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Experimental determination and Re-optimization of Ni-Ta binary system①

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[Abstract] Part of the phase boundaries of the solid phases in Ni-Ta system at 1 223 K, 1 173 K and 1 123 K were determined by the diffusion couple and EMPA methods. Based on the experiment data and the information from literatures, the phase equilibrium relationship and thermodynamic properties of the system were analyzed and a complete thermodynamic description were obtained. Apart from the intermetallic phase Ni_8Ta, All the inter-phases were treated with small homogeneity and the sub-lattice model was used to describe them. The calculated results reproduce the experiment information very well.

[Key words] diffusion couple; EMPA; thermodynamics; optimization; phase diagram
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1 INTRODUCTION

Ni-Ta binary alloy has been the subject of much research owing to a combination of scientific and technological interest. On one hand, the addition of tantalum into nickel can stabilize the γ' phase and therefore can improve the mechanical and chemical properties of nickel-based superalloys. On the other hand, the amorphous phases of refractory transition metals exhibit high crystallization temperature[1~4]. To understand and control behavior of Ni-Ta alloys demands a complete and accurate thermodynamic description of this system. Moreover, it is a basis to study some high order systems containing element Ni and Ta.

Since Therkelsen[5] published a paper about Ni-Ta system in 1933, many reports have been found[4, 6~12]. According to Nash's evaluation[13], there exist five inter-phases except liquid and two terminal solid solutions, FCC(Ni) and BCC(Ta) in this system. Kaufman[14] and Ansara et al[15] evaluated the system respectively by CALPHAD approach, but both of them treated all the intermetallic phases as stoichiometric compounds. Cui et al[16] re-evaluated the system with Ni_3Ta, NiTa and $NiTa_2$ treated as non-stoichiometric compounds, and Ni_8Ta and Ni_2Ta as line compounds. In addition, thermodynamic parameters used to describe the system are too many, which is not favorable for the extrapolation of some high order system. This contribution is committed to construct the phase diagram and their thermodynamic description of the Ni-Ta system more completely and accurately in order to facilitate our further study of Ni-Cr-Ta-Zr quater- nary system.

2 EXPERIMENTAL

Starting materials were electrolytic nickel (99.97%), tantalum bar (99.97%) provided by GE Company. The polished Ni and Ta buttons were welded in a GLEEBLE-1500-type Thermal Simulator under the condition of argon protection and a given force at 1 073 K for 10 min to form a well-contacted Ni-Ta binary diffusion couple. The assembled specimen were then cut into three pieces and sealed in silicon capsules filled with purified argon at a pressure of 0.13 Pa. These carefully prepared specimen were heat-treated in a GK-2B-type diffusion furnace, with one group at 1 223 K for 960 h, one group at 1 173 K for 1 200 h and the other at 1 123 K for 1 440 h. The heating temperature was controlled within ±5 ℃. After heat-treatment, the samples were air quenched and polished in the direction parallel to the diffusion direction.

All of the phases were first examined using scanning electron microscopy (SEM), especially the backscattered electron image (BEI). Quantitative EMPA analysis was performed on the JX8800R microprobe using 20 kV voltages and 20 μA current.

3 RESULTS

Fig. 1 shows the backscattered electron image of the diffusion couple specimen. The corresponding schematic diagram of the phase distribution in the specimen is presented in Fig. 2. Table 1 lists the

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phase equilibrium composition determined by EMPA. All of the data are directly obtained from the experiment. As we can see, five intermetallic phases are formed at 1223 K, which correspond to Ni_8Ta, Ni_3Ta, Ni_2Ta, NiTa and $NiTa_2$; but at 1173 K and 1123 K, only Ni_8Ta and Ni_3Ta are formed. The reason for the lack of the other phase are probably from the diffusion kinetics mechanism. Although the diffusion time was very long (960, 1200 and 1440 h respectively), the temperatures are too low for those interphases to form. As we can see, the experimental findings showed in Table 1 are in a very good agreement with the results from CUI et al[16], a little lower than those from Pimenov et al[11].

