Mechanical Properties of Reduced Activation Ferritic/Martensitic Steels after European Reactor Irradiations

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Abstract. The development of First Wall and Blanket materials which are capable of withstanding many years the high neutron and heat fluxes, is for material scientists a critical path to fusion power. In an energy generating fusion reactor, such structural materials will be exposed to very high levels of irradiation damage up to about 80 dpa. Regarding the radiation damage resistance of the considered Reduced Activation Ferritic/Martensitic steels, e.g. the European EUROFER and the Japanese F82H mod., as international reference material of this kind, broad European reactor irradiation programmes cover several steps from up to 5 dpa for ITER Test Blanket Modules, till up to 80 dpa for First Wall and Blanket of a DEMO fusion reactor.
The lower irradiation damage conditions until 15 dpa can be realized in European fission reactors like the HFR at JRC, Petten, but higher damages - in reasonable times - in fast reactors only. For this purpose the fast reactor BOR 60 of the State Scientific Centre of Russian Federation Research Institute of Atomic Reactors, Dimitrovgrad, has been utilized for irradiations up to 80 dpa.
Results from the lower damage irradiations like SIENA, MANITU and HFR-Ib, up to 2.4 dpa had been reported in the past frequently. Recent results of mechanical properties, like Ductile to Brittle Transition Temperatures from instrumented impact-V tests with sub size specimens and stress and strain values from tensile tests with miniaturized specimens will be presented from specimens of the HFR Phase-IIb (SPICE) irradiation project up to 15 dpa at different irradiation temperatures between 250 and 450°C.
The fast reactor irradiation project ARBOR 1 reached at a temperature ≤ 340°C an irradiation damage of 33 dpa. In the post irradiation instrumented impact-V tests a dramatic increase in the Ductile to Brittle Transition Temperature as an effect of irradiation has been detected. During tensile testing the strength values are increased and the strain values reduced due to irradiation hardening.
After first thermal recovery tests with 550°C annealing for three hours it could be demonstrated on EUROFER specimens irradiated at a temperature ≤ 340°C up to 15 dpa, that nearly virgin conditions could be achieved.

1. Introduction

In an energy generating fusion reactor, structural materials will be exposed to very high levels of irradiation damage of about 80 dpa. A simulation facility - like IFMIF - is not available in the nearer future to study the materials behavior under fusion relevant irradiation conditions, e.g. specific He/dpa-ratio of 10 – 15 appm/dpa. Therefore irradiations in high flux reactors (HFR, Petten) and fast reactors (BOR 60, Dimitrovgrad) have been realized. The irradiation to 16.3 dpa at different irradiation temperatures (250-450°C) was carried out in the Petten High Flux Reactor (HFR) in the framework of the HFR Phase-IIb (SPICE) (**Sam**p**le Holder for** **I**rradiation of Miniaturized Steel Spe**c**im**e**ns Simultaneously at Different Temperatures) irradiation project. Due to the fact that for higher dpa-rates fast reactor irradiation facilities in Europe are no longer available, a cooperation between Forschungszentrum Karlsruhe (FZK) and State Scientific Centre of Russian Federation Research Institute of Atomic Reactors (SSC

RF RIAR) has been implemented. The irradiation project "**A**ssociated **R**eactor Irradiation in **BOR** 60" is named "ARBOR 1" (Latin for tree).

