Quantitative Study on Dendrite Fragmentation Phenomenon in Aluminum Alloy Solidification Process (Postprint)

Authors: Bi Cheng, Guo Zhipeng, LIOTTIE, Xiong Shoumei, GRANTPS

Date: 2023-03-19T00:00:00+00:00

Abstract

Real-time observations of dendrite growth and fragmentation phenomena during the solidification of Al-15%Cu (mass fraction) alloy were conducted using X-ray synchrotron radiography. A large number of dynamic images under various experimental conditions were obtained by applying a pulsed electromagnetic field and altering the dendrite growth direction. Quantitative analysis of the experimental results was performed using Matlab software, and statistical measurement programs were developed to statistically characterize the variation of dendrite fragment count under different experimental conditions, and to measure the distribution relationship of dendrite fragment count along the mushy zone depth and mushy zone solid fraction. The results indicate that the application of an electromagnetic field, growth against the gravitational direction, and dendrites with higher growth velocities produce more dendrite fragmentation; the number of dendrite fragments exhibits a Gaussian distribution along both the mushy zone depth and mushy zone solid fraction, peaking at a solid fraction of approximately 0.45. Finally, the influence of necking fracture induced by the flow field, solute enrichment caused by the gravity field, and interdendritic convection induced by the electromagnetic field on the aforementioned quantitative results was analyzed.

Full Text

Quantification Study on Dendrite Fragmentation in Solidification Process of Aluminum Alloys

Bi Cheng[1], **GUO Zhipeng**[1], **LIOTTI E**[2], **XIONG Shoumei**[1], **GRANT P S**[2] [1] School of Materials Science and Engineering, Tsinghua University, Beijing 100084 [2] Department of Materials, University of Oxford, Oxford OX1 3PH, UK

ABSTRACT

Alloy solidification is an important process to control the mechanical properties of engineering products. During solidification, dendrite fragmentation occurs commonly as a key phenomenon to determine the microstructure and to obtain fine grain size. Recently, in situ synchrotron X-radiography technique was developed and applied to observe thermodynamic behaviors such as dendrite growth and fragmentation during solidification. External forces such as mechanical and electromagnetic stirring, and thermal shock were added into the solidification process to investigate their effects on the fragmentation behavior. However, most work conducted in literature focused on qualitative aspects e.g. morphology transition or solute distribution and quantitative investigation such as determining the specific relationship between fragmentation and solidification conditions was rather limited. In this work, the third generation synchrotron X-radiography technique was used to observe the solidification process of an Al-15%Cu (mass fraction) alloy. Experimental conditions including the strength of the pulsed electromagnetic fields, dendrite growth direction and the temperature gradients were varied and the subsequent effect on fragmentation was studied and quantified. A computer program was developed based on Matlab to perform the image processing and measurement. The fragmentation number according to experiments was counted and correlated to the mushy zone depth and local solid fraction. Results showed that a stronger electromagnetic field, growing against gravity and growing at higher velocity would significantly increase the fragmentation number. Furthermore, the fragmentation number followed a Gauss distribution as a function of either mushy zone depth or local solid fraction, and the maximum fragmentation occurred when the solid fraction was about 0.45. In the end, the extent to which caused those statistic results above were analyzed as the necking process due to the velocity field, the cumulative solid due to the gravity field and the liquid flow due to the electromagnetic field.

