Non-destructive synchrotron probes of optical floating zone crystal growth: cm-scale grain maps, interfaces, mosaicity, and temperature profiles

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Large single crystals grown using optical floating zone (OFZ) furnaces have had a transformative impact on many scientific fields over the last few decades. However, the OFZ crystal growth process remains more of an art than a science since it is typically only can be monitored visually with a video camera. Our integrated synthesis, characterization, and modeling team has developed novel synchrotron-based methods that provide unprecedented quantitative insights into the OFZ growth process. These methods exploit the ability of high energy X-rays to effectively penetrate through many mm (or even cm) of crystals.

The first thrust of this work was investigating temperature gradients present in tip-heated polycrystalline rods, which are used as the material feed in OFZ crystal growth. We designed a "mini-FZ" furnace capable of reaching temperatures exceeding 1500 °C that is compatible with synchrotron powder diffraction beamlines. This novel furnace allows temperature profiles to be mapped with both high precision and with high spatial resolution, allowing us to directly probe the effect of both the gas type and the gas flow on sample thermal gradients. Our *in situ* data were used to develop the first experimentally validated physical models that effectively describe both the position- and time-dependence of thermal gradients.

The second thrust of this work was developing methods and software for non-destructive grain mapping studies on OFZ crystal growth boules (typically about 6 mm in diameter and 50 mm in length) – samples that are far larger than can be accommodated using commercial lab instruments or even existing synchrotron beamline resources. Our approach utilizes high-energy synchrotron radiation that can fully penetrate through the diameter of our samples, providing comprehensive information its component grains. Furthermore, the highly parallel synchrotron beam enables us to resolve rocking curve features on the order of 0.01°, allowing us to explore correlations between the growth process and the crystal quality. Our fast grain-tracking algorithms follow both the position and orientation of grains whose maximum dimension ranges from sub-mm to 50 mm, allowing us to sensitively determine preferred growth directions. Furthermore, the use of a focused beam (~30 micron diameter) to collect high-resolution maps allow us to infer the shape of the solid-liquid interface and provides high sensitivity to anomalies that occur at the beginning of the crystal growth which may affect the final growth product. While the *ex situ* studies described here focused on the forensic analysis of crystal growths done prior to synchrotron beamtime, they can readily be adapted to study crystal growth *in situ* as soon as suitable furnace environments are available.