Influence of Light-Water-Reactor Working Medium on Brittlening of Cr-Mo-V Steels

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Summary

For reliability and residual time of life assessment of light-water-reactor /PWR/ pressure vessels the radiation embrittlement sensitivity and the influence of reactor coolant on the low-alloy steels behaviour has to be known. The failure of the internal corrosion resisting cladding of the reactor pressure vessel can result in important corrosion effects of the coolant caused by hydrogen embrittlement of steel.

The hydrogen embrittlement of Cr-Mo-V steel has been investigated in dependence on the electrolytic hydrogen charging parameters and on irradiation. It was observed that the degradation of plastic behaviour appears over a critical value 2.5 ppm of hydrogen, both for irradiated and unirradiated specimens. The superposition of radiation and hydrogen embrittlement can cause a full los of plasticity of the steel. In delayed fracture tests of unirradiated steel a limit threshold stress of 210 MPa was observed and below this value no specimen failure was ascertained neither for high current densities. The fractographical observation has shown, that the hydrogen embrittlement is characterized by quasi-cleavage fracture of hydrogen embrittlement and by combination of intergranular separation and transgranular cleavage.

1. Experimental

The experiments have been realized on the low alloy Cr-Mo-V steel which is used for light water reactor pressure vessels. The material was heat treated by oil quenching, high temperature tempering and long term stress relief annealing, which simulated the real thermal regimes of the pressure vessel production technology. The material composition in weight percents is as follows: C 0.15; Mn 0.36; Si 0,31; P 0.010; S 0.012; As 0.001; Cr 2.78; Ni 0.2; Mo 0.58; V 0.29; Cu 0.09.

Tensile tests were conducted at strain rate 8.3×10^{-4} s^{-1} on smooth cylindrical tensile specimens with diameter d = 4 mm and working section length 1 = 20 mm, the delayed fracture test on cylindrical 60 deg. V notched specimens with d_1 = 3.5 mm, d_2 = 5 mm, 1 = 17.5 mm, notch radius 0.05 mm. Both tests were performed at room temperature.

The specimens have been irradiated in an irradiation rig with He environment, located in the core of the VVR-S reactor at temperature of 130, 180, and 290 ^{o}C for a fast neutron fluence of 2.6 - 4.9×10^{23} $n.m^{-2}$ /E > 0.5 MeV/. Irradiated and unirradiated specimens were prior the tensile test hydrogen charged at room temperature in 1 N H_2SO_4 solution /with addition of 30 $ppm.dm^{-3}$ of As_2O_3/. During the test under constant loading the delayed fracture specimens have been charged at current densities of 10 to 3000 Am^{-2}. The hydrogen content in weight ppm has been determined by means of vacuum extraction of melted specimens in an BALZERS EAH-200 analyser, adapter for irradiated materials /1/.

2. Hydrogen and radiation embrittlement

The plastic behaviour of the unirradiated Cr-Mo-V steel depends on the content of hydrogen and can be influenced by the hydrogen charging parameters. Up to 2 ppm approximately of hydrogen by charging at current densities of 1 - 10 $A.m^{-2}$ no changes in plasticity have been observed. With hydrogen content reaching 2 - 2.5 ppm and at current densities 10 - 50 $A.m^{-2}$ there is a distinct decrease of elongation /Fig. 1/, reduction in area and fracture stress. The yield strength R_p 0.2 : 542 MPa and tensile strength R_m : 624 MPa remain unchanged in these described conditions.

The irradiation at temperatures 290 ^{o}C for $\phi = 3.4 \times 10^{23}$ $n.m^{-2}$ and 130 ^{o}C for $\phi =$ $= 4.9 \times 10^{23}$ $n.m^{-2}$ was followed by strengthening: R_p 0.2 increased by 19 % and 47 %, respectively, R_m increased by 12 % and 32 %, respectively. The irradiation embrittlement at 130 ^{o}C and 180 ^{o}C for $\phi = 2.6 \times 10^{23}$ $n.m^{-2}$ evolves a reduction of ductility /Fig. 1/. The effect of radiation damage on hydrogen embrittlement is inversely proportional to the irradiation temperature /2/.

