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EXPERIMENTAL METHOD FOR INVESTIGATING HELIUM EFFECTS IN IRRADIATED VANADIUM*

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ABSTRACT

Analyses have been performed which indicate that an effective method for experimentally investigating helium effects in neutron irradiated vanadium base alloys can be developed. The experimental procedure involves only modest modifications to existing procedures currently used for irradiation testing of vanadium-base alloys in the FFTF reactor. Helium is generated in the vanadium alloy by decay of tritium which is either preinjected or generated within the test capsule. Calculations indicate that nearly constant He/dpa ratios of desired magnitude can be attained by proper selection of experimental parameters. The proposed method could have a major impact on the development of vanadium base alloys for fusion reactor applications.

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1. Introduction

Vanadium-base alloys exhibit several properties that are favorable for fusion reactor first wall/blanket structure applications [1-3]. Vanadium and selected alloys qualify as low activation materials with regard to waste management. The alloys exhibit good high temperature strength and relatively low thermal stresses because of low thermal expansion and high thermal conductivity properties. Vanadium is compatible with high purity lithium. Limited data indicate that certain vanadium-base alloys are highly resistant to radiation induced swelling. Dominant concerns regarding the use of vanadium-base alloys for fusion reactor applications relate to: nonmetallic element interactions, fabricability issues, and sensitivity to radiation embrittlement.

Irradiation-induced embrittlement is a major concern for all structural alloys exposed to a fusion (14 MeV) neutron environment. Of particular importance are the effects of gaseous transmutation products, particularly helium. Since a high flux 14 MeV neutron source does not exist, information on these effects must be obtained by simulation techniques. An example of this is the well known ^{58}Ni(γ)^{59}Ni(n,α) reaction to generate helium in nickel-containing austenitic steels during exposure in a mixed-spectrum reactor such as ORR and HFIR [4]. Currently, a reliable method does not exist for simulating high helium generation rates in neutron irradiated vanadium-base alloys.

This paper describes a new method that can provide important information on the simultaneous effects of neutron displacement damage and fusion-relevant helium production rates on the properties of vanadium-base alloys. The proposed method provides for fusion relevant helium production rates during irradiation in a fast fission reactor (FBR) at displacement damage rates similar to those projected for commercial fusion reactor applications. This paper also identifies further analyses and supporting out-of-pile experiments that are needed to qualify the method before practical in-reactor tests can be conducted.

2. Experimental Concept

The proposed experimental method provides for simultaneous neutron displacement damage and helium generation in vanadium-alloy test specimens in the following manner.

a. Vanadium-base alloy specimens are encapsulated with lithium as shown schematically in Fig. 1 for irradiation in a fast fission reactor (e.g., FFTF). The lithium provides for uniform temperature control, and displacement damage in the vanadium-base alloy produced is by fast neutron interactions [5].

b. Helium is generated uniformly in the vanadium alloy by the following mechanisms.

- Tritium is generated in lithium by neutron reactions and/or by precharging tritium into the lithium.

- At elevated temperatures the tritium is highly mobile and diffuses rapidly from lithium into the vanadium alloy attaining predictable near-equilibrium conditions.

- The tritium in the alloy will decay to helium (^{3}He) in the alloy a rate of 5.5% per year, which is determined by the half-life of tritium.

- The helium generation rate in vanadium can be conveniently varied over a large range to obtain desired values by adjusting the ^{6}Li enrichment and/or the precharge tritium concentration.

Several inherent parameters in this technique have sufficient flexibility to provide variations in He/dpa ratios of interest. Since most of the data base required to verify this method is available, only modest development is required to refine the method for practical application. Quantitative features of the method are briefly summarized below.

2.1 Displacement Damage Rate

Fast reactor (FFTF) irradiations provide a neutron displacement damage rate of 30-60 dpa/year in vanadium-base alloys. This compares to a predicted displacement rate of ~45 dpa/year in a fusion reactor first wall with a nominal 5 MW/m^2 neutron wall load.

