EFFECT OF DYNAMICALLY CHARGED HELIUM ON TENSILE PROPERTIES OF V–4Cr–4Ti* H. M. Chung, B. A. Loomis, L. Nowicki, and D. L. Smith (Argonne National Laboratory)

OBJECTIVE

The objective of this work is to determine the effect of displacement damage and dynamically charged helium on tensile properties of V–4Cr–4Ti alloy irradiated to 18–31 dpa at 425–600°C in the Dynamic Helium Charging Experiment (DHCE).

SUMMARY

One property of vanadium-base alloys that is not well understood in terms of their potential use as fusion reactor structural materials is the effect of simultaneous generation of helium and neutron damage under conditions relevant to fusion reactor operation. In the present Dynamic Helium Charging Experiment (DHCE), helium was produced uniformly in the specimen at linear rates of ≈0.4 to 4.2 appm helium/dpa by the decay of tritium during irradiation to 18–31 dpa at 425–600°C in the Li-filled DHCE capsules in the Fast Flux Test Facility. This report presents results of postirradiation tests of tensile properties of V-4Cr-4Ti, an alloy identified as the most promising vanadium–base alloy for fusion reactors on the basis of its superior baseline and irradiation properties. Effects of helium on tensile strength and ductility were insignificant after irradiation and testing at >420°C. Contrary to initial expectation, room–temperature ductilities of DHCE specimens were higher than those of non–DHCE specimens (in which there was negligible helium generation), whereas strengths were lower, indicating that different types of hardening centers are produced during DHCE and non–DHCE irradiation. In strong contrast to tritium–trick experiments in which dense coalescence of helium bubbles is produced on grain boundaries in the absence of displacement damage, no intergranular fracture was observed in any tensile specimens irradiated in the DHCE.

INTRODUCTION

Vanadium-base alloys have significant advantages over other candidate alloys (such as austenitic and ferritic steels) for use as structural materials in fusion devices, e.g., the International Thermonuclear Experimental Reactor (ITER) and magnetic fusion reactors. These advantages include intrinsically lower levels of long-term activation, irradiation afterheat, neutron-induced helium- and hydrogen-transmutation rates, biological hazard potential, and thermal stress factor. Recent attention has focused on V-4Cr-4Ti for fusion reactor structural components because of its excellent combination of mechanical and physical properties before and after irradiation.[1–6] One property of vanadium–base alloys that is not well understood is the effect of helium; no tensile data have been reported on effects of simultaneous generation of helium and neutron displacement damage under fusion–relevant conditions (i.e., ≈5 appm He/dpa ratio), although helium effects on other vanadium alloys have been investigated by less–than–prototypical simulation techniques such as tritium-trick,[7–11] cyclotron-injection,[12–16] and boron-doping.[16–19] In the DHCE,[20–22] the fusion–relevant helium–to–dpa damage ratio is closely simulated by utilizing slow transmutation of controlled amounts of ^{6}Li and a tritium–doped mother alloy immersed in ^{6}Li + ^{7}Li. This report presents results of postirradiation examination of mechanical properties of V-4Cr-4Ti alloy, which has been identified as the most promising candidate alloy on the basis of its superior baseline and irradiation properties.

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MATERIALS AND PROCEDURES

The elemental composition of the V–4Cr–4Ti alloy, determined prior to irradiation, is given in Table 1. An alloy ingot, melted from low–chlorine titanium and high–purity vanadium, was extruded at 1150°C and annealed at 1050°C several times after 8–10 passes of warm (400°C) rolling between the annealings. Final forms of the product were annealed plates and sheets 3.8–, 1.0–, and 0.3–mm in thickness. SS–3 tensile specimens with a gauge length of 7.62 mm and a gauge width of 1.52 mm were machined from 1.0–mm–thick annealed (1050°C) sheets. The specimens were ≈95% recrystallized and exhibited an average grain size of ≈14 µm. The only secondary phase in the as–annealed specimen was Ti(O,N,C), which is normally observed in titanium–containing vanadium alloys with O+N+C >400 wppm. Tensile properties were measured at 23°C and at irradiation temperatures in flowing argon at a strain rate of 0.0011 s^{-1}. The thickness and gauge width of each specimen were measured individually after irradiation and before each tensile test.

