Bent dendrite growth in undercooled Fe-B alloy melts

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Abstract. Dendritic growth is the main solidification mode in alloy casting. In order to control dendrite growth for materials design from the melt it is important to fully understand the influence of process conditions. This study stands as an experimental note observing bent dendrite growth in Fe-B alloys and suggesting possible explanations as induced by fluid flow, thermal, and concentational diffusion or impurities. Electromagnetic levitation technique (EML) is used for containerless processing of undercooled melts under 1g and reduced gravity conditions in parabolic flight. Further investigations are needed to find a suitable explanation for the observed bent dendrite growth behaviour.

1. Introduction

The dendritic structure formed during the solidification process determines to some extent the final microstructure and therefore can have significant influence on material properties. In alloy casting dendritic solidification is the main growth mode, for example in steel production. To design materials from the melt it is important to understand the influence of fluid flow, convectional effects, and impurities on the dendrite growth process.

Electromagnetic levitation technique (EML) is applied to undercool droplets of metallic melt accessible for in-situ diagnostics of the solidification processes. In order to verify models for dendritic growth in undercooled melts we measured growth velocities of pure Fe and Fe-B (1, 5, and 10 at.% B) alloys. In the case of cubic metals such as Fe, dendrite tips grow along the preferred $<100>$ direction [1]. During our investigations on the Fe-B binary system, our measurements showed unexpected bent growing dendrites. This work shows empirically that at least the path of the dendrite tip can change during growth and deviates from a straight line.

2. Experimental

To achieve large undercoolings the use of high-purity raw materials is essential. Samples of Fe-1, 5, and 10 at.% B were prepared from commercial high-purity elements 6N Fe and 6N B purchased from ALFA AESAR. Components were alloyed in an arc melting furnace previously evacuated ($10^{-6}$ mbar) and subsequently filled with high purity Argon (6N) atmosphere (1 bar).

For in-situ studies of deeply undercooled Fe-B melts the electromagnetic levitation technique (EML) [3] is used as schematically shown in Figure 1. This containerless technique provides large undercoolings prior to solidification and allows direct observation of the solidification process. The transformation of the undercooled liquid phase into the solid phase leads to a visible contrast due to the release of latent heat during rapid solidification, which is recorded by a high-speed video camera.
The temperature is measured contactlessly by an infrared-pyrometer. High purity 6N He-gas is used to cool down the sample.

**Figure 1.** Schematic view of an electromagnetic levitation facility and levitated liquid sample with stream lines of fluid flow as indicated inside the melt [2].

### 3. Results

Investigations of Fe with 1, 5, and 10 at.% B alloys show bent dendrite growth up to certain undercoolings in ground-based 1g-EML. Figure 2 shows typical results for the trajectories of the growing dendrites for each composition. The dendrite growth velocities are in the order of $10^{-1}$ m/s, therefore in the same order of magnitude as the fluid flow velocities in 1g-EML (0.3 m/s) according to R.W. Hyers [4]. In Table 1 the undercooling regimes and velocity results are listed. Different types of bent patterns are observed for the three compositions. The trajectories of the dendrite tips show spiralling (Fe-1 at.% B), zigzagging (Fe-5 at.% B) and U-turn (Fe-10 at.% B) growing dendrites. This behaviour is reproducible and characteristic for each composition.

**Figure 2.** High-speed video images of electromagnetic levitated samples. The dark grey area is the undercooled liquid. The light grey region corresponds to the growing solid, which appears brighter due to the release of latent heat during rapid solidification. Arrows indicate the directions and trajectories of the growing dendrites showing straight (pure Fe), spiral like (Fe-1 at.% B), zigzagging (Fe-5 at.% B), and U-turn (Fe-10 at.% B) patterns in 1g-EML.
Table 1. Highest undercoolings $\Delta T$ and corresponding dendrite growth velocities $v$ for Fe-1, 5 and 10 at.% B where bent dendrite growth has been observed so far. Data for pure Fe is added for comparison.

|                | Undercooling $\Delta T$ [K] | Dendrite growth velocity $v$ [m/s] | Observed pattern   |
|----------------|-----------------------------|-----------------------------------|--------------------|
| Pure Fe        | 60                          | 0.6                               | Straight, no bending |
| Fe-1 at.% B    | 70                          | 0.2                               | Spiral growth       |
| Fe-5 at.% B    | 200                         | 0.3                               | Zigzagging          |
| Fe-10 at.% B   | 110                         | 0.05                              | U-turn              |
| TEMPUS Fe-1 at.% B | 100                     | 0.14                              | Zigzagging          |

3.1. Bent growing dendrites under reduced gravity conditions

In order to verify the influence of convection on the growth morphology described in this study, parabolic flight experiments were performed under reduced gravity conditions using the TEMPUS facility [5]. In $\mu$g-EML the fluid flow is about 0.05 m/s [4], one order of magnitude lower than in 1g-EML but still in the order of the slowest observed growth velocities. Figure 3 shows a bent growing dendrite under microgravity during parabolic flight. The undercooling $\Delta T$ is 98 K and the growth velocity $v$ is about 0.14 m/s.

![Figure 3](image1.png)

Figure 3. Snapshots of an undercooled solidifying Fe-1 at.% B liquid sample (dark grey) under reduced gravity conditions during parabolic flight showing bent dendrite growth (light grey).

