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Summary

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Abstract

Micrometer sized samples could offer a possibility for advanced condition based monitoring in advanced nuclear plants. Tiny sample volumes can be removed either directly from a component or from test coupons for mechanical and microstructural analyses. Modern micro-sample preparation techniques (e.g. focused ion beam) together with micro-deformation machines (nano-indenter) allow both, the preparation of small thin foils for micro structural analyses and the testing of micrometer sized samples. Samples from well characterized irradiated reactor pressure vessel materials were tested with nano-indentation and micro-pillars to obtain information on irradiation effects on mechanical properties. Qualitatively, the results from the micro-samples compared well with the expectations. Quantitatively it was found that for annealed conditions the yield stress measured with micro-pillars was below the expectations whereas for irradiated samples a quite good agreement with expectations existed. The results can be related to differences in the stress-strain behaviour of the material. For large samples a pronounced yield stress is expected for the annealed condition which is no longer present in the irradiated samples.

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Contents

1 Introduction

Materials in advanced reactors as discussed in the Generation IV initiative are exposed to service conditions which are different from light water reactors (higher temperatures, higher dose, fast neutrons, and coolants other than water). The safe and reliable operation of such plants needs information on the condition of its critical components. For this purpose, methods for condition based monitoring must be developed. Analyses of test coupons or small samples taken directly from highly exposed locations of components are currently being considered as an option to support non-destructive evaluation of the condition of a component. Similar attempts have been made also in the past. The influence of specimen size on impact testing was extensively studied (e.g. [1]) with respect to analysis of surveillance samples however, these samples were still in the millimeter range. Small punch test were also investigated for toughness testing (e.g. [2]) and ball indentation tests were also considered for toughness monitoring in nuclear environments [3,4]. Nano-indentation and micro-pillar testing for components exposed to nuclear environments was also proposed [5, 6]. The reactor vessel is a very important component with respect to safety and conventional low alloy reactor pressure vessel steels are considered for currently discussed high temperature reactors (such as the US-NGNP [7]). An assessment of the capabilities of miniaturized samples for determining the reactor pressure vessel condition is therefore of interest for current as well as for future nuclear power plants. The irradiation embrittlement of reactor pressure vessels of light water reactors is currently a topic which attracts much interest in materials research particularly with respect to life extensions to 60 years or more. As the same material is proposed as a pressure vessel of the high temperature reactor the results are interesting for advanced nuclear plants, too. Sample material exists from surveillance investigations and other related projects such as the JRQ-samples prepared in an IAEA action [8]. Material of the ASTM A533 grade B class 1 steel that was designated as ‘JRQ reference steel’ served for many years as a radiation/mechanical property correlation monitor in a number of international and national studies of irradiation embrittlement of reactor pressure vessel steel. A comprehensive listing of material test data obtained on the JRQ manufacturing history and material properties can be found in the literature [8]. One set of samples consisting of reference material in an irradiated as well as annealed condition was thoroughly investigated with different methods to investigate irradiation hardening and eventual recovery procedures [9]. With these samples, which were investigated in the present paper too, atom probe analyses [10], extended X-ray absorption (EXAFS) studies [11] and Seebeck effect investigations [12] were also performed. In the current work for this deliverable the response of these samples to nano-indentation and micro-pillar testing was investigated.

2 Material

The samples were manufactured from JRQ IAEA standard material [8]. The chemical composition in summarized in Table 1. This is an A533 B class 1 plate 225 mm thick. After rolling, the plates were heat treated normalizing at 900 ^{o}C, quenching from 880 ^{o}C and tempering at 66 5^{o}C for 12 hours, then stress relieving at 620 ^{o}C for 40 hours. Irradiations carried out by PSI were performed in the 10 MW (t) SAPHIR reactor at 290 °C and at different neutron flux. Additional specimens were thermally annealed at 460 °C for 18 h when 50 % of the target fluence was reached, followed by re-irradiation (IAR). The detailed irradiation/annealing procedures of the samples investigated are summarized in Table 2. In this table “I” means irradiated, “A” means annealed, “R” means re-irradiated. The set of samples has been well characterized in the literature. Not only mechanical data (yield stress, rupture stress, Brittle ductile transition temperature, KI_C) are available [9, 13] but also microstructural investigations with an atom probe [10] and Seebeck effect measurements were performed [12]. Details of these treatments and further information concerning the reasons for this choice can be found in the literature. The samples were radioactive and therefore testing had to be performed using the following experimental Procedure:

The (active) samples were prepared for nano-indentation and micropillar testing at the hot laboratory of PSI. In order to reduce the radiation exposure to around 10 μSv•h^{-1} in 5 cm distance, small platelets of around 5x5x1 mm^3 were extracted from the bulk material, significantly reducing the radiation exposure for the operators and the equipment.

Nano-indentation was performed with an MTS (today Agilent) Nano Indenter G200 using a diamond Berkovich tip. The measurements were performed in the continuous stiffness mode (CSM, see [14]) with a frequency of 45 Hz and an amplitude of 2 nm. The maximum depth of the indentation was 2000 nm approached at a strain rate of 0.05 s^{-1}.

Square micro-pillars with a 2 x 2 μm^2 cross section and a height of about 4 μm were cut with a focused ion beam (FIB) of the type NVision from Carl Zeiss NTS. Similar to the procedure described in [5], first a larger 6 x 6 μm^2 pillar was extracted, in order to form the later platform of the final pillar and to have better control in shaping it (visibility from side). Figure 1 shows a finished pillar prior to compression test.

The compressions of the pillars were also performed with the above mentioned MTS nanoindenter. However, in this case a diamond flat punch of 10 μm diameter was used.

The displacement rate was 5 nm•s^{-1} until a typical maximal compression of 1 µm was reached. Prior to the compressions, thermal equilibrium of the equipment was awaited until a drift rate of below 0.1 nm•s^{-1} was reached. The typical maximal loads were 5-10 mN. Figure 2 shows all the true stress strain curves demonstrating the reproducibility of the tests.

C	*Si*	*Mn*	*P*	*S*	*Cu*	*Ni*	*Cr*	*Mo*	*V*
0.18	*0.24*	*1.42*	*0.017*	*0.004*	*0.14*	*0.84*	*0.12*	*0.51*	*0.002*

Table 1: *Chemical compositions of JRQ steel plate.*

specimen designation	Fluence 1 (cm^{-2})	Annealing 1 T (K)	Annealing 1 t (h)	Fluence 2 (cm^{-2})	Annealing 2 T (K)	Annealing 2 t (h)
Ref.						
I	5.0x10^{19}	-	-	-	-	-
IA	5.0x10^{19}	735	168	-	-	-
IAR	0.85x10^{19}	735	18	0.85x10^{19}	-	-
IARA	0.85x10^{19}	735	18	0.85x10^{19}	735	168

Table 2: Summary of exposure conditions for the investigated samples

	Ref	I	IA	IAR	IARA
Yield stress [MPa]	470	580	490	500	n.a.

Table 3: Yield stress (derived from literature [9,13])

3 Results:

The results of the nano-indentation are shown in Figure 3. Between the reference material and the irradiated material (I) about 45% hardening was found. After annealing (IA) a clear reduction of the hardening could be determined. However, the hardness was still about 20% above the hardness of the reference condition.

For the IAR samples the irradiation hardening was much lower (17%) which would be consistent with the lower irradiation dose and the fact that an annealing treatment was done between the two irradiation steps. Additional heat treatment slightly reduces the hardening; however, also in this case the low value of the reference could not be reached.

Yield stresses were derived for the micro-pillar tests from the load-displacement curves using the exact geometry of the pillars. For the determination of the yield stress, the elastic line was determined and the first deviations from this line were taken as the yield points. Additionally the first derivatives were analyzed to obtain a consistent picture. The method was straight forward for almost all samples. In case of the IAR samples systematically two not very pronounced stress levels were found indicating deviation from the elastic line. In this case an average between the two was taken as yield stress. The results of the pillar tests are shown in Figure 4. Essentially a similar picture to the one of the nano-indenter measurements was obtained. Hardening at the highest dose was highest, though relative to the reference material considerably higher (about a factor of two) compared with the indentation.

