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Gibbs-Thomson effect as driving force for liquid film migration: Converting metallic into ceramic fibers through intrinsic oxidation

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ABSTRACT

Liquid film migration is of great practical importance in materials engineering. The phenomenon has been shown to depend on thermal gradients and coherency strain, but no single driving mechanism seems capable of justifying the whole array of experimental observations. On the other hand, the inevitable capillarity effects are often disregarded due to the unknown 3-dimensional geometry of the system. Here, we present evidence of liquid film migration governed primarily by capillarity through a microstructural setup of cylindrical interfaces that allows clear interpretation and modeling. The experiments rely on the strong oxygen-gettering ability of tantalum fibers dispersed in a tungsten matrix and on field-enhanced diffusivity provided by pulse plasma compaction. Tantalum scavenges the residual oxygen present in the *W* powder and, as a result, oxide films grow around the fibers. These oxide tubes, in liquid state during sintering, migrate toward the fiber axis and eventually become oxide rods surrounded by metallic Ta. The process is driven by the Gibbs-Thomson effect that generates the required composition gradient across the liquid film. An analytical description of the film evolution is implemented by combining the incoming *O* flux with capillarity-driven migration. Possible contributions from other mechanisms are examined and the relevance of the Gibbs-Thomson effect to the general phenomenon of liquid film migration is established.

1. Introduction

The importance of liquid film migration (LFM) is widely acknowledged in materials engineering (see [1] for a review). The phenomenon can be considered beneficial in liquid phase sintering of refractory metals [2], while it is severely detrimental in processes such as warm forming of aluminum [3]. A variety of driving forces can cause LFM but a consensual model, valid for all boundary conditions, remains elusive [4]. Regardless of the system configuration, some aspects are common to LFM episodes:

- The film comprehends a pair of interphase interfaces, coupled kinetically by the liquid cohesive forces but imparting individual thermodynamic contributions to the system.
- The movement is largely independent of enthalpic effects since the latent heat needed for melting at the leading interface is provided by solidification at the trailing interface.
- Migration is characteristically slower than the interfacial atomic kinetics; hence, the solid on either side of the film is in thermodynamic equilibrium with the adjacent liquid. Yet, the two different interfacial states result in a net flux of heat and/or solute across the film.
- The ensemble is highly mobile as the migration rate is essentially controlled by the velocity of the solid/liquid interfaces and by diffusion in the liquid phase.

Nonetheless, the fundamental causes of the migration can be multiple, and a quantitative understanding requires careful evaluation of the contributing mechanisms.

Coherency strain as driving mechanism

Under isothermal conditions, such as in liquid phase sintering, the prevailing theory is that LFM is driven by diffusional coherency strain [1] though this has also been refuted [5–8]. In this mechanism, solute atoms diffuse from the liquid film into the adjacent solid, where they generate coherency strain due to lattice misfit

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