Table 1. Chemical composition of V–4Cr–4Ti

	Nominal Composition	Impurity Composition (wppm)							
ANL ID	(wt.%)	O	N	C	Si	S	P	Nb	Mo
BL-47	V–4.1Cr–4.3Ti	350	220	200	870	20	<40	<100	<100

The alloy specimens were irradiated in the Fast Flux Test Facility (FFTF), a fast reactor located near Richland, Washington, at 420, 520, and 600°C to neutron fluences (E > 0.1 MeV) ranging from 3.7 x 10^{22} n/cm^2 (≈18 displacements per atom, or dpa) to 6.4 x 10^{23} n/cm^2 (≈31 dpa). Helium in the alloy specimens was produced by utilizing transmutation of controlled amounts of ^{6}Li and predetermined amounts of tritium–doped vanadium mother alloy immersed in ^{6}Li + ^{7}Li.[20–22] Table 2 summarizes the irradiation temperature, weight of the mother alloy, fraction of ^{6}Li, and tritium and lithium inventory charged in each of the seven DHCE capsules before irradiation.

Table 2. Summary of capsule–loading parameters of DHCE.

Capsule ID No.	Irradiation Temp. (°C)	Total Weight (g) Vanadium[a]	Specimen[b]	Lithium	Fraction of ^{6}Li (%)	Initial Tritium Charged[a] (Ci)	(mmol)
4D1	425	1.5468	5.86	0.765	5.0	99	1.70
4D2	425	1.5536	5.38	0.765	4.5	70	1.20
5E2	425	1.5657	5.38	0.670	1.0	26	0.45
5D1	500	1.5727	5.77	0.938	6.5	73.5	1.26
5E1	500	1.5651	5.82	0.952	1.0	57	0.98
5C1	600	1.5656	5.82	0.808	8.0	16.4	0.28
5C2	600	1.5466	5.95	0.955	8.0	18	0.31

[a] Letter from C. E. Johnson to K. Pearce, April 23, 1991; 1 mmol = 58.3 Ci.

[b] Excluding tritium–charged mother alloy.

Table 3 summarizes actual postirradiation parameters determined from tensile and TEM disk specimens of the V–4Cr–4Ti alloy, i.e., dose and helium and tritium contents measured ≈20–25 days after the postirradiation tests.

Table 3. Summary of irradiation parameters of Dynamic Helium Charging Experiment and helium and tritium contents measured in V–4Cr–4Ti specimens

Capsule ID No.	Irradiation Temp. (°C)	Total Damage (dpa)	Calculated Helium (appm) to dpa Ratio[a] at EOI[b] (Assumed k_a or k_w)[c] k_a=0.073 (k_w=0.01)	Measured Helium Content[d] (appm)	Actual Helium to dpa Ratio (appm/dpa)	Measured Tritium Content[e] (appm)
4D1	425	31	3.8	11.2–13.3	0.39	27
4D2	425	31	2.8	22.4–22.7	0.73	39
5E2	425	18	2.1	3.3–3.7	0.11	2
5D1	500	18	4.4	14.8–15.0	0.83	4.5
5E1	500	18	3.1	6.4–6.5	0.36	1.7
5C1	600	18	1.1	8.4–11.0	0.54	20
5C2	600	18	1.1	74.9–75.3	4.17	63

[a] L. R. Greenwood "Revised Calculations for the DHCE," April 30, 1993.
[b] Beginning of irradiation (BOI) May 27, 1991; end of irradiation (EOI) March 19, 1992; 203.3 effective full power days (EFPD), hot standby at ≈220°C until November 1992.
[c] Equilibrium ratio (k_a by atom, k_w by weight) of tritium in V alloy to that in the surrounding liquid lithium.
[d] Measured June 1994.
[e] Measured August 1994.

Helium and tritium contents were determined by mass spectrometry at Rockwell International Inc., Canoga Park, California. Two TEM disk or broken tensile specimens were selected from each capsule after multiple–bending (at –196°C to 50°C) or tensile tests (at room temperature) and analyzed to determine helium and tritium contents. For each specimen, four separate analyses of ^{3}He and ^{4}He were conducted. The tritium contents were determined on the basis of analysis of ^{3}He decay measured on the same specimens ≈50 days apart.

