ELSEVIER

Journal of Nuclear Materials 271&272 (1999) 349–355

journal of nuclear materials

Neutronics aspects of a DHCE experiment

Itacil C. Gomes [*], Hanchung Tsai, Dale L. Smith

Argonne National Laboratory, 9700 South Cass Ave. TD/207, Argonne, IL 60439-4841, USA

Abstract

The DHCE (dynamic helium charging experiment) irradiation experiment was conceived to simulate fusion-relevant helium production in a fission reactor irradiation. The main objective is to maintain the helium-to-dpa ratio at, roughly, the same level as expected in a fusion environment. The problem in fission reactor irradiation is that helium production is very low, because the fission neutrons, for basically all structural materials relevant for fusion applications, do not have enough energy to trigger the helium producing reactions. A DHCE experiment involves the decay of tritium to He-3 to produce the required helium during irradiation. This paper describes an analysis of the most important aspects of a DHCE experiment and compares different types of fission reactors and their suitability for performing such an experiment. It is concluded that DHCE experiments are feasible in a certain class of mixed-spectrum fission reactors, but a careful and detailed evaluation, for each facility and condition, must be performed to ensure the success of the experiment. © 1999 Published by Elsevier Science B.V. All rights reserved.

1. Introduction

The DHCE experiment is well suited for vanadium alloys due to the high solubility of tritium in these alloys. High tritium concentrations can be obtained in these alloys without demanding very high pressures to drive the tritium into the samples. This characteristic considerably simplifies the engineering and fabrication of the irradiation capsules. The required tritium is introduced into the capsule in a so-called mother alloy. This alloy sample has a very high tritium concentration while the test samples do not contain any tritium, simplifying their handling and loading into the capsule. The tritium is confined in the mother alloy until the temperature of the capsule is raised upon reactor start-up at which time the tritium migrates to the test alloys. As a result helium production in the test samples is initiated at the time that the irradiation capsule is brought up to the desired irradiation temperature.

The first DHCE experiment was performed in the MOTA irradiation vehicle of the FFTF (fast flux test facility), a fast breeder reactor. This proof-of-principle experiment demonstrated that helium concentrations approaching the desired He/dpa ratio could be obtained. Also, the microstructure of the irradiated vanadium alloys have been shown to be significantly different from similar alloys that have undergone irradiation without enhancing the helium production. The goal for the DHCE MOTA experiment was basically to be ‘a proof-of-principle experiment’, and a large range of parameters was adopted. For example, a range for the tritium distribution coefficient between the lithium thermal bonding (used to homogenize the temperature of the sub-capsule) and the vanadium alloys was assumed to account for variations in the composition of the alloys and irradiation temperatures, different capsules were irradiated at different temperatures and several alloy compositions were irradiated, sometimes, in the same capsule. After irradiation, the samples were analyzed for helium content. All specimens indicated helium concentrations much higher than in conventional irradiation, however, most specimens contained less than the desired 4–5 He/dpa.

Since FFTF and EBR-II fast-breeder reactors were shut down, no other fast-flux irradiation facility is available in US. The feasibility of conducting DHCE experiment in one of the available mixed spectrum reactors in US, mainly ATR (advanced test reactor, LMIT (Lockheed Martin Idaho Technologies), Idaho Falls) and HFIR (high flux irradiation reactor, ORNL, Oak Ridge), is being evaluated.

---

[*] Corresponding author. Tel.: +1-630 252 4665; fax: +1-630 252 5287; e-mail: icgomes@anl.gov

0022-3115/99/$ – see front matter © 1999 Published by Elsevier Science B.V. All rights reserved.
PII: S0022-3115(98)00719-3

This paper presents results of some calculations performed to assess the suitability of the mixed-spectrum reactors for a DHCE experiment. It is shown that it is possible to have DHCE experiments in a mixed-spectrum reactor provided that enough shielding for thermal neutrons is incorporated into the design. Also, a comparison of the performance of different reactors is included with this analysis.

