THREE-DIMENSIONAL DENDRITIC MORPHOLOGY AND BRANCHING MECHANISM IN DIRECTIONALLY SOLIDIFIED MG-ZN ALLOY

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Abstract. The three-dimensional α-Mg dendritic morphology and branching mechanisms of directionally solidified Mg-Zn alloy were studied. Different microstructures were obtained under different pulling velocities various from 30μm/s to70μm/s. Directional growth tissue of α-Mg dendrite was obseved in the specimen solidified under the pulling velocity of 30μm/s, while in the samples solifdified in other pulling velocities α-Mg dendrites tend to exhibit equiaxed morphologies without any directional growth structure. Coupled with a novel method X-ray synchrotron tomography and trational metallographic technique, the three-dimensional morphology and topology structure of the α-Mg in the sample with the pulling velocity of 30μm/s were characterized. The result show that the morphology of α-Mg dendrites are diversity and complicated in three-dimensional space. Through the analysis we demonstrated that the α-Mg dendrite grows with seven secondary dendrite arms around the trunk which is different from what have been observed in the directionally solidified AZ91[1][2] and non-directional solidified Mg-Zn[3] or Mg-Al[4] alloys. In addition, the angles between the secondary dendrite arms and the trunck are different, with an angle of 53~54 °for three arms, 80~82 ° for two and 60 ° for the rest of the two arms. A hypothetical model of topological structure in α-Mg dendrites is proposed and the crystallography of the branches and advancing dendrite is discussed.

1 INTRODUCTION

Mg-based alloys are known for their low density and high strength-to-weight ratio and gaining increasing technical importance in the transportation and aerospace industries [5]. Nowadays, even in biomaterial, magnesium alloys are widely used as implant materials due to its biodegradable. Solidification is a phenomenon which is widely existed in material processing, such as casting, welding, etc. Magnesium-Aluminium and Magnesium-Zinc alloys constitute important classes of high pressure die-casting lightweight alloys and solidification is an important formation route for Mg-based alloys. Due to the close relationship between the properties and microstructure of material, to understand the dendrite growth, topology, morphology and crystallographic orientation is crucial in material science and engineering. But to date, most of the research of dendrite orientation and morphology is focused on face-centred cubic or body-centred cubic alloys. The study related to dendrite growth during solidification of HCP alloys such as Mg is still relatively rare. In addition, to characterize the 3-D morphology and topology evolution of dendrites during solidification is important and full of challenge.

It is generally believed that the dendrites of FCC or BCC alloys grows with a preferred orientation of [100] [6-7], while for HCP alloys, usually there are a variety of preferred orientations such as $\langle 10\bar{1}0\rangle$ for Zn, Cd and H_2O, $\langle 11\bar{2}0\rangle$ for Mg alloys, and even $\langle 0001\rangle$ in $Co_{17}Sm_2$ and Zn [7]. According to the research of Rappaz [8-9] and co-works, there is a dendrite orientation transition (DOT) phenomenon in binary Al-Zn alloys. The preferred dendrite orientation of α-Al changed from <100> to <110> with the Zn composition various from 5% to 90%. Their experimental results also illustrate that the concentration of the solid solution elements is one of the important factors to determine the dendrite orientation during the solidification. But it is still unclear that whether the α-Mg will suffer the similar dendrite orientation transition in Mg-Zn alloys, and that the morphology or topology will be affected with different concentrations of Zn

The work of Wang [4]shows that in Mg-9Al alloy, the morphology of α-Mg tends to form a plate-like structure and the preferential growth orientation is $<11\bar{2}0>$, while in Mg-38wt.% Zn alloys the morphological and topologic structure show to be complex in three-dimension space, and the preferred growth direction is $<22\bar{4}5>$. They also demonstrated that the α-Mg(Zn) dendrites grow with a six secondary branched arms around the advancing dendrite tips [3]. Furthermore, Pettersen [1] et al. studied the crystallographic direction and the topological structure of Mg dendrites in directionally solidified AZ91 alloy (9.1 wt.% Al, 0.81 wt.% Zn, 0.27 wt.% Mn), and suggested that the dendrites grew in a $<11\bar{2}0>$ direction at a low temperature gradient and high growth velocity, while at higher temperature gradient and higher growth velocity, the growth direction was $<22\bar{4}5>$ [2]. They also found that dendrites with $<11\bar{2}0>$ stems have arms in six directions around the stem, while dendrites with $<22\bar{4}5>$ stems get only three secondary arm directions.

