Thermodynamic Database and the Phase Diagrams of the (U, Th, Pu)-X Binary Systems

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A thermodynamic database for nuclear materials, including U-Th, U-Pu, Th-Pu, and (U, Th, Pu)-X (X = Al, Co, Cr, Cu, Fe, Ga, Mg, Mn, Mo, Nb, Ni, Si, Ta, W, Zr) binary system has been developed by the Calculation of Phase Diagrams (CALPHAD) method. Thermodynamic parameters describing Gibbs free energies of different phases have been evaluated by optimizing experimental data on phase equilibria and thermodynamic properties. The present thermodynamic database can provide much-needed information such as stable and metastable phase equilibria, phase fractions, and various thermodynamic quantities that is important to the design of nuclear materials. This database is also an essential starting point to construct thermodynamic databases for the multicomponent systems.

1. Introduction

With ever increasing global demands for energy and rapid depletion of fossil fuel, nuclear energy will become an important part of the global energy portfolio.[1] To achieve nuclear energy's full potential, new nuclear materials need to be developed for new generations of reactors. The use of uranium (U), thorium (Th), and plutonium (Pu) in the production of nuclear energy has given considerable impetus to the investigation of nuclear materials in recent decades. The investigations of the U, Th, and Pu base alloys focus not only on the U, Th, and Pu compounds used in the fuels, but also on the structural materials and fission products of the U, Th, and Pu base alloys.[2-4] In order to develop nuclear materials with better performance, a thermodynamic database of the U, Th, and Pu alloys for reliable predictions of liquidus, phase fraction, equilibrium,

and nonequilibrium solidification behavior in multicomponent system is highly desirable.

The objective of the present study is to develop a thermodynamic database for the phase diagrams of the U, Th, and Pu containing systems including (U, Th, Pu)-X (U, Th, Pu, Al, Co, Cr, Cu, Fe, Ga, Mg, Mn, Mo, Nb, Ni, Si, Ta, W, Zr) binary systems using the Calculation of Phase Diagrams (CALPHAD) method.

2. CALPHAD Method and Thermodynamic Models

2.1 CALPHAD Method

The CALPHAD method is a powerful tool to reduce cost and time during development of materials.[5] The optimization of the thermodynamic parameters of various phases was performed using the PARROT module of the Thermo-Calc software package, which takes various types of experimental data for the optimization process. The experimental data on phase diagrams and thermodynamic properties were used as inputs to the program. A certain weight was given according to the importance of the data, which was changed by trial and error during the assessments until most experimental data were reproduced within the expected uncertainty limits. The thermodynamic database is developed from the assessed parameters. This database consists of 149 phases, which include 131 intermetallic phases, 16 solution phases, L and gas. The descriptions of thermodynamic models in the U, Th, and Pu containing binary systems are given as follows.

2.2 Solution Phases

The Gibbs free energies of solution phases in the (U, Th, Pu)-X binary systems are described by a subregular solution model using the Redlich-Kister formula.[6] Gibbs free

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energy of a solution phase in the (U, Th, Pu)-X binary system is expressed as:

$$G_m^\phi = \sum_i {}^0G_i^\phi x_i + RT \sum_i x_i \ln x_i + {}^EG_m^\phi + {}^{mg}G^\phi, \quad \text{(Eq 1)}$$

where G_i^ϕ is the molar Gibbs free energy of pure element i with the structure ϕ in a nonmagnetic state, which is taken from the lattice stability values for pure elements compiled by Dinsdale,[7] and the term ${}^EG^\phi$ is the excess free energy, which is expressed in the Redlich-Kister polynomial[6] as:

$${}^EG_m^\phi = x_i x_j \left({}^0L_{i,j}^\phi + {}^1L_{i,j}^\phi (x_i - x_j) + {}^2L_{i,j}^\phi (x_i - x_j)^2 + \cdots \right),$$
$$\text{(Eq 2)}$$

where ${}^mL_{i,j}^\phi$ is the interaction energy between i and j atoms, and is expressed as:

$${}^mL_{i,j}^\phi = a + bT + cT \ln(T), \quad \text{(Eq 3)}$$

the parameters of a, b, and c are evaluated based on the experimental data. The term ${}^{mg}G^\phi$ is the magnetic contribution to the Gibbs free energies for the fcc, bcc, and hcp phases in the (U, Th, Pu)-X (X = Co, Cr, Fe, Mn, Ni) systems, can be expressed as[8]:

$${}^{mg}G^\phi = RT \ln(\beta^\phi + 1) f(\tau^\phi), \quad \text{(Eq 4)}$$

where β^ϕ is a quantity related to the total magnetic entropy, which in most cases is set to the Bohr magnetic moment per mole of atoms; τ^ϕ is defined as T/T_c^ϕ, and T_c^ϕ is the critical temperature for magnetic ordering, T_c^ϕ and β^ϕ are described by the following expression:

$$T_c^\phi = {}^0T_{c,i}^\phi x_i + {}^0T_{c,j}^\phi x_j, \quad \text{(Eq 5)}$$

$$\beta^\phi = {}^0\beta_i^\phi x_i + {}^0\beta_j^\phi x_j, \quad \text{(Eq 6)}$$

where ${}^0T_{c,i}^\phi$ and ${}^0T_{c,j}^\phi$ are the curie temperatures for pure i and j (i = Co, Cr, Fe, Mn, or Ni, j = U, Th, or Pu) and ${}^0\beta_i^\phi$ and ${}^0\beta_j^\phi$ are the Bohr magnetic moments for i and j, respectively. The polynomial $f(\tau^\phi)$ is expressed as:

$$f(\tau^\phi) = 1 - \frac{1}{D}\left[\frac{79\tau^{-1}}{140P} + \frac{474}{497}\left(\frac{1}{P} - 1\right)\left(\frac{\tau^3}{6} + \frac{\tau^9}{135} + \frac{\tau^{15}}{600}\right)\right]$$
$$\text{for } \tau \le 1, \quad \text{(Eq 7)}$$
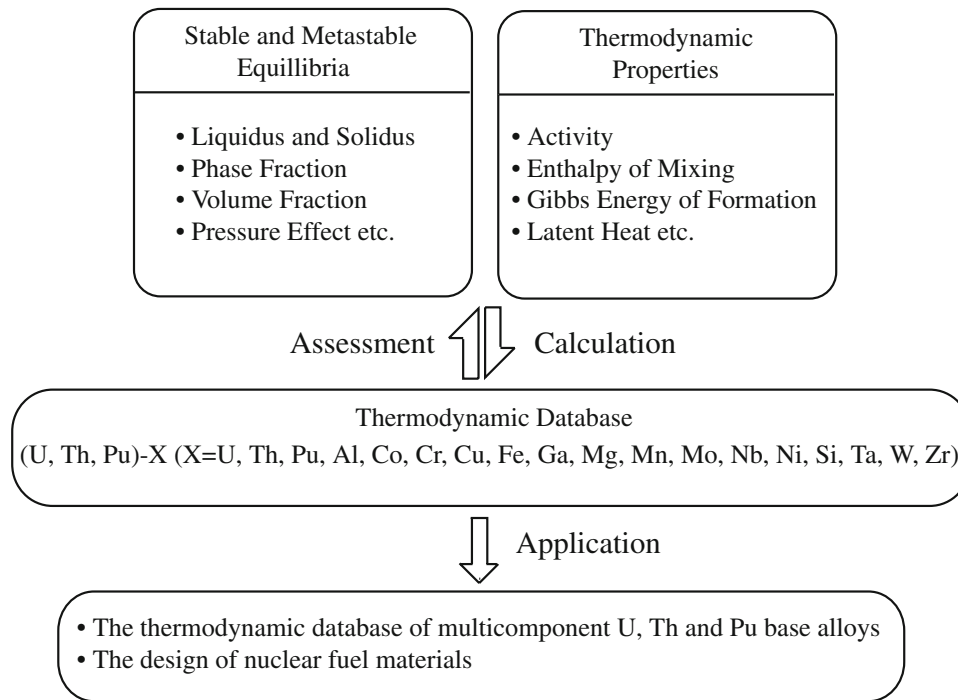
$$f(\tau^\phi) = -\frac{1}{D}\left(\frac{\tau^{-5}}{10} + \frac{\tau^{-15}}{315} + \frac{\tau^{-25}}{1500}\right) \quad \text{for } \tau > 1, \quad \text{(Eq 8)}$$

where $D = \frac{518}{1125} + \frac{11692}{15975}\left(\frac{1}{P} - 1\right)$, and P depending on the structure and is 0.4 for the bcc structure and 0.28 for the other structures.

2.3 Stoichiometric Intermetallic Compounds

The stoichiometric intermetallic compounds in some (U, Th, Pu)-X binary systems, for example, in the U-Ga, U-Mn, Pu-Cu, and Pu-Mg systems, are treated as a formula unit


Stable and Metastable Equillibria	Thermodynamic Properties
• Liquidus and Solidus • Phase Fraction • Volume Fraction • Pressure Effect etc.	• Activity • Enthalpy of Mixing • Gibbs Energy of Formation • Latent Heat etc.

