The solidification rate of high-strength aluminum alloys strongly impacts the cast structure. The size of eutectic cells could be reduced from several hundred micrometers, typical for ingot-cast materials, down to sub-micrometer size in rapidly solidified materials. Finer initial microstructures reduce the necessary high-temperature homogenization holding times, detrimental to some beneficial dispersoid-forming microalloying elements. A typical example is Sc, which positively influences the formation of fine-grained structures in post-processed materials. However, it is prone to coarsening and a loss of the beneficial effects at high temperatures. Two Al-Cu-Li-Mg-Zr-based alloys, one with an addition of Sc, were conventionally ingot-cast, twin-roll cast, and melt-spun. A relationship between solidification kinetics and the size of the eutectic cells was established, and a significant homogenization holding time reduction was confirmed. A model processing route based on physical metallurgy was proposed. The beneficial effects of Sc on texture, fine-grained structure, and mechanical properties were fully exploited and confirmed.