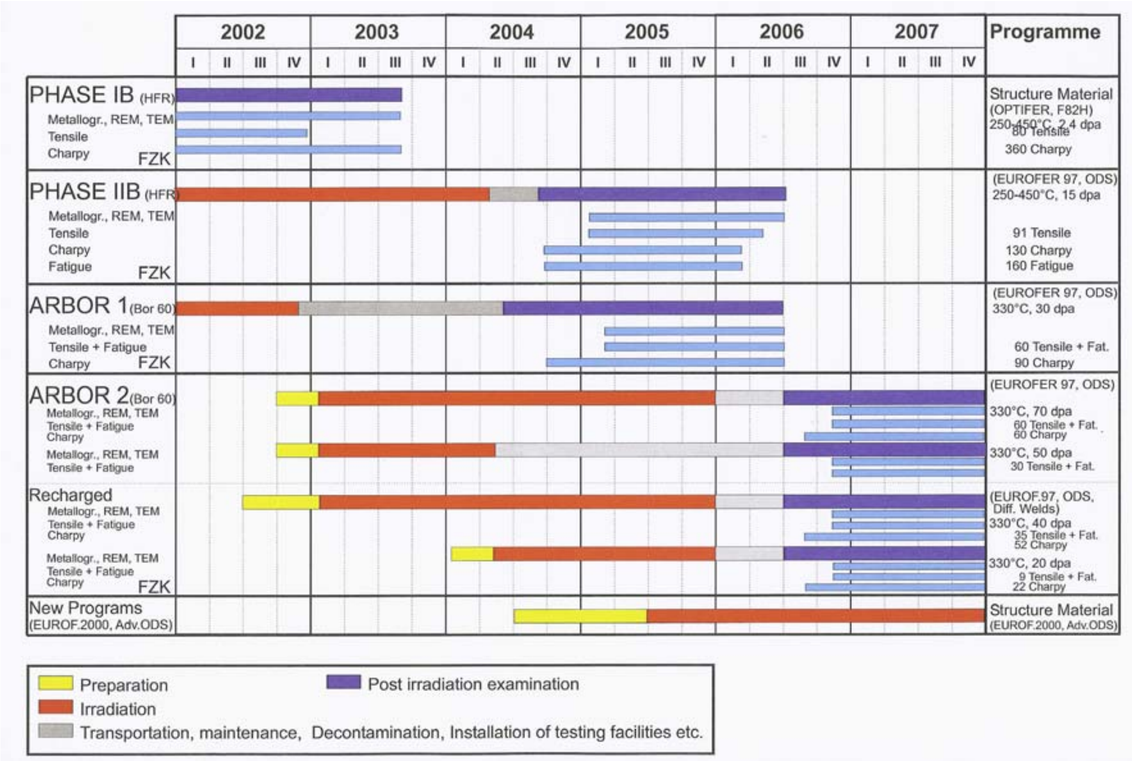

FIG. 1: Time schedule of the HFR and BOR 60 irradiations.

Both irradiation programmes are embedded in the European long term programme for the development of structural materials for DEMO-blankets. The time schedule for both irradiations is shown in *FIG. 1*.

2. Irradiation conditions

The technical details of the HFR reactor of JRC and specific features of the SPICE irradiation rig are reported in [1]. The BOR 60 experimental fast reactor of SSC RF RIAR is nowadays widely used as an irradiation facility for material science purposes. [2].
In the SPICE irradiation rig the irradiation dose of 16.3 dpa in steel was achieved as measured by activation detectors installed in the sample holder. The irradiation temperatures of 250,300,350,400, and 450°C were maintained by a balance between nuclear heating and cooling with liquid sodium [3].
The ARBOR 1 irradiation device with outer hexagon size of 45 mm and a specimen capsule diameter of 39 mm is heated by the direct flow of the sodium coolant from the reactor high-pressure chamber, which allows a coolant flow rate of 7 m^3/ h and a low gamma heating rate of 5 watts/g, which produces an increase of about 10-15°C over the length of the capsule. The irradiation rig was instrumented; neutron monitors in the central tube and three temperature detectors at three of ten levels of specimen positions were equipped. The calculation of the damage dose values for ferritic steel specimens was conducted using the SPECTER code [4]. In this case a neutron energy spectrum in cell D-23 of the BOR 60 core was used that had been measured in previous dosimetry experiments.

3. Irradiated materials

In case of the SPICE irradiation, specimens of an industrial heat of EUROFER97 steel, produced by Böhler Austria GmbH, and of other steels for comparison were implemented, see [5] for the chemical composition of the different steels.
Part of the SPICE specimens (labeled EUROFER97 ANL) was machined from the 25 mm thick EUROFER97 plates in the as-delivered state (i.e. austenized at 980°C, tempered at 760°C). In order to study the influence of higher austenitizing temperature on a laboratory scale, another part of the specimens (labeled EUROFER97 WB) was subjected to a heat treatment at the higher austenitizing temperature of 1040°C. An extensive micro structural investigation on EUROFER97 for different austenitizing temperatures was performed in [6]. For comparative purpose the heat treatments for reference alloys F82H-mod, GA3X, OPTIFER-Ia and martensitic steel MANET-I were identical to those used in previous experiments [7,8]. EUROFER97 ANL was irradiated at all irradiation temperatures. Because of the limited irradiation space, other materials were irradiated at selected temperatures only.
The ARBOR 1 irradiation included 150 mini-tensile/low cycle fatigue specimens and 150 mini-impact (KLST) specimens of 9 different RAFM steels, e.g. EUROFER 97, F82H mod., OPTIFER IVc, EUROFER 97 with different boron contents and ODS-EUROFER 97. They were irradiated in a fast neutron flux of 1.8×10^{15} n/cm²s (> 0.1 MeV) at a temperature < 340°C up to ~ 30 dpa [9]. Approximately 50 % of the specimens were unloaded for Post Irradiation Examinations (PIE). The other 50 % were reloaded into the ARBOR 2 rig for further irradiation in BOR 60 to reach a maximum irradiation damage of 70 dpa. The chemical composition of the different steels was reported in [10].
Part of the ARBOR 1 specimens (labeled EUROF 1) was in the as-delivered state (i.e. austenized at 980°C, tempered at 760°C). Another part of the specimens (labeled EUROF 2) was subjected to a heat treatment at the higher austenitizing temperature of 1040°C.

