

NEUTRONICS ANALYSIS OF THE DHCE EXPERIMENT IN ATR-ITV*, I.C.Gomes, D.L. Smith, and H.Tsai (Argonne National Laboratory)

OBJECTIVE

The object of this analysis was to assess the suitability of the irradiation test vehicle (ITV), under design, for Dynamic Helium Charging Experiment (DHCE) with vanadium alloys. To achieve this goal a preliminary analysis of the irradiation conditions inside the vehicle, reaction rates, tritium leakage, among others have to be calculated. The objective is to calculate the helium to dpa ratio and analyze its range of variation and stability during a given irradiation period.

SUMMARY

The preliminary analysis of the DHCE experiment in the ITV of ATR was performed and it was concluded that such a vehicle is suitable for this kind of experiment. It is recommended to place an extra filter material in the thermocouple sleeve (such as B-10), to improve the helium to dpa ratio profile during irradiation. Also, it was concluded that a preliminary estimation of period of time for replacement of the external filter would be around 5 dpa's.

INTRODUCTION

The DHCE irradiation experiment was conceived to simulate Helium production due to neutron interaction in a fusion environment through the tritium decay to Helium-3. The experiment consists in charging a mother alloy with tritium at a concentration that after diffusion into vanadium alloy specimens produces a net build-up of helium in the samples compatible with the helium to dpa ratio of a fusion environment. The use of lithium as thermal bonding in the experiment allows an extra production of tritium during irradiation.

The analysis of such an experiment is complex in nature due to the large number of intervening variables. For example, the net build-up of helium-3 is a function of neutron flux intensity, neutron flux spectrum, tritium leakage rate from the capsule, initial tritium charge, the distribution coefficient of tritium atoms between samples, thermal bonding material, and capsule walls. Also, the tritium concentration inside any of the component materials (mainly in the lithium bonding) has to be below saturation otherwise chemical compounds are created and the distribution of the atoms among the material regions would not follow the theoretical distribution coefficient.

From a neutronics point-of-view the characterization of the thermal and epithermal neutron flux is very important in this analysis because of the very high cross section of the ${}^3\text{He}(n,p) {}^3\text{H}$ reaction in those energy regions. Despite the lithium thermal bonding producing tritium through the ${}^6\text{Li}(n, \alpha) {}^3\text{H}$ reaction, the cross section for the ${}^3\text{He}(n,p) {}^3\text{H}$ is between 5 and 10 times larger for almost all neutron energies. This indicates that the reaction rate for both reactions should be reduced to a minimum to allow a reasonable growth of the ${}^3\text{He}$ concentration in the sample.

Another factor that is very important in the balance for the net ${}^3\text{He}$ production is the tritium leakage from the capsule. A large leakage rate means that a large fraction of the initial and produced ${}^3\text{H}$ is going to leak out of the capsule reducing the ${}^3\text{H}$ inventory and the ${}^3\text{He}$ generation rate.

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This paper presents a brief summary of the analysis performed for a DHCE experiment in the ATR (Advanced Test Reactor). The calculations are based on the current design of the ITV (Irradiation Test Vehicle) to be placed in the central flux trap of the reactor.

RESULTS AND DISCUSSION

Results of analyses conducted include neutron filtering, filter depletion, tritium leakage, and distribution coefficient. Results and discussions for each of these areas are presented.

1. Neutron Filtering.

The large thermal/epithermal neutron interaction cross section of the ^3He makes the use of thermal neutron absorbers, an imperative for DHCE experiments performed in mixed spectrum fission reactors. ATR is among the available mixed spectrum reactors suitable for fusion materials irradiation that can provide a relatively large fast to thermal neutron flux ratio. Despite this favorable characteristic, the requirement for the use of an effective thermal neutron filter is essential.