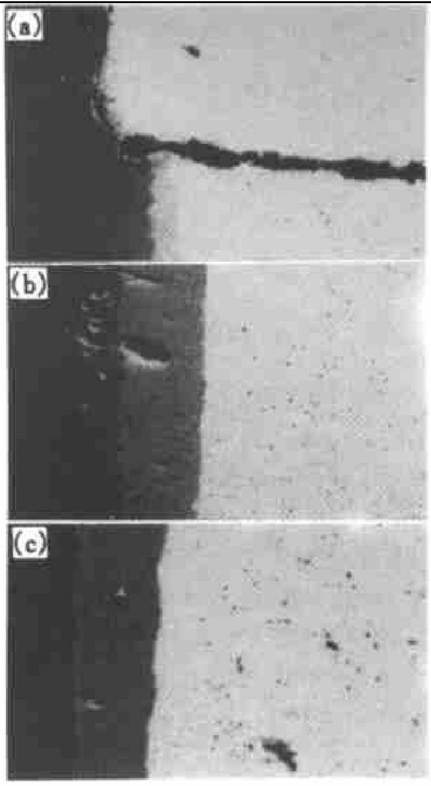

Fig. 1 Backscattered electron images of Ni-Ta diffusion couples
(a) —Annealed at 1223 K for 960 h;
(b) —Annealed at 1173 K for 1200 h;
(c) —Annealed at 1123 K for 1440 h

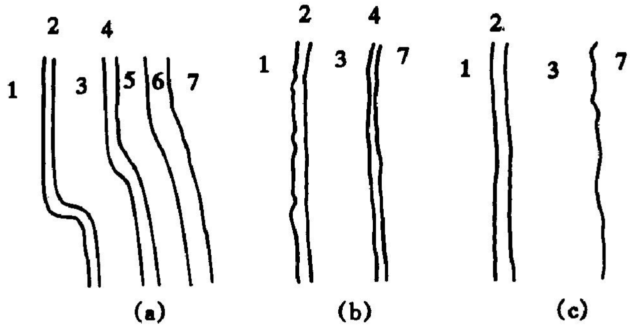

Fig. 2 Schematic diagram of phase distribution
1 —FCC(Ni); 2 —Ni_8Ta; 3 —Ni_3Ta;
4 —Ni_2Ta; 5 —NiTa; 6 —$NiTa_2$; 7 —BCC(Ta);
(a) —Annealed at 1223 K for 960 h;
(b) —Annealed at 1173 K for 1200 h;
(c) —Annealed at 1123 K for 1440 h

4 SELECTION OF EXPERIMENTAL DATA

4.1 Phase equilibria data

Nash et al[13], Ansara et al[15] and CUI et al[16] have evaluated the system. Therkelsen[5] studied the liquidus and solidus temperature by thermal and optical microscopy in the composition range of 0% ~ 55% Ta (mole fraction); while Kubaschewski[6] investigated these in the range of 0% ~ 75% Ta (mole fraction). Kornolov et al[8] investigated the entire system using similar techniques. From them we can see that the compound Ni_3Ta melts congruously at 1823 K. Up to 17.2% Ta (mole fraction) can solve in FCC (Ni) and the maximum solubility of Ni in BCC(Ta) is 20% (mole fraction) at 2061 K. The intermetallic phases NiTa and $NiTa_2$ are formed by peritectic reaction at 1843 K and 2058 K respectively. Nash et al[12] determined that the peritectic equilibrium $Ni(FCC) + Ni_3Ta \rightarrow Ni_8Ta$ takes place at 1593 ~ 1613 K. Pimenov et al[11] determined the homogeneity of the five intermetallic phases in the temperature range of 1423 ~ 1573 K by diffusion methods. All the experiment data mentioned above are taken into account in this assessment and they are shown in Fig. 3.

4.2 Thermodynamic data

Lyakishev et al[17] and Nesterenko et al[18] derived the Gibbs energy of formation for Ni_3Ta and Ni_2Ta from the enthalpy of formation of Ni_2Ta using high temperature solution calorimeter[19]. The enthalpies of formation of the solid phases were also estimated by Colinet et al[20] and Boer et al[21]. All of the data, which are shown in Fig. 4, are accepted in the present work but given different weight. Recently Schaeffers et al[22] measured the mixing enthalpy of the liquid at 2000 K in the Ni-rich with Ta content up to 0.313% (mole fraction) by levitation alloy calorimetry.

