

The Description of Additional Supplementary Files

Title: Supplementary Data (Energy of Molecular Structure by First-Principles Calculations)

Description: This folder contains the raw output files generated from CASTEP calculations using Materials Studio. Each file corresponds to a specific M-type ferrite model and includes detailed computational results such as total energy, forces on atoms, and convergence information. These files serve as the primary output of the first-principles calculation, from which key thermodynamic parameters. The data in these files directly support the energy analysis presented in the main text. The naming convention follows the table provided in Supplementary Data (Correspondence of Molecular Structure).

Title: Supplementary Data (Isosurface Plot of Molecular Structure)

Description: This folder contains isosurface plots of differential charge density for all M-type ferrite models, generated from CASTEP calculations in Materials Studio. Each PNG file visualizes the electron cloud distribution around atomic sites. These images were used to analyze polarization behavior and dipole formation induced by iron vacancies, high-entropy effects, and Zr^{4+} doping. The isosurface plots directly support the differential charge density analysis presented in Fig. 3e and Supplementary Fig. 14. File names follow the naming convention established in Supplementary Data (Correspondence of Molecular Structure).

Title: Supplementary Data (Molecular Structure)

Description: This folder contains structural visualizations of M-type ferrite models used in DFT calculations. The parent structure— $\text{BaFe}_{12}\text{O}_{19}$, was obtained from the Materials Project

database. Based on this parent structure, introduced iron vacancies and elemental doping to construct the derivative models. These images serve as visual references for computational models, supporting structural analysis in the main text. File names follow the naming convention in Supplementary Data (Correspondence of Molecular Structure).

Title: Supplementary Data (Optimization of Molecular Structure)

Description: This folder contains the raw output files from geometry optimization calculations performed with CASTEP in Materials Studio. Each file corresponds to a specific M-type ferrite model and records detailed computational results.

Title: Supplementary Data (Correspondence of Molecular Structure)

Description: This document maps abbreviated file names used in the computational data to their corresponding chemical formulas. For low-entropy ferrites, abbreviation like **Ba12** represent **BaFe₁₂O₁₉**. For high-entropy systems, **hem** denotes **(LaBaCaSrPb)_{0.2}Fe₁₂O₁₉**. This mapping ensures clear identification of data files for reproducibility.

Title: Supplementary Data (Gibbs Free Energy of Molecular Structure by a Combination of Software Calculations and Manual Calculations)

Description: This document summarizes the total energy results obtained from first-principles density functional theory calculations for all M-type ferrite models. It also details the calculation procedures for configurational entropy and Gibbs free energy, including key formulas and specific examples for high-entropy systems. The total energy

values for each composition are compiled in a table.