2. The basic quantities to be controlled in a DHCE experiment

The analysis of a DHCE experiment is complex in nature due to the large number of variables involved. For example, the net increase in the He-3 concentration in the sample during irradiation is a function of the tritium concentration in the sample, and of He-3 burn-up. The tritium concentration in the sample is a function of the initial tritium load, any tritium generation during irradiation, the tritium leakage through the capsule, and of the distribution coefficient between samples, thermal bonding material, and capsule walls. The He-3 burn-up depends on the He-3 concentration, on the neutron flux spectrum, and on the neutron flux intensity. Other concerns regarding the tritium solubility in the thermal bonding material and tritium permeation through the capsule wall add to the number of variables to be controlled during the experiment.

From a neutronics point-of-view, the characterization of the thermal and epi-thermal neutron flux is very important because significant ^{3}He burn-up can occur due to very high thermal and epi-thermal cross sections of the ^{3}He(n,p) ^{3}H reaction. Other factors that have to be considered are the tritium generation in the lithium bonding and the tritium leakage.

The relationship between the above parameters can be expressed as follows:

(a) Helium generation rate in the vanadium alloy is

$$S_{\mathrm{He}}(t) = \lambda_{\mathrm{T}} N_{\mathrm{T}}(t), \tag{1}$$

where $S_{\mathrm{He}}(t)$ is the helium generation rate per cubic centimeter at the time t from the decay of tritium, λ_{T} is tritium decay constant equal to $1.78 \times 10^{-9}\ S^{-1}$, and $N_{\mathrm{T}}(t)$ is the number of tritium atoms at the time t per cubic centimeter.

(b) Helium burn-up in the vanadium alloy is

$$D_{\mathrm{He}}(t) = N_{\mathrm{He}}(t)\sigma_{\mathrm{He}(n,x)}\phi(t), \tag{2}$$

where $D_{\mathrm{He}}(t)$ is the rate of disappearance of helium at the time t per cubic centimeter due to neutron induced transmutation reaction, $N_{\mathrm{He}}(t)$ is the number of ^{3}He atoms at the time t per centimeter-barn, $\sigma_{\mathrm{He}(n,x)}$ is the spectrum averaged ^{3}He transmutation cross section (basically the (*n,p*) reaction) in barns, and $\phi(t)$ is the energy integrated neutron flux in the sample region at the time t in the vanadium alloy.

(c) Tritium production in the lithium thermal bonding is

$$P_{\mathrm{T}}(t) = N_{\mathrm{Li}}(t)\sigma_{\mathrm{Li}(n,\alpha)\mathrm{T}}\phi(t), \tag{3}$$

where $P_{\mathrm{T}}(t)$ is the tritium production rate at the time t per cubic centimeter of lithium, $N_{\mathrm{Li}}(t)$ is the number of lithium atoms per centimeter-barn at the time t in the lithium region, $\sigma_{\mathrm{Li}(n,\alpha)\mathrm{T}}$ is the tritium-production cross section for lithium, in barns, averaged over the energy spectrum, and $\phi(t)$ is the energy integrated neutron flux in the lithium region at time t.

(d) Tritium atom distribution coefficient between thermal bonding material and vanadium alloy is

$$\mathrm{ADC} = (N_{\mathrm{T}}/N_{\mathrm{Li}})_{\mathrm{Li}}/(N_{\mathrm{T}}/N_{\mathrm{V}})_{\mathrm{V}}, \tag{4}$$

where ADC is the atom distribution coefficient between lithium and vanadium alloy, $(N_{\mathrm{T}}/N_{\mathrm{Li}})_{\mathrm{Li}}$ is the tritium atom fraction in lithium thermal bonding with N_{T} being the number of tritium atoms per cubic centimeter and N_{Li} the number of lithium atoms per cubic centimeter, $(N_{\mathrm{T}}/N_{\mathrm{V}})_{\mathrm{V}}$ is the tritium atom fraction in the vanadium alloy.

