

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

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Abstract

It is known that 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. 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 where at the mention of the American 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.

Introduction

Materials and Methods

Table 1 lists chemical composition of the RPV steel used (48TS type). A-543 type US steel takes for comparison.

Type	C	Si	Mn	P	S	Cu	Cr	Mo	Ni
48T	0,1	0,3	0,4	0,01	0,01	0,1	2,7	0,6	0,1
S	6	0	3	4	1	1	5	7	6
A54	0,1	0,1	0,2	0,01	0,01	0,0	1,6	0,5	3,0
3	4	8	0	1	5	7	0	0	1

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

4% solution of H_2SO_4 was used for additional electrolytic hydrogenation of the specimens (current density 0,1A/cm²). Hydrogen concentration was determined by thermal degassing 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. Results of the IPH concentration in steel analysis carried out in the USA are shown in Table 2. One can see that the greater the fast neutron fluence (FNF) the greater the hydrogen content.

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

Table 2: Dependence of the IPH concentration in steel versus FNF ($E > 1 \text{MeV}$)

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). Degassing kinetics are plotted in Figures 1, 2.

FNF, $\times 10^{18} \text{cm}^{-2}$; 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

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

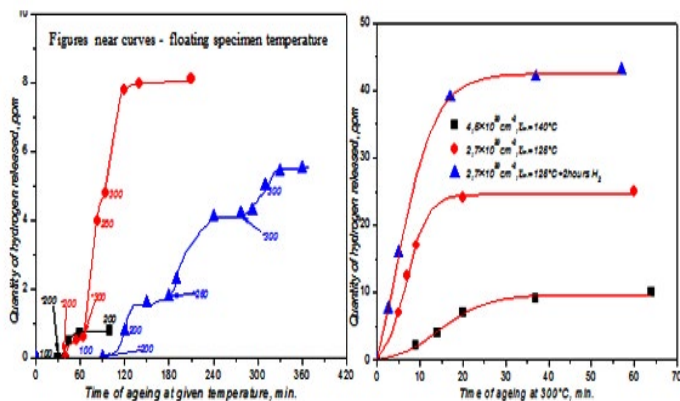


Figure 1: IPH degassing kinetics for irradiated steel ($4.5 \times 10^{20} \text{cm}^{-2}$ at 140°C)

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

As one can see from Figure 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 and high generations of hydrogen and helium in nickel. Surprisingly high hydrogen concentrations were revealed in irradiated graphite. The properties of graphite samples from the thermal column of a fabric testing reactor and from the internals of 2 completely different atomic power reactors were investigated relating to the thermal unharms of H and carbon 14. These reactor varieties are selected because of the various conditions beneath that graphite has been irradiated with neutrons throughout the in operation time of those reactors. supported the findings, choices for thermal treatment of irradiated graphite square measure mentioned so as to facilitate its final disposal.

Nuclear graphite is any grade of atomic number 6, typically artificial atomic number 6, specifically factory-made to be used as a moderator or reflector at intervals a apparatus. graphite is a crucial material for the development of each historical and fashionable nuclear reactors, because of its extreme purity and its ability to resist extraordinarily high temperatures. graphite has additionally recently been utilized in fusion reactors further, like the Wendelstein 7-X. As of experiments printed in 2019, the utilization of graphite in parts of the stellarator's wall and a graphite is land divertor have greatly improved the plasma performance at intervals the device; yielding higher management over impurity and warmth exhaust, and long high-density discharges.

Nuclear graphite parts square measure created from crystalline artificial graphite manufacture from a binder and filler coke with about 2 hundredth consistency. throughout the operational period of time, nuclear graphite moderator parts square measure subjected to quick nucleon irradiation that contributes to the modification of fabric and physical properties like thermal growth co-efficient, coefficient of elasticity and dimensional modification. These changes square measure directly driven by irradiation-induced changes to the crystal structure as mirrored through the majority microstructure. it's thus of important importance that these irradiation changes and there implication

on part property changes square measure absolutely understood. This work examines a variety of irradiated graphite samples off from country Experimental Pile Zero (BEPO) reactor; an occasional temperature, low fluence, cool Materials take a look at Reactor that operated within the kingdom. Raman spectroscopic analysis and high-resolution transmission microscopy (HRTEM) are utilized to characterise the impact of redoubled irradiation fluence on graphite microstructure and perceive temperature irradiation harm processes. HRTEM confirms the structural harm of the Bravais lattice caused by irradiation attributed to a high variety of defects generation with the buildup of dislocation interactions at nano-scale vary. Irradiation-induced crystal defects, lattice parameters and crystallization size compared to virgin nuclear graphite square measure defined exploitation selected space optical phenomenon (SAD) patterns in TEM and Raman spectroscopic analysis. The consolidated 'D' peak within the Raman spectra confirms the formation of in-plane purpose defects and mirrored as disordered regions within the lattice. The reduced intensity and broadened peaks of 'G' and 'D' within the Raman and HRTEM results make sure the looks of turbulence and disordering of the basal planes while maintaining their coherent superimposed graphite structure.

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. 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 ~ 15 min.).

References

- 1) A. Vainman, Hydrogen Embrittlement of the High Pressure Vessels. Kiev, Naukova Dumka (1990).
- 2) N. Alekseenko, Radiation Damage of Nuclear Power Plant Pressure Vessel Steels. ANS (1997).
- 3) E. Krasikov, Investigation of Hydrogen Embrittlement and Hydrogen Diffusion in Irradiated Steel, Ph.D Thesis, Moscow (1974).
- 4) C. Brinkmann, Effects of Hydrogen on the Ductile Properties of Irradiated Pressure Vessel Steels. Report IN-1359, NRTS, Idaho Falls (1970)
- 5) A.I. Jacobs, "Hydrogen Buildup In Irradiated Type-304 Stainless Steel". ASTM STP 956. F.A. Garner, and N. Igata, Eds. ASTM, Philadelphia, 239-244 (1987)
- 6) L. Greenwood, F. Garner and D. Oliver, Surprisingly Large Generation and Retention of Helium and Hydrogen in Pure Nickel. Journal of ASTM International, 1, 4. Paper ID JAI11365, 529-539 (2004)

7) A. Biriukov, E. Krasikov, "Impact of Neutron Irradiation on Graphite Dehydrogenations", VANT, ser. Thermonuclear Fusion, 1-2, 3-8. NRC "Kurchatov Institute, Moscow (1998)

8) I. Tweer, Flawed Reactor Pressure Vessels in the Belgian NPPS

Doel 3 and Tihange 2. Comments on the FANC 2015 Final Evaluation Report, 2016

9) Yu. Mostoyoi, Neutron Yesterday, Today, Tomorrow. Successes of Physics (UFN), 166, 9, 987-1022 (1996)