

Thermodynamic Database of the Phase Diagrams in Cu-Fe Base Ternary Systems

C.P. Wang, X.J. Liu, I. Ohnuma, R. Kainuma, and K. Ishida

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A thermodynamic database of the Cu-Fe-X [X: aluminum (Al), cobalt (Co), chromium (Cr), manganese (Mn), molybdenum (Mo), niobium (Nb), nickel (Ni), vanadium (V)] systems was developed by the CALPHAD (Calculation of Phase Diagrams) method, where the Gibbs energies of solution phases such as the liquid, face-centered-cubic (fcc), body-centered-cubic (bcc), and hexagonal-close-packed (hcp) phases are described by the subregular solution model, while the those of the bcc phase in the Cu-Fe-Al system and of all compounds are described by the sublattice model. The thermodynamic parameters describing Gibbs energies of the different phases in this database were evaluated by fitting the experimental data for phase equilibria and thermodynamic properties. On the basis of this database, much information concerning stable and metastable phase equilibria of isothermal and vertical sections, molar fractions of constituent phases, the liquidus projection, etc., can be predicted. This database is expected to play an important role in the design of Cu-Fe base alloys.

1. Introduction

Cu-Fe base alloys are of interest in the practical development of high-strength, high-electrical conductive Cu-rich alloys and age-hardened Fe-rich alloys. Recently, from the environmental point of view, Cu pickup from steel scrap is a problem for the recycling process, because it is difficult to remove Cu in the steel melting process. In face of this situation, a new process for recycling Fe and Cu from the mixed iron-copper scraps has been proposed using separation of two liquid phases between the Fe-rich and Cu-rich phases in the Fe-Cu-C ternary system [1998Yam, 1999Mar]. Investigations of the magnetic properties of the Cu-Fe spinodal-type system have also paid attention to the fundamentals of mechanical alloying and vapor-deposition techniques [1995Drb]. In addition, a new Fe-Cu base alloy containing aluminum (Al) has been developed as a glass die material [1995Tan]. In view of these practical applications of structural and functional Cu-Fe base alloys, a thermodynamic database for phase equilibria is a fundamental tool for alloy design.

The phase diagram of the Cu-Fe binary system reviewed by Okamoto [1993Oka] is shown in Fig. 1. This diagram shows a calculated metastable miscibility gap for the liquid phase [1995Che]. Several studies on this miscibility gap of the liquid phase in Cu-Fe base system have been carried out by various researchers [1938Iwa, 1958Nak, 1997Wil, 1997Tan, 1998Yam, 1999Ama]. Phase equilibria in the Cu-Fe base ternary systems have been investigated by the present group both experimentally and thermo-

dynamically [1997Oht, 1998Wan, 2000Wan, 2001Wan, 2002Wan].

The purpose of the present paper is to construct a thermodynamic database of the phase equilibria of Cu-Fe-X systems on the basis of the CALPHAD method.

2. Thermodynamic Models

The Gibbs energies of the liquid, face-centered-cubic (fcc), body-centered-cubic (bcc), hexagonal-close-packed (hcp), and other solid solution phases are described by the subregular solution model, except for the bcc phase in the Cu-Fe-Al system, with the Redlich-Kister formula [1948Red]. Their Gibbs energies can be expressed as

$$G = \sum_{i=\text{Cu,Fe,X}} {}^0G_i x_i + RT \sum_{i=\text{Cu,Fe,X}} x_i \ln x_i + L_{\text{CuFe}} x_{\text{Cu}} x_{\text{Fe}} + L_{\text{FeX}} x_{\text{Fe}} x_{\text{X}} + L_{\text{CuX}} x_{\text{Cu}} x_{\text{X}} + L_{\text{CuFeX}} x_{\text{Cu}} x_{\text{Fe}} x_{\text{X}} + \Delta^{\text{mag}} G \quad (\text{Eq 1})$$

where 0G_i is the Gibbs energy of pure component i in the respective reference state, x_i is the mole fraction of component i , and L_{ij} and L_{CuFeX} are the temperature and composition dependent interaction energy in binary and ternary systems, respectively. $\Delta^{\text{mag}}G$ is the magnetic contribution to the Gibbs energy. The Gibbs energy of pure component i in its different phase states is taken from the SGTE database [1991Din].

For the Cu-Fe-Al system, the Gibbs energy of the bcc phase is described by the two-sublattice model [1970Hil] based on the experimental results to describe the A2/B2 order-disorder transition, which was reported in our previous work [1998Wan].

C.P. Wang, Department of Materials Science and Engineering, Xiamen University, Xiamen 361005, P.R.China; X.J. Liu, I. Ohnuma, R. Kainuma, and K. Ishida, Department of Materials Science, Graduate School of Engineering, Tohoku University, Sendai 980-8579, Japan. Contact e-mail: ishida@material.tohoku.ac.jp.

