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TEM Foil Preparation from Irradiated Metallic Materials: A Practical Approach

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Abstract: This paper focuses on work related to post irradiation examination of 300-series austenitic stainless steel taken from reactor vessel internals of PWR. High neutron irradiation dose in NNP's leads to a degradation of microstructure of the material in a nano-metric scale. Hence, it is important to characterize the irradiated materials to understand the physical basis of the degradation mechanisms. Microstructural characterization of neutron-irradiated materials by TEM requires enhanced sample preparation methodologies, which commonly needs general improvements regarding particular experiment to be performed. In this study, the authors have developed methodology specialized in 1 mm TEM thin foil preparation from a deformed shank of a broken miniaturized tensile specimen. TEM foil size in current studies is smaller than standard because of the small shank diameter and high radioactivity of the studied material. The reduction of the TEM foil radioactivity to minimum is crucial to perform EDX chemical analysis and to increase the EDX detector lifetime. This paper describes whole process from bulk sample handling, including remote-controlled material cutting in shielded hot-cells and disc polishing in glow-boxes, up to the final procedure of electrolytic-polishing of electron transparent 1 mm TEM foils. Eventually, results of TEM microanalysis of radiation-induced defects were present.

Key words: Reactor vessel internals, irradiated sample preparation, microstructural characterization, irradiation induced defects.

1. Introduction

Same materials are used in primary and secondary coolant circuits of Pressurized Water Reactor (PWR) [1]. Austenitic stainless steels and Nickel based alloys are used as materials for the reactor internal components due to their relatively high strength, ductility and resistance to corrosion in water environment of PWR's. Tough environment in the reactor is due to irradiation, temperature, mechanical stress causes degradation of internal components (such as bolts, guides and springs etc.). Degradation is associated to change of materials structural and physical properties. Degradation mechanisms are divided broadly based on extent of radiation dose levels, temperature of surrounding, pre-load and stress conditions. Fig. 1 depicts the corresponding damage levels and respective failure modes [2].

Irradiation Assisted Stress Corrosion Cracking (IASCC) is one of the most significant environmental degradation in the internal components made from Austenitic stainless steel. Detailed description on IASCC can be referred [3]. IASCC damage mechanism in these materials is complex and involves simultaneous actions of neutron irradiation, stress/strain, temperature and coolant (H_2O). IASCC affected components include baffle bolts, in core shrouds and control rod cladding etc. of PWRs. Fig. 2 shows an image of the baffle bolt in VVER type reactor [4, 5]. There is a significant interest to perform the research activities to define the neutron fluence thresholds, where austenitic steels components are susceptible to IASCC. Such a data is crucial and critical for actual NPPs components aging management [6].

Post irradiation analysis of these materials in Transmission Electron Microscope (TEM) has been versatile way to understand the level of irradiation damage.

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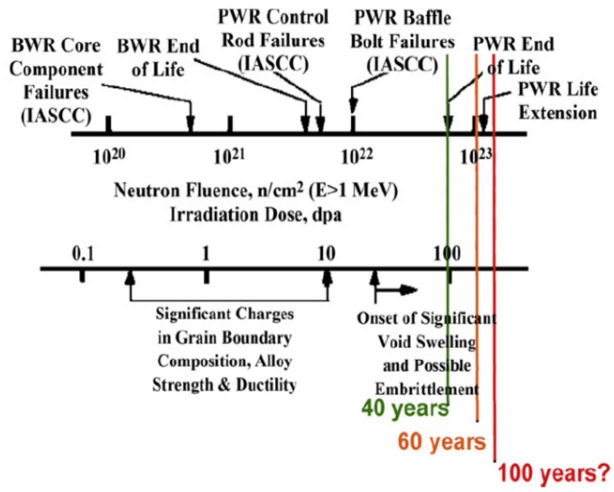


Fig. 1 LWR components failure modes corresponding displacement-fluence -time [2].

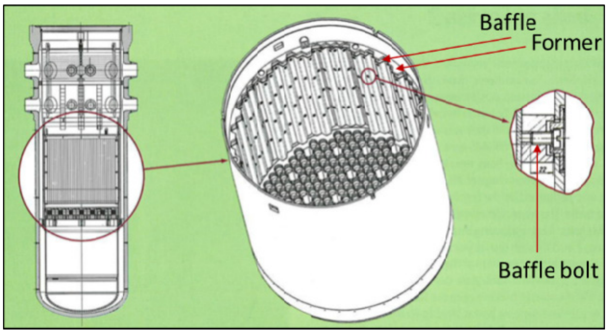


Fig. 2 Schematic representation of baffle bolt in VVER reactor type [3].

