

Supporting Information for

**Properties and reactivity of sulfidized nanoscale zero-valent iron prepared with
different borohydride amounts**

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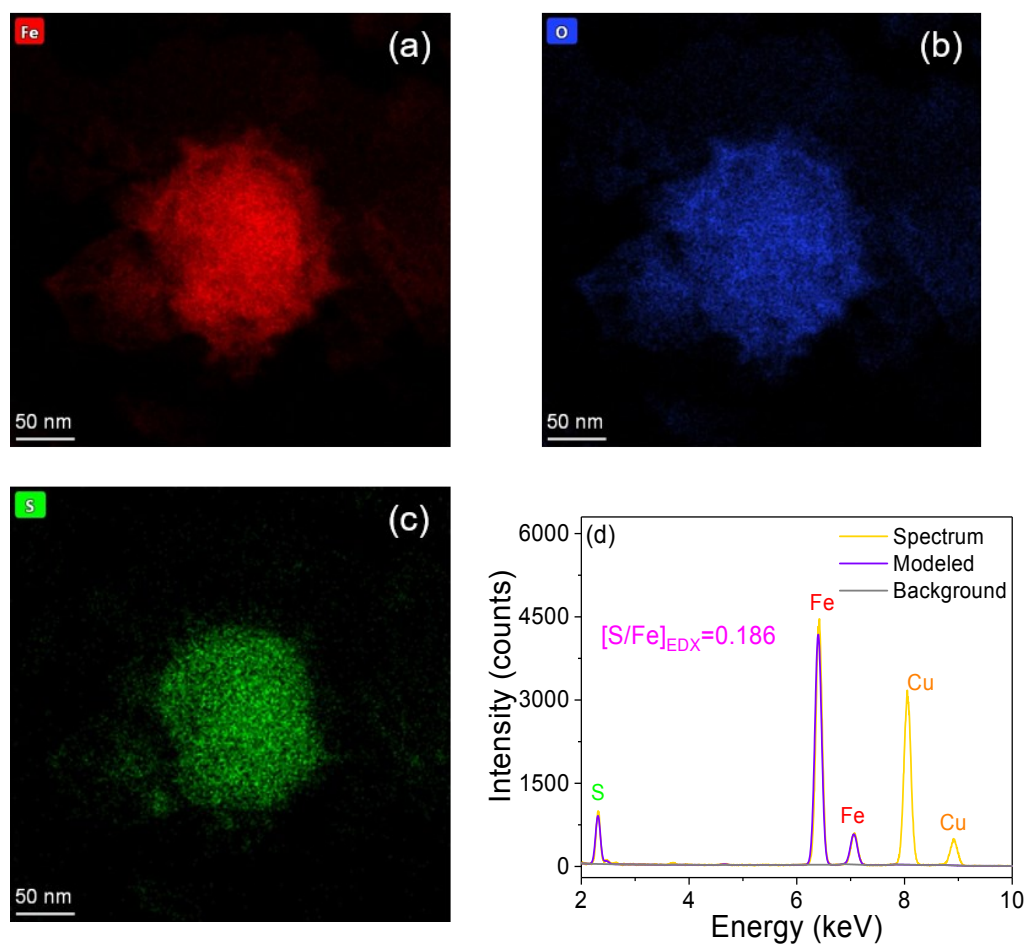


Figure S1 (a) Fe, (b) O, (c) S maps, and (d) EDX spectra of SNZVI synthesized with a dosed NaBH_4/Fe molar ratio of 1.5.