Assessment ⇕ Calculation

Thermodynamic Database
(U, Th, Pu)-X (X=U, Th, Pu, Al, Co, Cr, Cu, Fe, Ga, Mg, Mn, Mo, Nb, Ni, Si, Ta, W, Zr)

⇓ Application

• The thermodynamic database of multicomponent U, Th and Pu base alloys
• The design of nuclear fuel materials

Fig. 1 Outline of the process for constructing the thermodynamic database

Table 1 The assessment status of the (U, Th, Pu)-X binary systems

	U	Th	Pu	Al	Co	Cu	Cr	Fe	Ga	Mg	Mn	Mo	Nb	Ni	Si	Ta	W	Zr
U	...	Li et al.[11]	Kurata[12]	Wang et al.[13]	Wang et al.[13]	Y	Y	Kurata et al.[20]	Wang et al.[16]	Y	Liu et al.[17]	Y	Liu et al.[17]	F	F	F	Wang et al.[16]	Kurata[12]
Th	...	...	Y	Y	Wang et al.[14]	Y	Y	Wang et al.[14]	F	Y	Y	Wang et al.[18]	Y	Y	Y	F	Y	Li et al.[11]
Pu	...	...	...	Turchi et al.[19]	F	Wang et al.[15]	Y	Kurata et al.[21]	Turchi et al.[19]	Wang et al.[15]	Y	Y	Y	F	F	Y	Y	Kurata[12]

Binary systems of the (U, Th, Pu)-X alloy database. Earlier assessed systems are indicated by a reference code or by Y (done work) and those to be assessed are indicated by F (future work)
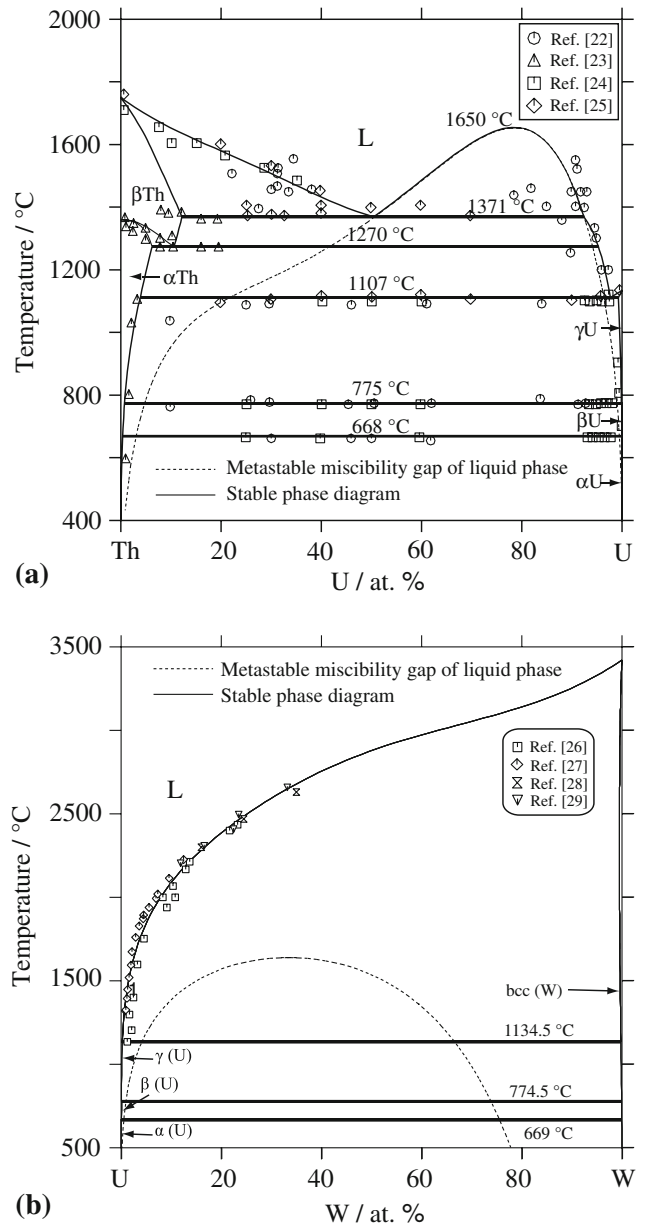
Fig. 2 Calculated U-X binary phase diagrams based on the present assessment in comparison with experimental data: (a) the U-Th system,[11] (b) the U-W system,[16] (c) the U-Nb system,[17] (d) the U-Cr system, (e) the U-Al system,[13] (f) the U-Co system,[13] (g) the U-Ga system,[16] and (h) the U-Mn system[17]

$(A)_a(B)_b$. The Gibbs free energy of formation per mole can be expressed by a two-sublattice model[9]:

$$\Delta G_f^{A_aB_b} = {}^0G_f^{A_aB_b} - a\,{}^0G_A^{ref} - b\,{}^0G_B^{ref} = a' + b'T, \quad \text{(Eq 9)}$$

where the $\Delta G_f^{A_aB_b}$ denotes the standard Gibbs free energy of formation of the stoichiometric compound from pure elements. The terms ${}^0G_A^{ref}$ and ${}^0G_B^{ref}$ are the molar Gibbs free energy of pure element A and B with its defined
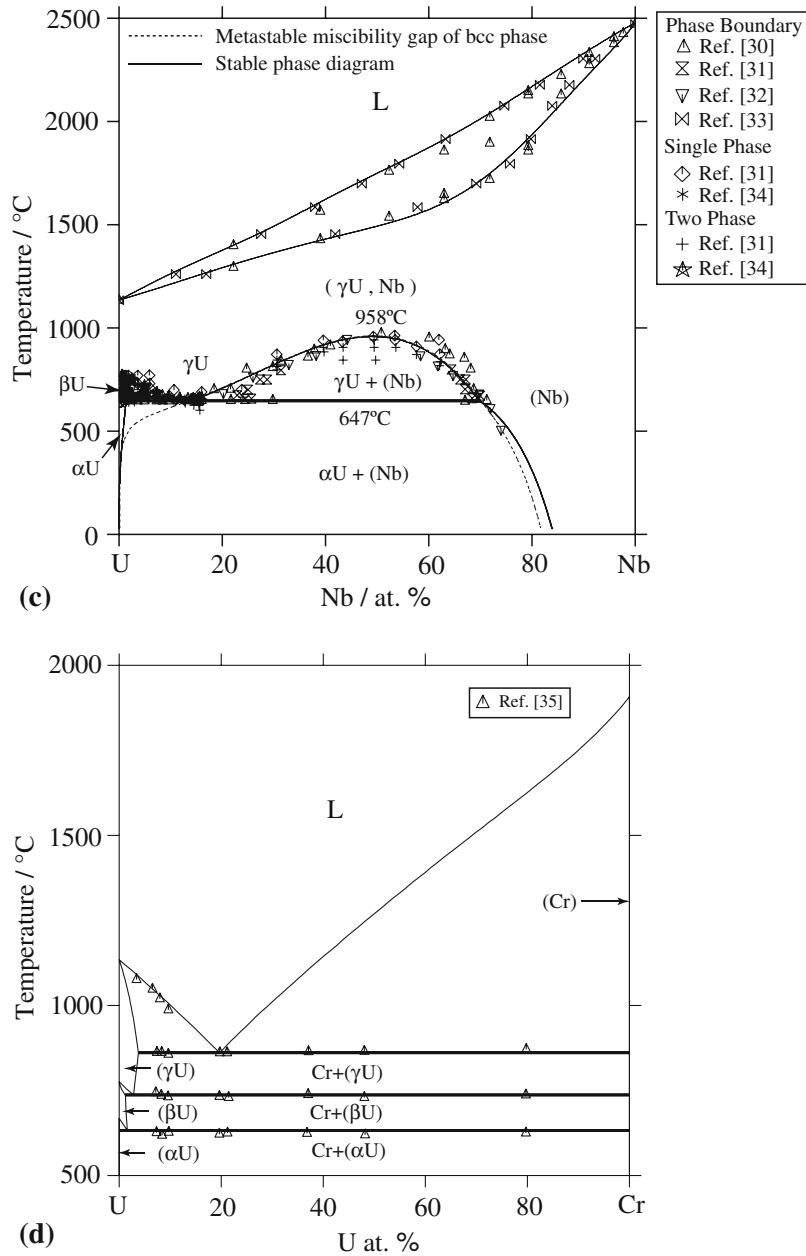
(c)

(d)

Fig. 2 Continued

reference structure in a nonmagnetic state. The parameters of a' and b' are to be evaluated in the present optimization.