TEM evaluation enables quantitative and qualitative analysis of irradiation induced microstructural changes/damage. This information will enable evaluation of the aging of material.

Microstructural analysis with modern tools requires enhanced sample preparation methodologies that could successfully prepare thin foils of about a few hundred nanometers. In addition, the success of the microstructural analysis critically depends on the quality of the thin foils prepared [7, 8]. Compared to TEM analysis of un-irradiated materials, for irradiated materials, the sample preparation methodologies are more complicated and are challenging task due to the activity of the samples. Henceforth, amended preparation tools and procedures are required to handle the specimens by manipulators in hot-cells or in semi hot-cells facilities.

2. Materials

The material examined is 15% Cold Worked (CW) 316 series fully austenitic stainless steel with chemical composition as given in the Table 1. This is the material used for baffle bolts in core internals.

The two SSRT tensile specimens were machined from 316 CW SS with a gauge length of 12 mm and a shank diameter of 2 mm in accordance with Fig. 3 [9, 10]. These tensile specimens were irradiated in the high-flux research reactor SM at a temperature close to 300 °C at two different positions. Specimen 1 was shielded against the thermal neutrons with a pure fast neutron spectrum. Specimen 2, without shielding with both fast and thermal neutron fluxes.

The difference between neutron spectra at both positions leads to different helium and hydrogen production by transmutation. For this study, the specimens will be differentiated by irradiation conditions as fast condition (specimen 1, with low He content) and as mixed condition (specimen 2, with higher He content).

2.1 TEM Foils Preparation from Irradiated CW316 Specimens

The critical aspect of the research work is to obtain the TEM foils from the shank of SSRT specimens with dose rates as given in the Table 2. The foremost concern was to perform TEM study of localized area adjacent to a fracture surface of broken tested specimen. Fig. 4 shows the broken specimen after the SSRT test.

The initial shank diameter of the specimen is 2 mm.

Table 1 Chemical composition of the studied 316 materials.

C	S	P	Si	Mn	Ni	Cr	Mo	Cu	Co	Nb-Ta	B	O	N	Al	Ti	Other
					(%)							(ppm)			(%)	
0.054	0.022	0.027	0.68	1.12	10.6	16.6	2.25	0.24	0.12	0.01	5	41	230	-	< 0.01	-

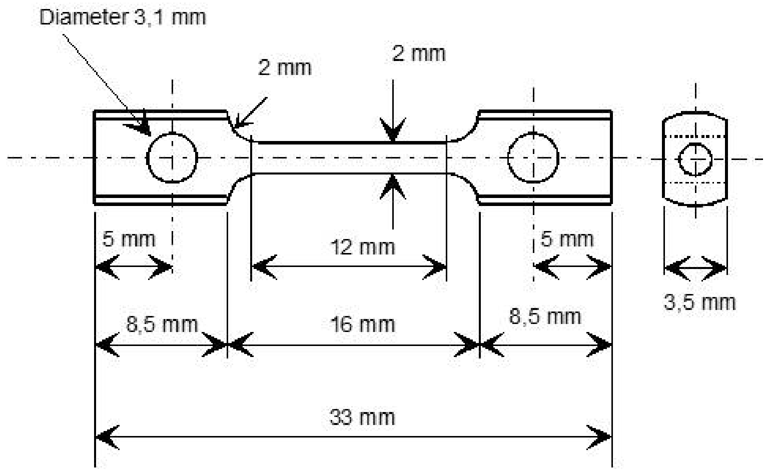


Fig. 3 Scheme of the mechanicaltest specimen.

Table 2 Dose rate accumulation data after irradiation.