The current design of the ITV calls for a replaceable external neutron filter 2.6mm thick made of borated aluminum. The boron concentration in the aluminum is about 4.3% by weight and 100% B-10 is used instead of natural boron. The diameter of the region inside the filter is about 7.4cm. Three holes drilled in an aluminum filler of this region contain the irradiation capsules, temperature control gas lines and thermocouple leads. A small water annulus is used as a coolant outside each of the irradiation capsules. The combination of the filling material, samples, and cooling water provides a potential for attenuating neutrons and producing an increase in the thermal and epithermal components of the flux inside the filter region. As an option for enhancing the filter performance the possibility of borating the thermocouple sleeves is also being considered. Since the thermocouple sleeves are a semi-permanent component of the irradiation vehicle, the depletion of the boron in this component has to be kept as low as possible. This is achieved by replacing the external filter periodically.

Figure 1 displays a comparison of the different filter configurations. These results are for a 400°C irradiation, which implies that the tritium leakage is small. The curve labeled "filter1" represents the configuration where only the outside filter is used, the curve "filter2" represents a configuration that both, outside filter and borated thermocouple sleeve is used, and the curve "no dep1" represents that case that the depletion of the external filter is not accounted. The results shown are for natural lithium, initial tritium charge equivalent to 1500 appm in the vanadium sample, temperature of irradiation 400°C, and distribution coefficient between the lithium and vanadium samples of 100 (in weight percent - weight per cent of tritium in lithium over weight per cent of tritium in vanadium). As can be seen, the helium to dpa ratio is maintained within an acceptable range. The performance of the configuration with outside and inside filter has a fairly good performance. The results indicate that using inside and outside filter the outside filter can be replaced after 5 dpa is reached without much of an impact to the helium to dpa ratio. The configuration with only outside filter suffers a clear reduction of performance above 5dpa, indicating the need for replacement of the external filter above this level.

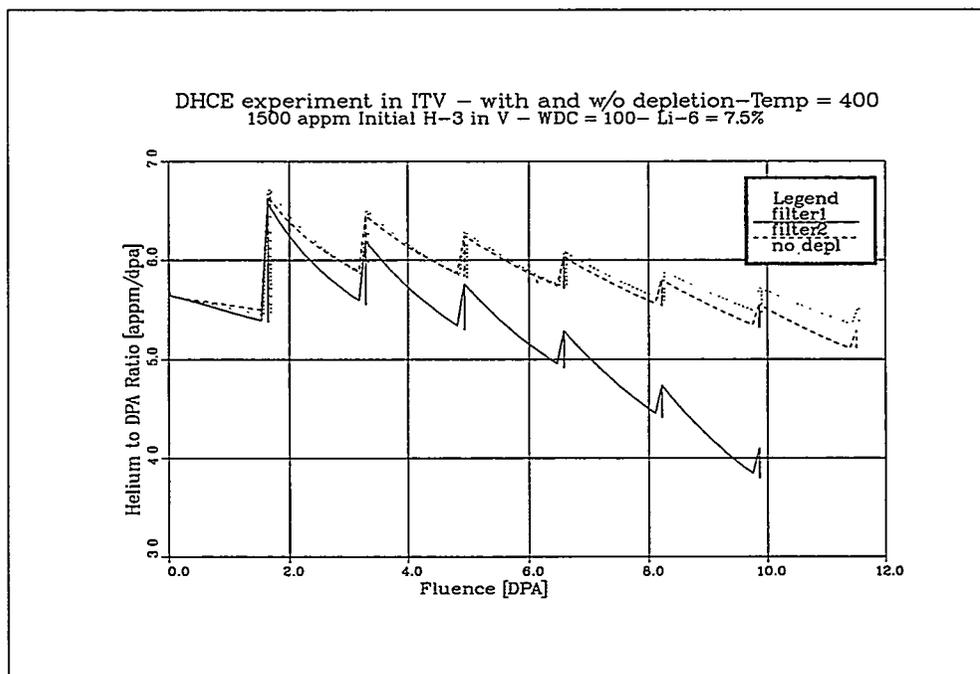


Figure 1. Calculated evolution of the helium to dpa ratio during irradiation of vanadium alloys in a DHCE experiment in the ITV position of ATR. The values plotted are for a temperature of 400°C, a weight distribution coefficient of 100, and lithium thermal bonding with natural enrichment.

