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Gravity effects on reactive settling of transition metals in liquid aluminum under SHS conditions

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Abstract

The reactive settling of W, Ta, Ni, and Ti in liquid aluminum was investigated under terrestrial and microgravity conditions. Settling occurred in some systems and total dissolution occurred in others. Analyses were made based on Stoke's settling time relative to dissolution time based on two approaches: dissolution based on a diffusion-controlled process and based on solubility limit. The experimental results are in agreement with the solubility model. The effect of solid volume fraction was also investigated. The results showed a critical volume fraction of about 20 vol.%, in agreement with literature values. Effect of particle size was investigated and the observations are consistent with the analysis of settling and dissolution times, indicated above. The absence of settling of the intermetallic phases in all systems was attributed to solid skeleton formation, and confirmed by results on low solid content experiments.

Keywords: Self-propagating high-temperature synthesis (SHS); Aluminum; Segregation; Intermetallic compounds; Reactive settling

1. Introduction

In the synthesis and processing of materials where a liquid phase is present, gravity can play an important role in the microstructural development of the product through phase segregation. Non-uniform microstructures in liquid-phase sintering containing relatively low solid-phase volume fraction are anticipated and attributed to segregation induced by gravitational forces. Experimental observations and modeling analysis have provided confirmation for these anticipations [1,2]. Segregation also occurs during melting; settling of the solid phase was observed under slow furnace heating conditions [3–6].

Settling (sedimentation) occurs when the solid content is less than that required to form solid phase contiguity. A critical volume fraction of the solid is required for the formation of the interconnected structure; its value has been reported to vary from 0.2 to 0.6 [7–9]. In systems with solid fractions higher than the critical value, sedimentation is inhibited by the formation of a solid skeletal structure. On the other hand, in systems where the volume fraction is less than the critical value, solid particles settle according to Stoke's law. Observations have been reported which show that, during settling, particles do not undergo dissolution in the liquid phase [5,6]. Thus, initially after settling, the liquid is the pure phase of the lower melting component and the settled particles are the pure form of the other, higher melting component. Interaction between the liquid and the solid occurs subsequently in the settled (mushy) region [5,6].

Phase segregation also occurs in the self-propagating high-temperature synthesis (SHS), a process used extensively to prepare a large number of material systems

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[10–12]. With few exceptions, the adiabatic combustion temperature in SHS is higher than the melting point of one or more reactants or the product itself [10,11]. Thus the presence of a liquid phase in the combustion zone is a common occurrence and segregation of phases is anticipated and is observed. This is exemplified by research whose objective is to separate product phases, either as a desired synthesis goal (e.g., casting) or as a means to lay down a protective coating on a substrate [13,14].

The effect of gravity on phase segregation and the resulting microstructure in SHS reactions has been investigated for various systems [15–18], and has been utilized in some cases to separate product phases under higher gravitational conditions [19]. Phase segregation was observed with the sedimentation of the heavier phase and the segregation of the lighter phase, including porosity [18,20]. Segregation of pores through buoyancy was observed under terrestrial conditions, while uniform distribution of pores was seen under 0*g* conditions [21].

Settling of a solid in a liquid is a well-understood phenomenon, governed by Stoke's law under isothermal conditions. However, under SHS conditions the process is complicated by the non-isothermal conditions, short process times (s), non-equilibrium chemical conditions, and by interference from other processes (e.g., new phase nucleation and growth). Therefore, settling under SHS conditions (i.e., reactive settling) is expected to depend on additional parameters arising from these conditions. In this study the reactive settling of solid transition metals in liquid Al is investigated under terrestrial and reduced gravity conditions, at two different reaction temperatures (1100 and 1600 °C). Experiments were performed with four transition metals: W, Ta, Ni, and Ti.

