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TARGET AND STRUCTURAL MATERIALS UNDER DUAL-BEAM IRRADIATION

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The European Spallation Source (ESS) is planed to have a beam power of 5 MW (3.75 mA of 1.334 GeV protons). This is by a factor 30 higher than the available most powerful spallation source ISIS at Rutherford-Appleton Laboratory (RAL) in Great-Britain. This causes a damage rate of approx. 10 dpa (*displacements per atom*) per month in target materials (W or Ta) or structural materials (steels for beam windows, vessels). The high energetic protons produce, beside damage, high levels of transmutation products. One main characteristics of the spallation reaction is the simultaneous production of high levels of H and He. H and He are produced at rates of approx. 3000 appm/dpa and 200–300 appm/dpa, respectively. Both damage and transmutation products have detrimental effects on mechanical properties of target and structural materials.

The problem of radiation damage by high energy protons in different alloys has been recently reviewed by Ullmaier and Carsughi [1]. Main conclusion of this bibliographical work is that available data set is, as far, too small to allow any reliable prediction of lifetime of the different components.

At HMI, simulation irradiations using heavy ions and helium or hydrogen ions at the dual-beam facility are being carried out. The decisive advantages of such irradiation experiments are:

- (i) high damage levels and high helium or hydrogen contents can be produced within few hours. Such levels cannot be reached in a reasonnable time in any spallation facility;
- (ii) the irradiation parameters (damage rate, implantation rate, temperature) can be varied easily. Such fundamental study is indeed necessary. As shown in Fig. 1, displacement rates and He production rates are very different from one radiation field to another, even if one considers only spallation irradiations. It will be necessary to extrapolate existing data on the ESS typical irradiation field. Effect of changing the damage rate is generally well understood, as it has been investigated extensively, both theoretically and experimentally, in the Fusion Reactor Program. Such extensive studies are not available concerning the influence of helium/hydrogen implantation rate, especially in the low temperature region in which the spallation source is expected to operate;
- (iii) the irradiated specimens are not radioactive and can be analysed with the conventional methods available at HMI such as transmission electron microscopy.

Here, we report on some preliminary results obtained on tantalum as target material and on ferritic/martensitic steels (HT9 type steels or comparable steels), which are proposed to be used as structural materials. Goal of these investigations is to correlate the results with those obtained at spallation irradiated specimens and to investigate the unexplored field of simultaneous production of damage, helium and hydrogen at high levels.

1 Evolution of ferritic/martensitic steel under dual-beam irradiation

Ferritic/martensitic steels are under discussion for use as beam window material and confinement material for the ESS project. As discussed very extensively in a recent publication by Dai [2] (PSI, Switzerland), ferritic/martensitic steels of HT9 type (HT9 steel, German DIN 1.4970 steel, Manet steel or comparable German DIN 1.4926 steel) have excellent properties in terms of strength, thermal conductivity, thermal strength resistance, corrosion resistance, void swelling, and irradiation creep resistance, as compared to austenitic steels. The composition of the Manet steel (fusion program NET) and of the DIN 1.4926 steel (PSI beam window) are given in Table 1. For comparison, the composition of the F82H steel is given. This steel is now under consideration in the fusion program ITER as a low activation steel. It would be meaningful to investigate its possible use for ESS. In the expected service temperature region of interest for ESS (i. e. between 473 K and 773 K after G. Bauer [3]), only few experimental data are available on these steels. We have investigated the microstructural evolution of the HT9 type steel under dual-beam irradiation with two experimental techniques: (i) with field-ion microscopy with atom probe (FIM–AP) to get information on phase stability, and (ii) with transmission electron microscopy (TEM) to get information on helium bubble formation.