Table 1 Phase eqilibrium composition determined by EMPA (mass fraction, %)

Temperature /K	FCC(Ni)	Ni_8Ta		Ni_3Ta		Ni_2Ta		NiTa		$NiTa_2$		BCC(Ta)
1223	3.86	11.04	11.88	23.47	25.12	33.17	33.56	51.12	54.23	66.97	66.97	98.64
1173	4.03	11.35	11.35	24.28	25.27	33.42	–	–	–	–	–	98.90
1123	3.72	11.31	11.31	23.66	24.62	–	–	–	–	–	–	98.50

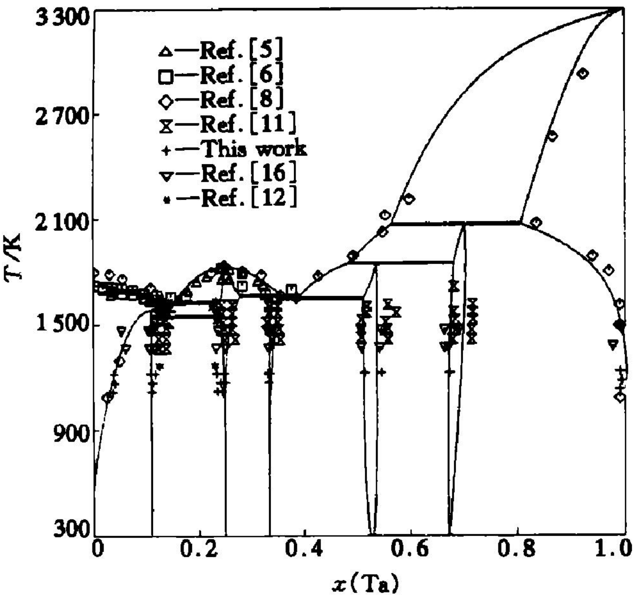

Fig. 3 Comparison between calculated phase diagram and available experimental data

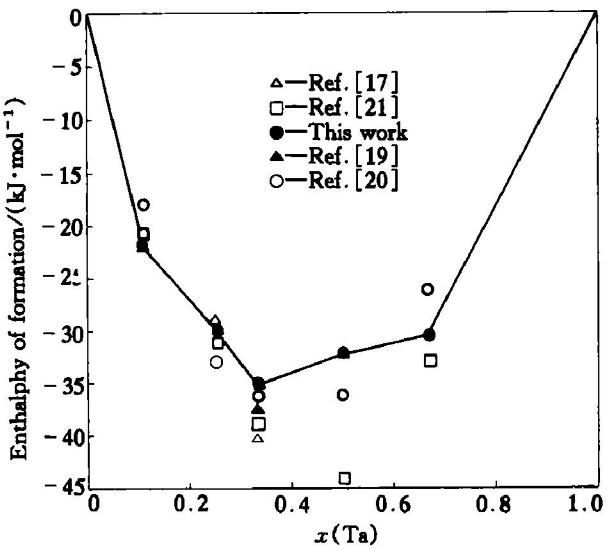

Fig. 4 Comparison between calculated and measured enthalpy of formation for intermetallic phases

4.3 Magnetism

The Curie temperatures of the terminal solution phase FCC(Ni) were measured by Therkelsen[5] and Kubaschewski[6]. The results given by them are very similar and both of them are used in this work.

5 THERMODYNAMIC MODEL

The stabilities of the pure elements are taken from the Scientific Group Thermodata Europe (SGTE)[23]. The reference states are referred to standard state. The substitution modal is used to describe liquid and the terminal solution phases FCC (Ni) and BCC(Ta); while the sublattice modal is used to describe the intermetallic phases. The detailed expressions for the Gibbs energy of the phases are given as follows, all in term of one atom.

5.1 FCC(Ni) and BCC(Ta)

$$G_m^{\varphi} = x_{Ni}{}^0G_{Ni}^{\varphi} + x_{Ta}{}^0G_{Ta}^{\varphi} + RT(x_{Ni}\ln x_{Ni} + x_{Ta}\ln x_{Ta}) + {}^EG_m^{\varphi} + \Delta^m G_m^{\varphi} \qquad (1)$$

where ${}^0G_{Ni}^{\varphi}$ and ${}^0G_{Ta}^{\varphi}$ denote the molar Gibbs energies of pure Ni and pure Ta with the structure of φ in a nonmagnetic state respectively.