2.4 Nonstoichiometric Intermetallic Phases

The Gibbs free energies of nonstoichiometric intermetallic phases in the (U, Th, Pu)-X systems, for example, the UAl$_4$ phase in the U-Al system and the Co$_2$U phase in the U-Co system, are also described by a two-sublattice model.[9] The Gibbs free energy of formation per mole of formula unit $(A, B)_a (A, B)_b$ can be expressed as the following equation referring to the pure elements in their nonmagnetic state:

$$G_{\mathrm{m}}^{(A,B)_a (A,B)_b}$$

$$= y_A^I y_A^{II} G_{A:A} + y_A^I y_B^{II} G_{A:B} + y_B^I y_A^{II} G_{B:A} + y_B^I y_B^{II} G_{B:B}$$

$$+ aRT \left(y_A^I \ln y_A^I + y_B^I \ln y_B^I \right) + bRT \left(y_A^{II} \ln y_A^{II} + y_B^{II} \ln y_B^{II} \right)$$

$$+ a y_A^I y_B^I \left[y_A^{II} \sum_n {}^n L_{A,B:A} \left(y_A^I - y_B^I \right)^n + y_B^{II} \sum_n {}^n L_{A,B:B} \left(y_A^I - y_B^I \right)^n \right]$$

$$+ b y_A^{II} y_B^{II} \left[y_A^I \sum_n {}^n L_{A:A,B} \left(y_A^{II} - y_B^{II} \right)^n + y_B^I \sum_n {}^n L_{B:A,B} \left(y_A^{II} - y_B^{II} \right)^n \right]$$

$$\text{(Eq 10)}$$

where y_i^I and y_j^{II} are, respectively, the site fractions of component i and j (i, j = A or B) in sublattices I and
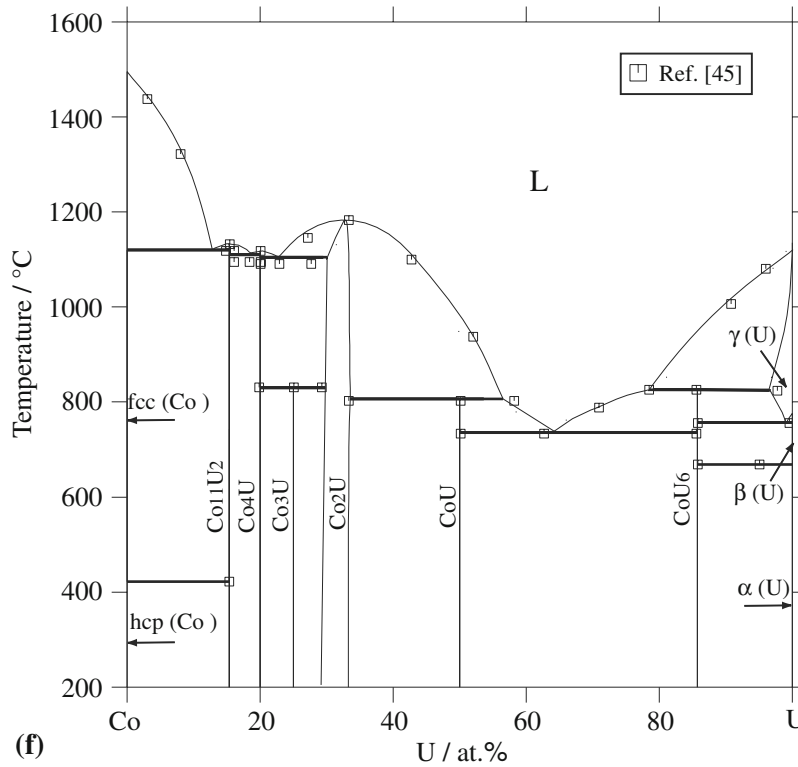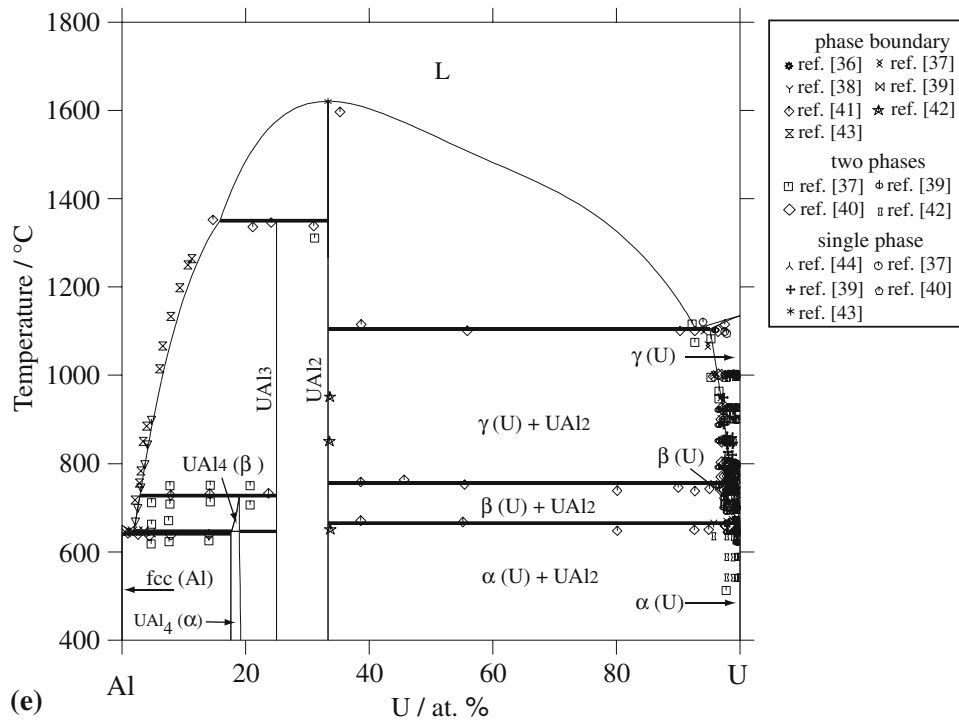
(e)

(f)

