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Studies of reactor irradiation effect on hydrogen isotope release from vanadium alloy V4Cr4Ti

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Abstract

Vanadium alloys are most promising materials being considered for lithium blanket-breeder in future fusion reactors. The primary reason for these stems from good combination of physical–mechanical and radiation properties of vanadium alloys. In operational conditions of fusion reactors the very important issue is behavior of vanadium alloy with respect to hydrogen isotopes under neutron and gamma irradiation. This paper shows results of the experimental studies of reactor irradiation influence on parameters of hydrogen release from vanadium alloys. Experiments were carried out for various levels of reactor irradiation and showed the effect of irradiation on parameters of hydrogen release from vanadium alloy V4Cr4Ti.

1. Introduction

Current interest in the mechanisms of the interaction of hydrogen isotopes with structural materials in fission and fusion applications is mainly motivated by safety problems and tritium generation in those materials. Since new designs of future fusion installations are systematically proposed using traditional and new structural materials there is the necessity for information about the interaction of these materials with hydrogen. One such class of materials is vanadium and its alloys. This paper presents the results of experimental studies of hydrogen release from V4Cr4Ti alloy (see Table 1). Experiments were carried out using differential and integral variants of the thermodesorption method.

2. Experiment outline

Some experiments on phase transitions were carried out using a differential variant in a so-called dynamic mode under continual pump-out of the volume containing the sample saturated with hydrogen. The procedure was as follows: after high-temperature degassing, the sample was saturated with hydrogen for fixed temperature, pressure and time. Hydrogen was then pumped out and the sample was cooled to room temperature. Using the curves of hydrogen yield from the vanadium sample, the following parameters were derived: (a) the amount

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Table 1
Chemical composition of V4Cr4Ti alloy (provided by Argonne National Laboratory)

Cr, wt%	3.72–3.83	Fe, ppm	180–270
Ti, wt%	3.79–3.80	Mo, ppm	260–350
V, wt%	Balance	N, ppm	80–93
Al, ppm	180–190	Nb, ppm	<50
B, ppm	5–7	O, ppm	280–360
C, ppm	64–94	P, ppm	<30
Ca, ppm	<10	S, ppm	<10
Cl, ppm	<2	Si, ppm	720–840
Cu, ppm	<50		

of released hydrogen and (b) the characteristic temperatures where the hydrogen yield rate was a maximum.

The integral method was used as follows: after high-temperature degassing, the sample was saturated with hydrogen as mentioned above. After the sample was cooled to room temperature it was gradually heated up. At each step the exposure time was sufficient to achieve an equilibrium flow of hydrogen to the vacuum system. Near the phase-transition points the temperature step was reduced to 5 °C to provide higher accuracy in the temperature determination. Using the measured temperature dependence of equilibrium pressure of hydrogen in the vacuum system of the analyzer, the amount of released hydrogen and its solubility was evaluated.

One sample ($1.5 \times 1.5 \times 0.1$ mm) of V4Cr4Ti alloy was studied by the differential method.

For the integral method the sample of V4Cr4Ti alloy used was a $30 \times 10 \times 0.3$ mm plate. A thermocouple was welded to the plate and the plate was rolled into a ring and installed in the test cell as shown at Figs. 1 and 2.

All experiments were carried out in two stages, (a) an out-of-pile (check experiment) and (b) an in-pile experiment. Irradiation was carried out at the IVG1M reactor. Values of neutron flux in the center of reactor's active zone are given in Table 2. A schematic diagram of the irradiation experiment is presented in Fig. 3.

The range of experimental conditions during gas release measurements (by the differential method)

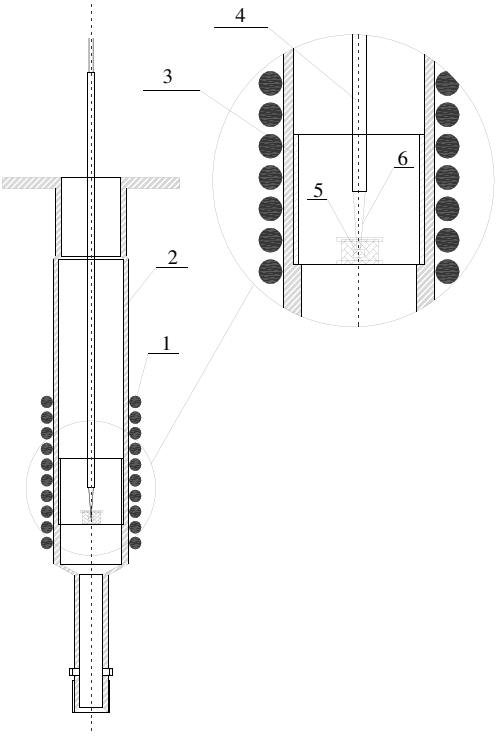

Fig. 1. Schematic view of test cell with installed sample (differential variant): 1 – heater, 2 – cell case, 3 – SS ring, 4 – ceramics tube for thermocouple, 5 – sample in a SS basket, 6 – thermocouple.