![Figure 4](image2.png)

Figure 4. SEM and EBSD images of a cross section of Fe-10 at.% B solidified at an undercooling $\Delta T$ of 110K in 1g-EML showing a bent dendrite. The bending occurs mainly in the light blue region and at its transitions to the dark blue and green region, respectively.
3.2. Microstructure

Figure 4 shows the cross section microstructure of a bent dendrite with SEM and EBSD of an Fe-10 at.\% B sample. The sample was processed in 1g-EML and solidified at an undercooling $\Delta T$ of 110 K.

4. Discussion

The trajectories of the dendrite tips show spiralling, zigzagging, and U-turn patterns for growing dendrites. So far we did not observe bent growing dendrites in any other metallic materials we investigated except multicomponent steel alloy. Our findings appear to be relevant only for Fe-based alloys.

In general, the dendritic solidification is a competition between heat/solute diffusion and surface energy [1]. B is poorly soluble in Fe [6]. According to growth models, a solute pile up exists at the solid-liquid interface of the dendrite tip, which slows down the dendrite growth velocity. Additionally, fluid flow influences the thermal and solute gradients ahead of the solid-liquid interface. In particular, the fluid flow velocity inside the melt in 1g-EML is in the same order of magnitude as the growth velocity itself. Figure 1 shows schematically the calculated fluid flow loops inside a laminar liquid sample according to P. Galenko et al. [2]. There seems to be a weak correlation between the fluid flow loops inside the liquid sample and the trajectories of the observed bent growing dendrites. More advanced investigations are needed to confirm this. Even under reduced gravity conditions with weaker fluid flow velocity the bent dendrite growth occurs for Fe-1 at.% B.

The microstructure of solidified samples showed that the effect of bent dendrites seems to be mainly sample-surface dominated. Up to now we were unable to find significant bent dendrites in the bulk microstructure of the samples. This could be explained due to coarsening and fragmentation. The cross section microstructure of solidified sample (Figure 4) indicates a stepwise breaking of the dendrite. Different crystal orientations of the grains are represented in different colours. A bending occurs mainly in the middle part (light blue region) and at its transitions to the dark blue and green region.

Regarding bent growing dendrites, one possible explanation was given by A. M. Mullis in 1999 [7]. He simulated dendritic bending and rosette formation during solidification under forced fluid flow perpendicular to the growth direction in a shearing flow. So far it is unclear whether our experiments show an actual bent growth or deformation during growth. Phase-field modelling could reproduce the measured bending/deformation during solidification as reported by M. Yamaguchi and C. Beckermann [8]. They computed elasto-viscoplastic deformation of growing solid under a linear shear velocity field using the material point method. According to them, a phase-field model for simultaneous solid deformation and liquid flow is still not available.

Another possible explanation for the observed bent dendrite growth is given by L. Gránásy et al. stating that impurities perturb the solidification process by deflecting the dendrite tip during growth [9]. These foreign particles act as orientation pinning centres. In the melt a random field of impurities may exist leading to zigzagging or spiralling dendrites.

Our study has several limitations for the analysis of the dendrite growth morphologies. First, the strong oscillation and rotation of the liquid droplet has to be taken into account. In particular, the shape of the sample changes during solidification. Second, the nucleation point appears statistically at a random point on the sample surface, so it is not possible to reproduce the same experimental conditions. Third, the fluid flow inside the melt is a nonlinear chaotic process and non-predictable. Fourth, the influence of impurities cannot be neglected.

5. Conclusion

Our research shows bent growing dendrites under different fluid flow conditions. This phenomenon has been observed in-situ, as far as we know, for the first time in solidifying metals during levitation. In the case of Fe-10 at.% B in 1g-EML the effect was stronger than for Fe-1 and 5 at\% B. Bent dendrite growth could also be observed for Fe-1 at.% B under micro-gravity conditions during
parabolic flight with weaker fluid flow compared to 1g-EML. Microstructure analyses of this sample are still in progress.

The cause of the observed bent dendrite growth is unclear. Impurities cannot be neglected and should be considered as a possible explanation to the growth behaviour. Unless it seems to be the influence of fluid flow on the thermal and/or concentration gradients at the growing dendrite tip. A more systematic approach with defined fluid flow conditions is necessary. It is well known that a growing dendrite follows the largest temperature gradient. For instance, future research using an infrared camera could help to clarify and visualise the thermal field around the growing dendrite.

Finally the effect of bent growing dendrites may be used to manipulate the microstructure development during solidification. For example, dendrites could be bent during growth to follow the curving of a turbine blade, or guided growing dendrites of semiconductor on substrates could lead to new technologies.

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References
[1] Dantzig J A and Rappaz M, Solidification 2009 Engineering Sciences, Materials (EPFL Press) 289
[2] Galenko P K, Binder S and Ehlen G J, 2012 Solidification of Containerless Undercooled Melts, ed. D.M. Herlach and D.M. Matson Wiley-VCH, 349-362
[3] Herlach D M, 1991 Annual Review of Material Science 21 23-44
[4] Hyers R W, 2005 Meas. Sci. Technol 16 394-401
[5] Lohöfer G and Piller J, 2002 Proceedings 40th AIAA 2002-0764
[6] Cameron T B and Morral J E, 1986 Metallurgical Transactions 17A 1481-1483
[7] Mullis A M, 1999 Acta Materialia 47 1783-1789
[8] Yamaguchi M and Beckermann C, 2013 Acta Materialia 61 4053-4065
[9] Gránásy L, Pusztai T, Warren J A, Douglas J F, Börzsönyi T and Ferreiro V, 2003 Nature Materials 2 92-96