RESULTS

Yield strength, ultimate tensile strength, uniform elongation, and total elongation measured on tensile specimens irradiated at 425°C–600°C to 18–34 dpa in the DHCE are summarized in Fig. 1. For comparison, similar properties measured on irradiated non–DHCE specimens are also plotted as a function of irradiation temperature.

After irradiation to ≈30 dpa in either a DHCE or a non–DHCE, ductility of the alloy remained significantly high, i.e., >8% uniform elongation and >10% total elongation. Tensile properties measured at 425°C, 500°C, and 600°C (the same as the irradiation temperatures) were essentially the same as those measured on non–DHCE specimens, showing that the effect of helium was insignificant. Room–temperature ductilities of the DHCE specimens (irradiated at 425°C, 500°C, and 600°C) were higher than those of the similar non–DHCE specimens, whereas strengths were lower. This was an unexpected finding. Although the mechanisms leading to the higher ductility and lower strength of the DHCE specimens are not understood at this time, the consistent observations indicate that different types of hardening centers are produced during DHCE and non–DHCE irradiation.

The dependence of uniform and total elongation on irradiation and test temperature, shown in Figs. 1C and 1D, respectively, is in sharp contrast to similar results obtained on specimens in which helium atoms were produced by the tritium–trick method. In the latter type of experiments, total elongation measured at room temperature and at 700–800°C was significantly lower than that measured at

500–600°C because of the strong susceptibility to intergranular cracking associated with extensive formation of grain–boundary helium bubbles.[10] However, no intergranular fracture surface morphology was observed in the tensile specimens irradiated in the DHCE and tested at 25–600°C (including the specimen irradiated in Capsule 5C2 at 600°C at a helium generation rate of 4.2 appm He/dpa), and no ductility degradation similar to that in tritium–trick experiments was observed. This is shown in Fig. 2, where the ratio of total strain in specimens with and without helium is plotted as a function of irradiation and test temperature for tritium–trick and dynamic helium charging experiments.