The term ${}^EG_m^{\varphi}$ is the excess Gibbs energy expressed in Redlich-Kister polynomials.

$${}^EG_m^{\varphi} = x_{Ni}x_{Ta}\sum_{i=0}^{n} L_{Ni,Ta}^{i}(x_{Ni} - x_{Ta})^{i} \qquad (2)$$

where $L_{Ni,Ta}^{i} = a^{i} + b^{i} * T$ is the interaction parameter and a^{i}, b^{i} need to be evaluated from experimental information.

While the term $\Delta^m G_m^{\varphi}$ represents the magnetic contribution to the molar energies of FCC(Ni) and BCC(Ta):

$$\Delta^m G_m^{\varphi} = RT\ln(\beta + 1)\zeta(\tau) \qquad (3)$$

where τ is defined as T/T_c and T_c is the Curie temperature; β is the Bohr magnetic moment per mole of atom. The function of $\xi(\tau)$ is a polynomial given by Hillert et al[24] as:

$$\xi(\tau) = 1 - 0.86034\tau^{-1} - 0.17499\tau^{3} - 0.007755\tau^{9} - 0.001745\tau^{15} \quad (\tau < 1)$$

$$\xi(\tau) = 0.04269\tau^{-5} - 0.001355\tau^{-15} - 0.0002846\tau^{-25} \quad (\tau > 1) \qquad (4)$$

The concentration dependence of T_c and β is described as follows.

$$V = x_{Ni}{}^0V_{Ni} + x_{Ta}{}^0V_{Ta} + \sum_{i=0}^{n} W_{Ni,Ta}^{i}(x_{Ni} - x_{Ta})^{i} \qquad (5)$$

where V represents either T_c or β, W is the interaction parameter evaluated from the available experimental data.

5.2 Intermetallic compounds

All the intermetallic phases except Ni_8Ta were treated to have some homogeneity. For Ni_8Ta, due to its uncertainty, we treat it as stoichiometric phase. Its Gibbs energy can be expressed as

$$G(Ni_8Ta) - H^{ser} = 0.889{}^0G_{Ni}^{FCC} + 0.111{}^0G_{Ta}^{BCC} + A + BT$$

where A and B are the adjustable parameters to be evaluated for experimental information.

According to Kripyakevich et al[25], the NiTa phase has a rhombohedral structure containing five independent sites in the ratio of 1∶2∶2∶2∶6. We used a four sublattice model $(Ni, Ta)_1(Ta)_4(Ni, Ta)_2(Ni)_6$ to described NiTa phase[6]. This model yields the following expression for the Gibbs energy per mole of atom:

$$G_m = y_{Ni}^{1}(y_{Ni}^{3}\,{}^0G_{Ni:Ta:Ni:Ni} + y_{Ta}^{3}\,{}^0G_{Ni:Ta:Ta:Ni}) + y_{Ta}^{1}(y_{Ni}^{3}\,{}^0G_{Ta:Ta:Ni:Ni} + y_{Ta}^{3}\,{}^0G_{Ta:Ta:Ta:Ni}) + 0.0769RT(y_{Ni}^{1}\ln y_{Ni}^{1} + y_{Ta}^{1}\ln y_{Ta}^{1}) + 0.1538RT(y_{Ni}^{3}\ln y_{Ni}^{3} + y_{Ta}^{3}\ln y_{Ta}^{3}) +$$

$$\begin{aligned}&y^1_{\mathrm{Ni}}y^1_{\mathrm{Ta}}(y^3_{\mathrm{Ni}}L_{\mathrm{Ni,Ta:Ta:Ni:Ni}}+\\&y^3_{\mathrm{Ta}}L_{\mathrm{Ni,Ta:Ta:Ta:Ni}})+\\&y^3_{\mathrm{Ni}}y^3_{\mathrm{Ta}}(y^1_{\mathrm{Ni}}L_{\mathrm{Ni,Ta:Ni,Ta:Ni}}+\\&y^1_{\mathrm{Ta}}L_{\mathrm{Ta:Ta:Ni,Ta:Ni}})\end{aligned}\tag{6}$$

where y^j_i is the site fraction of the element i in the j sublattice, ${}^0G$ represents the Gibbs energy of the compound in the virtual state of NiTa. L is the interaction parameter to be evaluated.

