



Characteristics of spreading dynamics for adsorption wetting at high temperatures



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ABSTRACT

The characteristics of adsorption wetting for metallic fluids at high temperatures were linked to the theories for the room temperatures. Reaction product controlled model is a special form of molecular dynamic model actually. The contribution of adsorption for spreading in a reactive wetting system at high temperatures should not be neglected, and may be in dominant. The relatively large ΔG_w in the molecular dynamic model is a remarkable difference from organic fluid, which is caused by high interaction at solid–liquid interface.

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1. Introduction

Adsorption is an important phenomenon, not just for the catalysis process at room temperature [1], but also for the fabrication of metal matrix composites and brazing process at high temperatures. Recently, the main researches focus on the adsorption at the solid–vapor (S–V) interface or liquid–vapor (L–V) interface [2]. Indeed, it is easy to be characterized for these two interfaces. For S–L interface, the research is usually based on the discussion of equilibrium interface energy and together with the method of microcalorimetry [3]. However, the dynamic behavior of adsorption which may determine the practical process should be paid more attention, especially for the effect on the spreading behavior in the wetting process.

However, the role of adsorption for the improvement of wettability at high temperatures is still controversy. Eustathopoulos [4] emphasized the limited contribution of adsorption for improving wettability, but the precipitation of reaction products at S–L interface should be the main factor. Saiz and Tomsia [5] considered that the moving of the triple line could be ahead of the nucleation of the new phase at S–L interface, and the adsorption of reactant (or interface tensioactive elements) could be a main factor conversely. Both of them admit the contribution of adsorption for improving wettability, but the former focused on initial stages (before nucleation) and limited contribution [6,7], and the latter emphasized the

continuous and non-ignorable contribution throughout the whole spreading. Therefore, whether adsorption contribution could be characterized from spreading dynamics of metallic fluids at high temperatures need to be further confirmed.

The purpose of this study is to reveal the characteristics of spreading dynamics for adsorption wetting at high temperatures, base on the analysis of wetting data at high temperatures and the spreading theories.

2. Theory and method

As suggested by Saiz and Tomsia [5], it is necessary to establish a connection with well developed theories for low-temperature systems, identifying the similarities and pointing out the fundamental differences. As known, the relationship for description of spreading dynamics with well developed theories at low-temperature systems usually base on the variation of $\cos\theta_d - v$, where θ_d is the dynamic contact angle and v is the velocity of the moving of triple line, such as hydrodynamic (HD) model [8] and Molecular dynamic (MD) model [9,10]. $\theta_d - t$ or $R_b^n - t$ are the common relationship for description of high temperature spreading, such as Reaction product controlled (RPC) model [11] and diffusion-limited (DL) model [12]. Here, we assumed that the droplets in all the wetting systems have the same volume and keep constant as spherical cap during spreading. The relationship of $\theta_d - t$ can be transformed to $\cos\theta_d - v$ or $\theta_d - v$, as following,

$$v = Cg(\theta_d) \frac{d\theta_d}{dt}, \quad (1)$$

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where C is a constant about volume, $-\frac{1}{3}\left(\frac{3V}{\pi}\right)^{\frac{1}{3}}$, and $g(\theta_d)$ is a function about dynamic contact angle, $\frac{3(1-\cos\theta_d)^2}{(2-3\cos\theta_d+\cos^3\theta_d)^{\frac{3}{2}}}$, and then all well developed spreading dynamic models can be reorganized in the same relationship, i.e., $\cos\theta_d - v$ or $\theta_d - v$.

The HD model, i.e., viscous dissipation-limited model, the velocity dependence of the contact angle is given by [8,13,14] (for contact angles less than 135°)

$$\theta_d^3 = \theta_e^3 + 9 \frac{\eta v}{\sigma_{lv}} \ln(L/L_s), \quad (2)$$

where θ_e is the final contact angle, η represents the liquid viscosity and σ_{lv} represents the liquid surface tension. L is the capillary length, equal to $\sqrt{2\sigma_{lv}/\rho g}$ (where ρ and g represent the liquid density and the gravitational acceleration, respectively), and L_s is known as slip length. We set $9 \frac{\eta}{\sigma_{lv}} \ln(L/L_s)$ in Eq. (2) as fitting parameter.

The MD model, the energy dissipation is mainly due to adsorption/desorption of fluid elements in the immediate vicinity of the triple line. The velocity dependence of the dynamic contact angle is given by [15],

$$v = 2\lambda\kappa_e \sin h\left(\frac{\sigma_{lv}(\cos\theta_e - \cos\theta_d)}{2nk_B T}\right), \quad (3)$$

where κ_e and λ are the quasi-equilibrium rate constant and the distance between two adsorption or desorption sites, respectively, k_B is the Boltzman constant, and n represents the number of adsorption/desorption sites per unit area (provided that the distribution of the sites is isotropic, $\lambda \approx n^{-1/2}$). We set $2\lambda\kappa_e$ and $\frac{\sigma_{lv}}{2nk_B T}$ in Eq. (3) as the fitting parameters. It is worthwhile to further indicate that κ_e is given by [15],

$$\kappa_e = \frac{k_B T}{h} \exp\left(-\frac{\Delta G_w}{RT}\right), \quad (4)$$

where h is the Planck constant and ΔG_w is the molar activation free energy of wetting that is derived mainly from the solid-liquid interactions.