(e) Tritium leakage through the capsule walls is

$$\mathrm{d}N_{\mathrm{T}}/\mathrm{d}t = -LN_{\mathrm{T}}(t), \tag{5}$$

where $N_{\mathrm{T}}(t)$ is the number of tritium atoms at the time t and L the leakage coefficient. The leakage coefficient can be represented by

$$L = D^{\mathrm{c}}K_{a}^{\mathrm{c}}(\Omega_{\mathrm{L}}/\Omega_{\mathrm{c}})(S/dV_{\mathrm{L}}), \tag{6}$$

where D^{c} is the diffusion coefficient of tritium in the capsule material, $K_{\mathrm{a}}^{\mathrm{c}}$ the distribution coefficient of tritium between the capsule and lithium (in atomic percent), Ω_{L} the atomic volume of lithium, Ω_{c} the atomic volume of the capsule material, S the surface area of the capsule, d the capsule wall thickness, and V_{L} the volume of lithium in the capsule. This formulation does not take into account the additional surface-controlled mechanisms. The diffusion coefficient of tritium, for TZM (a molybdenum alloy) capsules, is strongly dependent on the temperature, which makes the leakage coefficient also strongly dependent on the temperature. It is well known that a thin oxide (or other compound) layer can have a notable inhibiting effect in the tritium leakage. Also, desorption of tritium at low pressures was not considered in this formulation. This means that, the leakage coefficient is assumed to be diffusion controlled, without taking credit for the use of surface barrier mechanisms for the tritium or desorption controlled kinetics.

The final relationships obtained from factoring together the above equations are the following:

Helium concentration variation with the time in the vanadium alloy sample

$$\mathrm{d}N_{\text{He-3}}/\mathrm{d}t = S_{\text{He}}(t) - D_{\text{He}}(t); \quad (7)$$

this equation is dependent on the tritium content in the samples. The variation of the tritium concentration inside the vanadium alloy can be equated by

$$\mathrm{d}N_{\mathrm{T}}/\mathrm{d}t = \{N_{\mathrm{Li}}\sigma_{\mathrm{Li}(n,\alpha)\mathrm{T}}\phi - \lambda N_{\mathrm{T}} - LN_{\mathrm{T}}\}_{\mathrm{Li}} / \{\mathrm{ADC} * (N_{\mathrm{Li}}/N_{\mathrm{V}})\}. \quad (8)$$

The coupled equations (7) and (8) can be either directly integrated or solved by any of the available techniques. In this work the REAC [1] code was slightly modified to solve those equations. The 'leakage coefficient' as a function of the temperature was introduced in the decay library of the code in such a way that the leakage is seen as a tritium decay mode which produces the disappearance of the atom. Further, the tritium production from lithium was simulated through the use of an 'equivalent' concentration of lithium, in the vanadium alloy, based on the distribution coefficient of tritium between lithium and vanadium alloy.

3. Physical constraints to the experiment

The most important physical constraints for the integrity of the irradiation capsules is the pressure build-up inside the capsule, during irradiation due to gas production. The gas pressure build-up is largely due to the helium production when lithium is used as the thermal bonding material. Helium is a by-product of the tritium producing reaction Li-6(n,α)^{3}H. This tritium producing reaction has been proposed as a way to increase tritium inventory during irradiation to make-up for the He-3 burn-up and tritium leakage. A higher tritium concentration inside the capsule would allow a higher He-3 burn-up due to the higher He-3 production. Leaving aside the merits of allowing He-3 to be burned, this approach has as the main constraints two points: The first is that a higher tritium concentration creates a more noticeable saw-shaped profile of the Helium to dpa ratio, due to rapid increase in the helium concentration during the shut-down periods followed by the fast burn-up of the excess of He-3 when the reactor resumes the operation (as will be seen in the results presented in Section 4). The second limiting constraint is the pressure build-up due to associated helium atoms produced by the tritium generating reaction. To define the maximum enrichment that can be used at a given irradiation temperature one can calculate the maximum number of atoms of helium into the plenum (region inside the capsule, fill with inert gas, to accommodate the gases produced during irradiation) of the capsule.