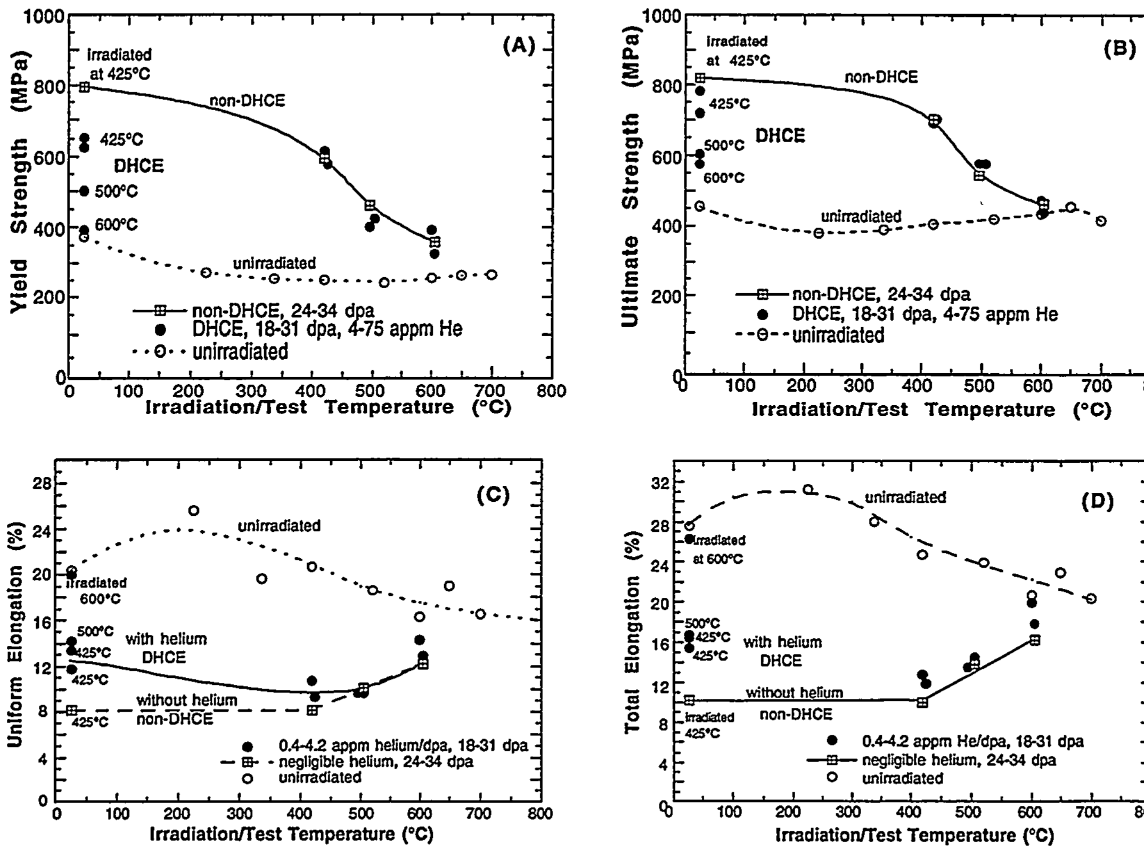

Figure 1. Yield strength (A), ultimate tensile strength (B), uniform elongation (C), and total elongation (D) of V–4Cr–4Ti after irradiation at 420–600°C to 18–34 dpa in DHCE and non–DHCE conventional irradiation (negligible helium generation).

DISCUSSION

An important finding from the DHCE was that the actual (measured) contents of helium and tritium in the V–4Cr–4Ti specimens were significantly lower than those calculated previously (see Table 3) on the basis of assumed equilibrium ratio ($k_W = 0.01$) of tritium in the alloy to that in the liquid lithium (Table 2). Except for specimens iradiated in Capsule 5C1 and 5C2 at 600°C, actual helium/dpa ratios (i.e., 0.36–0.83) were several times lower than those calculated on the basis of an equilibrium ratio of $k_W = 0.01$ (i.e., 2.1–4.4). This indicates that the level of hydrogen and tritium in the Li–cooled V–4Cr–4Ti first wall/blanket structure, and hence the effect of hydrogen and tritium on fracture toughness, will be significantly lower than previously assumed.

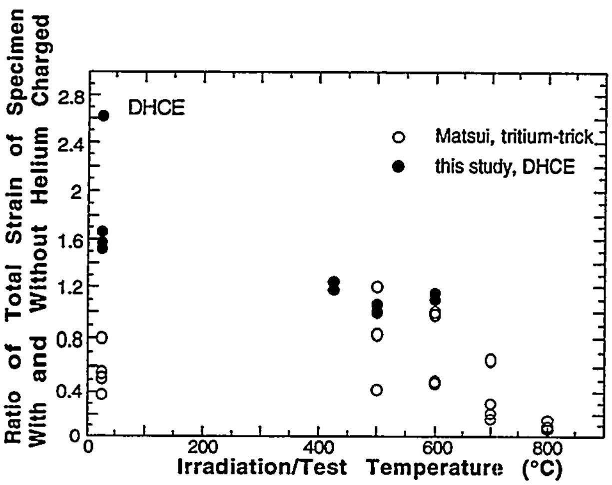

Figure 2.
Ratio of total strain in specimens with and without helium as a function of irradiation and test temperature. Results obtained from tritium–trick experiment and DHCE are shown for comparison.

As described in a separate report,[23] helium microvoids were negligible in all specimens irradiated in DHCE except for those irradiated at 425°C and retrieved from Capsule 4D1 and 4D2; only a few helium bubbles were observed at the interface between the grain matrix and some Ti(O,N,C) precipitates that are normally present in V–Ti and V–Cr–Ti alloys. Even in specimens irradiated at 600°C at the highest helium generation rate of ≈4.2 appm helium/dpa (Capsule 5C2), no microvoids could be detected in either grain matrix or grain boundaries. For specimens irradiated to 31 dpa at 425°C (retrieved from Capsules 4D1, ≈0.4 appm helium/dpa and 4D2, ≈0.73 appm helium/dpa), moderate number densities of diffuse helium bubbles were observed in localized grain matrix and near a limited fraction (≈15%) of grain boundaries.[23] The number density of helium bubbles, observed near the limited region of grain boundaries, was significantly lower than those in other alloys tested in the tritium–trick experiments, where extensive coalescence of helium bubbles occurred on all grain boundaries.[7–11] The absence of intergranular fracture morphology in any of the tensile specimens irradiated in the present DHCE seems to be consistent with the microstructural characteristics described above. However, a more comprehensive data base is needed for irradiation at <400°C to determine the effects of higher helium–dpa ratio (i.e., the fusion–relevant ratio of 4–5 appm helium/dpa) at the lower temperatures.

