



Short-range structural signature of transport properties of Al–Ni melts



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ABSTRACT

Ab initio molecular dynamics simulations are performed to investigate dynamic properties of Al–Ni melts. Along the $T = 1795$ K isotherm, we show that self-diffusion coefficients and the viscosity exhibit a strong non-linear dependence as a function of composition. We further demonstrate that this behavior has an underlying origin in the composition-dependent local structural ordering characterized by a strong interplay between icosahedral short-range order and chemical short-range order in the Ni-rich composition range. Finally we evidence that this structural evolution is at the origin of the breakdown of the Stokes–Einstein relation.

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1. Introduction

Structural and dynamic properties of Al–Ni melts are of technological interest to design commercial superalloys or Al-rich metallic glasses. Understanding diffusion phenomena and viscosity in metallic liquids is of great importance since they represent the key parameters governing solidification and glass formation processes [1–3]. The question of influence of local structural properties on these dynamic properties has attracted much attention in the recent years, but this line of research remains largely open [4,5].

Liquid Al–Ni alloys are known to exhibit appreciable chemical short-range order [6] (CSRO). Therefore, they can be seen as a model system for studying the influence of CSRO on dynamic properties. Indeed, recent quasielastic neutron scattering (QNS) measurements evidence a nonlinear dependence of the Ni self-diffusion coefficient with a strong increase on the Al-rich side [7–9]. Moreover, measurements of the viscosity display also a non-linear dependence as a function of composition [10]. In both cases, these variations are attributed to a pronounced CSRO even if the connection is only established on the basis of classical molecular dynamics (MD) simulations, with an embedded atom model (EAM) potential [7,9,11,12]. It should be mentioned that MD simulations are also of interest for the determination of self-diffusion coefficients for Al since their accurate measurements remain an experimental challenge. In the Ni-rich alloys, the calculated self-diffusion coefficients of Ni and Al, namely D_{Ni} and D_{Al} , are found to be different, D_{Al} being smaller than D_{Ni} . In the Al-rich alloys, D_{Ni} and D_{Al} have similar values and, moreover, they display a strong increase with increasing Al

content, in concordance with the experimental Ni self-diffusion coefficient. For D_{Ni} , there is an overall agreement with the experimental data, within 20%, and the computed viscosity of $\text{Al}_{80}\text{Ni}_{20}$ is also in correct agreement with experimental data.

In a very recent contribution [13], we have studied the structure–dynamic relationship in liquid $\text{Al}_{90}\text{Ni}_{10}$ and $\text{Al}_{80}\text{Ni}_{20}$ alloys using *ab initio* molecular dynamics (AIMD) simulations. We have found that self-diffusion coefficients and viscosity are strongly composition-dependent while their temperature dependence follows an Arrhenius-type behavior in close agreement with experimental data. However, our *ab initio* calculations lead to different values of D_{Al} and D_{Ni} for each composition, contrary to values obtained in classical MD simulations. We have shown that this difference becomes smaller with increasing Ni composition and is mainly due to different degrees of the icosahedral short-range order (ISRO) in local structures of both species.

The present work aims to extend our previous results by performing AIMD simulations to explore the composition dependence of dynamic and structural properties of liquid $\text{Al}_{1-x}\text{Ni}_x$ alloys. We consider the whole range of composition along the $T = 1795$ K isotherm, which corresponds to the experimental study of Refs. [7,8]. First, on the Al-rich side, we obtain a strong composition dependence of D_{Ni} and D_{Al} characterized by a strong decrease with increasing Ni content up to $x_{\text{Ni}} = 0.5$. On the Ni-rich side, *i.e.*, for $x_{\text{Ni}} = 0.5$ to 0.9, we find that self-diffusion coefficients are nearly composition independent, D_{Ni} being close to the value in pure liquid Ni [14]. Moreover, the ratio $D_{\text{Al}}/D_{\text{Ni}}$ becomes close to 1 in this composition range. We obtain also a non-linear behavior of the composition dependence of viscosity. Then, we provide a detailed, microscopic picture of the structural evolution in liquid Al–Ni alloys that can be responsible for the peculiar composition dependence of their dynamic properties. More particularly, we show that this

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structural evolution can be discussed on the basis of a strong interplay between ISRO and CSRO and is at the origin of the breakdown of the Stokes–Einstein relation.

2. Simulation methodology

2.1. First-principle molecular dynamics simulations

AIMD simulations of liquid Al–Ni alloys were performed using the *ab initio* total-energy and molecular-dynamics program VASP [15] developed on the basis of the density functional theory. The VASP code makes use of the projected augmented-wave method to describe the electron–ion interaction [16]. Simulations were performed in the local density approximation [17] using a plane-wave cutoff of 270 eV. Newton's equations of motion were integrated using the Verlet algorithm in the velocity form with a time step of 1.5 fs. All of the simulations were performed in the NVT ensemble and a Nosé thermostat to control temperature. As the Verlet algorithm associated with the small time step used guarantees an excellent accuracy [21] and the liquids, simulated here above the melting point, are ergodic systems, the Nosé thermostat will sample efficiently the canonical ensemble. We used a cubic box of 256 atoms whose size has been adjusted to reproduce the experimentally measured liquid densities [10]. It is worth mentioning that, using these densities, the calculated average pressures for all the compositions studied here were less than 1 GPa which is of the same order of the pressure fluctuations in the simulation boxes. The initial configurations were taken from a well-equilibrated liquid Al₉₀Ni₁₀ alloy at $T = 1795$ K [13] in which the appropriate number of Al atoms was randomly substituted with Ni atoms to mimic studied compositions of Al_{1-x}Ni_x alloys, *i.e.*, $x = 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.75$ and 0.9 . For each composition, the total simulation time was 80 ps, including a 10 ps equilibration period.

