

Disclosure of the Generation and Accumulation of the Hydrogen in Steel and Graphite Irradiated by Neutrons in Inert Environment



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Introduction

It is known that in traditional power engineering hydrogen may be one of the first primary source of equipment damage [1]. This problem has high actuality for both nuclear and thermonuclear power engineering [2]. Particularly reactor pressure vessels (RPV) of the WWER-440/230 project were manufactured without stainless cladding that were in contact with primary circuit water and accessible for hydrogen as a product of RPV wall corrosion. Analysis of the combined radiation-hydrogenation embrittlement of the 48TS type vessel steel was performed in [3] where at the mention of the American [4] and own data question concerning unknown source of hydrogen in metal that was irradiated in nuclear reactor in hermetic ampoules (was named as "irradiation-produced hydrogen" (IPH) was raised.

Material and Methods

Table 1 lists chemical composition of the RPV steel used (48TS type). A-543 type US steel takes for comparison. 4% solution of H_2SO_4 was used for additional electrolytic hydrogenation of the specimens (current density $0,1A/cm^2$). Hydrogen concentration was determined by thermal degassing method at temperatures up to $1000\text{ }^{\circ}C$ with gas chromatograph (thermal conductivity detector) registration of gas released.

Table 1: Chemical composition of the RPV steels 48TS and A-543 (%mass).

Type	C	Si	Mn	P	S	Cu	Cr	Mo	Ni
48TS	0,16	0,30	0,43	0,014	0,011	0,11	2,75	0,67	0,16
A543	0,14	0,18	0,20	0,011	0,015	0,07	1,60	0,50	3,01

Experimental Result and Discussion

Determination of the hydrogen content in the irradiated steel fulfilled in the USA went to unexpected result: hydrogen content noticeably exceeded the quantity rated at (n,p) transmutation reaction: less than 0,1ppm. Results of the IPH concentration in steel analysis carried out in the USA are shown in Table 2 [4]. One can see that the greater the fast neutron fluence (FNF) the greater the hydrogen content.

Table 2: Dependence of the IPH concentration in steel versus FNF ($E > 1MeV$).

FNF, $\times 10^{18}cm^{-2}$; $t_{irr}=225-300\text{ }^{\circ}C$.	0	7	200	400
IPH concentration, ppm; $t_{degassing}^{\circ}=1000\text{ }^{\circ}C$.	0,2	0,9	1,7	2,1

Ageing of the steel at $100-325\text{ }^{\circ}C$ for 48 hours revealed that IPH is not diffusible up to irradiation temperature that is IRH are in the irradiation produced traps. Inasmuch as IPH at temperatures of mechanical tests was immovable indicated values were subtracted from total quantity of hydrogen measured.

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In I.V. Kurchatov Institute at several experiments was determined that steel specimens irradiated at relatively low (100-140 °C) temperatures in sealed Ar contained ampoules hydrogen content was many times higher relatively initial content but was independent on FNF (Table 3) [3]. Degassing kinetics are plotted in Figures 1 & 2.

Table 3: Dependence of the IPH concentration in steel versus FNF ($E > 0,5 \text{ MeV}$).

FNF, $\times 10^{18} \text{ cm}^{-2}$; tirr.=100-140 °C.	0	100	170	190	270	450	500
IPH concentration, ppm; $t^{\circ}_{\text{degassing}} = 300 \text{ }^{\circ}\text{C}$.	0,2	2,9	4,3	13,3	24,9	9,8	3,1

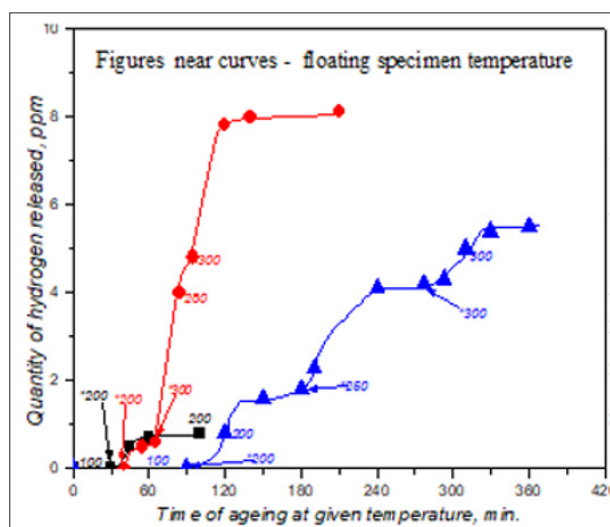


Figure 1: IPH degassing kinetics for irradiated steel ($4,5 \times 10^{20} \text{ cm}^{-2}$ at 140 °C).

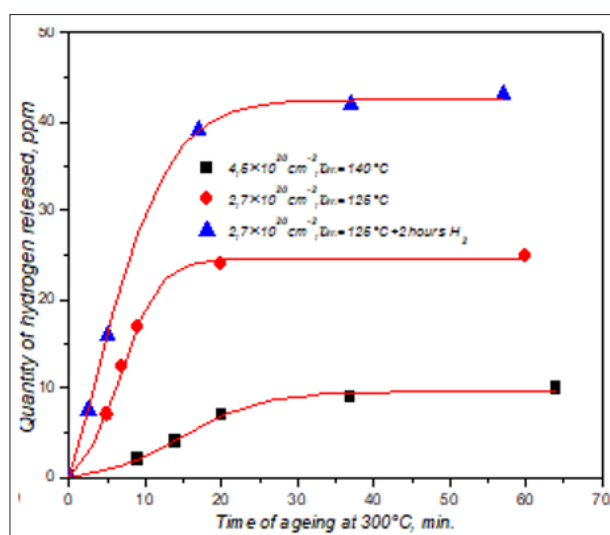


Figure 2: Hydrogen degassing kinetics for irradiated and irradiated+hydrogenated steel.

As one can see from table 1 that RIH discharge starts when heating temperature exceeds the irradiation temperature. It means that RIH is accumulated in radiation defects (traps). Rather later data appear on unexpectedly high hydrogen concentrations in stainless steels irradiated in BWR type reactors [5] and high generations of hydrogen and helium in nickel [6]. Surprisingly high hydrogen concentrations were revealed in irradiated graphite [7].

Conclusion

It is necessary to look for enigmatic source of hydrogen especially because in frame of inspections numerous flows were detected in the forged rings of the reactor pressure vessels in the Belgian nuclear power plants Doel 3 and Tihange 2 [8]. The owner Electrabel claimed that flaws were “most likely” hydrogen flakes. One of the unobvious but probable initial hypothesis on enigmatic source of the hydrogen in operating nuclear reactor is generation of protons as a product of beta-decay of free neutrons (lifetime $\sim 15 \text{ min.}$) [9].

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