4. Testing techniques

The mechanical post irradiation examination (PIE) of irradiated SPICE specimens was performed in the newly built materials hot cells of the fusion material laboratory of FZK. The impact testing facility is identical to that one used for testing of unirradiated specimens. The test and evaluation procedures are identical to those employed in previous investigations [7,8]: 25 J pendulum impact hammer; distance between supports 22 mm; impact velocity 3.85 m/s; automatic specimen cooling, heating and transporting system; test temperature between -180°C and 600°C. The tensile tests are performed with a ZWICK universal testing machine equipped with a vacuum furnace and extensometer.
The mechanical PIE of irradiated ARBOR 1 specimens is performed at a material science laboratory of the SSC RF RIAR. The impact and tensile testing results reported here are the first part of the PIE that also includes fatigue testing. The impact tests are performed with a modern instrumented impact testing facility of ZWICK 5113-HKE type, equipped with a pendulum hammer of 15 J impact energy. The tensile tests are performed at a modernized electromechanical testing machine INSTRON 1362 DOLI with a rigidity of 450kN/mm. This machine was equipped with a three-zone furnace and high-temperature extensometer of MAYTEC.

4.1. Impact testing

For both PIE impact series of SPICE as well as for ARBOR 1 specimens the force vs. deflection curve was recorded. The impact energy (E) was than determined by the integration of force vs. deflection curves. The impact energies were plotted vs. test temperature (T) and analyzed with respect to the characteristic values of the Charpy upper shelf energy (USE, i.e. maximum in the energy vs. temperature diagram) and the ductile-to-brittle transition temperature (DBTT). Between 5 and 6 specimens for each material and each irradiation condition ensured a sufficient number of measurement points for drawing Charpy energy vs. test temperature curves.

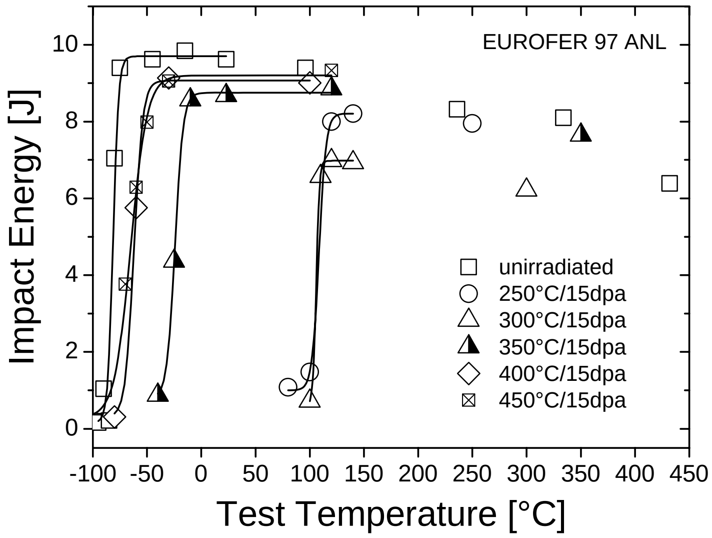

FIG. 2 Impact properties of unirradiated and irradiated SPICE/EUROFER97 ANL with the curves fit by a function [5].

