Installation of a Scanning Electron Microscope in the Hot-cell Laboratory of NRG Petten

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Abstract. In 2010 a new scanning electron microscope (SEM), equipped with several detectors (EDS, WDS and EBSD) is installed in a new hot cell. The SEM is modified for use in a radioactive environment. Therefore the irradiation sensitive parts are removed or protected. In addition changes have been made to the SEM to allow remote handling and to allow maintenance of the important parts. This paper describes the new facility at the NRG hot cell laboratories and gives some examples of the first microscopy results.

1. INTRODUCTION

The Hot Cell Laboratories (HCL) of the Nuclear Research and consultancy Group (NRG) are equipped with both glove boxes and hot-cells to perform research on structural materials and nuclear fuels. This research covers non-destructive testing, testing of physical and mechanical properties and microstructural investigations. This complete package of pre- and post-irradiation testing in combination with the High Flux Reactor (HFR) allows irradiation testing at one location, avoiding complex and costly transports.

Recently NRG improved the microscopy facilities by installing a new scanning electron microscope (SEM). Besides the standard secondary electron (SE) and Back-scatter Electron detectors, this SEM is equipped with additional detectors for energy dispersive spectrometry (EDS), wavelength dispersive spectrometry (WDS) and electron back-scatter diffraction (EBSD). The equipment is placed in alpha tight hot-cell to allow analysing radioactive samples emitting α, γ and β radiation, therefore both nuclear structural materials and nuclear fuels can be analysed. The new equipment did not fit in the existing hot-cell of the previous SEM. As a result the hot-cell is dismantled, the old SEM removed and a new hot-cell engineered and built. This paper describes the equipment, modifications for using the SEM in a radioactive environment, the installation of this equipment and gives some examples of the first results.

2. EQUIPMENT

2.1. Microscope and detectors

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In 2005 and 2006 several SEM suppliers were contacted to have discussions on specifications, possibilities of adding extra detectors and possibilities to install the equipment in hot cell to allow analysing radioactive samples.

In 2007 it was decided to buy a Jeol JSM-6490 SEM, equipped with Oxford EDS/WDS/EBSD detectors. The SEM has a Tungsten filament and an acceleration voltage between 0.3–30 keV. It can be operated in both low and high vacuum mode. Low vacuum mode is useful for bad conducting samples, the resolution of 3.0 nm is reached in the high vacuum mode at 30 keV. Fig. 2.1 gives of summary of the equipment.

2.2. Modifications to the SEM for use in radioactive environment

The purpose of this SEM is to analyse radioactive samples, emitting γ/β radiation, as well as α radiation. To protect the operators and environment the SEM is placed in alpha tight box that itself is placed in a hot cell with lead walls. During operation the pressure in the box is lower than the surrounding, to keep the radioactive material in the box.

Besides the operators and environment, the SEM has to be protected as well. The parts of the equipment that are sensitive for radiation damage (electronics) were positioned outside the hot cell, if possible. This means that most cables are extended to allow separation of electronics and vacuum chamber. Vacuum tight connectors are used to lead the cables through the glove box wall to prevent leakage of potential radioactive air.

The vacuum chamber is placed, together with the detectors, inside the hot-cell. Electronics and equipment that was possible to separate was placed outside the hot-cell. This concerns electronics of the SEM (placed under the hot-cell), controllers and vacuum pumps (placed next the hot-cell) and operating equipment (placed at some distance from the SEM). Fig. 2.2 shows a drawing of the SEM and hot-cell. The green unit below the cell contains the electronics that is separated from the cell.

Some sensitive parts could not be placed outside the hot-cell, for instance the detectors and motor stage. Therefore local shielding by Tungsten is applied near the sample or near the sensitive equipment, considering the limited space available for shielding, and without compromising the functionality of the microscope.