The uniform and total elongations determined from the room–temperature tensile tests on DHCE specimens were significantly greater than similar room–temperature elongations measured on specimens irradiated in either non–DHCE (Fig. 1) or tritium–trick experiments (Fig. 2). This is also consistent with the absence of continuous aggregation of helium bubbles on grain boundaries in the specimens irradiated in DHCE. In addition, the observation indicates that different types of hardening centers are produced in the alloy during DHCE and non–DHCE irradiation at 425–600°C.

Although the nature of the hardening centers produced during DHCE is not understood at this time, helium atoms are believed to be associated with them. In a series of studies on thermal desorption behavior of helium from unalloyed vanadium and V–5Ti irradiated with helium ions of various energy levels, van Veen et al.[24] and Buitenhuis et al.[25] concluded that helium–oxygen–vacancy and helium–nitrogen–vacancy (and probably helium–vacancy–carbon as well) complexes formed in the irradiated material. These investigators further deduced that the complexes are stable at low temperatures (<230°C) but dissociate into helium atoms and oxygen–vacancy and nitrogen–vacancy complexes at 270–310°C, leading to a prominent helium desorption peak at ≈290°C that was observed consistently in their experiments. Desorption peaks at ≈770°C and ≈1250°C, observed only after irradiation with helium ion to higher doses, were attributed to clusters of helium atoms and helium bubbles, respectively. The clusters and bubbles of helium are believed to be unstable only at the high temperatures. During the degassing treatment in the present study in which DHCE specimens were heated to 400°C at a rate of ≈0.2°C/s, desorption peaks were observed consistently at ≈290°C, although helium desorption was not positively identified by mass spectroscopy, as as done by van Veen et al. and Buitenhuis et al.

Based on these observations, it is likely that stable helium–vacancy–impurity complexes are also present in the specimens irradiated in DHCE during tensile tests at room temperature. In contrast, in specimens irradiated in non–DHCE under similar conditions, vacancies and impurities (such as oxygen, nitrogen, and carbon) are not expected to form complexes in the absence of appreciable helium atoms. Rather, the impurity atoms in solution and vacancies or vacancy clusters will be scattered more or less randomly in interstitial and vacancy sites, respectively. Dislocation motion would then be more difficult, and hence ductility would be lower in the non–DHCE than in the DHCE specimens; this is in accordance with the results shown in Fig. 1.

CONCLUSIONS

1. Tensile ductility of the V–4Cr–4Ti alloy, irradiated to 18–31 dpa at 425°C to 600°C in the Dynamic Helium Charging Experiment (DHCE) at helium generation rates of 0.4–4.2 appm helium/dpa, remained significantly high at 25–600°C, i.e., >8% uniform elongation and >10% total elongation. Tensile properties measured at >400°C were essentially the same as those measured on non–DHCE specimens (negligible helium), showing that effects of helium were insignificant. Room–temperature ductilities of the DHCE specimens (irradiated at 425, 500, and 600°C) were higher than those of the similar non–DHCE specimens, whereas strengths were lower. These observations indicate that different types of hardening centers are present at room temperature in the DHCE specimens (helium–vacancy–impurities complex, impurities being oxygen, nitrogen, and carbon) and in non–DHCE specimens (defects and defect clusters, impurities in interstitial sites).

2. The dependence of uniform and total elongation on irradiation and test temperature and fracture morphology were in sharp contrast to similar results obtained on specimens in which helium atoms were produced by the tritium–trick method. Neither partial nor predominantly intergranular fracture was observed in tensile specimens irradiated in the DHCE and tested at 23 to 600°C.

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REFERENCES

[1] B. A. Loomis, L. Nowicki, and D. L. Smith, "Effect of Neutron Irradiation on Tensile Properties of V–Cr–Ti Alloys," in Fusion Reactor Materials, Semiannual Prog. Report, DOE/ER-0313/15, Oak Ridge National Laboratory, Oak Ridge, TN (1994), pp. 219–222.

[2] B. A. Loomis, H. M. Chung, L. Nowicki, and D. L. Smith, "Effects of Neutron Irradiation and Hydrogen on Ductile-Brittle Transition Temperatures of V–Cr–Ti Alloys," ibid., pp. 253–257.

