

MEASUREMENTS OF DENDRITIC GROWTH VELOCITIES IN UNDERCOOLED MELTS OF PURE NICKEL UNDER STATIC MAGNETIC FIELDS

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Abstract

Dendritic growth velocities in undercooled melts of pure Ni have been intensively studied over the past fifty years. However, the literature data are at marked variance with the prediction of the widely accepted model for rapid dendritic growth both at small and at large undercoolings. In the present work, bulk melts of pure Ni samples of high purity were undercooled by glass fluxing treatment under a static magnetic field. The recalescence processes of the samples at different undercoolings were recorded using a high-speed camera, and were modeled using a software to determine the dendritic growth velocities. The present data confirmed the effect of melt flow on dendritic growth velocities at undercoolings below 100 K. A comparison of the present data with previous measurements on a lower purity material suggested an effect of impurities on dendritic growth velocities at undercoolings larger than 200 K as well.

Introduction

Many studies have been carried out to measure dendritic growth velocities in undercooled melts for an understanding of the mechanism of dendritic growth [1]. Pure nickel has attracted constant attention over the past fifty years [2-16]. The main reason is because its materials properties both in the liquid and in the solid states are well known so that the measured growth velocities can be referred to for a rigorous test of physical models for rapid dendritic growth. A review of previous measurements on pure nickel could be found elsewhere [12,16]. The latest studies [9,15,16] showed reduced scatter as well as an improved agreement with the widely accepted dendritic growth model (known as LKT model [17]) owing to the use of a series of advanced techniques including those for undercooling and those for in-situ observations. However, the measured data were yet at marked variance with the predictions of the LKT model. A positive and a negative deviations were observed for the small (below 100 K) and for the large undercooling regions (above 200 K), respectively [9,15]. Efforts were already made to account for these discrepancies. Eckler et al. [8,9] first proposed two alternative explanations for the positive deviation: impurity and fluid flow effects. Galenko et al. [18] extended the LKT model by taking into account the contribution of fluid flow and showed a reduction of the discrepancy. Herlach and Galenko [19] further incorporated the impurity effect and explained the residual discrepancy. Using a phase field method, Bragard et al. [20] and Nestler et al. [21] explored effects of capillary and kinetic anisotropy on dendritic growth to understand the deviation in the large undercooling region. The former group suggested that the deviation might also be related to fluid flow or impurities in liquid samples. Thus, it is of great interest to determine the effects of fluid flow and impurities on dendritic growth velocities in undercooled Ni samples experimentally.

A fluid flow effect on dendritic growth velocities has been observed for electromagnetically levitated samples of few intermetallic compounds under reduced gravity conditions [22-23]. However, no experimental data for pure nickel samples processed under similar experimental conditions are available. Recently, a few studies [24-26] have shown that a static magnetic field of few Tesla is able to damp convective flow in electromagnetically levitated samples under terrestrial conditions. Such advancement opens a new opportunity for investigations of fluid flow effects on dendritic growth velocities in undercooled metals. Very recently, the present authors designed a novel glass-fluxing facility, which combined an inducting heating furnace with a superconducting magnet. A preliminary study on nickel samples of 99.99% purity (referred to 4N Ni below) showed evidence for a detectable effect of fluid flow on dendritic growth velocities in the small undercooling region [16]. In the present work, the measurements were extended to samples of higher purity, 99.999% purity (referred to 5N Ni below) to investigate the effects of fluid flow and impurities simultaneously.