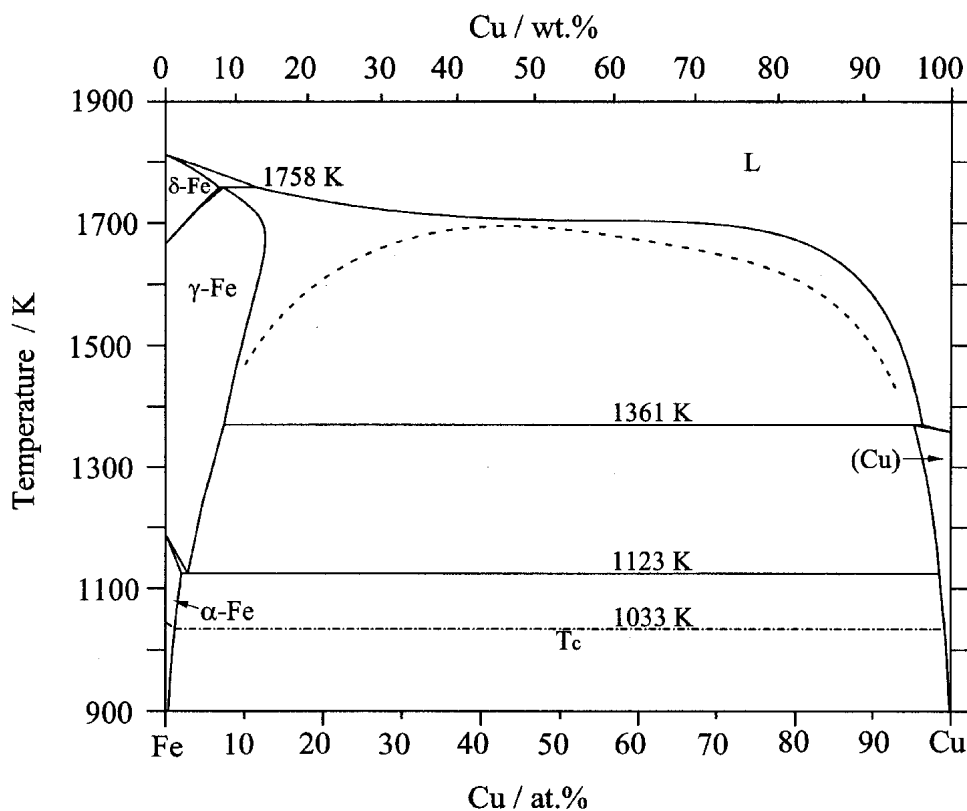


Fig. 1 Fe-Cu phase diagram compiled by Okamoto [1993Oka]

$\Delta^{\text{mag}}G$ is the magnetic contribution to the Gibbs energy, which is described by [1978Hil]:

$$\Delta^{\text{mag}}G = RT \ln(\beta + 1) f(\tau) \quad (\text{Eq 2})$$

The function $f(\tau)$ is formulated by the polynomial of the normalized temperature, τ , where τ is defined as T/T_c^ϕ . T_c^ϕ is the Curie temperature of solution for ferromagnetic ordering, and β is the Bohr magneton number.

Due to the lack of experimental information on the intermetallic phases in the Cu-Fe-X ternary system, no solubilities of the intermetallic phases in the ternary systems are considered in the present assessment.

3. Thermodynamic Database

The present thermodynamic database contains thermodynamic parameters for calculating the phase equilibria in binary and ternary alloys of the Cu-Fe-X [X: Al, cobalt (Co), chromium (Cr), manganese (Mn), molybdenum (Mo), niobium (Nb), nickel (Ni), vanadium (V)] system. The thermodynamic parameters for describing the Gibbs energy of each phase are optimized by fitting the experimental data pertaining to the phase boundary compositions and thermodynamic properties such as activity, heat of mixing, and

Table 1 Assessed Fe-X and Cu-X Binary Systems Used in the Present Calculation

Fe-X	Reference	Cu-X	Reference
Fe-Cu	[1995Che]	Cu-Co	[2000Kub]
Fe-Co	[1988Gui1]	Cu-Cr	[1990Ham]
Fe-Cr	[1987And]	Cu-V	[1990Ham]
Fe-V	[1991Hua]	Cu-Al	[1998Liu]
Fe-Al	[1998Ohn]	Cu-Mn	[1998Liu]
Fe-Mn	[1993Lee]	Cu-Mo	[2000Wan]
Fe-Mo	[1988And]	Cu-Nb	[1990Ham]
Fe-Nb	[1990Hua]	Cu-Ni	[1992Mey]
Fe-Ni	[1988Gui2]		

enthalpy of formation. For some alloy systems for which there was little or no experimental data, the phase equilibria on the Cu-Fe side were experimentally determined in order to obtain a better estimation of the thermodynamic parameters with good agreement between the calculated and experimental data [1938Dan, 1970Ahm, 1997Oht, 1998Wan, 2000Wan, 2000Kim, 2001Wan, 2002Wan, 2003Wan]. The thermodynamic parameters were evaluated by Thermo-Calc software, which was originally developed by Sundman et al. [1985Sun]. The thermodynamic descriptions for Cu-X and Fe-X subsystems are taken from the literature and are summarized in Table 1.

