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Addition of EBSP equipment to the Studsvik HCL SEM

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ABSTRACT

The principles of and the components necessary for obtaining electron backscattering patterns (EBSP) are outlined. The information obtained by the EBSP method is discussed. Applications in the nuclear fuel field include texture in Zircaloy, phase identification and characterisation of high burnup structure in UO_2. The computerised measurements and presentation of results are discussed.

The EBSP method can obtain information on the grain lattice that is not readily available by any other method. The method fills a gap between X-ray diffraction and transmission electron microscopy.

By moving the beam and observing the pattern it is possible to measure the extension of different grains. When the beam moves into a grain with a different orientation, the pattern changes. The resolution in orientation is approximately 0,5°.

By making 150x150 measurements in a grid of 25 µm side length it is possible to create a map showing several properties related to the lattice itself. The first is the orientation of the individual grain that can be presented as a grain map where each orientation is represented by a specific colour. The second property that is presented is the pattern contrast, called the "band contrast" which is a measure of the disturbance of the lattice in that spot. This property is sensitive to cold deformation, grain boundaries, pores or other phenomena that change the lattice symmetry or size. It is possible to make calculations in order to find e.g. mismatch orientations of different types. If the orientation changes between two measurements more than 10° it can be considered a grain boundary. These mismatch positions can be collected into a map that shows the grain boundaries. Other, more complicated relations can also be calculated.

A sample of UO_2 fuel irradiated to a burnup of 67 MWd/kgU was examined. The fuel had developed a high burnup structure near the periphery, gradually vanishing towards the centre. It was demonstrated that the EBSP method was able to characterise the extent of the sub-grain formation at a given radial position.

1 INTRODUCTION

The scanning electron microscope located at the Studsvik Hot Cell Laboratory is mainly used for fuel and cladding investigations. The secondary electron image mode is used for general characterisation of surfaces. The backscatter electron image mode provides information on porosity, composition and grain orientation. It is used to e.g. characterise the grain structure of UO_2 or the hydride structure in cladding materials.

By using EDS and WDS a versatile analytical capability is added to the instrument. The WDS is used for e.g. detailed study of the fission products in UO_2 and the EDS for characterisation of the cladding inside.

However, some of the significant changes in the microstructure that occur during normal irradiation or under defect conditions can not be characterised by any of the above mentioned methods. By using diffraction information from backscattered electrons the orientation of the crystal lattice within a grain can be measured. Furthermore, information on the quality of the lattice in this point can be obtained. By using modern computer methods many such measurements can be performed and analysis of the whole group of data provides further information on the relation between different areas.

The method is well suited to investigate e.g. texture in Zircaloy, grain growth and grain sub-division in UO_2 or to identify unknown phases with a composition similar to the matrix. Other tasks such as measurement of grain size can of course also be carried out.

In a first attempt to evaluate the method an investigation on high burnup UO_2 fuel was carried out [1].

High burnup UO_2 fuel exhibits the formation of a high burnup structure (HBS) above a local burnup of 40-50 MWd/kgU. This structure is characterised by the formation of small pores and the sub division of the grains into smaller units. The structure starts to form at some grain boundaries near the periphery where the burnup reaches a maximum. The structure forms further and further into the pellet as the burnup increases.

The present report deals with the implementation of the EBSP method and its ability to characterise the extent of the high burnup structure in a high burnup UO_2 sample.

2 EXPERIMENTAL

2.1 The EBSP method

The electron backscattering pattern (EBSP) method can obtain information on the grain lattice that is not readily available by any other method. The competing methods are X-ray diffraction and transmission electron microscopy (TEM). The first method gains information from a relatively large area/volume whereas the TEM always looks at very small samples. The EBSP can cover the range in between (and overlapping) the two methods.

The electron backscattering pattern (EBSP) is a pattern formed on a phosphor screen located near a highly tilted specimen that is bombarded by electrons, see Figure 1. The electrons are diffracted inside the specimen crystal and escape from the crystal with varying intensities in different planes. This variation in intensity forms a pattern similar to a Kikuchi pattern on the phosphor screen. The screen is observed by a CCD TV camera and the image is improved by a background subtraction before it is transferred to the computer. The phosphor screen and the camera are built in one unit and are attached to the JEOL 840 SEM by using the microscope port. The computer automatically interprets the image by detecting a number of diffraction "bands". These bands are compared with a simulated set of bands by using an assumed crystal type and lattice spacing. The simulation with the smallest mismatch (mean angular deviation) between the detected bands and the best fitting simulated crystal orientation is used. The location of the point, the contrast in the image (band contrast), the orientation of the crystal (three angles) and the mean angle deviation (MAD) are stored.