It is obvious that both the solid solution elements in magnesium alloys and the growth conditions such as temperature gradients and cooling rates can greatly affect the dendrites orientation [10-11] and morphologies during solidification. During the practical solidification, the heat release in front of the solid-liquid interface can be various by the perturbation, and then resulting in a variety of complicated morphologies which can only be quantitatively characterized in 3-D space. In addition, the effect of alloying elements on dendrite growth is also fundamental for the anisotropy of the crystal as they can directly determine the preferred orientation of dendrite during the evolution. In Mg-Al alloys, the preferred orientation of the α-Mg dendrites is $<11\bar{2}0>$ as predicted for the reason that aluminum is relatively isotropic so that the solid-liquid interface free energy doesn't vary dramatically in different crystallographic orientations [12]. The addition of Al into Mg does not change the growth orientation of α-Mg dendrites due to the isotropic property of Al. While for Zn, the anisotropy different between the base plane and c-axis direction can be as high as 30% [13], which means the addition of Zn in Mg will affect the preferred orientation of α-Mg dramatically during the dendrite growth due to the large anisotropy of Zn. This brings great challenge in predicting the complex 3-D morphologies and topological structure of α-Mg dendrite in Mg-Zn alloys during solidification and to characterize it quantitatively.

Up to now, the research related to the basic understanding of 3-D morphology and topology of dendrite growth in directionally solidified Mg-Zn alloys is still rare, and most of the work was focused on the 2-D microstructures which cannot provide the complicated and diversity of dendrite growth in Mg-Zn alloys. Thanks to the development of Serial-Sectioning [14], FIB [15] and Synchrotron radiation x-ray tomography [16] et al, we can get the access to the three-dimension visualization of α-Mg dendrite morphology and topological structure in directionally solidified Mg-38wt.%Zn and characterize them qualitatively and quantitatively. In this paper, we combined the x-ray tomography with traditional metallographic methods to characterize the 3-D microstructures of α-Mg in Mg-Zn alloys. The crystallography morphologies and topological structure were studied in detail.

2 EXPERIMENTAL METHODS

2.1 Sample preparation

Cylindrical Mg-38 wt. % Zn specimen (ϕ = 6.6 mm, L = 100 mm) was remelted at 720 ℃. After kept at the temperature of 720 ℃ for 30 minutes, the specimen was solidified in a Bridgeman directional solidification furnace with different pulling velocities of 30um/s, 50um/s and 70um/s respectively. The temperature gradient was 10~20 ℃/mm. These specimens were quenched to liquid metal bath to preserve the columnar structure. After the solidification, the specimens were machined into two pieces from the center of the longitudinal section by electrical discharge method for the metallographic and subsequent EBSA experiment. The regions with an integral developed dendrite were chosen according to the metallographic observation and were cut again by the electrical discharge cutting method for the Synchrotron radiation x-ray tomography. The diameter of the specimen for the x-ray tomography experiments is ~1.2mm. During the x-ray microtomography, a spatial resolution of 0.74μm with the total pixels of 2048 for each projection was selected. The size of the full field during imaging was 1.4mm, so that the size of specimen should be smaller than that of the full imaging field in order to reconstruct the microstructures successfully.

2.2 Three-dimensional characterization by Synchrotron radiation x-ray tomography

The x-ray microtomography experiments were carried out at the BL13W1 beamline of the Shanghai Synchrotron Radiation Facility (SSRF). This Beamline offers fast acquisition of tomographic data at sub-micron spatial resolution. An x-ray energy of 30~35 keV was used to penetrate a cylindrical Mg-38 wt.% Zn specimen of approximately 1.2 mm in diameter, ~5 mm in height, with a voxel size of approximately (3.7μm and 0.74μm). A YAG:Ce scintillator screen was used to convert the transmitted x-rays to visible light. This was coupled with 2048 × 2048 pixels from a CCD camera with typical exposure times 400ms for resolution of 3.7μm and 4s~6s for 0.74μm per projection. In this configuration we collected a projection every 1/4 degree between 0° and 180°. The 2D projections were reconstructed in 3D using a filtered-back-projection algorithm. The ImageJ and Amira software were used to process and reconstruct 3D visualization after the acquisition of the 2D slice.

