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Liquid-like interface complexion: From activated sintering to grain boundary diagrams

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ABSTRACT

This article reviews recent experimental and theoretical results that elucidate the mysterious subsolidus activated sintering mechanism. A related new concept of liquid-like interface complexion and its broad applications in comprehending microstructural evolution and materials properties are discussed. A recently proposed thermodynamic model is discussed and further elaborated with new analyses that have not been published before. A long-range scientific goal is to develop quantitative and predictive grain boundary diagrams as a new tool for realizing predictable fabrication of materials by design.

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1. Introduction

Subsolidus (solid-state) activated sintering refers to the phenomenon whereby densification rates are significantly improved upon minor addition of sintering aids at temperatures when the bulk liquid phase is not yet stable [1–6]. For example, sintering of tungsten can be significantly enhanced with the addition of <1% of Ni, Pd, Co or Fe, and the accelerated sintering can initiate at as low as 60–85% of the corresponding bulk eutectic or peritectic temperatures [1,2,4,5]. Subsolidus activated sintering has also been observed in various ceramic systems [3,6]. The exact mechanism of subsolidus activated sintering has puzzled the materials community for decades.

In parallel, geophysicists have long suspected that "sintering" of snow is related to the premelting of ice [7]. Premelting, also known as "surface melting", refers to the stabilization of quasi-liquid interfacial layers below the bulk melting temperature in a unary system [7]. Since the consolidation of ice results in densification, enhanced grain boundary (GB) transport is inferred. While surface premelting has been extensively studied, the existence and importance of GB premelting in unary systems are somewhat controversial [8,9]. On the other hand, in binary and multicomponent systems, "premelting like" films can in principle be stabilized at GBs over wider ranges of undercoolings, because GB structural disorder can be promoted by concurrent solute adsorption [10,11]. Specifically, ice premelting can be significantly enhanced by minor ionic impurities via electrostatic interactions [7,12,13]. Phenomenological similarities among ceramics, metals and ice clearly exist.

Recent high-resolution transmission electron microscopy (HRTEM) studies revealed the stabilization of impurity-based, quasi-liquid, interfacial films well below the bulk eutectic temperatures in both ceramic [6,11,14,15] and metallic [4,16] activated sintering systems (Fig. 1), thereby suggesting that subsolidus activated sintering is due to enhanced diffusion in these quasi-liquid (premelting like) interfacial films. Furthermore, related concepts and theories of GB complexion (phase) transitions (see Section 3 for elaboration) were developed [10,17,18]. Most recently, CALPHAD (CALculation of PHAase Diagrams) methods and statistical thermodynamic models were combined to predict the stability of subsolidus quasi-liquid intergranular films (IGFs) and the related onset activated sintering temperatures in doped tungsten [19]. This interfacial thermodynamic model has broad applications for understanding GB-controlled materials processing and properties. This review examines these recent developments and future opportunities.

2. Impurity-based quasi-liquid interfacial films

Nanometer-thick, impurity-based, intergranular "glassy" films or IGFs have been widely observed in ceramics [11,20–22]. Recently, similar interfacial films have been found at metallic GBs

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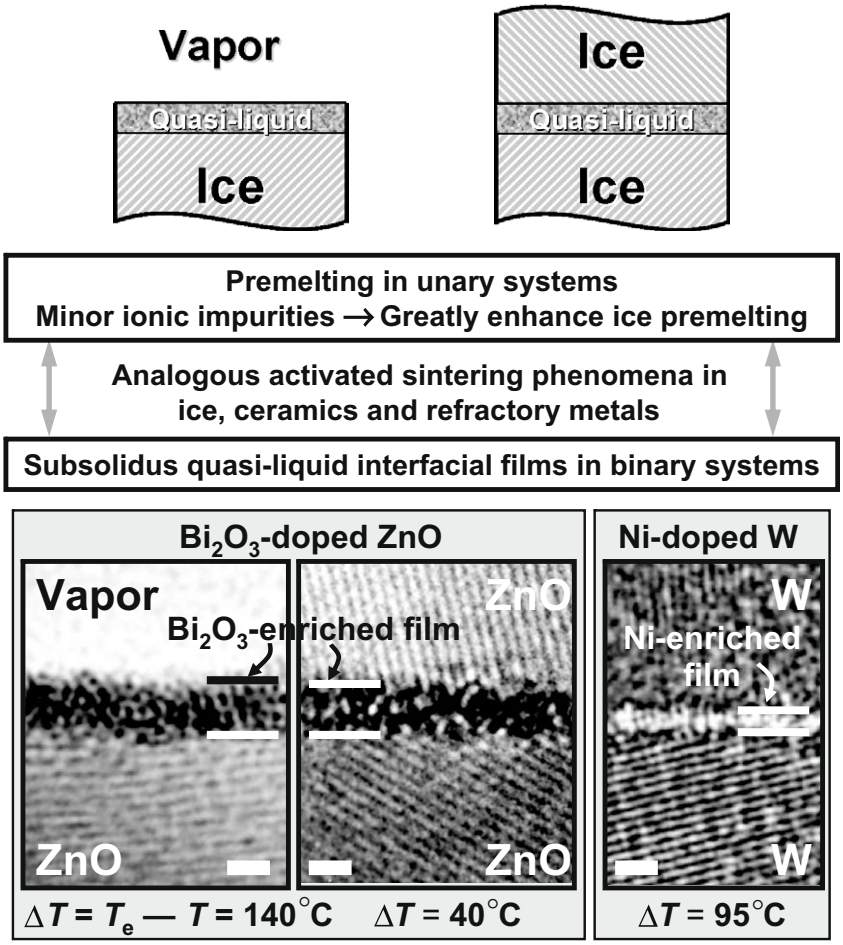