Material	Specimen	Irradiation condition	Acquired dose (dpa)
CW316	1	Fast	16.1
	2	Mixed	15

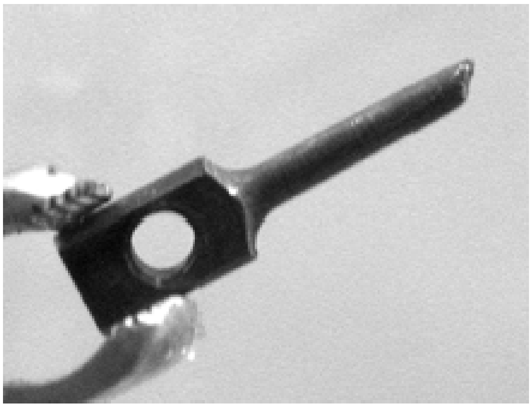


Fig. 4 Broken SSRT specimen after test.

After the tensile test, the shank diameter is reduced to < 2 mm due to specimen necking and fracture. Preparing TEM foils from such active specimens is critical task, requires extreme sample preparation skills and precise sample preparation methodology to identify the irradiation induced damage or damage mechanisms.

2.2 Applied Methodology to Attain 1 mm Samples in Diameter

A methodology can be considered as a standard when it can repeatedly produce the same results. Due to simplicity and find possible difficulties with active materials, TEM foil preparation was tested for the un-irradiated SSRT specimens with exactly same dimensions like active ones. Upon several repeated preliminary tests on un-irradiated specimens, the developed methodology was applied to the irradiated specimens in a semi hot-cell. The methodology of foil preparation for TEM analysis from irradiated SSRT specimens comprised of following steps in a shielded semi hot-cell.

2.2.1 Remote-Controlled Cutting

The cutting of the broken tensile specimens at the crack area was performed in the semi hot-cell using a diamond saw machine with a special sieve attached in the bottom to collect the sliced discs. A custom-made specimen holder manufactured was able to fix the broken specimen accurately as well as for fixation in the manipulators tongue. This special holder avoids any unexpected movement of the specimen during cutting. The sample was cooled by distilled water instead oil to avoid contamination during cutting. This also avoids cleaning step in ultrasonic baths. A camera coupled with the attached mirror was able to see the specimen exact position, control the cutting procedure and monitor the sample thickness as well as the cutting depth. The details of custom-made specimen holder and related fixation steps are shown in Figs. 5(a)-(d).

Two slices were cut from each specimen as shown in Fig. 6. One slice from the closer end of the broken shank and the other 300 μm away in the direction of specimen head, as in Fig. 7.

2.2.2 Mounting and Polishing

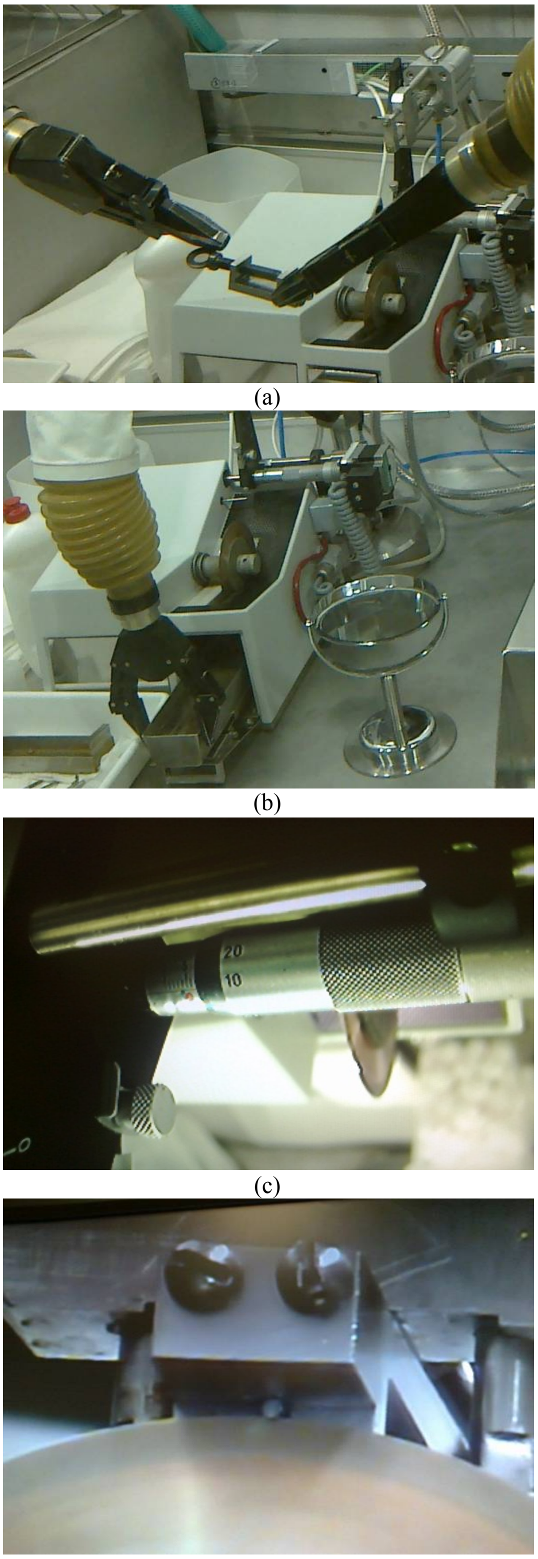
Mounting as well as mechanical polishing of cut slices performed in the shielded glove boxes. This procedure generally performed in the same way as for classical disc specimens of 3 mm in diameter. Firstly, the cut slices were mounted on metallographic sample holders to get the desired thickness by grinding. SiC papers of grit 1,000 were used to make the same flat and to reduce its thickness. The slices were polished firstly from one side down to 200 μm and thereafter from the other side to obtain final thickness of 60 μm.