1.1 Phase stability

Concentration depth profiles of chromium as obtained by FIM–AP after dual-beam irradiation at temperatures between 673 K and 773 K are shown in Fig. 2. The samples were irradiated to a fluence of 50 dpa (displacement rate of 10^{-2}dpa s^{-1}) with a He/dpa implantation rate of 200 appm dpa^{-1}. In total, 1 at.% He were implanted. This corresponds to an ESS service time of approx. 1 year (full power). After irradiation at 673 K and 698 K, the chromium distribution is found to be statistically different from a random distribution we would expect for a solid solution. A detailed analysis yields a variation of the concentration amplitude ΔC of approx. 2.2 at.% to 2.8 at.%, respectively. At the temperatures of 723 K and 773 K, chromium clusters with diameters of about 4 nm with concentrations up to 25 at.% are observed. These results are reported extensively elsewhere [4]. We conclude from this study that the observed decomposition is caused by radiation-induced segregation in the lath structure of the annealed martensite. This effect has the detrimental consequence on mechanical properties to reduce the ductility and to increase the yield strength of the material [5,6].

1.2 Helium bubble formation

As helium is insoluble in metals, it will precipitate as bubbles. These bubbles, especially when they form at grain boundaries, have a detrimental effect on the mechanical properties and fatigue time of the materials. Experimental data on temperature dependencies of bubble densities observed under different experimental conditions have been compiled by Singh and Trinkaus [7] and demonstrate the key role of the helium generation rate for microstructural evolution. As the helium generation rate is different, even from one spallation environment to another, correlation of data is necessary. However, as pointed out by Singh and Trinkaus, lack of data in the low temperature regime, where ESS should operate, makes it impossible to confirm or infirm the prediction of theoretical models. This requires further studies. We have started respective investigations at HMI. Figure 3 contains, as examples, two TEM micrographs of the DIN 1.4926 steel implanted with He at 473 K and 773 K. Note that the size of the bubbles decreases as the temperature decreases. It renders such studies difficult as bubble size is at the limit of resolution of TEM. Investigations with high resolution electron microscopy (HREM) are planed, but are also difficult due to the high density of defects produced by the irradiation.

2 Hardness of tantalum and steel after helium implantation

Helium has been implanted at room temperature in both tantalum and DIN 1.4926 steel with energies up to 400 keV. The penetration depth of such ions is approx. 1 μm. Hence, the hardness must be measured with a nanoindenter (TU Berlin). The results are shown in Fig. 4, where the Vickers hardness is plotted as a function of the He concentration. In both cases, increase of the He concentration results in a drastic increase of the hardness of the specimens. This hardening might be interpreted in terms of a shift of the ductile-brittle transition temperature ΔDBTT. Such transition temperature appears in bcc alloys. It is possible to correlate the change in hardness with ΔDBTT. In the case of the HT9 type alloys, it has been done by Chen and Jung [8]. The observed increase in hardness of ΔHV$=$ 150 would correspond to a shift ΔDBTT$=$ 150 to 300 K. As the DBTT of such an unimplanted/unirradiated steel is just below room temperature, the DBTT under working condition would be around 400 to 570 K. Similar shifts have been observed under neutron irradiation [2].

3 Conclusion

The investigations carried out for environments which are different from the ESS spallation conditions (ISIS, Los Alamos, dual-beam irradiations) must be correlated to ESS conditions in order to be useful for the ESS design. Thus, the influence of the parameters, temperature, damage rate, implantation rate of helium and hydrogen, on bubble morphologies and, in turn, on the mechanical properties of the gas-containing materials must be investigated. It is clear from the

wt.%	ITER F82H steel	NET Manet steel DIN 1.4914	PSI window Steel DIN 1.4926
Cr	8.0	10.3	11.8
C	0.1	0.1	0.21
Si	0.1	0.14	0.36
Mn	0.1	0.75	0.47
P	0.005	0.005	0.016
S	0.002	0.0045	0.003
Ni	0.03	0.65	0.63
Mo	0.01	0.57	0.90
V	0.2	0.19	0.29
Nb	0.0002	0.14	<0.005
⋮			
W	2.0	—	0.02
Ta	0.04	—	—
Fe	bal		

Table 1: Composition of different Fe-Cr steels investigated in the fusion project and in the spallation project

experimental studies cited above that the combination of all damage sources, i.e. introduction of (i) defects, (ii) hydrogen, and (iii) helium at high levels and their interaction plays a determinant role in the evolution of the microstructure. In particular, the interaction of helium and hydrogen has to be studied [9]. Experiments at our dual-beam facility are in progress.

The morphologies and microstructures obtained under these simulation experiments must then be compared to the morphologies observed and to the mechanical properties of the spallation irradiated materials (investigations done at KFA Jülich by F. Carsughi and Prof. Ullmaier).