The effect of radiation damage and hydrogen charging can be evaluated with help of a nondimensional factor Δ , determined by the difference of the original elongation of unirradiated and hydrogen uncharged steel A_5 and the elongation $A_{5,H,\phi}$ of the steel after irradiation and hydrogen charging, related to the original value of A_5. The dependence of the factor Δ on the current density /Fig. 2/ shows, that the decrease of the elongation up to the value of 5 $A.m^{-2}$ is determined by radiation embrittlement and beyond this value the contribution of radiation embrittlement is not uniform and a superposition begins of hydrogen and radiation embrittlement.

The hydrogen charging of specimens irradiated at 130 °C with current densities over 100 $A.m^{-2}$ causes a drastic hydrogen embrittlement, the fracture of specimens appear before the material tensile strength is reached /2/.

In the delayed fracture test the influence of the initial stress R on the time to fracture has been investigated with running hydrogen charging of unirradiated specimens at 10, 30, 100, 300, and 3000 $A.m^{-2}$ /3/. The lower threshold stress R_i was determined from the obtained curves and R_i as a function of current densities has been constructed /Fig. 3/. This function enables to determinate the magnitude of the limit threshold stress R_{id}, being 210 MPa. Below this limit the specimens are no failure even with higher current densities. The values given on the curve on Fig. 3 represent the contents of hydrogen in nondestructed specimens, which have been tested with threshold stresses R_i for 167 h.

3. Trapping and release of hydrogen

The Cr-Mo-V steel contains in the as delivered state 0.4 - 0.6 ppm and after irradiation 1.2 - 2.0 ppm of hydrogen. The dependence of the hydrogen content on current density of charging /Fig. 4/ presents two regions. In the first part of the curves the hydrogen content in irradiated and unirradiated specimens reaches 2 - 3 ppm. The following course of the hydrogen charging curve is determined by structural and radiation enhanced defects. With decreasing irradiation temperature, i.e. with increasing radiation damage of steel the hydrogen content is growing up after irradiation at 290 °C approximately 1 - 1.5 times, at 130 °C 3 - 4 times. In the same time the break point on the curves moves to lower values of current density.

The hydrogen release at room temperature has been studied after charging at 20 and 300 $A.m^{-2}$ for 1 h. The hydrogen content after 72 h degassing time decreased for unirradiated specimens nearly up to the original value /below 1 ppm/, for specimens irradiated at 290 °C up to half to the initial value after charging; at 130 °C and given conditions the content remained approximately unchanged /2/.

4. Fractography

4.1 Tensile test specimens /2/

- Ductile cup fracture /Fig. 5/, practically the only fracture mechanism is the ductile dimple rupture. It has been observed on unirradiated specimens up to 2 ppm H_2 and on specimens only irradiated at 130 - 290 °C.
- "Hydrogen" fracture, the dominating fracture mechanism being the quasicleavage caused by hydrogen emgrittlement /4/ - QC_{HE} /Fig. 6/. The fracture is initiated simultaneously in several points of the specimen cross-section, the initiation centres being the inhomogeneities of the rough inclusion type, of cracks along the grain boundaries and at high

contents of intergranular separation in the boundary intersection of three grains /Fig. 7/. Starting from those inhomogeneities the failure propagates radially in a plane approximately perpendicular to the load axis and forms characteristical oval facets /Fig. 6/ on specimens charged over 3 ppm, unirradiated and irradiated at 290 °C.

- Brittle fracture, fracture plane is perpendicular to the load axis /Fig. 8/. The failure is initiated on the specimen surface /or near the surface/. The initial stade is characterized by intergranular separation, the later one by the combination of cleavage and intergranular rupture. The fracture occured on specimens irradiated at 130 °C and hydrogen charged at current densities of 100 $A.m^{-2}$ and more.
- The surface embrittlement, it is manifested by cracking of the sample surface /Fig. 9/ and has been ascertained on specimens irradiated at 130 °C and charged at 5 – 50 $A.m^{-2}$. The maximum of cracks was observed for charging at 5 $A.m^{-2}$, the deepest failures /up to 0.5 mm/ for charging at 50 $A.m^{-2}$. With increasing hydrogen content the fracture changes from ductile cup form through shear-oblique to brittle fracture perpendicular to load axis.

