

Research Article

Hydrogen Generation and Accumulation in Steel and Graphite Irradiated in Inert Environment

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Abstract

In traditional power engineering hydrogen may be one of the first primary source of equipment damage. This problem has high actuality for both nuclear and thermonuclear power engineering. Study of radiation-hydrogen embrittlement of the steel raises the question concerning the unknown source of hydrogen in reactors. Later unexpectedly high hydrogen concentrations were detected in irradiated graphite.

It is necessary to look for this source of hydrogen especially because hydrogen flakes were detected in reactor vessels of Belgian NPPs.

As a possible initial hypothesis about the enigmatical source of hydrogen one can propose protons generation during beta-decay of free neutrons поскольку inasmuch as protons detected by researchers at nuclear reactors as witness of beta-decay of free neutrons.

Keywords: Nuclear Reactor, Steel, Graphite, Source of Hydrogen

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 was manufactured without stainless cladding that is 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.

Materials 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 H₂SO₄ was used for additional electrolytic hydrogenation of the specimens (current density 0,1A/cm²).

Hydrogen concentration (C) was determined by thermal degasation method at temperatures up to 1000°C with gas chromatograph (thermal conductivity detector) registration of gas released.

Experimental Results 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 0,1 ppm.

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

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.

Ageing of the steel at 100-325°C during 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.

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 is plotted in Figure 1, 2.

As one can see from Fig.1 that RIH discharge starts when heating temperature exceeds the irradiation temperature. It means that RIH is accumulated in radiation defects (traps).

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

FNF, ×10 ¹⁸ cm ⁻² ; t _{irr} = 225-300°C.	0	7	200	400
IPH concentration, ppm; t ^o _{degassing} = 1000°C.	0,2	0,9	1,7	2,1

Table 3: Dependence of the IPH concentration in steel versus FNF (E>0,5MeV)

FNF, ×10 ¹⁸ cm ⁻² ; t _{irr} = 100-140°C.	0	100	170	190	270	450	500
IPH concentration, ppm; T _{degassing} = 300°C.	0,2	2,9	4,3	13,3	24,9	9,8	3,1

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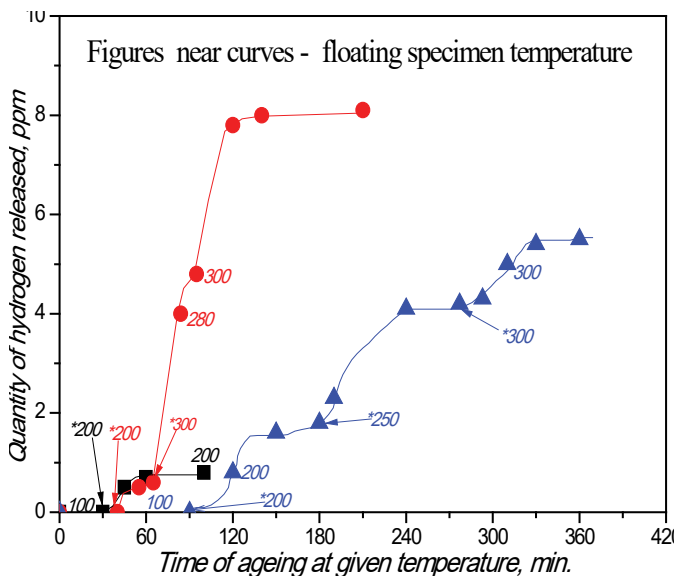


Figure 1: IPH degassing kinetics for irradiated steel ($4,5 \times 10^{20} \text{cm}^{-2}$ at 140°C). kinetics

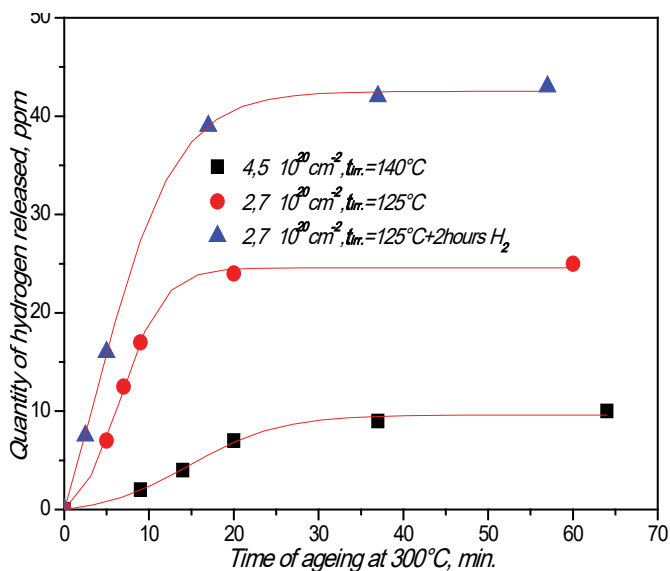


Figure 2. Hydrogen degassing for irradiated and irradiated+ hydrogenated steel.

Rather later data appear on unexpectedly high hydrogen concentrations in stainless steels irradiated in BWR type reactors (Figure 3 [5]) and high generations of hydrogen and helium in nickel [6].

Surprisingly high hydrogen concentrations were revealed in irradiated graphite [7]. Table 4 presents the results of the thermal degasation of the GR-280 type graphite.

As it is seen hydrogen concentrations in irradiated specimens is one-two orders of magnitude higher than in unirradiated ones.

It is necessary to look for enigmatic source of hydrogen especially because in frame of inspections numerous flaws were detected in the forged rings of the reactor pressure vessels in the Belgian nuclear power plants Doel 3 and Tihange 2 [8,9]. The owner Electrabel claimed that flaws were “most likely” hydrogen flakes.

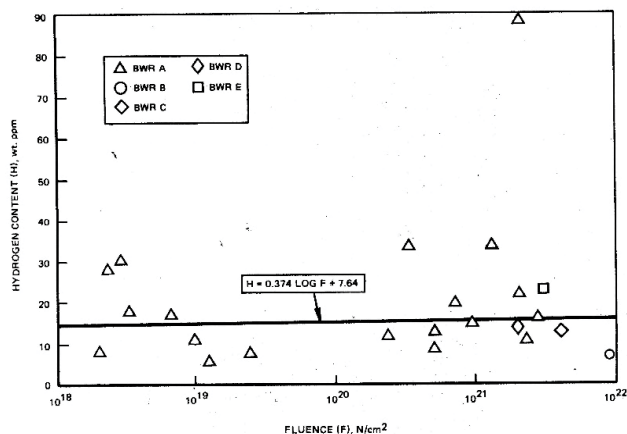


Fig. 2—Hydrogen content of irradiated Type-304 SS as a function of fast ($E > 1 \text{ MeV}$) neutron fluence.

Figure 3: IPH content in 304 SS versus FNF ($E > 1 \text{ MeV}$).

Table 4. Results of the thermal degassing of the GR-280 type graphite

FNF, cm^{-2} ($E > 0,5 \text{ MeV}$)	t_{irr} , °	Quantity of hydrogen released, ppm		
		$t_{\text{degassing}}$, °		Total ppm
		1000	1800	
0		2,2	4,2	6,4
		1,5	2,9	4,4
		20,8	-	20,8
1'	500	49,7	-	49,7
		41,2	1342	1383
		19,5	864	883
		-	369	369
2'	1100	-	327	327
		-	-	-

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 $\sim 15 \text{ min.}$) [10].

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