Fig. 2 Continued

II; the parameters $G_{i,j}$ represents the Gibbs free energy of the compound phase when the two sublattices are occupied by element i or j, respectively; $L_{i:A,B}$ or $L_{A,B:j}$ is the interaction parameter between A and B in the second or first sublattice, when the first or second sublattice is occupied by element i or j.
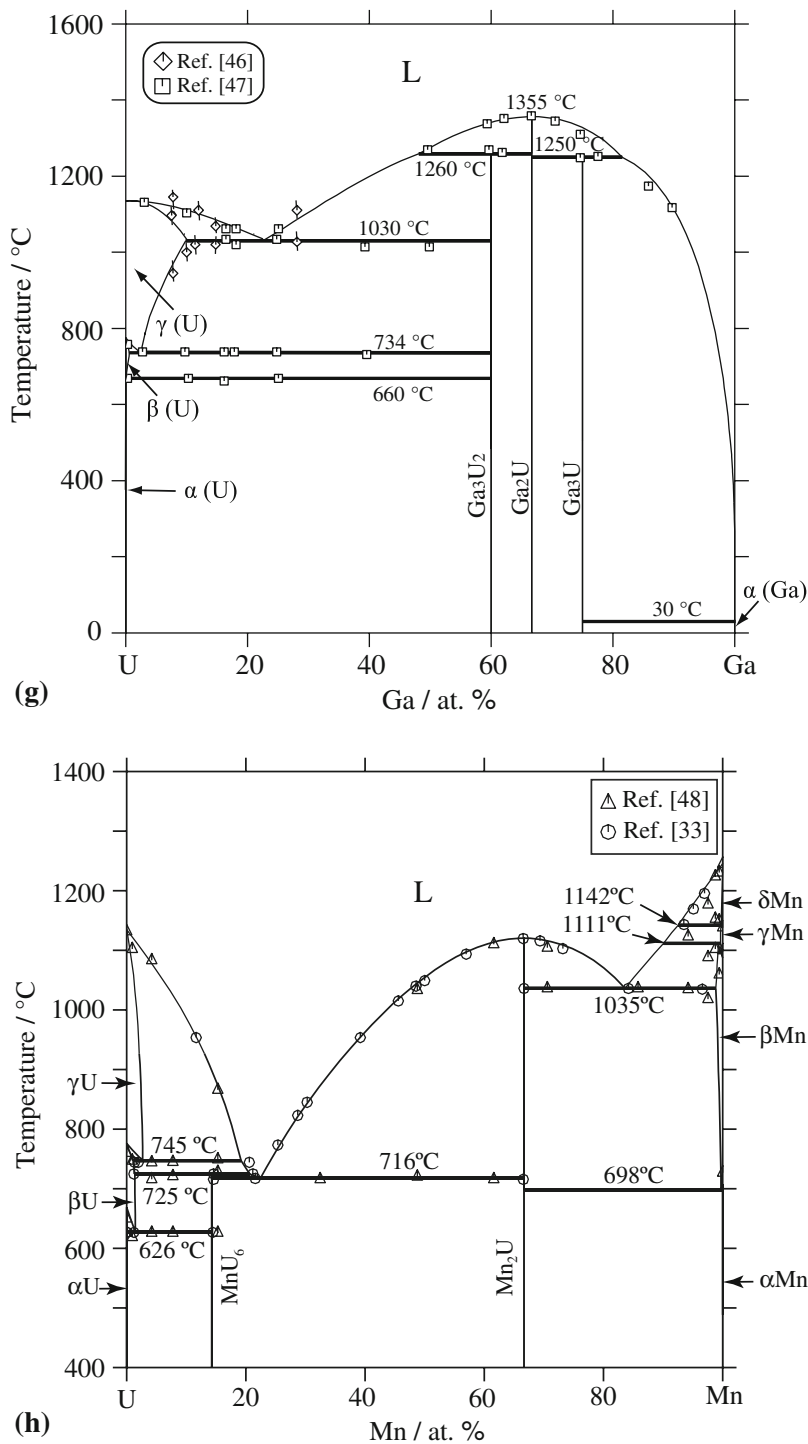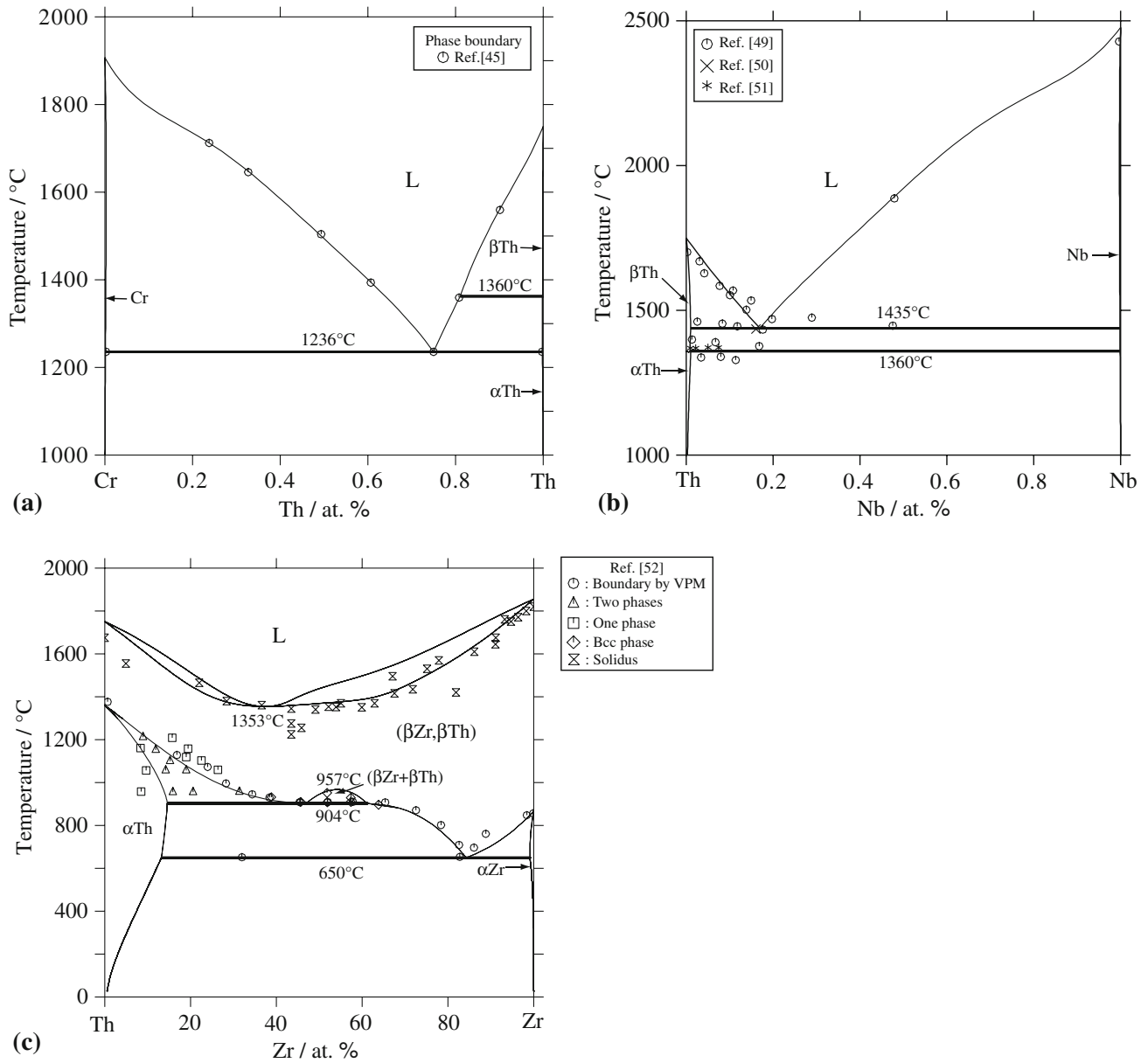
(g)

(h)

Fig. 2 Continued

2.5 Gas Phases

The gas phase in Pu-Mg system is described as an ideal mixture containing the gaseous species Mg, Mg$_2$, and Pu. The Gibbs free energy of the species in the gas phase is given as:
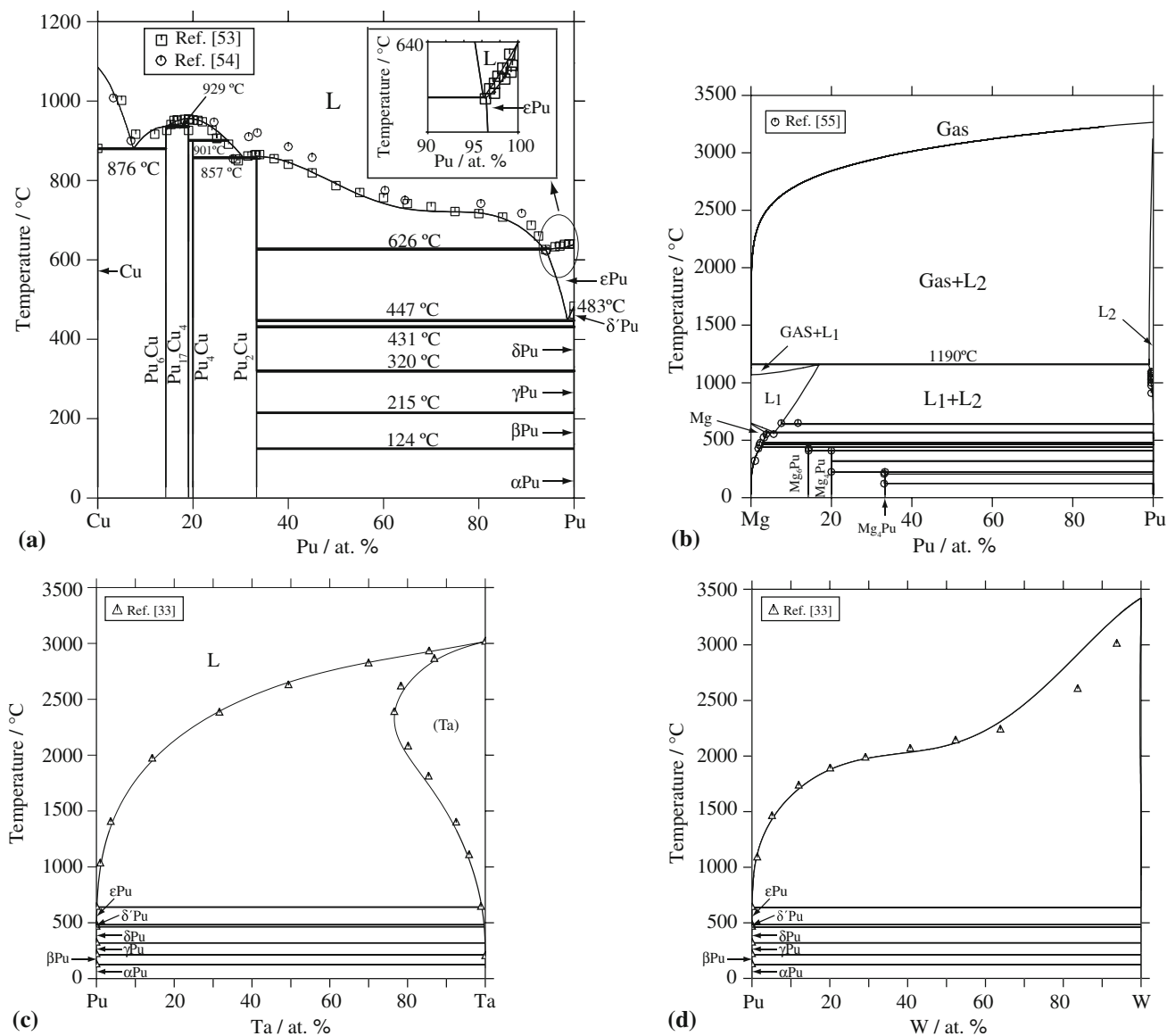
$$G^{\mathrm{gas}} = \sum_i x_i \,^0 G_i^{\mathrm{gas}} + RT \sum_i x_i \ln x_i + RT \ln \left(\frac{P}{P_0} \right), \quad \text{(Eq 11)}$$