2.2.3 Punching of 1 mm TEM Discs

The standard punching device used for preparing classical TEM foils was modified to obtain precisely TEM disc of 1 mm in diameter. A new punch and die developed from quenched martensitic steel with optimized diameters and clearance. This novel equipment shown in Fig. 7 with a 2D view in Fig. 8 which was able to punch out the 1 mm discs without mechanical deformations required for electrolytic polishing.

2.2.4 Electrolytic Polishing (EP) and TEM Instrumentation

Fischionne Twin-jet electrolytic-polisher [11] is used to polish the 1 mm dia. discs with thickness of 300 nm in the solution of 5% perchloric acid ($HClO_4$) in 95% methanol (CH_3OH). A new 1 mm disc holder had been developed and used. Electrolytic polisher is shown in Fig. 9 along with holder. Figs. 10 and 11 show the foil assembly with electrode and modified holder. The electro-polishing procedure was optimized based on extensive testing of all parameters on non-irradiated specimens made from an austenitic stainless steel. In addition, parameters of each



(d)

Fig. 5 (a) Tailored sample holder with broken specimen, (b) optimized diamond saw machine and the mirror, (c) control of cutting deviceby the camera and mirror and (d) monitoring of cutting process by the camera and mirror.

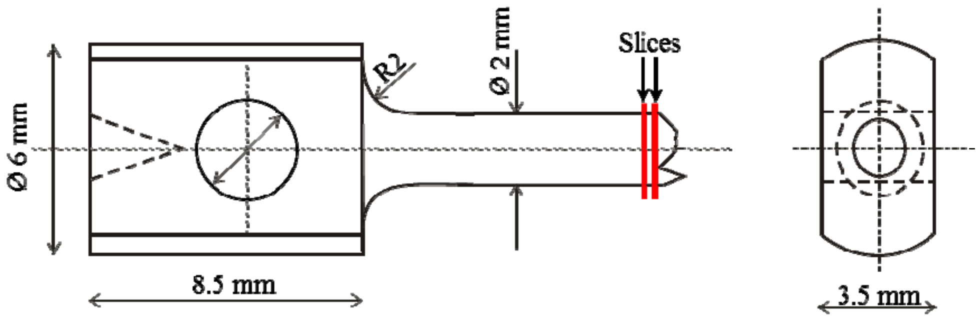


Fig. 6 **Cutting scheme of tensile specimen shank and slice position details.**

Fig. 7 **Novel punching device for 1 mm TEM thin foils.**

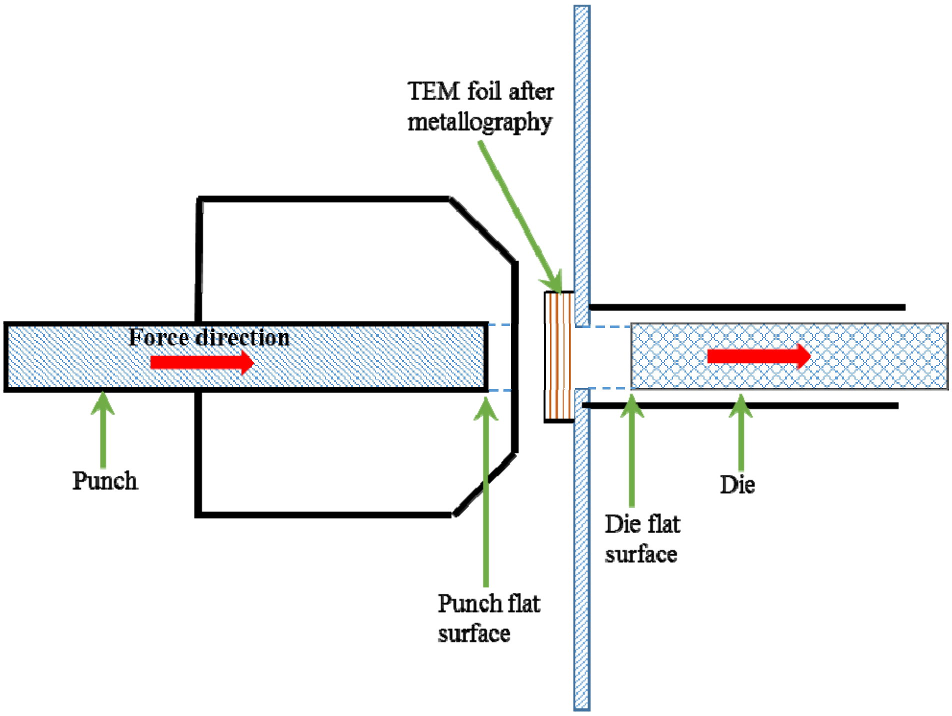


Fig. 8 **Amended punching device.**

Fig. 9 Fischionne twin-jet electrolytic-polisher.

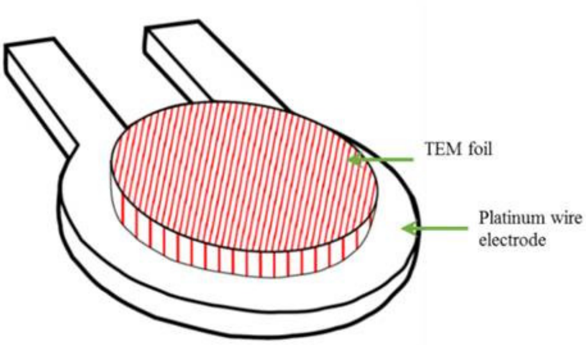


Fig. 10 Assembly of platinum electrode and TEM foil.

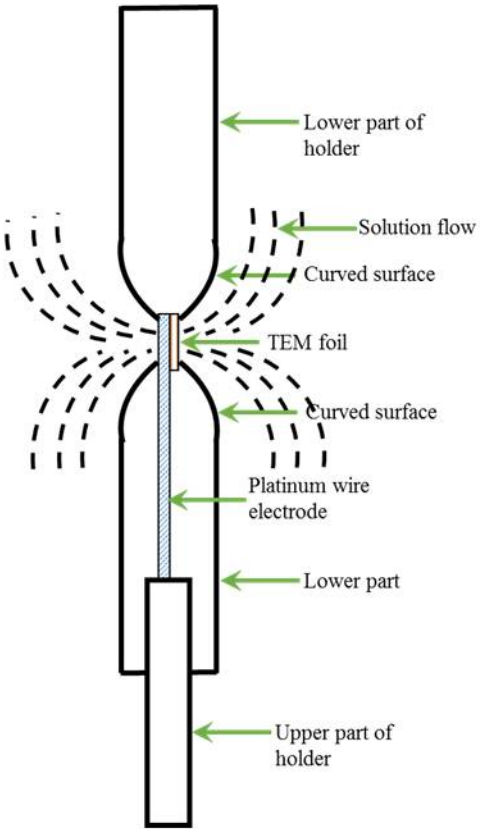


Fig. 11 Modified EP holder.