The electronics of the motor stage is very close to the sample position and in this region of the SEM limited volume is available to add tungsten shielding. These parts are unavoidably exposed to γ radiation, in spite of the Tungsten shielding that is applied where possible. To investigate the sensitivity of the electronics of the motor stage parts of the same type are deliberately exposed to gamma radiation in the HFR. The results showed that the electronics can withstand enough γ radiation (at 250 Gy no degradation in resistance values, at 2500 Gy minor degradation in resistance values), to be operated for a significant period. However, because only one device is tested, we anticipated that during the life time of the SEM it may happen that this motor stage may fail. A plan is in place to repair or exchange the motor stage when this will happen.

Besides the radiological protective measures, a lot of effort is taken on the handling of the sample in the hot cell. This handling is limited by the fact that the cell is equipped with tong manipulators (manoeuvrable rods with a handle outside the hot-cell, that the operator can use to position and tweak the grips at the end of the rod in the hot-cell). These tongs are restricted in their movements with respect to the more common master/slave power manipulators. The hot-cell has two tong manipulators (Fig. 2.2). One tong is positioned opposite to the posting port that is located between the hot-cell of the SEM and the adjacent hot-cell (in this second cell the sample is placed in the sample holder). This tong is used for getting the sample holder (including the sample) from that cell to the SEM hot-cell.

	JEOL 6490 LV SEM • Placed in hot-cell for radioactive specimens. • High resolution (3.0 nm). • Possibility of low vacuum mode for badly conducting surfaces. • Enhanced SE and BSE detection. • Five-axis motorised stage. • Equipped with EDS, WDS and EBSD detectors.
	Oxford Instruments energy-dispersive spectrometer (EDS) • Able to operate with high count rates. • Accurate element identification. • Accurate element quantification. • Superior light element analysis. • Fast mapping.
	Oxford Instruments wavelength dispersive spectrometer (WDS) • Trace analysis possible due to high sensitivity (0.01 wt%). • Excellent energy resolution. • Quantitative elemental analysis. • Five crystals (detection from Be to Pu). • Extra shielding for radioactive specimens.
	HKL Nordlyss electron back-scatter diffraction (EBSD) • Phase analysis and identification. • Texture analysis. • Can be combined with EDS. • Indexed mapping at speeds up to 100 Hz.

FIG. 2.1. Summary of description of the scanning electron microscope with the installed detectors.

The second tong is positioned in front of the door of the vacuum chamber. The door of the SEM vacuum chamber is redesigned to be able to open and close the door by using this tong. Opening and closing is done vertically, by using a counterweight. The same tong manipulator is used for placing the sample holder in the vacuum chamber by a specially engineered loading system.

Another limitation is the lack of space in the hot-cell, because the SEM and its detectors are filling up most of the work space. To ensure good visibility the windows are therefore placed under an angle. Another important item is accessibility of the system in case of servicing, replacing the filament and repairing or removing detectors etc. The glove box is equipped with openings for gloves on all necessary locations (Fig. 2.2). The access to the electronics is provided by the relocation of parts that need to be serviced to the side of the electronics unit that can easily be reached.

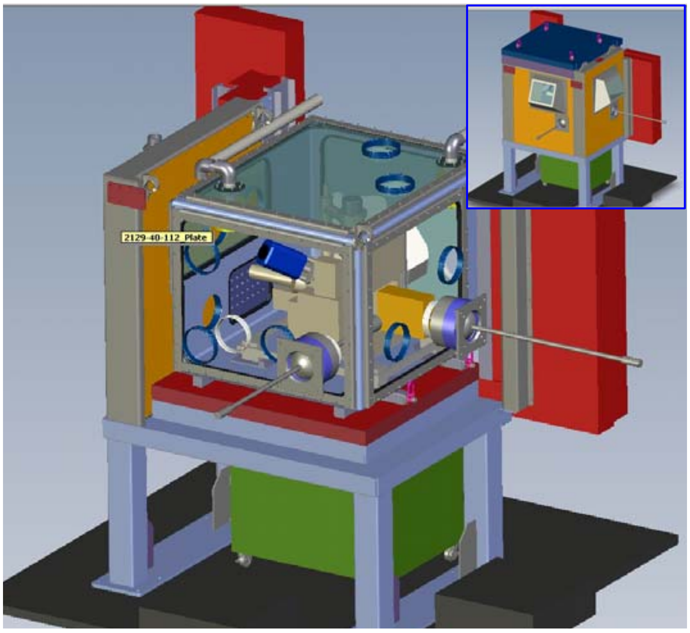

FIG. 2.2. Drawing of the hot-cell (without two lead walls and roof), showing the part of the SEM inside the glove box and the separated electronics unit under the cell (green). The positions of the openings for gloves are visible in this figure as well. The insert shows the complete hot-cell with special lead windows and two tong manipulators.

