Supporting Information

Regulating Ordered Structure and MOR Catalytic Performance of $L1_0$-FePt Nanoparticles through Mn Doping and Magnetic-Field Assistance

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First-principles calculations

DFT calculations were performed using VASP with PAW pseudopotentials and PBE functional [1-3]. We employed a 450 eV cutoff energy and ensured convergence within 10^{-5} eV for energy and 0.01 eV/Å for ionic forces, using a 5×5×1 Γ-centered k-mesh. The catalyst was modeled as a 6-layer $L1_0$-FePt (111) [4] supercell (96 atoms), with the bottom four layers constrained to bulk coordinates and a 15 Å vacuum layer applied. To accurately capture the Fe magnetic properties, spin-polarized ferromagnetic

configurations were enforced throughout the optimization process.

References:

[1] G. Kresse, J. Furthmüller, *Phys. Rev. B*, 1996, 54, 11169-11186.

[2] P. E. Blöchl, *Phys. Rev. B*, 1994, 50, 17953-17979.

[3] J. P. Perdew, A. Ruzsinszky, G. I. Csonka, O. A. Vydrov, G. E. Scuseria, L. A. Constantin, X. L. Zhou, K. Burke, *Phys. Rev. Lett.*, 2008, 100, 136406.

[4] H. Hong, D. X. Liu, J. Li., *Rare Met.*, 2024, 43, 1108-1115.

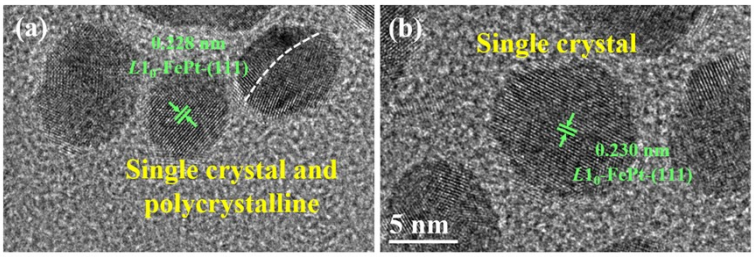

Fig. S1 HRTEM image of a single crystalline FePtMn-0 T (a) and FePtMn-2 T (b) samples

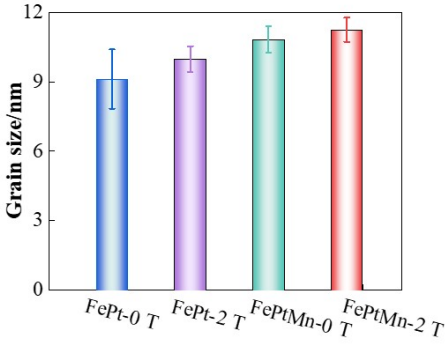

Fig. S2 Grain size of FePt and FePtMn NPs synthesized under 0 T and 2 T magnetic fields

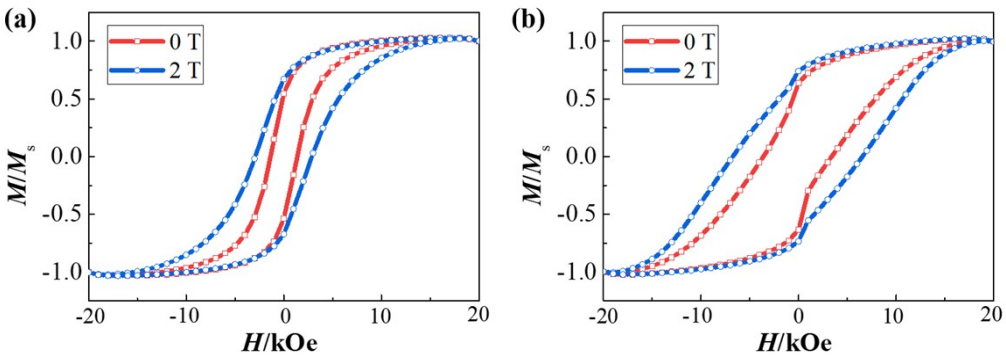

Fig. S3 Room temperature hysteresis loops of FePt NPs (a) and FePtMn NPs (a) synthesized under 0 T and 2 T magnetic fields