3 RESULT AND DISCUSSION

Through the analysis from x-ray tomography result and metallographic graphs, we present the dendrites orientation and 3-D microstructures of α-Mg in directionally solidified Mg-38wt.%Zn alloys. For directionally solidified alloys with pull velocities various from 30μm/s to 70μm/s, the metallographic images (figure1) show that directional growth dendrites can only be observed when the pull velocity is below 30μm/s. Samples solidified under the pull velocities above 30μm/s tend to show equiaxed morphologies as can been seen in figure b,c.

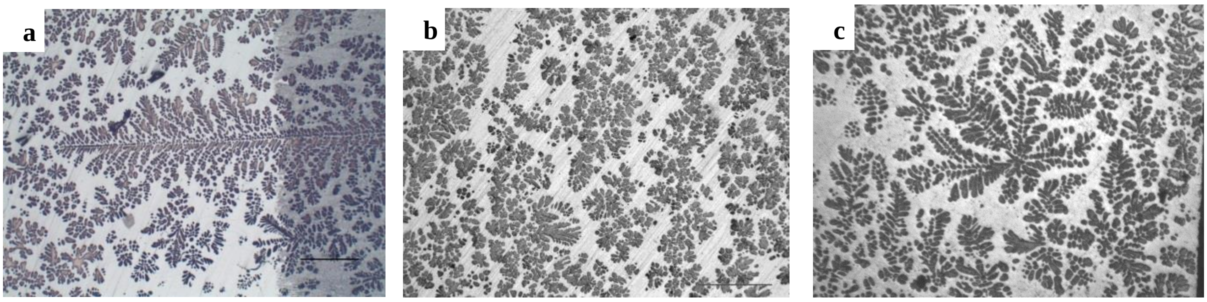

Figure 1 The longitudinal section of Mg-38wt.%Zn (a.30μm/s, b.50μm/s and c.70μm/s)

3.1 Morphologies of α-Mg in quenched and directionally solidified Mg-Zn alloys

The 3D dendrite morphology evolution during solidification can be a fundamental factor which determines the mechanical properties of magnesium alloys. The nucleation and growth of secondary phases such as eutectic phase or precipitations following the primary phase is affected by the morphology of primary dendrites, which makes the distribution of the secondary phases more complicated.

For the specimen quenched at temperature of 10℃ above the eutectic point, the growth condition of α-Mg was relatively homogeneous, without distinctive temperature gradient among different directions and the α-Mg dendrites were equiaxed. The preferred growth orientation was more likely to be determined by the anisotropy of interface free energy which is dependent on the crystalline anisotropy of solid solute element Zn as Mg is relatively isotropic [17]. From the morphologies images in figure 2, the structure shows to be various and complicated in comparison with the six-symmetric structure, therefore it can be deduced that the preferential growth direction of α-Mg is obviously different from the predicted $<11\bar{2}0>$ and that the addition of Zn in Mg alloys will affect the growth direction of α-Mg dendrite due to the large anisotropy of Zn element, but how it affects the dendrite growth direction and whether there are different effects for different content of Zn will be discussed in another paper in preparation.

