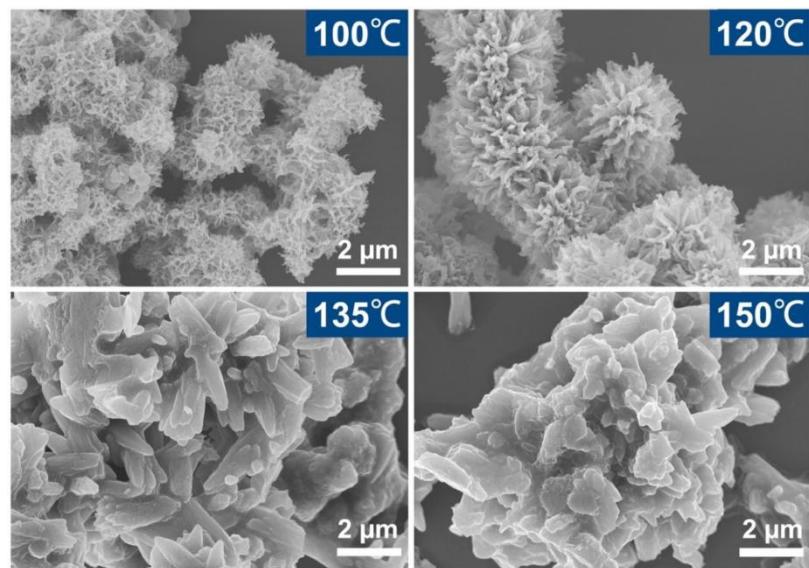


Figure S7. The photographs of aFMM-100, aFMM-120, FMM-135 and FMM-150after oxidation/adsorption of arsenic.

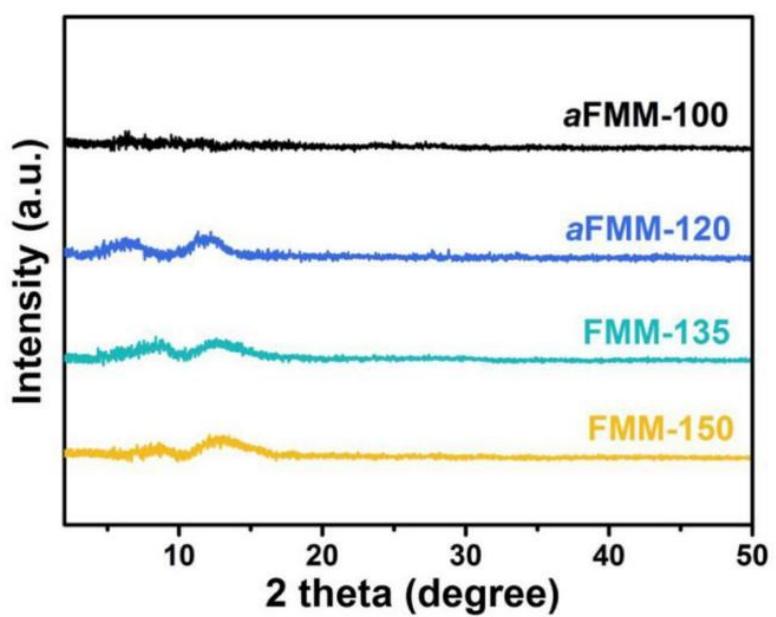


Figure S8. XRD patterns of aFMM-100, aFMM-120, FMM-135 and FMM-150 after oxidation/adsorption of arsenic.