2.2. Inherent structures

We base our study of the local ordering on the concept of inherent structures (IS) proposed by Stillinger and Weber [18], which is now commonly used. In this approach, the objective is to bring the configurations in local minima of potential energy landscape (PEL) [19], so called the inherent structure energies (ISE). For this purpose, we have selected ten independent configurations from the molecular dynamics simulations to which we apply a conjugate gradient energy-minimization in order to suppress the kinetic energy, but leaving the configuration in the same basin of the PEL than the high temperature liquid. Therefore, the structural properties can be studied without thermal noise, and the genuine features of the high temperature liquid structures can be better revealed.

2.3. Structural properties

First, we have considered the structural properties through the partial pair-correlation functions $g_{ij}(r)$ ($i, j = \text{Al or Ni}$) determined from 1000 independent equilibrated configurations. We refer the reader to Refs. [20,21] for a detailed description of standard techniques to extract them. We have then calculated the partial nearest-neighbor coordination numbers by integrating the appropriate partial radial distribution functions $RDF_{ij}(r) = x_j 4\pi\rho r^2 g_{ij}(r)$ (ρ being the atomic density) up to the first minimum of the corresponding $g_{ij}(r)$.

Several methods have been proposed to go beyond the angular averaged properties obtained from pair-correlation functions and the coordination numbers, being their integrated counterparts. A first class of methods is based on the bond-orientation order, proposed initially by Steinhardt et al. [22], and recently improved [23]. In this method a development in spherical harmonics about vectors joining all first neighbors of a central atom is carried out. The method can discriminate between liquid and solid local orders, nevertheless it is very sensitive to

the structure distortion which is a drawback for a finer identification of the local ordering of liquids and glasses. Another class of methods to give a three-dimensional view of atomic arrangements is based on a direct geometrical identification of the SRO. The Voronoi tessellation method [24] is the first method proposed in this spirit. It consists in dividing the simulation box into polyhedra of bisecting planes along the lines joining each atom to its first neighbors. The characterization of the local topology of clusters surrounding each atom is obtained by identifying the geometrical properties of these polyhedra with a four index classification. Recently, a method based on collective alignment of clusters around each atom has been proposed [25] that overcomes the difficulty of identifying specific distorted local structures in the Voronoi method due to thermal effects. Another interesting method is the common-neighbor analysis (CNA) [26] in which the local symmetry is determined on smaller basic units, namely atomic pairs. The CNA is considered as simple and robust and is well suited for AIMD simulations [27,28]. These methods are widely used to reveal the icosahedral ordering of liquids and glass-forming materials [1,2,29].

In the present work, we have chosen the CNA which makes it possible to discriminate between various local topologies like fcc, hcp, bcc and icosahedral, as well as more complex polytetrahedral environments. The classification of each pair of atoms according to the number and topology of their common neighbors is done by means of a set of four indices whose detailed description is given in our preceding paper [28]. Here, we apply the CNA to the inherent structures and we focus on the most abundant pairs namely the 15xx = 1551 + 1541, which represent the icosahedral and distorted icosahedral symmetry. The 1431 pair is also found with a significant abundance and can be either classified as distorted icosahedral or distorted closed-packed structures [27].

2.4. Dynamic properties

In the present work, we focus on the diffusion and viscosity Al–Ni alloy. Self-diffusion coefficients are characterized by the individual atom displacement through the mean-square displacement (MSD) for each species i ($i = \text{Al or Ni}$)

$$R_i^2(t) = \frac{1}{N_i} \sum_{k=1}^{N_i} \left\langle [\mathbf{r}_k(t + t_0) - \mathbf{r}_k(t_0)]^2 \right\rangle_{t_0}. \quad (1)$$

In Eq. (1) $\mathbf{r}_k(t)$ is the position of atom k of species i at time t , and angular brackets represent the average over time origins t_0 . The self-diffusion coefficient D_i can be determined from the long time slope of $R_i^2(t)$, namely:

$$D_i = \lim_{t \rightarrow \infty} \frac{R_i^2(t)}{6t}. \quad (2)$$

Alternatively D_i can be calculated from the well-known Green–Kubo time integral of $\Psi_i(t)$

$$D_i = \frac{1}{3} \int_0^{\infty} \Psi_i(t) dt \quad (3)$$

with

$$\Psi_i(t) = \frac{1}{N_i} \sum_{k=1}^{N_i} \langle \mathbf{v}_k(t + t_0) \cdot \mathbf{v}_k(t_0) \rangle_{t_0} \quad (4)$$

is the velocity auto-correlation function for species i obtained from the velocity $\mathbf{v}_k(t)$ of atom k of species i at time t . Consistency of the numerical values of the self-diffusion coefficients calculated from Eqs. (2) and (3) is a strong indication of the correctness of the simulations.

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