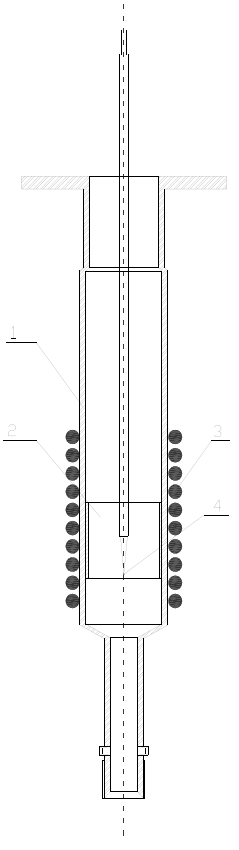

Fig. 2. Schematic view of test cell with installed sample (integral variant): 1 – cell case, 2 – sample, 3 – heater, 4 – thermocouple.

Table 2
Neutron flux in the center of the ampoule system of the IVG.1M reactor operating at 6 MW power

Energy group	0–0.67 eV	0.67 eV to 0.1 MeV	0.1–10 MeV	Total flux
Neutron flux in $n/cm^2\,s$	0.87×10^{14}	0.42×10^{14}	0.22×10^{14}	1.5×10^{14}

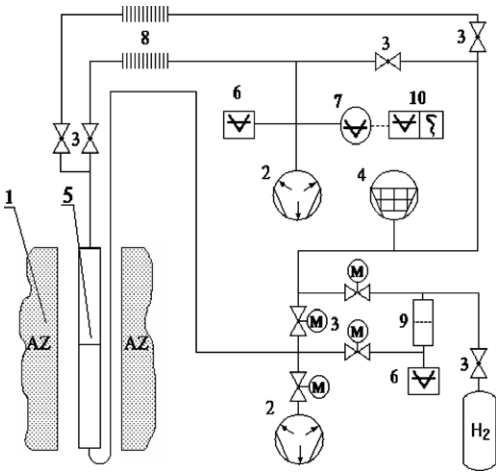

Fig. 3. Schematic of irradiation experiment: 1 – center of active zone; 2 – ion sputtering pump; 3 – vacuum valve; 4 – mechanical pump; 5 – ampoule system with sample; 6 – vacuum gage; 7 – mass-spectrometer sensor; 8 – metal-reinforced tubing; 9 – oxygen filter; 10 – data recording system.

were as follows: duration of sample saturation with H_2, −5 h; hydrogen saturation pressure, 1–10 Torr; sample heating rate, 1–20 °C/min; residual pressure in measurement system, 10–6 Torr.

The differential method experiments were carried out under continuous oil-free pump-out. The sample surfaces were cleaned with alternating exposures to oxygen and hydrogen, accordingly to a special procedure. Reactor power was 6 MW. The experimental conditions during gas release measurements (by the integral method) were as follows: duration of sample saturation with H_2, 1–5 h; equilibrium pressure in cell with the sample at 573E−1.5 Torr.

The integral method experiments were carried out under continuous oil-free pump-out. The sample surfaces were cleaned with alternating exposures to oxygen and hydrogen, accordingly to a special procedure. Reactor power levels were 1, 2 and 6 MW.

3. Out-of-pile experimental results

The main results on hydrogen yield from V4Cr4Ti sample for out-of-pile experiments [1–5] are presented in Tables 3 and 4 and Fig. 4. The temperatures of the phase transition of the hydride (deuteride) of V4Cr4Ti alloy are 454 ± 5E and 394 ± 5E, respectively. These temperatures are slightly lower than the corresponding temperatures for pure vanadium. This difference is apparently connected to the presence of chromium. It is known that Cr in V leads to a systematic increase of the solution enthalpy (the alloy became less exothermic)

Table 3
The results of experimental studies of the temperature of phase transitions in vanadium and the vanadium alloy V4Cr4Ti

	H-V (method)		D-V	H-V4Cr4Ti	D-V4Cr4Ti
Temperature of phase transition (K), (V4Cr4Ti) experiment	462 ± 5 (Differential) 472 ± 5 (integral)		402 ± 5	454 ± 5	394 ± 5
Temperature of phase transition (K), (vanadium) published data	473 [1,3]	423 [1,3]	393 [1,3]	–	–

Table 4
Solubility of hydrogen in V4Cr4Ti alloy

	S_0	E_S (eV)
Our data (experiment)	(3 ± 1)E−7	-0.370 ± 0.020
Reference data for pure vanadium [4] (440–730) K	2.2E−6	−0.331
Reference data for pure vanadium [5] (473–723) K	2.1E−6	−0.330