2. Experimental procedure

Experiments in 0*g* were performed on board NASA's KC-135 Reduced Gravity Aircraft at the Johnson Space Center (JSC). The plane can sustain weightlessness with gravitational disturbances on the order of $\pm 10^{-2}\,g$ [22]. Experiments were done in Combustion Synthesis of Materials™ (COSYM) equipment from Guignè Int. Ltd. Canada. Modified versions of COSYM and the reaction chambers were used. The most important aspect of the modification is the introduction of an electric field. Details of these modifications are presented in recent publications [22,23]. An experimental technique, commonly known as chemical oven, was used. In this approach the sample is embedded within a SHS reacting mixture, whose SHS reactions produce the heat to propagate the reaction in the sample. The chemical oven and the sample, referred to here as the specimen, are shown schematically in Fig. 1. Each specimen was loaded into a reaction chamber, which was sealed and then evacuated and refilled with ultra-high-purity argon gas to about 1 atm pressure.

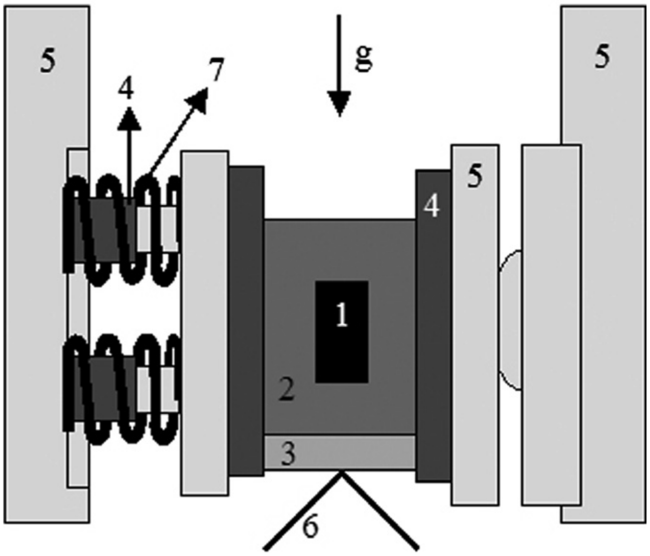

Fig. 1. Schematic of specimen setup: (1) sample; (2) chemical oven; (3) igniter layer; (4) graphite electrodes; (5) copper electrodes; (6) tungsten igniter wire; (7) stainless steel springs.

Two different chemical oven reactions were used: Nb + 2Si and Ti + Al. The reaction between Nb (~1–5 μm, <20 μm) and Si (−325 mesh) resulted in a self-sustaining combustion wave (without field activation [24]) with a peak temperature of about 1600 °C. However, the Ti (−325 mesh) + Al (~20 μm) reaction is not self-sustaining due to its relatively lower adiabatic temperature, so it was necessary to use an electric field for activation. A current of 300 A was applied during each reaction. With this current the reaction between Ti and Al reached a peak temperature of 1100 °C. In both types of chemical oven the peak temperature is achieved at the combustion wave front and decreases after the passage of the wave at a rate of about 47 °C s^{-1}.

Four different sample systems were prepared using elemental powders of W, Ta, Ni, and Ti with powders of Al. Characteristics of the powders used are given in Table 1. Similar particle sizes (<45 μm) were used for all the transition metals in order to normalize the size effect and thus only compare effects of density and solubility. Since aluminum melts at the combustion temperatures, its particle size should not play a role in the process. The four transition metals were mixed with Al to give about 9 vol.% (W, Ta, Ni or Ti) concentrations. Powders were mixed in a glass jar with alumina balls (1:1 powder to ball weight ratio) for 1 h in a Turbula® mixer (Glen Mills Inc. T2C) to form

Table 1
Characteristics of reactant powders

Material	Particle size (μm)	Density (g cm^{-3})	Purity (%)	Supplier
Al	~20	2.40	99+	Aldrich
W	<45	19.35	99.9	Alfa
Ta	<45	16.4	99.9	Alfa
Ni	<45	8.90	99.8	Alfa
Ti	<45	4.51	99.7	Alfa

Densities are given at room temperature. Density of Al is given for liquid at melting point.