First-Principles Assessment of Nd-Fe-B Thermodynamic Properties for Application in CALPHAD

The Nd-Fe-B system is of great importance in the field of magnetism. Currently commercially available, powerful permanent magnets use the ternary phase $Nd_2Fe_{14}B$ as a base due to its high magnetisation and relatively robust coercivity. The phase diagram of Nd-Fe-B is therefore of great interest to researchers and engineers. The CALPHAD method of computational thermodynamics can calculate the phase diagram of a system from the thermodynamic data of its constituent phases. An extensive field of study has developed around the CALPHAD method, and it has contributed greatly both in the academic and the industrial world.

While CALPHAD traditionally exclusively utilises reliable experimental data, first-principles evaluation of thermodynamic properties can be implemented to provide these data to CALPHAD. They are, specifically, thermodynamic properties which contribute to the mathematical model of Gibbs energy constructed within CALPHAD. In this work, both formation energy and specific heat (under constant pressure, C_p) of Nd-Fe-B constituent phases are calculated by computational material science techniques. Most Nd-Fe-B constituent phases are classified as 'strongly correlated' materials, in which electronic correlation plays an important role in determining its properties.

Density Functional Theory (DFT) is used as a general framework to calculate total energy of compounds and to calculate forces from finite displacements, used for phonon calculation. A simplified Hubbard correction (DFT+U) [1] is used to correct for strong correlation, and the quasi-harmonic approximation (QHA) is used in phonon calculations to include volume dependence from fitting an energy-volume curve to the Birch Murnaghan equation of state. The resulting formation energies and C_p data from these calculations achieve good agreement with available experimental data. C_p data is reliable for finite temperatures between $20\ K < T < 300\ K$. Both sets of data are valid for use in CALPHAD for calculation of Nd-Fe-B phase diagram.

Keywords: CALPHAD, DFT+U, phonon, Nd-Fe-B, magnets, phase diagram

Reference

[1] M. Cococcioni and S. de Gironcoli, “Linear response approach to the calculation of the effective interaction parameters in the LDA+U method” Phys. Rev. B 71, p. 035105 (2005)