where x_i is the mole fraction of the specie in gas phase, $^0 G_i^{\mathrm{gas}}$ is the standard Gibbs free energy of the gaseous
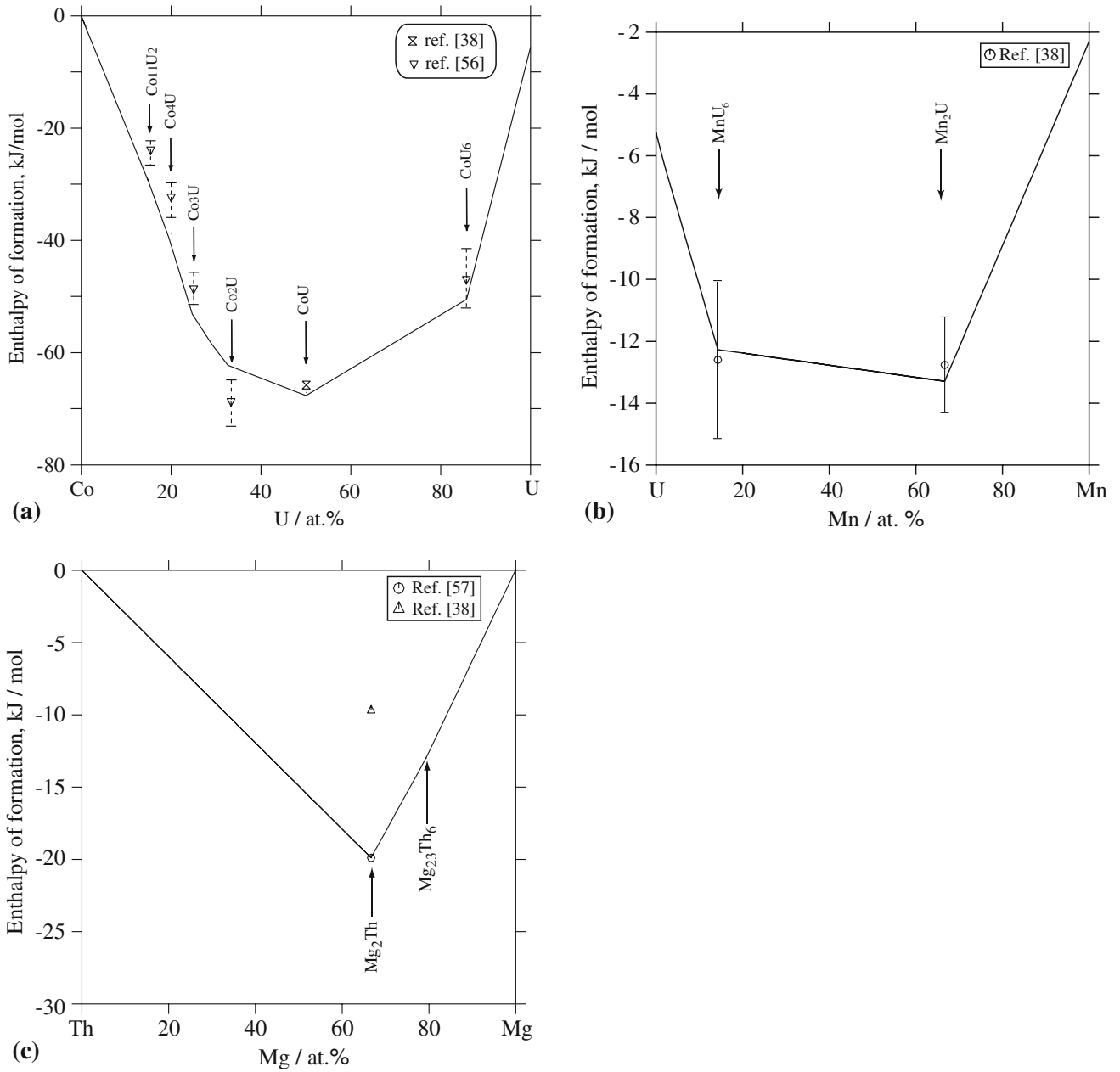
Fig. 3 Calculated Th-X binary phase diagrams based on the present assessment in comparison with experimental data: (a) the Th-Cr system, (b) the Th-Nb system, and (c) the Th-Zr system[11]

Fig. 4 Calculated Pu-X binary phase diagrams based on the present assessment in comparison with experimental data: (a) the Pu-Cu system,[15] (b) the Pu-Mg system,[15] (c) the Pu-Ta system, and (d) the Pu-W system
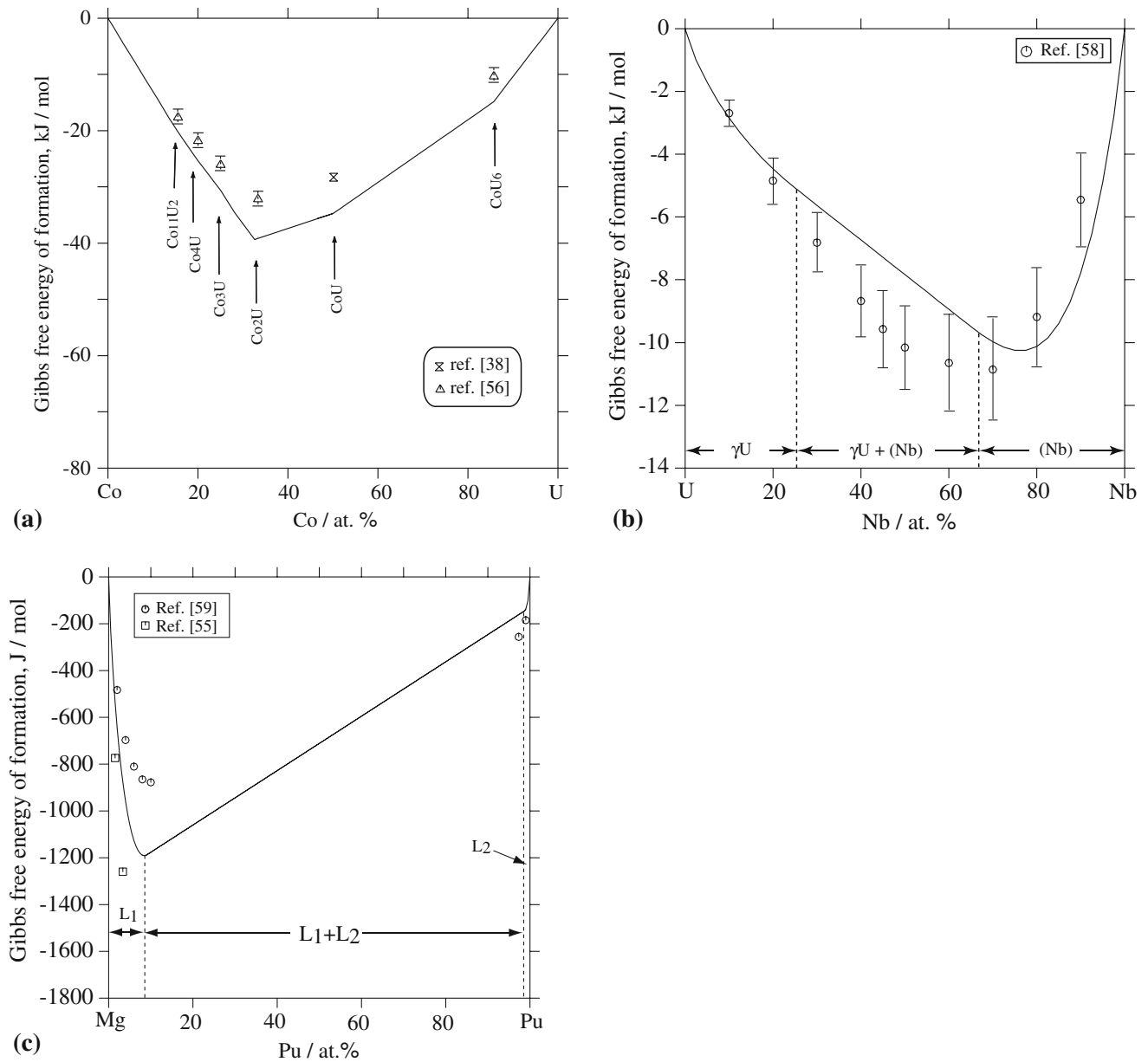
Fig. 5 Calculated enthalpies of formation of intermetallic compounds in several binary systems in comparison with experimental data: (a) the U-Co system at 727 °C[38,56] (The reference states are the γ (U) phase and the fcc (Co) phase), (b) the U-Mn system at 667 °C[38] (The reference states are the γ (U) phase and the β (Mn) phase), and (c) the Th-Mg system at 700 K[38,57] (The reference states are the α (Th) phase and the liquid (Mg) phase)
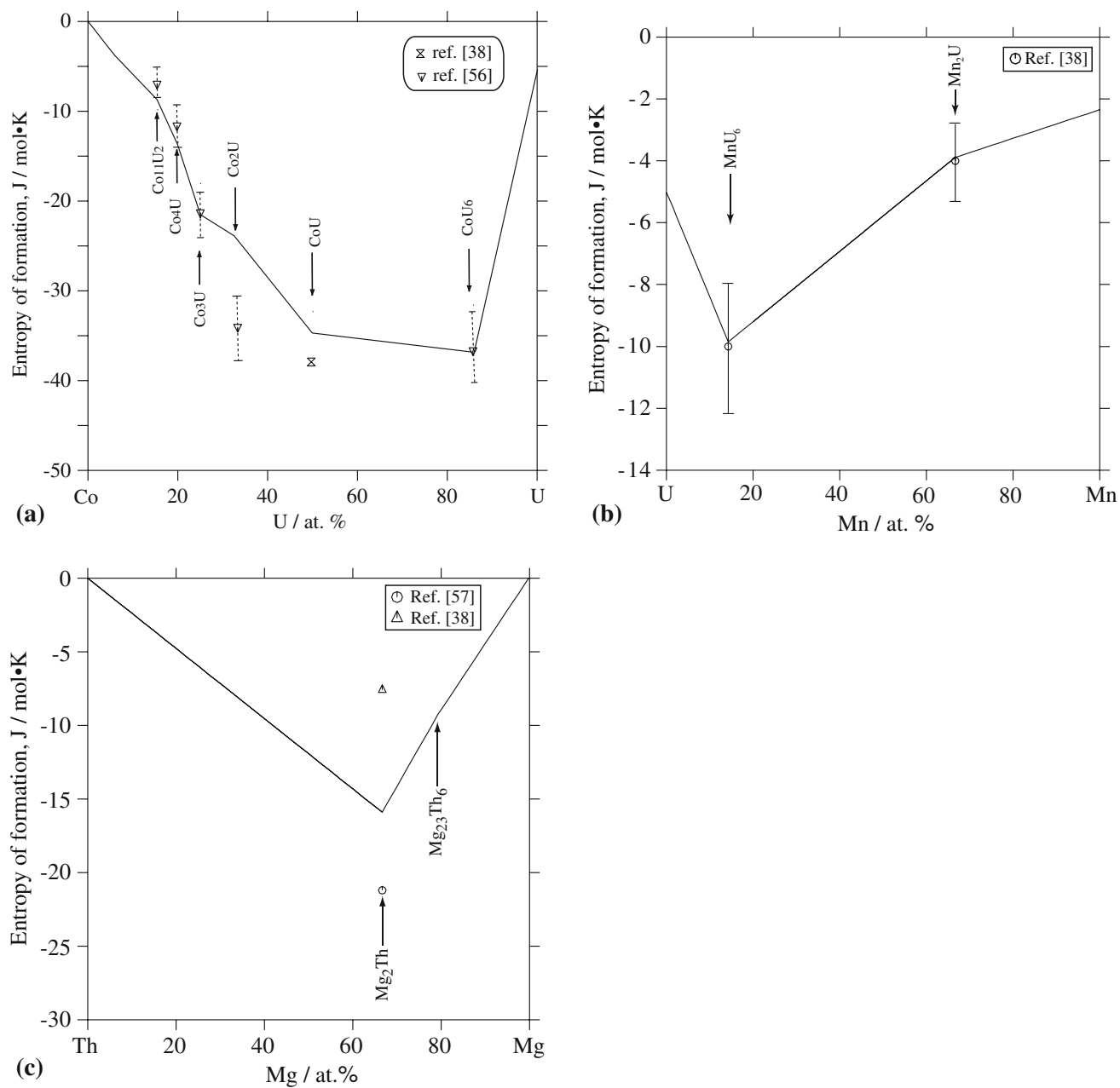
Fig. 6 Calculated entropies of formation of intermetallic compounds in three binary systems in comparison with experimental data: (a) the U-Co system at 727 °C[38,56] (The reference states are the γ (U) phase and the fcc (Co) phase), (b) the U-Mn system at 667 °C[38] (The reference states are the γ (U) phase and the β (Mn) phase), and (c) the Th-Mg system at 700 K[38,57] (The reference states are the α (Th) phase and the liquid (Mg) phase)