2. Filter Depletion.

The depletion of the thermal neutron absorbing atoms of the filter material is an important parameter to be analyzed in the design of an experiment. During irradiation the number of absorbing atoms is reduced and the effectiveness of the filter decreases. As indicated in Figure 1, the filter depletion was taken into account and the neutron flux, at the sample position, was calculated each cycle to reflect the new filter atomic composition. Except for small local variations the neutron flux represents fairly well what would be expected during the irradiation length of an experiment.

3. Tritium Leakage

The permeation of tritium through the subcapsule wall is a function of the tritium concentration inside the capsule and the leakage coefficient. An equation that describes the variation of the number of tritium atoms inside the capsule as a function of the time can be written as follows:

$$dN_T/dt = -L N_T \quad (1)$$

where N_T is the number of tritium atoms at the time t and L the leakage coefficient. The leakage coefficient is a function of the diffusion coefficient of tritium in the capsule material, distribution coefficient of tritium between capsule material and lithium, surface area of the capsule in contact with lithium, among others. A way to represent this coefficient is given as follows:

$$L = D^c K_a (\Omega_L/\Omega_c) (S/dV_L) \quad (2)$$

where D^c is the diffusion coefficient of tritium in the capsule material, K_a^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 is basically theoretical and does not take in account the effects of the surface, this means that the tritium is considered to be free to leak out of the surface. The diffusion coefficient of tritium 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 strong effect in inhibiting the tritium leakage. Also, desorption of tritium at low pressures was not considered in this calculation. In the analysis performed, the leakage coefficient was assumed to be diffusion controlled, without taking credit for the possible improvements represented by the use of surface barriers for the tritium.

Figure 2 displays a comparison of the helium to dpa ratio for different temperatures considering all other variables constant. The results presented are for the configuration with only outside filter, for a distribution coefficient of 100, natural lithium enrichment, and initial tritium charge equivalent to 1500 appm in the vanadium samples. The values presented in this plot are calculated based on the diffusion controlled leakage, without any credit for the use of some kind of auxiliary tritium barrier or for H desorption rate control. As can be seen, for 600°C the helium to dpa ratio decreases considerably over the irradiation time, being at 5 dpa roughly 4 and at 10 dpa roughly 2. In this case filter replacement would not improve the performance. The possible solution is to have a more effective filter associated with a higher lithium enrichment, to reduce the leakage through the use of a tritium barrier in the outside capsule wall, or to include possible benefits of desorption controlled permeation.

