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SANS investigations on the solidification of aluminum alloys

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Abstract

Small-angle neutron scattering (SANS) measurements have been carried out on pure Al and on Al–0.3Ti–0.02B (wt%) alloy during solidification with different cooling rates. The experimental data have been fitted to the Porod law ($d\Sigma/dQ(Q) = K_p Q^{-4}$), where the increase in Porod constant K_p during solidification is expected to be proportional to the specific surface S_V of the grain boundaries. For increasing cooling rates the specific surface of the grain boundaries is found to increase for both samples, indicating a decrease in grain size. The presence of TiB₂ grain refiners in the Al–0.3Ti–0.02B alloy further reduces the grain size compared to pure aluminum at a given cooling rate.

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1. Introduction

The process of crystal nucleation and growth in liquid aluminum alloys during solidification plays an important role in the mechanical properties of the solid phase. These mechanical properties can be significantly improved by a decrease in the average grain size. This grain refinement is generally achieved by the addition of micron-size TiB₂ particles to the melt, which act as a substrate for heterogeneous nucleation of solid aluminum grains upon solidification. The average grain size can be reduced further by excess titanium in the

melt, leading to a reduced grain-growth velocity [1–4]. In this paper, we present small-angle neutron scattering (SANS) experiments for both the pure aluminum and aluminum containing grain refiners with composition Al–0.3Ti–0.02B (wt%) to estimate the grain size during solidification for different cooling rates. The advantage of SANS is that a relatively large sample volume can be studied in situ.

2. Experimental

The samples used in this study were 99.999% pure aluminum (Goodfellows) and an Al–0.3Ti–0.02B (wt%) alloy prepared from an Al–5Ti–0.2B

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(wt%) commercial master alloy (KBM AF-FILIPS). The particle size distribution of TiB_2 precipitates in the Al–0.3Ti–0.02B alloy was determined by optical microscopy and showed a particle size distribution in the range from 0.6 to $2.2\ \mu\text{m}$ with a maximum around $1.2\ \mu\text{m}$ and the number density $3 \times 10^8\ \text{cm}^{-3}$. The samples were placed in a cylindrical single-crystalline sapphire container with a height of 22 mm, an inner diameter of 10 mm, and a wall thickness of 1 mm. The in situ SANS measurements with fixed neutron wavelength of $\lambda = 10.0\ \text{\AA}$ ($\Delta\lambda/\lambda = 10\%$) were performed on D11 at the Institute Laue-Langevin.

In order to achieve a higher temperature stability ($\Delta T < 100\ \text{mK}$) for our solidification experiments, a furnace insert consisting of a nickel cylinder with a mass of 252 g was placed around the sample in a standard radiation furnace under vacuum.

3. Results and discussion

The influence of grain refiners on the solidification behavior of aluminum was studied by systematic time-dependent SANS measurements on pure aluminum and an Al–0.3Ti–0.02B alloy. For each of the measurements the sample was heated to a temperature of 943 K for 1 h to obtain a homogeneous liquid phase, followed by a continuous cooling, with rates ranging from 0.3 to 14 K/min. During the continuous cooling, the scattered intensity was continuously monitored in time steps of 30 s. During the liquid to solid phase transformation the scattered intensity gradually increased, but depended on the cooling rate and sample composition.

In Fig. 1 the macroscopic differential scattering cross-section $(d\Sigma/d\Omega)(Q)$ of the liquid Al–0.3Ti–0.02B sample as a function of wave-vector transfer, Q , is compared to that of the sample solidified at a cooling rate of 0.3 K/min. The scattering intensity in Fig. 1 has been corrected for the scattering of the empty sapphire container. The wave-vector transfer $Q = (4\pi/\lambda) \sin(\theta)$ is directly related to the scattering angle 2θ . The relatively strong scattering observed in the liquid state is