Two-sublattice model: $(\mathrm{Ni},\ \mathrm{Ta})_u(\mathrm{Ni},\ \mathrm{Ta})_v$ was used to described all the other compounds, with the Gibbs energy expressed as:

$$\begin{aligned}G_{\mathrm{m}}=\ &y^1_{\mathrm{Ni}}(y^2_{\mathrm{Ni}}\,{}^0G_{\mathrm{Ni:Ni}}+y^2_{\mathrm{Ta}}\,{}^0G_{\mathrm{Ni:Ta}})+\\&y^1_{\mathrm{Ta}}(y^2_{\mathrm{Ni}}\,{}^0G_{\mathrm{Ta:Ni}}+y^2_{\mathrm{Ta}}\,{}^0G_{\mathrm{Ta:Ta}})+\\&RT[u(y^1_{\mathrm{Ni}}\ln y^1_{\mathrm{Ni}}+y^1_{\mathrm{Ta}}\ln y^1_{\mathrm{Ta}})+\\&v(y^2_{\mathrm{Ni}}\ln y^2_{\mathrm{Ni}}+y^2_{\mathrm{Ta}}\ln y^2_{\mathrm{Ta}})]+\\&y^1_{\mathrm{Ni}}y^1_{\mathrm{Ta}}(y^2_{\mathrm{Ni}}L_{\mathrm{Ni,Ta:Ni}}+y^2_{\mathrm{Ta}}L_{\mathrm{Ni,Ta:Ta}})+\\&y^2_{\mathrm{Ni}}y^2_{\mathrm{Ta}}(y^1_{\mathrm{Ni}}L_{\mathrm{Ni:Ni,Ta}}+y^1_{\mathrm{Ta}}L_{\mathrm{Ta:Ni,Ta}})\end{aligned}\tag{7}$$

For the compound $\mathrm{Ni_3Ta}$, u and v have the value of 0.75 and 0.25 respectively. For the compounds $\mathrm{Ni_2Ta}$ and $\mathrm{NiTa_2}$, the second sublattice is occupied alone by the element Ta. Thus we have the following equations:

$$\left.\begin{aligned}&y^2_{\mathrm{Ni}}\equiv 0\ \text{and}\ y^2_{\mathrm{Ta}}\equiv 1\\&u\equiv 1/3\ \text{and}\ v\equiv 2/3\ (\text{for NiTa}_2)\\&u\equiv 2/3\ \text{and}\ v\equiv 1/3\ (\text{for Ni}_2\text{Ta})\end{aligned}\right\}\tag{8}$$

6 EVALUATION OF THERMODYNAMIC PARAMETERS

The optimization was performed by the Parrot module in the Thermo-Calc computer program cell developed by Sundman et al[26], which works by minimizing the square sum of the differences between experimental and computed values. Each of the selected data was given a certain weight based on the data uncertainties given by the original publication or judged by the modeler, until most of the selected experimental information was reproduced within the expected uncertainty limits.

During the assessment, the thermodynamic description for liquid, FCC and BCC were developed firstly. As mentioned above, it is extremely difficult to fit the only available information from Schaeffers et al[22]. Then the interphases $\mathrm{Ni_3Ta}$, $\mathrm{NiTa_2}$, $\mathrm{Ni_8Ta}$, $\mathrm{Ni_2Ta}$ and NiTa were added one by one according to this order. Finally all the parameters were optimized together to give a better description.

Similar to the treatment of Cui et al[16], we give a value of 5 kJ/mol per atom and 2 kJ/mol per atom to ${}^0G^{\mathrm{Ni_3Ta}}_{\mathrm{Ni:Ni}}$ and ${}^0G^{\mathrm{Ni_3Ta}}_{\mathrm{Ta:Ta}}$ respectively to reduce the number of parameters and simplify the assessment process. Meanwhile, considering the interaction parameter in one sublattice independent of the occupation of the another sublattices, some other constrains are imposed here for $\mathrm{Ni_3Ta}$ phase. That is

