

Cluster structure in Al-Si eutectic melt with solid Ni particles

S. MUDRY¹, I. SHTABLAVYI^{1*}

¹ Ivan Franko National University of Lviv, Physics of Metals Department, Kyryla and Mephodiya St. 8, 79005 Lviv, Ukraine

* Corresponding author. Tel.: +380-322-964594; e-mail: sihor@ukr.net

Received October 17, 2007; accepted June 18, 2008; available on-line September 10, 2008

The structure of liquid Al_{0.88}Si_{0.12} eutectic alloy upon addition of Ni powder was studied by means of X-ray diffraction. The main structure parameters as a function of the Ni-content were analyzed. It is shown that the atomic distribution in these alloys is inhomogeneous, revealing the existence of chemically ordered structural units.

Liquid eutectic alloy / Metallic powder / Short range order / Cluster

Introduction

Metal matrix composites (MMC) reinforced with metallic, intermetallic or ceramic particles are of great importance nowadays. Many reviews are available on processing, properties and interfacial phenomena in MMC (see [1-4]). Although there are several techniques for processing composite materials, on a large scale these are produced by liquid metallurgy (LM) or powder metallurgy (PM) routes. In the former, solid particles or powders are mechanically dispersed in the liquid before solidification of the melt. The composites obtained this way can be used in different areas of application: liquid magnetic systems, solders, new porous alloys. In this work results on structure changes induced by addition of Ni-powder to Al-Si eutectic melts are presented. The aim of the investigation was to study the structure, which are commonly formed via diffusive way when metallic powder is mixed with metallic alloy having a low melting point. Nickel is commonly used to reinforce Al-based alloys [5].

The Al-Si alloy system, which is characterized by high specific strength, excellent corrosion resistance, as well as good thermal and electrical conductivities, is widely used to supplant other alloys in the areas of transportation, packaging, construction, and machinery to achieve great weight reduction [6]. The properties of the alloys depend on processing variables that are interrelated, such as the rate of solidification, dissolved gases, level of porosity, alloying elements, etc. [7].

Experimental

The studies were carried out by means of X-ray diffraction and scanning electron microscopy. The samples were prepared in an arc-furnace filled with pure argon. The purity of the initial metals was 99.999%. The diffraction studies were carried out using a high-temperature diffractometer with a special attachment that allows to investigate solid and liquid samples at different temperatures up to 1800 K. Cu-K_α radiation, monochromatized by means of a LiF single crystal, and Bragg-Brentano focusing geometry were used. The scattered intensities as a function of the scattering angle were recorded within the range $1\text{\AA}^{-1} < k < 7\text{\AA}^{-1}$, with the angular step equal to 0.05° and 0.5° for the region of the principal peak and for the remaining values of the wave vectors, respectively. The accuracy of the measured scattered intensities was estimated to be better than 2%. In order to obtain more accurate scattered intensities, the scan time was equal to 100 s. The diffracted intensity was recorded using a NaI(Tl) scintillator detector in conjunction with an amplification system. The sample was placed in a rounded cup of 20 mm diameter. The intensity curves were corrected for polarization, absorption and incoherent scattering [8]. After this procedure they were normalized to electron units by the Krogh-Moe method [9]. The obtained intensity curves were used to calculate the structure factors (SF) and then the pair correlation function (PCF). The main structure parameters, obtained from SF and PCF, were analyzed.

Computer simulation methods (Reverse Monte Carlo) were also used to analyze the X-ray diffraction data.

Results and discussion

Solid $\text{Al}_{0.88}\text{Si}_{0.12}$ eutectics show a discontinuous microstructure in which the Si-phase is dispersed in the Al-matrix as discrete particles (needle-like morphology). Chill casting of eutectic Al-Si alloys generally produces a refinement of the eutectic. The shape of the Si particles is the same as in slowly cooled alloys, but they are scaled down to considerably smaller dimensions [10]. There exist many works devoted to the investigation of the structure of liquid Al-Si alloys [11-15]. On the basis of these works one can draw conclusions on the existence of preferred interaction between unlike atoms at low Si content. At higher Si content microgroups with the structure of pure Si appear.