Assuming that the maximum acceptable pressure inside the capsule is 20 atm, and given the volume of the capsule's plenum, one can use the ideal gas law to calculate the number of moles that can be in the plenum for the irradiation temperature. Then, using

$$PV = nRT, \quad (9)$$

where P is the pressure given in atm, V the volume is given in liters, n the numbers of moles, R the universal gas constant equal to 0.0821 atm l/(mol K), and T the temperature given in Kelvin. Now, assuming that the volume of samples plus lithium bonding inside the capsule is roughly the same as the volume of the gas plenum, the maximum number of moles of helium in 1 cm^3 of plenum, as a function of the temperature, is as follows:

$$n = 0.244/T = N/6.023 \times 10^{23}, \quad (10)$$

where N is the number of atoms of ^{4}He per cubic centimeter.

The number of ^{4}He atoms produced is a function of the neutron flux, flux averaged ^{6}Li(n,α) cross section, and number of ^{6}Li atoms in the thermal bonding material. The number of helium atoms produced per cm^3 of lithium is as follows:

$$N_{\text{He-4}} = N_{\text{Li-6}} \times \sigma_{\text{Li-6}} \times \phi \times t, \quad (11)$$

where t is total irradiation time, ϕ is the total neutron flux at the position considered, $\sigma_{\text{Li-6}}$ is the spectrum averaged ^{6}Li(n,α) cross section, $N_{\text{Li-6}}$ is the number of atoms of ^{6}Li per cm^3, and $\mathrm{N}_{\text{He-4}}$ the number of atoms of ^{4}He produced per cubic centimeter of plenum. Considering that the plenum volume is twice the lithium volume in the capsule, the total number of ^{4}He atoms produced per cubic centimeter of lithium must be divided by 2 to enter in Eq. (10).

Table 1 presents some values for the flux averaged ^{6}Li(n,α) cross section. The ^{6}Li content considered in these calculation was the natural abundance in Li (7.5%). The reactor positions considered were the ATR-A10 (irradiation position A-10 of the advanced test reactor) with a neutron thermal filter 3 mm thick made of Eu_2O_3 powder 50% dense, the ATR-ITV (irradiation

Table 1
Flux averaged ^{6}Li(n,α) cross section for different reactor and thermal filter configurations

ATR		HFIR – RB4 position		MOTA
A10 – 3 mm Eu_2O_3 (50% dense)	ITV – 3 mm Al with 4.0 wt% ^{10}B	4 mm Eu_2O_3 (50% dense)	2 mm Eu_2O_3 (50% dense)	no filter (mid-plane)
10.2 barns	13.6 barns	56.0 barns	110.0 barns	1.0 barns

Table 2
Time and associated number of dpa necessary to reach 20 atm. inside the irradiation capsule (plenum volume equal to samples plus lithium volume) using natural lithium as thermal bonding material

Temp. (°C)	ATR				HFIR-RB4 position				MOTA	
	A10 – 3 mm Eu_2O_3 (50% dense)		ITV – 3 mm Al with 4.3 wt% ^{10}B		4 mm Eu_2O_3 (50% dense)		2 mm Eu_2O_3 (50% dense)		no filter (mid-plane)	
	T(fpd [1])	dpa	T(fpd [1])	dpa	T(fpd [1])	dpa	T(fpd [1])	dpa	T(fpd [1])	dpa
400°C	185.0	7.6	124.0	5.1	32.5	0.8	16.5	0.41	160.0	24.0
500°C	161.0	6.6	108.0	4.4	28.3	0.7	14.4	0.36	140.0	21.0
600°C	143.0	5.9	95.6	3.9	25.1	0.6	12.7	0.31	124.0	19.0

[1] fpd = full power days.

test vehicle of the advanced test reactor) with outside borated aluminum (4.3% weight of ^{10}B) 3 mm-thick-thermal filter, the HFIR RB4 (removable beryllium position) with outside Eu_2O_3 thermal filter (50% dense) with the thickness of 4 and 2 mm, and the MOTA mid-plane (−0.30 cm) position of FFTF with no filter.