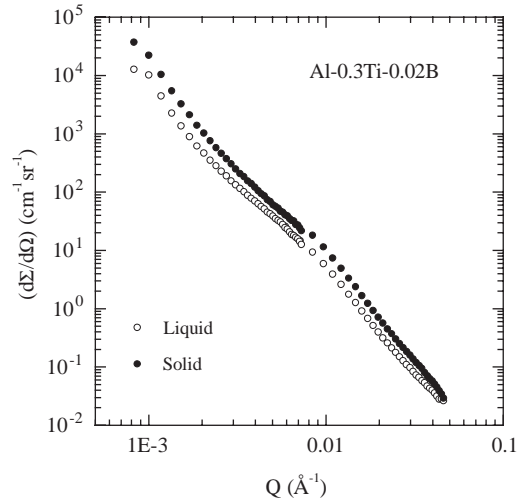


Fig. 1. Macroscopic differential scattering cross-section $(d\Sigma/d\Omega)(Q)$ as a function of wave vector transfer Q of the liquid (open circles) and solid (solid circles) Al–0.3Ti–0.02B alloy for a cooling rate of 0.3 K/min.

expected to originate predominantly from the TiB_2 particles in the melt. A weak increase in intensity is observed in the solid state compared to the liquid state. The observed change in intensity due to grain boundaries, evolved during solidification can be described by an additional contribution to the measured intensity $\Delta(d\Sigma/d\Omega)(Q)$ such that

$$\Delta\left(\frac{d\Sigma}{d\Omega}\right)(Q) = \left(\frac{d\Sigma}{d\Omega}\right)_{\text{solid}}(Q) - \frac{(\rho_{\text{solid}} - \rho_{\text{TiB}_2})^2}{(\rho_{\text{liquid}} - \rho_{\text{TiB}_2})^2} \left(\frac{d\Sigma}{d\Omega}\right)_{\text{liquid}}(Q), \quad (1)$$

where $(\rho_{\text{solid}} - \rho_{\text{TiB}_2})^2 / (\rho_{\text{liquid}} - \rho_{\text{TiB}_2})^2$ accounts for the relative change in the scattered intensity caused by the variation in contrast between scattering length density of the aluminum matrix and the TiB_2 particles as the aluminum density changes during solidification in the Al–0.3Ti–0.02B alloy. When the structural inhomogeneities in the material are relatively large ($QR \gg 1$), the scattering cross-section of the sample can be described by Porod law [5]

$$\Delta\left(\frac{d\Sigma}{d\Omega}\right)(Q) = K_p Q^{-4} + B, \quad (2)$$

where K_p is the Porod constant and B is the (incoherent) background. In the following, we have assumed that B is negligible in the Q range of interest.

The difference in scattering cross-section $\Delta(d\Sigma/d\Omega)(Q)$ between the solid and the liquid state for pure aluminum and that of Al–0.3Ti–0.02B, is studied as a function of Q and for different cooling rates. For increasing cooling rate, a significant increase in scattered intensity is observed which can be analyzed in terms of a fit to the Porod law. This difference in scattering cross-section as a function of cooling rate and the corresponding Porod fit for Al–0.3Ti–0.02B is shown in Fig. 2. The calculated Porod constants both for pure aluminum and Al–0.3Ti–0.02B are shown in Fig. 3 (a), as a function of cooling rate. The derived Porod constant was found to increase for increasing cooling rates. The Porod constant is defined as $K_p = 2\pi(\Delta\rho)^2 S_v$ where S_v is the specific surface area of grain boundaries and $\Delta\rho$ is the contrast in scattering length density between the grain boundary and the solid aluminum grain. For the difference in scattering between the solid and the liquid phase, K_p is expected to be proportional to the specific surface of the grain boundaries and describes the decrease in grain size with increasing cooling rate. The solid lines are a fit to the Porod