2.2 Tritium Generation in Lithium

Tritium generation rates in lithium can be accurately calculated by current neutronic and dosimetry methods [6]. The total tritium generation rate from the $^6Li(n,\alpha)T$ reaction can be varied by over two orders of magnitude by adjusting the ^6Li enrichment (<1 to 90%). This will provide for a similar variation in the helium generation rate in vanadium. Minor predictable variations will also occur because of neutron spectrum changes associated with reactor core location. Small amounts of tritium preinjected in the lithium can be used to adjust the tritium source in the initial stages. Since the preinjected tritium can be retained in the lithium until the temperature is raised at the time of insertion into the reactor, helium generation in vanadium can be delayed almost until the displacement damage is initiated.

2.3 Tritium Redistribution into Vanadium

Tritium is highly mobile at temperatures of interest (>300°C) and will redistribute into the vanadium in very short times (<< hour). Therefore, nearly equilibrium conditions will exist at temperatures above 300°C. The equilibrium distribution coefficients for hydrogen in the vanadium-lithium system have been calculated as a function of temperature from temperature dependent Sieverts' constants (K_s) for the vanadium-hydrogen and lithium-hydrogen systems [7].

$$\ell n\ K_s = -5.653 + \frac{3490}{T\ (^0K)} \quad \text{for vanadium.}$$

$$\ell n\ K_s = -5.449 + \frac{6460}{T\ (^0K)} \quad \text{for lithium.}$$

Calculated distribution coefficients (K_A) based on these Sieverts' constants where K_A = appm H in V/appm H in Li are given below.

T (°C)	K_A
650	0.033
550	0.022
450	0.013

Preliminary experiments to determine the equilibrium distribution of hydrogen between lithium and V-15Cr-5Ti have been conducted at temperatures of 600-650°C [8]. The measured distribution coefficient is relatively insensitive to temperature with a value of approximately

$$K_A \sim 10 \frac{\text{appm H in V}}{\text{appm H in Li}} ,$$

which is considerably larger than the calculated value for pure vanadium. Additional measurements are required to resolve these discrepancies and to provide a data base for other vanadium alloy compositions.

2.4 Helium Generation Rates in Vanadium

A tritium concentration in vanadium of the order of 400 wppm will provide the desired helium generation rate that corresponds to a He/dpa ratio of ~5 at a damage rate of ~50 dpa/year. This concentration of hydrogen (tritium) in vanadium is not expected to produce significant effects on mechanical properties of the alloys at the temperatures of interest. It is necessary to consider the neutron burnup of ^{3}He in the vanadium to obtain an accurate measure of the helium generation rate. For the FFTF spectrum, the ^{3}He burnup rate is higher than the tritium decay rate. Therefore, the concentration of tritium in the alloy must exceed the desired concentration of helium.

2.5 Tritium Mass Balance

Other factors that affect the tritium mass balance in the system must be considered to obtain desirable helium generation rates in the vanadium alloy. These include tritium permeation through the capsule wall and tritium inventory in the capsule wall. In order to minimize these effects the capsule should be constructed of a material with low hydrogen (tritium) permeability and solubility. Molybdenum alloy TZM is currently used as a capsule material. Tungsten exhibits lower permeability characteristics and would be preferred in this respect; however, this material is extremely difficult to weld. The importance of the permeation factor is evaluated by comparison of the leakage from capsules of these two materials.

The magnitude of the leakage Q_L can be evaluated as follows if the leakage is controlled by diffusion of tritium through the wall.

$$Q_L = \frac{D\ S\ N_c}{d\ V_L}$$

where D is the diffusivity, S is the surface area, N_c is the tritium concentration immediately below the inner surface of the capsule, d is the wall thickness, and V_L is the volume of lithium. The equilibrium distribution coefficient K_A^c for tritium in the wall material/lithium system is given by

$$K_A^c = \frac{N_c}{N_L}$$

where N_C and N_L are the concentrations of tritium in the capsule material and lithium, respectively. These two equations can be combined to provide an expression for the leakage rate as a function of the tritium concentration in lithium. For a representative capsule size (10 mm-diameter × 20 mm-long × 1 mm-wall), the leakage coefficients for molybdenum and tungsten capsules are 4.0×10^{-12} and 3.7×10^{-9} s^{-1}, respectively. The value for the molybdenum capsule in this case is comparable to the tritium decay constant of 1.8×10^{-9} s^{-1}. Therefore, leakage from the molybdenum capsule must be considered when determining the helium production rate in the test alloy. However, leakage from a tungsten capsule (or equivalent) is insignificant.