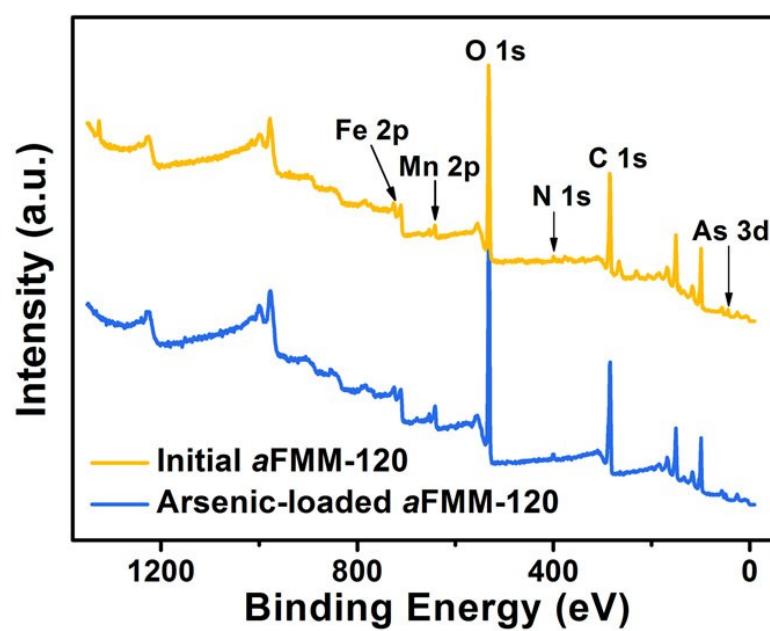


Figure S9. XPS wide scan spectrum of aFMM-120 before and after oxidation/adsorption of arsenic.

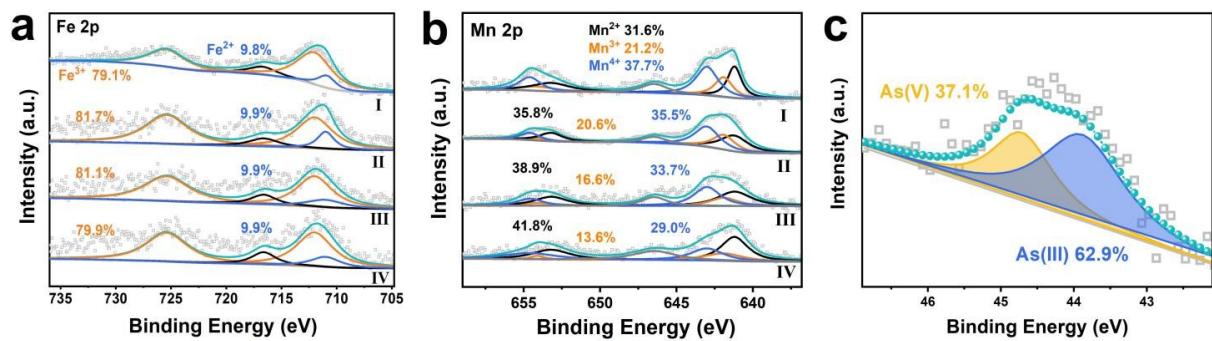


Figure S10. High resolution XPS spectra of **a)** Fe 2p and **b)** Mn 2p of regenerated aFMM-120 in four adsorption-desorption cycles from I to IV. **c)** High resolution XPS spectra of As 3d in the eluent.

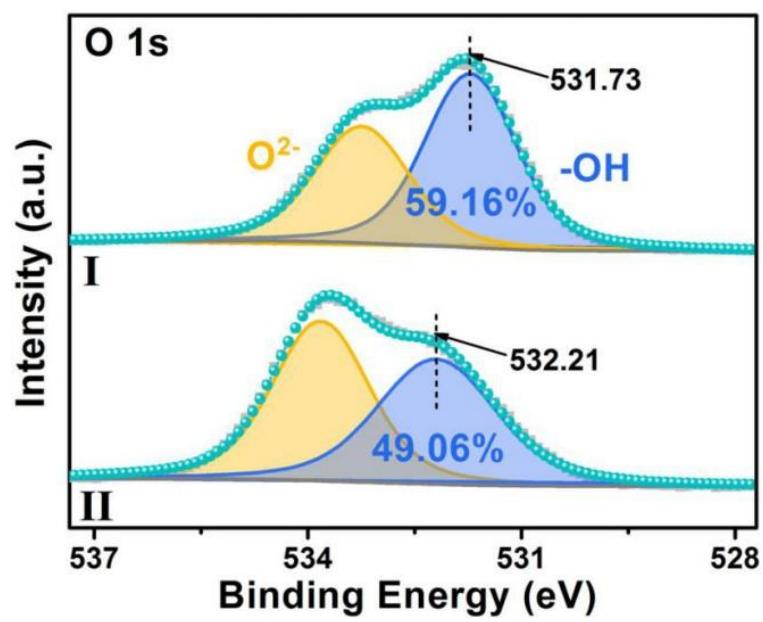


Figure S11. O 1s XPS spectra of aFMM-120 before and after oxidation/adsorption of arsenic.