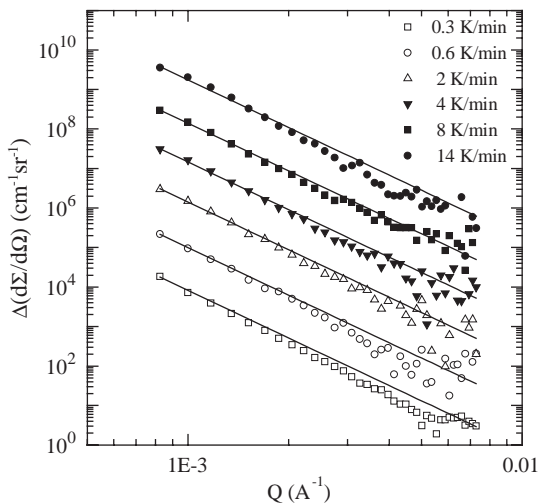


Fig. 2. Difference in macroscopic differential scattering cross-section $\Delta(d\Sigma/d\Omega)(Q)$ between the liquid and the solid phase of Al–0.3Ti–0.02B alloy for different cooling rates.

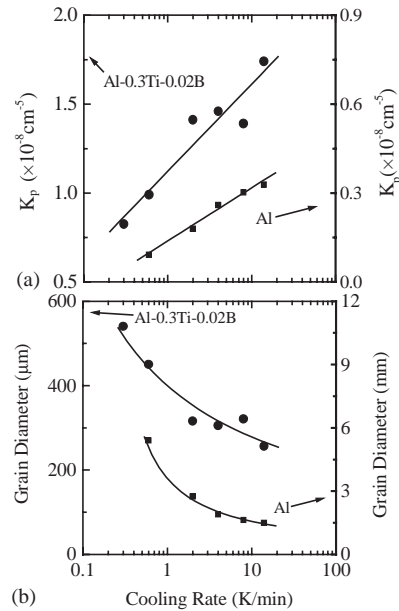


Fig. 3. (a) Calculated Porod constant K_p for Al–0.3Ti–0.02B alloy (circle) and pure aluminum (square) and (b) the corresponding grain diameter as a function of cooling rate. The size of the symbol is equal to the size of error bar.

law. For clarity, the curves for higher cooling rates are multiplied by 10, compared to immediate lower cooling rate. Assuming the solid aluminum grains to be spherical with radius R , $S_v = 3/R$. Due to uncertainty in $\Delta\rho$ the microscopic grain structure for the last measured solid aluminum and Al–0.3Ti–0.02B sample, was also investigated by optical microscopy. The average grain size of pure aluminum for a cooling rate of 14 K/min is about 2 mm and that of Al–0.3Ti–0.02B, measured for a cooling rate of 0.3 K/min is about 460 μm . By combining the SANS data on the variation in Porod constant as a function of cooling rate with the metallographic measurements, the average grain size of solid aluminum and Al–0.3Ti–0.02B alloy as a function of cooling rate is determined, as shown in Fig. 3(b). The fits to the experimental data yield information about the change in average grain size in the solid aluminum, with and without grain refiners and as a function of cooling rate. The time resolved small-angle neutron scattering measurements were also carried out during the liquid to solid phase transformation. The results of these measurements will be presented in future.

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References

- [1] M. Easton, D. Stjohn, *Met. Mater. Trans. A* 30 (1999) 1613.
- [2] M. Easton, D. Stjohn, *Met. Mater. Trans. A* 30 (1999) 1625.
- [3] N. Iqbal, N.H. van Dijk, V.W.J. Verhoeven, T. Hansen, L. Katgerman, G.J. Kearley, *Proceedings of the 6th International ESAFORM Conference On Material Forming, European Scientific Association for Material Forming, 2003*, p. 411.
- [4] N. Iqbal, N.H. van Dijk, V.W.J. Verhoeven, T. Hansen, W. Montfrooij, L. Katgerman, G.J. Kearley, *Acta. Mater.* 51 (2003) 4497.
- [5] G. Porod, *Z. Kolloid.* 124 (1951) 83.