Where do Crystals Start to Grow and How is the Microstructure Evolved? – Solidification of Zn-Al Alloys for Improving Zn-coated Steel Sheets –

Steel sheets coated with Zn-based alloys such as Zn-Al and Zn-Al-Mg, have been widely used for various applications because of their excellent corrosion resistance. In the hot-dip coating process, a steel sheet is dipped in a molten Zn bath for certain duration and is then pulled from the bath. The molten layer on the steel sheet solidifies, producing the coating layer. The hot-dip process for Zn coating is a type of solidification process.

Uniformity of the solidified structure in the coating layer is an important issue in improving the quality of products. Despite this importance from the industrial viewpoint, the solidification phenomenon in a molten Zn layer is still not well understood. It is valuable to clarify how the microstructure of Zn-Al alloys is evolved during solidification.

SPring-8, a third generation synchrotron radiation facility, enables us to use monochromatized hard X-rays, which are advantageous for observing microstructure evolution *in situ*. Using synchrotron radiation, we performed time-resolved X-ray imaging to observe crystal growth and solidification of Sn-based and Al-based alloys [1-4]. In this project, an experimental technique was developed for observing the solidification of Zn-Al alloys *in situ*.

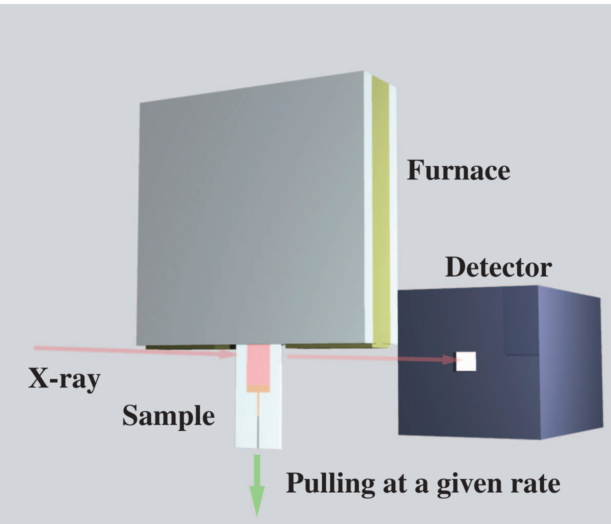


Fig. 1. Set-up of time-resolved X-ray imaging for solidification of metallic alloys. The alloy specimen was melted in the furnace and was pulled down at a given rate. Solidification occurred from the bottom to the top. The X-ray beam passing the specimen was detected using the image detector.

The experiments were performed at beamline **BL20B2**. The setup of the observation system is shown in Fig. 1. An image detector consisting of an X-ray direct-sensing pickup tube “SATICON” was used (pixel size: 4 or 10 μm/pixel, 1024 pixels × 1024 pixels; depth: 10-bit resolution; frame rate: 32 fps max). A sample-to-detector distance was 2.5-3.0 m, which gave rise to the phase contrast along with the absorption contrast. The specimen was melted in the furnace and was pulled down at a given rate.

Figure 2 shows the sequence of transmission images (1 fps and 10 μm/pixel) for the solidification of the Zn-10mass%Al alloy at a pulling rate of 10 μm/s. Dendrites of the primary Al-rich phase were clearly observed by the absorption contrast, and the eutectic interface of the Al-rich and the Zn-rich phases was also detected by the phase contrast (arrow a). Nucleation ahead of the growing Al-rich dendrite and fragmentation of the Al-rich dendrites (arrows b and c) occurred during the unidirectional solidification. The