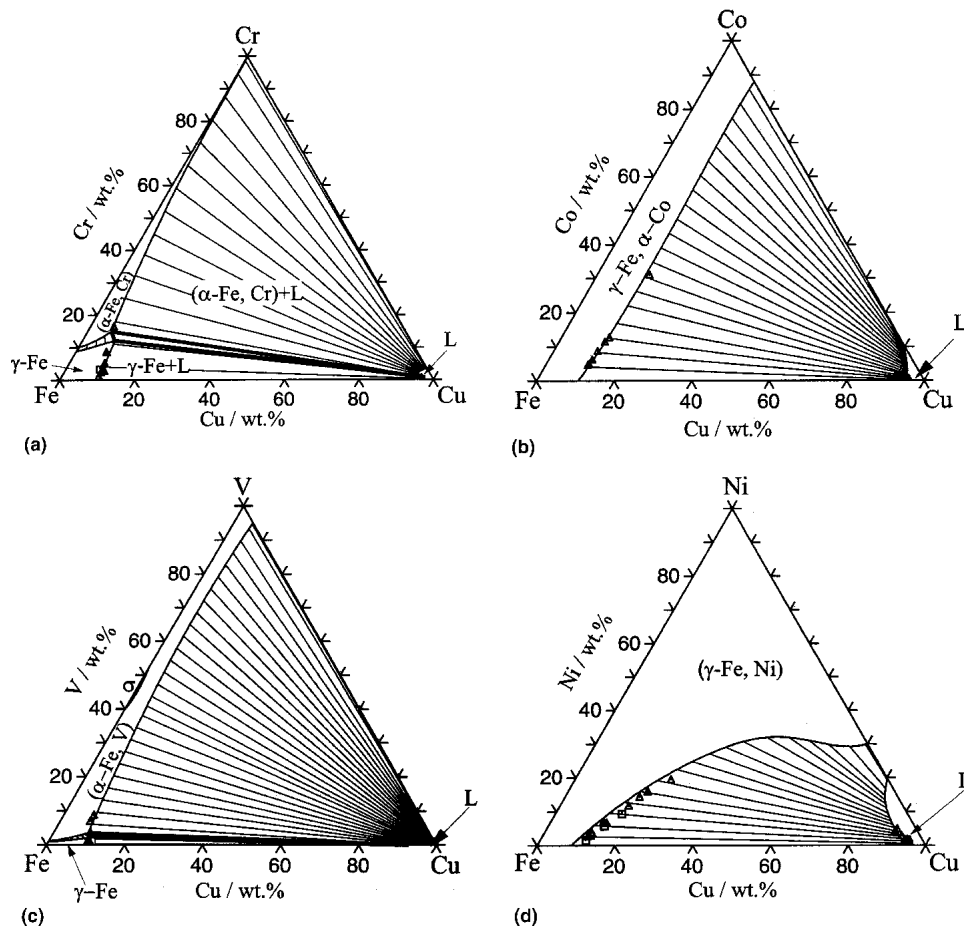


Fig. 2 Calculated isothermal section diagrams at 1473 K in the (a) Fe-Cu-Cr, (b) Fe-Cu-Co, (c) Fe-Cu-V, (d) Fe-Cu-Ni systems

The present thermodynamic database can provide much information on stable and metastable phase equilibria, phase fraction, and various thermodynamic quantities such as activity, mixing enthalpy, Gibbs energy of formation, driving forces for phase transformation, simulation of solidification, etc. In addition, *A2/B2* order-disorder transition in the region of the bcc phase can also be predicted in the Cu-Fe-Al system.

4. Information Obtained from the Database

4.1 Phase Equilibria

On the basis of the evaluated thermodynamic parameters, the phase equilibria in the Cu-Fe-X (X: Al, Co, Cr, Mn, Mo, Nb, Ni, V) ternary system in the whole range of composition and temperature above 673 K can be calculated. As an example, the calculated isothermal sections for the Cu-Fe-X systems at 1473 K are shown in Fig. 2, where the calculated results show good agreement with the experimental data for the Cu-Fe side. The *A2/B2* transition in the Cu-Fe-Al system is discussed in the following section. In the Cu-Fe-Mo and Cu-Fe-Nb systems, there is a three-phase region composed of the liquid, α - or γ -Fe, and the intermetallic compound of the Fe-X system.

The calculated vertical sections at 10 wt.% Mo and 10 wt.% Co in the Cu-Fe-Mo and Cu-Fe-Co systems are shown in Fig. 3, where a metastable miscibility gap in the liquid phase appears due to the addition of Mo, while no such gap results from the addition of Co.

4.2 Order-Disorder Transition

The order-disorder transition of the bcc phase should exist in the Cu-Fe-Al system because there are stable and metastable order-disorder transitions of the bcc phase in the Fe-Al [1993Kat] and Cu-Al [1998Kai] binary systems. As previously reported on the phase equilibria in the Cu-Fe-Al system [1998Wan], some typical examples of isothermal and vertical sections related to the ordering are represented in Fig. 4. Figure 4(a) shows the isothermal section at 1273 K, where the stable *B2* ordered bcc phase region exists in a wide composition range from the Fe-Al side to the Cu-Al side, and the miscibility gaps observed in the bcc phase region of the Cu-Fe-Al ternary can be divided into three regions, namely, *A2* + *A2*, *A2* + *B2*, and *B2* + *B2*. The calculated vertical section at 15 wt.% Al contents is shown in Fig. 4(b), where the stable *A2/B2* transition with a critical temperature of about 1400 K appears.