Fig. 1. Quasi-liquid interfacial films in ice, ceramics and refractory metals. Recent HRTEM experiments revealed the stabilization of impurity-based, quasi-liquid, interfacial films at subsolidus temperatures in Bi_2O_3-doped ZnO [6,15,32] and Ni-doped W [4,16], which are model ceramic and metallic systems, respectively, for studying subsolidus activated sintering. These HRTEM images were taken from cooled specimens, and these films are likely more disordered and wider at the sintering temperatures. The HRTEM image for Ni-doped W was enhanced by FFT image filtering [4,16] to clearly distinguish the thin IGF. All scale bars represent 1 nm.

[11,16], metal-oxide interfaces [11,23,24], and free surfaces [15,24–29]. These impurity-based interfacial films exhibit the following distinct characteristics [11]:

- self-selecting or "equilibrium" thickness on the order of 1 nm;
- large structural disorder (yet with partial order imposed by abutting crystals); and
- an average film composition that markedly differs from that of the corresponding (stable or metastable) bulk liquid/glass phase (even if these quasi-liquid interfacial films are in equilibrium with the bulk liquid/glass phase).

These nanoscale interfacial films are non-wetting films [11,15,21,22], and they can alternatively be understood to be

- equilibrium thickness liquid-like films in a high-temperature colloidal theory [11,15,20–22,30]; or
- multilayer adsorbates formed from coupled premelting [7] and prewetting [31] transitions [10,11,15,22].

Recently, stabilization of impurity-based, quasi-liquid, intergranular and surficial films below the bulk eutectic/solidus temperatures has been observed for binary ceramics [11,15,25–29,32] and metals [16] (Fig. 1), where an analogy to premelting in unary systems [7] can be made. A recent critical review of these quasi-liquid interfacial films can be found in Ref. [11].

In the literature, these "quasi-liquid" or "liquid-like" interfacial films were often termed as "glassy" or "amorphous" films, which is not rigorously correct because some partial structural order generally exist in these films. Impurity-based films of similar character in metallic alloys were also called "prewetting" or "premelting" films [33–35]; further discussion of the rather subtle usages of these terminologies can be found in Section 4.5.4 in Ref. [11].

3. Grain boundary transitions and complexions

Like bulk phases, GBs can undergo first-order or continuous transitions. GB transitions in unary systems include premelting [7,17] and roughening/faceting [36,37]. In particular, a recent atomistic simulation demonstrated temperature-induced GB disordering and widening in Si [38]. For multicomponent systems, GB wetting transitions have been investigated for both metals [33] and ceramics [22]. The well-known Fowler–Guggenheim model also forecasts a first-order GB adsorption transition (i.e. a discontinuous jump in GB excess with increasing chemical potential) for strong adsorbate–adsorbate interactions. GB transitions and kinetics are often influenced by interface anisotropy [39,40].

In 2006, Tang, Carter and Cannon predicted "GB transitions in solute adsorption" that are "coupled with localized structural order–disorder transitions" in binary alloys using a diffuse-interface model [10]. These transitions can be considered as coupled GB prewetting [31] and premelting [7] transitions. In the original

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