The time and the number of dpa to reach 20 atm pressure inside the capsule is given in Table 2, for the same configurations presented in Table 1. The results presented in Table 2 indicate that for longer irradiation time than calculated, the plenum volume has to be increased by the ratio of the target irradiation time divided by the values presented in the table. This leads us to conclude that the use of lithium as thermal bonding material in HFIR is very difficult due to the high thermal flux and associated ^4He production inside the capsule, unless depleted lithium (1% ^6Li) is used. It is also clear that a higher allowable pressure, or a larger gas plenum, or even a more detailed analysis of the distribution of the produced ^4He between gas phase and that dissolved in lithium phase can make the use of natural or slightly enriched lithium possible in ATR.

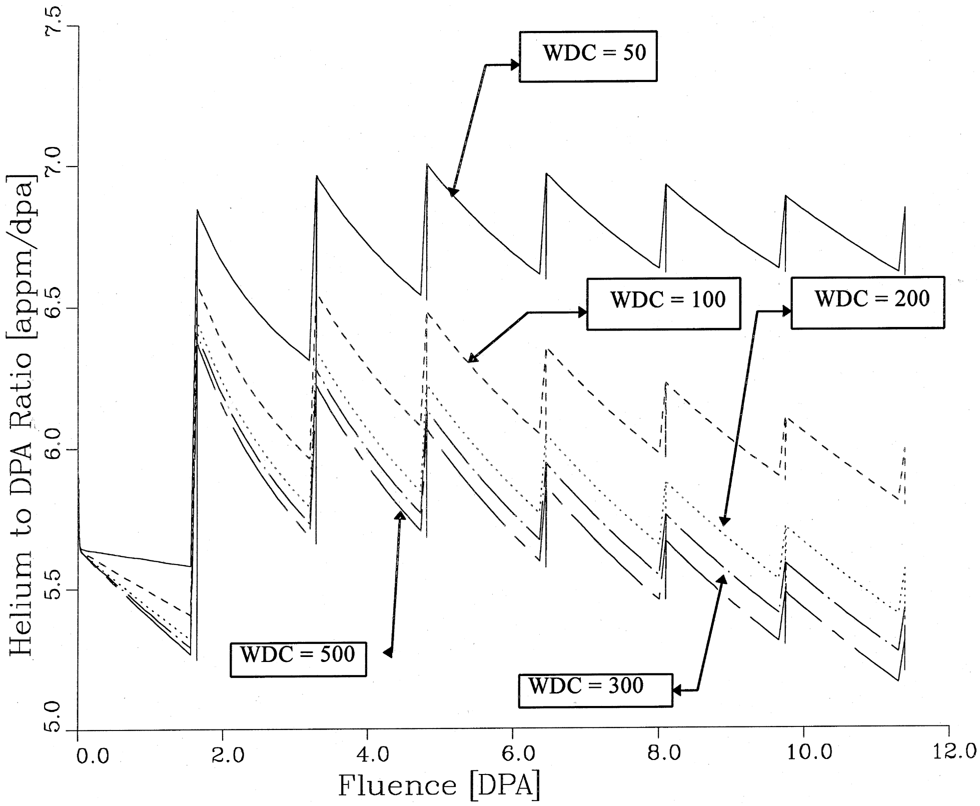

Fig. 1. Effect of changing the distribution coefficient on the helium to dpa ratio profile for the ATR A10 position using a 3 mm Eu_2O_3 thermal neutron filter.

4. Results for MOTA, ATR, and HFIR

During the course of analyzing DHCE experiments several fission environments were considered. It is worth mentioning that one of the most challenging aspects of designing an experiment such as DHCE is to determine a reliable neutron flux description, mainly for the thermal and epithermal energy regions. The use of thermal neutron absorbers and their efficiency in removing thermal neutrons and shaping the flux to be compatible with the fusion spectrum is deeply affected by the correct characterization of the neutron flux at the irradiation position.