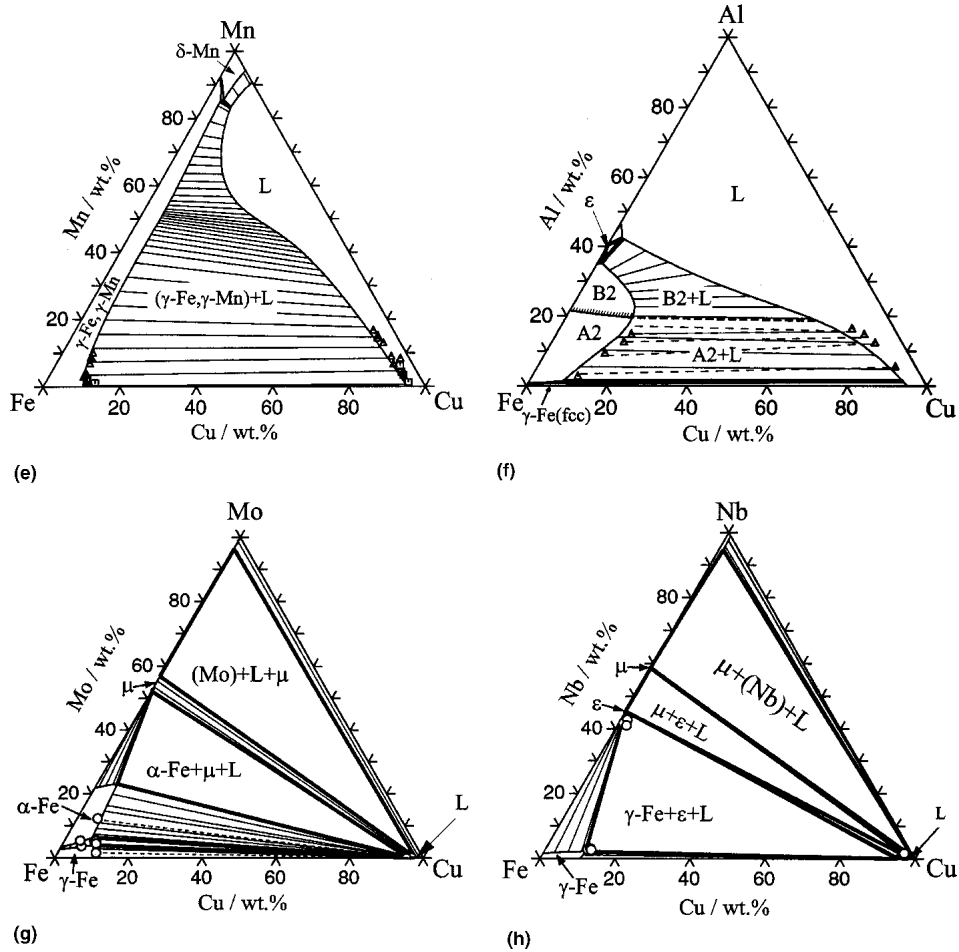


Fig. 2 cont. Calculated isothermal section diagrams at 1473 K in the (e) Fe-Cu-Mn, (f) Fe-Cu-Al, (g) Fe-Cu-Mo, and (h) Fe-Cu-Nb systems

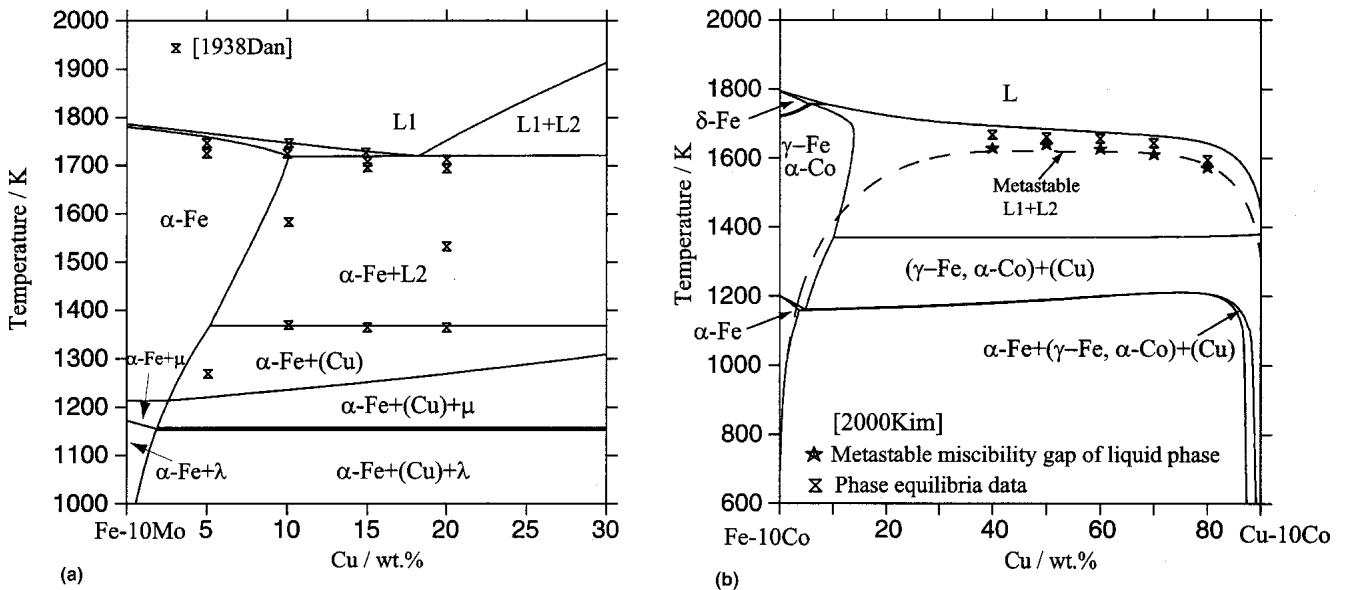


Fig. 3 Calculated (a) 10 wt.% Mo and (b) 10 wt.% Co vertical section diagrams including the stable and metastable miscibility gaps of the liquid phase in the Fe-Cu-Mo and Fe-Cu-Co systems

