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Short and medium-range order in liquid binary Al–Ni and Al–Co alloys

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ABSTRACT

A local short-to-intermediate range structure of liquid Al–Ni and Al–Co alloys was examined by X-ray diffraction technique and the reverse Monte Carlo modeling. The existence of a prepeak in the $S(Q)$ function of the liquid Al–Ni and Al–Co alloys reached with Al caused by specific arrangement of 3*d*-transition metal atoms in dense packed polytetrahedral clusters.

1. Introduction

Modern materials science is directed on preparation and examination of quasicrystalline (QC) metallic alloys, since they possess unique physical, chemical and mechanical properties. This feature of the QC metallic alloys concerns with the essential difference in atomic ordering between QC and crystalline phases of equal composition. As the QC phases can be prepared by rapid quenching from the liquid state, therefore, searching for similarities in atomic ordering in liquid, solid and QC states is the major problem, which has as scientific as well as technological importance. The comparison of local atomic structure of liquid metallic alloys and corresponding QC phases as well as the systematic study on structural prerequisites of quasicrystals formation are the most prominent direction to develop scientific base for preparation of QC metallic alloys.

Starting with the pioneering work of Shechtman [1], the studies on quasicrystals (QCs) have shown that the majority of the QC materials can be obtained from the Al-based binary and ternary liquid alloys like Al–Co(Ni) and Al–Ni(Co)–TM (TM = transition metal) [2–4].

The results of X-ray diffraction studies on local atomic structure in the liquid binary Al–Co and Al–Ni alloys as a function of the alloy composition have been reported by us earlier [5,6]. These studies are supported with modeling in the framework of the reverse Monte Carlo method (RMC). The concentration range of QC phases formation and the concentration range of extreme values of the Voronoi metric and topological parameters, which characterize the local atomic structure in the liquid state, correlate well for these binary alloys.

The liquid Al–Ni and Al–Co alloys are characterized by considerable compression of the molar volumes [7], negative deviations of activity from the Raoult's law [8], and extrema in the isotherms of viscosity [9] and of electrical resistance [10,11] for the equiatomic compositions. A microinhomogeneous structure [7–10] was supposed as the main reasons of phenomena observed for these liquid alloys.

The present paper was devoted to the comparative analysis of local structure in the liquid Al–Ni and Al–Co alloys. For this reason X-ray diffraction studies supported by the RMC simulations were performed on the liquid $Al_{85.7}Co_{14.3}$, $Al_{80}Co_{20}$ and $Al_{75.43}Co_{28.57}$ alloys (all concentrations of the alloys constituents in this paper are given in at.%).

2. Experimental methods

The binary $Al_{100-x}Co_x$ (x = 10, 14.3, 18, 20, 23.5, 26, 28.57 and 80.7) and $Al_{100-x}Ni_x$ (10, 18, 25, 28, 32, 40, 75 and 90) alloys were prepared from high purity Al (A999) and Co (99.8%) or Ni (99.8%) metals by arc-welding in an Ar gas atmosphere. The preparation route was similar to that reported earlier in [5,6]. The binary alloys were placed into the alumina crucibles and transferred in a stainless steel chamber. The chamber inner volume was vacuumized and filled with pure helium gas. The special resistant heater inserted into the chamber was used for progressive heating of the binary alloys up to experimental temperature in helium atmosphere. The X-ray diffraction experiments for the liquid Al–Ni and Al–Co alloys was carried out at temperatures approximately 50 K higher than the liquidus temperature. The liquid binary Al–Ni and Al–Co alloys were studied by means of X-ray diffraction with a θ–θ diffractometer (MoK_α-radiation, λ = 0.71069 Å) [12]. Axis of the goniometer oriented horizontally on the θ–θ diffractometer and the chamber axis were adjusted. The Zr–Y differential filters were used for monochromatization of X-ray radiation, which was reflected from the free surface of the liquid alloy. The magnitude of experimental diffraction vector ($Q = 4\pi\sin\theta/\lambda$, where θ is the half of the scattering angle) was ranged from 0.9 to 12.5 Å^{-1}. Three sets of diffraction data were collected for each of the liquid alloy in order to minimize random errors in the diffraction experiment. The experimental scattering intensity functions were corrected for the polarization and the angular dependence of incoherent scattering in accordance with methodology proposed in

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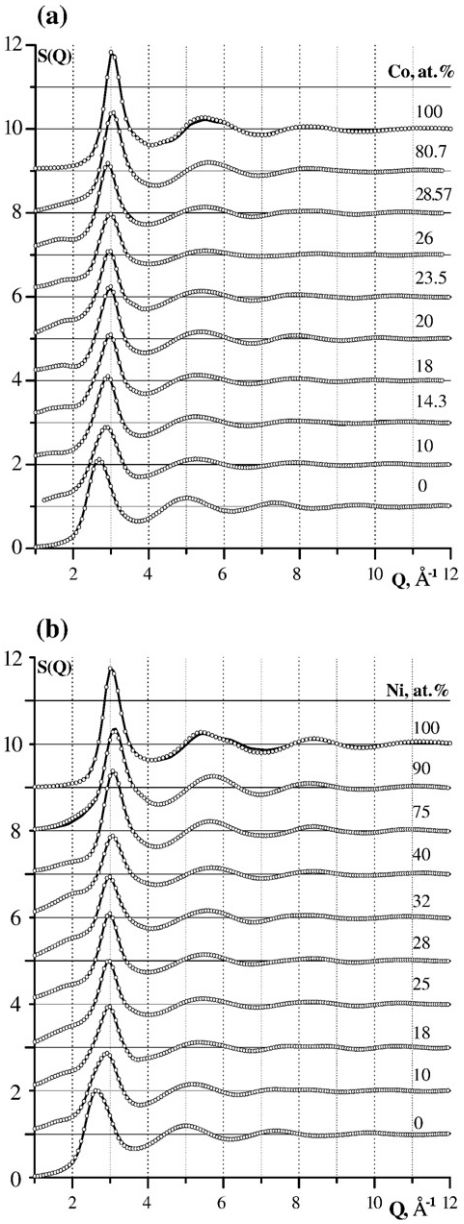