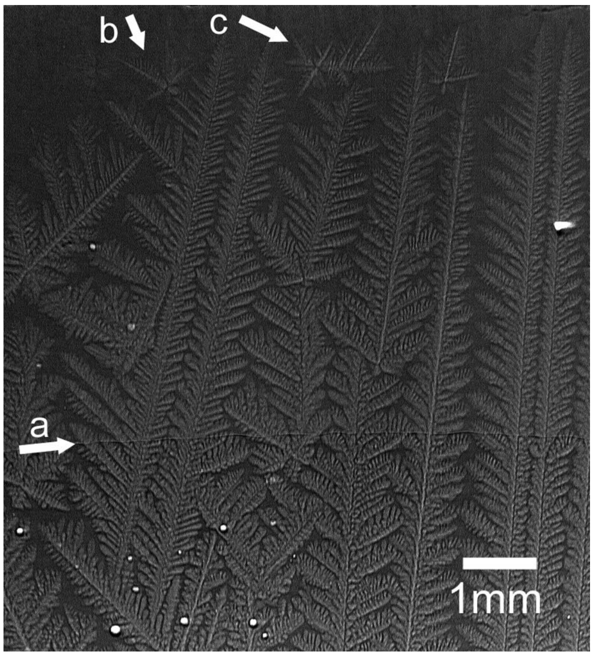
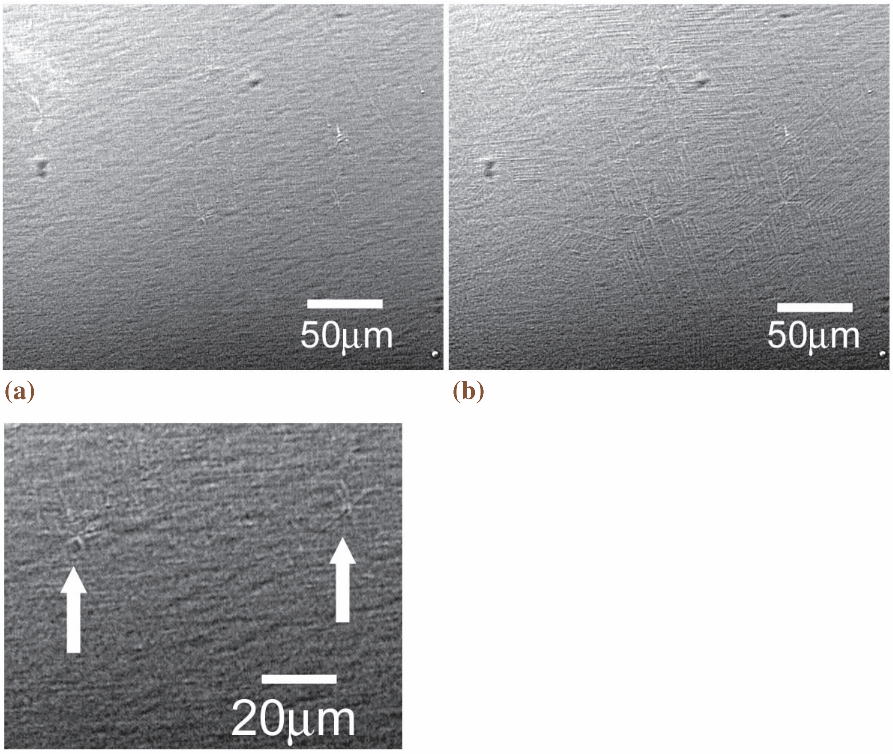


Fig. 2. Development of Al-rich dendrites (gray) in Zn-10mass%Al alloy. The pulling rate was 10 μm/s in the vertical direction. The interface of eutectic solidification (Al-rich and Zn-rich phases) was realized by the phase contrast **a**. Fragmentation (**b** and **c**) and nucleation ahead of the dendrite tips resulted in the formation of equiaxed grains.

observation showed that the grain size of the Al-rich phase was determined by the frequency of both nucleation and fragmentation.

Time-resolved imaging (16 fps and 4 μm/pixel) was also performed for the solidification of the Zn-10mass%Al alloy at a higher cooling rate of about 5 K/s. Figure 3(a) shows a transmission image obtained after the nucleation of the Al-rich phase in the Zn-Al alloy was realized. Development of the Al-rich dendrites is also shown in Fig. 3(b). As shown in Fig. 3(c), the crystals (envelope size, 10 - 20 μm; width of the branches, several μm) formed just after nucleation can be detected using the present setup.

The frequency of nucleation in the Zn-Al alloy on the steel sheet was higher that that in the Zn-Al alloy without the steel sheet. The increase in nucleation frequency suggests the presence of preferred nucleation sites on the steel sheet. The *in situ* observation at a high frame rate (32 fps max) and a high spatial resolution (10 μm) enables the determination of where the crystals start to grow and how the microstructure is evolved, and provides valuable information for improving the microstructure control of Zn-coated steel products.



(a) (b) (c)

Fig. 3. Observation of solidification in Zn-10mass%Al alloy. **(a)** After nucleation of the Al-rich phase occurred, **(b)** development of Al-rich dendrites, and **(c)** close-up view of the crystals after the nucleation. The present setup enabled to observe where the crystals nucleated and how they grew during the cooling procedure. The in situ observation provides valuable information for controlling the microstructure of Zn-coated steel products.

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