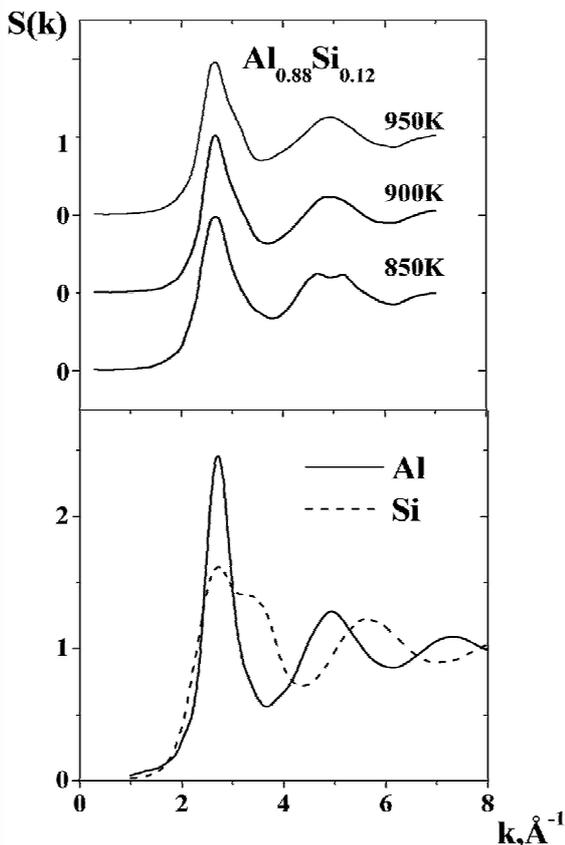


Fig. 1 The structure factors for $\text{Al}_{0.88}\text{Si}_{0.12}$ eutectic melts in comparison with SF for liquid Al and Si.

The phase equilibria in the Ni–Al–Si ternary system have been studied using diffusion couple experiments with pure Ni and an Al–Si eutectic alloy [16]. All the binary Ni–Al intermetallics, NiAl_3 , Ni_2Al_3 , NiAl , Ni_5Al_3 and Ni_3Al were observed in the

temperature interval 830–1170 K. The ternary intermetallics were, however, observed in specimens annealed at lower temperatures.

The structure factor for the $\text{Al}_{0.88}\text{Si}_{0.12}$ eutectic melt near the eutectic temperature ($T=850\text{K}$) is shown in Fig. 1 in comparison with SF for liquid Al and Si.

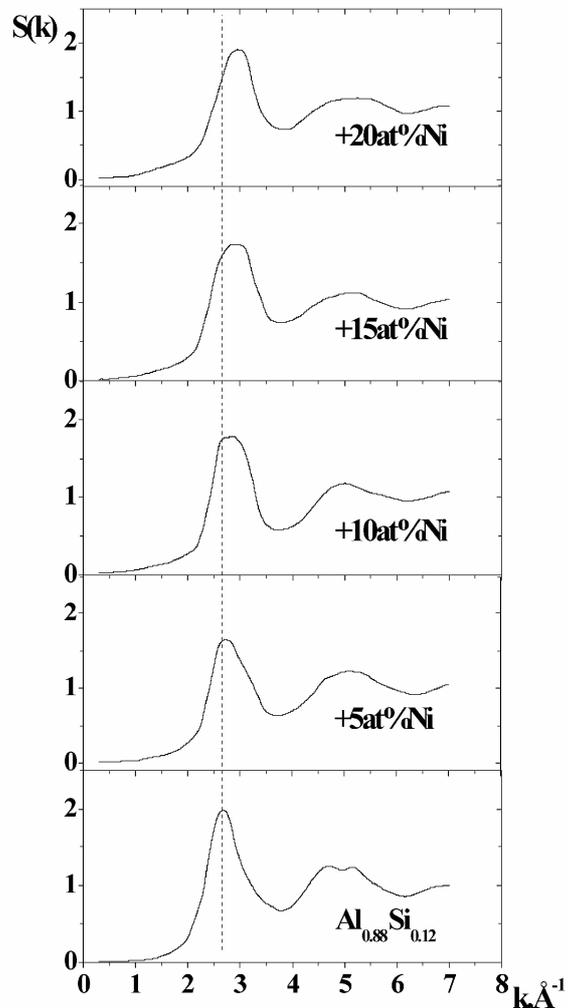


Fig. 2 The structure factors for $\text{Al}_{0.88}\text{Si}_{0.12}$ melts with 5; 10; 15 and 20 at.% of Ni ($T= 850, 860, 910$ and 950K).