RPC model, has two forms, one the velocity for the moving of triple line is almost constant, and another is described as following which the too strict requirement of "linear spreading" is removed,

$$\cos\theta_e - \cos\theta_d = (\cos\theta_e - \cos\theta_0) \exp(-k_r t), \quad (5)$$

where k_r is a kinetic constant in s^{-1} . We assumed that the volume of droplet kept constant, and then Eq. (5) can be transferred as following,

$$v = \left(\frac{3V}{\pi}\right)^{\frac{1}{3}} \frac{(\cos\theta_e - \cos\theta_d)}{\sin\theta_d(\cos\theta_d - 1)^{\frac{2}{3}}(\cos\theta_d + 2)^{\frac{1}{3}}} k_r. \quad (6)$$

Here, we set $\left(\frac{3V}{\pi}\right)^{\frac{1}{3}} k_r$ as fitting parameter.

DL model, as suggested by Mortensen et al. [12], the relationship of v and θ_d is

$$v = \frac{2DF_{(T)}(C_0 - C_e)}{e_p n_{v,b}} \theta_d, \quad (7)$$

where D is the diffusion coefficient, e_p is the thickness of the reaction product that forms a wettable surface ahead of the triple contact line, $n_{v,b}$ is the number of moles of reactants per unit volume of the reaction product, $F_{(T)}$ can be approximated as 0.04 [12], C_0 and C_e can be approximated as the concentration of reactive element in reaction product and in the bulk drop. Here, we set $\frac{2DF_{(T)}(C_0 - C_e)}{e_p n_{v,b}}$ as fitting parameter.

3. Results and discussion

Base on the above unified treatment, all reported $\theta - t$ data in the references can be transferred to $\cos\theta_d - v$, and then can be further fitted by the above models. Fig. 1(a) shows the fitting results which base on the reported $\theta - t$ data. It can be found that the MD model (the range reported by Saiz and Tomsia [16], for simple metal/metal systems), RPC model and DL model have overlapped area in the certain range of velocities. For RPC and DL model, both of them are all used for the description of reactive wetting at high temperatures, and thus such overlapped area is appreciable. However, a large number of reactive systems cannot only be fitted by the RPC model but also by MD model (the black data in Fig. 1(a)). The calculated parameters in the MD model were shown in Table 1.

For typical reactive systems (metal/ceramic systems), the λ is less than that organic fluid at room temperature, but ΔG_w is relatively larger which is also larger than that simple metal/metal systems [16]. It may be interesting, although Eustathopoulos and Saiz have the some divergences of opinion; the calculated λ and ΔG_w for the almost same systems have very close values, respectively, e.g., λ and ΔG_w are 2.39 Å and 177.87 kJ mol⁻¹ (calculated from Saiz's data [17]) for Ag-Cu-Ti/Al₂O₃ systems corresponding to 3 Å and 140 kJ mol⁻¹ [6] (reported by Eustathopoulos's group). Eremenko et al. [27] proposed an expression for reactive wetting (Al/Fe (Co and Ni) systems in his study),

$$v = v_\infty \exp(A F_{dr}), \quad (8)$$

where A and v_∞ are constants, F_{dr} means the driving force for spreading in dimensionless form, equals to $\sigma_{lv}(\cos\theta_e - \cos\theta_d)$. We noted that when $\sigma_{lv}(\cos\theta_e - \cos\theta_d) \gg 2nk_B T$, the MD model can be approximated as [15],

$$v = \lambda\kappa_e \exp\left(\frac{\sigma_{lv}(\cos\theta_e - \cos\theta_d)}{2nk_B T}\right). \quad (9)$$

Two expressions are same but with different physical meaning for the parameters. Kang proposed another expression for the description of the wetting behavior of Sn-Pb/Cu and Sn-Ag-Cu/Cu systems (typical reactive wetting) [28],

$$v = M_0 \exp\left(\frac{-\Delta G_w}{RT}\right) \sigma_{lv}(\cos\theta_e - \cos\theta_d), \quad (10)$$

where M_0 is a constant value determined by the material properties of solder and substrate. Further, when $\sigma_{lv}(\cos\theta_e - \cos\theta_d) \gg 2nk_B T$, MD model can be approximated as [15],

$$v = \lambda k^0 \frac{\sigma_{lv}(\cos\theta_e - \cos\theta_d)}{nk_B T} = \lambda \left(\frac{kT}{h}\right) \exp\left(\frac{-\Delta G_w}{RT}\right) \frac{\sigma_{lv}(\cos\theta_e - \cos\theta_d)}{nk_B T}. \quad (11)$$

Two expressions are also same. Ruckenstein and Dunn considered that the gradient of the chemical potential along the interface induced a force per molecule as a driving force (F), and the velocity for the moving of triple line can be expressed as [29,30],

$$v = \delta v_0 \exp\left(\frac{-E_a}{k_B T}\right) \sin h\left(\frac{\delta F}{2k_B T}\right), \quad (12)$$

where δ is a distance for a liquid molecule which moves along solid surface by jumping from one potential well to the next, E_a is energy barrier for a liquid molecule moving ($E_a = \Delta G_w/N$, N is the Avogadro constant), v_0 is the vibration frequency of a molecule in the potential well, and F can be expressed as [29],

$$F = -\frac{1}{n_l} \frac{\partial \mu}{\partial x}, \quad (13)$$

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