FIG. 3 shows the DBTT vs. irradiation temperature of the SPICE irradiation. For all investigated materials the DBTT is most strongly influenced at low irradiation temperatures (T_{irr}≤ 300°C). Remarkably, the DBTT of EUROFER97 WB at T_{irr}=250°C is smaller than the DBTT of EUROFER97 ANL by 50°C. The difference in the DBTT between EUROFER97 materials austenized at the two different temperatures decreases with increasing T_{irr}, completely vanishing at 450°C. The DBTT of F82H mod. and OPTIFER-Ia is comparable to that of EUROFER97 ANL at T_{irr}=300°C. However, for F82H mod. the slope of the impact energy vs. test temperature curve in transition region is smaller than the corresponding slopes for EUROFER97 ANL and OPTIFER-Ia. The DBTT's of the materials irradiated above 400°C remain below -30°C and thus well below the expected material application temperature [11].

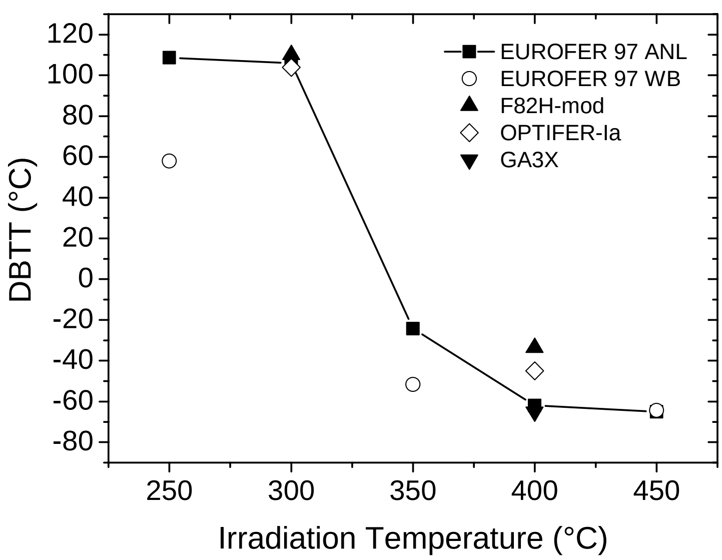

FIG. 3 DBTT vs. irradiation temperature of irradiated SPICE materials (they are indicated in legend)

In *FIG. 4* the impact curves for EUROF 1 (as received) and EUROF 2 (annealed) are shown for the unirradiated condition and for material irradiated at 332°C to 31.8 dpa as an average value. The DBTT-values for unirradiated materials lie very close together (EUROF 1: – 81°C and EUROF 2: - 90°C), the USE-values for both materials are 9.84 J. For comparison, the DBTT and USE of NRG impact tests had been found at – 68°C and 9.5 J, respectively. The DBTT for 31.8 dpa irradiated EUROF 1 is 137°C (NRG-DBTT-data: 10°C (SUMO-04, 300°C, 2.46 dpa) and 115°C (SUMO-02, 300°C, 8.9 dpa) [12]) and for EUROF 2 is 107°C. The USE is reduced in the irradiated state, to 7.01 J for EUROF 1 (NRG-USE-data: 8.5 J (SUMO-04, 300°C, 2.46 dpa) and 7.07 J (SUMO-02, 300°C, 8.9 dpa))

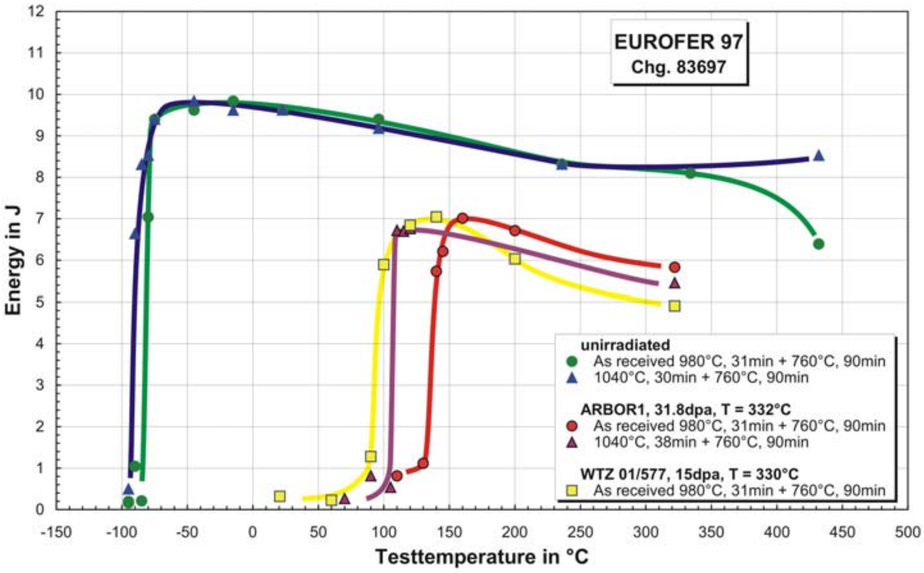

FIG. 4 Impact properties of unirradiated and irradiated ARBOR 1/-EUROFER97in the as received and annealed condition and compared to another 15 dpa irradiation performed in BOR 60 (WTZ 01/577)