$$\left.\begin{aligned}&{}^0G^{\mathrm{Ni_3Ta}}_{\mathrm{Ta:Ni}}={}^0G^{\mathrm{Ni_3Ta}}_{\mathrm{Ni:Ni}}+{}^0G^{\mathrm{Ni_3Ta}}_{\mathrm{Ta:Ta}}-{}^0G^{\mathrm{Ni_3Ta}}_{\mathrm{Ni:Ta}}\\&{}^0L^{\mathrm{Ni_3Ta}}_{\mathrm{Ni,Ta:Ni}}={}^0L^{\mathrm{Ni_3Ta}}_{\mathrm{Ni,Ta:Ta}}\\&{}^0L^{\mathrm{Ni_3Ta}}_{\mathrm{Ni:Ni,Ta}}={}^0L^{\mathrm{Ni_3Ta}}_{\mathrm{Ta:Ni,Ta}}\end{aligned}\right\}\tag{9}$$

likewise, the following restraints are introduced for the NiTa phase:

$$\begin{aligned}{}^0G^{\mathrm{NiTa}}_{\mathrm{Ta:Ta:Ni:Ni}}=\ &{}^0G^{\mathrm{NiTa}}_{\mathrm{Ta:Ta:Ta:Ni}}+{}^0G^{\mathrm{NiTa}}_{\mathrm{Ni:Ta:Ni:Ni}}-\\&{}^0G^{\mathrm{NiTa}}_{\mathrm{Ni:Ta:Ta:Ni}}\end{aligned}\tag{10}$$

And as a further simplification, ${}^0G^{\mathrm{NiTa}}_{\mathrm{Ni:Ta:Ni:Ni}}$ and all the other interaction parameters were fixed to zero. Thus there are only three independent adjustable parameters left for the phase $\mathrm{Ni_3Ta}$ and two for the phase NiTa.

7 DISCUSSION

By means of the computerized optimization, a complete and self-consistent thermodynamic description for the Ni-Ta system is obtained and listed in Table 2. From Fig. 3, it can be seen that the assessments in present work and that of Cui et al are quite alike except the liquidus and solidus of BCC(Ta) at the Ta-rich region and the invariant reaction temperature. The comparison between the calculated phase diagram and the experimental information are in agreement when in consideration of the scatter in the experimental data. Table 3 lists the calculated invariant equilibria together with those reported by Ansara et al[15], Nash et al[13] and Cui et al[16]. As we can see, considering both aspect of the invariant reaction temperature and the equilibria composition, the results from the present work fit the evaluated values by Nash et al[13] very well, superior to those by Ansara et al[15] and Cui et al[16]. Nonetheless much attention should be paid to the invariant reaction: L → $\mathrm{Ni_2Ta}$+ NiTa and L+ $\mathrm{NiTa_2}$ → NiTa. The above two reactions show less temperature difference, while in Nash et al's[13] assessment, these two reactions take place at 1678 K and 1623 K respectively. The temperature difference between the two reactions is up to 55 K, which is thermodynamically impossible as pointed out by Okamoto[27]. Meanwhile, more importance should also be attached to the invariant reaction: FCC(Ni) → $\mathrm{Ni_8Ta}$+ $\mathrm{Ni_3Ta}$. According to Nash et al[13], the compound $\mathrm{Ni_8Ta}$ is supposed to be formed from a peritectoid reaction: FCC(Ni) + $\mathrm{Ni_3Ta}$ → $\mathrm{Ni_8Ta}$, different from the assessment of Ans et al[15] and the present result. Therefore further study is need to classify the truth.

Considering the scatter of the measured and the predicted data, the enthalpies of formation for the intermetallic compounds from the present work are acceptable. Fig. 5 presents the Curie temperature for

Table 2 Thermodynamic descriptions from this work

BCC(Ta):

$$^{0}L^{\mathrm{BCC}}_{\mathrm{Ni,Ta}} = -47\,705.31 - 14.247\,7\,T$$

$$^{1}L^{\mathrm{BCC}}_{\mathrm{Ni,Ta}} = -39\,120.8$$

$$T^{\mathrm{BCC}}_{\mathrm{c_{Ni:Ta}}} = 575$$

$$B^{\mathrm{BCC}}_{\mathrm{Ni:Ta}} = 0.85$$

FCC(Ni):