The main feature of this SF is the splitting of the second peak at 850 K into two subpeaks. The first of these subpeaks corresponds to the second peak of liquid Al, while the second one appears at the same position as for liquid Si. This allowed us to conclude that the short range order structure in the eutectic melt cannot be described by a random atomic distribution, nor by the self association model. One can suppose that part of the Si-atoms are stronger bonded to Al-atoms, forming chemically ordered structural units (clusters). Other Si-atoms are bonded via covalent bonding and form self-associated structural units. On heating the splitting disappears, and the peak shape becomes similar to SF for simple liquids, confirming

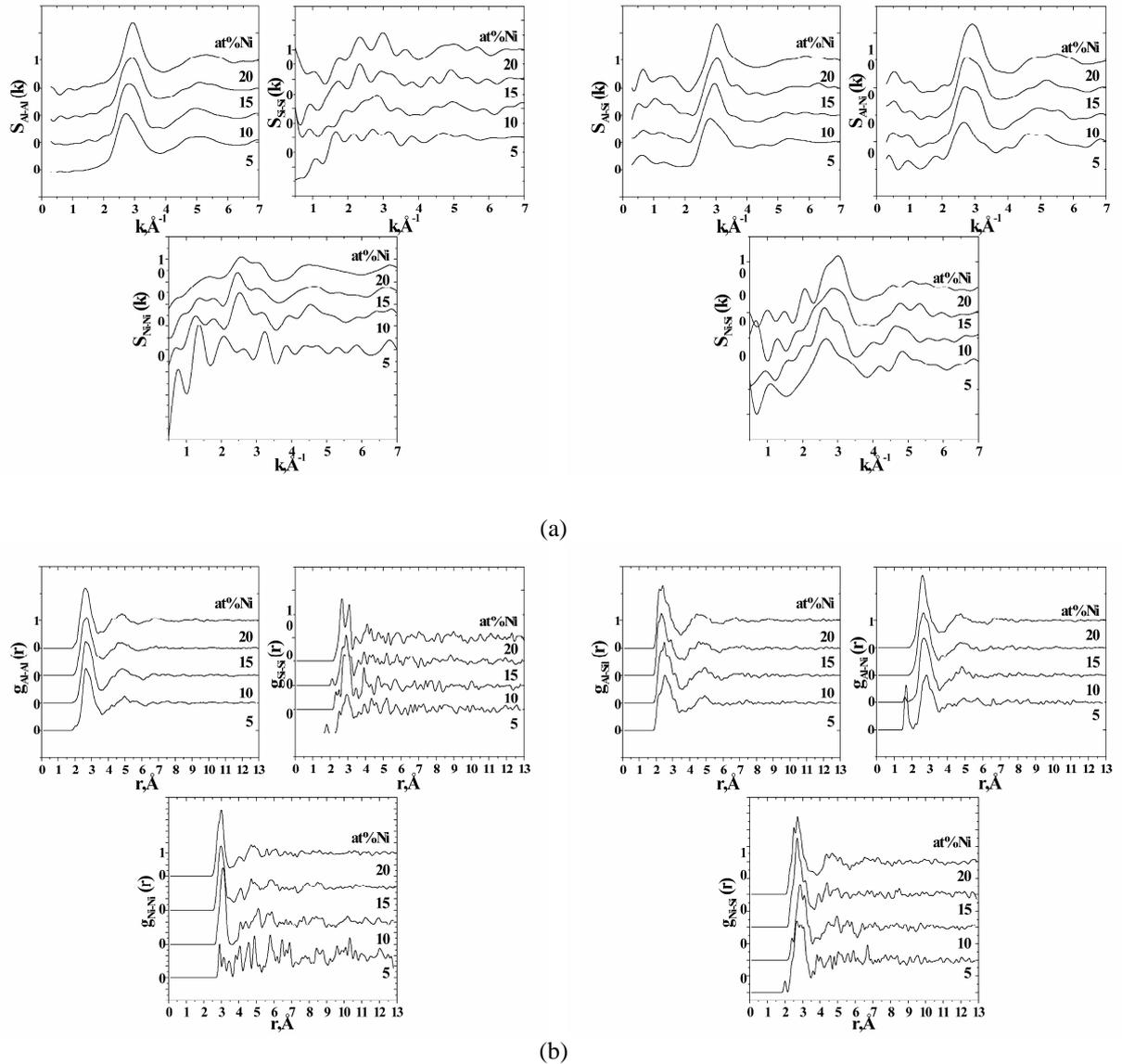


Fig. 3 Partial structure factors (a) and partial pair correlation functions (b) for $\text{Al}_{0.88}\text{Si}_{0.12}$ melts with 5, 10, 15 and 20 at.% Ni ($T = 850, 860, 910$ and 950 K).

the dominant role of thermal disorder in the atomic rearrangement at higher temperatures. The results obtained on the structure of $\text{Al}_{0.8}\text{Si}_{0.12}$ eutectic melts are in accordance with other diffraction studies and physical property measurements [11-15].

