CALPHAD: Computer Coupling of Phase Diagrams and Thermochemistry 58 (2017) 138–150

Contents lists available at ScienceDirect

CALPHAD: Computer Coupling of Phase Diagrams and Thermochemistry

journal homepage: www.elsevier.com/locate/calphad

Thermodynamic description of the Al–Ge–Ni system over the whole composition and temperature ranges

Chenyang Zhou, Jiaxin Cui, Cuiping Guo, Changrong Li, Zhenmin Du*

Department of Materials Science and Engineering, University of Science and Technology Beijing, Beijing 100083, PR China

ARTICLE INFO

Keywords:
Al–Ge–Ni system
CALPHAD method
Thermodynamic modeling

ABSTRACT

The phase equilibria and thermodynamic properties of the Al–Ge–Ni system are useful for understanding the diffusion process during the transient liquid phase (TLP) bonding. In this work, the thermodynamic description of the Al–Ge–Ni system over the whole composition and temperature ranges was performed by means of the CALPHAD (CALculation of PHAse Diagrams) method. The enthalpies of mixing of the liquid phase, three isothermal sections at 973, 823, and 673 K and nine vertical sections at 10, 20, 35, 55, 60, 70, 75, and 80 at% Ni and at a constant Al:Ni ratio of 1:3 were taken into account in the present optimization work. A set of self-consistent thermodynamic parameters of the Al–Ge–Ni system was first obtained. The liquidus projection and reaction scheme were constructed according to the thermodynamic parameters obtained in this work. The phase equilibria and thermodynamic properties calculated by the present thermodynamic description show satisfactory agreement with the available experimental information.

1. Introduction

During the last few decades, Ni-based superalloys have attracted considerable attention owing to their striking properties at elevated temperatures, such as high tensile and compressive yield strength, and superior creep and corrosion resistance [1,2] and thus these alloys are extensively found in the aerospace, nuclear power, gas turbine and chemical processing industries [3]. However, conventional fusion welding process is not possible for the formation of most precipitation hardening Ni-based superalloys with a large number of Al and Ti due to their high susceptibility to heat affected zone (HAZ) cracking.

TLP bonding process, first developed by Duvall et al. [4], is one of the most suitable and cost-effective bonding process of the superalloys [5] and has been widely used for jointing Ni-based superalloys [6–11]. In general, the filler materials for the TLP bonding process ideally possess low melting point and similar chemical composition in comparison to the parent materials. Germanium, forming eutectics with Al and Ni, is an ideal candidate as melting point depressant (MPD) elements. For a better understanding of diffusion processing and development of new filler materials, the detailed information about the related phase equilibria and thermodynamic properties is essential, which require reasonable thermodynamic description of the Al–Ge–Ni system.

In the present work, the thermodynamic modeling of the Al–Ge–Ni system over the whole composition and temperature ranges with the CALPHAD method is presented on the basis of the available experimental information. A set of self-consistent and reliable thermodynamic parameters of the Al–Ge–Ni system is first obtained.

2. Literature review

2.1. Al–Ge system

Ansara et al. [12], McAlister and Murray [13] and Srikanth and Chattopadhyay [14] thermodynamically assessed the Al–Ge system and their calculated results were in good agreement with the available experimental data. However, the optimized thermodynamic parameters for the pure elements in their work were not completely taken from the SGTE (Scientific Group Thermodata Europe) database, which could not be easily extrapolated to the multicomponent system. In order to overcome above shortcoming and further assess the Al–Ge–Mg system, Islam et al. [15] re-optimized the Al–Ge system. So the thermodynamic description of the Al–Ge system of Islam et al. [15] is accepted and the calculated Al–Ge phase diagram is shown in Fig. 1.

2.2. Al–Ni system

The thermodynamic assessment of the Al–Ni system was firstly carried out by Kaufman and Nesor [16] and revised by Ansara et al. [17], but the phase Al_3Ni_5 was not included in their work. Then, Du

* Corresponding author.
E-mail address: duzm@ustb.edu.cn (Z. Du).

http://dx.doi.org/10.1016/j.calphad.2017.06.006
Received 28 March 2017; Received in revised form 23 June 2017; Accepted 24 June 2017

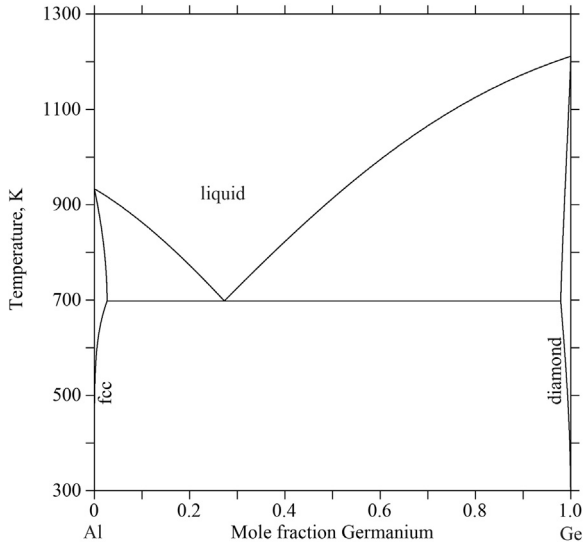

Fig. 1. Calculated Al–Ge phase diagram using the thermodynamic parameters from [15].