4.2 Delayed fracture samples

The fracture of notched specimens which have been used starts always from the surface or near from it. The controlling mechanism is the intergranular fracture /Fig. 10, area A/, together with QC_{HE} micromechanisms, dimple failure /area B/ and transcrystalline fracture /area C/. The percentage of the intercrystalline fracture face IR as a function of applied load in the range $R_m - R_{id}$ can be in the first approximation expressed as follows:

$$IR\ /\%/ = 10.5 - 0.98\ R \qquad /MPa/$$

5. Conclusion

Cr-Mo-V steel both in irradiated and unirradiated state is sensitive to hydrogen embrittlement at hydrogen content above 2.5 ppm. A strong decrease of reduction in area and elongation over this value is determined by hydrogen content, resp. current density of hydrogen charging. The irradiation at 130 °C and 180 °C /for $> 2.6 \times 10^{23}\ n.m^{-2}$, $E > 0.5$ MeV/ followed by charging gives the possibility of a mutual superposition of hydrogen and radiation embrittlement. It is proved by the decrease of elongation and reduction in area with hydrogen content over 8 ppm, when a full loss of plasticity may appear.

The irradiated and unirradiated specimens fail with ductile fracture. Hydrogen contents over 2.5 ppm in unirradiated specimens and those irradiated at 290 °C leads to a fracture characterized by QC_{HE} and at hydrogen charged samples irradiated at 130 °C the fracture is brittle and the controlling mechanism is the intercrystalline cleavage or the combination of the intercrystalline and transcrystalline cleavage.

The decrease of irradiation temperature from 290 to 130 °C substantially increases the hydrogen content. It means, that the effect of radiation induced defects on trapping and maintaining the hydrogen appears to be substantially higher in comparison to structural defects of unirradiated steel.

The time to fracture depends on the current density of charging and initial stress. The influence of hydrogen appears at hydrogen contents over 3 ppm. The determining mechanism of fracture is the intercrystallic separation and his relative area on the fracture face depends on the initial stress.

The content of hydrogen of cca 2.5 ppm has an analog effect on embrittlement of irradiated and unirradiated stell A 302B /5/, unirradiated steel A 533B and A 542 /6/.

References

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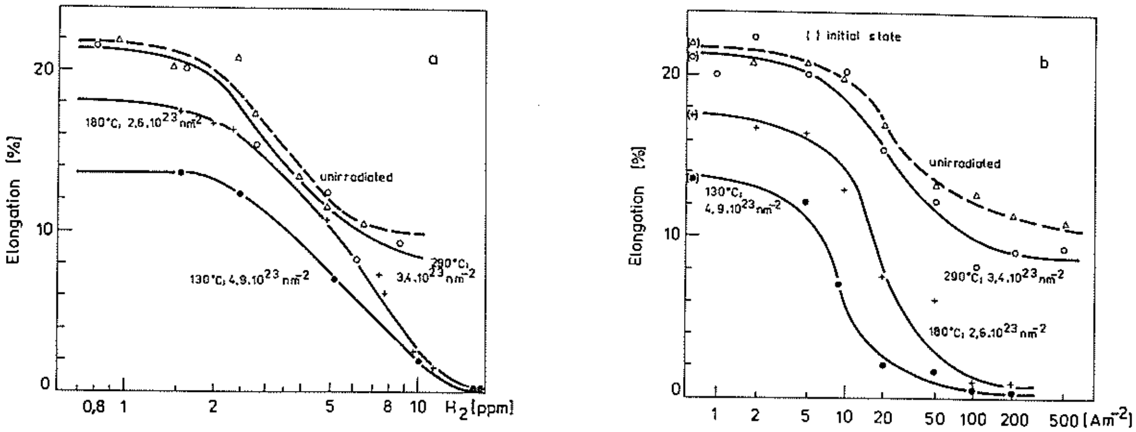

Fig. 1: Elongation of irradiated and unirradiated steel /charging time 1 h/ as a function of hydrogen content /a/ and current density /b/.