Starting from such a structure, it seemed of interest to understand how admixtures will interact with the structural units. In case of Ni, taking into account that the binary Ni-Si and Al-Ni systems contain chemical compounds of higher thermal stability [17], one can expect an increase of the tendency to interaction of unlike atoms. This assumption is based on thermodynamic properties of binary and ternary Al-Si-Ni alloys [18-20]. For instance, the enthalpy of mixing shows negative values over the entire concentration range in the Al-Ni, Al-Si and Ni-Si binary systems, indicating preferred interaction

between unlike atoms.

The structure factors for $\text{Al}_{0.88}\text{Si}_{0.12}$ melts with 5, 10, 15 and 20 at.% Ni at the temperatures 850, 860, 910 and 950 K, respectively, are shown in Fig. 2. The main feature of these functions is the shift of the principal peak to larger k -values and its significant increase. In addition, the broad peak at higher k -values becomes more asymmetric. Such changes cannot be explained by a simple dilution of Ni atoms similar to the formation of classical solid solutions.

Experimental SF were used to calculate the partial SF by means of the RMC-method. We computed the $S_{Al-Al}(k)$, $S_{Si-Si}(k)$, $S_{Ni-Ni}(k)$ partial structure factors, which describe like atoms, and $S_{Al-Si}(k)$, $S_{Al-Ni}(k)$, $S_{Ni-Si}(k)$, which give information about chemical ordering (Fig. 3a). The partial pair correlation functions are shown in Fig. 3b. One can see that

$S_{Al-Al}(k)$ is similar to the corresponding SF for pure liquid Al, whereas the partial SF describing the Ni-Ni and Si-Si atomic distribution are significantly different from the *typical* profile of SF. Therefore, an Al-like distribution has higher probability to exist in the structure of molten alloys. We can also note that all kinds of atomic distribution strongly depend on the Ni content. The most significant changes *versus* the Ni content are observed for the Ni-Ni distribution.

The partial SF describing the distribution of unlike atoms shows evident maxima. Their profile is in many aspects very similar to the SF commonly observed for molten intermetallics [21,22]. With increasing Ni content, the principal peaks arise, showing significant correlation in the distribution of unlike atoms. A similar behaviour is observed for the partial pair correlation functions, which describe the atomic distribution in real space.

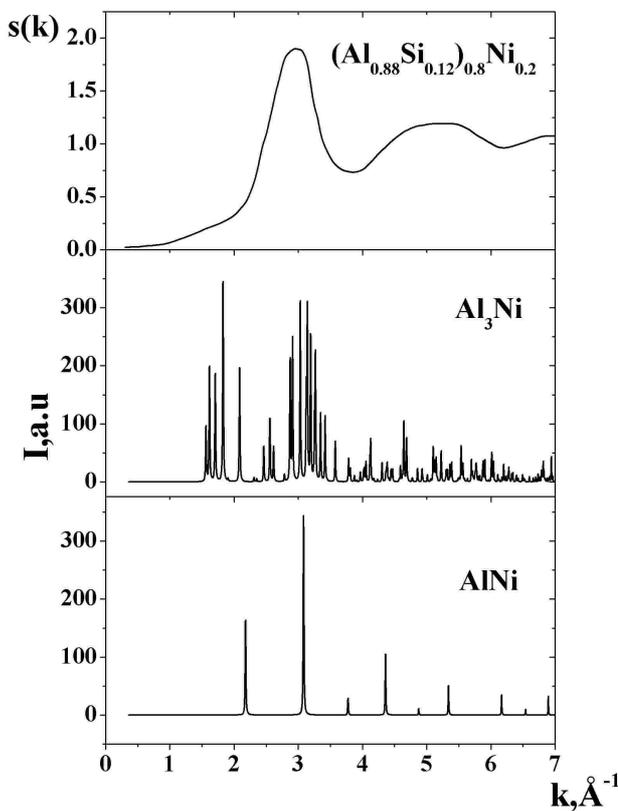


Fig. 4 Structure factor for liquid $(Al_{0.88}Si_{0.12})_{0.8}Ni_{0.2}$ and diffraction patterns for Al_3Ni and $AlNi$ intermetallics.

The comparison of the structure factor profile with the diffraction patterns of the $AlNi$ and Al_3Ni intermetallics (Fig. 4) shows a fairly good agreement. This allowed us to conclude that $AlNi$ and Al_3Ni -like chemical ordering mainly determines the structure features in $(Al-Si)_{1-x}Ni_x$ alloys. Chemically ordered structure groups become the nuclei of intermetallic phases at crystallization (Fig. 5).