Fig. 7 Calculated Gibbs free energies of three binary systems in comparison with experimental data: (a) the U-Co system at 727 °C[38,56] (The reference states are the γ (U) phase and the fcc (Co) phase), (b) the U-Nb system at 775 °C[58] (The reference states are the γ (U) phase and the bcc (Nb) phase), and (c) the Pu-Mg system at 927 °C[55,59] (The reference states are the liquid (Pu) phase and the liquid (Mg) phase)
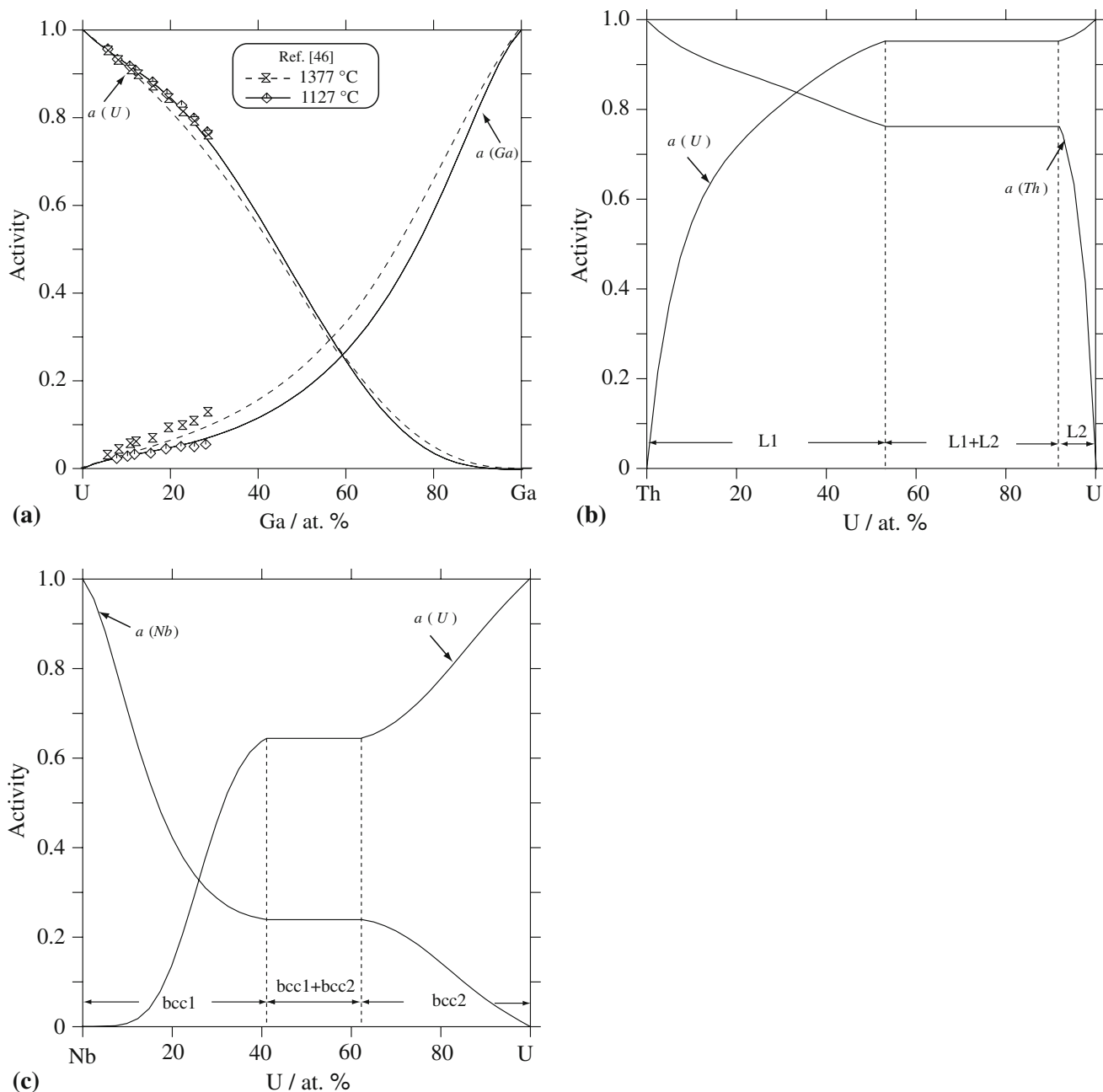
Fig. 8 Calculated activities of three binary systems: (a) activities of Ga and U in the liquid phase at 1127 °C and 1377 °C in the U-Ga system in comparison with experimental data (The reference states are the liquid (Ga) phase and the liquid (U) phase), (b) activities of Th and U in the liquid phase at 1400 °C in the U-Th system (The reference states are the liquid (Th) phase and the liquid (U) phase), and (c) activities of Nb and U in the bcc phase at 900 °C in the U-Nb system (The reference states are the bcc (Nb) phase and the bcc (U) phase)
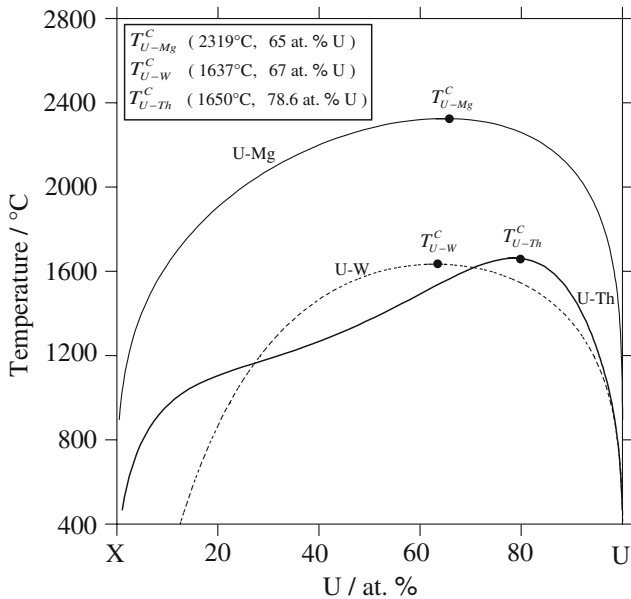
Fig. 9 Calculated miscibility gaps of the liquid phase in the U-X systems