3. BUILDING THE FACILITY

The situation at the start of the project was a contaminated hot-cell (F6 cell) with the old SEM. This hot-cell was connected to a contaminated cell (F5 cell) that was connected to the other cells used for sample preparation. Building of the new facility was carried out in different phases:

Phase 1: Removing old SEM and hot-cell

All activity from the old hot-cell was removed. When the activity in the hot-cell was low enough, a lead wall was removed to have better access to the glove box. The glove box is further decontaminated by using the gloves. After this the lead walls were dismantled (Fig. 3.1). The glove box and SEM were transported to the decontamination facilities of NRG, where the box and equipment is decontaminated. Parts that could not be decontaminated were treated as waste. The old F6 cell was placed on a concrete block. For the new set-up this space is needed for the electronics, as a result this block was removed. A maquette of the SEM and detectors was made, at the same size of the original SEM. After the old hot-cell was removed this maquette was placed in the same position of the new SEM. This maquette was used for the design of the new hot-cell (determining sizes, position of tongs, windows, openings for gloves and position of posting port).

Phase 2: Modification of F5 hot-cell

The new F6 cell is connected to the existing F5 cell. To achieve this in the new situation a posting lock needed to be designed and build. Therefore the F5 cell needed to be decontaminated and modified as well. This opportunity is taken to remove old equipment (that is not being used anymore) from this hot-cell. Size and position of the lead window and tong positions of this cell were optimised for the new situation as well.

Phase 3: Building the new hot-cell

During the manufacturing of the hot-cell the modified SEM is tested in another lab at NRG. Due to the modifications of the electronics some measures had to be taken to reach the specifications of the microscope. This time was also used to train the operators and to develop the final modifications in local shielding and handling tools.

FIG. 3.1. Dismantling the old hot-cell.

The hot-cell was built in the HCL after all its parts were available, initially without equipment. After acceptance test of the hot-cell the SEM is placed (Fig. 3.2) and tested. After a period of optimising the performance of the SEM a final acceptance test was performed. After this milestone the hot-cell was connected to the other cells, off-gas system and HCL alarm system. In August 2010 the SEM was ready for analyses on radioactive samples (Fig. 3.3).

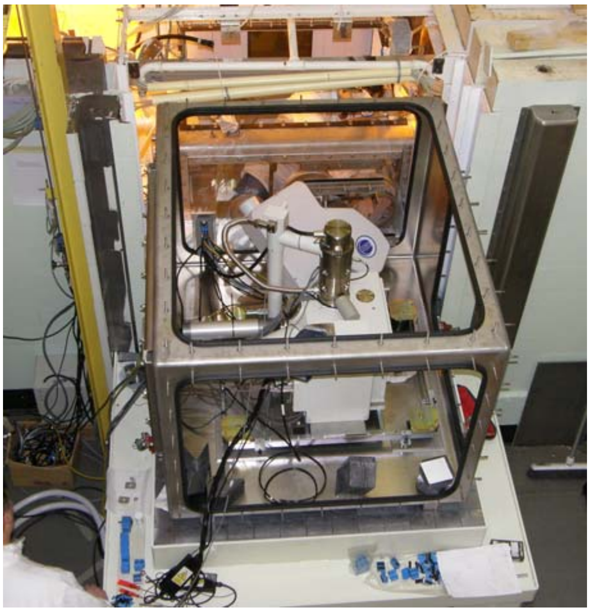

FIG. 3.2. Installation of the SEM in the hot-cell.