The band contrast, which can be imagined as the standard deviation of the grey scale in the pattern, is a measure of the perfection of the crystal lattice that causes the diffraction. A polishing sequence that leaves a cold deformed layer causes a low band contrast (blurred pattern). A lattice strain gradient causes the pattern to become more blurred also. Another factor that can cause a weak contrast is the occurrence of two patterns at the same beam position. This is the case on a grain boundary and more frequently in the areas of a high burnup fuel that is subjected to sub-grain formation.

The orientation of the grain is defined by the three Euler angles [2]. In the present case the co-ordinate system is chosen arbitrarily. However, it is very easy to relate the orientation co-ordinate system to any desired co-ordinate system e.g. the rolling, transversal and normal directions in plate a material.

In a mapping operation a grid of measurements are performed, typically between 10000 and 25000 measurements. The grid size can be chosen within wide limits. In the present case with a grain size of the order of 5-8 μm. A grid size of 25 μm including 150x150 measurements was chosen. If a measurement fails to meet certain criteria the measurement is rejected and called a zero solution. The most important criterion is the mean angular deviation. If the MAD is to high it means that the solution is poor and perhaps false. The second most important factor is band contrast which is a measure of the pattern quality. A very low band contrast almost always means that the measurement takes place in a pore. If it is a bit higher it may be that the beam spot is located on a grain (or sub-grain) boundary and reflections from both lattices are forming the (double) pattern.

The image formed by using the band contrast shows the areas with a high lattice disturbance, notably cold deformation, the grain boundaries and the zones with subgrains.

A convenient way of representing the orientation is to assign each of the RGB colours to one of the three Euler angles. Each colour represents a unique orientation. This means that all measurements within one grain will have the same colour, since all the measurements have approximately the same orientation. A small problem is encountered when representing orientations near the 0-360° for the Euler 1 and 0-90° transition for the Euler 2 and 3 angles. The image formed by using such a colour scheme shows an image with grain field contrast, i.e. an image which is similar to the image formed by electron channelling contrast (also caused by diffraction).

By comparing the orientation of two (neighbouring) measurements a map of misorientations can be found. If the misorientation is above a certain level it can be considered to be a grain boundary. By using appropriate threshold levels, high-angle, low-angle and twin grain boundaries can be outlined.

By making measurements at random positions on a sample and collecting the results into a pole figure any non-randomness can be found. The pole figure is commonly used for describing the texture of highly textured materials such as Zircaloy and aluminium.

2.2 Specimen preparation

The specimen used was originally prepared for SEM/EPMA analysis. The surface was covered by a thin layer of gold using an Emithech gold sputter apparatus. According to the manufacturer the gold layer should be around 4 nm thick at the current settings. The EBSP method usually demands an uncoated surface so a careful manual polishing of the specimen was performed using colloidal silica. The polishing is not thought to have removed the coating completely. An accelerating voltage of 20 kV was used since it was not possible to operate the instrument at low accelerating voltages, due to the remaining coating. A specimen tilt of 70° was used. After calibrating the system, the beam control mode was used performing measurements in a grid pattern of 100x100 or 150x150 points at 0,2 μm spacing.

3 RESULTS AND DISCUSSION

The patterns obtained on UO_2 exhibit a very high contrast. The areas containing sub-grains can easily be identified using the EBSP. In these areas, the grain size is of the same order as or smaller than the beam spot size (actually: beam interaction volume). Two patterns are visible simultaneously on the phosphor screen, which means that it is not possible to automatically index the patterns. In the automatic measurement these points show up as not indexed and having a relatively low band contrast. Pores are also not indexed, but have an even lower band contrast (close to 0). The orientation of the sub grains appear to be completely random.

The maps of band contrast and the true Euler angle can be seen in Figure 2a and 2b. In Figure 2c the grain boundaries have been identified by the program. In Figure 2d the conditions for two types of grain boundaries are set up: i) A normal grain boundary with $>10^\circ$ mismatch between two adjacent measurement spots. This grain boundary is coloured yellow. ii) "Sub-grain boundary" with a mismatch between two adjacent measurements in the range 1 to 10°. These boundaries are coloured white. The orientation resolution of the system is of the order of $0{,}5^\circ$ which means that differences less than $0{,}5^\circ$ should not be considered.

In parts of the specimen where the sub grain formation has not yet started, the lattice is strained but the orientation remains approximately the same, see Figures 2c. Points measured in these areas show up as having the same or only slightly distorted orientation as the rest of the grain it belongs to and a band contrast that is decreasing towards the grain edge. The difference in orientation between positions on one side of a grain to the other can be as large as 6°.