2.6 Calculated Time Dependence of Helium Generation

The time dependence of the helium generation rate in the vanadium alloy can be controlled over a wide range by adjusting the ^{6}Li enrichment and the precharged tritium concentration in lithium. Figures 2-4 show a comparison of the predicted helium-to-displacement damage rate for a V-15Cr-5Ti alloy exposed to a fusion neutron environment (dashed curve: appm He/dpa = 5) with calculated values for several experimental conditions. All three sets of calculated values are based on the experimentally determined distribution coefficients and equal volumes of vanadium alloy and lithium in each capsule.

Figure 2 shows results for two base cases. The open circles represent a case where tritium is generated only by 16% enriched ^{6}Li. this curve is similar to the values obtained for Type 316 stainless steel exposed in ORR or HFIR as a result of the ^{59}Ni reaction to form helium. The squares represent a case where the only tritium source is 3% tritium preinjected in the lithium prior to the experiment. The irregular pattern of the calculated values arises from the projected irradiation cycle for FFTF, which is assumed to be

100 days on and 30 days off. The ^{3}He increases rapidly when the reactor is turned off and ^{3}He generation is controlled by tritium decay. The ^{3}He generation rate is slower when the reactor is on because of the significant burnup of ^{3}He back to tritium.

Figure 3 indicates effects on helium generation rates that correspond to projected tritium leakage from the capsule for case 1 above. Predicted tritium leakage rates that correspond to a tungsten capsule are negligible. Leakage rates corresponding to those for a molybdenum capsule result in slightly lower helium generation rates.

Figure 4 indicates calculated values obtained for two cases where helium is generated both from preinjected tritium and from tritium produced from enriched ^{6}Li. The case with 1 at. % tritium and 13% ^{6}Li (open squares) illustrates that nearly constant He/dpa ratios appear attainable with appropriate selection of two primary variables; viz., preinjected tritium concentration and ^{6}Li enrichment. The other case shows that nearly desired helium levels can be obtained with natural lithium if larger amounts (2-3%) of tritium are preinjected.

2.7 Other Considerations

A significant amount of tritium will also be generated within the lithium in the capsule. This helium will collect in the plenum region at the top of the capsule. The plenum must be sized so as to accommodate the pressure buildup from the helium, which is more important at high fluences. As an example, at 450°C the tritium concentration in lithium based on calculated distribution coefficients is about 50 times the concentration in vanadium (for alloys with titanium this value is expected to be less). For a He/dpa ratio of 5, the amount of helium generated at 100 dpa is 500 appm in vanadium and

25000 appm in lithium. This corresponds to ~90 cm^3 (STP) of helium generated per gram of lithium. The pressure buildup in a 1 cm^3 plenum corresponds to about 100 MPa. Several more detailed aspects must be considered, which tend to ameliorate this problem. Since the calculated distribution coefficient decreases as the temperature increases, the pressure will be less at higher temperature. The distribution coefficients for the alloys are expected to be less than for vanadium and the ratio of lithium to vanadium can be reduced within limits. Although pressure buildup must be considered in the capsule design, this does not appear to be a prohibitive problem.

3. FURTHER DEVELOPMENT REQUIRED

Additional analyses and experiments must be conducted to refine the method, to improve the reference base, and to verify the analyses before the method can be validated. The following information must be obtained before the method can be used for routine in-reactor tests.

- Experimental confirmation of the thermodynamics and kinetics of hydrogen (tritium) redistribution between lithium and vanadium alloys of interest is required. The differences between calculated and measured distribution coefficients for conditions of interest must be resolved.

- Details of the capsule design and analysis of tritium inventory/leakage for specific experiments are required. Effective tritium barriers will be required for the higher tritium partial pressures predicted from the Sieverts' constants.

- More detailed evaluation of the tritium handling aspects is required. Procedures for preinjection of tritium and for post-irradiation examination must be developed.

- Additional analytical calculations are needed to optimize the reference test parameters for selected alloy compositions, exposure conditions, and test capsule design.