In this section three different reactors are considered, the FFTF (fast flux test facility), the ATR (advanced test reactor), and the HFIR (high flux irradiation reactor). The FFTF is not currently in operation, and it is shown here for the sake of comparing fast flux reactor with mixed spectrum reactors, such as ATR and HFIR. The results presented were obtained from neutron flux spectra derived from published data [1,3], by the introduction of thermal filter material (when applicable) and neutron transport in a simplified MCNP [2] geometric model of the reactor.

Fig. 1 presents a comparison of the helium to dpa ratio for different distribution coefficients, keeping constant the temperature (400°C), the tritium concentration in the vanadium alloy (1500 appm) and the lithium thermal bonding enrichment (7.5% ^{6}Li). It can be noted that the lower the distribution coefficient the larger is the helium to dpa ratio. This is due to the larger number of tritium atoms generated in the lithium that can diffuse to the vanadium samples. The distribution coefficient would have a much greater impact if one had maintained the initial tritium charge of the capsule constant but this would mask the only physical effect (migration of tritium atoms) due to different distribution coefficients among different materials.

Fig. 2 shows comparisons of the development of the helium to dpa ratio during the irradiation for the three reactors mentioned earlier. The temperature of irradiation in Fig. 2 is 400°C. A figure for results of 500°C irradiation is not shown because of its similarity with the 400°C. In the Fig. 2 the distribution coefficient was set

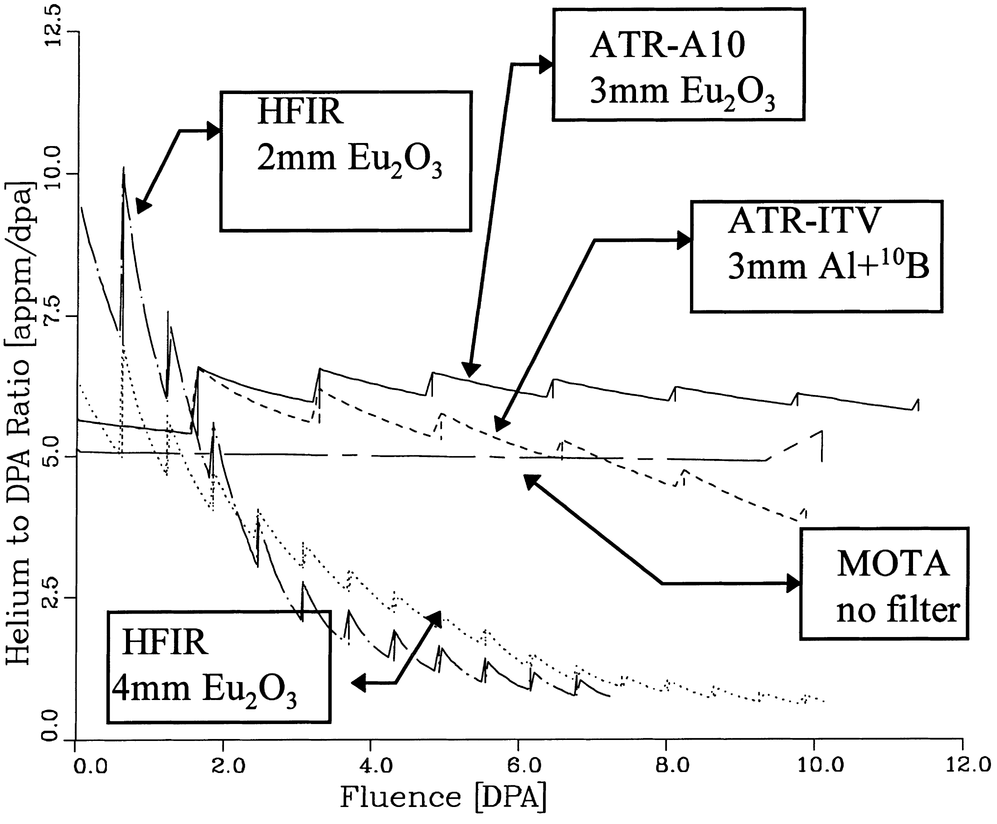

Fig. 2. Calculated helium to dpa profile during irradiation for a temperature of 400°C, natural ^{6}Li abundance (except for HFIR which has 1% ^{6}LI), distribution coefficient of 100, and 1500 appm (ATR and HFIR) and 5000 appm (MOTA) tritium concentration in the vanadium alloy.

