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### Determination of thermodynamic properties of aluminum based binary and ternary alloys



ALLOYS AND COMPOUNDS

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#### 1. Introduction

ABSTRACT

In the present work, the Gibbs-Thomson coefficient, solid-liquid and solid-solid interfacial energies and grain boundary energy of a solid Al solution in the Al-Cu-Si eutectic system were determined from the observed grain boundary groove shapes by measuring the thermal conductivity of the solid and liquid phases and temperature gradient. Some thermodynamic properties such as the enthalpy of fusion, entropy of fusion, the change of specific heat from liquid to solid and the electrical conductivity of solid phases at their melting temperature were also evaluated by using the measured values of relevant data for Al-Cu, Al-Si, Al-Mg, Al-Ni, Al-Ti, Al-Cu-Ag, Al-Cu-Si binary and ternary alloys.

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Aluminum alloys have many advantages that include high thermal and electrical conductivities, low density, high specific strength, ease of casting, high strength to weight ratio and reasonable corrosion resistance. By using appropriate heat treatment techniques [1–6] we can achieve better features. Aluminum alloys have a wide range of applications in the aerospace and automotive industries. In particular, the automotive industry uses aluminum alloys for engine blocks and cylinder heads [6,7].

One of the most important aluminum based alloys is Al-Cu-Si. Because of its low melting temperature and good fluidity, the Al-Cu-Si eutectic alloy has been used in the application of braze welding. Although a lot of research has been carried out on the Al–Cu–Si non-eutectic allov in the literature [8–10], only a few studies [11] have been done on ternary eutectic Al-Cu-Si alloy systems. From the Al–Cu–Si phase diagram [12], the eutectic composition of Al-Cu-Si alloy is Al-26.82 wt.% Cu-5.27 wt.% Si and the eutectic temperature is 797 K. The eutectic phases of Al-26.82

Corresponding author. E-mail address: kesli@erciyes.edu.tr (K. Keşlioğlu). wt.% Cu-5.27 wt.% Si eutectic alloy are Al solution, Si and  $\theta$ (CuAl<sub>2</sub>). The aim of this study is to evaluate some of the thermophysical properties of Al-26.82 wt.% Cu-5.27 wt.% Si ternary eutectic alloy such as the Gibbs-Thomson coefficient, solid-liquid interfacial energy, solid-solid interfacial energy, enthalpy of fusion, entropy of fusion, change of specific heats from liquid to solid and the electrical conductivity of solid phases at their melting temperatures.

#### 2. Experimental procedure

#### 2.1. Sample preparation

The composition of the alloy was chosen as Al-26.82 wt.% Cu-5.27 wt.% Si [12] to grow a single solid Al solution phase on the eutectic casting phase. The Al-26.82 wt.% Cu-5.27 wt.% Si alloy was prepared in a vacuum furnace by using Al, Cu and Si metals with 99.99, 99.9 and 99.999% purities, respectively. The molten alloy was poured into a graphite crucible at the casting furnace and then directionally solidified. The sample was then ready to place in the radial heat flow apparatus.

In order to obtain the grain boundary groove shapes (GBGS) in metallic alloy systems, a radial heat flow apparatus was firstly



designed by Gündüz and Hunt [13]. The block diagram of the radial heat flow apparatus is shown in Fig. 1. The details of the experimental technique are given in Refs. [13–19]. Similar instruments were used in this study to observe the GBGSs in the Al–Cu–Si.

### 2.2. Microstructure of Al-Cu-Si eutectic system

The microstructure of the alloy was observed through optical microscope and SEM (Scanning Electron Microscope) in different areas of the samples. The specimen was prepared for microscopic observation using standard metallographic techniques. In order to obtain a good image an etchant (2.5 ml nitric acid, 1.5 ml hydrochloric acid, 1 ml hydrofluoric acid in 95 ml water) was used for 15 s. The optical microscope and SEM photographs of the microstructure of the Al-Cu-Si allov are shown in Fig. 2. The solid phases existed in the Al-Cu-Si systems were clearly shown in Fig. 2c. The photograph of Fig. 2c was taken from cross-sectional area of cvlindrical sample which was melted around the central heating element and then annealed a sufficient period in a constant temperature gradient to obtain the GBGS. Fig. 2c has shown the microstructures of solid phases with casting phase away from the solid–liquid interface. According to Fig. 2c the solid  $\alpha$ -Al phase grown on the eutectic structure during the annealing period.

EDX composition analysis was used to determine the three phases of the Al–Cu–Si alloy. The EDX result is illustrated in Fig. 3. As shown in Figs. 2 and 3, the black phase is the Al solid solution, the gray phase is the Si solid solution and the white phase is  $\theta$  (CuAl<sub>2</sub>). While the  $\alpha$ -Al and  $\theta$ -CuAl<sub>2</sub> phases grew alongside in accordance with each other, the Si phase disrupted this harmony with its needle like appearance in Fig. 2a, and with its blade like appearance in Fig. 2c. Because the unstable Si phase has a faceted structure, the  $\alpha$ -Al and  $\theta$ -CuAl<sub>2</sub> phases have unfaceted structures.

### 2.3. Determination of Gibbs–Thomson coefficients for solid Al solutions in the Al–Cu–Si eutectic system

In order to determine the Gibbs–Thomson coefficient ( $\Gamma$ ) from

the numerical method firstly constructed by Gündüz and Hunt [13], the groove coordinates of the GBGSs, the thermal conductivity ratio, and the temperature gradient of the solid phase must be known. In the measurement of temperature gradient and groove coordinates the total experimental errors were determined as 6.5% and 0.1%, respectively. Thus the total experimental error in the determination of the Gibbs—Thomson coefficient is about 7%.

### 2.3.1. Determination of Gibbs–Thomson coefficient for solid Al solution in equilibrium with Al–Cu–Si liquid

The GBGS for the Al solid solution in equilibrium with the Al–Cu–Si liquid (Al-26.82 wt.% Cu-5.27 wt.% Si) were observed and a typical GBGS was shown in Fig. 4. To determine the Gibbs–Thomson coefficients ( $\Gamma$ ) for the Al solid solution with the numerical model we used ten equilibrated GBGSs. We determined the Gibbs–Thomson coefficients for both sides of these ten groove shapes. The determined values of  $\Gamma$  for the Al solid solution are given in Table 1. The average value of  $\Gamma$  from Table 1 is found to be  $(2.11 \pm 0.15) \times 10^{-7}$  Km.

### 2.3.2. Determination of Gibbs–Thomson coefficient for solid Al solution in equilibrium with the solid CuAl<sub>2</sub>

For the first time, the GBGSs for a solid Al solution in equilibrium with solid CuAl<sub>2</sub> were also observed in the present study and a typical GBGS is shown in Fig. 5. As can be seen from Fig. 5, the solid CuAl<sub>2</sub> phase has grown in front of the solid Al phase. To determine the Gibbs—Thomson coefficients for the solid Al solution in equilibrium with the solid CuAl<sub>2</sub>, we used four GBGSs. The values of  $\Gamma$  for the solid Al solution in equilibrium with the solid CuAl<sub>2</sub> are given in Table 2. The average value of  $\Gamma$  from Table 2 is found to be  $(2.23 \pm 0.16) \times 10^{-7}$  Km.

## 2.4. Determinations of entropy of fusion per unit volume for solid Al solutions

The entropy change per unit volume for an alloy is given by Refs. [13],



Fig. 1. Block diagram of radial heat flow apparatus.

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