4. CONCLUSIONS

Analyses have been performed which indicate that an effective method for experimentally investigating helium effects in neutron irradiated vanadium base alloys can be developed. The experimental procedure involves only modest modifications to existing procedures currently used for irradiation testing of vanadium-base alloys in the FFTF reactor. Helium is generated in the vanadium alloy by decay of tritium which is either preinjected or generated within the test capsule. Calculations indicate that nearly constant He/dpa ratios of desired magnitude can be attained by proper selection of experimental parameters. The proposed method could have a major impact on the development of vanadium base alloys for fusion reactor applications.

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FIGURE CAPTIONS

Figure 1 Schematic diagram of capsule for investigating helium effects in neutron irradiated vanadium alloys.

Figure 2 Comparison of He/dpa ratio for two base cases with desired value of 5 for fusion neutron environment.

- squares represent helium produced by tritium preinjection (3% T) only
- circles represent helium produced by neutron reactions only (16% ^{6}Li enrichment)

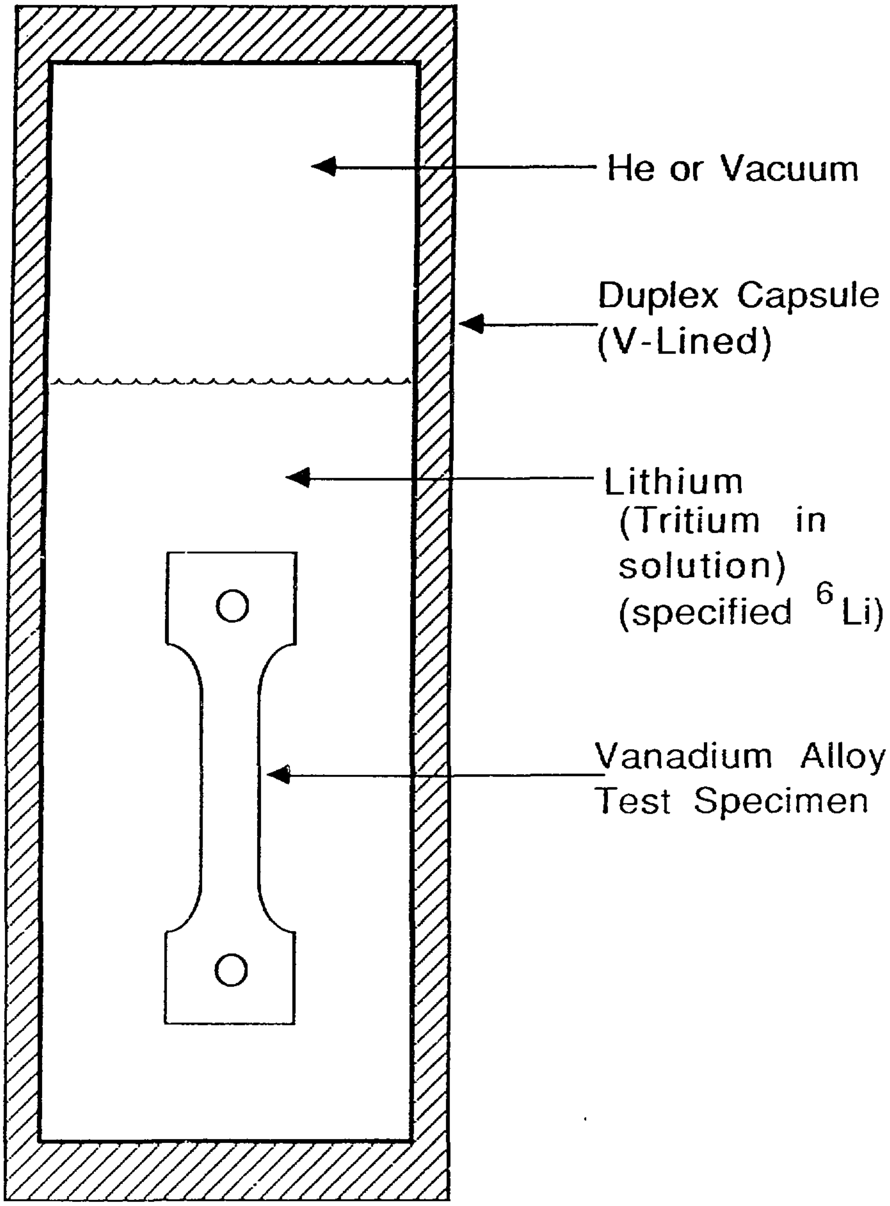
Figure 3 Comparison of predicted helium generation rates for different capsule leakage rates.