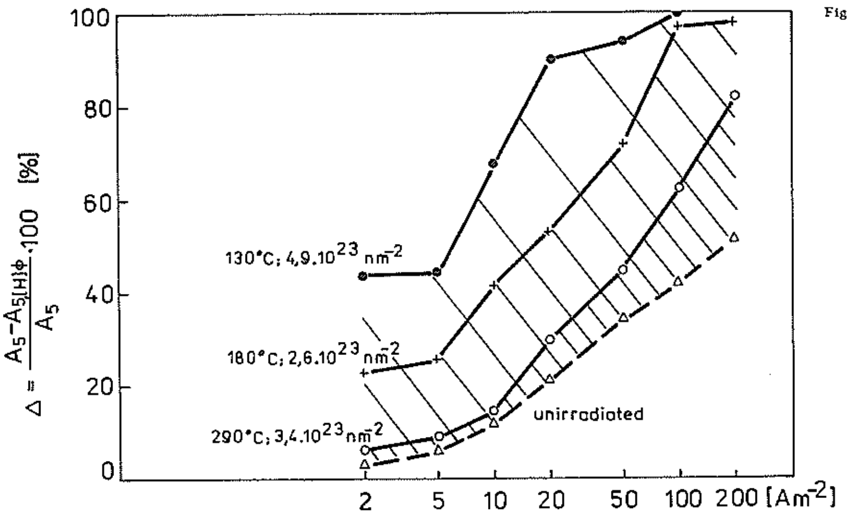

Fig. 2: The parameter Δ as a function of current density of irradiated and unirradiated specimens charging.

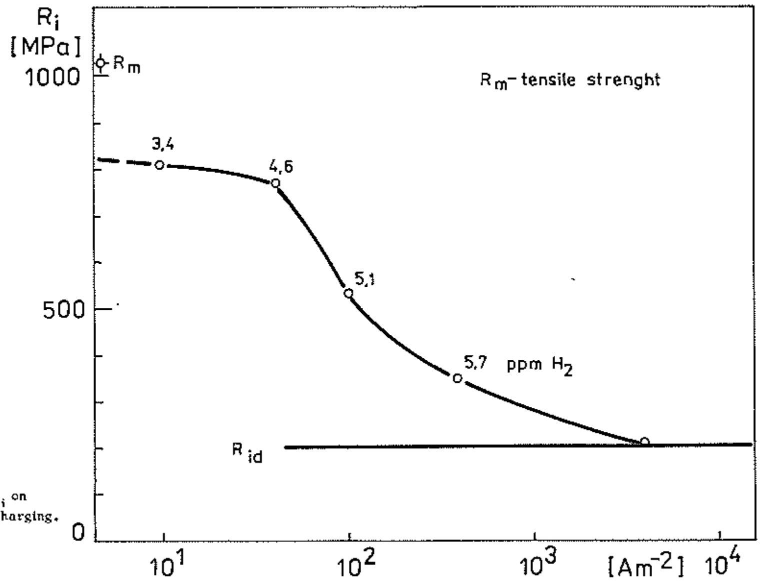

Fig. 3: The dependence of lower threshold stress R_i on current density of unirradiated specimens charging.

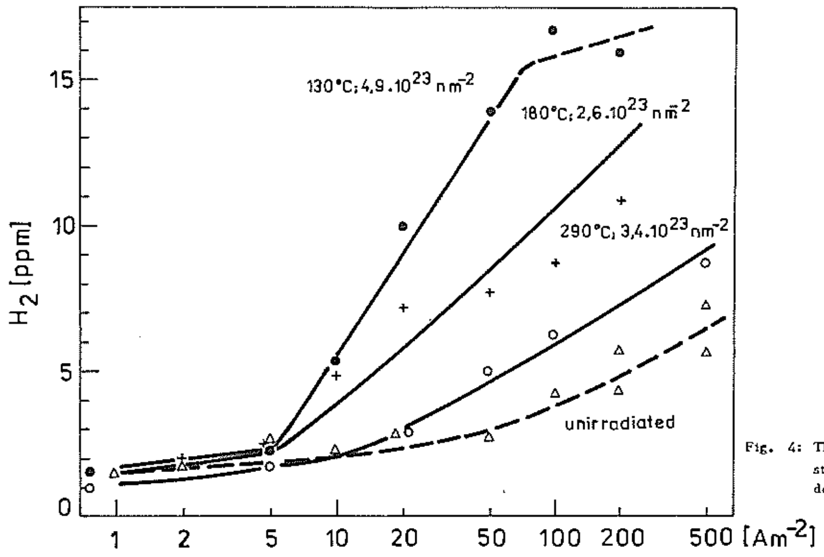

Fig. 4: The hydrogen content in irradiated and unirradiated steel /charging time 1 h/ as a function of current density.

