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Coarsening of grain-refined semi-solid Al–Ge32 alloy: X-ray microtomography and in situ radiography

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Abstract

The Lifshitz, Slyozov and Wagner theory (LSW) describes the coarsening of low volume fraction dispersed particles in a supersaturated solution as governed by a $t^{1/3}$ power law, while stating that ripening occurs in a self-similar manner. Only a few experiments have reported three-dimensional (3D) coarsening in binary semi-solid alloys, which differs from the LSW theory. We report here on in situ coarsening of Al–Ge32 (wt.%), which is used as a model system for a large variety of technical alloys. Numerical analysis of 2D and 3D images of the microstructure measured by X-ray radiography and microtomography reveals the evolution of the solid particles during annealing. Ripening of a grain-refined particle network is found to be quite well described by LSW theory, although somewhat smaller exponents ($t^{1/4}$–$t^{1/5}$) are found. Changes in coarsening behavior are observed in samples which are thinner than 0.5 mm, as well as in non-equiaxed alloy microstructures, characterized by anisotropic dendrites.

Keywords: Microtomography; Radiography; 2D/3D image analysis; Technical binary alloys; LSW

1. Introduction

The microstructure of binary metallic alloys such as Al–Si and Al–Ge is characterized by a primary solid phase and an eutectic phase: during solidification the primary solid phase nucleates at the liquidus temperature and continues to grow until the solidus temperature is reached and the remaining melt freezes, forming the fine-structured eutectic phase. The resulting primary solid phase microstructure depends to a large extent on the cooling rate and the distribution of nucleation sites, and it is desirable that a globular microstructure forms. Whenever prealloyed metals are partially remelted and are then annealed at a temperature above the solidus, as often occurs in industrial casting processes, coarsening of the primary solid is observed [1,2]. This coarsening occurs due to dissolution of the eutectic phase, which results in additional "ripening" of the solid phase [3,4]. In particular, during rheo- and thixocasting, the alloy is heated to the semi-solid state before it is cast [5,6] (for reliable and successful casting processes and to obtain the designed final components, stringent control over the microstructure is crucial). The morphology of the primary phase can coarsen in various ways when an alloy is held in the semi-solid state during processing [7]. Many parameters contribute to this variation, including flow of the slurry, temperature and possibly sample dimensions [8,9]. Under the assumption that the primary phase

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forms an agglomeration of many particles, coarsening is considered to be the result of two processes: isolated particles growing at the expense of smaller particles [10,11] (cf. Fig. 1a) or connected "touching" particles merging into larger particles by coalescence [12,13] (Fig. 1b). These descriptions are of course only models, because in reality many technical alloys form semi-solid skeletons that coarsen in a different way (see Fig. 1c).

The partition of solid (S) and liquid (L) phases in binary alloys can be expressed in terms of volume fractions g_S and g_L (note that $g_S + g_L = 1$). During annealing, the solid particles change their volume and grow. The average volume, which for sufficiently spherical particles can be approximated to be proportional to the cube of the average diameter, increases linearly with the annealing time, according to

$$d(t) = K(t - t_0)^{1/3} \tag{1}$$

Here, $d(t)$ is the average diameter at time t, t_0 the time when $d = 0$ and K is the growth rate and the coarsening exponent is 1/3 according to Lifshitz, Slyozov and Wagner (LSW) theory [14,15]. Eq. (1) is considered a valid description for growth and coarsening of spherical particles in a supersaturated solution. The LSW theory also claims that at any time during coarsening, the average particle diameter maintains a constant ratio to the width of the particle diameter distribution at any given time. Consequently, self-similarity is an expected characteristic of the structure as it coarsens.

One of the assumptions of the LSW theory is that g_S has to be small, so that individual particles may be considered "non-interacting". A small g_S is fundamental to the interpretation of many of the dynamic processes that are related to coarsening. This is because moderate g_S values result in interactions between neighboring grains, that bring about higher concentration gradients in the liquid while increasing K [16–18]. For semi-solids we note that with g_S exceeding a percolation threshold (all particles become interconnected), the particles start to form a three-dimensional (3D) skeleton which is characterized by agglomerates that make identification of single particles difficult [17,19].

The liquid film migration model [20] extends LSW theory further to include significantly higher solid fractions than originally proposed, accounting for connected particles whereby mass transfer occurs along the connection necks. Most experimental models explicitly predict broader size distributions of particles, compared to the ideal dispersion that was considered in the LSW theory. Theoretical predictions of the dynamics of the process have provided possible explanatory mechanisms for deviations from classical LSW theory [21]. Despite these differences, coarsening theories confirm Eq. (1) and most assumptions result in similar linear relationships between average particle volume and annealing time. By using scaling functions and assuming self-similar statistical ensembles, the universality of the $t^{1/3}$ growth has been demonstrated in three dimensions for a variety of phase-separating processes [22]. Recently it has been shown that even for anisotropic metallic microstructures consisting of dendrites, the $t^{1/3}$ relation holds, if $d(t)$ in Eq. (1) is replaced by a length scale characteristic of the solid–liquid interface [23]. Exponents of 1/4 and 1/5 are thought to be related to the coarsening of precipitates located at grain boundaries and dislocations, respectively. However, even these cases are only different from LSW if volume diffusion is truly negligible [21].

To the best of our knowledge, only few in situ measurements have provided direct evidence for any of the theoretical predictions of coarsening of a semi-solid network of binary metallic alloys [24]. What is known about the evolution of the particle size distribution is mainly based on measurements obtained from 2D images [2,25,26] and much remains unknown about the 3D coarsening. Current state-of-the-art technology allows the visualization of the 3D characteristics of the particles of the primary solid, enabling us to provide direct answers to many questions. In this work, the coarsening of a non-agitated semi-solid network of particles (cf. Fig. 1c) is monitored in grain-refined Al–Ge32 (wt.%) alloy. When this alloy is heated to a temperature above the solidus and below the liquidus, a dispersion of solid Al-rich particles in a liquid Al–Ge solution takes place. In contrast to the destructive and time-consuming serial sectioning methods [17,19,23,27] we use non-destructive X-ray imaging to produce dynamic radiographs and static tomograms of the microstructure in

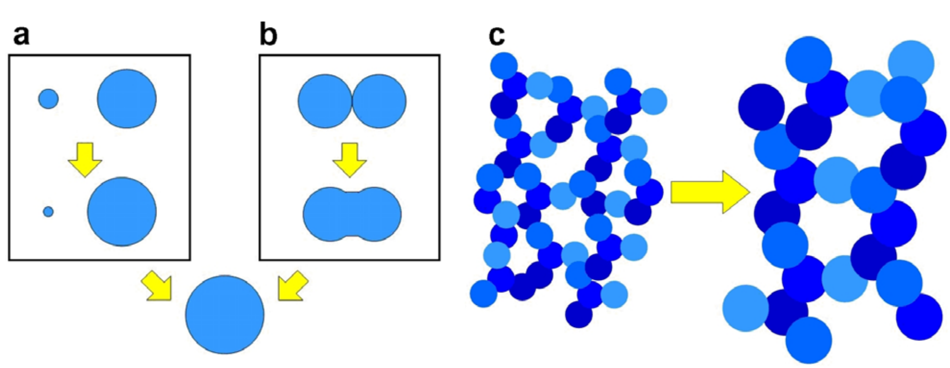

Fig. 1. (a) Coarsening of dispersed particles. Larger spheres grow at the expense of smaller ones, (b) coarsening of connected particles of similar size via coalescence and (c) coarsening of a network of agglomerated particles preserving its self-similar morphology.