

# Enhanced wetting of dual-phase metallic solids by liquid metals: A new effect of interfacial reaction

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## Abstract

The wetting of Cu–Fe two-phase composites by molten Sn is studied by the sessile drop technique under high vacuum at 400 °C. In this system Sn reacts with both solid components, forming intermetallic compounds. It is found that the curve of contact angle vs. the surface fraction of components passes through a minimum, behaviour that cannot be interpreted by existing models describing wetting of heterogeneous surfaces and/or reactive wetting. It is shown that the observed enhanced wetting can be explained by the dissolution contrast of Cu and Fe phases, leading to interfacial microroughness, thus providing an additional driving force for wetting. In order to take into account this new effect of interfacial reactions on wettability, an equation similar to Wenzel's equation is established. It is shown that this equation can explain the change in wettability of composites when Sn is replaced by SnPb eutectic presenting a lower reactivity than pure Sn, as well as the effect on wettability observed when the scale of composite microstructure is changed with the surface fraction of components remaining constant.

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## 1. Introduction

Several studies have been published on the effect of structured surfaces (patterned surfaces or surfaces with controlled microroughness) on their wettability properties (see e.g. Refs. [1–3]). Most of these studies are for room temperature liquids such as water, alcohol or silicon oil (for a review see Ref. [4]). As for wetting in high temperature systems involved in various practical processes (soldering in microelectronics, brazing, tip coating, etc.) it has long been known that grain boundary grooves forming on solid metallic surfaces provide an additional driving force for wetting and microchannels for liquid flow [4–8].

This investigation focuses on the wettability of dual-phase metallic solids by reactive liquid metals. The

selected system consists of molten Sn (and a liquid Sn–Pb alloy) and Cu–Fe composites. The Cu–Fe system presents an immiscibility gap in the solid state and its dual-phase microstructure can be modified by changing both the alloy composition and processing parameters [9]. Wetting of heterogeneous surfaces consisting of B particles dispersed in a continuous A phase by a non-reactive liquid is described by Cassie's equation, according to which the cosine of the equilibrium contact angle is equal to the average of cosines of equilibrium contact angles on the individual A and B solids weighted by the corresponding surface fraction  $f$ :

$$\cos \theta = f_A \cos \theta_A + (1 - f_A) \cos \theta_B \quad (1)$$

However, the Sn/Cu [10] and Sn/Fe [11] systems are both reactive, leading to the formation of intermetallic compounds (IMCs). For this type of system it has been shown [12,13] that both thermodynamics (driving force)

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and kinetics (spreading rate) are controlled by the reaction product formed at the interface. According to this model, referred to in the following as the RPC model, the steady contact angle observed in a reactive system is nearly equal to the equilibrium contact angle in the corresponding non-reactive system, i.e. the contact angle that should be formed by the same liquid on the reaction product. When more than one IMC is formed at the interface, the final contact angle is on the IMC in direct contact with the liquid at the substrate–liquid–vapour line (referred to as the triple line).

This study focuses on the following question: is it possible to apply Cassie's equation to a dual-phase substrate taking for  $\theta_A$  and  $\theta_B$  the steady contact angles measured for the reactive liquid on pure A and B? In other words, could Cassie's equation also be applied to reactive systems through the predictions of the RPC model? In order to answer this important question, sessile drop experiments are performed with the same liquid (pure Sn) by varying the surface fraction of copper between 0 and 1. Although the main aim of the study is to measure steady contact angles concerned by Eq. (1), some information will also be given on spreading kinetics. Experiments are also conducted varying the scale of composite microstructure, keeping the surface fraction of copper constant. Moreover, the steady contact angle vs. copper surface fraction curve was determined for the SnPb eutectic presenting a lower reactivity with both Cu and Fe than pure Sn.