- squares assume Mo capsules
- circles assume W capsules

Figure 4 Comparison of predicted helium generation rates for refined case with both preinjected tritium and ^{6}Li enrichment.

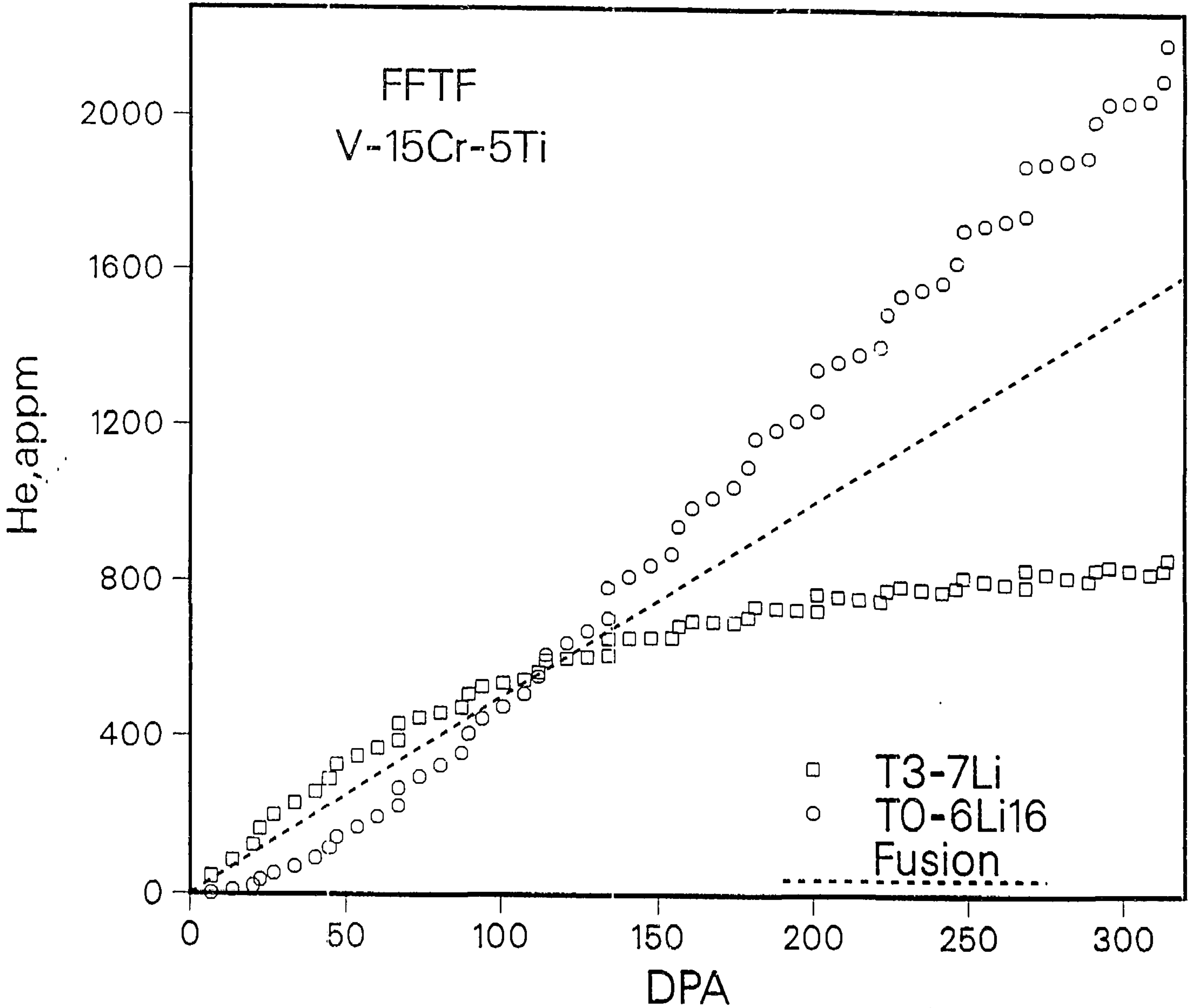
- squares: 1% T preinjected with 13% ^{6}Li enrichment shows good correlation with desired ratio of 5
- irregular plot of symbols indicates effects of projected reactor run cycles