$$^{0}L^{\mathrm{FCC}}_{\mathrm{Ni,Ta}} = -105\,912.6$$

$$^{1}L^{\mathrm{FCC}}_{\mathrm{Ni,Ta}} = -76\,913.11$$

$$T^{\mathrm{FCC}}_{\mathrm{c_{Ni:Ta}}} = 633$$

$$T^{\mathrm{FCC}}_{\mathrm{c_{Ni:Ta}}} = -3\,820.736\,5$$

$$B^{\mathrm{FCC}}_{\mathrm{Ni:Ta}} = 0.52$$

Ni_3Ta:

$$^{0}G^{\mathrm{Ni_3Ta}}_{\mathrm{Ni:Ta}} = {}^{0}G^{\mathrm{FCC}}_{\mathrm{Ni}} + 5\,000$$

$$^{0}G^{\mathrm{Ni_3Ta}}_{\mathrm{Ni:Ta}} = {}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} + 2\,000$$

$$^{0}G^{\mathrm{Ni_3Ta}}_{\mathrm{Ni:Ta}} = 0.75\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ni}} + 0.25\,{}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} - 31\,485.89 - 1.263\,5\,T$$

$$^{0}G^{\mathrm{Ni_3Ta}}_{\mathrm{Ta:Ni}} = 0.25\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ni}} + 0.75\,{}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} + 7\,000 + 31\,485.89 + 1.263\,5\,T$$

$$^{0}L^{\mathrm{Ni_3Ta}}_{\mathrm{Ni,Ta:Ni}} = {}^{0}L^{\mathrm{Ni_3Ta}}_{\mathrm{Ni,Ta:Ta}} = -3\,405.169$$

$$^{0}L^{\mathrm{Ni_3Ta}}_{\mathrm{Ni:Ni,Ta}} = {}^{0}L^{\mathrm{Ni_3Ta}}_{\mathrm{Ta:Ni,Ta}} = -1\,209.865$$

Ni_8Ta:

$$^{0}G^{\mathrm{Ni_8Ta}}_{\mathrm{Ni:Na}} = 0.889\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ni}} + 0.111\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ta}} - 23\,728.2 + 3.055\,236\,T$$

Ni_2Ta:

$$^{0}G^{\mathrm{Ni_2Ta}}_{\mathrm{Ni:Ta}} = 0.666\,7\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ni}} + 0.333\,3\,{}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} - 36\,367.1 + 1.444\,672\,T$$

$$^{0}G^{\mathrm{Ni_2Ta}}_{\mathrm{Ta:Ta}} = {}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} + 6\,270.021$$

$$^{0}G^{\mathrm{Ni_2Ta}}_{\mathrm{Ni,Ta:Ta}} = {}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} - 0.258\,156$$

NiTa:

$$^{0}G^{\mathrm{NiTa}}_{\mathrm{Ni:Ta:Ni:Ni}} = 0.538\,462\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ni}} + 0.153\,846\,{}^{0}G^{\mathrm{BCC}}_{\mathrm{Ni}} + 0.307\,692\,{}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}}$$

$$^{0}G^{\mathrm{NiTa}}_{\mathrm{Ta:Ta:Ni:Ni}} = 0.461\,538\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ni}} + 0.153\,846\,{}^{0}G^{\mathrm{BCC}}_{\mathrm{Ni}} + 0.307\,69\,{}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} + 0.076\,923\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ta}} - 3.055\,6\times10^{-3} - 0.65\,T$$

$$^{0}G^{\mathrm{NiTa}}_{\mathrm{Ni:Ta:Ta:Ni}} = 0.538\,461\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ni}} + 0.461\,538\,{}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} - 33\,887.459 + 1.231\,656\,8\,T$$

$$^{0}G^{\mathrm{NiTa}}_{\mathrm{Ta:Ta:Ta:Ni}} = 0.461\,538\,4\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ni}} + 0.461\,538\,{}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} + 0.076\,923\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ta}} - 33\,918.015 + 0.581\,650\,87\,T$$

$NiTa_2$:

$$^{0}G^{\mathrm{NiTa_2}}_{\mathrm{Ni:Ta}} = 0.333\,{}^{0}G^{\mathrm{FCC}}_{\mathrm{Ni}} + 0.667\,{}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} - 31\,081.333 + 2.972\,060\,8\,T$$

$$^{0}G^{\mathrm{NiTa_2}}_{\mathrm{Ta:Ta}} = {}^{0}G^{\mathrm{BCC}}_{\mathrm{Ta}} + 40\,590.107$$