## 2. Experimental procedure

Dual-phase Cu–Fe composites with fine microstructure were fabricated by vacuum induction melting and quenching. To obtain a coarse microstructure, the composites were made by induction melting followed by cooling down in the furnace at a controlled rate. For comparison purposes, pure iron and copper substrates were also used. Prior to the experiments, all the substrates were polished using 1  $\mu\text{m}$  diamond paste, cleaned ultrasonically with purified water and acetone and dried with an air gun. Pure (99.9%) Sn balls weighing  $2.7 \pm 0.1$  mg and SnPb eutectic (63 wt.% Sn, 37 wt.% Pb) solder balls weighing  $3.1 \pm 0.2$  mg were etched in 5% HCl + 95% H<sub>2</sub>O for 10 min and then cleaned in the same way as the substrates.

Wetting was studied by the sessile drop technique under high vacuum ( $10^{-5}$  Pa). The wetting couples were heated at a rate of  $5^\circ\text{C min}^{-1}$  to  $400^\circ\text{C}$  and held at that temperature. The wetting process was recorded in situ with a camera through a side-view window of the chamber. From the drop profile the cap diameter was measured and the contact angle determined by the tangent method. The values of steady contact angles given in Figs. 3 and 6 are the average of three independent experiments.

After the sessile drop test, cross-section samples were made by cutting through the solder caps. Scanning electron microscopy (SEM) with associated energy dispersive spectroscopy (EDS) was used to characterize the substrate

microstructure and reactive interface. The surface fraction of Cu phase and the average linear size of the particulate phase of dual-phase substrates were measured by image with Image-Pro Plus 6.0. The surface fraction of the Cu phase was measured with a proper threshold value in a binarized black–white image. The average linear size was obtained by measuring the linear size of particles intersected by a straight line of unit length.

## 3. Results

### 3.1. Microstructure of Cu–Fe composite

The microstructure of a quenched Cu–Fe composite with 50 wt.% Cu (50Cu50Fe) is shown in Fig. 1, in which the iron phase is dark and the copper phase is bright. It can be seen that this composite has a fine dual-phase microstructure consisting of Fe particles embedded in interconnecting Cu matrix. The surface fraction of Cu matrix was  $\sim 0.47$ .

Similarly, through image analysis, the surface fraction of the Cu phase in 19Cu81Fe, 31Cu69Fe, 67Cu33Fe and 82Cu18Fe composites was 0.10, 0.24, 0.63 and 0.75, respectively.

### 3.2. Wetting of Sn on Cu–Fe composites

The variation in contact angle and cap diameter with time during temperature rise to plateau temperature  $T_{pl} = 400^\circ\text{C}$  and subsequent holding period at  $T_{pl}$ , are shown in Fig. 2 for three types of substrate, pure Cu (a), 50 Cu–50 Fe composite (b) and pure Fe (c). In all the experiments steady contact angles were reached but with different spreading rates. Despite the higher temperature range within which spreading on iron occurred, the average spreading rate on iron, equal to  $0.58 \mu\text{m s}^{-1}$ , is 3–4 times lower than in the case of copper. Spreading on 50 Cu–50

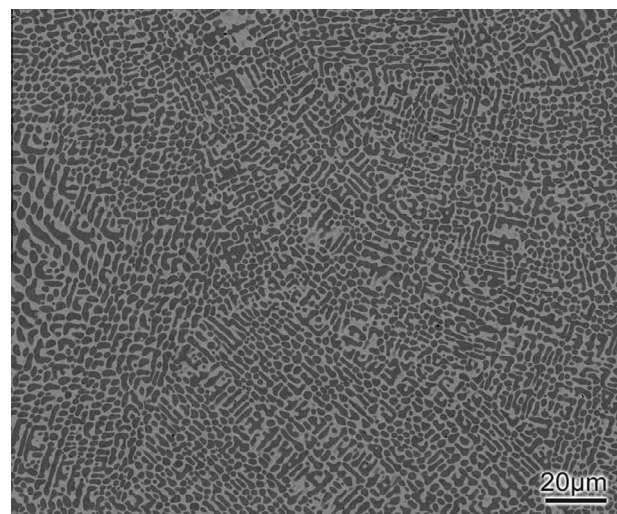


Fig. 1. Microstructure of a quenched Cu–Fe alloy with 50 wt.% Cu. Iron is dark, copper is grey.

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