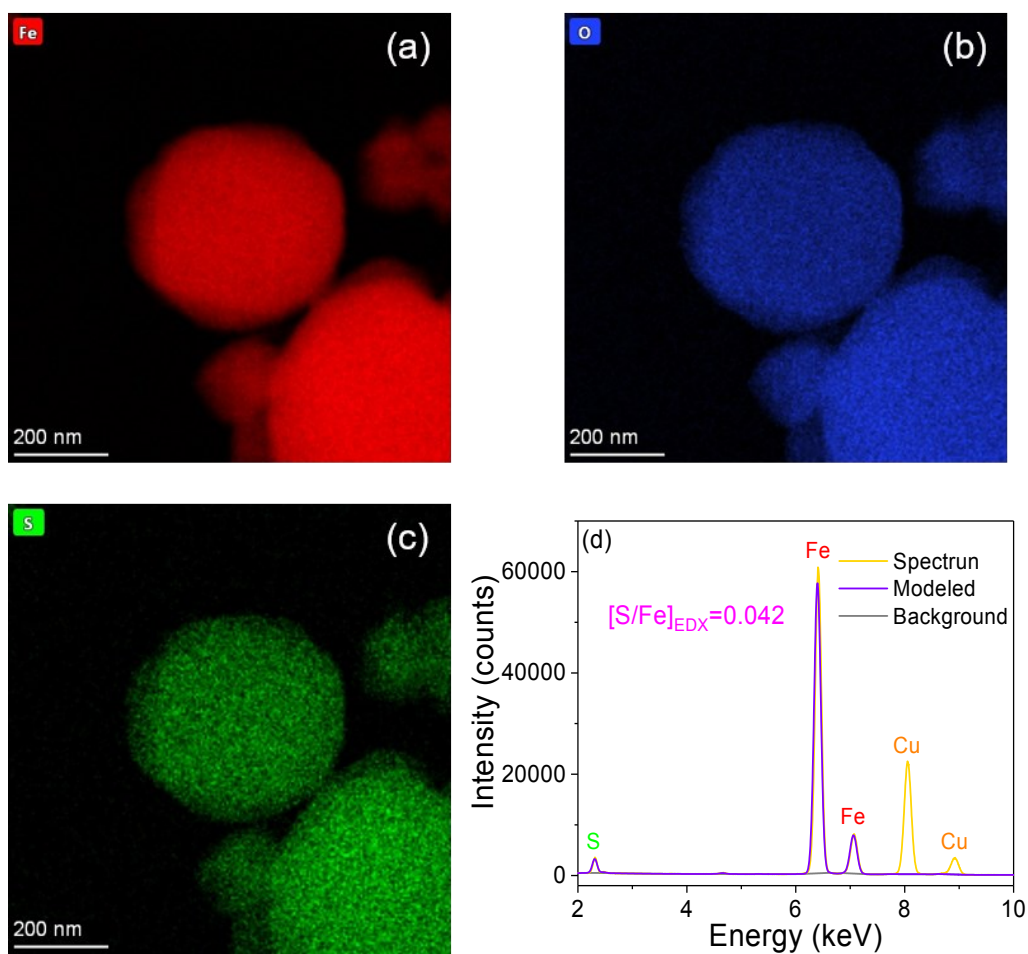


Figure S2 (a) Fe, (b) O, (c) S maps, and (d) EDX spectra of SNZVI synthesized with a dosed NaBH_4/Fe molar ratio of 3.

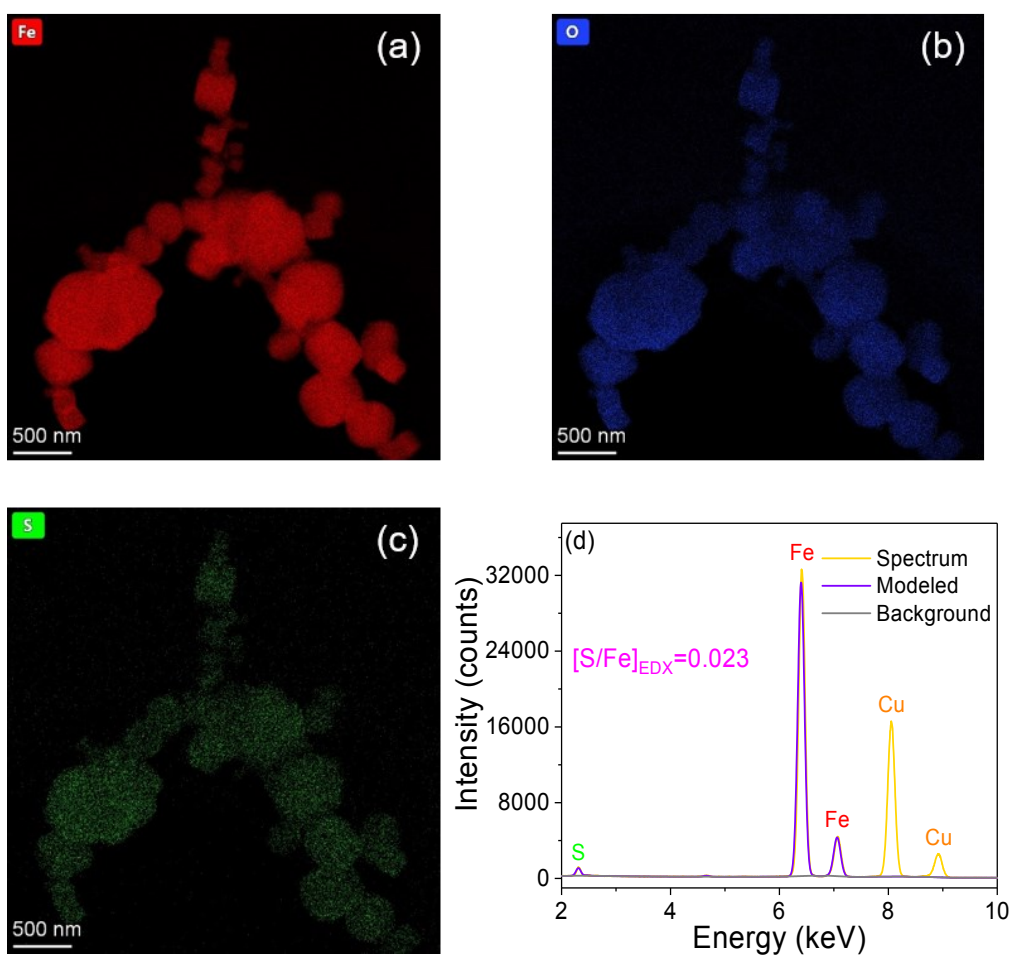


Figure S3 (a) Fe, (b) O, (c) S maps, and (d) EDX spectra of SNZVI synthesized with a dosed NaBH_4/Fe molar ratio of 5.