to 100, the lithium enrichment to the natural abundance of ^{6}Li (except for HFIR, for which the ^{6}Li enrichment was set to 1%, for the reasons explained in Section 3), and the initial tritium charge equivalent to 1500 appm (for ATR and HFIR) and 5000 appm (for MOTA) of tritium in the vanadium alloy.

It is clear that in ATR it is possible to maintain a relatively flat helium to dpa profile during irradiation for the temperatures presented (also, for 500°C), even when using borate aluminum as a filter. HFIR, due to the high thermal flux, can not achieve a flat profile even using a 4 mm Eu_2O_3 filter. MOTA as presented in these pictures does not have the same saw-shaped profile as the other reactors because 10 dpa is about the length of one cycle of the reactor, so the shut down time, which is responsible for the increase in helium concentration, is not shown in the MOTA curve. It is worthwhile mentioning that the tritium leakage term, calculated based on a diffusion driven mechanism, is not large at these temperatures. This makes them very similar to each other. The helium to dpa ratio at the end of the 10 dpa irradiation campaign is calculated to be for 400°C 6.0 for ATR-A10 with 3 mm of Eu_2O_3 filter, 4.1 for ATR-ITV with 3 mm of borate (4.3 wt% 10B) aluminum filter, 0.6 for HFIR-RB with 4 mm of Eu_2O_3 filter, 0.7 (at 7.2 dpa) for HFIR-RB with 2 mm of Eu_2O_3 filter, and 4.9 for MOTA mid-plane position. At 500°C the results after 10 dpa are as follows: 6.0 for ATR-A10, 4 for ATR-ITV, 0.5 for HFIR-4 mm, 0.6 for HFIR-2 mm (at 7.2 dpa's), and 4.8 for MOTA.

Fig. 3 presents the helium to dpa profile for the same positions and irradiation parameters as Figs. 2 and 3 except for a irradiation temperature which is set to 600°C in this case. This figure is included to emphasize the effect of a diffusion driven leakage term for tritium on the helium production. The only barrier to the permeation of tritium out of the capsule is the use of a TZM capsule wall, but as can be seen, the leakage has an important impact on the helium production due to reduction of the tritium inventory inside the capsule.

As it was mentioned earlier, other mechanisms, such as desorption and an oxide layer can significantly reduce the tritium permeation when compared with diffusion controlled leakage. However, the final helium to dpa ratio after 10 dpa of irradiation are not completely un-