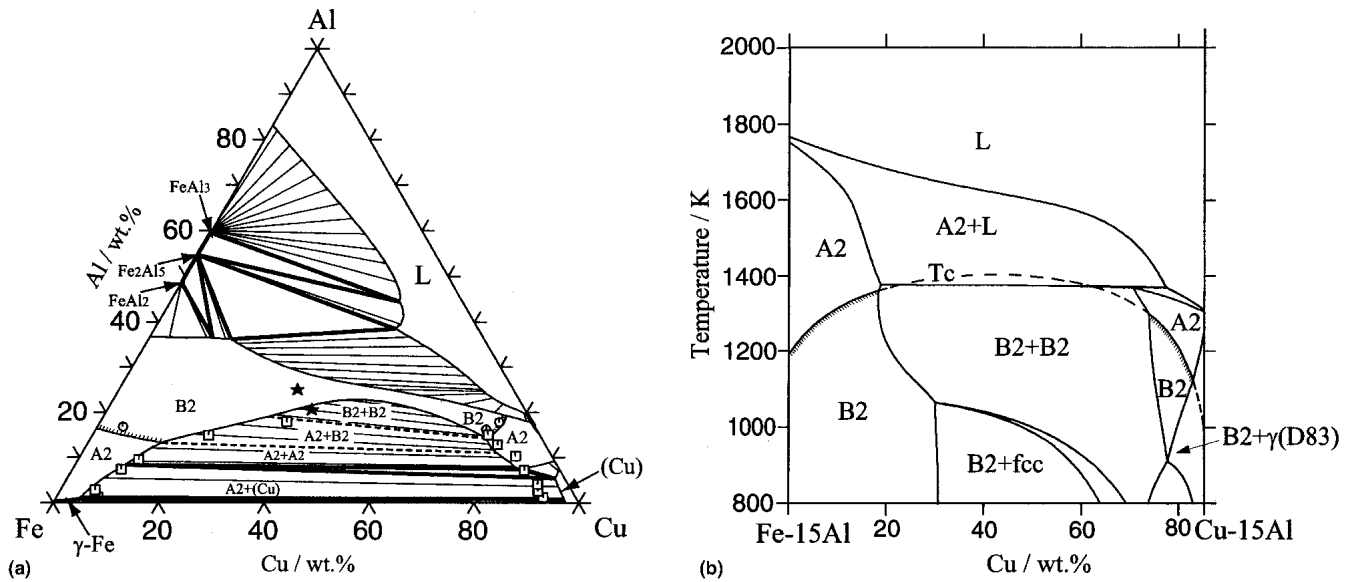


Fig. 4 Calculated isothermal and vertical section diagram at (a) 1273 K and (b) 15 wt.% Al with the experimental data [1998Wan]

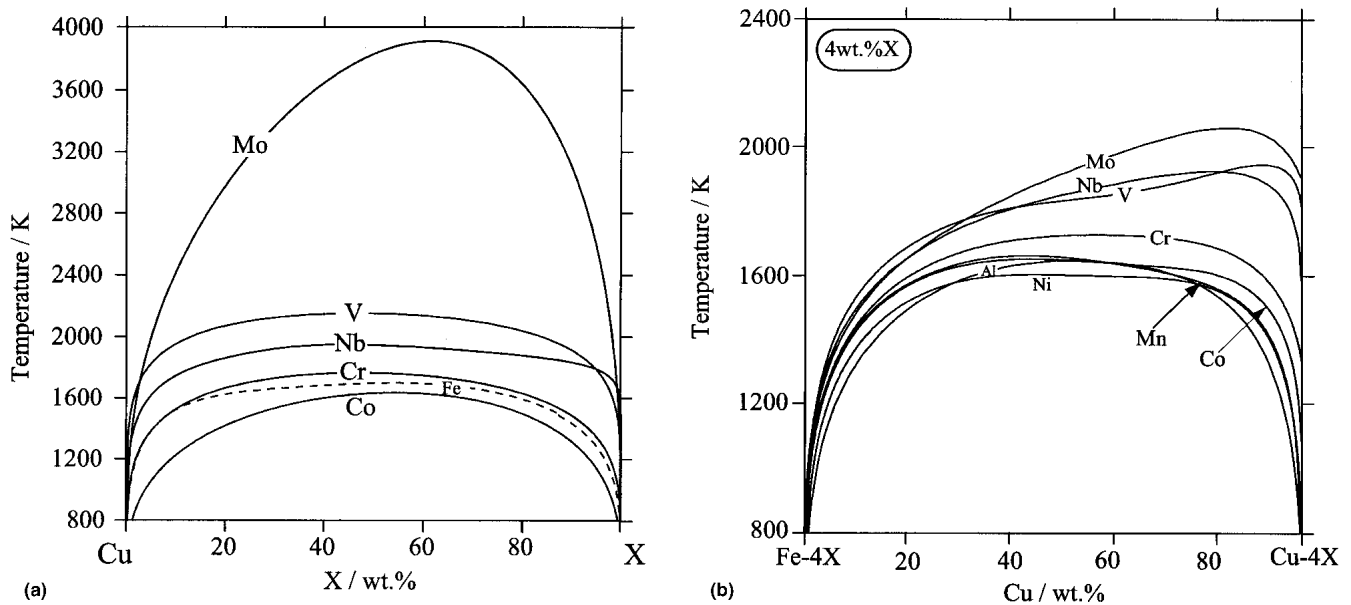


Fig. 5 Calculated metastable and stable miscibility gaps of the liquid phase in the (a) Cu-X binary system, and (b) (Fe-4X)-(Cu-4X) quasi-binary system

4.3 Miscibility Gap in the Liquid Phase

In the Cu-X (X: Co, Cr, Fe, Mo, Nb, V) binary system, stable and metastable miscibility gaps in the liquid phase exist; however, no liquid phase separation occurs in the Cu-Al, Cu-Mn, and Cu-Ni systems. Figure 5(a) shows the calculated results for the miscibility gap in the liquid phase of the Cu-X systems, where there are stable miscibility gaps in the Cu-V and Cu-Mo systems and metastable ones in other alloy systems; the highest critical temperature of the metastable miscibility gap in the Cu-Fe system is about 1694 K at 53.8 wt.% Fe.

The effect of the different alloying elements on the miscibility gaps of the liquid phase in the (Fe-4X)-(Cu-4X) (X: Al, Co, Cr, Mn, Mo, Nb, Ni, V) quasi-binary system are shown in Fig. 5(b). It can be seen that the addition of Cr, Mo, Nb, or V increases the critical temperatures of the miscibility gap of the Fe-Cu binary system, while the addition of Al, Co, Mn, or Ni decreases it.