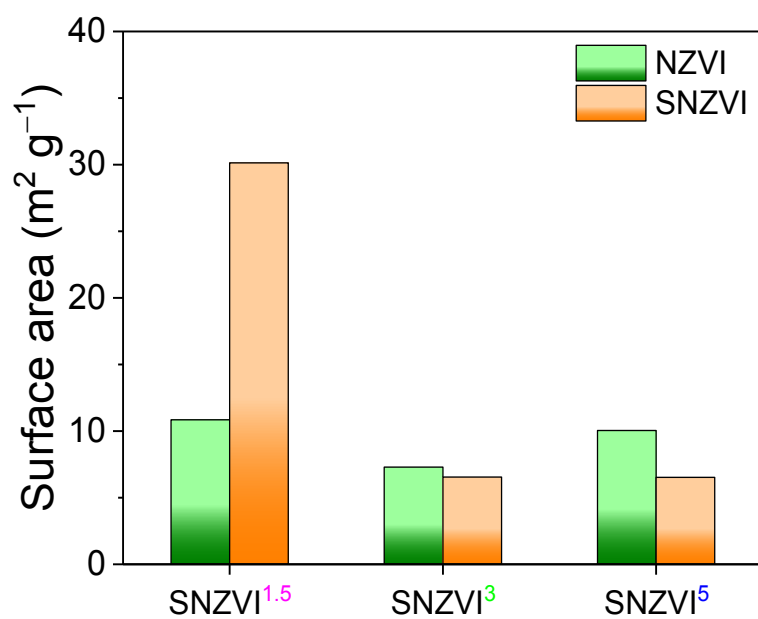


Figure S4 Specific surface area of NZVI and SNZVI

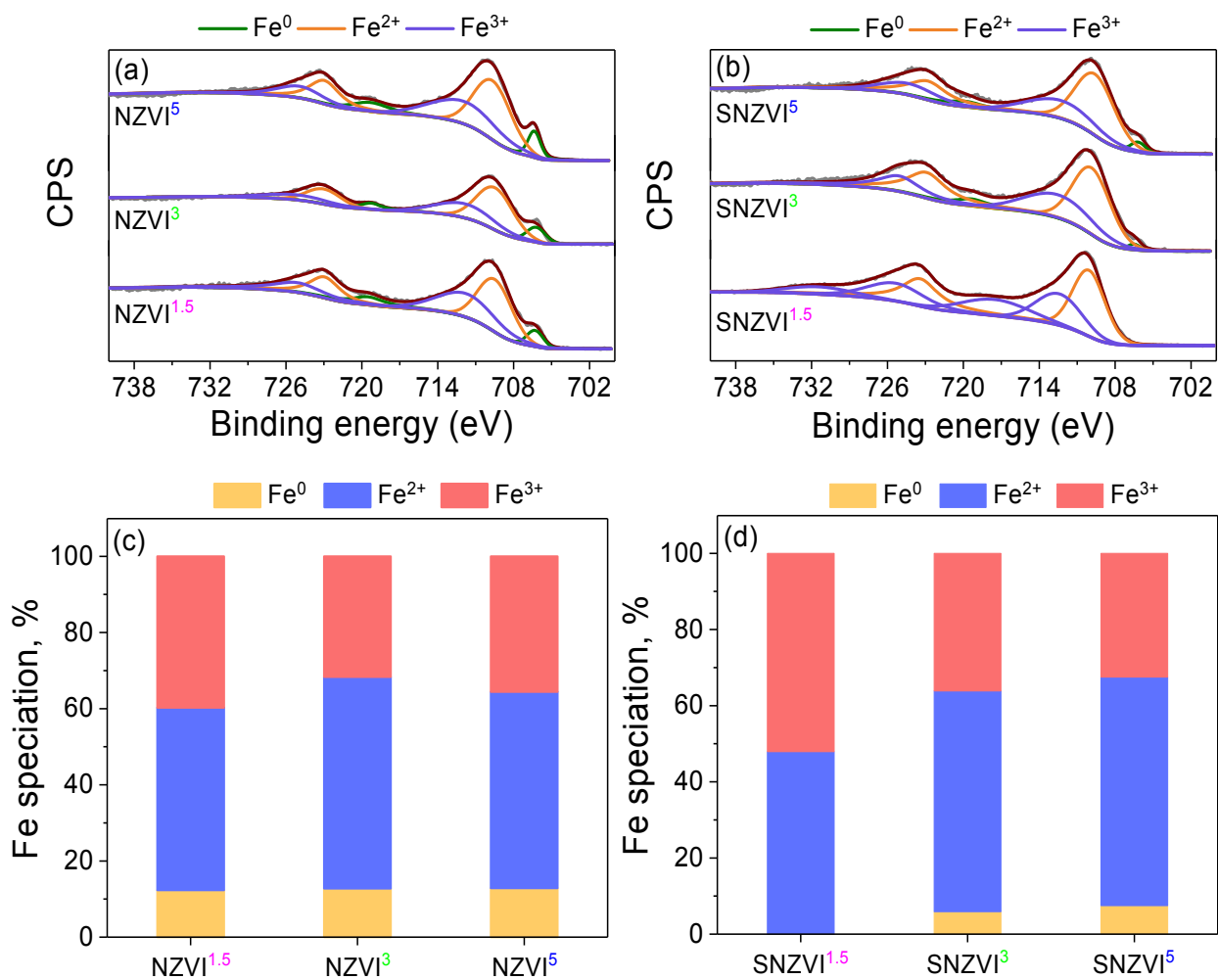


Figure S5 (a, b) Fe 2p XPS spectrum