Liquid:

$$^{0}L^{\mathrm{L}}_{\mathrm{Ni,Ta}} = -11\,744.79$$

$$^{1}L^{\mathrm{L}}_{\mathrm{Ni,Ta}} = -67\,189.94$$

* Values are given in J/mol of formula unit; value for T_c is given in K and for B in Bohr magnetron, μ_B

Table 3 Invariant reactions

Reaction	Reaction type	T/K	Composition			Ref.
L $\leftrightarrow$ FCC(Ni) + Ni_3Ta	Eutectic	1641	0.146	0.135	0.250	[15]
		1633	0.172	0.140	0.233	[13]
		1639	0.137	0.113	0.237	[16]
		1625	0.154	0.140	0.242	This work
L $\leftrightarrow$ Ni_3Ta	Congruent	1820	0.250	0.250		[15]
		1823	0.250	0.250		[13]
		1820	0.252	0.252		[16]
		1824	0.251	0.251		This work
FCC(Ni) $\leftrightarrow$ Ni_8Ta	Congruent	1578	0.111	0.111		[15]
		1580	0.111	0.111		[16]
		1584	0.111	0.111		This work
FCC(Ni) $\leftrightarrow$ Ni_8Ta + Ni_3Ta	Eutectic	1555	0.128	0.111	0.250	[15]
	Peritectic	1603	0.110	0.125	0.225	[16]
	Eutectic	1547	0.136	0.111	0.242	This work
L + Ni_3Ta $\leftrightarrow$ Ni_2Ta	Peritectic	1676	0.357	0.25	0.333	[15]
		1678	0.350	0.285	0.340	[13]
		1677	0.363	0.275	0.333	[16]
		1663	0.357	0.280	0.341	This work
L $\leftrightarrow$ Ni_2Ta + NiTa	Eutectic	1674	0.362	0.333	0.500	[15]
		1623	0.360	0.375	0.350	[13]
		1668	0.500	0.385	0.333	[16]
		1644	0.350	0.500	0.509	This work
L + $NiTa_2$ $\leftrightarrow$ NiTa	Peritectic	1843	0.480	0.666	0.500	[15]
		1843	0.489	0.667	0.510	[13]
		1843	0.495	0.669	0.500	[16]
		1841	0.481	0.678	0.534	This work
L + BCC(Ta) $\leftrightarrow$ $NiTa_2$	Peritectic	2059	0.578	0.788	0.666	[15]
		2061	0.543	0.800	0.665	[13]
		2065	0.599	0.792	0.693	[16]
		2060	0.561	0.800	0.697	This work

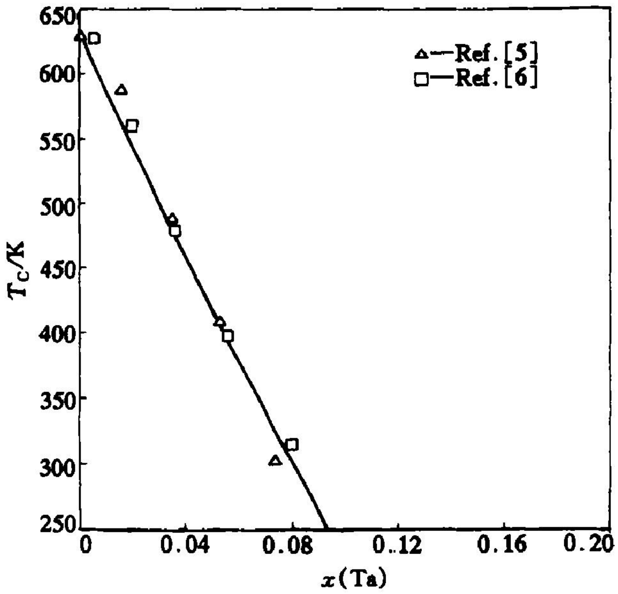

Fig. 5 Comparison between calculated and measured curie temperature of FCC(Ni) solid solution

FCC(Ni) solid solution. As we can see, the calculated results reproduce the measured value perfectly.

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