KEY WORDS aluminum alloy, X-ray synchrotron radiation, solidification, dendrite fragmentation, mushy zone, electromagnetic field

Introduction

The solidification process of liquid alloys determines the final microstructure morphology, which plays a crucial role in casting performance. During solidification, a series of phenomena including dendrite arm fracture, floating, and subsequent growth occur concurrently with dendrite growth, collectively referred to herein as dendrite fragmentation. An appropriate amount of dendrite fragmentation can increase heterogeneous nucleation during metal solidification, thereby achieving grain refinement and performance improvement. In the past, due to the high-temperature sealed environment of metal solidification, internal processes could not be directly observed. To better investigate the formation mechanism of dendrite fragmentation, its relationship with external conditions, and its influence on the final solidified structure, practical observation methods for metal solidification have gradually been developed. Since the 1980s, scholars have used transparent organic materials to simulate metal solidification processes [1], but property differences limit their ability to completely substitute for metals. Other researchers have employed conventional X-rays to observe metal solidification under equilibrium conditions [2], but resolution limitations prevented grain-level observation. In recent years, synchrotron X-ray imaging technology has entered its third generation, with research facilities established in the United States, Japan, United Kingdom, China, and other countries. This technology offers high radiation intensity, high collimation, high coherence, and high optical sensitivity, meeting researchers' needs for dynamic real-time observation of dendrite growth and fragmentation during solidification.

Based on this observation technology, scholars have conducted in-depth investigations of solidification processes. In experimental observation, Mathiesen et al. [3-5] studied the directional solidification of Al-30%Cu (mass fraction) alloys, obtaining α-dendrite fronts and solute boundary layers in the solidification mushy zone through post-image processing. In Al-20%Cu directional solidification experiments, they directly observed dendrite arms fracturing and drifting to the columnar dendrite growth front, causing the columnar-to-equiaxed transition (CET), and found that the probability of dendrite root remelting fracture depended on buoyancy. Other researchers [6-8] observed the three-dimensional evolution of dendrite growth in Al-10%Cu alloys, detailing the coarsening mechanisms of small dendrite degeneration between large dendrites and dendrite coalescence at high solid fractions, and observed real-time evolution of individual dendrites. Regarding external force fields, Wang Tongmin et al. [9] observed the dynamic regulation of dendrite growth morphology by electric current, noting that direct current significantly inhibited dendrite growth and flattened dendrite tips, while pulsed current refined dendrite arm spacing and affected solidification progress. Other scholars [10-12] observed dendrite fragmentation under pulsed electromagnetic fields, marked the zigzag movement of fragmented dendrites, and statistically compared fragmentation results with and without electromagnetic fields. In theoretical analysis, researchers [13,14] proposed through observation experiments that solute accumulation caused by dendrite arm remelting is the primary cause of dendrite fragmentation. Liu et al. [15] statistically analyzed secondary dendrite arm numbers and growth velocities, suggesting that solute accumulation caused by decreasing growth velocity made dendrite arms more susceptible to necking and fracture.

The experimental results presented in the aforementioned studies are often images of specific processes, with analyses being qualitative. To further reveal the specific relationships between dendrite fragmentation phenomena and solidification itself and external control factors during solidification, systematic quantitative studies are essential. This work focuses on quantitative research, performing image processing and large-scale statistical analysis of experimental results to obtain statistically significant distribution patterns. Based on this, we attempt to quantify the relationships between dendrite fragmentation, mushy zone depth, mushy zone solid fraction, dendrite growth velocity, and other variables.