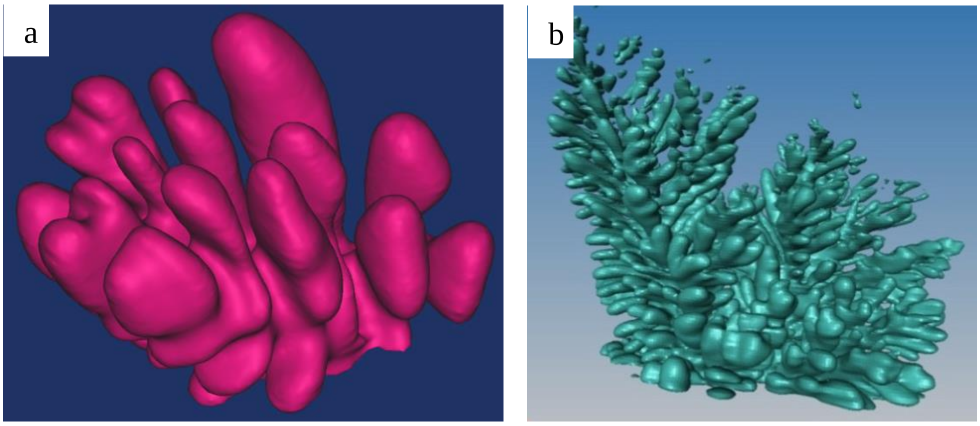

Figure 2 3D reconstructed results of Mg-38wt.% Zn quenched at temperature of 10 ℃ above the eutectic point

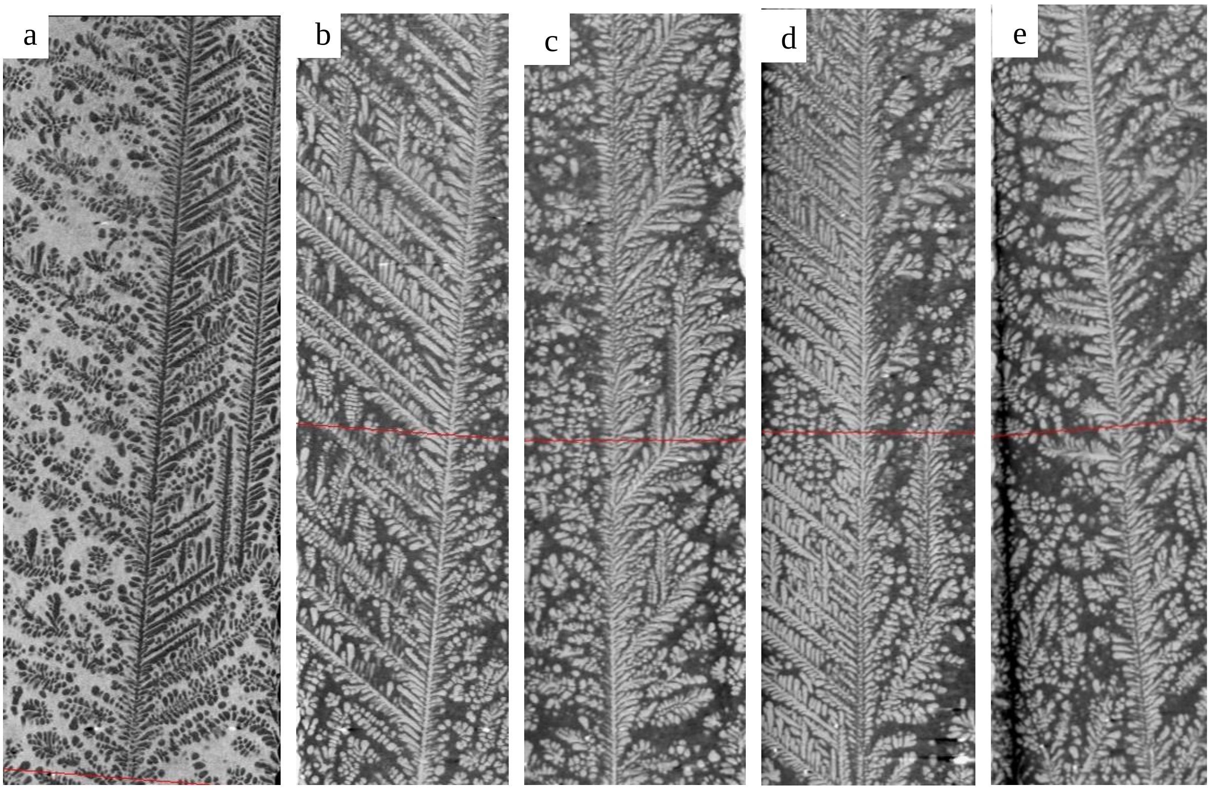

Figure 3 Selected longitudinal sections from x-ray tomography (figures from (a) to (e) illustrate the different dendrite morphologies in an extracted integral directionally growth dendrite with abundant secondary and higher -order dendrites branches. The morphologies vary in different dendrite branches growth directions)

While in directionally solidified Mg-Zn alloys, the morphology and topological structure of the trunk and secondary arms (SA) tend to be more complicated than that in the equiaxed α-Mg. The different longitudinal sections with well-developed secondary arms were extracted from the x-ray tomographic reconstructed result and were shown in figure 3. As can be seen, different morphologies of the secondary arms were well developed. Some arms tend to grow along a certain direction while some SAs evolved like seaweed (figure 3(c)) without definite direction. Besides, the angles between SAs and the stem vary from ~53 ° to ~80 °for different growth directions.