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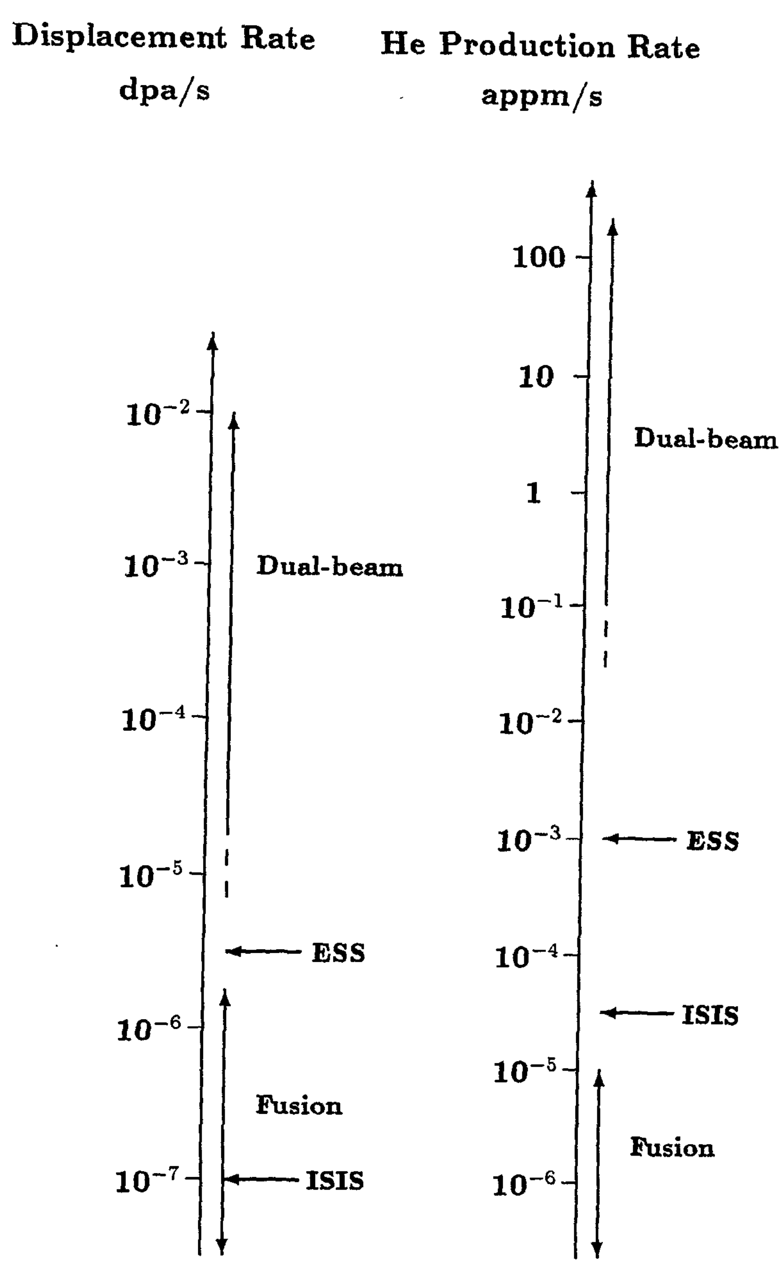


Figure 1: Comparison of different environments in terms of displacement rate and helium production rate (ESS: European Spallation Source; ISIS: Spallation Source at RAL).

