Acta Materialia 103 (2016) 184–191

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Dendritic growth velocities in an undercooled melt of pure nickel under static magnetic fields: A test of theory with convection

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ARTICLE INFO

Article history:
Received 25 June 2015
Received in revised form 6 October 2015
Accepted 9 October 2015
Available online 6 November 2015

Keywords:
Dendritic solidification
Growth kinetics
Undercooling
Static magnetic field
Theory

ABSTRACT

Dendritic growth velocities in an undercooled melt of pure nickel under static magnetic fields up to 6 T were measured using a high-speed camera. The growth velocities for undercoolings below 120 K are depressed under low magnetic fields, but are recovered progressively under high magnetic fields. This retrograde behavior arises from two competing kinds of magnetohydrodynamics in the melt and becomes indistinguishable for higher undercoolings. The measured data is used for testing of a recent theory of dendritic growth with convection. A reasonable agreement is attained by assuming magnetic field-dependent flow velocities. As is shown, the theory can also account for previous data of dendritic growth kinetics in pure succinonitrile under normal gravity and microgravity conditions. These tests demonstrate the efficiency of the theory which provides a realistic description of dendritic growth kinetics of pure substances with convection.

1. Introduction

Dendritic growth of crystals occurs in nature and metallurgy. Many studies were devoted to an understanding of the growth kinetics and pattern selection issues of this process. A number of theories were proposed and compared with experimental observations. The currently accepted theories [1,2] include Ivantsov's solution to the heat and mass transport issue at a needle-like dendritic tip and a microsolvability analysis for unambiguous selection of a tip radius at a given undercooling in terms of the anisotropy of crystal-melt interfacial energy. A third accepted theory is a linear approximation of non-equilibrium thermodynamics which becomes evident at high undercoolings and depends on the anisotropy of interface attachment kinetics [3]. Such theories are often combined and referred to as an assembled theory in literature. The LKT/BCT theory [4–6] is one such example and represents a full combination of the three theories. Although some studies concluded a general agreement between the LKT/BCT theory and experiment [7–9], a discrepancy remains in the low undercooling region. The measured dendritic growth velocities in pure substances were found to deviate from the predictions of the LKT/BCT theory significantly [10]. This is also true for dendritic tip radii measured in a transparent substance [11]. A microgravity experiment [12,13] and phase field modeling [14–17] suggested that the discrepancy is likely to arise from an interaction of dendritic growth with fluid flow in an undercooled bulk melt. Thus, theoretical efforts have been made to bridge this discrepancy by the introduction of a rigorous treatment of the fluid flow effect on dendritic growth.

An early attempt was made by Boussiou and Pelce [18]. They extended Ivantsov's theory by applying the Navier–Stokes equation to a tilted flow in a pure substance. They proposed a solution to the flow-modified heat transport issue in two-dimensional dendritic growth. They also worked out the microsolvability condition at low growth Peclet numbers and presented a selection criterion for the dendritic tip that relies on the anisotropy of the crystal-melt interfacial energy and on a longitudinal component of the tilted flow as well. Recently, Alexandrov and Galenko [19] extended this treatment to the dendrite tip growing at arbitrary growth Peclet numbers in a pure substance or a dilute alloy system. The selection criterion for the dendritic tip is dependent on the growth Peclet number. On the basis of this advancement, they proposed a combined three-dimensional theory for dendritic growth with convection [20]. For convenience, this new theory is termed as the Alexandrov–Galenko theory (AG theory for short). A preliminary test of the AG theory

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http://dx.doi.org/10.1016/j.actamat.2015.10.014
1359-6454/

has already been conducted using a two-dimensional formalism [20]. It was shown that the AG theory can provide a satisfactory explanation of the dendritic growth velocities observed in containerlessly undercooled intermetallic compounds in the presence of a convective flow in a bulk volume [20,21]. However, phase field modeling suggested that the influence of melt convection on a three-dimensional tip is more pronounced than on a two-dimensional tip [17,22]. Thus, it is of great interest to perform a test of the three-dimensional AG theory using data measured under conditions of controllable melt convection.

In this paper, we report measurements of dendritic growth velocities of pure nickel under static magnetic fields for a test of the three-dimensional AG theory [20]. The utilization of the static magnetic fields allowed us to tune forced convection in an undercooled melt continuously, thus providing much freedom for the test. Then we present a complementary test of the AG theory using literature data of dendritic growth velocities and dendritic tip radii of pure succinonitrile measured under normal gravity and microgravity conditions [13]. Finally, we compare the AG theory with phase field modeling of dendritic growth at high growth Peclet numbers [23].

