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Miscibility gap of B2 phase in NiAl to Cu₃Al section of the Cu–Al–Ni system

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Abstract

The phase separation in the bcc phase of the Cu–Al–Ni system at 600–700 °C was investigated mainly by energy dispersion X-ray spectrometry (EDS) and differential scanning calorimetry (DSC). The compositions of the β_1 (A2 or B2: Cu-rich), β_2 (B2: NiAl-rich) and γ (γ -brass type) phases in equilibrium were determined. It was found that there is a $\beta_1 + \beta_2$ miscibility gap in the β phase region as previously reported by Alexander. It was confirmed by means of high temperature in situ TEM observation that this miscibility gap consists of the B2+B2 phases but not the A2+B2 phases which is sometimes observed in many other Ni–Al and Co–Al base ternary bcc alloys. Thermodynamic calculation was performed which indicates that this characteristic feature suggests that the β_1 (B2) + β_2 (B2) miscibility gap is a part of a Cu-rich B2+NiAl-rich B2 miscibility gap island formed around the center of the composition triangle of the isothermal section. The phase separation in the β phase region and the stability of the ordered bcc aluminide are presented and discussed. © 2004 Elsevier Ltd. All rights reserved.

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1. Introduction

The Cu–Al system, one of the basic systems of Albronze and Cu-base shape memory alloys, has acquired importance not only practically but also basically as a system with two-stage order–disorder transitions, A2 (disordered bcc Cu)–B2 (CuAl)–D0₃ (Cu₃Al) and martensitic transformation in the bcc β phase [1,2]. While the Cu–Al binary alloy shows the shape memory (SM) effect, immediate precipitation of the γ (γ -brass type) phase results in a decrease of the SM property and restricts the practical application as a SM material. Cu–Al base alloys containing several at% Ni are well-known as one type of Cu-base SM alloys, where the addition of Ni is effective to suppress the precipitation of the γ phase [3].

Recently, the present authors have determined the ternary phase equilibria in Ni–Al and Co–Al rich portions of the Ni–Al–Fe, Ni–Al–Mn, Co–Al–Cr, Co–Al–Fe and

Co–Al–Mn systems and found that all of these systems have an A2 (Fe-, Mn- or Cr-rich bcc) + B2 (NiAl- or CoAl-rich bcc) closed miscibility gap due to ordering in the bcc phase region [4–7]. In the Cu–Al–Ni system, Alexander reported in 1938 that a miscibility gap in the bcc phase appears along the Cu₃Al–NiAl pseudo-binary section at temperatures below 740 °C, as shown in Figs. 1 and 2 [8]. However, information on the phase equilibria of the Cu–Al–Ni system is limited [9,10].

The purpose of this paper is to report the results on phase stability of the β and γ phases in the Cu–Al rich portion of the Cu–Al–Ni system and to clarify the characteristic features of the miscibility gap appearing in the bcc phase by thermodynamic analysis with the Bragg–Williams–Gorsky approximation (BWG) [11–13].

2. Experimental procedures

Alloys were prepared by induction melting of Ni (99.9%), Cu (99.9%) and Al (99.7%) in an Al_2O_3 crucible

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Fig. 1. Cu-Al-Ni phase diagram of 700 °C reported by Alexander [8].

in an Ar gas. The specimens cut from the ingot were sealed in a quartz capsule and equilibrated at 600–750 °C for 336–720 h. Microstructural examination of the specimens after heat-treatment was carried out by optical microscopy. The equilibrium compositions were determined by analyzing the two-phase specimens by energy dispersion X-ray spectrometry (EDS) using a standard calibration method.

Transmission electron microscopy (TEM) examinations both at room temperature and at about 700 °C were carried out with a JEM 2000EX electron microscope. Thin foil specimens were prepared by electro-polishing in a solution consisting of 30 ml of HNO₃, 10 ml of HCl, 10 ml of H₂PO₄



Fig. 2. Vertical diagram along the Cu₃Al–NiAl pseudobinary section reported by Alexander [8].

and 10 ml of CH₃COOH. Critical temperatures corresponding to the decomposition reactions were determined by differential scanning calorimetry (DSC) at heating and cooling rates of 5 °C/min.

3. Results and discussion

3.1. Determination of phase equilibria

Fig. 3(a)–(c) show typical microstructures of the β_1 (Cu₃Al-rich bcc)+ β_2 (NiAl-rich bcc), $\beta_2 + \gamma$ and $\beta_2 + \alpha$ (fcc) two-phase alloys, respectively. These two-phase alloys show different types of morphology. The microstructure of the $\beta_1 + \beta_2$ two-phase alloy has a massive shape with habit planes, that of the $\beta_2 + \gamma$ alloy is also massive but without habit planes and that of the $\beta_2 + \alpha$ alloy exhibits a typical Widmanstaetten structure. The phase constitution of the two-phase alloys can be easily distinguished by such characteristic morphologic features. The equilibrium composition of each phase was quantitatively determined by the EDS analysis. All the experimentally determined results on



Fig. 3. Optical micrographs of various two-phase structures taken from (a) the Cu-37.5 at%Al-25 at%Ni, (b) Cu-40 at%Al-20 at%Ni and (c) Cu-35 at%Al-25 at%Ni alloys annealed at 700 °C after quenching from 900 °C.

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