and (c, d) Fe speciation of NZVI and SNZVI

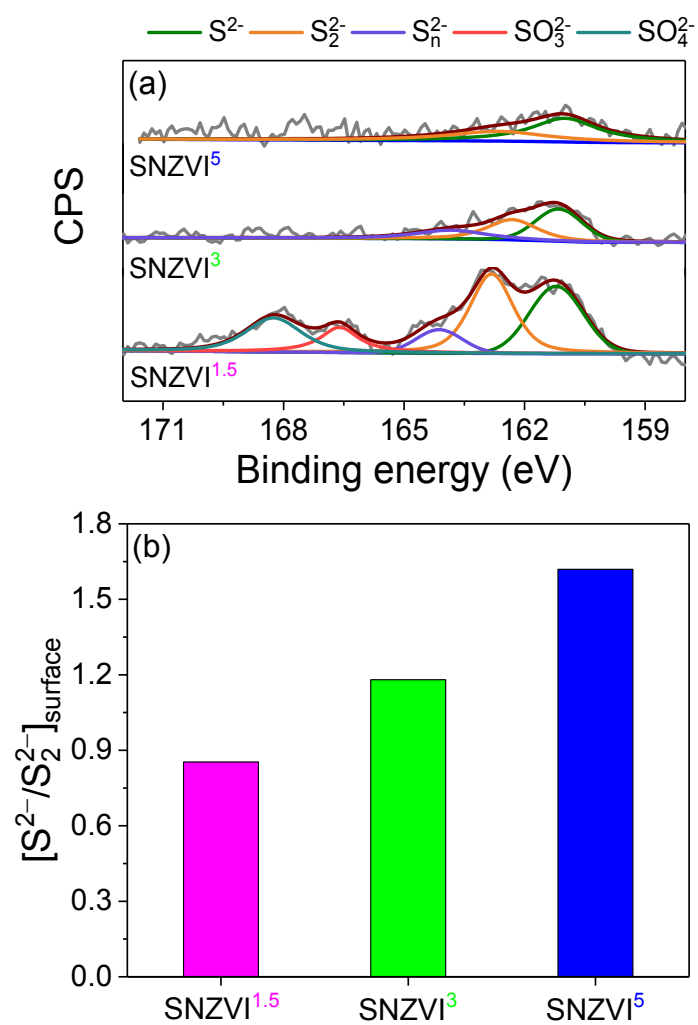


Figure S6 (a) S 2p XPS spectrum and (b) $[S^{2-}/S_2^{2-}]_{\text{surface}}$ of SNZVI.