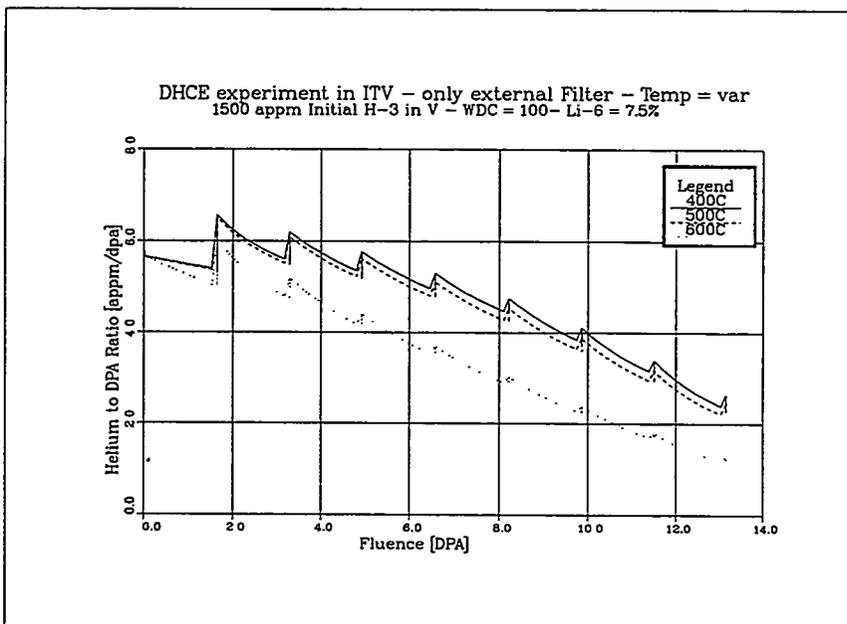


Figure 2. Calculated helium to dpa ratio during the irradiation for different irradiation temperatures. The lithium thermal bonding has natural enrichment, only outside filter is used, and initial tritium concentration is equivalent to an equilibrium tritium concentration in the vanadium samples of 1500 appm.

4. Distribution Coefficient

Distribution coefficient is defined as the ratio of the equilibrium concentration of an element able to diffuse through two media that are in contact. In this case the term distribution coefficient is used to represent the ratio of the tritium concentration in lithium to that in vanadium. This quantity is given the symbol WDC (weight distribution coefficient) or ADC (atomic distribution coefficient). The quantity which is the inverse (concentration of tritium in vanadium over concentration in lithium) is given the symbol K_w or K_a , depending on being weight or atomic distribution coefficient, respectively.

In this analysis the WDC was let to vary from 50 to 500. Studies performed by Park et.al. [1] indicated that this value should be in the range of roughly 100-150 depending on temperature and alloy composition.

Figure 3 and 4 show a comparison of the estimated helium to dpa values for a 500°C irradiation, with natural lithium. Figure 3 displays calculated values using only outside filter and Figure 4 using inside and outside filter. Figure 3 presents the worst scenario where only outside filter is used and the filter is not replaced for the full length of the irradiation. As it was shown in Figure 4, the use of inside and outside filter improves considerably the overall performance of the configuration. Important information that can be extracted from these plots is that the distribution coefficient has some impact on the helium to dpa profile during the irradiation but its impact is not as large as the temperature.