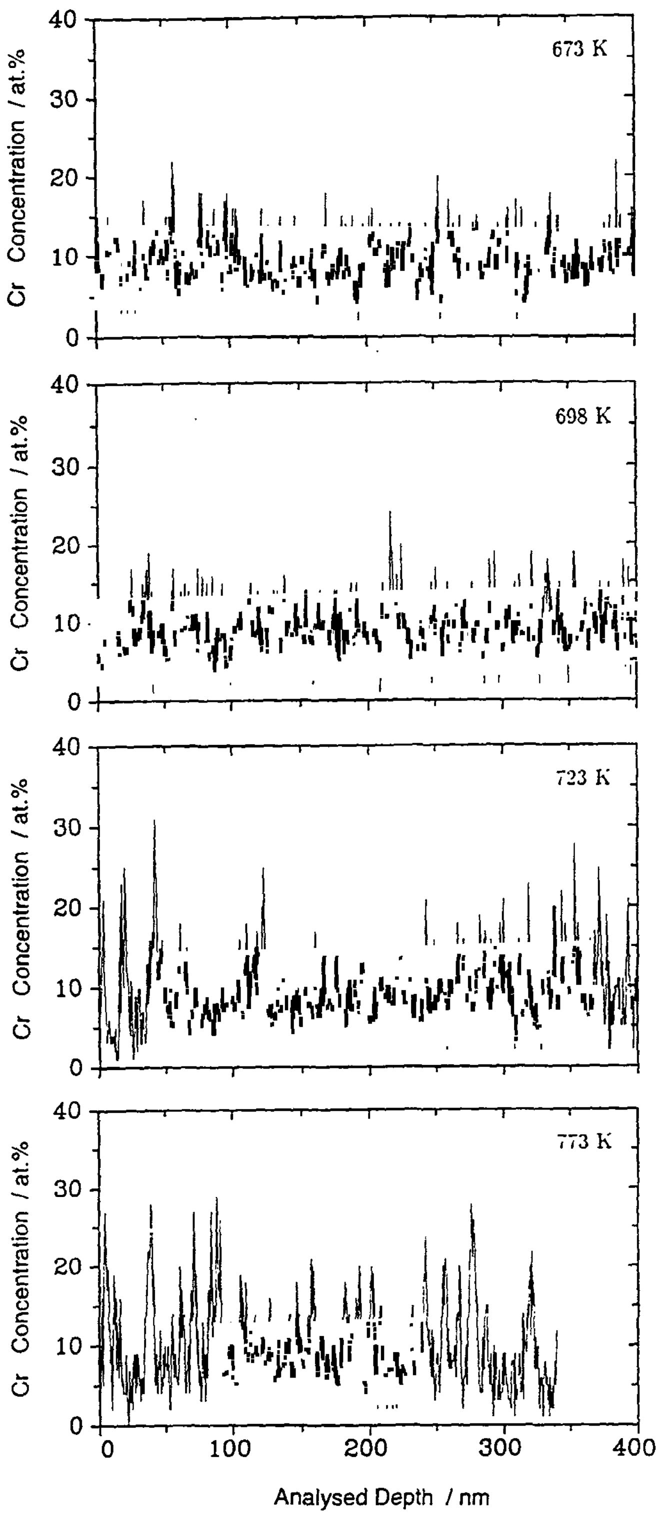


Figure 2: Chromium concentration profiles of Manet steel obtained after irradiation to 50 dpa implanted with 1 at.% helium with an implantation rate of 2 appm s^{-1} at temperatures between 400°C and 500°C. The implanted level of 1 at.% helium corresponds to approx. 1 year ESS service time (full power).

