

“AlSiCu\_unmodified” shows microstructure evolution of a non-modified Al-Si eutectic in an Al-8wt%Si-15wt%Cu alloy during directional solidification with the imposed temperature gradient parallel to gravity. A relatively high alloying with Cu is done to provide adequate X-ray transmission contrast to study the Al-Si eutectic, since the difference in X-ray attenuation between Al and Si alone is far too low to allow for appreciable contrast at the X-ray energies required for sample transmission. The partition of Cu in Si is practically zero, and in  $\alpha$ -Al  $\sim$  3-4wt%Cu at relevant temperatures and accordingly Cu rejection at the eutectic front is substantial, providing appreciable transmission contrast between a Cu-enriched liquid and the Al-Si-solid. Although the  $\alpha$ -Al and Si-crystals cannot be distinguished directly from one another via the X-ray transmission contrast alone, they are easily identified by large differences in growth dynamics and interface morphologies. Due to gravity-induced settlement of the Cu-rich melt, the sample has already segregated slightly, and become hypereutectic in the region where the image sequence is taken from, evidenced by the presence of two faceted primary Si-crystals. These crystals formed before they appeared in the camera field of view at higher temperatures, presumably inside the hot compartment of the Bridgman furnace which was operated at 858 K. As the eutectic interface approaches the primary Si-crystals, new non-faceted Al-crystals nucleate at the interfaces of the primary Si-crystals and start to grow as dendrites into the intercolonial melt. This is consistent with observations of halo formation of  $\alpha$ -Al surrounding primary silicon crystals in Al-Si alloys, and confirms that Si can act as a nucleant for  $\alpha$ -Al. Later, several new Si-needles is seen to form at irregular intervals and shoot off quickly in directions distinctively different from those compatible with side-branching of the Al-dendrite. It is not possible to determine directly from the images whether these needles really are new Si-crystals, or if they have an orientation relationships with pre-existing Si-crystals. Yet, the Si-needles always form in the vicinity of the  $\alpha$ -Al dendrites, and may well nucleate directly on the  $\alpha$ -Al at adequately high undercooling. It is notable, that frequency for formation of silicon needles is relatively high. Potentially, also the high Cu-content could have an effect on nucleation mechanisms. Observing the growth morphology of the Si crystals in greater detail indicates that they have needle-like tip morphologies, probably due to the high Cu-solute undercooling. In other video sequences taken with this sample the Si-crystals were found to attain a more plate-like morphologies when they grew in directions pointing away from the front, inwards the eutectic mush, while Si-crystals propagating with larger growth velocities more parallel to the imposed gradient, remained needle-shaped. The coupled nature of growth of this eutectic is readily observed, as dendritic  $\alpha$ -Al quickly evolves from the Si interfaces. Another observation is the disordered nature of eutectic growth in this alloy resulting in liquid pockets surrounding solid eutectic as solidification progresses. Although similar observations can be made with quenched samples from binary Al-Si alloys, the pockets here are due to Cu-accumulation in the mushy zone liquid which will remain molten until the Al-Cu eutectic reaction is reached. Clearly the Al-Si eutectic solid-liquid interface can be described as irregular and highly non-isothermal, suggesting that a good mathematical description of such an interface is very challenging, and perhaps not properly represented by a non-isothermal modification of the Jackson-Hunt model.

“AlSiCu\_Sr-modified” shows results from a directional solidification experiment with the imposed temperature gradient parallel to gravity in a Al-9wt%Si-15wt%Cu-0.015wt%Sr

alloy. As for the Al-8wt%Si-15wt%Cu, there is no appreciable absorption contrast available between Al and Si at relevant X-ray energies, and the role of the Cu addition is mainly to provide the necessary image contrast for the experiments. It is readily apparent that the Al-Si eutectic solidification is very different in this Sr-modified alloy compared to the unmodified alloy shown in "AlSiCu\_unmodified". In the Sr-modified alloy, nucleation of a eutectic cell is seen to occur at or near the surface of a secondary branch of the rightmost  $\alpha$ -Al dendrite. Nucleation of the eutectic cell is restricted beyond nucleation of the first silicon. Watching the video sequence carefully from the start, a silicon particle can be seen moving in from the rightmost interdendritic column over the  $\alpha$ -Al dendrite in the location where the eutectic cell forms. Nevertheless, nucleation of the Al-Si eutectic does not occur before the Si particle has reached the Cu-enriched melt surrounding the  $\alpha$ -Al dendrite, indicating that additional undercooling beyond the Si nucleation point is required for the Al-Si eutectic reaction to occur. It is assumed, both with the unmodified and modified alloys that Si nucleation occurs on potent intermetallic particles suspended in the melt. Potentially, nucleation of Si could also occur on the sample container/oxide skin, but due to the observations made from several repeated solidification sequences made with these alloys, that can be considered as quite unlikely, although it cannot be completely excluded [Ref: R.H. Mathiesen, et al., Met. Mat. Trans A. 2010, submitted]. The eutectic cell is initially quite globular, but gradually evolves into a coral-like six-folded equiaxed cellular rosette. The solid-liquid interface is quite smooth and it is not possible to distinguish a leading phase, although small protrusions to the interface can be seen and are presumed to be Si since this phase will have to reject more Cu and consequentially adapt to a higher undercooling. Post-mortem metallography investigations have revealed the typical fibrous Si consistent with Sr- and Na-modified Al-Si eutectic microstructures in castings with commercial Al-Si alloys. The rosette envelope branches of the eutectic grow with reasonably steady tip velocities which is quite uniform in all directions. It is presumed that the rosette-like eutectic envelope is caused by the far-field transport of rejected Cu from the fine-structured fibrous eutectic. As the eutectic evolves the short-range variations in Cu-concentration along the eutectic interface caused by the difference in Cu-solubility between Al and Si is not sufficient to redistribute all the rejected Cu into the liquid adjacent to the eutectic front, and in consequence a far-field Cu concentration gradient establishes normal to the interface, giving rise to Mullis-Sekerka type interface perturbations that evolve into the equiaxed envelope morphology. As no other eutectic cells are seen to nucleate within the field of view during the video sequence, it seems justified to claim that Sr-additions has profound effects on the nucleation frequency, and consequentially the subsequent growth process and thereby also the final eutectic microstructure. . [Refs. to original work: RH Mathiesen, L Arnberg, Y Li, V Meier, PL Schaffer, I Snigireva, A Snigirev and AK Dahle. Met. Mat. Trans. A, 42A, 2011, 170-80; RH Mathiesen, L Arnberg, Y Li, I Snigireva, A Snigirev and AK Dahle, ISIJ Intl. 50, 2010, 1936-40].