Investigating structure-stability relationship of $SmFe_{12-\alpha-\beta}X_\alpha Y_\beta$ structures X, Y={Mo, Zn, Co, Cu, Ti, Al, Ga}

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We introduce a *query-and-learn* active learning to search for stable crystal structure, clarifying the stabilization mechanism of $SmFe_{12}$-based compounds which exhibits prominent magnetic properties. The proposed method aims to (i) accurately estimate formation energies with limited first-principles calculation data, (ii) visually monitor the progress of the structure search process, (iii) extract correlations between structures and formation energies, and (iv) recommend the most beneficial candidates of $SmFe_{12}$-substituted structures for the subsequent first-principles calculations.

3307 structures of $SmFe_{12-\alpha-\beta}$ are created by substituting X, Y elements–Mo, Zn, Co, Cu, Ti, Al, Ga–with $\alpha + \beta < 4$ into iron sites of the original $SmFe_{12}$ structures. Using the optimized structures and formation energies obtained from the first-principles calculations after each active learning cycle, we construct an embedded two-dimensional space to rationally visualize the set of all the calculated and noncalculated structures for monitoring the progress of the search. The optimal model attained a prediction error for the formation energy of 1.25×10^{-2} (eV/atom) using one-sixth of the training data and the recall rate of potentially stable structures was nearly 4 times faster than the random search. The formation energy landscape visualized using the embedding representation revealed that the substitutions of Al and Ga have the highest potential to stabilize the $SmFe_{12}$ structure. Finally, the correlations between the distortion in coordination number and the corresponding formation energy are revealed.

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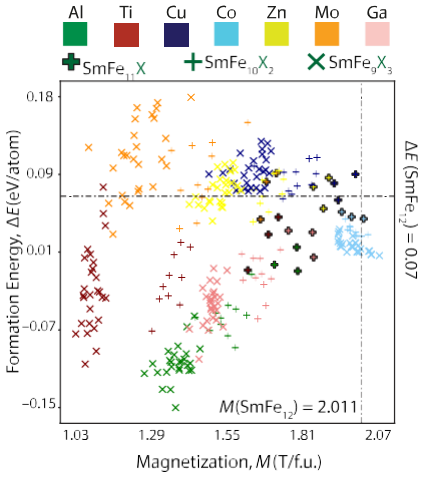

Figure 1: Formation energy and magnetization of SmFe12−αXα used as the initial training data of the active learning circle.

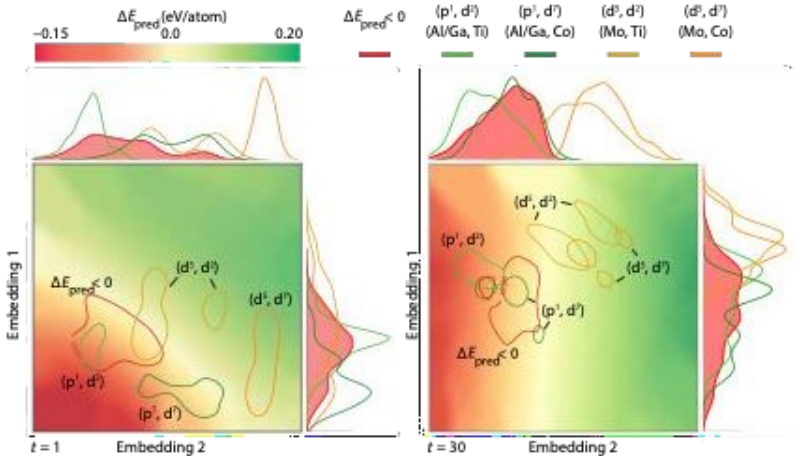

Figure 2: Distribution $SmFe_{12-\alpha-\beta}X_\alpha Y_\beta$ on the formation energy landscape at the initial (left) and the last (right) query time.

Ref: [1] *MRS Bulletin Impact Section*, 1-14, 10.1557/s43577-022-00372-9 (2022). [2] *IUCrJ* 7 (6), 1036-1047, 10.1107/S2052252520010088 (2021). [3] *J. Chem. Phys.* 153, 114111; 10.1063/5.0015977 (2020). [4] *J. Chem. Phys.* 148, 204106 10.1063/1.5021089, (2018).