[3] H. M. Chung, B. A. Loomis, L. Nowicki, J. Gazda, and D. L. Smith, "Irradiation-Induced Density Change and Microstructural Evolution of Vanadium-Base Alloys," ibid., pp. 223–231.

[4] H. Matsui, M. Tanno, J. Gazda, and H. M. Chung, "Microstructural Stability of V-4Cr-4Ti During Neutron Irradiation," ibid., pp. 240–246.

[5] H. M. Chung, J. Gazda, L. Nowicki, J. E. Sanecki, and D. L. Smith, "Effects of Fabrication Variables on Impact Properties and Microstructure of V–Cr–Ti Alloys," ibid., pp. 207–218.

[6] H. M. Chung, B. A. Loomis, and D. L. Smith, "Thermal Creep Behavior of V–5Cr–5Ti and V–10Cr–5Ti Alloys," in Fusion Reactor Materials, Semiannual Prog. Report, DOE/ER-0313/14, Oak Ridge National Laboratory, Oak Ridge, TN (1993), pp. 309-317.

[7] D. N. Braski, in Influence of Radiation on Material Properties, ASTM-STP 956 (1986), pp. 271-290.

[8] D. N. Braski, J. Nucl. Mater. 141-143 (1986) 1125.

[9] D. N. Braski, in Reduced Activation Materials for Fusion Reactors, ASTM-STP 1047 (1988), pp. 161-178.

[10] H. Matsui, M. Tanaka, M. Yamamoto, and M. Tada, J. Nucl. Mater. 191-194 (1992) 919.

[11] M. Satou, K. Abe, and H. Matsui, J. Nucl. Mater. 191-194 (1992) 938.

[12] W. van Witzenburg, A. Mastenbroek, and J. D. Ellen, J. Nucl. Mater. 103-104 (1981) 1187.

[13] M. P. Tanaka, E. E. Bloom, and J. A. Horak, J. Nucl. Mater. 114 (1981) 895.

[14] M. L. Grossbeck and J. A. Horak, in Influence of Radiation on Material Properties, ASTM-STP 956 (1986), p. 291.

[15] J. M. Vitek, D. N. Braski, and J. A. Horak, J. Nucl. Mater. 141-143 (1986) 982.

[16] W. van Witzenburg and E. de Vries, in Effects of Radiation on Materials, ASTM-STP 1125 (1990).

[17] L. L. Horton and K. Farrell, J. Nucl. Mater. 122-123 (1984) 687.

[18] H. Kawanishi and S. Ishino, in Reduced Activation Materials for Fusion Reactors, ASTM-STP 1047 (1988), pp. 179-189.

[19] H. Kawanishi, Y. Arai, and S. Ishino, J. Nucl. Mater. 191-194 (1992) 933.

[20] D. L. Smith, H. Matsui, L. R. Greenwood, and B. A. Loomis, J. Nucl. Mater. 155-157 (1988) 1359.

[21] D. L. Smith, B. A. Loomis, H. Matsui, M. L. Hamilton, K. L. Pearce, J. P. Kopasz, C. E. Johnson, R. G. Clemmer, and L. R. Greenwood, in Fusion Reactor Materials, Semiannual. Prog. Rep. DOE/ER-0313/10, Oak Ridge National Laboratory, Oak Ridge, TN (1991), p. 159.

[22] H. Tsai, H. M. Chung, B. A. Loomis, and D. L. Smith, "Status of Dynamic Helium Charging Experiment (DHCE)," in Fusion Reactor Materials, Semiannual Prog. Report, DOE/ER-0313/15, Oak Ridge National Laboratory, Oak Ridge, TN (1994), pp. 247–252.

[23] H. M. Chung, B. A. Loomis, L. Nowicki, J. Gazda, and D. L. Smith, "Void Structure and Density Change of Vanadium–Base Alloys Irradiated in the Dynamic Helium Charging Experiment," in this report.

[24] A. van Veen, H. Eleveld, and M. Clement, "Helium Impurity Interactions in Niobium and Vanadium," in Proc. 6th Intl. Conf. on Fusion Reactor Materials, September 27–October 1, 1993, Stresa, Italy, in press.

[25] T. Buitenhuis, A. Fedorov, and A. van Veen, "Thermal Desorption Investigation of V–5Ti Alloy," IRI–131–94–005, Delft University of Technology, The Netherlands, 1994.