

# Experimental determination of the nucleation rate of melt in a solid solution

Stephanie Lippmann\*, Marcel Fink, Markus Rettenmayr

*Otto Schott Institute of Materials Research, Friedrich Schiller University, Löbdergraben 32, 07743 Jena, Germany*

Received 30 November 2013; received in revised form 20 February 2014; accepted 24 February 2014

Available online 18 April 2014

## Abstract

An experimental method for measuring the nucleation rate of liquid in a solid solution using partially melted and resolidified samples is presented. The size distributions of liquid droplets that formed in the grain interior during annealing in a temperature gradient are determined from the size distribution of secondary-phase particles resulting from resolidification of the droplets. Quantitative microstructural analysis considering correcting sectioning artifacts is used. In combination with numerical simulations that calculate the droplet growth on the basis of the experimental temperature history, the nucleation rate of liquid is determined as a function of time. Depending on the location in the temperature gradient, both the size distributions and the nucleation rates exhibit either one or two maxima. The transition from monomodal to bimodal distributions at a temperature less than 10 K above the solidus temperature indicates the existence of two different types of nucleation sites for melting in the grain interior. The transferability of the time-dependent nucleation rate of liquid on the nucleation rate of solid is discussed in view of the differences from the established concepts of athermal, thermal and instantaneous nucleation.

© 2014 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.

*Keywords:* Nucleation rate; Phase transformation kinetics; Microstructure

## 1. Introduction

The nucleation rate of solid from the melt is one of the key parameters that determine the grain size distribution and thus various mechanical and functional properties of crystalline materials. However, it is by no means trivial to assess nucleation rates, either experimentally or theoretically.

The fundamental approach for predicting nucleation rates is based on the classical nucleation theory (e.g. [1–3]). According to the work of Turnbull [4], thermal fluctuations in the melt induce the formation of a number of clusters  $N^*$  with a size that reaches the critical size for nucleation  $r^*$ :

$$N^* = N \cdot e^{-\frac{\Delta G^*}{k_B T}} \quad (1)$$

$N$  is the number of potential clusters,  $\Delta G^*$  is the change in the Gibbs free energy when the cluster size equals the critical radius  $r^*$ ,  $k_B$  is the Boltzmann constant, and  $T$  is the temperature.

In Ref. [4], the authors distinguish between thermal and athermal nucleation mechanisms. The thermal mechanism assumes first a cooling step to a temperature below the melting point, followed by isothermal holding at this temperature. After an incubation time, nucleation starts, and the nucleation rate increases up to a given value, which then remains constant. This approach does not consider the consumption of nucleation sites while the nuclei grow, or the latent heat release.

Athermal nucleation is assumed to occur when the temperature decreases continually and a constant nucleation rate will not be observed [4]. In this case, there is insufficient

\* Corresponding author. Tel.: +49 3641 947768; fax: +49 3641 947792.  
E-mail address: [stephanie.lippmann@uni-jena.de](mailto:stephanie.lippmann@uni-jena.de) (S. Lippmann).

time for nucleus formation in the classical sense. However, at low temperatures, the clusters retained from higher temperatures exceed the critical size and become viable nuclei. It has been pointed out that the athermal nucleation mechanism can only be effective during rapid quenching [5].

Rappaz [6] suggests a model for the prediction of the mean grain size that assumes instantaneous heterogeneous nucleation without incubation time on a distribution of inoculant particles that act as heterogeneous nucleation sites. It is assumed that the potency of the nucleation sites follows a Gaussian distribution, and the activation of a particular class of sites depends on the undercooling achieved. In this model, the former unrealistic assumption of a constant nucleation rate is amended by an extinction factor that limits the final grain number.

In principle, direct experimental observation of nucleation is possible using calorimetric or radiation (protons or X-ray) methods. Bokeloh et al. [7] determine the nucleation rate of pure Ni using differential scanning calorimetry. They observe nucleation with respect to the achieved undercooling in small samples that allow only one single nucleation event. Numerous repetitions of the heating/cooling cycles eventually allow nucleation rates corresponding to homogeneous nucleation in pure substances to be determined.