Automatic measurements were performed at 0,34 and 1 mm from the periphery. At the map from 0,34 mm from the periphery not many "normal" grain boundaries can be found. The sub grains have caused the band contrast to be quite low and the automatic indexing has not been successful in large parts. Further into the fuel pellet, at 1 mm from the periphery, the extent of the sub grain formation is not so severe, see Figure 2. It can also be seen that the sub grain formation starts at some grain boundaries, while others are left unaffected.

4 CONCLUSIONS

The EBSP method includes the following features:

- The orientation of the grains can be directly measured.
- Qualitative information on the strain gradient in the lattice can also be obtained by the "band contrast" parameter.
- Maps of orientation and pattern quality information can be used to correlate lattice parameters to specific locations in the microstructure.
- Grain size and grain boundary type can be investigated by the method.

The EBSP is capable of characterising the UO_2 high burnup structure (HBS) in the following aspects:

- The EBSP method is capable of identifying the sub-grains.
- At 0,34 mm from the periphery of the analysed sample the HBS formation is only partial. However, almost all grain boundaries are affected
- At 1 mm from the periphery the HBS of the analysed sample has only developed on certain grain boundaries. It is estimated that approximately 50 % of the grain boundaries are affected.

REFERENCES

[1] BENGTSSON, S., Examination of sub-grain formation in high burnup UO_2 fuel using the EBSP method, Presented at the IAEA Technical Committee Meeting on Advances in Pellet Technology for Improved Performance at High Burnup, Tokyo, 28 October - 1 November, 1996.

[2] Preferred Orientation in Deformed Metals and Rocks: an Introduction to Modern Texture Analysis, ed. H.R. Wenk, Academic Press Inc. (London), (1985), ISBN 0-12-744020-8, p79.

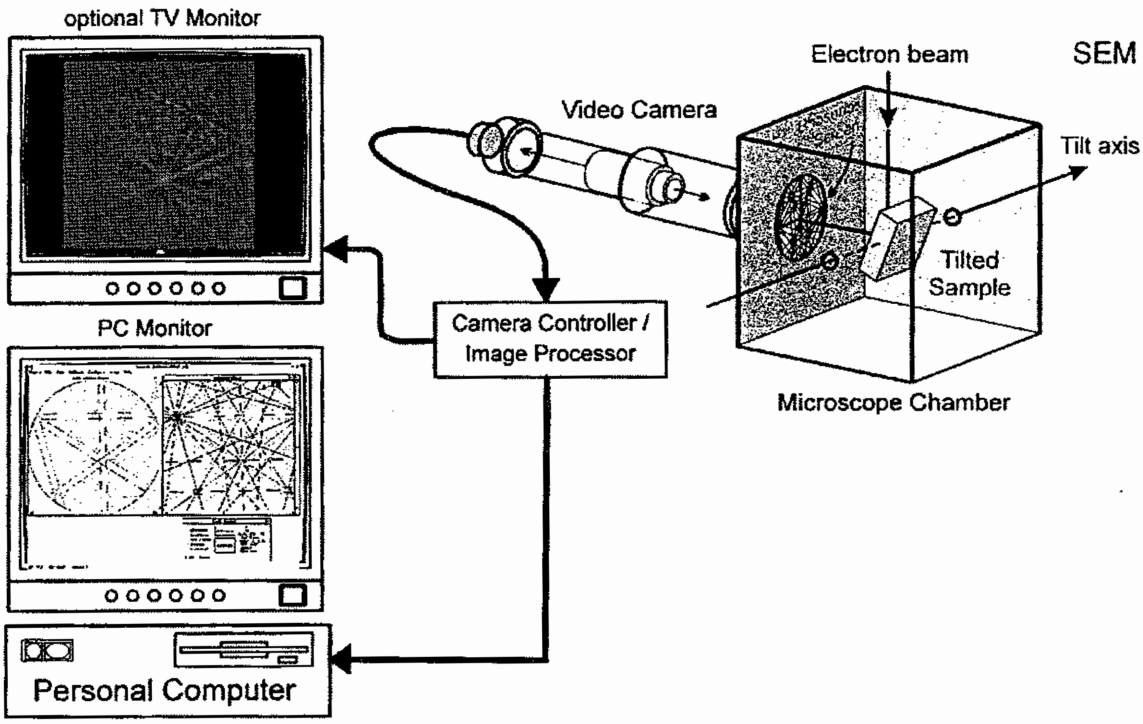
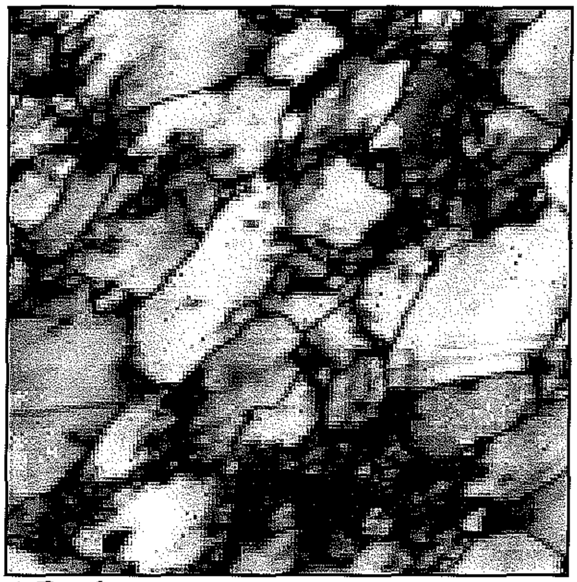
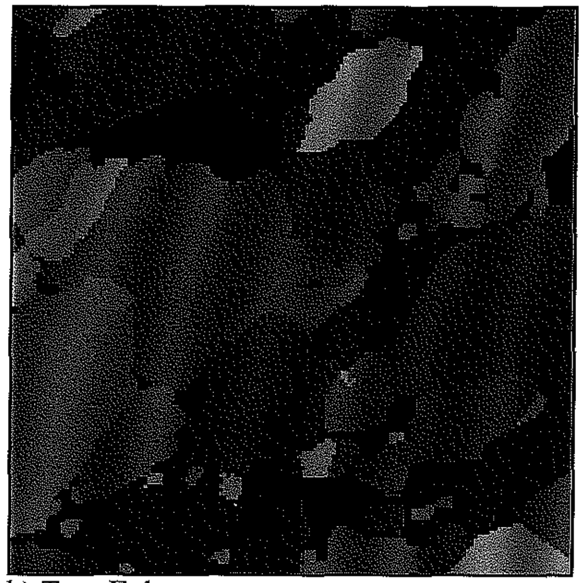