3.2 Topological structure of α-Mg dendrites

The results of the three-dimensional microstructure, cross-section and different longitudinal sections are shown in figure 4. One interesting phenomennon found is that almost all the fully developed dendrits tend grow with seven secondary arms aroud the trunk which is different from the topological structure reported in the literatures [1-3].

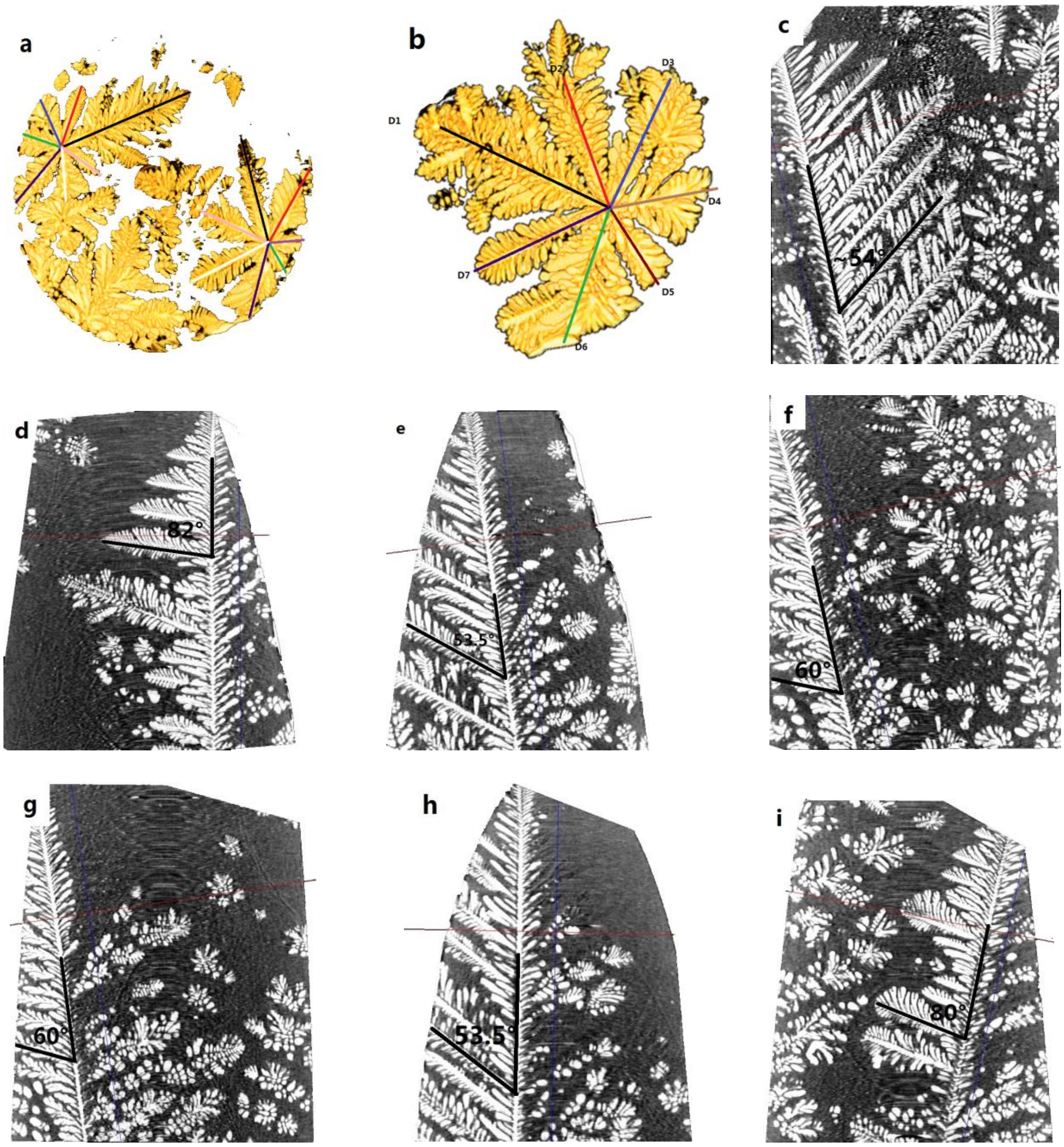

Figure 4 The morphology and topological structure of dendrites in the directionally solidified Mg-38wt.% Zn alloys (pulling velocity: 30μm/s)

In order to quantitatively analyze the topological structure, all of the angles between the SAs and the trunk were measured and the mearsured values are listed in table 1. In total, five angles of each direction were measured and the averages were taken as the final angle of each direction to enhance the reliablity of the result. From the measured angle for the seven dendrite arms listed in table 1,it is confirmed that three of the dendirte arms (SA1, SA3 and SA6) grow in a angle of ~54 °which is much close to the angle between $\langle 22\bar{4}5\rangle$ and $\langle 11\bar{2}0\rangle$ directions for the h.c.p structure. While two of the SA arms (SA4 and SA5) with seaweed-like arms are ~60 °between the trunk which is the angle between $\langle 22\bar{4}5\rangle$ and$\langle \bar{4}225\rangle$ or $\langle 2\bar{4}25\rangle$. The remaining two (SA2 and SA7) are ~82 ° with their thirdly arms like seaweed as the SA4 and SA5. 82 ° is an angle which is rare reported in the literature, but it is very close to 90 ° which means that the secondary arms is perpendicular to the stem and the orientation can supposed to be $\langle 1\bar{1}00\rangle$ if the direction of the trunk is $\langle 22\bar{4}5\rangle$. Therefore, in our case, the main dendrite is more likely to grow in a $\langle 22\bar{4}5\rangle$ direction with 7 dendrite arms around the it and the growth directions of the secondary arms are $\langle 11\bar{2}0\rangle$, $\langle \bar{4}225\rangle$, and $\langle 1\bar{1}00\rangle$, respectively.