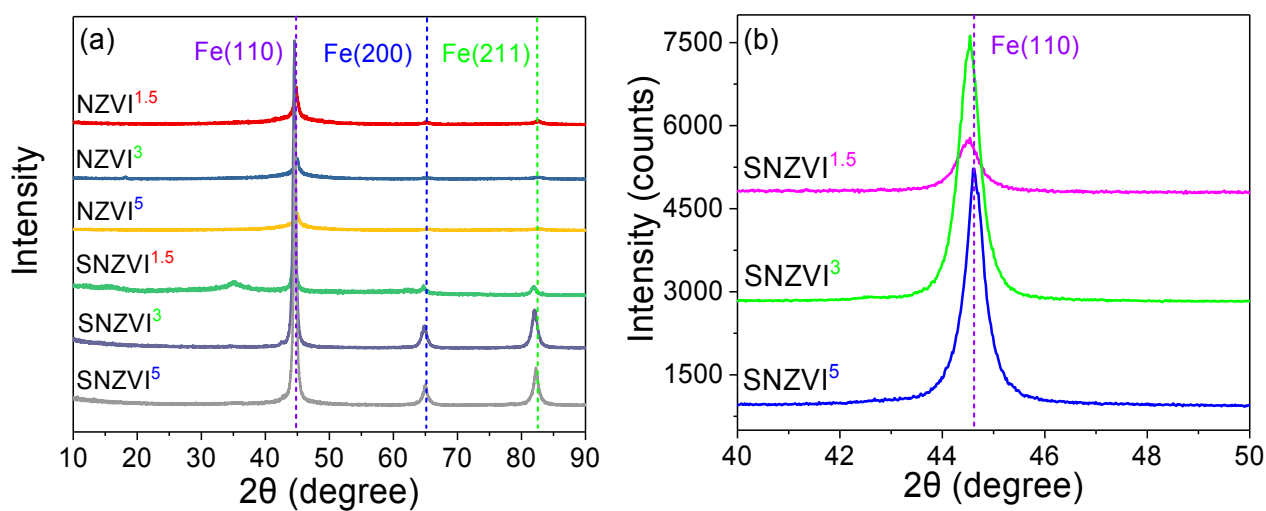


Figure S7 (a) XRD spectrum of NZVI and SNZVI and (b) The shift of SNZVI at Fe(110) lattice plane in XRD spectrum (Position of Fe(110) plane for NZVI was $2\theta = 44.98^\circ$)