After annealing a slightly higher yield stress was found compared with the reference. The differences between IAR and IARA are small for both cases and relative changes could be explained with data scatter.

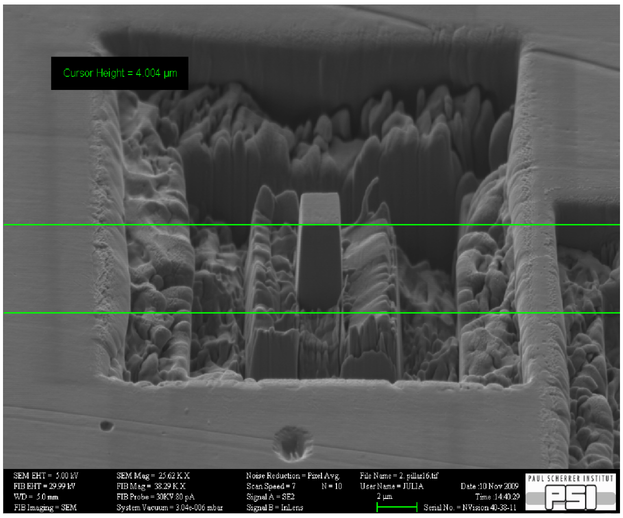

Figure 1: Micropillar used for the investigations described

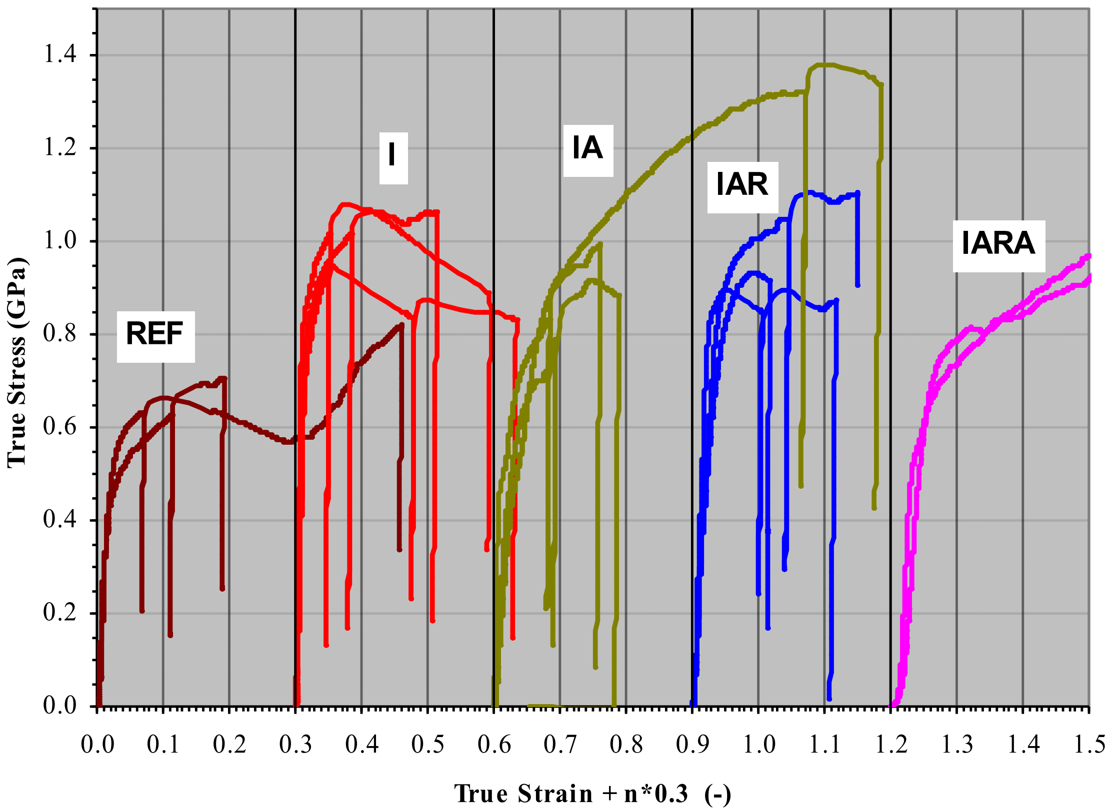

Figure 2: True strain and true stress of the pillar compression tests. The abscissa denotes the true strain plus n times 0.3, where n = 0 for the reference sample REF and n = 1, 2, 3 and 4 for I, IA, IAR and IARA respectively.