Table 1: The angles between secondary dendrite arms and the trunk

Directions	**angles between trunk and the secondary arm/ °**					
	angle1	angle2	angle3	angle4	angle5	average
SA1	53.87	53.13	57.25	56.23	54.18	54.93
SA2	79.41	75.62	80.33	85.49	85.41	81.25
SA3	53.53	53.25	53.08	53.51	53.38	53.35
SA4	58.82	60.48	60.32	60.40	59.14	59.83
SA5	60.79	60.14	59.62	61.31	61.35	60.64
SA6	52.73	53.11	54.16	53.77	53.43	53.44
SA7	80.88	80.39	79.53	80.94	79.69	80.29

4 CONCLUSION

Coupled with a novel method x-ray tomographic microscopy and traditional metallographic techniques, the 3D dendritic morphology and topological structure in Mg-Zn directionally solidified alloys were studied. The experimental results indicate that in both quenched and directionally solidified Mg-38wt.%Zn alloys, the α-Mg dendrite morphology and topological structure are more complicated than those in Mg-Al alloys, with a denser branches with secondary, thirdly or higher-order arms around the trunk in the Mg-Zn alloys than in the Mg-Al alloys. For the quenched specimen with an equiaxed growth α-Mg, it is difficult to tell precise number of secondary dendrite arms growing round the trunk as he dendrites are not well developed, while in directional solidified specimen, it is unambiguous that seven dendrite arms developed around the stem, however it is not sure whether the case is ubiquitous or there are other kinds of topological structures as it is very hard to investigate all the isolated dendrites in the solidified specimen. In addition, the morphologies of arms in different directions are various and there are three different angles between the main dendrite and the secondary arms. For the detail of all the angles, three of which are ~54 °, two ~60 °and the others are ~81 °. The hypothetical model can reflect the topological structure of α-Mg dendrite in directional solidified Mg-38wt.%Zn alloys in a certain extent.

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