Figure 6 shows the miscibility gaps of the liquid phase in the Cu-Fe-X ternary system. On the basis of Fig. 6 and the calculations in other systems [1998Wan, 2000Wan, 2001Wan, 2002Wan], it is clear that the miscibility gaps of

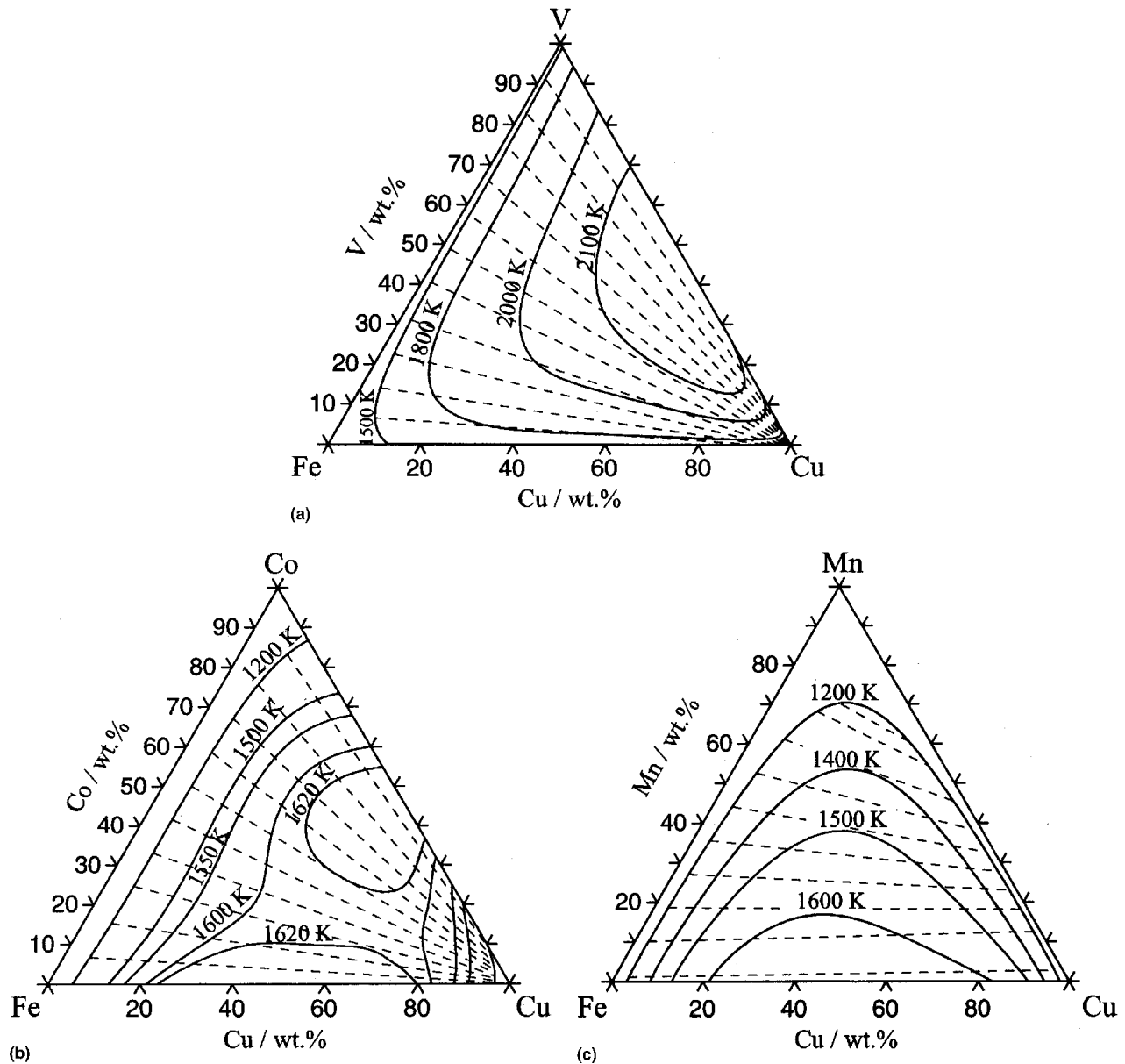


Fig. 6 Calculated miscibility gap of the liquid phase in the (a) Fe-Cu-V, (b) Fe-Cu-Co, and (c) Fe-Cu-Mn systems

the liquid phase in the Cu-Fe-X ternary system can be classified into three types: (1) The tie lines run in the direction from the Fe-X side to the Cu corner in the Cu-Fe-X (X: Cr, Mo, Nb, V) system. A typical example of this miscibility gap is given in Fig. 6(a). (2) The miscibility gap of the liquid phase continually exists from the Cu-Fe side to the Cu-Co side as shown in Fig. 6(b), where the tie lines run in the direction from the Fe-Co side to the Cu corner. (3) The miscibility gap of the liquid phase exists only on the Cu-Fe side. This type of miscibility gap appears in the Cu-Fe-X (X: Al, Mn, Ni) systems as shown in Fig. 6(c). The experimental and calculated information on the miscibility gap of the liquid phases in Cu-Fe-X system is summarized in Table 2.