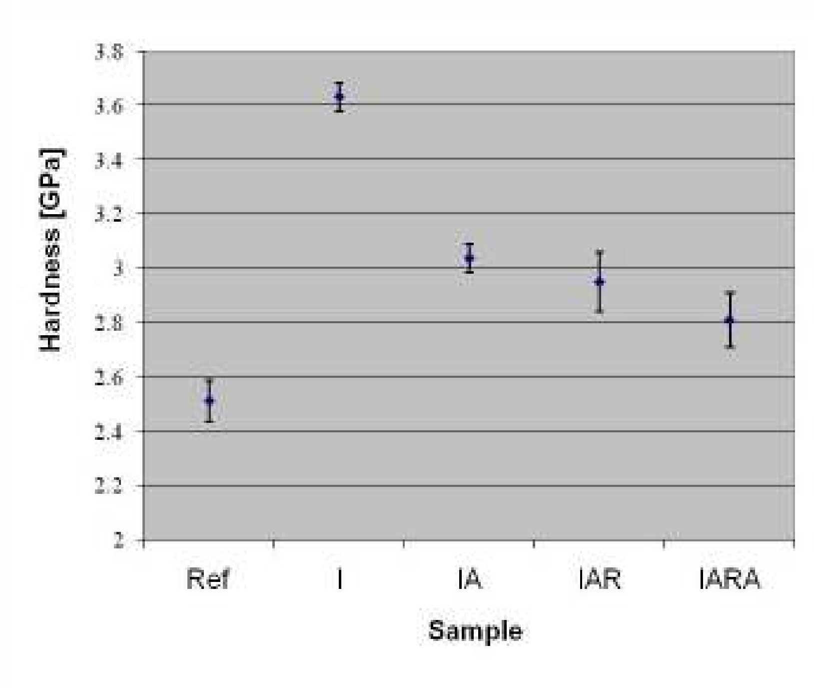

Figure 3: Hardness as a function of treatment of the samples. “Ref”-reference material, “I”- irradiated, “IA”- irradiated and annealed, “IAR” – irradiated, annealed, re-irradiated, “IARA” - irradiated, annealed, re-irradiated, annealedFor the IAR samples the irradiation hardening was much lower (17%) which