1.1 X-ray Synchrotron Radiation Experiments

X-ray synchrotron radiation experiments were conducted at the B16 beamline of the Diamond Light Source in the United Kingdom. As shown in [Figure 1: see original paper] [10], the experimental platform consisted of the following components: (1) X-ray source: 18 keV power, penetrating directly through the sample. (2) Alloy sample: Al-15%Cu composition, dimensions approximately 20 mm × 7 mm × 0.2 mm, polished on both sides, sprayed with BN, sealed in a quartz layer with upper and lower wires, mounted on an adjustable sample holder. (3) Heating equipment: Heating devices placed at both ends of the sample with externally controlled temperature, maximum cooling rate of 2 ℃/s. After temperature curve stabilization, error could be controlled within approximately 0.7 ℃. Although sample lengths varied, heating equipment positions were fixed, allowing a stable temperature gradient of 48 K/mm, with dendrite growth direction controlled by changing the temperature gradient direction. (4) Pulsed electromagnetic field: A permanent ring magnet fixed 3 mm in front of the sample generated a pulsed electromagnetic field by applying pulsed current to the upper and lower wires, producing Lorentz forces. Pulse frequency was 1 Hz, pulse current intensity was \$±\$300 mA, and magnetic field strength was set to 0.1 and 0.3 T in experiments. The effective length of the sample in the electromagnetic field was 10 mm. According to the Lorentz force equation, this pulsed electromagnetic field could generate forces of \$±\$0.3 and \$±\$0.9 mN. Current direction, magnetic field direction, and Lorentz force direction are indicated by arrows in [Figure 1: see original paper]. The Lorentz force alternated back and forth perpendicular to the dendrite growth direction within the sample plane, disturbing dendrites and interdendritic liquid. (5) Imaging system: A Phantom 7.3 CMOS high-speed camera received converted X-ray source signals, with image resolution of 5.5 mm/pixel, imaging area of 4.4 mm × 3.3 mm, and frame rate up to 30 fps. (6) Control system: PID control integrated temperature, current, position, and other control terminals into a single panel.

Before experiments, the temperature of both heating blocks was increased to melt the sample, and X-ray imaging was initiated. During experiments, the temperature of one block was decreased to create a temperature gradient, causing directional solidification. After stabilization, the pulsed electromagnetic field was activated, and the solidification dynamic process after temperature stabilization was recorded. All experiments were divided into five groups based on growth direction and magnetic field strength, as shown in . Growth directions included bottom-to-top (against gravity) and top-to-bottom (with gravity).

1.2 Quantification Methods for Experimental Results

After preliminary image processing of experimental observations, clear solidification process image sequences were obtained with 1-second intervals between images. Matlab programming was used to extract various information from images for subsequent quantitative analysis. The parameters requiring quantification for each experiment and their measurement methods were as follows: (1) Cumulative dendrite fragmentation number over time: For each frame, the moment of dendrite fragmentation was manually located, and the program marked and recorded its coordinates, occurrence time, and sequence number. Manual positioning through preliminary training and multiple reviews controlled error within approximately 5%. Finally, marked images and record arrays of dendrite fragmentation at each moment were obtained [10]. (2) Solid fraction of mushy zone during stable growth: The mushy zone in each image was divided into several regions of equal width. After image binarization, the proportion of white pixels in each region was measured as the solid fraction of the mushy zone at different depth ranges, as shown in [Figure 2: see original paper]. Solid fractions measured at different times for the same region were averaged as the final result. (3) Depth of dendrite fragmentation within mushy zone: The Y-direction distance from the dendrite fragmentation moment to the tip of the fastest-growing primary dendrite trunk at that moment was measured, as indicated by arrows in [Figure 3a: see original paper]. (4) Primary dendrite trunk growth velocity: Three primary dendrite trunks with complete growth processes were selected. The positions of dendrite tips at different times were located, and displacements over several time intervals were measured [16], as shown in [Figure 3b: see original paper], where arrows indicate tip displacement after several time intervals. Displacement per unit time interval was calculated as the growth velocity at that moment [17], and the average value yielded a velocity curve composed of several points.

2.1 Variation of Dendrite Fragmentation with Experimental Conditions

Based on the grouping in , cumulative curves of dendrite fragmentation over time were plotted for all experiments, with timing starting after temperature curve stabilization, as shown in [Figure 4a: see original paper]. Linear fitting was performed for each experimental group, yielding five average curves, as shown in [Figure 4b: see original paper]. Due to different effective recording times for each group, this work primarily compared the fragmentation generation rate, i.e., curve slope. As seen in [Figure 4: see original paper], Group B showed the highest total fragmentation number and rate, followed by Group E, with Group A slightly higher than Group C, and Group C slightly higher than Group D. If the outlier sequences (maximum in Group C and minimum in Group D) were removed, results for Groups C, A, and D became essentially similar. Combined with experimental conditions: (1) Dendrites growing from the bottom produced the most fragmentation under electromagnetic fields, several times more than results without electromagnetic fields for the same growth direction. Dendrites growing from the top also produced substantial fragmentation under strong electromagnetic fields, several times more than results with weak or no electromagnetic fields for the same growth direction. Thus, electromagnetic field application promotes dendrite fragmentation. (2) Although

