Application of X-ray Synchrotron Topography to in situ Studies of Recrystallization

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It is demonstrated that X-ray topography with synchrotron radiation provides an excellent means of simultaneously measuring the size and orientation of grains in polycrystalline materials. The technique, which also provides data on the lattice strains, is particularly suited to studies of grain growth at high temperatures and preliminary studies of recrystallization in iron–silicon alloy sheet are reported. The initial radial-growth rate is constant and approximately equal for all grains studied in one recrystallization.

1. Introduction

X-ray topography with synchrotron radiation is now well established as an important experimental technique for rapid assessment of crystal perfection (Tanner, 1977). It has great potential for studying time-dependent changes in the microstructure of single crystals (Bordas, Glazer & Hauser, 1975; Tanner, Safa, Midgley & Bordas, 1976; Safa & Tanner, 1977). The topographic technique used in these experiments was originally developed by Guinier & Tennevin (1949) using the *Bremsstrahlung* from a conventional X-ray generator and is closely related to the transmission Laue technique for crystal orientation. In their original article, Guinier & Tennevin (1949) demonstrated that in a polycrystalline material each grain produced its own set of Laue topographs. The grain size could be measured from the dimensions of the image and the misorientation between grains determined from the coordinates of the image on the film. Similarly, regions with a different crystal structure will give their own characteristic set of Laue topographs, and this has recently been exploited by Steinberger, Bordas & Kalman (1977) in a study of the polytypes in ZnS. These authors used synchrotron radiation and were able to study changes in the polytypes as a function of temperature. In this communication we report the application of synchrotron radiation to the original example of a polycrystalline, single-phase material.

The kinetics of recrystallization of a cold-worked metal have been studied by Mehl and co-workers by annealing numerous equivalent samples for increasing time periods and examining the statistical distribution of grain sizes in the quenched material. For example, Stanley & Mehl (1942) studied the kinetics of Fe–1% Si, measuring the area of material recrystallized with a planimeter, whilst Decker & Harker (1950), working with rolled copper, determined the proportion of the recrystallized material by an X-ray technique developed by Decker, Asp & Harker (1948). These statistical experiments are not only tedious to perform but also require the questionable assumption that the grain-growth rate is the same for all grains. Clearly, it would be advantageous to be able to observe the growth of individual recrystallized grains directly during the heat treatment. We describe here how such experiments can be carried out with X-ray synchrotron topography.

2. Measurement of grain size and orientation

Just as in the laboratory-based Laue technique, each grain in a polycrystalline foil produces its own Laue pattern of diffracted beams. However, because the synchrotron radiation beam is of large area at the sample whilst the source is small and situated some 50 m away, each Laue ‘spot’ becomes a topograph of the grain. Within the topograph, contrast arising from the strains associated with lattice defects can be observed. For transmission work with a polycrystalline foil, the beam passes normally through the specimen and a film placed behind the specimen records the Laue topographs for all grains in the beam area. The exact orientation of the foil is not critical.

Fig. 1(a) shows an example of the images obtained from a commercial sheet of recrystallized silicon steel. The dimensions of each image, corrected for the geometrical image distortion, correspond directly to the grain size, and different reflections for the same grain can be identified from the grain shape. In this example the boundaries of the images are quite sharp, indicating a discrete mosaic block structure with little strain within each grain. There is, however, a large spread in the orientation of the grains.

It has been demonstrated that the geometrical resolution obtainable in synchrotron topographs is typically a few μm with a specimen to plate distance of several cm (Hart, 1975; Tanner, Midgley & Safa, 1977). Thus it is possible to make accurate assessment of the grain size of much finer-grained material: Fig. 1(b) shows an example of the images from an iron foil whose average grain size was measured optically to be about 50 μm. There is more strain associated with this material, as evidenced by the streaking of some images normal to the Bragg planes. The technique has also been used successfully in back reflection to measure...
the grain size in samples of recrystallized niobium. However, it is when the specimen is in a hostile environment and standard metallographic techniques become inapplicable that synchrotron topography has most to offer.

3. **In situ** recrystallization experiments on iron–silicon

3.1. Apparatus and experimental techniques

The sample, $25 \times 20$ mm in superficial area, was cut from a 370 $\mu$m thick sheet of commercial Fe–3.5\% Si, cold rolled so that the thickness was reduced by 85\%. It was supported vertically in a graphite holder inside a long alumina tube filled with high-purity argon. The alumina tube was inserted in a resistance-heated furnace which allowed the specimen temperature to be maintained at around 1000 °C. The synchrotron radiation beam entered the alumina tube through a mylar window at the cold end and passed along the length of the tube. Diffracted beams emerged through the closed end wall of the alumina tube and were recorded on Polaroid film situated 200 mm from the sample. The alumina produced a halo around the direct beam but images of individual grains were not observed, the speckle on the micrographs being due to emulsion grains. The film was cooled with a stream of cold air as the heat radiated from the furnace was substantial. A little care was necessary in arranging the furnace geometry so that the angle of divergence of the emerging X-ray beams was sufficient to enable a reasonable fraction of the Laue pattern to be recorded. The arrangement is shown schematically in Fig. 2.