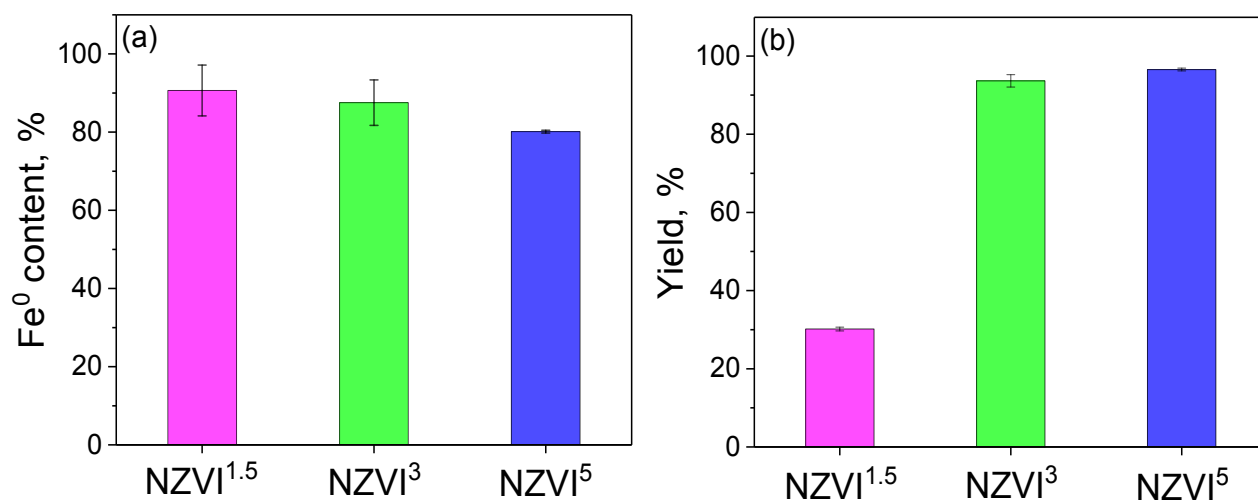


Figure S8 (a) Fe^0 content and (b) Yield of NZVI

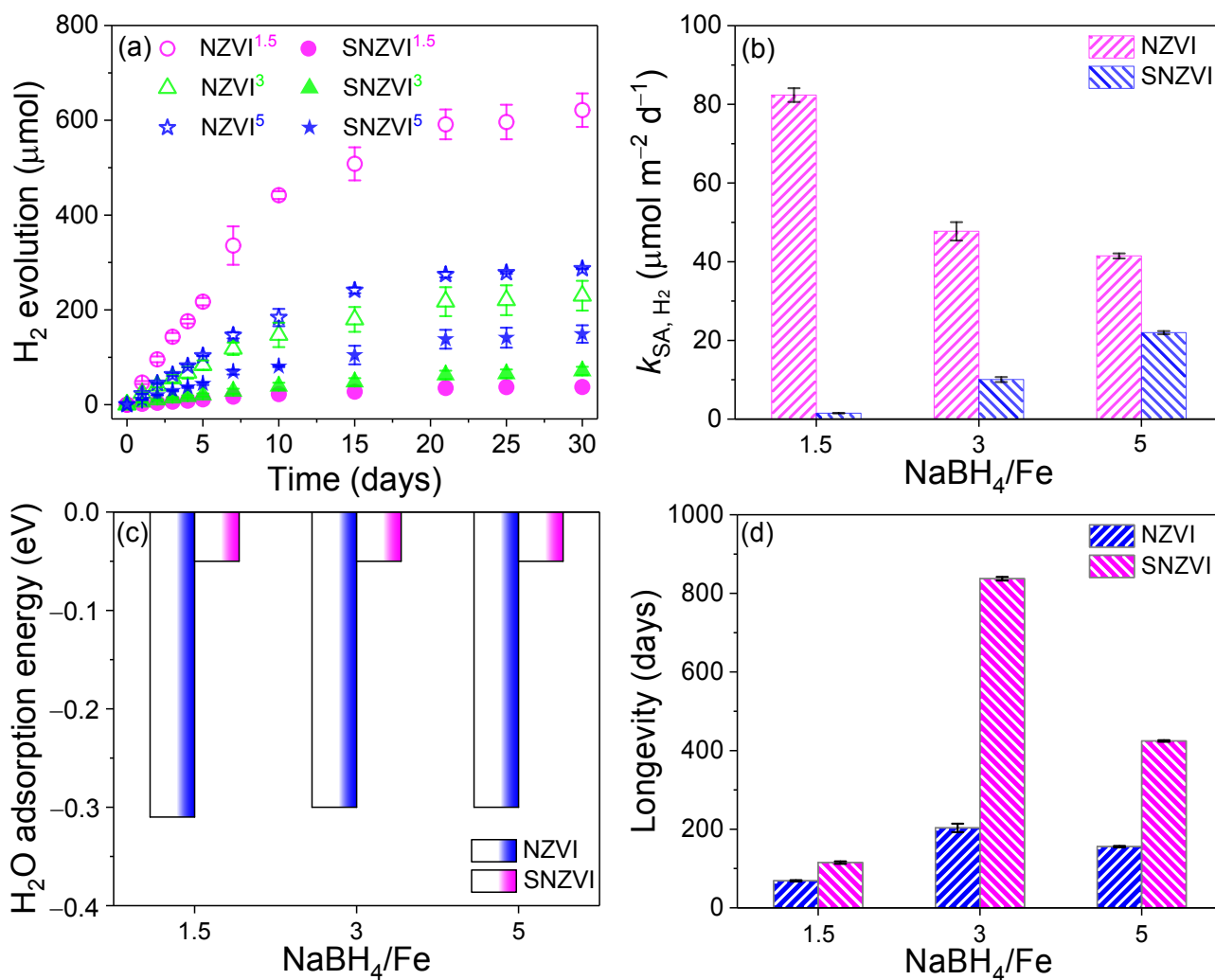


Figure S9 (a) H_2 evolution and (b) Surface area-normalized zero-order kinetics rate of H_2 evolution (k_{SA, H_2} , H_2 evolution of NZVI in first 5 days was calculated for comparison), (c) DFT-calculated $H_2\text{O}$ adsorption energy and (d) Estimated longevity of NZVI and SNZVI (The longevity was estimated when the accumulated H_2 reached to 100% of actual Fe^0 content). (T=298 K, initial pH=7.0, 1.0 g L^{-1} NZVI or SNZVI).