and 6.76 J for EUROF 2. So, on one hand the earlier finding [7] was confirmed that the higher austenizing temperature of EUROF 2 at 1040°C reduces the irradiation damage in respect to impact properties, mainly on DBTT. On the other hand the DBTT increases with increasing irradiation damage and a saturation value, as it was expected for RAF/M steels in [10] has not been reached at the irradiation damage of around 30 dpa.

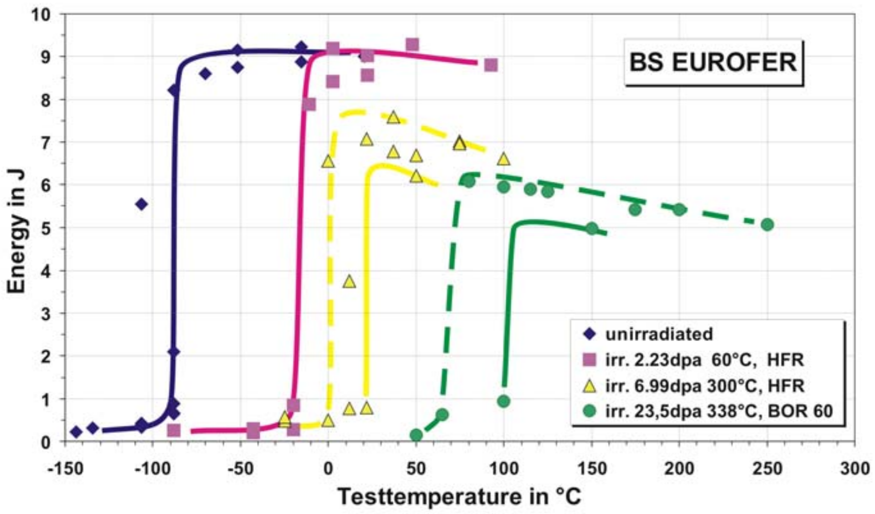

FIG. 5 Impact properties of unirradiated and irradiated ARBOR 1/BS EUROFER and compared to other 2.23 and 6.99 dpa irradiations, SIWAS-09 and SUMO-02, resp.[12,13]

The performance of the NRG lab heat BS EUROFER, which was produced in accordance with the EUROFER 97 specifications, is good, as shown in *FIG. 5*. It was tested after 6.9 dpa irradiation the DBTT is 10°C and the USE reduced by 2.2 J. Before, this heat was in the SIWAS-09 irradiation at 60°C up to 2.23 dpa. It is evident that no USE reduction occurs at this dose and irradiation temperature, while DBTT shift is 70°C [12]. The ARBOR 1 irradiation (23.5 dpa at 338°C) damaged the material considerably to reach a high DBTT value of 70°C and a low USE of about 6 J, that lies under the EUROFER 97 values for 31.8 dpa damage.

The state of knowledge on the impact behaviour of RAF/M steels is illustrated in *FIG. 6.*. This figure shows the irradiation damage dependence of DBTT of EUROFER 97, F82H mod. and OPTIFER's, and compares them to the conventional steel MANET-I. The increase in DBTT with increasing irradiation damage in the RAF/M's is actually half of that of MANET-I. However the hope to have reached a saturation state is not confirmed. Further information will come from the joint ARBOR 2 irradiation with specimens from FZK and CEA irradiated to a damage of 70 to 80 dpa, respectively.