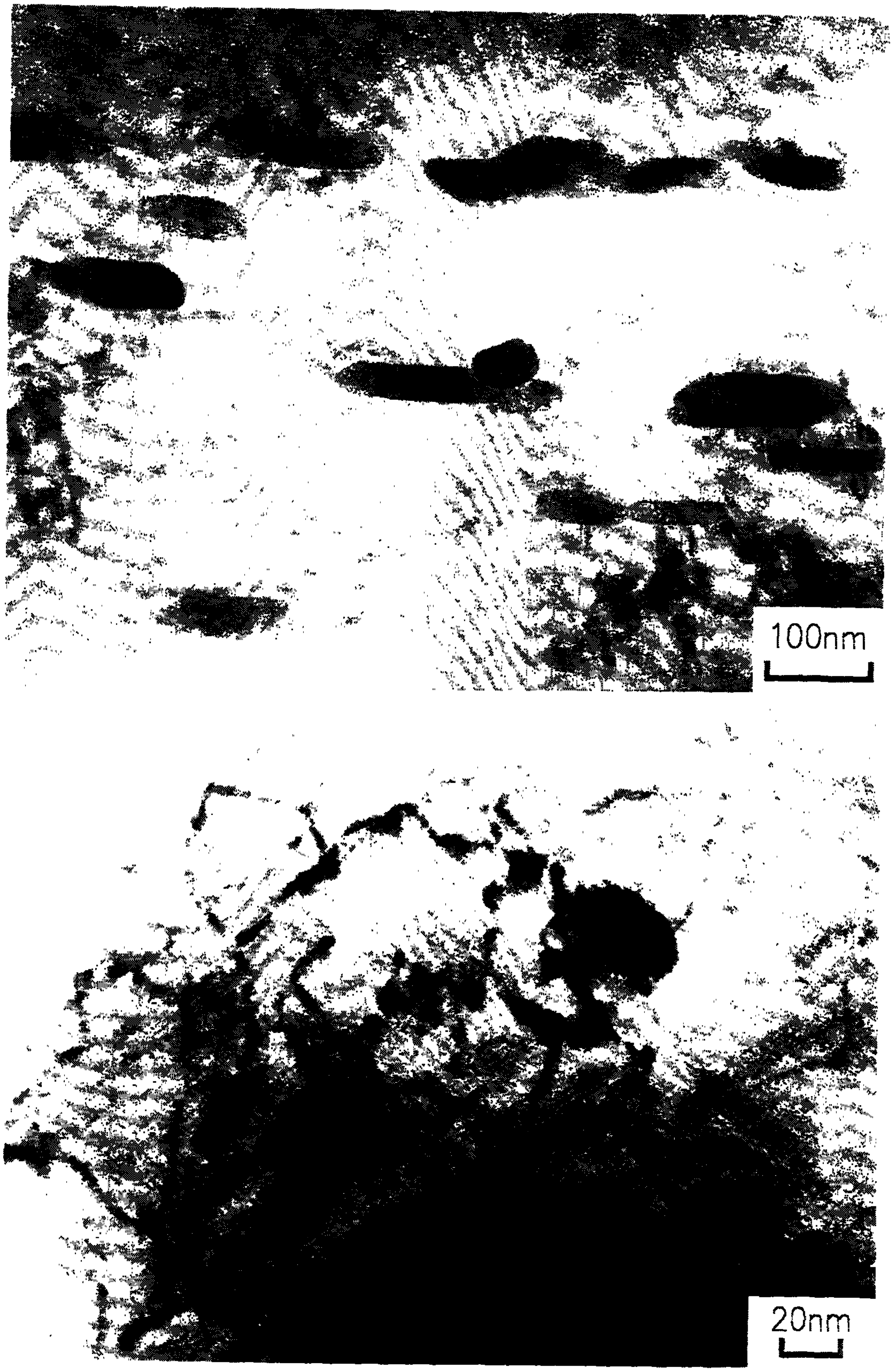


Figure 3: TEM micrographs of DIN 1.4926 steel obtained after irradiation to 30 dpa implanted with 0.6 at.% helium at a) 773 K and b) 473 K with an implantation rate of 2 appm s^{-1}. The implanted level of 0.6 at.% helium corresponds to approx. 6 months ESS service time (full power). In the upper image, helium bubbles are clearly visible and precipitate preferentially at grain boundaries, lath boundaries and precipitate/matrix interfaces. In the lower image, a high density of very small helium bubbles is visible.