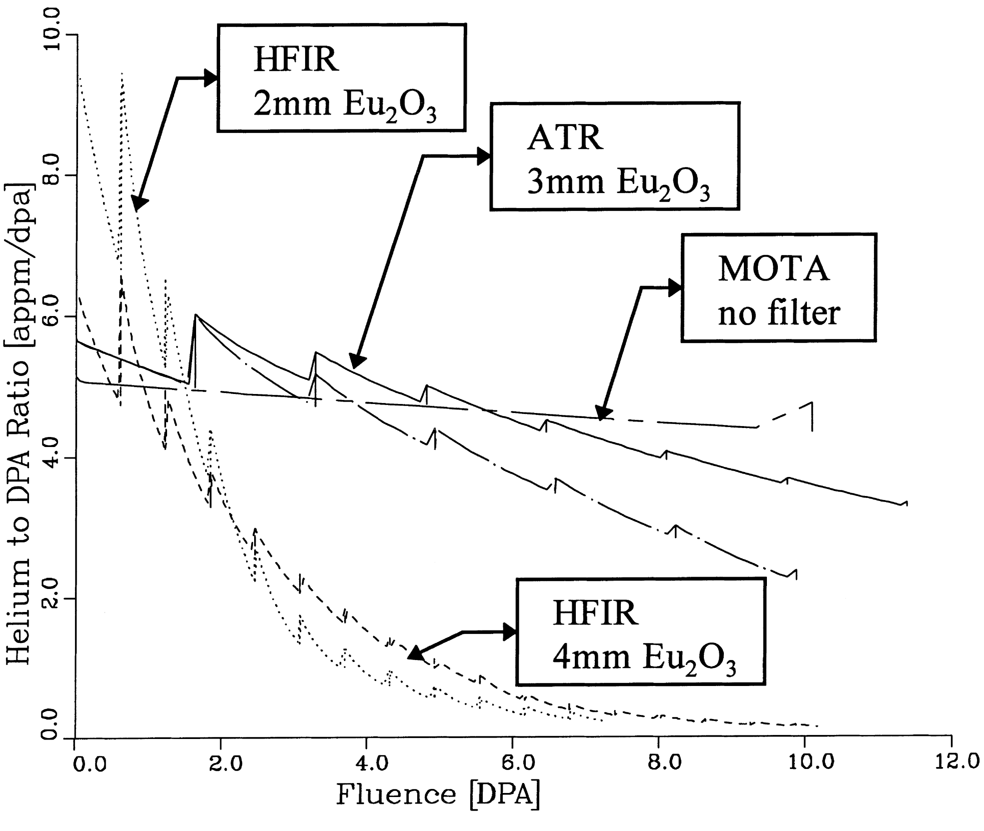

Fig. 3. Calculated helium to dpa profile during irradiation for a temperature of 600°C, natural ^{6}Li abundance (except for HFIR which has 1% ^{6}Li), distribution coefficient of 100, and 1500 appm (ATR and HFIR) and 5000 appm (MOTA) tritium concentration in the vanadium alloy.

acceptable for ATR with 3 mm Eu_2O_3 (he/dpa = 3.3), MOTA (he/dpa = 4.7), and ATR-ITV (he/dpa = 2.4). On the other hand, the values for HFIR are extremely low, being 0.14 for 4 mm of Eu_2O_3 and 0.23 (at 7.2 dpa) for 2 mm Eu_2O_3 filter.

5. Conclusions

The calculations presented in this paper indicate that the DHCE experiment is possible in mixed-spectrum reactors, and that a significant amount of helium will be produced during irradiation. Furthermore, the results indicate that even for high temperatures, using the most pessimistic assumptions for tritium leakage, it is possible to have fusion relevant helium concentrations in vanadium alloys after an irradiation campaign of 10 dpa. The amount of helium generated should be enough to demonstrate the influence of helium production in vanadium alloys under intense neutron bombardment.

The calculations also indicate that mixed-spectrum reactors with very high thermal component in the neutron energy spectrum are expected to have a poor performance in this kind of experiment. The case analyzed for the HFIR reactor shows that an external filter in the RB position does not produce good results. As an alternative to reduce the thermal neutron flux inside the neutron thermal filter, it is conceivable to borate the filling material of that position. However the helium production in the filler due to the neutron absorption in the boron may cause serious problems.

In the ATR-ITV, despite the relatively good performance, it was recommended to reduce the thermal neutrons to further borate the thermocouple slaves. This would produce a similar performance to the Eu_2O_3 external filter.

Finally, the calculations indicate that with a relatively small margin of uncertainty, it is possible to acquire valuable data from the helium effect on vanadium alloys using the available mixed spectrum reactors.

References

[1] F.M. Mann, REAC-3 – A Computer Code System for Activation and Transmutation, Hanford Engineering Development Laboratory, Report HEDL-TME-81-37, 1993.

[2] J.F. Briesmeister (Ed.), MCNP – A General Monte Carlo N-Particle Transport Code, Version 4A, Report LA-12626, Los Alamos National Laboratory, Los Alamos, New Mexico, 1993.

[3] B.G. Schnitzler, LMIT (Lockheed Martin Idaho Technologies), Idaho Falls, private communication.