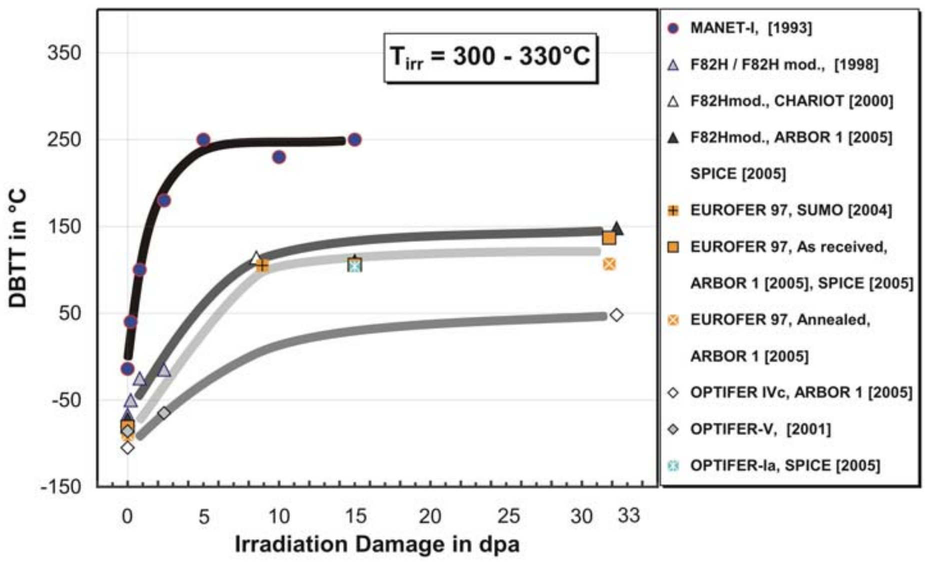

FIG. 6 Comparison of irradiation dependence on the Ductile to Brittle Transition Temperature behavior for different technically relevant RAF/M steels compared to conventional 12% Cr steel MANET-I.

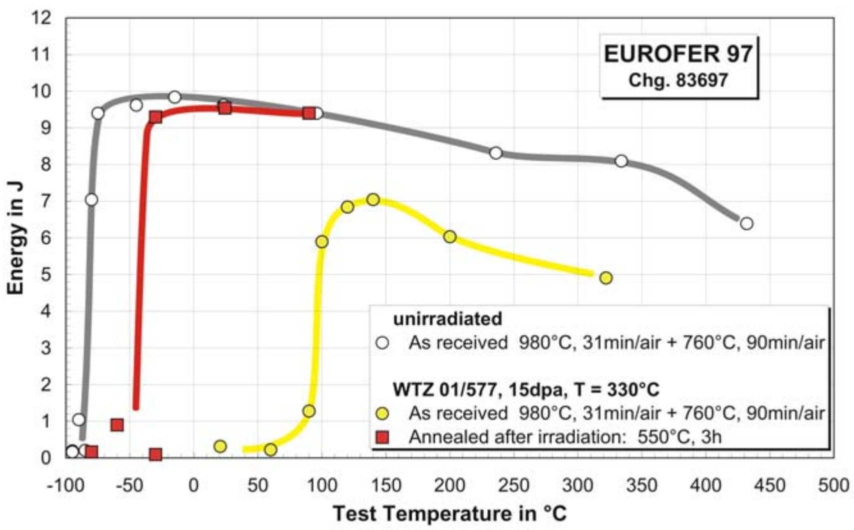

FIG. 7 Impact properties of unirradiated and irradiated (15 dpa irradiation in BOR 60 (WTZ 01/577)) EUROFER97 in the as received condition and compared to a postirradiation annealed material.

To explore the Reduced Activation Ferritic/Martensitic materials possibilities exemplary, postirradiation annealing experiments after the 15 dpa BOR 60 irradiation had been performed. An annealing for 3 hours at 550°C lead, as can be seen from *FIG. 7*, to the possibility to heal irradiation damage and to reach again nearly virgin Ductile to Brittle Transition Temperatures in the desired temperature range below – 30°C.

4.2. Tensile testing

The tensile tests have been performed on four different kinds of specimens types utilized in the different irradiations. NRG irradiated cylindrical specimens of 20 mm gauge length and 4 mm diameter and performed the tests with a strain rate of $5x10^{-4}$ s^{-1}. In the SPICE irradiation cylindrical specimens of 18 mm gauge length and 3 mm diameter are tensile tested under vacuum with a strain rate of 10^{-4} s^{-1}. In the 15 dpa BOR 60 irradiation cylindrical specimens of 15 mm gauge length and 3 mm diameter are tensile tested with a strain rate of $3x10^{-3}$ s^{-1}. In the ARBOR 1 irradiation cylindrical specimens of 7 mm gauge length and 2 mm diameter are tensile tested with a strain rate of $3x10^{-3}$ s^{-1}. From the load-displacement curves strength and strain data are calculated: 0,2% offset Yield Stress ($R_{p0,2}$), Ultimate Tensile Strength (R_m), Uniform Strain (A_g) and Total Strain (A). Since the most considerable changes due to irradiation are found in the ($R_{p0,2}$)- and (A_g)-values only these quantities are reported here. Even if one takes into account that the tensile testing conditions are slightly different, from *FIG's. 8 and 9* an increase of the Yield Stress and a decrease in Uniform Strain with increasing irradiation damage can be detected.