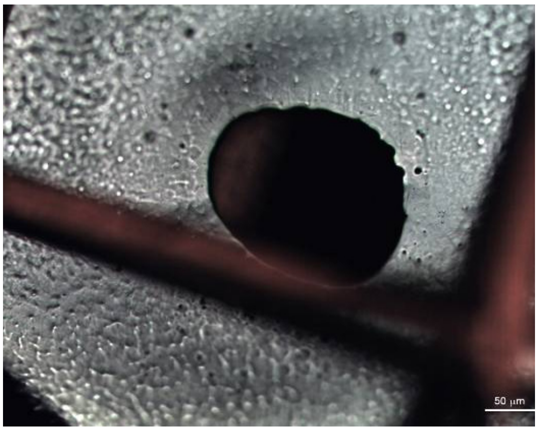


Fig. 12 Active TEM thin foil in a double grid mesh.

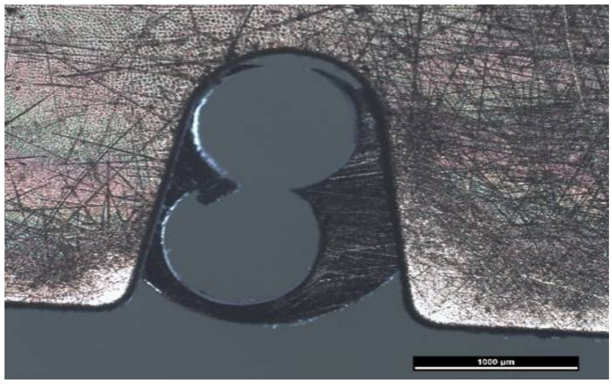


Fig. 13 Slice with two 1 mm diameter punches.

electro-polishing procedure of irradiated TEM disc were checked by testing on non-irradiated specimens to assure successful accomplishment of the unique procedure. The optimized electro-polishing conditions used for irradiated material were T (-25 °C) and I (10-20 mA). The light-optical micrograph of a typicalthin foil and left over of slice are shown in Fig. 12 and Fig. 13 respectively.

The TEM foil preparation procedure from 2.2.1 to 2.2.4 appears to be simple and straightforward. However, to have reproducible sample, conditions of the EP parameters (voltage, current density, temperature, time and flow rate) and specimen characteristics (thickness, conductivity and its nature as brittle or ductile) has to be taken into concern. The EP rates and polishing conditions vary from material to material. Microscopic examinations performed on JEOL JEM-2010 transmission electron microscope operated at 200 kV with W-filament.

Microstructure was analysed using bright-field and centered dark-field two-beam dynamical conditions, kinematical conditions and weak beam imaging. Size and density of frank loops were evaluated from centred dark-field rel-rod images. Cavities were identified and quantified from through-focal series under bright-field kinematical and/or down-zone conditions in regions thin enough to limit defect overlapping. Densities of defects were evaluated using conventional foil thickness 150 nm, established as an average value from several Convergent Beam Electron Diffraction (CBED) measurements on as-received material.

3. Results and Discussions

The use of typical TEM discs of 1 mm in dia. instead of the standard discs in dia. 3 mm resulted in substantial reduction of dose rates of final 1 mm TEM thin foils. Fig. 14 shows dose rates measured at different distances on 3 mm dia. foils and on 1 mm dia. foils prepared from the past and current studies respectively. From Fig. 14, it can be read that the dose rate of the 3 mm dia. foil was about 6 mSv/h and the dose rate of the 1 mm dia. foil decreased to about 0.34 mSv/h.

Dose rates of samples irradiated in mixed neutron spectrum are higher than samples irradiated in fast neutron spectrum. One of the prominent point here from the 1 mm dia. thin foils preparation is the reduction in the dose rates. This is so beneficial for further chemical analysis by EDS, because an additional noise, increased background of energy spectrum caused by γ-radiation will be reduced and for long-term safe operation of detectors.

In the Figs. 15-22, which revealed are TEM results from 1 mm dia. active thin foils. Microstructural analysis provided full characteristic details of the irradiated sample. Deformed microstructure, cavities, precipitates, frank loops and cavities at grain boundaries on the irradiated samples were clearly visible. The most important and noticeable evidences