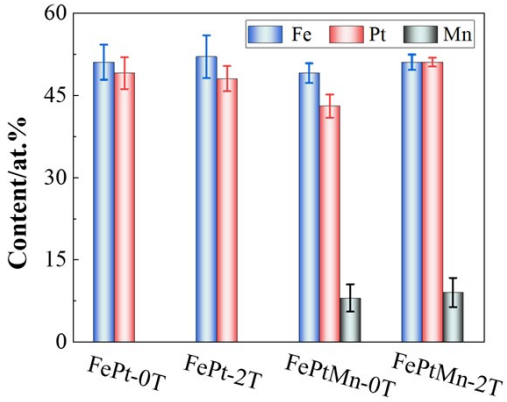

Fig. S4 The elemental composition of FePt and FePtMn NPs synthesized under 0 T and 2 T magnetic fields

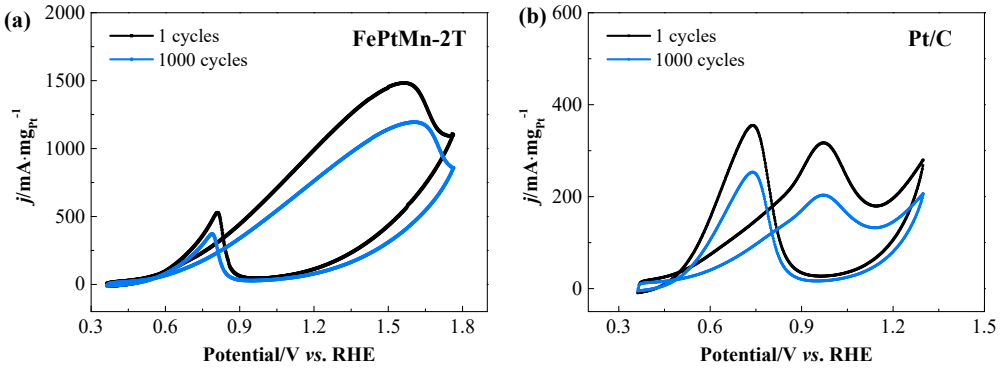

Fig. S5 The electrocatalytic durability of (a) FePtMn-2 T sample and (b) commercial Pt/C catalysts for MOR in 0.1 M HClO4 solution containing 0.1 M methanol for 1000 CV cycles

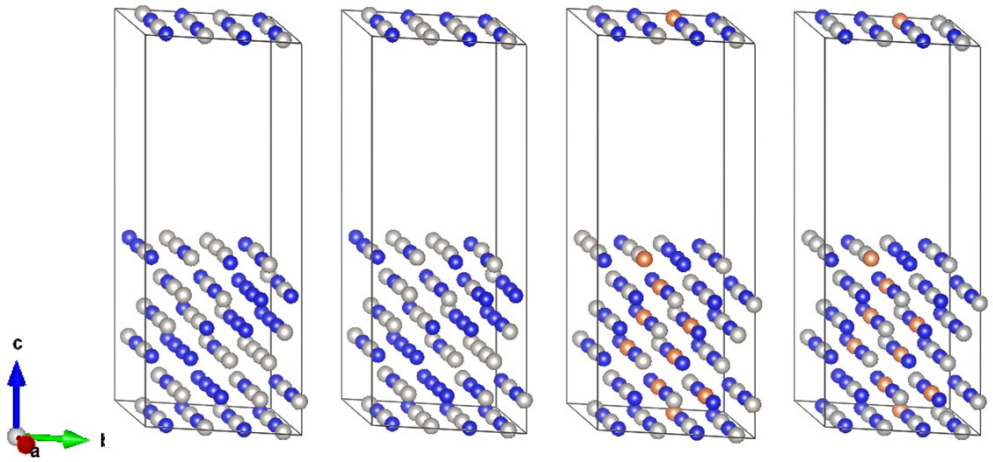

Fig. S6 DFT calculation models of (a) FePt-0T, (b) FePt-2T, (c) FePtMn-0T, and (d) FePtMn-2T samples