Structural Energies and Phase Stability in Ni-Cr Alloys

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The structural energy difference between stable bcc and metastable fcc structures of Cr are investigated via a detailed first-principles and phenomenological study of the thermodynamic properties of Ni-Cr alloys. Our work focuses on the large discrepancies between the first-principles calculations of structural energies and those obtained from phase diagram studies. For the Ni-Cr system, we find that the existing phase diagram and thermochemical data are insufficient to assess accurately the structural energies of Cr. Electronic structure calculations based on a tight-binding (TB) approximation for the alloy Hamiltonian are used, together with available thermochemical data, to obtain more reliable estimates of structural energies.

Introduction

In the late 1960's, Kaufman and his co-workers [70Kau] developed a formalism to compute phase diagrams and to evaluate thermochemical quantities in metallic systems. The basis for their approach was a regular- or subregular-solution model for the Gibbs energy of mixing, together with a linear temperature dependence approximation for the Gibbs energy differences of pure elements in difference crystal structures. These Gibbs energy differences, consisting of structural energy and vibrational entropy contributions, are commonly referred to as lattice stability parameters. Lattice stability parameters are key ingredients in the computation of phase diagrams and as such have been the subject of close scrutiny by a number of investigators, either through *ab-initio* and first-principles calculations of structural energies or through the assessment of thermochemical data and phase diagrams.

Pettifor [70Pet] used a hybrid nearly-free-electron TB scheme to calculate the structural energy differences among the fcc, bcc, and cph structures of the transition metals as a function of the *d* band filling. His results were the first to point out that the first-principles calculations gave structural energy differences between the fcc and bcc structures considerably larger than those estimated from the phase diagram studies. The discrepancies were particularly pronounced for elements with nearly half-filled *d* bands, such as Cr, W, and Mo. Essentially the same results and conclusions were obtained by Skriver [85Skr] using the force theorem and a self-consistent linear muffin-tin orbital (LMTO) method within the atomic sphere approximation (ASA).

Subsequently, Miodownik [86Mio] compared and reassessed the lattice stability parameters obtained through first-principles calculations and those proposed by Kaufman [70Kau]. He showed that the gap between the two sets of structural energy differences could be narrowed down appreciably by a suitable revision of the vibrational entropies. The latest semiempirical assessment of lattice stability parameters [88Sau] shows for the 4-*d* and 5-*d* transition metal series considerably better agreement with the first-principles calculations of [85Skr]. However, for the 3-*d* transition metals, and in particular for Cr, the discrepancy between calculated and estimated values of structural energy differences remains large.

Here we use the Ni-Cr system to reexamine the outstanding discrepancies between the first-principles structural energies of Cr in the bcc and fcc structures and those obtained from phase diagram studies. Following Kaufman [73Kau], our phenomenological approach is based on a subregular-solution model for the Gibbs energies of mixing and on a simple linear temperature dependence for the Gibbs energies of the pure elements. Three difference values for the structural energy difference of Cr are examined—the value calculated by Skriver [85Skr], the value assessed by Kaufman [73Kau], and an intermediate value. For all three, we fit our thermodynamic model to thermochemical data, particularly activity data, and to the experimental phase diagram [86Nas]. Finally, we compare the results of the fitting to a first-principles calculation of the enthalpies of formation based on a TB description of the Ni-Cr system [89Tso, 89Kos].

Phenomenological Approach

A subregular-solution model works well for the description of liquid phases. In general, solid phases require that the short-range order (SRO) effects be included in the configuration entropy of formation, using for example the cluster variation method (CVM) [51Kik,

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