Figure 1

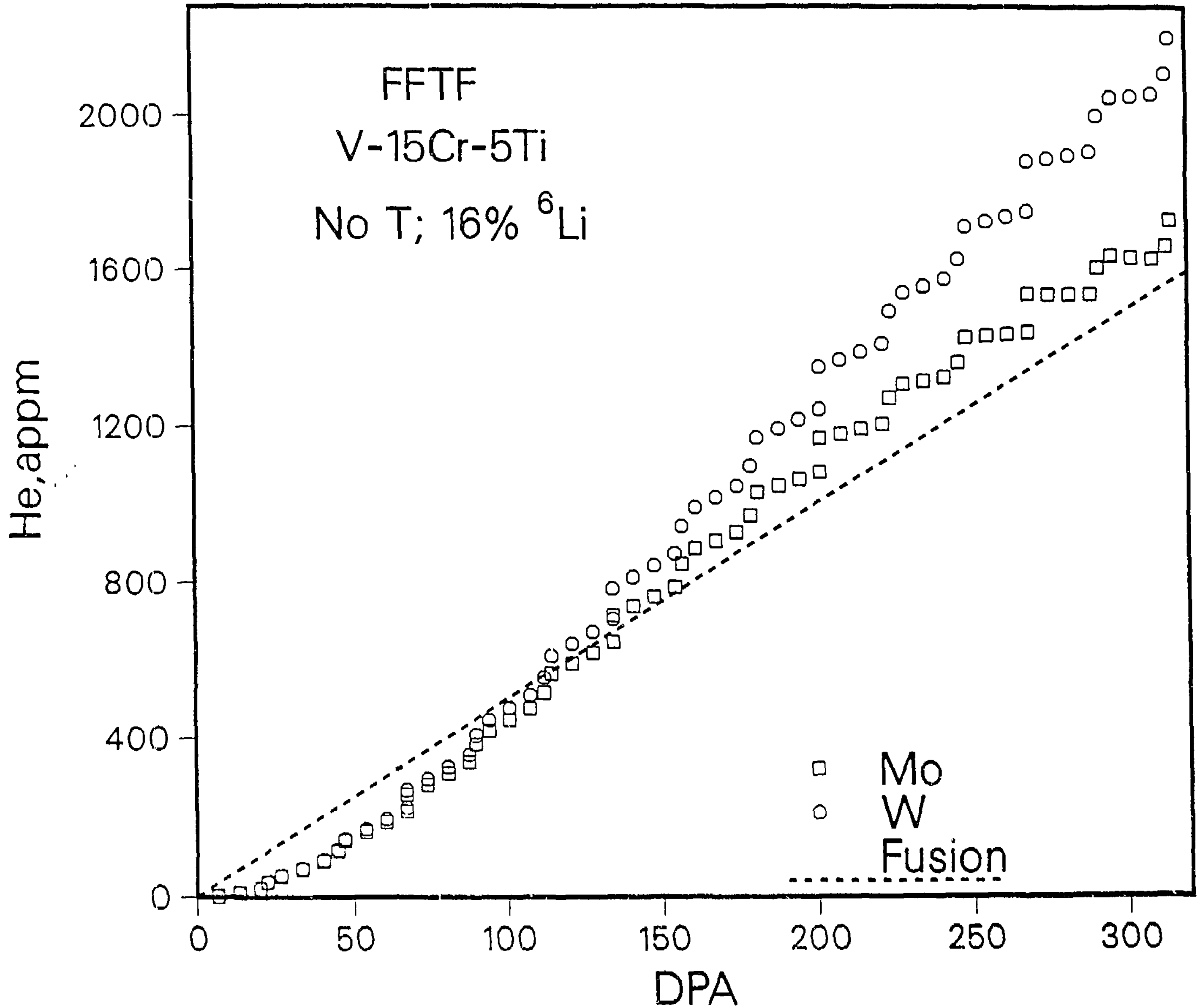


Tritium in lithium diffuses into vanadium where it decays to helium at predictable rate to provide desired He/dpa ratio.