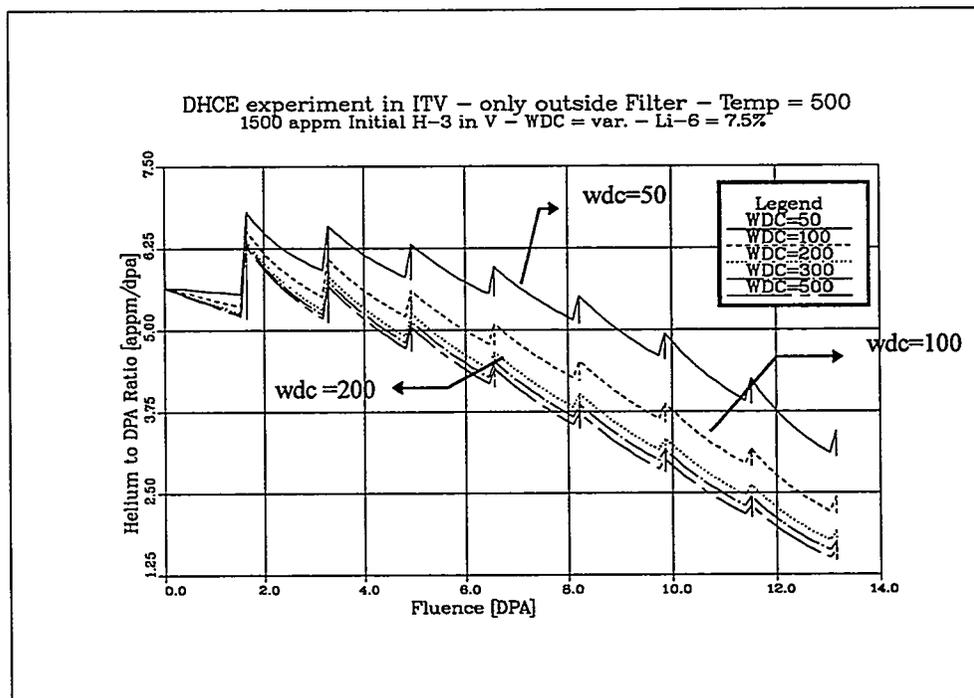


Figure 3. Calculated values of the helium to dpa ratio during irradiation of vanadium alloys in the ITV position of ATR. The temperature of irradiation is 500°C, the lithium thermal bounding has natural enrichment, only outside filter is used, and initial tritium concentration is equivalent to an equilibrium tritium concentration in the vanadium samples of 1500 appm.

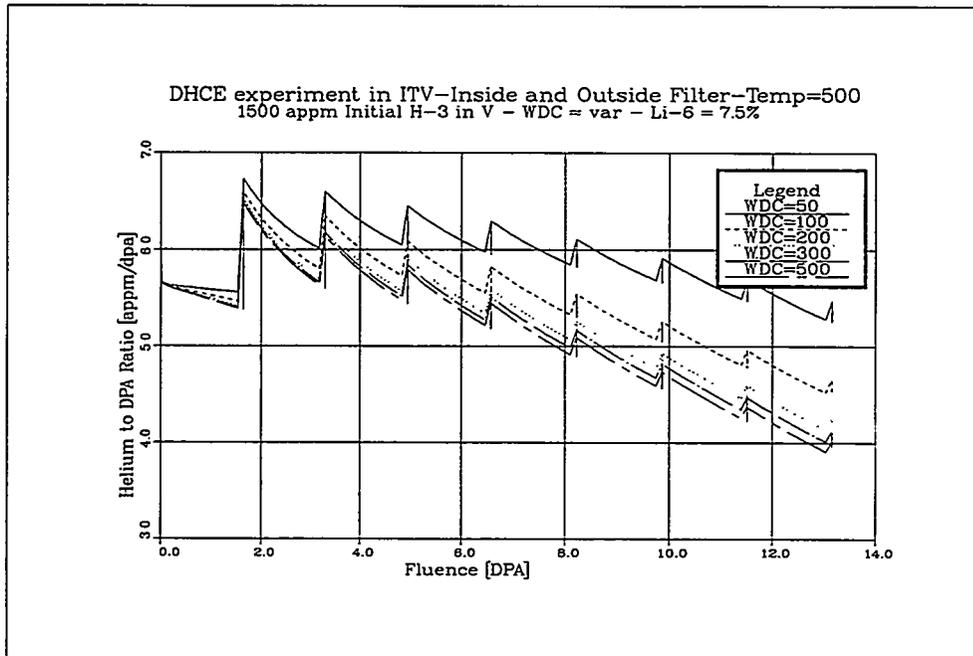


Figure 4. Calculated values of the helium to dpa ratio during irradiation of vanadium alloys in the ITV position of ATR. The temperature of irradiation is 500°C, the lithium thermal bounding has natural enrichment, outside and inside filter is used, and initial tritium concentration is equivalent to an equilibrium tritium concentration in the vanadium samples of 1500 appm.

FUTURE WORK

The future activities in the analysis of the DHCE experiment in the ITV include refinement of the current geometric module of the irradiation train, inclusion of the axial profile of the neutron flux into the model with, if possible, the use of the full model of the ATR core to better represent local and global variations of the neutron flux, interaction with the ITV designers to achieve the design goals of developing an operational vehicle suitable for fusion materials irradiation. Also, calculations for the total tritium charge for each capsule (as soon as a final design of the capsule is available), the total tritium inventory and estimated tritium leakage, as well as other parameters of the experiment are to be performed to support the design of a DHCE experiment in the ITV position of ATR.

REFERENCES

- [1] J.-H. Park, R. Erck, S. Crossley and F. Deleglise, Fusion Materials Semiannual Progress Report for ending period December 31, 1996, DOE/ER-0313/21, April 1997, pp. 45-51.