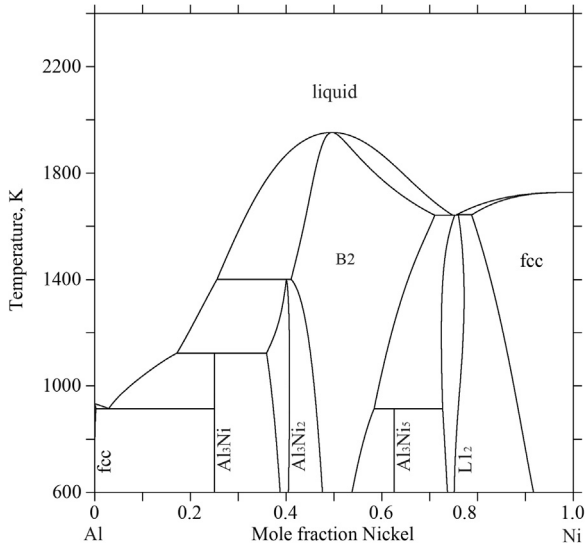

Fig. 2. Calculated Al–Ni phase diagram using the thermodynamic parameters from [21] after refinement.

and Clavaguera [18] considered the phase Al_3Ni_5, used the associate model to describe liquid phase and re-assessed the Al–Ni system, but they did not considered the order-disorder transformation between the phases fcc-A1 and fcc-$L1_2$. Later, Ansara et al. [19] used a single model to represent the transformation from the disordered phase fcc-A1 to ordered one fcc-$L1_2$ for the first time, but Huang and Chang [20] thought a single model with too many parameters was not beneficial to the establishment of the Ni-based database. However, Dupin et al. [21] disagreed with the opinion of Huang and Chang [20] because additional thermodynamic parameters were still used for the ordered phase in their work [20], and thus they revised some thermodynamic parameters on the basis of the assessment of Ansara et al. [19] to correspond with the new experimental data and also used a single function to model the disorder phase bcc-A2 and its ordered one bcc-B2. Recently, Wang and Cacciamani [22] introduced the Al_4Ni_3 phase, but it was not found in the Al–Ge–Ni system. So the thermodynamic modeling of the Al–Ni system of Dupin et al. [21] after refinement is accepted and the calculated Al–Ni phase diagram is presented in Fig. 2. Detail information of the refinement is discussed in Section 4.

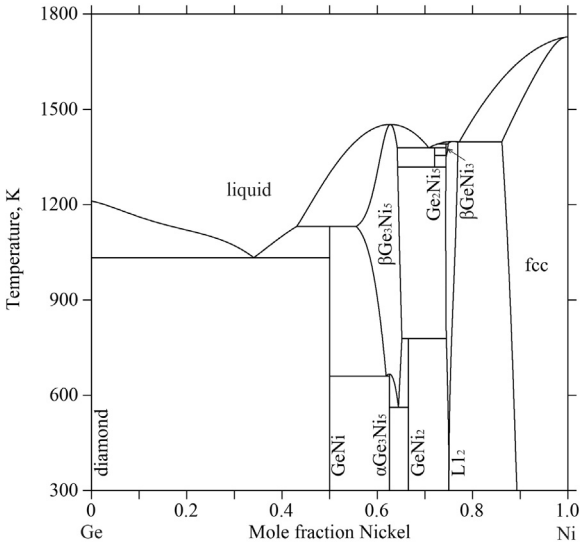

Fig. 3. Calculated Ge–Ni phase diagram using the thermodynamic parameters from [24].

2.3. Ge–Ni system

The thermodynamic description of the Ge–Ni system was performed by Liu et al. [23]. Fundamental consistency was shown between the experimental data and their calculated results, except for the enthalpies of formation of the compounds. Furthermore, the thermodynamic models of the compounds Ge_2Ni_3, $Ge_{12}Ni_{19}$, βGe_3Ni_5 and $\alpha GeNi_3$ in their work were only selected according to the homogeneity ranges. For these reasons, Jin et al. [24] determined and calculated the enthalpies of formation of the partial compounds by direct reaction calorimetry and first-principles calculations, respectively. And then they improved thermodynamic models to match with the corresponding crystal structure, and re-assessed the Ge–Ni system on the basis of the available experimental information. The thermodynamic parameters optimized by Jin et al. [24] are adopted in the present work and the calculated Ge–Ni phase diagram is presented in Fig. 3.

2.4. Al–Ge–Ni system

Ochiai et al. [25] determined the solid solubility between $AlNi_3$ and $\alpha GeNi_3$ by means of metallographic and X-ray diffraction (XRD) methods, and confirmed that a complete solid solution was formed at 1273 K. Yanson et al. [26] used same method as [25] to measure the isothermal section at 770 K in the Al–Ge–Ni system. A ternary phase $AlGeNi_4$ was first found, but its crystal structure was still unknown. These early experimental information were reviewed by Villars et al. [27].

Recently, the Ni-poor part of the Al–Ge–Ni system was investigated using optical microscopic, different thermal analysis (DTA), XRD, scanning electron microscope (SEM) and electron probe microanalysis (EPMA) techniques by Reichmann et al. [28], who detected three ternary phases τ_1, τ_2 (Detailed information of its crystal structure was reported separately [29].) and τ_3. Two isothermal sections at 973 and 673 K and three vertical sections at the $Al_{0.90}Ni_{0.10}$–$Ge_{0.90}Ni_{0.10}$, $Al_{0.80}Ni_{0.20}$–$Ge_{0.80}Ni_{0.20}$, $Al_{0.65}Ni_{0.35}$–$Ge_{0.65}Ni_{0.35}$ joints were constructed, and eleven reactions and the corresponding liquidus projection were derived. Later on, Jandl et al. [30] determined the Ni-rich part of the Al–Ge–Ni system by optical microscopic, DTA, XRD and SEM with energy dispersive X-ray spectroscopy (EDX) measurements, in which two ternary phases τ_4 and τ_5 were confirmed, two isothermal sections at 973 and 823 K were obtained, six vertical sections at 55, 60, 70, 75 and 80 at% Ni and at a constant Al:Ni ratio of 1:3 were determined, and four invariant reactions, the partial liquidus projec-