Once the furnace had reached 1000 °C (after about 100 min), micrographs were taken sequentially. Exposure times of 30 s were adequate but, because of the beam-area search procedure in operation in the Synchrotron Radiation Facility at Daresbury Laboratory, a total interval of 2 min between each data point was unavoidable.

3.2. Results

Rather conveniently, no diffraction pattern was obtained from the sample initially, because of the large amount of deformation introduced by the cold rolling. In run 1, 31 min elapsed after reaching 1000 °C before any recrystallized grains appeared on the film (Fig. 3). They then continued to grow for 18 min before all had impinged on one another and recrystallization ceased. We note, Fig. 3(b) and (c), that the incubation period for all the grains covered by the beam area was very similar, all grains appearing after 31–33 min. We note that there is very little strain in the recrystallized grains at any stage during grain growth, and the resulting grains are large and display a small spread around a preferred direction.

Precise measurements of growth rates are complicated by the fact that images of different grains tend to overlap. However, images of several grains could be easily identified throughout the sequence. The square root of the grain area, which is a fair measure of the grain diameter of the irregular grains, is plotted as function of time in Fig. 4. At small grain sizes, the rate of radial growth is approximately constant and nearly equal for all five grains, but as the grains begin to impinge, the growth velocity falls gradually to zero.

![Fig. 1. (a) Laue topographs of grains in recrystallized iron–silicon. Images of a selected grain are indicated with arrows. (b) Laue topographs of small-grained iron foil. Recorded on 10 $\mu$m thick Ilford L4 nuclear emulsions. Exposure 14 min, 4 GeV, 15 mA.](image)

![Fig. 2. Schematic diagram of apparatus.](image)
Fig. 3. Sequence of micrographs showing grain growth during recrystallization. After (a) 25 (b) 31 (c) 35 (d) 37 (e) 43 (f) 49 min at 1000°C. Recorded on Polaroid 4 × 5 Land Film, type 52; 5 GeV, 7 mA, 30 s exposure.

The general shape of the curve is very similar to that obtained for Fe–1% Si (Stanley & Mehl, 1942). Values of the initial growth velocity, final grain size and relative orientation are given in Table 1. P. Spychal (private communication) reports a rate of change of diameter of 5 μm s⁻¹ for similar material at 1000°C, using a standard metallographic and statistical technique.

This is very close to our measured value for run 1. Unfortunately, with the unsophisticated furnace used here, we were unable to reproduce the results.

Table 1. Recrystallization data at a nominal temperature of 1000°C

<table>
<thead>
<tr>
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<th>Run 1</th>
<th>Run 2</th>
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<tbody>
<tr>
<td>Initial rate of change of diameter</td>
<td>5.5 ± 0.3 μm s⁻¹</td>
<td>—</td>
</tr>
<tr>
<td>Average final grain area</td>
<td>15 ± 2 mm²</td>
<td>10 ± 4 mm²</td>
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<tr>
<td>Orientation spread of grains</td>
<td>3°</td>
<td>&gt; 7°</td>
</tr>
<tr>
<td>Lattice strain within grains</td>
<td>Small</td>
<td>Small</td>
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</tbody>
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Fig. 4. Grain dimension as a function of time for run 1.
on successive runs. For example, in run 2, at the same nominal temperature of 1000°C, the recrystallization commenced almost immediately after reaching the maximum temperature and was complete after only 8 min. The recrystallized grains were much smaller and more widely distributed in orientation. We assume that the temperature in the second run was somewhat higher than in the first. It was found that a temperature gradient of 5°C mm⁻¹ existed at the position of the sample and hence small changes in the location of sample and thermocouple could have resulted in a significant temperature difference.

4. Discussion and conclusions
To the authors' knowledge there has been only one previous successful attempt to study crystal growth in situ with X-ray topographic techniques. Chikawa (1974) used a 60 kW rotating-anode generator and a PbO vidicon fitted with a beryllium window to view Lang topographs directly. He observed the generation and annihilation of dislocations as a molten zone was formed and moved in a silicon single crystal. The experiments reported in this paper represent the first in situ X-ray topographic study of crystal growth under normal growth conditions. As has been demonstrated (Tanner, Safa & Midgley, 1977), because of the high intensity of synchrotron radiation and because a good geometrical resolution can be obtained with the photographic plate a considerable distance from the sample, one can perform experiments impossible with a conventional X-ray generator. In situ crystal-growth experiments are potentially some of the most exciting.

We hesitate to attach too much significance to the exact numerical values of the recrystallization parameters displayed in Table 1 because of the uncertainty in the temperature measurement. For complete data, it is essential to construct a furnace in which the specimen is directly heated by the passage of an electric current and in which thermocouples are welded directly to the specimen. Such an arrangement will also eliminate the troublesome warm-up period. However, we have demonstrated that X-ray synchrotron topography can be used for studies of grain size and orientation at high temperature and that the data obtained is in accord with the previous experimental studies of recrystallization. We are convinced that the technique has a wide range of applicability.

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References