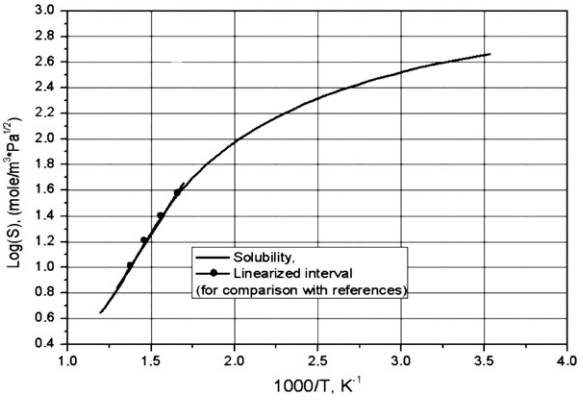

Fig. 4. Solubility of hydrogen in vanadium alloy V4Cr4Ti.

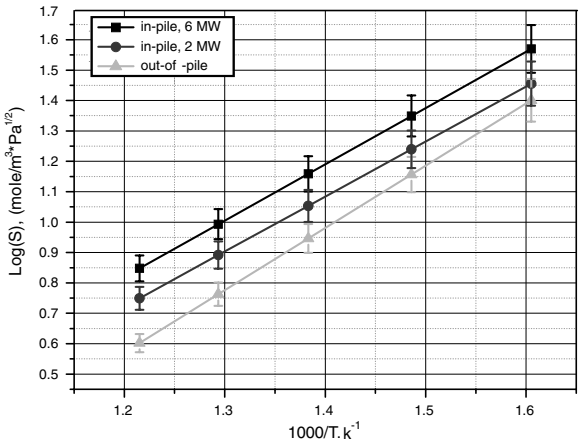

Fig. 5. Temperature dependence of hydrogen solubility in V4Cr4Ti.

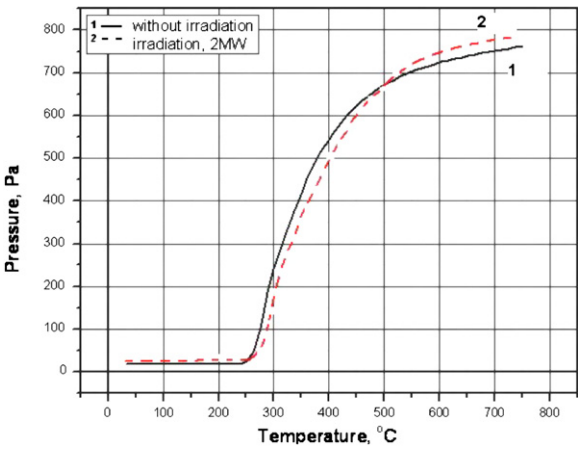

Fig. 6. Temperature dependence of hydrogen pressure over the sample of V4Cr4Ti during isochronous heating.

while the addition of Ti to V decreases the solution enthalpy.

The value of hydrogen solubility in the V4Cr4Ti alloy was also derived for the temperature range 293E–873E (see Fig. 5), which is in satisfactory agreement with published data.

Comparison of experimental results obtained with different methods showed that they are close and within the accuracy range, indicating their reliability.

Table 5
Parameters for hydrogen solubility in V4Cr4Ti (in-pile and out-of-pile experiments)

Experiment	S_0, mole/(m^3 Pa$^{1/2}$)	E_S, kJ/mole
In-pile, 6 MW	3.98E−02 ± 5%	35.4 ± 5%
In-pile, 2 MW	3.55E−02 ± 5%	34.6 ± 5%
In-pile, 1 MW	3.16E−02 ± 5%	36.9 ± 5%
Out-of-pile	1.30E−02 ± 5%	39.2 ± 5%

4. In-pile experimental results

Investigations of hydrogen yield from the vanadium alloy V4Cr4Ti under reactor irradiation with different power levels (1, 2 and 6 MW of reactor power) allowed us to obtain the temperature dependence of the hydrogen pressure over the sample during isochronous heating. Typical dependence obtained for the 2 MW power level is shown in Fig. 6. Using these dependencies, the values of S_0 and activation energy E_S were derived (see Table 5).

5. Conclusions

1. The results show the visible effect of reactor irradiation on hydrogen solubility in the V4Cr4Ti alloy. During irradiation, the solubility of hydrogen increases 1.5–2 times while the activation energy increases by 10%.
2. In these experimental studies it was established that reactor irradiation changes the temperature of the phase transition and the solubility of hydrogen in a V4Cr4Ti alloy. We suggest that this effect is linked to radiation-stimulated segregation of impurities at grain boundaries of the alloy.

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