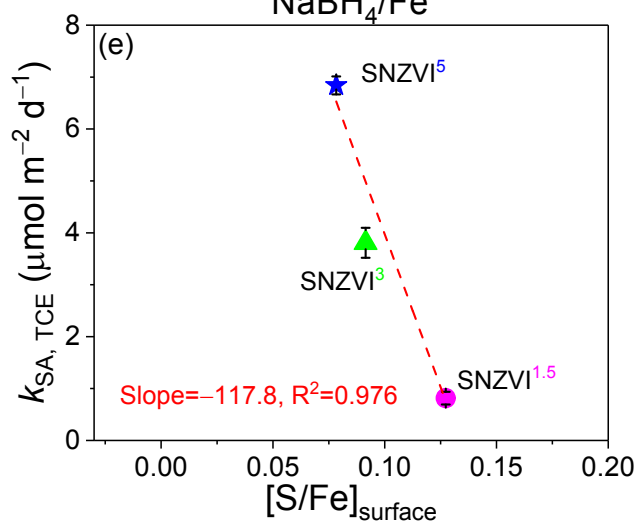
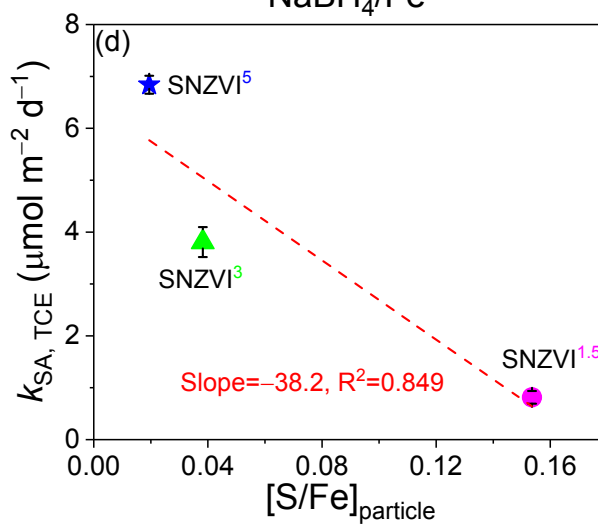
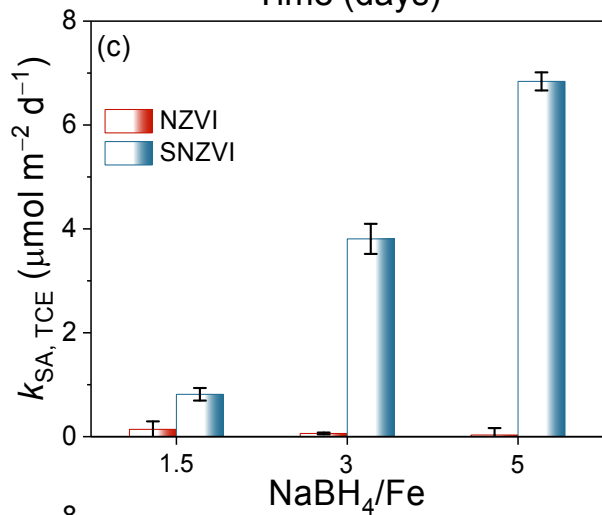
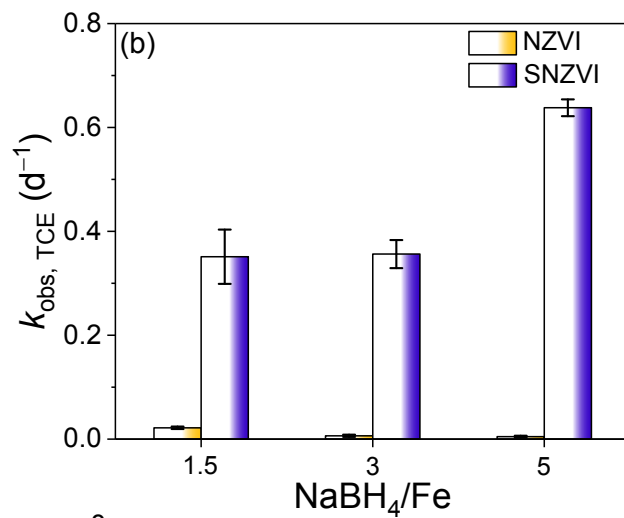
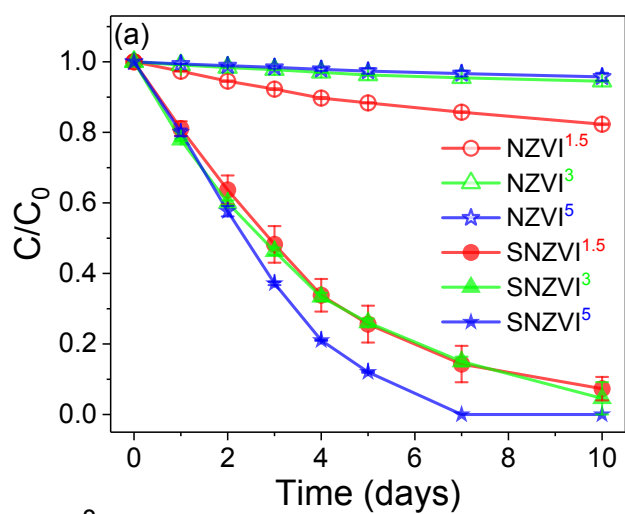


Figure S10 (a) Removal of TCE, (b) Pseudo first-order kinetics rate of TCE removal ($k_{\text{obs, TCE}}$) and (c) Surface area normalized rates of TCE removal ($k_{\text{SA, TCE}}$) by NZVI and SNZVI, Correlations of $k_{\text{SA, TCE}}$ to (d) $[\text{S/Fe}]_{\text{particle}}$ and (e) $[\text{S/Fe}]_{\text{surface}}$. (T=298 K, initial pH=7.0, 1.0 g L⁻¹ NZVI or SNZVI, 70 μM TCE)

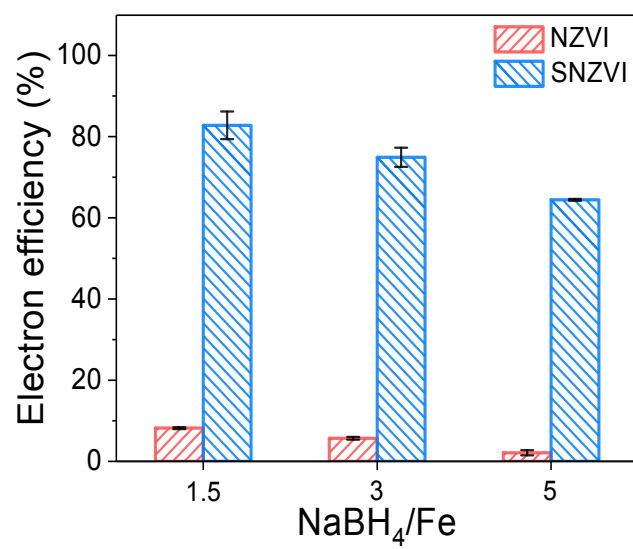


Figure S11 Electron efficiency of NZVI and SNZVI for TCE removal.