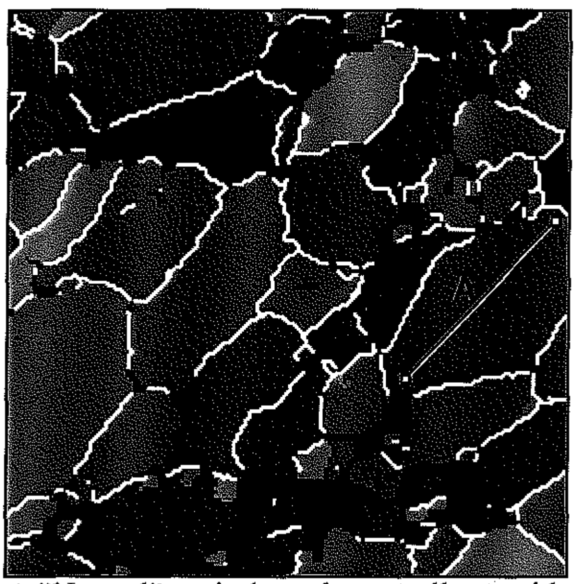
Figure 1. Components and schematic layout of the EBSP system.



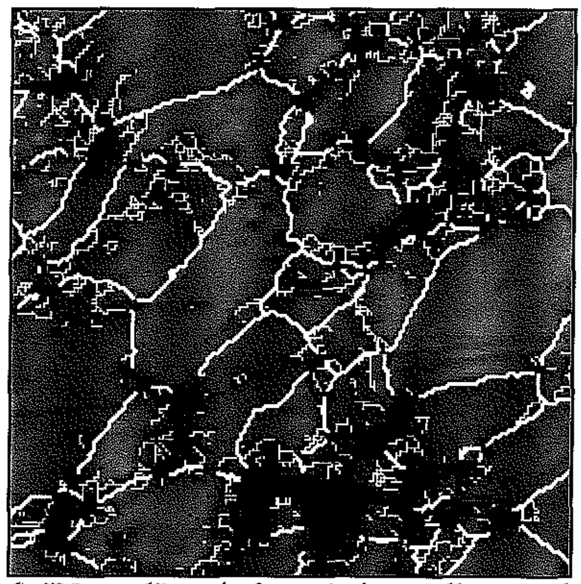
a) Band contrast map.



b) True Euler



c) "Normal" grain boundary (yellow) with >10° mismatch between two adjacent measurement points. A 6° orientation mismatch between two sides of the same grain is indicated at "A".



d) "Normal" grain boundaries (yellow) and "Sub-grain" boundaries (white) with a mismatch between adjacent measurements in the range 1 and 10°.

Figure 2. Electron backscattering pattern measurement maps. Measurements where the indexing failed are colored black in maps b to d. Image side length is 25 μm. Irradiated UO_2 specimen (pellet average burnup of 67 MWd/kgU) located approximately 1 mm from the pellet periphery.