Group B had weaker electromagnetic fields than Group E, its total fragmentation number exceeded Group E, indicating that growing against gravity yields more fragmentation under electromagnetic fields. (3) Although Group A grew against gravity, its results were similar to Group C growing with gravity. Although Group D had electromagnetic fields, its results were similar to Group C without electromagnetic fields. Thus, under conditions where dendrite fragmentation is difficult to produce, the effects of growth direction and electromagnetic field conditions are not obvious or independent but result from combined action.

2.2 Distribution of Dendrite Fragmentation Along Mushy Zone Depth

The depth of all dendrite fragmentation events within the mushy zone at the moment of occurrence was measured. Groups D and E from were combined, and fragmentation numbers within the same depth range were accumulated for each group. Since total numbers differed among groups and had been compared previously, accumulated results were normalized by dividing by the maximum cumulative value within each group to plot normalized histograms of fragmentation distribution along mushy zone depth, as shown in [Figure 5: see original paper]. All groups showed a distribution pattern with more fragmentation in the middle and less at both ends. Gauss distribution fitting [18] was applied to the four groups, with bimodal Gauss fitting used for Group 4. Fitting results are shown in . Combined with experimental conditions: (1) In the deepest mushy zone region, all four groups showed minimal fragmentation distribution. However, in the shallowest region near the dendrite tips, groups growing against gravity showed some fragmentation distribution, while groups growing with gravity showed almost no distribution near tips. (2) From the R^2 values, all four groups matched the Gauss distribution well. (3) Comparing expected values revealed that electromagnetic field application reduced expected values for both growth directions, indicating an overall trend of fragmentation distribution moving toward dendrite tips. For gravity-direction growth, fragmentation simultaneously showed trends toward both shallower and deeper positions (forming bimodal distribution).

2.3 Distribution of Dendrite Fragmentation with Mushy Zone Solid Fraction

Fragmentation numbers in each divided region of the mushy zone were correlated with measured solid fractions of that region. After similar intra-group accumulation and normalization, histograms of fragmentation distribution with mushy zone solid fraction were plotted, as shown in [Figure 6: see original paper]. The distribution also showed a middle-high, ends-low trend with relatively uniform values. Peak fragmentation for Groups A, B, C, and D+E occurred near solid fractions of 0.5, 0.45, 0.35, and 0.45, respectively. Averaging yielded a peak distribution at approximately 0.45 solid fraction. Combined with experimental conditions, electromagnetic field application enhanced the central tendency of fragmentation distribution, with more fragmentation occurring within the 0.4-

0.5 solid fraction range and greater differences between peak and other values.