4.4 Miscibility Gap in the fcc and bcc Phases

The miscibility gaps of the fcc and bcc phases show tendencies similar to those of the liquid phases. The stable miscibility gap of the fcc phase exists in the Cu-Fe-X system because the miscibility gap between γ -Fe (fcc) and Cu (fcc) appears over a wide temperature range. Figure 7(a) shows the calculated miscibility gap of the fcc phase in the Cu-Fe-V system. It can be seen from Fig. 7(a) that the fcc phase separation extends from the Cu-X side to the Cu-Fe side with the temperature decreasing. This type of miscibility gap is formed in the Cu-Fe-X (X: Co, Cr, Mo, Nb) system. The miscibility gap of the fcc phase in the Cu-Fe-Al system only exists on the Cu-Fe side as shown in Fig. 7(b).

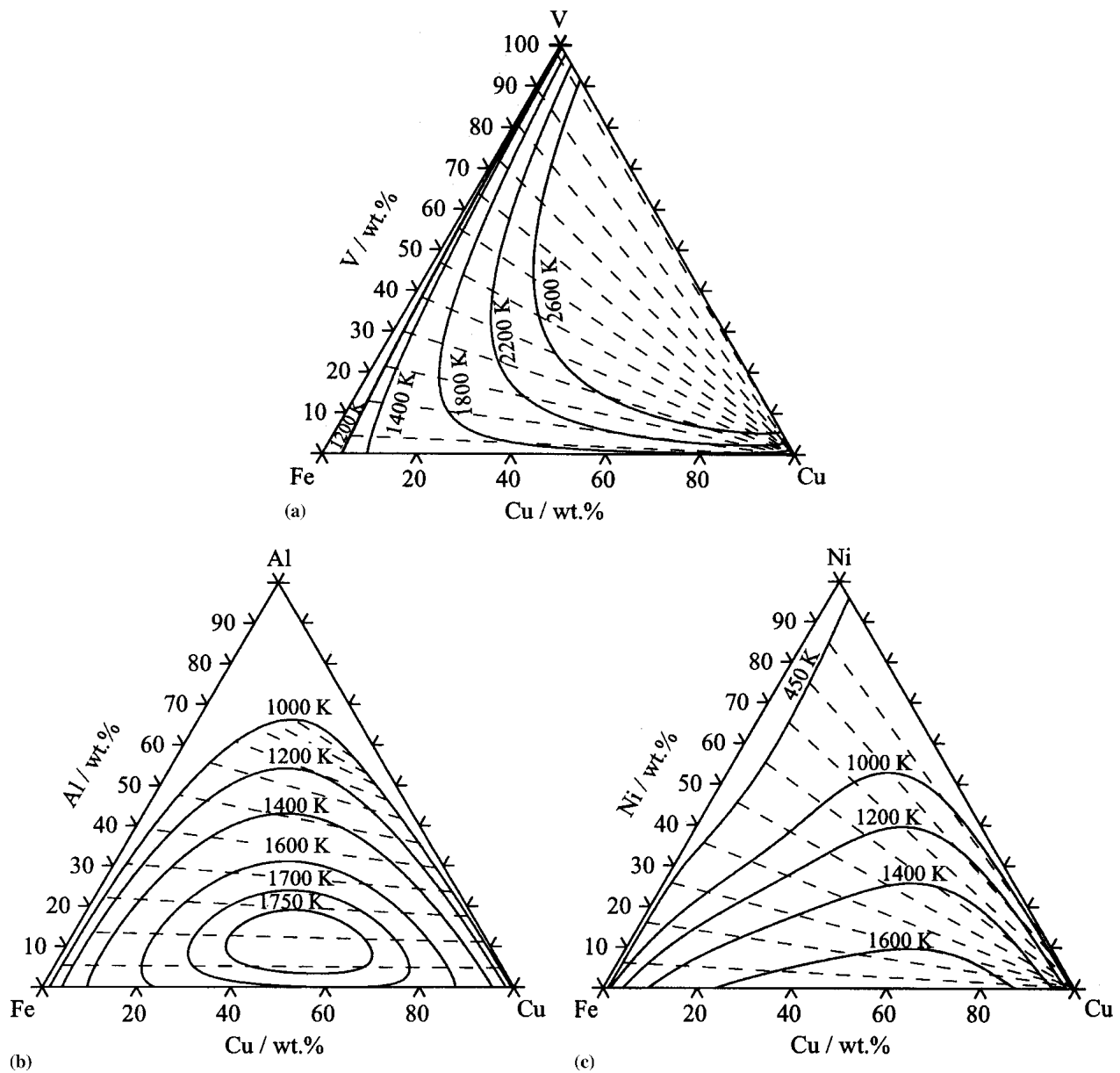


Fig. 7 Calculated miscibility gap of the fcc phase in the (a) Fe-Cu-V, (b) Fe-Cu-Al, and (c) Fe-Cu-Ni systems

Table 2 Miscibility Gap of Liquid Phase in Cu-Fe-X Ternary System

System	Miscibility Gap of Liquid Phase in Cu-Fe-X Ternary System				Difference in Critical Temperature of Miscibility Gap Between 50Cu-50Fe Binary and 50Cu-46Fe-4X Ternary Alloys ($T_{CuFeX}^c - T_{CuFe}^c$)
	Experiment		Calculation		
	Stable	Metastable	Stable	Metastable	
Cu-Fe-Al	No [2003Wan]	No report	...	Yes [1998Wan]	-
Cu-Fe-Co	No [2003Wan]	Yes [2000Kim]	...	Yes [2002Wan]	-
Cu-Fe-Cr	Yes [2003Wan]	...	Yes [2002Wan]	...	+
Cu-Fe-Mn	No [2003Wan]	No report	...	Yes [2001Wan]	-
Cu-Fe-Mo	Yes [1938Dan], Yes [2003Wan]	...	Yes [2000Wan]	...	+
Cu-Fe-Nb	Yes [2003Wan]	...	Yes [2000Wan]	...	+
Cu-Fe-Ni	No [2003Wan]	No report	...	Yes [2001Wan]	-
Cu-Fe-V	Yes [2003Wan]	...	Yes [2002Wan]	...	+