Experimental Details

The experimental facility used for measurements of dendritic growth velocities comprised a vacuum induction melting furnace, a superconducting magnet, a shielding tube, an Ultima-APX type digital high-speed camera (HSC), a METIS MS09 type single-color pyrometer, and two computers. The details of the facility can be found elsewhere [16]. In the present measurements, individual samples of 1.0 g were cut from a 5N purity Ni rod of 5 mm in diameter offered by Alfa Aesar. Under each condition, the measurements were made on one sample only. The cut samples were polished using an abrasive paper and cleaned in an ultrasonic bath for few minutes. Before melting, a single sample was placed onto an alumina crucible containing soda lime glass lumps. The crucible was positioned in a heating coil. After evacuation to a vacuum pressure of 5×10^{-3} Pa, the chamber of the facility was back-filled with argon of 99.999% purity to a pressure of 5×10^4 Pa. The superconducting magnet was ramped to a magnetic field of 1.0 T. Then, the sample was heated and melted inductively. The sample temperature was measured using a single-color pyrometer at a frequency of about 100 Hz. The glass powders were fused by conducted heat. The sample was overheated and soaked for a few minutes. After soaking, the power to the coil was switched off. The sample was cooled and solidified spontaneously. A recalescence event took place during solidification of the undercooled sample due to rapid release of latent heat. For measurements of dendritic growth velocities, the recalescing sample surface was in-situ observed using a high-speed camera with a resolution of 256×32 pixels at a frame rate of 87600 fps. A pixel corresponded to an area of $100 \mu\text{m} \times 100 \mu\text{m}$. The sample was subjected to more than 40 melting-solidification cycles. In order to determine the nucleation site and therefore the growth velocity of each recalescence process, the recorded images of the sample surface was modeled using computer software named POV Ray 3.7 [23]. The recalescence front was assumed to travel through the bulk volume of the undercooled sample at a constant rate like a spherical wave. Thus, the three-dimensional recalescence front traveling through the sample volume could be reconstructed by a continuous analysis of the recorded loci of the recalescence front sweeping across the sample surface with the aid of the software. The dendritic growth velocity was calculated by dividing the total traveling distance of the three-dimensional recalescence front by the consumed time in a given recalescence process.

Results and Discussion

Figure 1 illustrates cooling curves of a Ni sample of 5N purity. The recalescence temperature, i.e., the temperature reached at the end of recalescence, often showed a deficit with respect to a

well-determined melting temperature of 1728 K. The deficit generally increases with increasing undercooling. A similar phenomenon was observed by the present authors for the Ni sample of 4N purity [16] and by other researchers for glass-fluxed Ni-Cu single-phase alloys [27] and Ni-Sn eutectic alloys [28]. The most probable reason for the deficit is a thermal loss of the sample to the crucible through the molten glass during recalescence. Other reasons include creation of numerous defects and stresses in the solid phase, which might reduce the energy difference between the solid and the liquid phases during recalescence [27]. Because of this deficit, the measured sample temperature was calibrated with respect to a maximum recalescence temperature derived from a statistical analysis of all data.

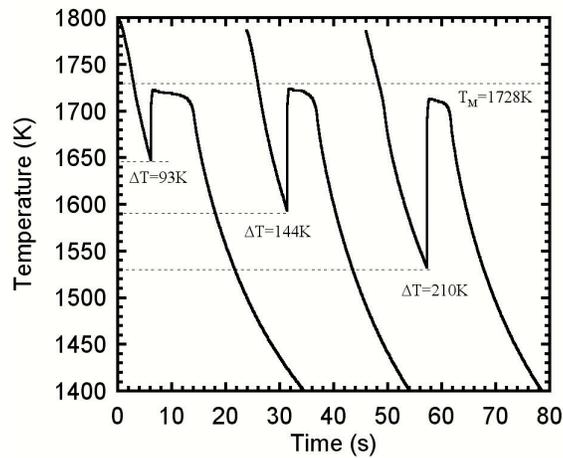


Figure 1 Illustration of cooling curves of 5N Ni sample at different undercoolings

Figure 2 illustrates the corresponding sequential images of the top surface of the recalescing sample recorded by the high-speed camera. In agreement with many previous studies (e.g. Refs. [12] and [15]), the observed recalescence front shows an angular morphology and a smooth morphology for small and large undercoolings, respectively. The transition does not correspond to a sharp undercooling, but occurs over a span of 15 K. The imposition of the static magnetic field did not produce any visible changes either in the morphology or in the transition. It is