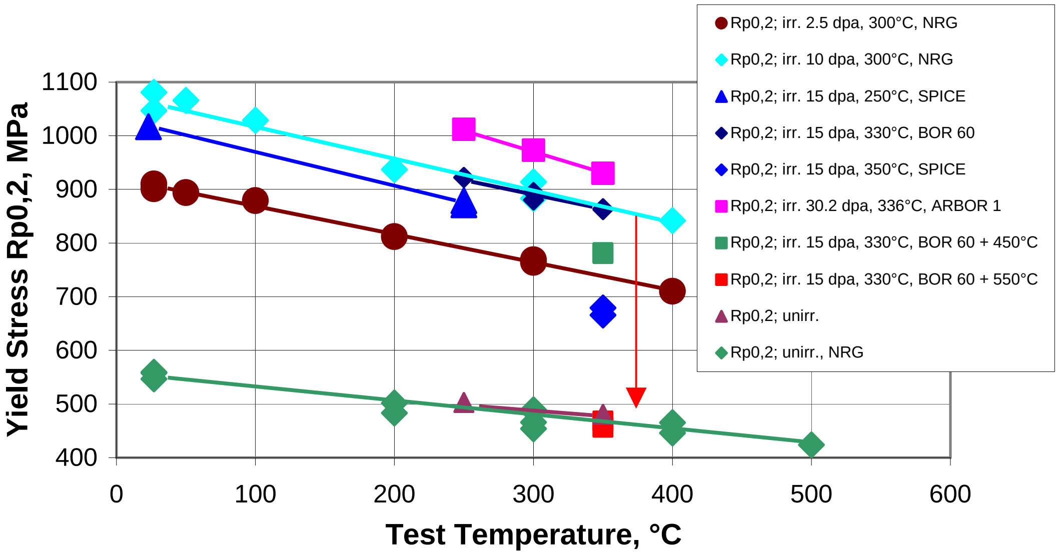

FIG. 8 Yield Stress ($R_{p0,2}$) behaviour of irradiated EUROFER 97 on dependence of test temperature compared to unirradiated data (the temperature in the legend indicates the irradiation temperature).