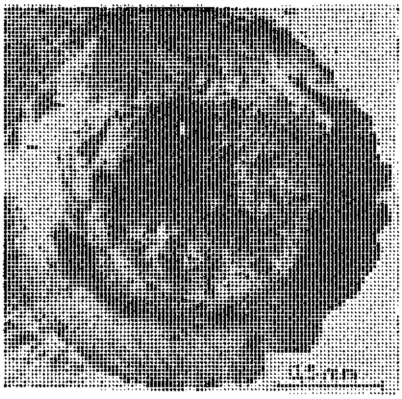

Fig. 5: Ductile fracture, specimen irradiated for 4.9×10^{23} $n.m^{-2}$, $T_{ir} = 130$ °C, not hydrogen charged.

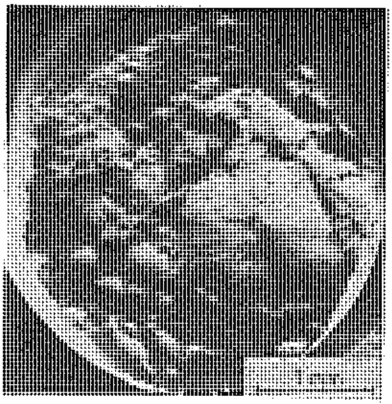

Fig. 6: "Hydrogen" fracture, unirradiated sample, charged - 6 ppm H_2.

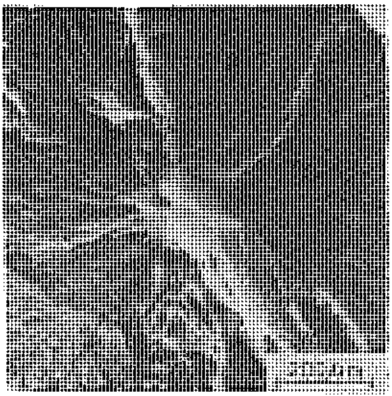

Fig. 7: QC_{HE} facets, unirradiated specimen, hydrogen charged - 6 ppm.

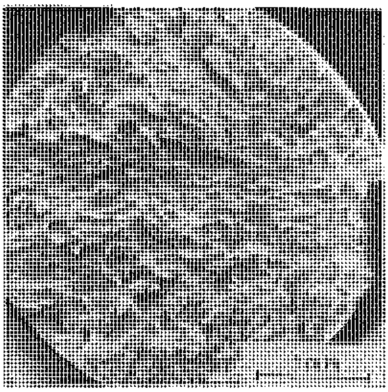

Fig. 8: Brittle fracture, irradiated specimen for 4.9×10^{23} $n.m^{-2}$, $T_{ir} = 130$ °C, charged 100 $A.m^{-2}$.

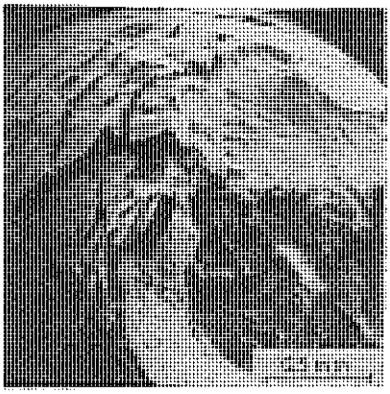

Fig. 9: Surface cracks, irradiated specimen for 4.9×10^{23} $n.m^{-2}$, $T_{ir} = 130$ °C, charged 5 $A.m^{-2}$.

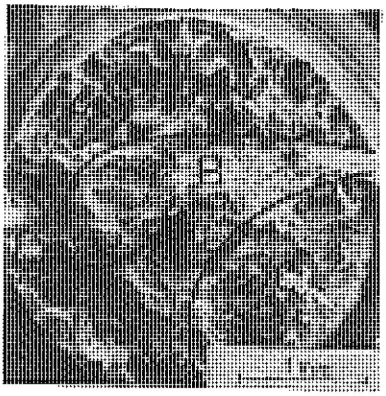

Fig. 10: Unirradiated specimen, hydrogen charged 300 $A.m^{-2}$ with load of R = 713 MPa, time to fracture 0.8 h.