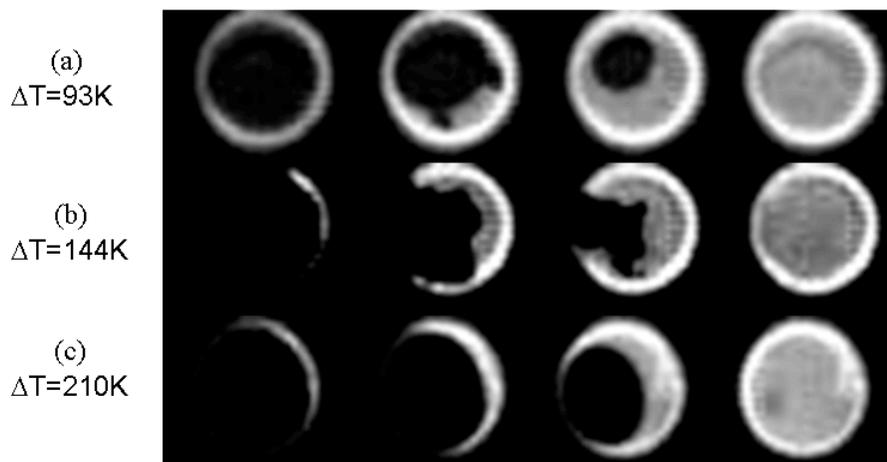


Figure 2 Illustration of HSC images of the 5N Ni samples at different undercoolings

worthy of mentioning that the irregular morphology of the recalescence front for small undercoolings brought about a difficulty in modeling of the recalescence process. However, this difficulty could be overcome by modeling the major contour of the recalescence front instead of the whole front. In this sense, the derived traveling distance of the recalescence front represented a mean value, rather than a specific value for local regions.

Figure 3 shows the measured dendritic growth velocities of the 5N Ni samples as a function of undercooling. As seen in Figure 3a, the data measured under the static magnetic field show a reduced scatter with respect to those measured without the static magnetic field. Apart from this aspect, it is hardly to see a difference between the two sets of the data in the normal-scale plot. In agreement with many previous studies, the present data show a power-law behavior and a linear law behavior for small and large undercoolings, respectively. Such a transition of the growth velocities, however, is sharper than the morphological transition of the recalescence front is. The former occurs at a well-defined undercooling of 200 K. If compared with the previous data for the 4N purity samples [16], one can find a striking difference in the large undercooling region. The velocities measured on the 5N purity samples are considerably higher than those measured on the 4N purity sample. Such a difference highlights that the effect of unknown impurities on growth velocities of undercooled nickel melts is tremendous. In the following, attention is focused onto the data measured at small and medium undercoolings.

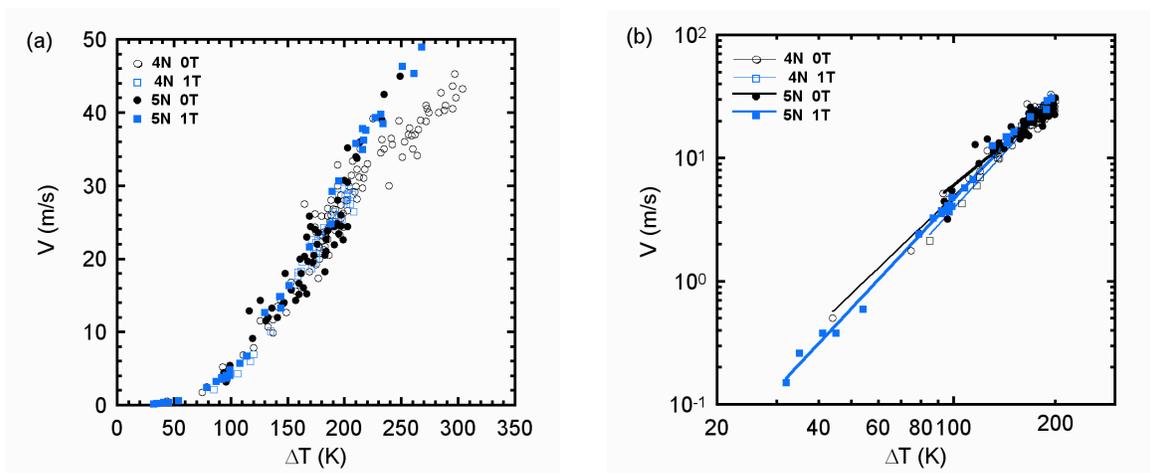


Figure 3 Growth velocity – undercooling relationship measured on the 5N Ni samples with and without imposition of a static magnetic field of 1 T. For comparison, two sets of the previous data measured on 4N purity samples [16] are also shown.

