Metastable equilibria unveiled by DFT calculations and CALPHAD extrapolations used in phase field microstructure simulations

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Abstract – The large amount of experimental data establishing phase stability for relevant multiphase, multicomponent materials done in the last 100 years can be captured by the CALPHAD method. Many applications, however, require extrapolations into metastable regions. With the help of the density functional theory (DFT) and the compound energy formalism (CEF) thermodynamic potentials can be calculated for stable as well metastable states. Relevant properties can be provided as input in simulations using the phase field method. The approach is predictive for metastable regions as based in the DFT energetics.

Multicomponent databases created by the CALPHAD method can be visualized as sets of hypersurfaces of the Gibbs energy as each phase in the database is described over a wide range of temperatures and compositions (and pressure). Most of the phases are described for all compositions between the pure elements covering regions far from the stable range of the individual phases.

The Gibbs energy for the pure element, in a given crystallographic configuration (phase) relative to the stable state (phase) of the element is called "lattice stability" [1]. The effect of mixing two or more elements in the same phase is described by interactions or ``excess'' parameters. These have normally been evaluated from the experimental information related to the stable range of the phase in binary and ternary systems. Many different models for these interactions have been developed [2, 3]. Because the Gibbs energies can be extrapolated to areas where no experimental data is known, or are not measurable, the description of the metastable regions can present features that are mainly connected to some artifact created by the model parameters. This fact makes the modeling discussion an important issue in the CALPHAD community even nowadays.

First-principles calculations using the DFT can now provide very accurate values for many ordered compounds at $0~K$. This approach can also be extended to finite temperatures which aim to predict the whole Gibbs energy surface for a binary and ternary system. It is very impressive to be able to predict a transformation temperature with an error $< 200~K$ purely from DFT calculations but the experience from CALPHAD assessments are that a difference of $1~K$ is related to an energy difference of $1~J/mol$ in the Gibbs energies. Such precision in DFT calculations it is very difficult to obtain . Even if the phase diagram is not as the experimentally known, DFT gives a very valuable information about the topology of metastable phase in particular the ordered and disordered configurations on a given lattice. Recent experience shows that using a mean field theory as provided by the CALPHAD method, the combination of experimental and DFT data results into a much improved predictive characterization of metastable regions.

The combination of the best features of DFT together with up-to-date experimental information is used in the project SAPIENS to create an open database for steels which is well documented and sustaintable.

The use of these databases in phase-field simulations of microstructure evolution and kinetics [4], where metastable states are real, provides a step forward to realistic simulations. Detailed illustration of the whole procedure will be reported.

References
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