Documented radiation methods that bear potential for direct observation of phase transformations are limited in spatial and/or temporal resolution and show microstructural details only after a considerable increase in size of the former nuclei. For experiments using proton radiation, a resolution in the order of 1 mm is achieved [8], several orders of magnitude above the size of a critical radius. For experiments using synchrotron radiation, the currently employed third generation of detectors attains a resolution of 100 nm at a frequency of 20 Hz [9]. However, for a finite sample size and maintaining a sensible temporal resolution, a spatial resolution that does not exceed 2  $\mu\text{m}$  [10] is realistic. The sample geometry in synchrotron experiments (flat thin samples with a thickness of  $\sim 200 \mu\text{m}$ ) can further complicate the evaluation of the nucleation rate. Heterogeneous nucleation at the crucible wall or at the surface oxide layer, on the one hand, and buoyancy that causes motion of the nuclei, on the other hand, can significantly affect the measured nucleation rates.

Investigations on the formation of liquid in a solid solution are scarce and mostly do not focus on the nucleation stage. Recently, the present authors [11] presented an experimental method in which partially liquid samples are quenched, and traces of early stages of melting remain visible. In experiments where a sample consisting of a homogeneous single-phase alloy is exposed to a temperature gradient for a short time and partially melted, former liquid regions can be located in the sample after resolidification, owing to the formation of a secondary phase. In a temperature gradient, different intermediate stages of melting are present in the same samples, depending on the local temperature that is reached in the gradient. Small spherical

secondary-phase particles with sizes between 50 nm and 1  $\mu\text{m}$  clearly stem from single nucleation events and are accessible to analysis [12].

The present paper introduces a method to determine the (temporally resolved) nucleation rate of liquid droplets in a solid solution by a combination of experimental observations using the above-mentioned gradient annealing experiment and simulation calculations of droplet growth.

## 2. Method

### 2.1. Experimental

Rods 8 mm in diameter of an Al–Cu alloy with 3.7 wt.% Cu were cast and annealed for 100 h at 813 K to make a homogeneous, coarse-grained sample. After homogenization annealing, the sample diameter was reduced to 4 mm. The purpose was, on the one hand, to generate a defined surface of the sample and, on the other hand, to allow for uniform heating of the sample in a middle frequency induction coil. The penetration depth  $\delta$  of the induction field according to Ref. [13] is

$$\delta = \frac{1}{2\pi} \sqrt{\frac{\rho}{f \cdot \mu}} \approx 0.6 \text{ mm} \quad (2)$$

where  $\rho$  is the specific resistance ( $\rho(\text{Al}) = 2.7 \times 10^{-7} \text{ V m A}^{-1}$ ),  $\mu$  is the magnetic permeability ( $\mu = \mu_r \cdot \mu_0$ ;  $\mu_r(\text{Al}) \approx 1$ , thus  $\mu \approx \mu_0 = 4\pi \times 10^{-7} \text{ V s (A m)}^{-1}$ ), and  $f$  is the frequency of the induction furnace ( $f = 16 \text{ kHz}$ ).

Precipitates that possibly formed during previous processing steps (which were conducted at room temperature) were redissolved during a second annealing for 1 h at 813 K. To prevent further precipitate formation, the samples were then stored in liquid nitrogen and retrieved shortly before the annealing in the temperature gradient.

By cooling one end and heating the other end of the samples in the coil of a middle frequency induction furnace (see Fig. 1a) for a few seconds, the samples were exposed to a temperature gradient. During annealing, the temperature gradient was measured using an IR 512 pixel line camera,

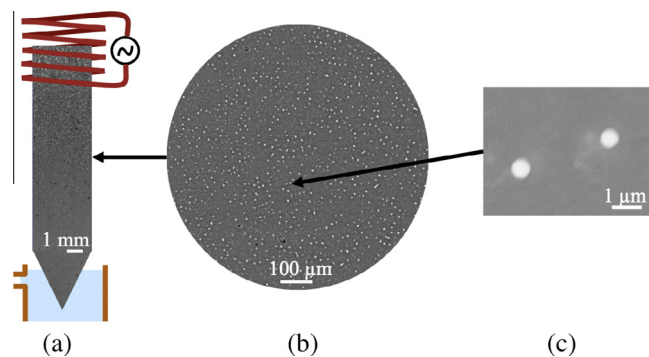


Fig. 1. (a) Experimental setup for the annealing in the temperature gradient for a short time interval; (b) part of a cross section of the sample at a given position with a known maximum temperature; (c) secondary single  $\theta$ -phase particles in the cross section.

Download English Version:

<https://daneshyari.com/en/article/7881820>

Download Persian Version:

<https://daneshyari.com/article/7881820>

[Daneshyari.com](https://daneshyari.com)