2.4 Variation of Dendrite Fragmentation with Dendrite Growth Velocity

Since primary dendrite trunks were measured for growth velocity calculations, corresponding fragmentation events were also primary dendrite arm fractures. Five experiments with complete primary dendrite trunk growth from Group A were selected, counting only trunk fractures and plotting fracture numbers over time. As image sizes were identical across the five experiments, observation region volumes were also identical, allowing direct comparison of cumulative fracture numbers without unit volume normalization, as shown in [Figure 7a: see original paper]. Average growth velocities of primary trunks were measured and plotted as velocity-time curves, as shown in [Figure 7b: see original paper]. As seen in [Figure 7a: see original paper], Experiment 1 showed the steepest slope and highest fracture generation rate (though the total number did not increase later due to heating and remelting). Experiments 5 and 1 had extremely similar slopes, with only slight decreases later. Experiments 2 and 3 had smaller slopes, while Experiment 4 had the smallest slope, producing fractures slowest and least. As seen in [Figure 7b: see original paper], Experiment 1 had the highest initial velocity with overall high curve position, followed by Experiment 5, with Experiments 2 and 3 at similar lower positions, and Experiment 4 at the lowest position. Velocity curves could be divided into two stages: early high-velocity stage and later slow-velocity stage. According to [Figure 7a: see original paper], trunk fractures occurred more obviously during the early high-velocity stage. Linear fitting of initial fracture curves (before platform formation) in [Figure 7a: see original paper] yielded approximate slopes. Velocities exceeding the average in [Figure 7b: see original paper] were considered high-velocity stage velocities, and their average was taken as the high-velocity stage average velocity, with results shown in . The two parameters corresponded well to the order 1>5>2>3>4. Combined with experimental conditions, faster primary dendrite trunk growth velocity during the initial stage made trunks more prone to fracture.

Analysis and Discussion

The quantified phenomena can be discussed and explained from three aspects affecting dendrite fragmentation: (1) Solute accumulation: Previous studies [4,10] indicate that appropriate solute enrichment causes remelting at dendrite-dendrite arm connections, promoting fragmentation. In this experiment, solute deposited toward the bottom due to gravity, allowing dendrites growing from the bottom to contact this solute and undergo fragmentation, while dendrites growing from the top had no such opportunity. This aligns with observations that anti-gravity growth produced more fragmentation than gravity-direction growth. Consequently, fragmented dendrites from anti-gravity growth showed some distribution near dendrite tips, while gravity-direction growth showed almost no tip distribution. (2) External influences: Interdendritic liquid flow impacts and entrains dendrites, promoting fragmentation. Pulsed electromagnetic fields generate alternating Lorentz forces that enhance interdendritic liquid convection [19-21], consistent with observed increases in fragmentation with electromagnetic fields. Additionally, dendrite tips have less solid and more liquid than mushy zone bottoms, making fragmentation promotion more obvious after electromagnetic field application, thus causing overall fragmentation distribution to shift toward dendrite tips. (3) Fracture mechanism: According to the necking fracture mechanism, larger connection surface area between dendrites and arms generates higher probability of concave curvature, thinner connections, and easier fracture. Dendrite growth simulations show that faster primary dendrite trunk growth produces thinner trunks [22,23], which according to the fracture mechanism should fracture more easily, consistent with the result that higher trunk growth velocity yields higher fracture rates. Combined with the previous phenomenon that growth direction and electromagnetic field effects are not obvious under conditions where fragmentation is difficult to produce, it can be inferred that dendrite growth velocity determines the thickness of dendrite-arm connections, thereby determining fracture difficulty. If many vulnerable connections exist, anti-gravity growth or electromagnetic fields better promote fracture; conversely, if few vulnerable connections exist, these factors have limited effect.

For fragmentation distribution with solid fraction, solute accumulation effects can be represented by solute concentration. According to the Scheil equation, Al-Cu alloy solute concentration increases with solid fraction. Interdendritic liquid flow can be expressed by permeability. According to the Blake-Kozeny equation [24,25], permeability decreases with increasing solid fraction. These two external factors have opposite trends with solid fraction, counteracting each other. Only at the intersection of the two trend curves can both exert maximum effect, which falls within the 0.4-0.6 solid fraction range, basically consistent with the experimentally observed peak fragmentation distribution near 0.45 solid fraction.

When dendrite fragmentation is abundant, pulsed electromagnetic field application and anti-gravity growth promote fragmentation; when fragmentation is scarce, these promotion effects are not obvious. Total dendrite fragmentation follows a Gauss distribution along mushy zone depth, and fragmentation approximately follows a Gauss distribution with mushy zone solid fraction, with an average peak at 0.45 solid fraction. Faster primary dendrite trunk growth velocity produces more fragmentation. Among factors affecting fragmentation, necking at dendrite-dendrite arm connections plays a decisive role, while solute accumulation and interdendritic liquid flow play promotional roles.