Due to relatively small values in magnitude, the differences of the growth velocities in the small undercooling region cannot be readily distinguished on a normal-scale plot. For this reason, the data are re-plotted on a dual-logarithm scale. However, the data are too few to allow us to directly extract the effect of impurities and residual fluid flow on the growth velocities in the small undercooling region of high interest. Thus, the data are all truncated at 200 K and fit to different power laws. As seen in Fig. 3b, each power law is represented by a straight line. Two clear tendencies are shown by these lines. One is that the imposition of the static magnetic field decreases the slope of the power law fitting line independent of the sample purity. The other is that the improvement of sample purity increases the slope of the power law fitting line under identical magnetic field conditions. The first tendency means that damping of fluid flow in the undercooled samples lowers the dendritic growth velocities. Reversely, the introduction of fluid flow increases the dendritic growth velocities as expected. Such a tendency agrees reasonably

with the prediction of the extended LKT model [18]. The second tendency means that the reduction of impurities increases the dendritic growth velocities. Reversely, the presence of the impurities lowers the dendritic growth velocities. This tendency, however, is in contrast to the theoretical prediction for a strongly partitioning impurity [8,19]. Probably the species or the amount of the impurity is different from that assumed in the previous studies [8,19]. Consequently, the reversed effect of the impurities occurs to the present samples.

It is also noted in Figure 3b that two power law fitting lines for the samples of different purities intersect at an undercooling of 168 K in the absence of the static magnetic field. However, the other lines never intersect. This difference between the fitting lines implies that the lower purity sample can reach a growth velocity comparable to that of the higher purity sample with the aid of fluid flow, but never can reach the same velocity without the aid of fluid flow. For the higher purity 5N sample, the two power law fitting lines intersect at an undercooling of 145 K. In contrast, the two power law fitting lines of the lower purity 4N sample intersect at an undercooling of 405 K, where the power laws do not apply any longer. Such a difference suggests that the fluid flow effect becomes negligible at a medium undercooling for the higher purity 5N sample, but does not vanish for the lower purity 4N samples for all accessible undercoolings.

In order to produce more insight into the effects of impurities and fluid flow, dendritic growth velocities of the Ni samples are calculated for five different undercoolings in terms of the above power law fittings. Relative changes of the growth velocities are determined. The results are listed in Table 1. At an undercooling as small as 10 K, the imposition of the static magnetic field decreases the growth velocities by 76% and by 85% for the 4N and 5N purity samples, respectively. With increasing undercooling, the effect of the static magnetic field on the 4N purity sample shows a continuous decrease. For the 5N purity sample, the effect of the static magnetic field decreases faster and reaches a minimum at an undercooling of 150 K. After that, the effect resumes but changes its direction. On the other hand, the impurities produce the strongest effect on the growth velocities at small undercoolings. In the absence of the static magnetic field, the effect of the impurities decreases quickly with increasing undercooling and changes its direction at 200 K. Under the action of the static magnetic field, the effect of impurities decreases slightly with increasing undercooling. Such different behavior highlights that the impurities interact strongly with fluid flow during dendritic growth at small undercoolings. In many of the previous studies, these two quantities were not controlled strictly. Thus, it is not surprising that a large discrepancy had arisen between the measured growth velocities and the prediction of the LKT model. A detailed analysis of the present data within the framework of the LKT model is under way. In the following, the present data measured under the static magnetic field are further discussed with respect to two recent studies and one phase field modeling.

Table 1 Relative changes of dendritic growth velocities of pure nickel in response to altering of the experimental conditions

| Sample conditions | | Relative change of growth velocity (%) | | | | |
|-------------------|----|--|------|-------|-------|-------|
| | | 10 K | 50 K | 100 K | 150 K | 200 K |
| 0T→1T | 4N | -76 | -55 | -42 | -32 | -24 |
| | 5N | -85 | -53 | -23 | 2 | 26 |
| 5N→4N | 0T | -67 | -38 | -19 | -5 | 7 |
| | 1T | -47 | -41 | -38 | -37 | -35 |

