Excess Volume in Al-Au Binary Liquid Alloys

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Aluminum-based liquid alloys containing one or more transition metals are found to exhibit pronounced negative excess volumes. This effect is investigated in a combined experimental and simulation study on the example of liquid Al-Au. Al-Au received some interest as it played a role in the context of wire bonding between semiconductor devices and conducting surfaces. Here, it is treated as a model system in order to generally study the excess volume on both, a macroscopic and an atomic scale. Density and excess volumes of the liquid pure components, Al and Au, as well as of alloys samples were measured as functions of temperature and composition. These measurements were carried out by electromagnetic levitation using optical dilatometry. In addition, viscosities of the pure components were measured using a high temperature oscillating cup viscometer. Molecular dynamics (MD) simulations were carried out using the LAMMPS package. Existing embedded atom potentials of pure Al and Au were further developed and applied to the liquid pure components. In addition, the two body part of potential is adjusted for the Al-Au interaction using existing excess free energy data. Good quantitative agreement between simulation and experimental data is obtained for all compositions. In particular, the measured negative excess volumes which are larger than 5% of the ideal volume, are reproduced as well the as the experimentally determined thermal volume expansion coefficients. Structural analysis in the MD simulation shows that medium range order emerges in the alloys, accounting for more efficient local packing and a large negative excess volume in this binary system. Furthermore, it is shown that the excess volume cannot be explained by a simple geometric packing argument. In fact, the effective atomic radii become composition dependent contradicting the conventional hard-spheres picture.