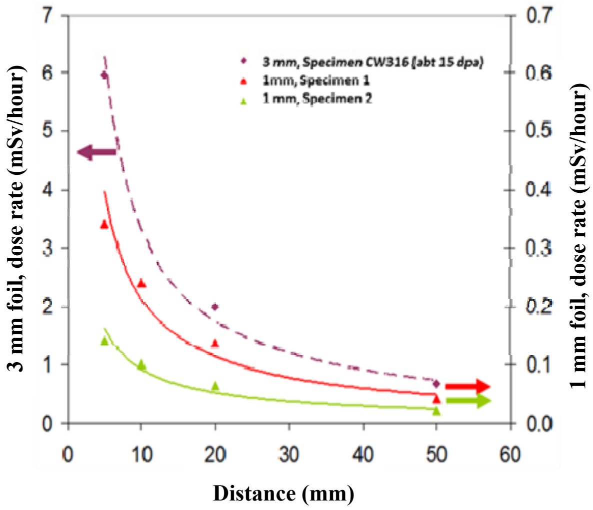


Fig. 14 Dose rate from classical 3 mm and currently developed 1 mm TEM foils.

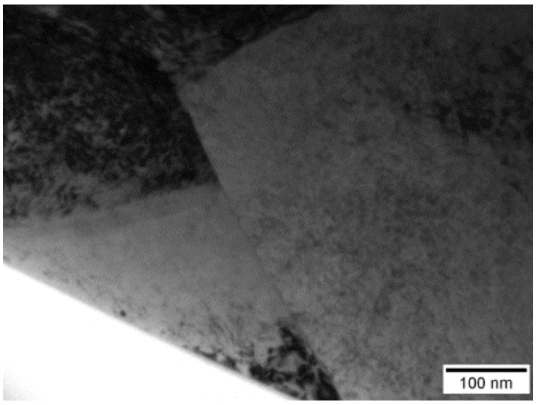


Fig. 15 Sample after irradiation.

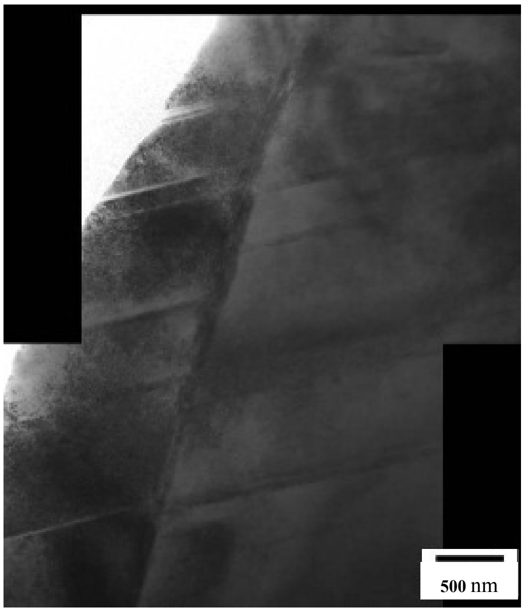


Fig. 16 Deformation bands in irradiated specimens.

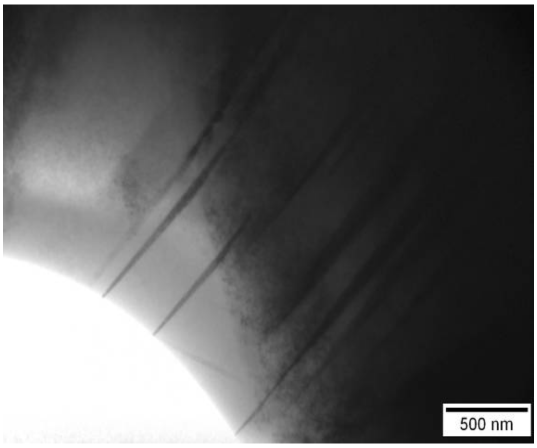


Fig. 17 Deformation twins, bright-field image.

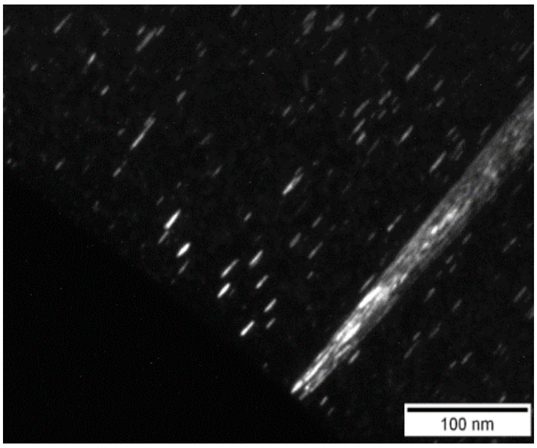


Fig. 20 Frank loops imaged in rel-rod dark-field.