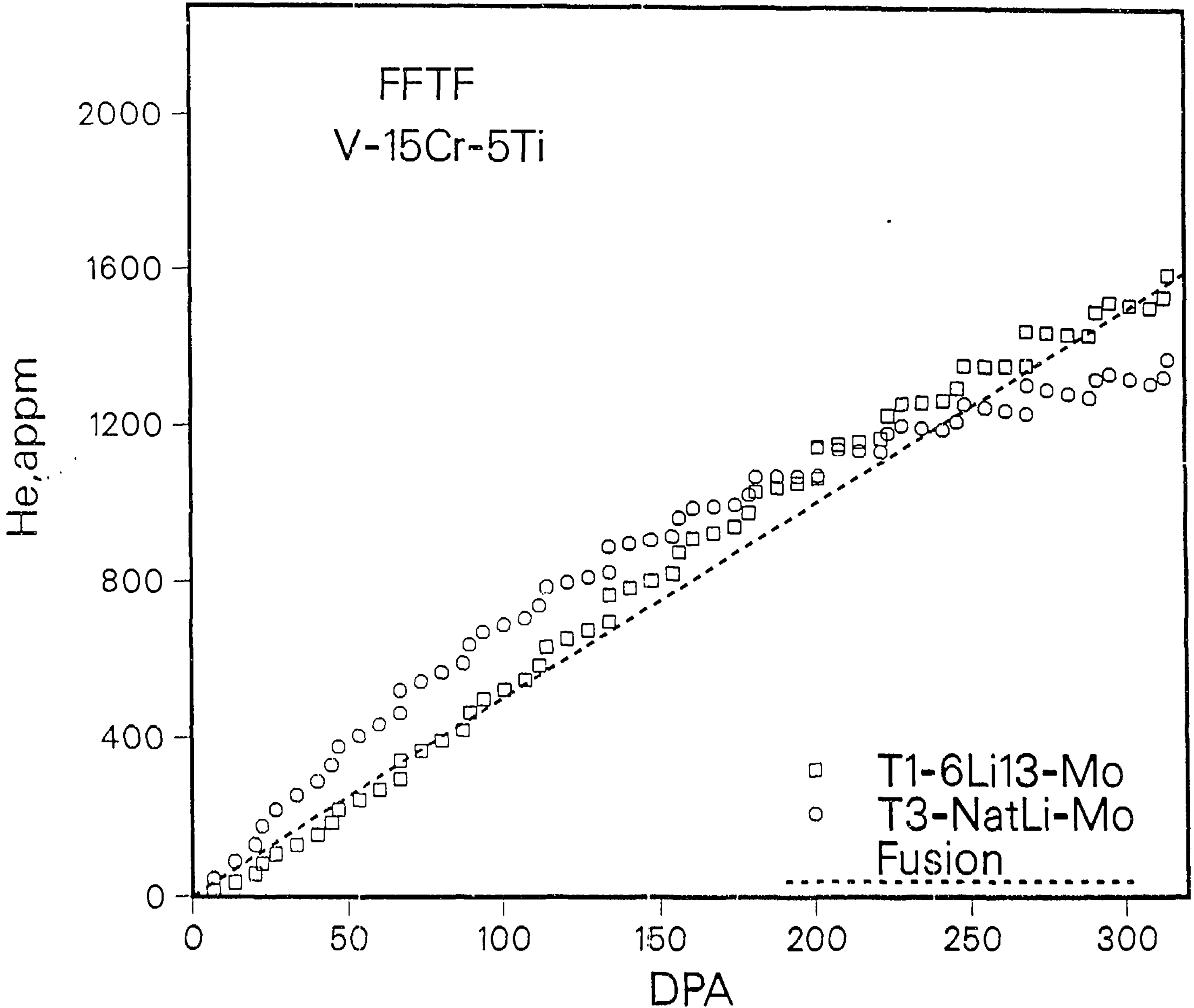
Schematic Diagram of Capsule for Investigating Helium Effects in Neutron Irradiated Vanadium



Preinjected Tritium (3%) vs Enriched ^{6}Li (16%)



Comparison of T Leakage from Mo and W Capsules



Simultaneous ^{6}Li Enrichment and T Preinjection