As shown in Figure 4a, the present data measured under the static magnetic field are in good agreement with the previous measurements by Lum et al. [12] for undercoolings larger than 180 K. However, there is a large discrepancy in the medium undercooling region. Since those researchers employed the same glass-fluxing method for undercooling, the convection in their samples is assumed to be comparable to that in the present samples. Under this assumption, the large discrepancy may be ascribed to a difference in the method for calibration of the temperature measurements. Actually, if the deficit of the recalescence temperature of the present samples with respect to the melting temperature was eliminated deliberately, the derived growth velocity-undercooling relationship would show an excellent agreement with those researcher's data over the whole undercooling regime. However, such calibration may not be necessary. Note that the present data coincide with the lower limits of the latest data measured by Funke et al. [15] for electromagnetically levitated samples. As concluded from the above discussion, residual fluid flow in the present samples can enhance growth velocities of pure nickel. Convective flow in the electromagnetically levitated samples is much stronger [24,29] and should make a larger contribution to the growth velocities than in the present samples. From this point of view, the present method for calibration of the sample temperature is reasonable. More importantly, the present data show an excellent agreement with the recent phase field modeling [21] up to an undercooling of 220 K (see Fig. 4b). The contributions of fluid flow and impurities were not incorporated in the modeling. Therefore, the conditions hypothesized therein were well satisfied through the present experiments under the static magnetic field. The discrepancy remaining at extremely large undercoolings can be ascribed to the impurity effect because it is smaller for the higher purity sample. It will vanish if the material purity can be improved further. In other words, the deviation of the growth velocities from a power law at large undercoolings is largely due to the impurities dissolved in the pure Ni samples.

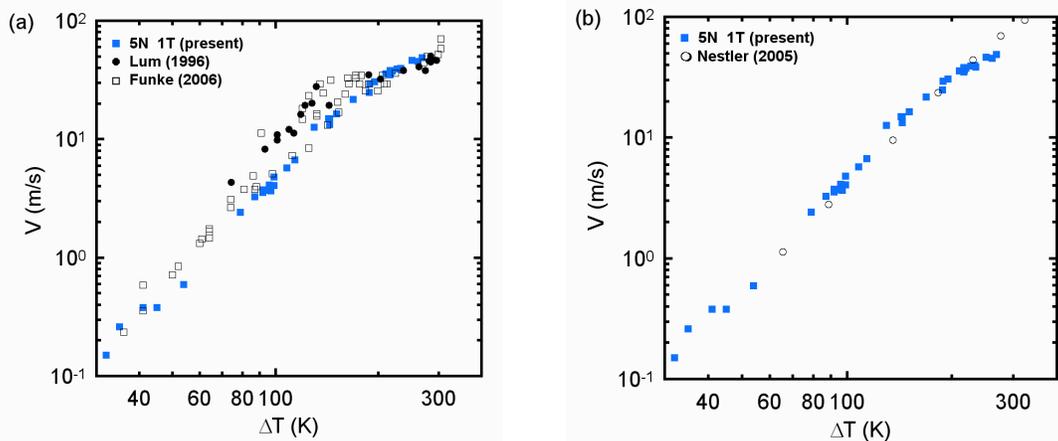


Figure 4 Comparison of the present data with two sets of literature data [12,15] (a) and the results of the recent phase-field modeling [21] (b). For clearance, the data measured in the present of the static magnetic field are shown only.

Conclusions

Dendritic growth velocities in undercooled melts of high purity nickel samples have been measured with and without the imposition of a static magnetic field of 1 T. Similar to the latest measurements on the samples of lower purity, the present data have confirmed the long-term suspected effect of fluid flow on growth velocities in the small undercooling region. The present data have also shown that the impurity effect plays a role both in the small and in the large

undercooling regions. Although the species of the impurities are unknown, they tend to lower the dendritic growth velocities under the present experimental conditions. Additionally, the present data measured under the static magnetic field show an excellent agreement with the prediction of the recent phase field modeling [21] up to an undercooling of 220 K. The remaining discrepancy at larger undercoolings has been attributed to the impurity effect.