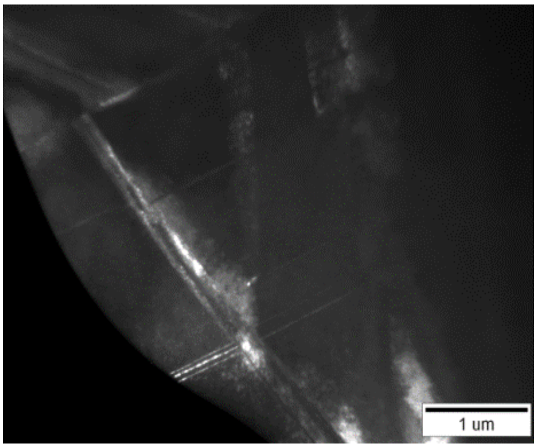


Fig. 18 Deformation twins, dark-field image.

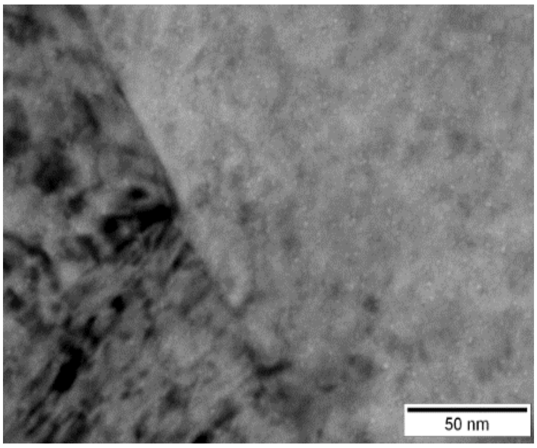


Fig. 21 Cavities in the irradites sample.

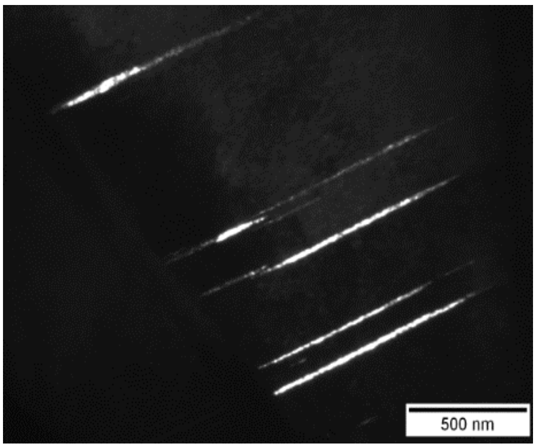


Fig. 19 Micro-twins observed in dark-field.

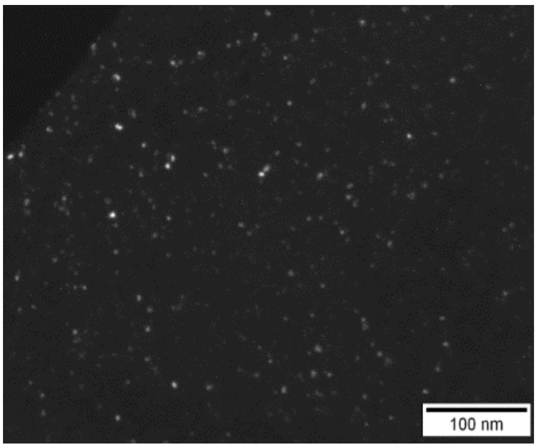


Fig. 22 Precipitates in the diffraction pattern.

are that the 1 mm foils provided outstanding micrographs as compared to classical 3 mm dia. TEM foils.

4. Conclusions

New methodology and instrumentation for 1 mm dia. TEM thin foils preparation developed to study the microstructure from broken shanks of neutron irradiated tensile specimens. The methodology reduced the radioactivity of TEM foil to minimum that is crucial to perform EDX chemical analysis and to increase detector lifetime.

Numerous deformation twins, deformation bands and probably transformation phases in interactions of twins and bands were determined. Frank dislocation loops are primary type of damage in the irradiated specimens. On irradiated specimens, cavities and fine rounded particles of radiation-induced precipitates were identified.

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