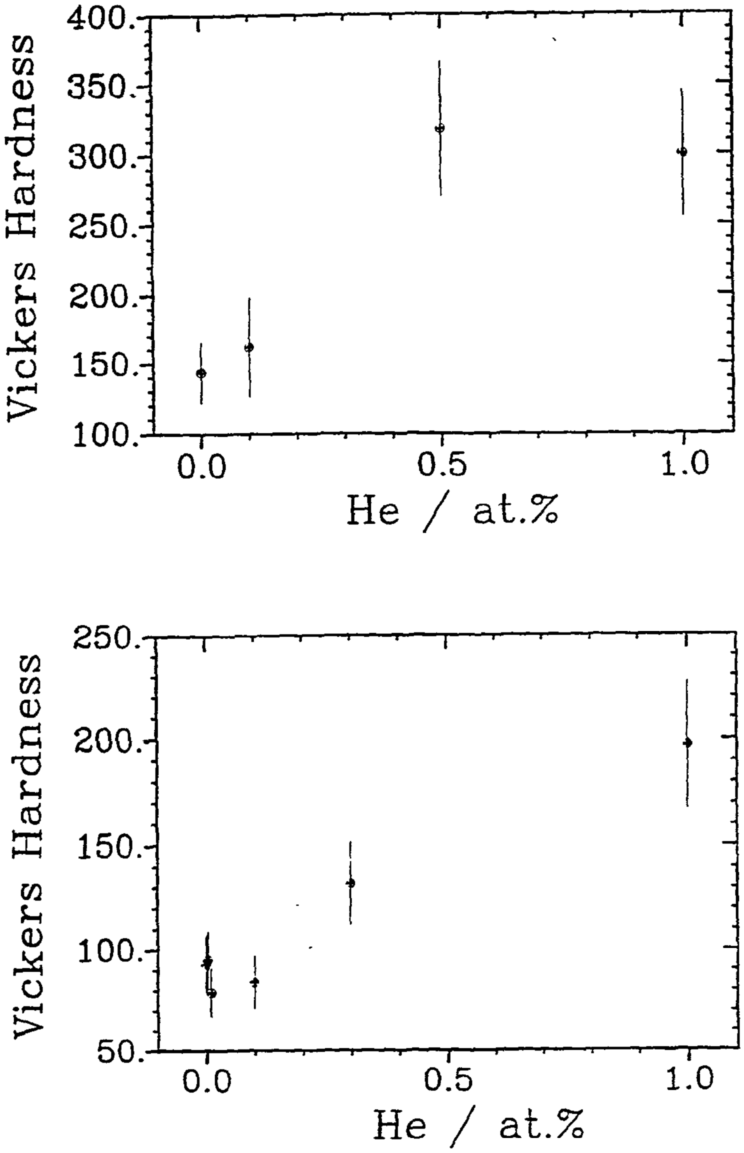
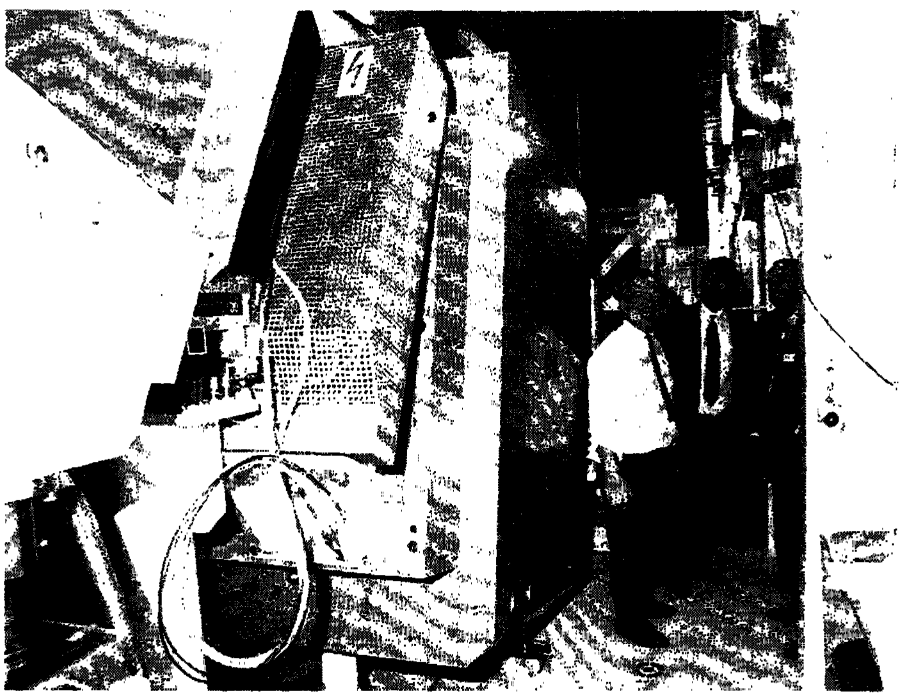


Figure 4: Vickers hardness of a) HT9 type steel and b) tantalum as a function of implanted He concentration. The materials were implanted at room temperature with an implantation rate of 10 to 50 appm s^{-1}. The implanted level of 1 at.% helium corresponds to approx. 1 year ESS service time (full power).



Accelerator Systems and Components