FIG. 3.3. Hot-cell with SEM. One lead wall and the roof is removed to show the equipment.

4. EXAMPLES OF FIRST RESULTS

Since August 2010 several projects on irradiated samples are carried out. The section gives some examples.

Irradiated Fuel pebbles

In the European HTR (High Temperature Reactor) programmes HTR-F and RAPHAEL-IP an irradiation experiment (HFREU1BIS) is carried out on HTR fuel pebbles [1,2]. In the post irradiation examinations the pebbles are analysed with the new SEM. Fig. 4.1 gives an overview and detail by SEM-imaging of one HTR TRISO fuel particle from an HTR fuel pebble, after irradiation. UO_2 kernel, buffer, inner Pyrocarbon, SiC and outer Pyrocarbon layers and surrounding graphite matrix can be clearly distinguished. The images show fission gas bubble formation at the grain boundaries, metallic fission product precipitates, and a peculiar kernel surface, indicating fission gas release. On these samples many WDS measurements are performed to get information on the present elements. Fig. 4.2 shows an example of a Cs mapping in the layers around UO_2 kernel. The red dots indicate presence of Cs.

Irradiated SiC/SiC composites

Different material types were irradiated in the HFR within the EXTREMAT Integrated Project (6^{th} Framework programme EU). The aim of this project was to investigate materials in extreme environments like intense neutron fields. Carbon Fibre Reinforced Ceramics (SiC/SiC, SiC/C, C/C) are envisaged to be used as structural or functional materials in high temperature applications, i.e. up to about 1100–2000°C in fusion reactors and up to about 850–1500°C in advanced fission reactors, respectively. These materials were, among other materials, investigated in the EXTREMAT project. The SEM is an excellent good tool to study fracture surfaces and this is valuable information to the strength test results. Figs 4.3–4.4 show two examples of fracture surfaces of SiC/SiC samples, fractured in four point bending tests, after irradiation in the HFR.

EBSD

In the first period of the SEM the EDS and WDS detectors were successfully used on irradiated samples. In this half year no experience is gained with the EBSD detector. This detector gives crystallographic information, like crystal structure and orientation [3]. This can be used for example for phase identification, texture, orientation relationships or grain boundary definition. Fig. 4.5 gives an example of EBSD result on unirradiated Si samples. The Kikuchi lines on the image are detected and identified by the software. The plan is to test the EBSD technique on irradiated samples this year.

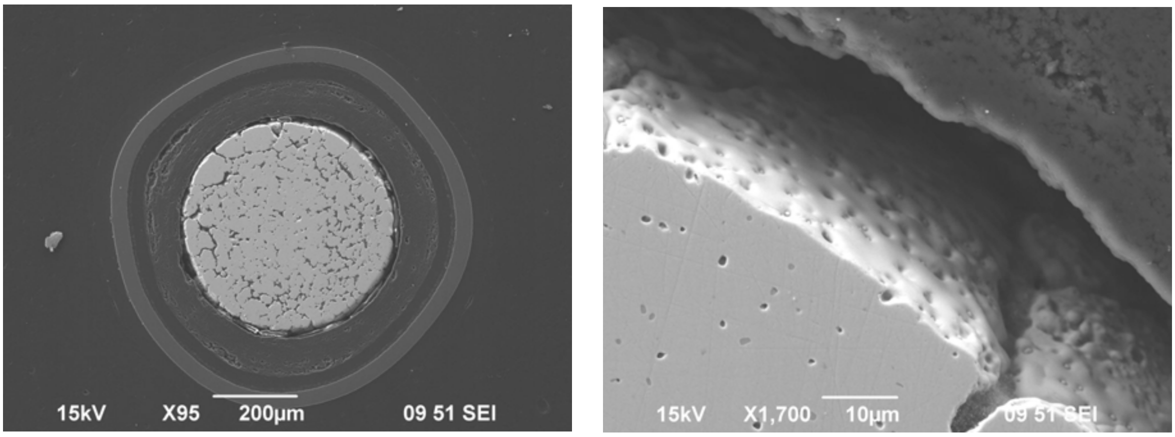

FIG. 4.1. SEM images of HTR TRISO fuel particle (left: overview, right: detail between UO_2 kernel and buffer layer).