Supporting Tables

Table S1. Arsenic adsorption kinetics parameters using pseudo-second-order model on aFMM-120.

| Concentration (mg/L) | pseudo-second-order | | | |
|-------------------------|--------------------------------|-------------------------|--------------------------------|--------|
| | $q_e, \text{exp}(\text{mg/g})$ | $k_2 (\text{g/mg min})$ | $q_e, \text{cal}(\text{mg/g})$ | R^2 |
| 5 | 5.518 | 0.02587 | 5.924 | 0.9926 |
| 10 | 13.97 | 0.01668 | 14.54 | 0.9985 |
| 20 | 28.89 | 0.01751 | 29.42 | 0.9994 |
| 30 | 46.36 | 0.009612 | 46.64 | 0.9964 |
| 50 | 79.05 | 0.003961 | 81.28 | 0.9936 |

Table S2. Kinetic fitting parameters using pseudo-first order and intra-particle diffusion models of As(III) adsorption on aFMM-120.

| Concentration (mg/L) | pseudo-first-order | | | intra-particle diffusion | | |
|----------------------|----------------------------|--------------|----------------|--------------------------|--------|----------------|
| | k_1 (min ⁻¹) | q_e (mg/g) | R ² | k_i | C_i | R ² |
| 5 | 0.1067 | 5.298 | 0.9907 | 0.8918 | 0.5918 | 0.9956 |
| 10 | 0.1549 | 13.24 | 0.9803 | 0.9457 | 7.663 | 0.9889 |
| 20 | 0.2613 | 27.75 | 0.9921 | 2.202 | 16.98 | 0.8246 |
| 30 | 0.2484 | 43.57 | 0.9745 | 3.213 | 26.75 | 0.2942 |
| 50 | 0.1957 | 74.86 | 0.9807 | 7.300 | 36.82 | 0.6721 |

Table S3. Isotherms parameters using Langmuir and Freundlich isotherm model of As(III) adsorption on aFMM-120.

| Temperatu re (K) | $q_{\max,}$ exp(mg/g) | Langmuir isotherm model | | | Freundlich isotherm model | | |
|---------------------|--------------------------|-------------------------|------------------------------------|--------|---------------------------|-------|--------|
| | | k_L | $q_{\max,cal}(\text{mg}/\text{g})$ | R^2 | k_F (mg/g) | n | R^2 |
| 298 | 138.0 | 0.009529 | 230.4 | 0.9592 | 4.374 | 1.433 | 0.9901 |
| 308 | 145.7 | 0.009077 | 258.4 | 0.9831 | 4.340 | 1.386 | 0.9934 |
| 318 | 161.6 | 0.01439 | 233.6 | 0.9782 | 7.465 | 1.595 | 0.9903 |

Table S4. Comparison of maximum As(III) adsorption over different adsorbent materials.

| Adsorbents | Adsorption capacity to As (III) (unit: mg/g) | Equilibrium time | Reference |
|--|--|------------------|-----------|
| UIO-66 | 40 | 24 h | 1 |
| MnO ₂ @ZIF-8 | 147.28 | 24 h | 2 |
| CoFe ₂ O ₄ @MIL-100(Fe) | 143.6 | 2 min | 3 |
| ZIF-8nps | 49.49 | 13 h | 4 |
| Fe ₃ O ₄ @MIL-101(Cr) | 121.5 | 24 h | 5 |
| Cubic ZIF-8 | 122.6 | 10 h | 6 |
| Leaf-shaped ZIF-8 | 108.1 | 10 h | 6 |
| Dodecahedral ZIF-8 | 117.5 | 10 h | 6 |
| Fe mesh@MIL-100(Fe) | 35.2 | 6 h | 7 |
| Fe ₃ O ₄ @ZIF-8 | 100.0 | 4 h | 8 |
| Fe ₃ O ₄ -RGO-MnO ₂ | 14.04 | 40 min | 9 |
| Fe-Mn binary oxide | 100.4 | 16 h | 10 |
| Ce-Mn binary oxide | 97.7 | 14 h | 11 |
| Ceria associated manganese oxide | 34.89 | 40 min | 12 |
| Zr-Mn binary oxide | 104.5 | / | 13 |
| Magnetic Fe–Mn binary oxide | 56.1 | 60 min | 14 |
| aFMM-120 | 161.6 | 70 min | this work |

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