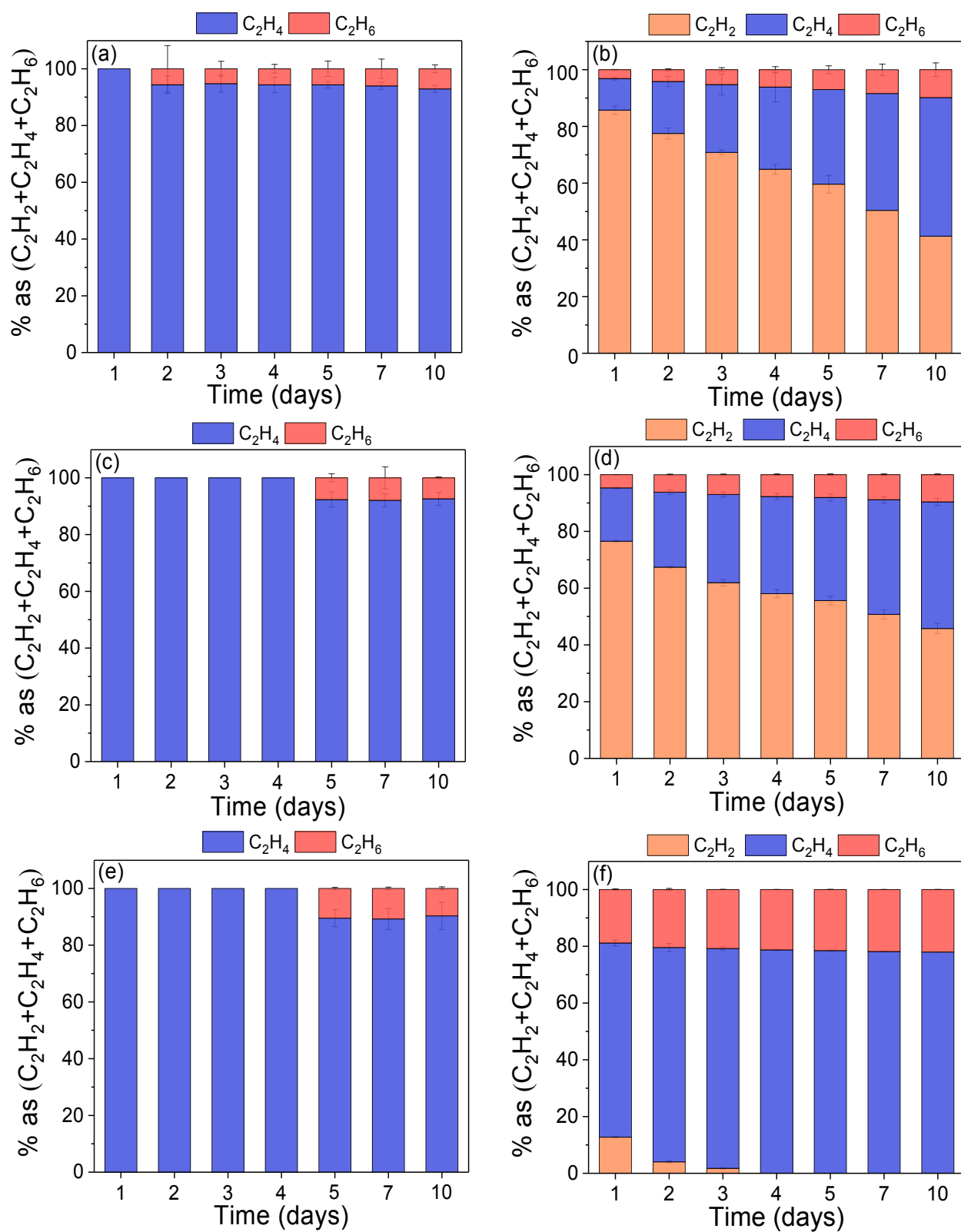


Figure S12 Main products fraction of TCE by NZVI and SNZVI synthesized with

$NaBH_4/Fe$ ratio of (a, b) 1.5, (c, d) 3, (e, f) 5

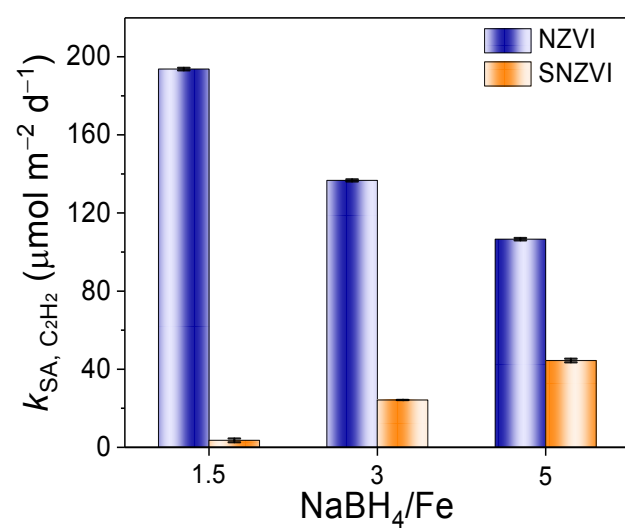


Figure S13 Surface area-normalized pseudo first-order kinetics rate of C_2H_2 removal (k_{SA, C_2H_2}).