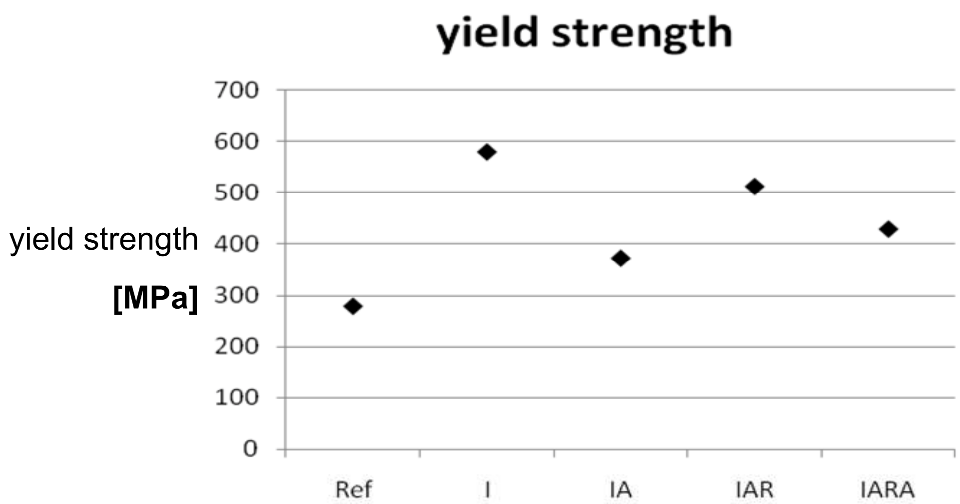

Figure 4: Yield stress determined with micro-pillars as a function of treatment of the samples. “Ref”-reference material, “I”- irradiated, “IA”- irradiated and annealed, “IAR” – irradiated, annealed, re-irradiated, “IARA” - irradiated, annealed, re-irradiated, annealed

4 Discussion

The results gained with different testing procedures are compared in Figure 5. They were plotted normalized to the values measured for the reference material. Yield stress values given in Table 3 were also included. The three different methods give qualitatively fairly consistent results. It can be seen that the values determined with the nano-indenter agreed very well with the values determined with large samples. The relative changes determined with the micro-pillars were considerably higher than the values determined from the large samples. For further explanation the absolute values between the yield stresses determined with large samples and with the pillars were made as shown in Figure 6.

The results are surprising. After irradiation the yield stress values were comparable (I, IAR). The reference state (Ref) and the state after irradiation with subsequent annealing (IA) showed significantly lower yield stress for the pillar tests. For IARA no yield stress value could be found in the literature. This result is considered very surprising because usually pillar tests are expected to give yield stresses either higher or comparable to large samples [15]. A possible explanation could be that low alloy steels show a pronounced yield stress but micro-plastic effects occur also below this value [16] which could be the reason for the low yield stresses found for the pillars. Samples after irradiation having a high defect density do not show this pronounced yield stress [17] and therefore the agreement between the yield stresses measured with pillars and the one expected from large samples disappears. Further examinations of pillar deformation in the scanning microscope will be performed in the future to prove this assumption and to better quantify the yield stress values determined for low alloy steel with micro-pillars.

It is considered, however, even without a quantitative explanation, that important condition based information could be derived from the miniature sample testing. Neutron irradiation with $5.0x10^{19}$ n/cm^2 causes strong irradiation hardening and consequently a considerable embrittlement of the material. A quantitative assessment of the shift in KI_C from the shifts in yield stress is currently difficult, as long as the influence of a pronounced yield stress on the results of the micro-sample tests is not fully understood. As the differences between irradiated and non-irradiated material are higher for the micro-samples the expected KI_C-shift would be higher and it would therefore be at least on the conservative side. Other conclusions which can be drawn from the micro-sample tests are consistent with the Atom probe analyses shown in [9]. The applied heat treatment recovers the radiation damage but not to a full 100 percent. The remaining hardening could be explained by copper-enriched precipitates formed during irradiation which are not completely dissolved during annealing [9]. After the irradiation, annealing and re-irradiation (IAR) treatment, a high number density of small copper-enriched precipitates was observed [9]. The presence of these small precipitates correlates with the re-hardening and consequently re-embrittlement of the steel. A significantly lower number density of ultrafine copper-enriched precipitates was observed after the second annealing treatment (IARA).

However, some larger copper-rich precipitates were also observed after this treatment. These results indicate that the ultrafine precipitates did not re-dissolve into the matrix but coarsened.