References

[1] Jie W Q, Zhou Y H. J Northwest Polytech Univ, 1988; 6(1): 29
[2] Grange G, Gastaldi J, Jourdan C, Billia B. J Cryst Growth, 1995; 151: 192

[3] Mathiesen R H, Arnberg L, Ramsoskar K, Weitkamp T, Rau C, Snigirev A. Metall Mater Trans, 2002; 33B: 613
[4] Mathiesen R H, Arnberg L. Mater Sci Eng, 2005; A413: 283
[5] Mathiesen R H, Arnberg L, Bleuet P, Somogyi A. Metall Mater Trans, 2006; 37A: 2515
[6] Limodin N, Salvo L, Suery M, DiMichiel M. Acta Mater, 2007; 55:
[7] Campanella T, Charbon C, Rappaz M. Metall Mater Trans, 2004; 35A: 3201
[8] Li M J, Tamura T, Omura N, Miwa K. J Alloys Compd, 2010; 494:
[9] Wang T M, Zhu J, Chen Z N, Xu J J. Sci Sin Phy, Mech Astron, 2011; 41: 23
[10] Liotti E, Liu A, Vincent R, Kumar S, Guo Z, Connolley T, Dolbnya I P, Hart M, Arnberg L, Mathiesen R H, Grant P S. Acta Mater, 2014; 70: 228
[11] Shu D, Sun B D, Mi J W, Grant P S. Phys Metall Mater Sci, 2012; 43A: 3755
[12] Mapelli C, Gruttadauria A, Peroni M. J Mater Process Technol, 2010; 210: 306
[13] Ruvalcaba D, Mathiesen R H, Eskin D G, Arnberg L, Katgerman L. Acta Mater, 2007; 55: 4287
[14] Pataric A, Mihailovic M, Gulisija Z. J Mater Sci, 2012; 47: 793
[15] Liu S, Lu S Z, Hellawell A. J Cryst Growth, 2002; 234: 740
[16] Yasuda H, Ohnaka I, Kawasaki K, Suglyama A, Ohmichi T, Iwane J, Umetani K. J Cryst Growth, 2004; 262: 645
[17] Stransky K, Kavicka F, Sekanina B, Stetina J, Gontarev V, Dobrovska J. Mater Technol, 2011; 45: 163
[18] Arnberg L, Mathiesen R H. JOM, 2007; 59(8): 20
[19] Ananiev S, Nikrityuk P, Eckert K. Acta Mater, 2009; 57: 657
[20] Schenk T, Thi H N, Gastaldi J, Reinhart G, Cristiglio V, Manglinck-Noel N, Klein H, Hartwig J, Grushko B, Billia B. J Cryst Growth, 2005; 275: 201
[21] Zhu J, Wang T M, Chen Z N, Xu J J, Xie H L, Xiao T Q, Li T J. In: Ludwig A ed., IOP Conf Ser: Mater Sci Eng, 2012; 33: paper No.012039
[22] Guo Z, Mi J, Xiong S M, Grant P S. J Comput Phys, 2014; 257:
[23] Rack A, Weitkamp T, Riotte M, Grigoriev D, Rack T, Helfen L, Baumbach T, Dietsch R, Holz T, Kramer M, Siewert F, Meduna M, Cloetens P, Ziegler E. J Synchrotron Radiat, 2010; 17: 496
[24] Puncreobutr C, Phillion A B, Fife J L, Lee P D. Acta Mater, 2014; 64: 316
[25] Nielsen O, Arnberg L, Mo A, Thevik H. Metall Mater Trans, 1999; 30A: 2455

Note: Figure translations are in progress. See original paper for figures.

Source: ChinaXiv — Machine translation. Verify with original.