Fig. 10 Calculated miscibility gaps of bcc phase in the U-X systems

species i, which is taken from the SGTE pure element database,[10] R the gas constant, and P_0 the standard pressure at 1 bar.
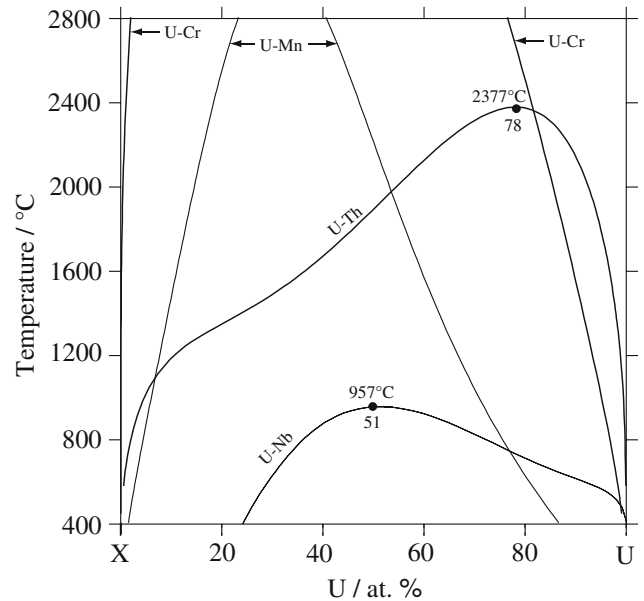
3. Thermodynamic Database and its Applications

3.1 Thermodynamic Database

The thermodynamic optimization of phase equilibria was carried out using available experimental data. Figure 1 shows the scheme of the present thermodynamic database of the U, Th, and Pu base alloys, which provides information on phase diagrams and thermodynamic properties such as liquidus, solidus, mole fraction of constituent phases, activity, enthalpy of mixing, and Gibbs free energy of formation. These data are essential information for the calculation of multicomponent systems. Table 1 summarizes the assessment status of the (U, Th, Pu)-X (X = U, Th,[11] Pu,[12] Al,[13,19] Co,[13,14] Cr, Cu,[15] Fe,[14,20,21] Ga,[16,19] Mg,[15] Mn,[17] Mo,[18] Nb,[17] Ni, Si, Ta, W,[16] Zr[12,13]) binary systems.

3.2 Calculation of Phase Equilibria

Several phase diagrams of the (U, Th, Pu)-X binary systems were calculated using the present binary thermodynamic database for nuclear materials, as shown in Fig. 2, 3, 4. The metastable miscibility gap is presented as broken lines in Fig. 2(a) to (c). It can be seen that miscibility gaps of the liquid phase exist in the U-Th and U-W systems, and a miscibility gap of the bcc phase exists in the U-Nb system. The complex phase diagrams of the U-Cr, U-Al, U-Co, U-Ga, U-Mn, Th-Cr, Th-Nb, Th-Zr, Pu-Cu, Pu-Mg,

Pu-Ta, and Pu-W systems with many intermetallic compounds and invariant reactions can also be calculated using this database, as shown in Fig. 2(d) to 4(d). The calculated results are in good agreement with the experimental data.[22-59]

3.3 Calculation of Thermodynamic Properties

As an example, the calculated thermodynamic properties in some (U, Th, Pu)-X binary systems are shown in Fig. 5 to 8. The calculated enthalpies and entropies of formation of intermetallic compounds in the U-Co, U-Mn, and Th-Mg systems at different temperatures are shown in Fig. 5 and 6. The Gibbs free energies of formation of the U-Co, U-Nb, and Pu-Mg systems at different temperatures are shown in Fig. 7. The activity of the liquid phase in the U-Ga and U-Th system is shown in Fig. 8(a) and (b), and that of the bcc phase in the U-Nb system is shown in Fig. 8(c). And the activity of the liquid phase in the U-Ga system is compared with experimental data, which is in good agreement. From the calculated results, it is obvious that the thermodynamic parameters in the database can be used to calculate both phase diagrams and thermodynamic properties of different phases in the (U, Th, Pu)-X systems.

3.4 Calculation of Metastable Phase Equilibria

Based on the binary thermodynamic database of nuclear materials, metastable phase equilibria, such as the metastable miscibility gap of the liquid and the bcc phases can be estimated. For instance, the calculated metastable miscibility gaps of the liquid phase in the U-Th, U-W, and U-Mg binary systems are shown in Fig. 9, and the calculated metastable miscibility gaps of the bcc phase in the U-Cr, U-Nb, U-Mn, and U-Th binary systems are shown in Fig. 10. The
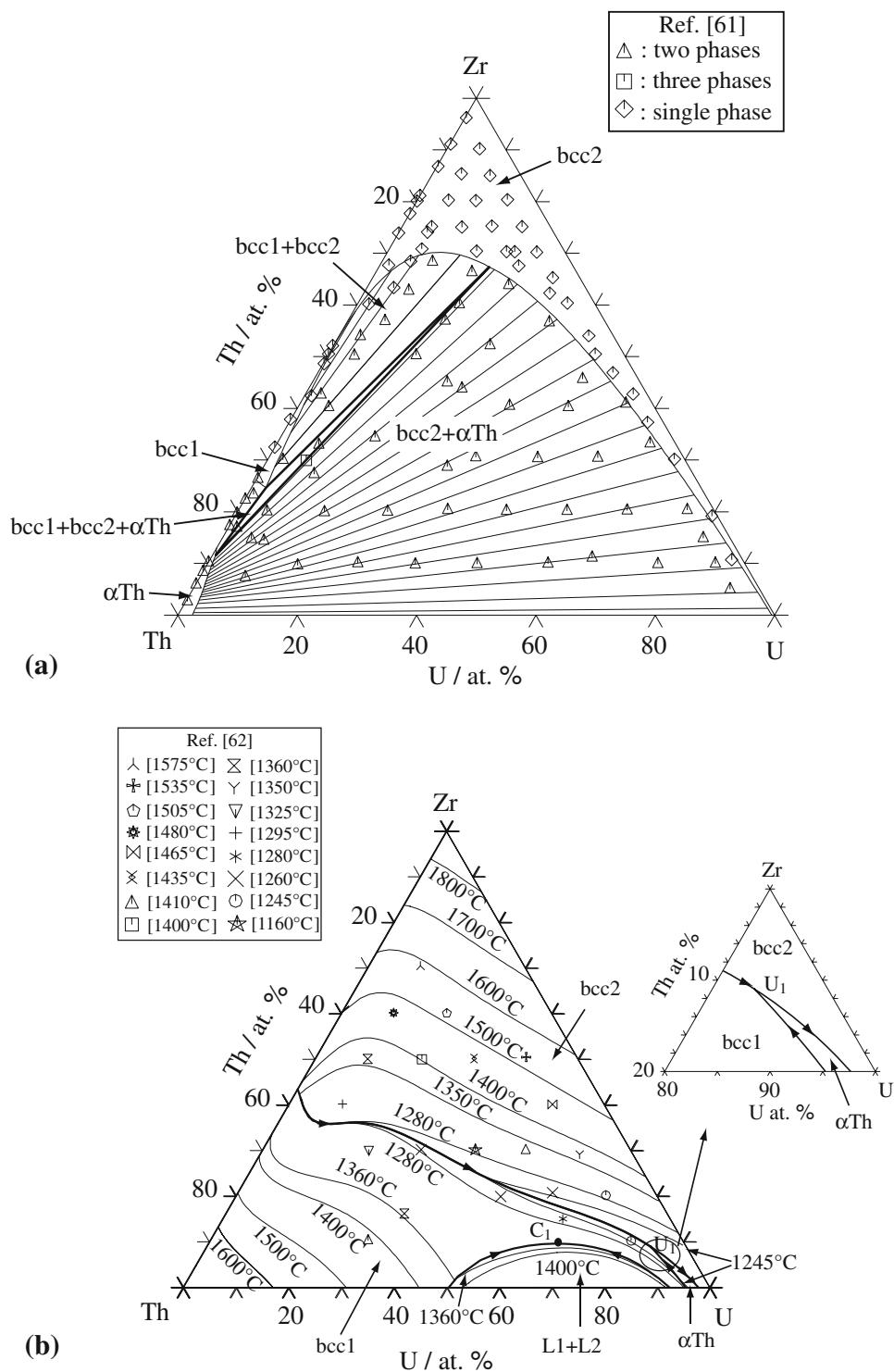
Fig. 11 Calculated ternary diagrams of the Th-U-Zr system: (a) isothermal section at 1000 °C, (b) the liquidus projection, (c) isopleth at U = 1 at.%, and (d) isopleth at U = 4 at. %