2. Experimental details

The measurements were performed on a single glass-fluxed sample of pure nickel. The sample had a purity of 99.99% and a mass of about 1 g. A radio-frequency induction furnace was combined with a superconducting magnet to melt and solidify the sample under static magnetic fields up to $B = 6$ T. The sample was supported by a pan-like ceramic holder containing a small amount of soda lime glass, and was fixed between two opposite windings of a heating coil of the furnace. The vacuum chamber of the furnace was pumped to a vacuum pressure of 5.7×10^{-3} Pa and was backfilled with argon of 99.999% purity to a pressure of 5×10^4 Pa. The sample was inductively heated, melted, and overheated under the protection of the argon atmosphere. Then, the heating power to the coil was reduced to about 14% of the initial power. The sample was cooled and solidified spontaneously. In solidification, crystal nucleation occurred preferentially on the lower surface of the sample, which was in intimate contact with the fluxed glass. A recalescence process followed due to instantaneously released latent heat. Under each of the static magnetic fields ranging between $B = 0$ T and $B = 6$ T, the melting-solidification cycle was repeated 20 to 30 times to produce a wide spectrum of undercooling. The surface temperature of the sample was measured using a single-color pyrometer with an accuracy of ±6 K at a sampling rate of 100 Hz. Meanwhile, the recalescence process was monitored using a high-speed video camera at a frame rate of 87,600 fps. The video images were analyzed using an executable program running in the environment of the commercial software Matlab to determine the speed of an advancing recalescence front. The recalescence front was assumed to travel like a spherical wave starting from the nucleation site and advancing towards the other side of the sample surface at a constant speed. With the aid of the program, the location of the nucleation site on the sample surface was determined first. Then, the traveling distance of the recalescence front away from the nucleation site was determined as a function of time by analyzing the traces of the recalescence front. A linear law was fitted to the distance versus time relationship, and the slope of the linear law was taken as the traveling speed of the recalescence front. The speed of the recalescence event gave a good approximation of dendritic growth velocities in the undercooled sample due to a thermal diffusion distance shorter than a one-dimensional resolution of 100 μm of each pixel of the video images. The algorithm used in the program was exactly the same as that established by Binder using a free software, POV Ray 3.6, for the same purpose [24] but with an improved efficiency in determining the speed.

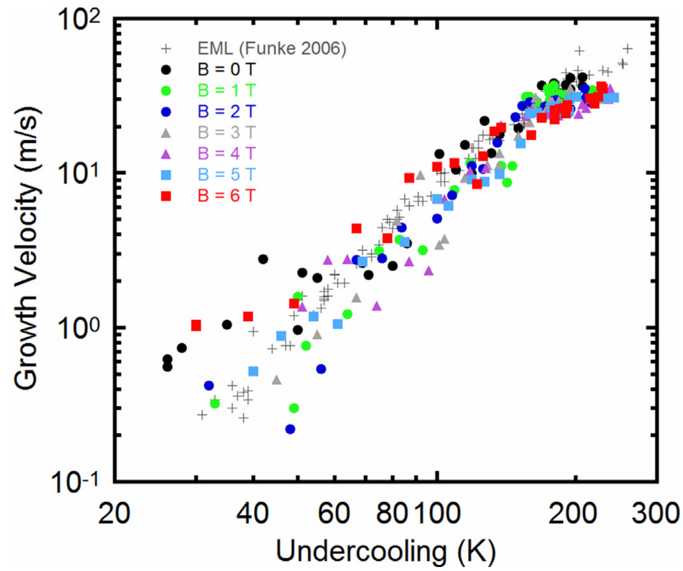

Fig. 1. Measured dendritic growth velocities of pure nickel as a function of undercooling under static magnetic fields between $B = 0$ T and $B = 6$ T. The relative errors of individual measurements are less than 10%. Error bars of data points are not shown for clarity. Previous data measured by Funke et al. [10] in electromagnetically levitated samples of higher purity (99.999%) is also shown for comparison.

3. Results and discussion

3.1. Dendritic growth velocities of pure nickel under forced convection conditions

The measured dendritic growth velocities are plotted in Fig. 1 as a function of undercooling, ΔT. A power law is observed over a wide range of undercooling for each static magnetic field. The growth kinetics appears to be decelerated abruptly at a critical undercooling of $\Delta T_{crit} = 165$ K, as evidenced by a negative deviation from the power law.[1] Such observations are in good agreement with the latest measurements on electromagnetically levitated nickel samples of higher purity [10]. However, there are distinct differences in the magnitude of the measured dendritic growth velocities. As displayed in Fig. 1, the present data for undercoolings below ΔT_{crit} shows a positive or a negative deviation depending on the static magnetic fields. These deviations are related to forced convection in the undercooled sample as explained below. Another deviation is observed for undercoolings above ΔT_{crit}. It is negative and independent of the magnetic fields. It is assumed to arise from the higher impurity concentration of the present sample because a reduced deviation was observed by one of the present authors using a purer material [28].

Forced convection is very common in inductively heated metallic melts and is active in the present sample. Unlike the electromagnetically levitated samples, the present sample was fixed on the sample holder. The forced convection inside it was expected to be weaker than in the electromagnetically levitated samples as suggested by Battersby et al. [26]. But, as seen in Fig. 1, the present data measured with no static magnetic fields shows higher growth velocities for undercoolings below 60 K than those

[1] A similar deviation of dendritic growth kinetics from a power law was also observed for dilute Ni–B and Cu–O alloys [25,26] and can be interpreted by considering local non-equilibrium at a rapidly advancing crystal/liquid interface [27].