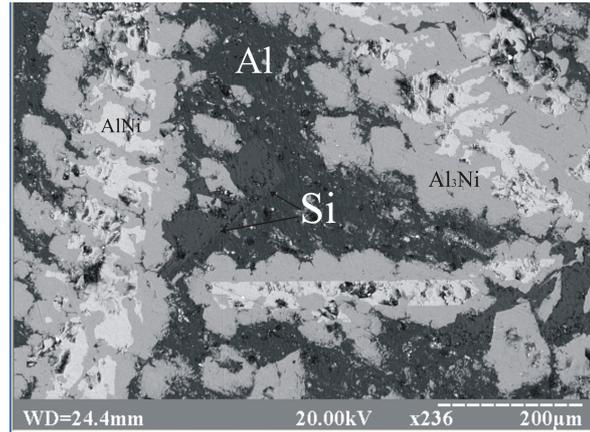


Fig. 5 Microstructure of a solid $(Al_{0.88}Si_{0.12})_{0.8}Ni_{0.2}$ alloy.

Conclusions

The structure of Al-Si eutectic melts with additions of Ni revealed changes corresponding to the formation of chemically ordered microgroups with $AlNi$ and Al_3Ni topology. The fraction of such structural units increases with increasing Ni content.

References

- [1] S.J. Cytron, *J. Mater. Sci. Lett.* 1 (1982) 211–213.
- [2] J.M. Torralba, C.E. da Costa, F. Velasco, *J. Mater. Process. Technol.* 133 (2003) 203–206.
- [3] K. Kristiakova, P. Svec, M. Deanko, *Mater. Sci. Eng. A* 375–377 (2004) 136–149.
- [4] V.C. Srivastava, S.N. Ojha, *Bull. Mater. Sci.* 18 (2005) 125–130.
- [5] J.W. Kaczmar, K. Pietrzak, W. Wlosinski, *J. Mater. Process. Technol.* 106 (2000) 58–67.
- [6] L. Lu, A.K. Dahle, *Mater. Sci. Eng. A* 435–436 (2006) 288–296.
- [7] B. Suarez-Pena, J. Asensio-Lozano, *Mater. Charact.* 57 (2006) 218–226.
- [8] D.T. Cromer, J.T. Waber, *Acta Crystallogr.* 18 (1965) 104–109.
- [9] J. Krogh-Moe, *Acta Crystallogr.* 9 (1956) 951–953.
- [10] B. Chalmers, *Progress in Materials Science*, Vol. 12, Pergamon Press, Oxford, 1965.
- [11] Ya.I. Dutchak, V.S. Frenchko, *Russ. Phys. J.* 18 (1975) 101–102.
- [12] A. Prygunova, E. Rastovskaya, A. Romanova, *Ukr. Fiz. Zh.* 24 (1979) 685–687 (in Russian).
- [13] A. Prygunova, V. Mazur, Ju. Taran, *Metallofizika* 5 (1983) 88–94 (in Russian).
- [14] Bian Xiufang, Wang Weimin, Yuan Shujuan, Qin Jingyu, *Sci. Technol. Adv. Mater.* 2 (2001) 19–23.

- [15] N.M. Keita, S. Steinemann, *J. Phys. C: Solid State Phys.* 11 (1978) 4635-4641.
- [16] M. Jain, S.P. Gupta, *Mater. Charact.* 51 (2003) 243–257.
- [17] T. Massalsky, *Binary Alloy Phase Diagrams*, American Society for Metals, Metals Park, OH, 1998, V.1,2, 2233 p.
- [18] K.V. Grigorovich, A.S. Krylov, *Thermochim. Acta* 314 (1998) 255–263.
- [19] D.S. Kanibolotsky, O.A. Bieloborodova, N.V. Kotova, V.V. Lisnyak, *J. Therm. Anal. Calorim.* 70 (2002) 975–983.
- [20] V.T. Witusiewicz, I. Arpshofen, H.-J. Seifert, F. Sommer, F. Aldinger, *J. Alloys Compd.* 305 (2000) 157–171.
- [21] Li Wang, Yu Qing Wang, Chuanxiao Peng, Yanning Zhang, *Phys. Lett. A* 350 (2006) 405–409.
- [22] J. Brillo, A. Bytchkov, I. Egry, L. Hennem, G. Mathiak, I. Pozdnyakova, D.L. Price, D. Thiaudiere, D. Zanghi, *J. Non-Cryst. Solids* 352 (2006) 4008–4012.

Proceeding of the X International Conference on Crystal Chemistry of Intermetallic Compounds, Lviv, September 17-20, 2007.