FIG. 4.2. Example of element mapping (Cs) in the layers around the UO_2 kernel. Red dots indicate presence of C.

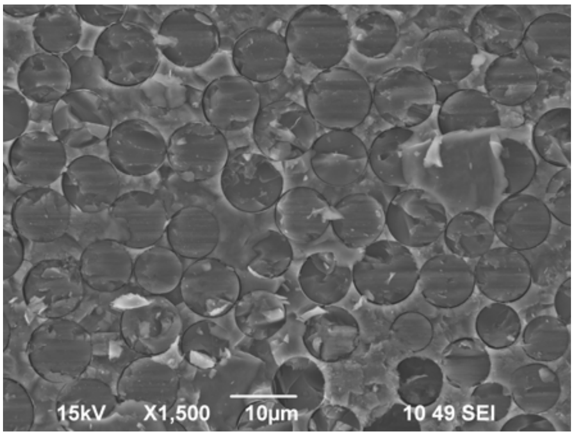

FIG. 4.3. Fracture surface of SiC/SiC bonded type 1, irradiated to 2.4 dpa (steel) at 600ºC.

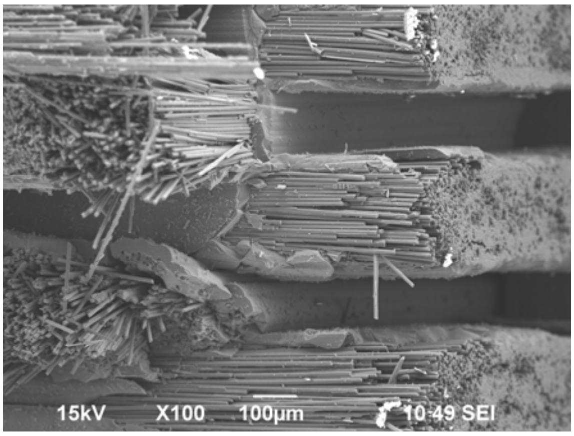

FIG. 4.4. Fracture surface of SiC/SiC 3D woven type 1, to 4.5 dpa (steel) between 800–900ºC.

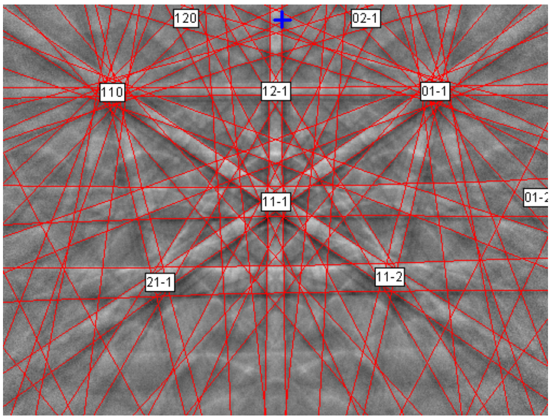

FIG. 4.5. EBSD pattern of a Si sample (unirradiated).

5. CONCLUSIONS

A Jeol JSM-6490 SEM, equipped with Oxford EDS/WDS/EBSD detectors, was successfully installed in a new hot-cell at the NRG Hot-Cell Laboratory. The first radioactive samples were investigated by this equipment in August 2010.

EDS and WDS detectors were successfully used to analyse irradiated fuel and structural materials.

The EBSD detector is not used on irradiated materials yet.

REFERENCES

[1] FÜTTERER, M.A., et al., Irradiation results of AVR fuel pebbles at increased temperature and burn-up in the HFR Petten, Proc. HTR2006, Johannesburg (2006).
[2] DE GROOT, S., et al., Fission product behaviour during irradiation of TRISO-coated particles in the HFREU1bis experiment, Proc. HTR2008, Washington DC (2008).
[3] DINGLEY, D.J., RANDLE, V., Microtexture determination by electron backscatter diffraction, Journal of Materials Science **27** (1992) 4545–4566.