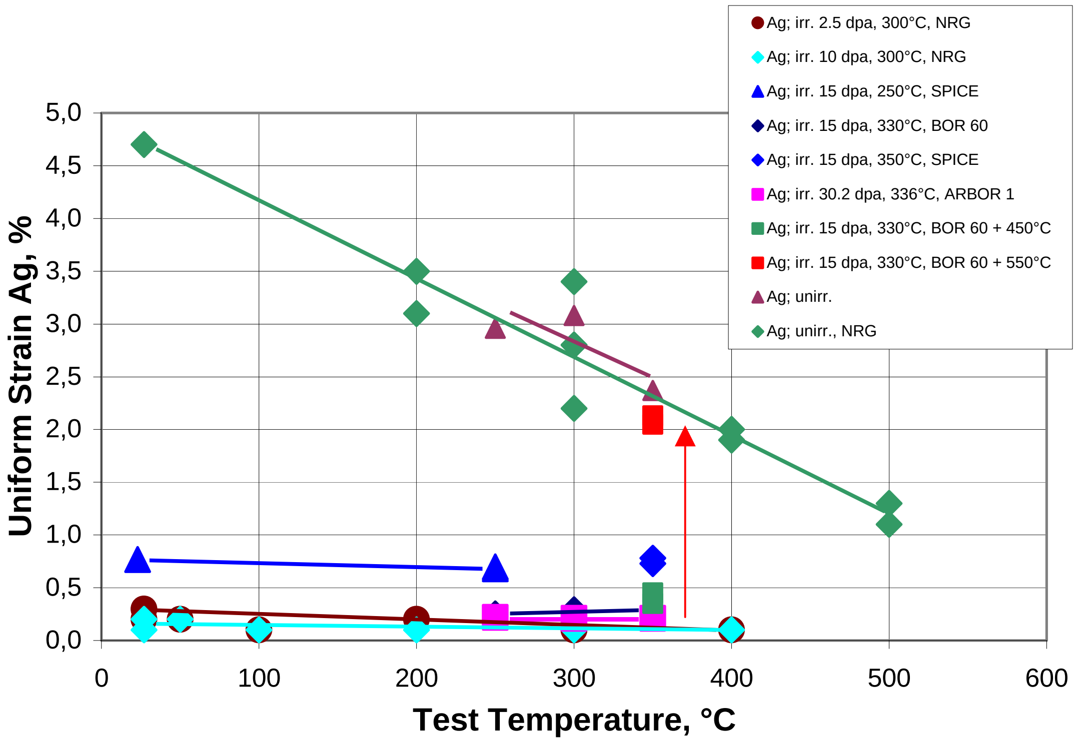

FIG. 9 Uniform Strain (A_g) behaviour of irradiated EUROFER 97 on dependence of test temperature compared to unirradiated data (the temperature in the legend indicates the irradiation temperature).

So the 2.5 dpa damage has the lowest increase of around 300 MPa in Yield Stress and the 30.2 dpa damage the highest increase of around 460 MPa in Yield Stress, that is nearly a du-

plication of the unirradiated quantity. The effect of the irradiation damage on the Uniform Strain is also considerable - mostly A_g - values below 0.5 % are reached - but does not depend so much of the damage dose as the stress values.
An exemplary postirradiation annealing experiment after the 15 dpa BOR 60 irradiation for 3 hours at 450°C or 550°C lead to the possibility to heal irradiation damage. But only at 550°C the Yield Stresses and Uniform Strains reached again virgin values (see red arrows in the above figures).

Acknowledgement

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References:

[1] AHLF J., ZURITA A., “High Flux Reactor (HFR) Petten – Characteristics of the Installation and the Irradiation Facilities”, Nucl. Sci. and Techn., EUR 15151 EN, (1993).
[2] SULABERIDZE V.S., et al., “Testing Techniques, Facilities and Devices for Irradiation Test Performance in the RIAR Research Reactors”, Status and Prospects for Irradiation Experiments in Russia/CIS, Proc. 32. Japan Workshop on 17. 11. 2004 at Oarai, Japan.
[3] SCHNEIDER H.-C., et al., “HFR irradiation programme”, Scientific Report FZKA 7117, (2004).
[4] GREENWOOD L.R., et al. “SPECTER: Neutron Damage Calculations for Materials Irradiations”, ANL/FPP/TM-197, 1985.
[5] GAGANIDZE E., et al., “Embrittlement Behavior of Neutron Irradiated RAFM Steels”, Contribution P04-9 to the 12th ICFRM, Santa Barbara, (2005), submitted for publication in J. Nucl. Mater..
[6] RIETH M., et al., “EUROFER97, Tensile, Charpy, Creep and Structural Tests“, Scientific Report FZKA 6911, October 2003.
[7] SCHNEIDER H.-C., et al., “Embrittlement behaviour of different international low activation alloys after neutron irradiation“, J. Nucl. Mater., **295** (2001) p. 16 – 20.
[8] SCHNEIDER H.-C., et al., “ Embrittlement behaviour of low activation alloys with reduced boron content after neutron irradiation“, J. Nucl. Mater. **321** (2003) 135 - 140.
[9] PETERSEN C., et al., “Fast reactor irradiations”, Scientific Report FZKA 7117, (2004).
[10] PETERSEN C., et al., “The ARBOR irradiation project”, J. Nucl. Mater. **307 - 311** (2002) 1655 - 1659.
[11] JITSUKAVA S. et al. “Development of an extensive database of mechanical and physical properties for reduced-activation martensitic steel F82H”, J. Nucl. Mater., **307 - 311** (2002) 179 – 186.
[12] RENSMAN J., et al., “Characteristics of unirradiated and 60°C, 2.7 dpa irradiated Eurofer 97”, J. Nucl. Mater., **307 - 311** (2002) 250 – 255.
[13] RENSMAN J.W., “NRG irradiation testing: report on 300°C and 60°C irradiated RAFM steels”, Final report on the EFDA Tasks TW2-TTMS-001a D 6 and TW2-TTMS-001b D 12, (2005).