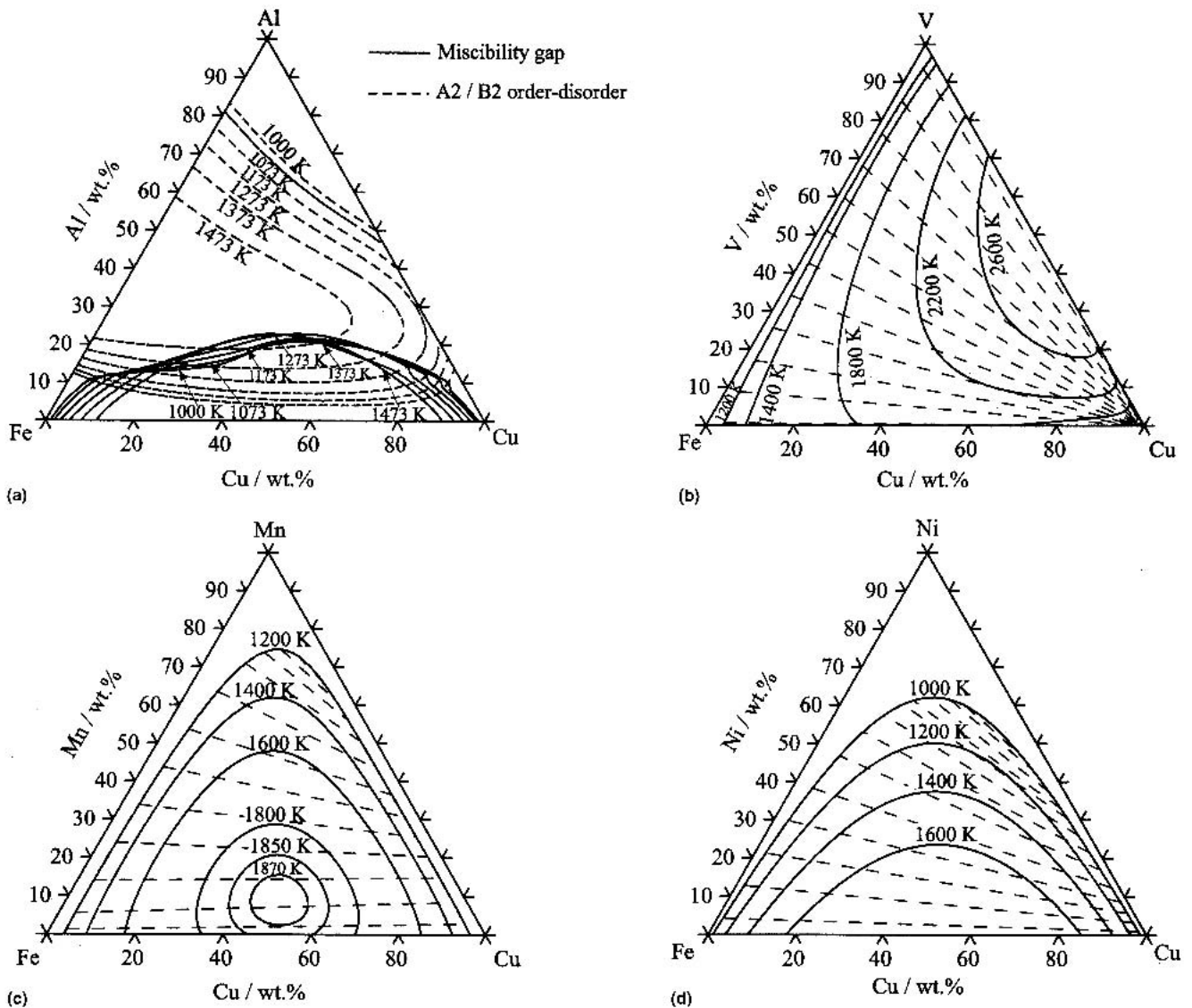


Fig. 8 Calculated miscibility gap of the bcc phase in the (a) Fe-Cu-Al system with the calculated A2/B2 ordering temperature, (b) Fe-Cu-V system, (c) Fe-Cu-Mn system, and (d) Fe-Cu-Ni system

It may also be noted that a miscibility gap island is formed at a higher temperature in the Cu-Fe-Al system. The miscibility gap of the fcc phase in the Cu-Fe-Ni system, which is important for a spinodal type of magnet, is given in Fig. 7(c). A phase separation similar to that in Fig. 7(c) is also found in the Cu-Fe-Mn system, where the miscibility gap extends from the Cu-Fe side to the Cu-X (X: Mn, Ni) side with decreasing temperature.

On the other hand, a stable miscibility gap of the bcc phase only exists in the Cu-Fe-Al system as shown in Fig. 8(a), where the A2/B2 ordering temperature is also plotted. It is shown that the miscibility gap of the bcc phase shrinks due to the formation of the B2 phase. Typical examples of the metastable miscibility gaps of the bcc phases in the other systems are shown in Fig. 8(b)-(d). The calculated results for the Cu-Fe-X (X: Co, Cr, Mo, Nb) systems are similar to those for the Cu-Fe-V system as

shown in Fig. 8(b). A metastable miscibility gap island of the bcc phase is also found in the Cu-Fe-Mn system, as shown in Fig. 8(c).

5. Summary

A thermodynamic database of Cu-Fe-X system was developed on the basis of the CALPHAD method. Examples are given to demonstrate the great utility of this database in the design of Cu-Fe base alloy.

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Section I: Basic and Applied Research

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