calculated critical temperatures and compositions of the bcc miscibility gap in the U-Cr and U-Mn systems are 3072 °C, 31.8 at.% U and 4880 °C, 30.5 at.% U, respectively. It can be seen that the critical temperature of the liquid miscibility gap

in the U-Mg system is higher than that of U-Th or U-W system (e.g., $T^C_{U-Mg} > T^C_{U-Th} > T^C_{U-W}$). Thus, it can be concluded that ΔH of the liquid phase in the U-Mg system is greater than that in the other systems, which is consistent
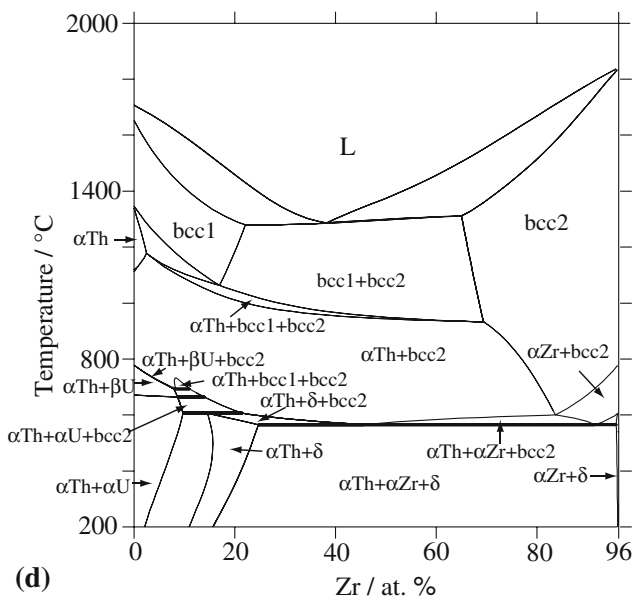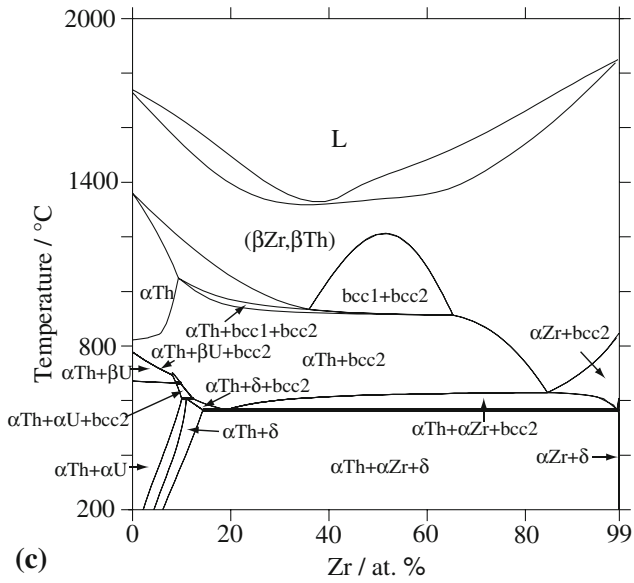
(c)

(d)

Fig. 11 Continued

(a)

(b)

(c)

Fig. 12 Calculated 1000 °C isothermal sections of the Th-U-X systems: (a) Th-U-Cr system, (b) the Th-U-Cu system, (c) the Th-U-Mg system, and (d) the Th-U-Mn system, and (e) the Th-U-Pu system

with the data reported by Niessen et al.[60] (e.g., $\Delta H_{\text{U-Mg}}$ (53 kJ/mol) > $\Delta H_{\text{U-Th}}$ (19 kJ/mol) > $\Delta H_{\text{U-W}}$ (4 kJ/mol)).

3.5 Calculation of Ternary System Diagrams

The present database can be used to estimate ternary phase diagrams when there are no ternary compounds and the solubility of a third element in the binary compounds is negligible. Figure 11(a) and (b) shows a calculated isothermal section and the liquidus projection of the Th-U-Zr system in comparison with experimental data.[61,62] The calculation isopleths at 1 U at.%, and 4 U at.% of the Th-U-Zr system are shown in Fig. 11(c) and (d). The calculations show that the miscibility gap of the bcc phase in
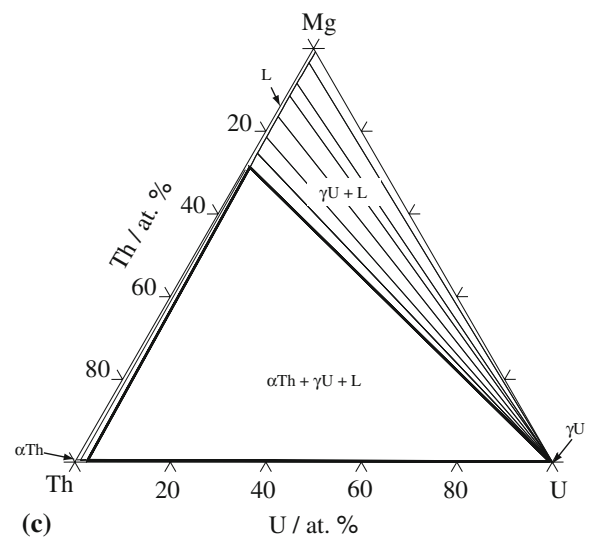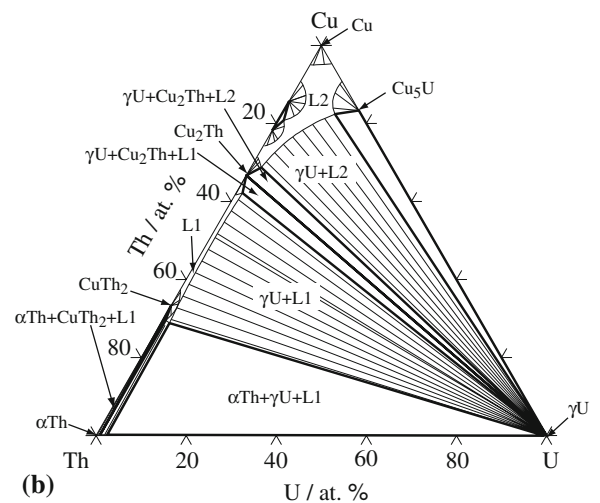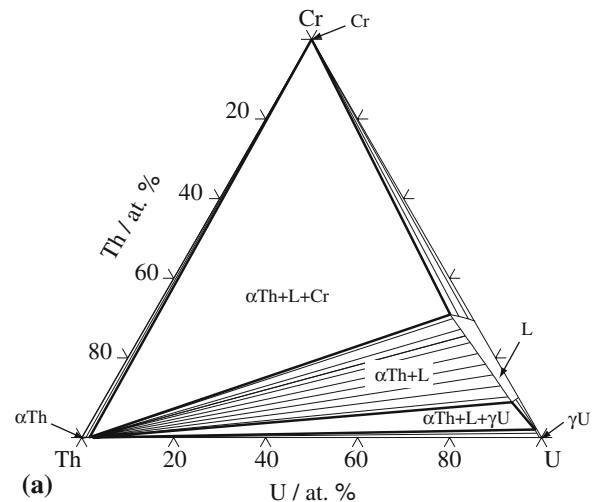
(d)

(e)

Fig. 12 Continued

the Th-U-Zr system becomes more stable as the content of U increases. The calculated isothermal sections in the Th-U-X (X = Cr, Cu, Mg, Mn, Pu) systems without any experimental data are shown in Fig. 12. These calculated results may provide information for the design of U, Th, and Pu base alloys. One should be cautioned that these predictions need to be validated before being used for practical applications since assumptions were made that there are no ternary compounds and little solubility of third elements in binary compounds.

4. Summary

A thermodynamic database for the U, Th, and Pu base alloy systems including (U, Th, Pu)-X (X = U, Th, Pu, Al, Co, Cr, Cu, Fe, Ga, Mg, Mn, Mo, Nb, Ni, Si, Ta, W, and Zr) binary systems was developed. This database can provide much-needed information such as phase diagrams, thermodynamic properties, and metastable phase equilibria for designing U, Th, and Pu base alloys.

Acknowledgments

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