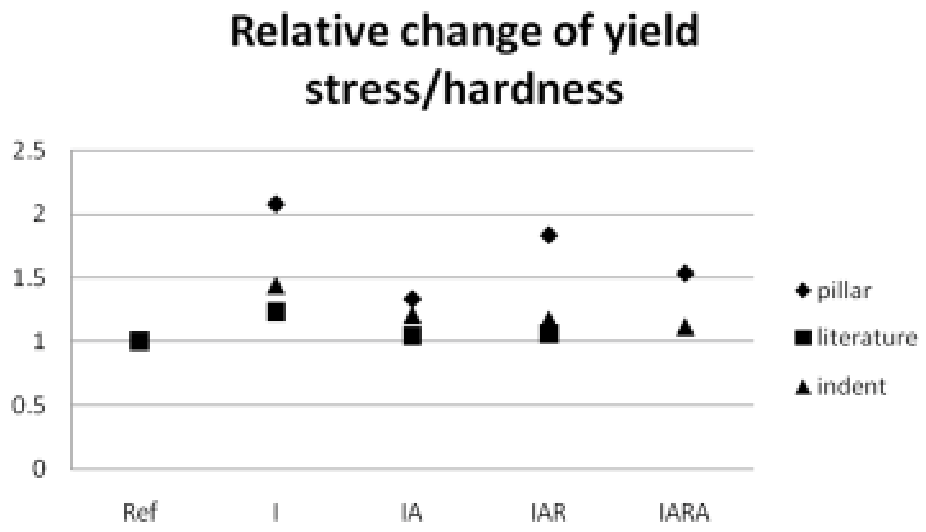

Figure 5: Relative change of (yield) stress for different test procedures. “Ref”-reference material, “I”- irradiated, “IA”- irradiated and annealed, “IAR” – irradiated, annealed, re-irradiated, “IARA” - irradiated, annealed, re-irradiated, annealed. Each measurement set is compared to its corresponding "Ref" sample result, being different for each method/source.

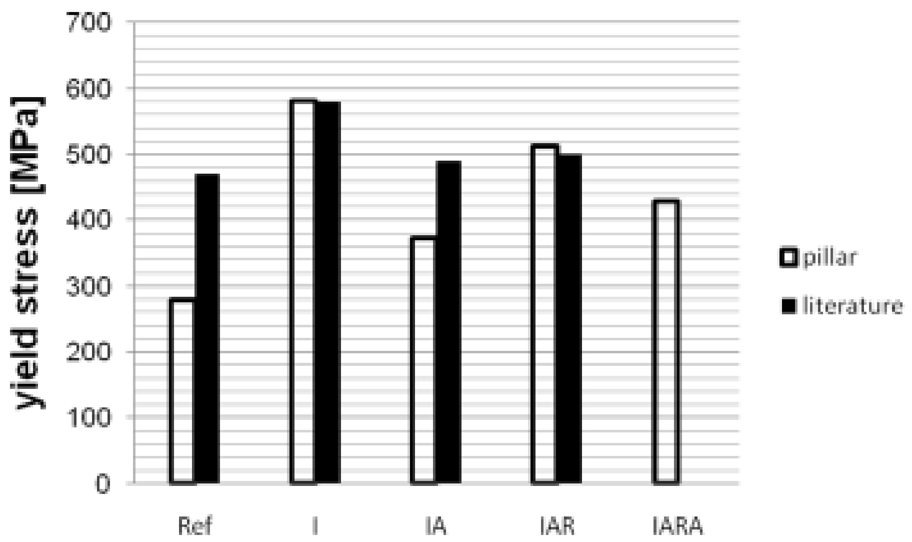

Figure 6: Comparison of yield stresses determined with micro-pillars and with conventional samples. “Ref”-reference material, “I”- irradiated, “IA”- irradiated and annealed, “IAR” – irradiated, annealed, re-irradiated, “IARA” - irradiated, annealed, re-irradiated, annealed

5 Conclusions

Irradiated and heat treated samples from an IAEA reference reactor pressure vessel material (JRQ) were analyzed with micro-sample techniques (nano-indentation, micropillar compression). The purpose was to determine the capabilities of these methods with respect to condition based monitoring of nuclear plants.

- The relative changes of yield stress from literature are well mapped with nano-indenter tests
- The yield stress of the samples after irradiation (without adjacent annealing) was in very good agreement with the yield stress determined with micro-pillars.
- The yield stress expected from micro-sample testing for the reference material and for the material after annealing is considerably below the values measured using large samples which could be typical for low alloy steel in conditions showing a pronounced yield stress.

The results encourage further evaluation of micro-sample testing for condition based monitoring of components in current and particularly in advanced nuclear plants. Further work is necessary to understand the discrepancy between the yield stress measured with micro-pillars and the ones determined with conventional samples for annealed material.

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