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References

- [1] D. M. Herlach, P. Galenko and D. Holland-Moritz, *Metastable Solids from Undercooled Melts*, Pergamon Materials Science, Amsterdam, 2007.
- [2] J. L. Walker, in: *Principles of Solidification*, ed. B. Chalmers, Chapter 4, Wiley, New York, 1964.
- [3] G. A. Colligan and B. J. Bayles, *Acta Metall.* 10 (1962) 895.
- [4] E. Schleich, R. Willnecker, D. M. Herlach and G. P. Görlner, *Mater. Sci. Eng. A* 98 (1988) 39.
- [5] T. J. Piccone, Y. Wu, Y. Shiohara and M. C. Flemings, in: *Solidification Processing 1987*, The Institute of Metals, London, 1988, p. 268.
- [6] R. Willnecker, D. M. Herlach and B. Feuerbacher, *Phys. Rev. Lett.* 62 (1989) 2707.
- [7] W. H. Hofmeister, R. J. Bayuzick and M. B. Robinson, *Rev. Sci. Instrum.* 61 (1990) 2220.
- [8] K. Eckler, D. M. Herlach, R. G. Hamerton and A. L. Greer, *Mater. Sci. Eng. A* 133 (1991) 730.
- [9] K. Eckler and D. M. Herlach, *Mater. Sci. Eng. A* 178 (1994) 159.
- [10] B. T. Bassler, W. H. Hofmeister, G. Carro and R. J. Bayuzick, *Metall. Mater. Trans. A* 25 (1994) 1301.
- [11] S. Walder and P. L. Ryder, *Mater. Sci. Eng. A* 203 (1995) 197.
- [12] J. W. Lum, D. M. Matson and M. C. Flemings, *Metall. Mater. Trans. B* 27 (1996) 865.
- [13] B. T. Bassler, W. H. Hofmeister and R. J. Bayuzick, *Mater. Sci. Eng. A* 342 (2003) 80.
- [14] P. R. Algosio, W. H. Hofmeister and R. J. Bayuzick, *Scrip. Mater.* 51 (2004) 539.
- [15] O. Funke, G. Phanikumar, P. K. Galenko, L. Chernova, S. Reutzel, M. Kolbe and D. M. Herlach, *J. Cryst. Growth* 297 (2006) 211.
- [16] J. Gao, Z. N. Zhang, Y. K. Zhang and C. Yang, in: *Solidification of Containerless Undercooled Melts*, ed. D. M. Herlach and D. Matson, Chapter 13, Wiley, in press.
- [17] J. Lipton, W. Kurz and R. Trivedi, *Acta Mater.* 39 (1994) 49.
- [18] P. K. Galenko, O. Funke, J. Wang and D. M. Herlach, *Mater. Sci. Eng. A* 375-377 (2004) 488.
- [19] D. M. Herlach and P. K. Galenko, *Mater. Sci. Eng. A* 449-451 (2007) 34.
- [20] J. Bragard, A. Karma, Y. H. Lee and M. Plapp, *Interface Sci.* 10 (2002) 121.
- [21] B. Nestler, D. Danilov and P. Galenko, *J. Comput. Phys.* 207 (2005) 221.
- [22] S. Reutzel, H. Hartmann, P. K. Galenko, S. Schneider and D. M. Herlach, *Appl. Phys. Lett.* 91 (2007) 41913.
- [23] S. Binder, Ph.D. Thesis of Ruhr University of Bochum, 2010.
- [24] V. Bojarevics and K. Pericleous, *ISIJ Inter.* 43 (2003) 890.

- [25] H. Yasuda, I. Ohnaka, Y. Ninomiya, R. Ishii, S. Fujita and K. Kishio, *J. Cryst. Growth* 260 (2004) 475.
- [26] Y. K. Zhang, J. Gao, D. Nagamatsu, T. Fukuda, H. Yasuda, M. Kolbe and J. C. He, *Scrip. Mater.* 59 (2008) 1002.
- [27] W. Yang, F. Liu, H. F. Wang, Z. Chen, G. C. Yang and Y.H. Zhou, *J. Alloys Compd.* 470 (2009) L13.
- [28] J. F. Li, W. Q. Jie, S. Zhao and Y. H. Zhou, *Metall. Mater. Trans. A* 38 (2007) 1806.
- [29] R. W. Hyers, *Meas. Sci. Technol.* 16 (2005) 394.