Fig. 1. Structural factor $S(Q)$ of the liquid $Al_{100-x}Co_x$ (a) and $Al_{100-x}Ni_x$ (b) alloys. The open circles are the experimental data, solid lines are the data calculated for the RMC configuration.

[13]. The scattering intensity measured in arbitrary units was converted into coherent scattering intensity per atom in electron units using the Vainshtein equation and atomic scattering factors corrected on anomalous dispersion [14]. The structure factors $S(Q)$ and radial distribution function (RDF) of atoms $g(R)$ were calculated according to procedure reported in [15].

The configuration models of the liquid Al–Ni and Al–Co alloys were reconstructed from the experimental structure factors using the RMC method [16]. A basic cubic cell was arranged with 5000 spherically symmetric particles with the imposed periodical boundary conditions. The experimentally measured density of the liquid alloys [7] at the temperature of X-ray diffraction study defines dimensions of the cubic cell. The cut-off distances of $d_{Al-Al}=2.3$ Å, $d_{Ni-Ni}=2.1$ Å and $d_{Co-Co}=2.1$ Å obtained for one-type atoms from experimental $g(R)$ functions of the pure liquid metals were used as restrains in the RMC simulations. The cut-off distances between different kinds of the metal atoms were calculated as $d_{Al-Me}=(d_{Al-Al}+d_{M-M})/2$, where M is Co or Ni.

According to the RMC algorithm, the iterative process proceeds until the discrepancy χ^2 between experimental $S^{exp}(Q)$ and simulated $S^{sim}(Q)$ structure factors becomes smaller than equilibrium value:

$$\chi^2 = \sum_{i=1}^{N} \left(S^{sim}(Q_i) - S^{exp}(Q_i)\right)^2 / \sigma^2(Q_i), \quad (1)$$

where i is the number of points in the diagram of the structure functions, $\sigma^2(Q_i)$ is the experimental error as a function of diffraction vector Q. Statistical error of the $S^{exp}(Q)$ function counting is found to be <2 %.

The analysis of local structure of the RMC simulated configuration was carried out using tessellation of the models configuration space into the Voronoi polyhedra by means of the "radical planes" method. During the tessellation procedure metals atomic radii were taken as a half of cut-off distances. The tessellation was realized using the algorithm described by Medvedev [17]. The Voronoi polyhedron (VP) is defined as the volume of space, which is the closest to a given atom center then to any other atom centers in the configuration model. In other words, the VP is the geometrical image of the local environment of given atom. VP metric and topological characteristics i.e. the distributions of VP's volumes, face areas, sphericity coefficient ($K_{sph}=36\pi V^2/S^3$, where V is the volume, and S is the surface area of

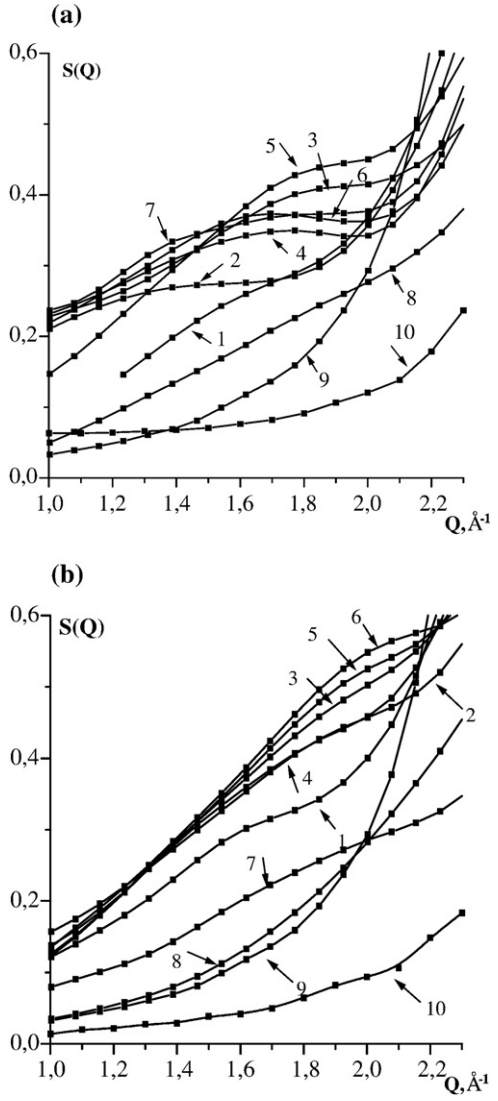

Fig. 2. The structure factor function in the low-Q range of the liquid $Al_{100-x}Co_x$ alloys (a): $1-x=10$, 2–14.3, 3–18, 4–20, 5–23.5, 6–26, 7–28.57, 8–80.7, 9-pure Al, 10-pure Co; and $Al_{100-x}Ni_x$ (b): $1-x=10$, 2–18, 3–25, 4–28, 5–32, 6–40, 7–75, 8–90, 9-pure Al, 10-pure Ni.