

Thesis Summary

The wetting dynamics at high-temperature have despite their apparent simplicity defied generalization of what was developed for room-temperature systems. The aggressive experimental conditions are responsible for this as well as, for liquid metals, the high oxidation, and compound formation that can possibly occur during wetting processes (ridges, miscibility, etc.)

In this thesis, we investigate liquid metal drops spreading on a solid surface. We combine molecular dynamics simulations with new experiments to investigate the underlying microscopic mechanisms.

The spontaneous spreading of liquid metal (Cu, Ag, Au) and oxide drops on Mo substrates is first studied experimentally using a drop transfer setup combined with high-speed video. Under the experimental conditions used, spreading occurs in the absence of interfacial reactions or ridging making the system non-reactive. The analysis of the spreading data indicates that dissipation at the triple junction can be described in terms of a triple-line friction playing a dominant role in the movement of the liquid front. This is due, in part, to the much stronger atomic interactions in high-temperature systems when compared to organic liquids. The wetting of low melting point metals and Si–Ca–Al–Ti–O glasses on molybdenum is also investigated experimentally. By controlling the oxygen activity in the furnace, glass spreading can take place under reactive or nonreactive conditions. As a result of these experiments, a comprehensive view of spreading emerges in which the strength of the atomic interactions (solid-liquid and liquid-liquid) determines the relative roles of viscous impedance and dissipation at the triple junction in spreading kinetics.

The spreading for Cu, Au and Ag droplets on top of a rigid solid surface modeling Mo (a fixed Ni solid surface mimicking a nonreactive substrate) is then considered via Molecular Dynamics (LAMMPS code) for a more detailed understanding of the spreading process from an atomistic point of view. The dynamics of the base radius and the contact angle are recorded and fitted using the Molecular-Kinetic Theory. A method is described to calculate in the simulations the parameters appearing in this theory. The microscopic parameters of the Molecular-Kinetic model are compared to the fitted values. An agreement between the fitted values and the calculated ones is shown and it gives an atomistic validation that the dissipation of energy within the drop is caused primarily by the friction of liquid atoms over the substrate. Different behaviors are observed between Cu, Ag, Au which are supposed to be linked to the different atomic sizes and inter-atomic distance. To prove this, the effect of the lattice commensurability of the solid surface during the wetting of Ag and Au droplets is then studied by molecular dynamics simulations. The liquids become ordered in a region of 10 Å from the solid due to the liquid/solid interactions and the liquid atoms in contact with the Ni support adopt a solid-like ordering. Different fixed Ni surfaces with different interatomic distances are thus considered to make substrates that are commensurate or not with the liquid. A commensurate substrate is shown to inhibit the diffusion and to increase the friction at the triple-line. After the use of simple atomic metallic liquids, a generalization is made to binary metallic alloys (Cu-Ag above the eutectic point) to observe the influence of composition on the dynamic properties of the liquids and the wetting kinetics. The alloys are more disordered and this disorder enhances the spreading processes. Surface segregation is observed where Ag atoms preferentially diffuse in a monolayer of atoms and Cu wet the solid at the base of the bulk drop. After the studies with rigid substrates, non-rigid Ni substrates are used. A more realistic thermostat is implemented in the simulations. The validation of the Molecular-

Kinetic Theory is also recovered by comparison of fitted versus calculated values of the fitted parameters.

This work thus validates at the microscopic level the mechanisms controlling the dynamics of wetting for high temperature liquid metals on non-reactive substrates.

Papers

Published

- (1) E. Saiz, A.P. Tomsia, N. Rauch, C. Scheu, M. Rühle, M. Benhassine, D. Seveno, J. De Coninck, S. Lopez-Esteban. "Non-Reactive Spreading at High-Temperature: Molten Metals and Oxides on Molybdenum." *Phys. Rev. E*, **2007**, 76:041602.
- (2) A.P. Tomsia, E. Saiz, S. Lopez-Esteban, M. Benhassine, J. De Coninck, N. Rauch and M. Rühle. "Wetting of metals and glasses on Mo." *International journal of materials research*, **2007** 98(12): pp 1238-43.

Submitted or in preparation

- (3) M.Benhassine, E.Saiz, A.P. Tomsia and J.De Coninck. "Non-reactive spreading at high-temperature revisited for metal systems via molecular dynamics." Submitted to *Langmuir*, **2009**:la200901064q.
- (4) M.Benhassine, E.Saiz, A.P. Tomsia and J.De Coninck. "Non-reactive wetting kinetics of liquid metals versus the commensurability of the substrate." In preparation, **2009**.
- (5) M.Benhassine, E.Saiz, A.P. Tomsia and J.De Coninck. "Wetting kinetics of binary alloys using molecular dynamics." In preparation, **2009**.
- (6) M.Benhassine, M.Hou and J.De Coninck. "Wetting of Au on Ni using molecular dynamics: A molecular-kinetic description." In preparation, **2009**.