High-Energy Diffuse Scattering on the 1-ID Beamline

T. R. Welberry,[1] D. J. Goossens,[1] D. R.Haeffner,[2] P. L. Lee,[2] J. Almer[2]

[1] *Research School of Chemistry, Australian National University, Canberra, Australia*

[2] *User Program Division, Advanced Photon Source, Argonne National Laboratory, Argonne, IL, U.S.A.*

Introduction

We report here on experiments to record the detailed diffuse diffraction patterns of a number of ceramic materials using high-energy (65.35 keV) x-rays. Although diffuse scattering can easily be observed by directing the incident beam onto a stationary crystal and recording the scattering on a two-dimensional detector, this scattering corresponds to a curved section through reciprocal space. There is much to be gained from an interpretation and analysis point of view by having data from plane sections of reciprocal space.

Estermann and coworkers[1] have achieved this by collecting large numbers of curved 2D sections of data as the crystal orientation is varied systematically, with subsequent retrieval of plane sections from the resulting full 3D data set. Such a strategy involves an enormous quantity of data, only a small fraction of which is ever likely to be used. In our home laboratory, we use a flat-cone Weissenberg geometry technique that allows a desired plane section of data to be recorded directly,[2] and, in a previous study,[3] we have adapted this method for use on the 1-ID beamline.

In that preliminary experiment, it was found that, using the smallest feasible Weissenberg slit width of 0.5 mm, while good resolution was obtained in the direction of the diffraction angle, ξ, insufficient resolution in the direction corresponding to the crystal rotation, ω, was obtained. One of the prime aims in the present experiments was to devise modifications of the method to achieve improved resolution in ω.

Methods and Materials

The samples on which experiments were carried out were an yttria-stabilized zirconia (Y-CSZ) of composition $Zr_{0.61}Y_{0.39}O_{1.805}$, a calcia-stabilized zirconia (Ca-CSZ) of composition $Zr_{0.875}Ca_{0.125}O_{1.875}$ and the nonstoichiometric iron oxide wüstite, $Fe_{1-x}O$.

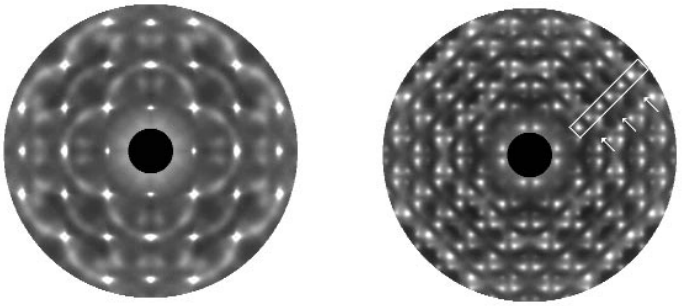
Improvement in the resolution in ω was obtained by performing exposures of the stationary crystal at incremental steps of 0.25° in ω. With a 0.5 mm slit placed in front of (and close to) the recording plate, a single exposure resulted in a 0.5-mm-wide line of data on the image plate. Before the next exposure, the image plate was translated normal to the slit length by 0.5 mm so that each exposure resulted in an independent record with (virtually) no overlap. On one image plate, 300 such exposures were recorded over a total width of 150 mm corresponding to a rotation of 75.0° in ω. To record the total 360° of ω rotation, five image plates were required.

Each exposure was made for a time in which a constant number of counts was monitored in the primary beam. This time was ~5 seconds on average but varied with the state of the ring current by up to a factor of two. For a complete section of data, ~2 hours was required. In all, eleven layers of data were collected for Ca-CSZ, eight for Y-CSZ, and five for wüstite.

Results

The spatial resolution of the recorded data was much improved on that obtained in the prototype experiment[3] and now exceeds that available from our own laboratory-based experiments. Figure 1 shows two example sections of the data for the Ca-CSZ sample.

The diffuse peaks that are seen extending in rows parallel to [110] in Fig. 1b (indicated by the white rectangle) and which occur in pairs straddling the dark lines (indicated by arrows) are of prime importance in understanding the disorder in CSZs. They originate from the local relaxations of the cations around the vacancy defects in the oxygen array. For Y-CSZ, the peak on the high-angle side of the dark line is invariably more intense than that on the low-angle side, showing the majority of this asymmetric scattering must originate from a third-order term in the basic scattering equation,[4] because Y and Zr have very similar scattering factors. In the Ca-CSZ pattern shown, it is clear that for the first pair of peaks the lower-angle peak is the more intense, for the second pair the intensities are fairly similar, while for the third pair the peak on the high-angle side is the more intense.



(a) (b)

FIG. 1. Diffuse scattering patterns for (a) the 0.05c and (b) the 0.5c* sections of Ca-CSZ in the relatively low scattering-angle range $\sin\theta/\lambda \leq 0.67$.*

Discussion

The present results indicate that the method is a viable means of extracting high-quality 2D plane sections of diffuse scattering data from disordered crystals and is particularly useful for highly absorbing materials.

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