# EFFECTS OF NON-STEADY IRRADIATION CONDITIONS ON FUSION MATERIALS PERFORMANCE

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#### Abstract

During startup of fusion reactors, materials are exposed to neutron irradiation under non-steady temperature condition. Since the temperature of irradiation has decisive effects on the microstructural evolution, the non-steady temperature will have important consequences in the performance of fusion reactor materials. In the present study, a series of vanadium based alloys have been irradiated with neutrons in a temperature cycling condition. It has been found from this study that cavity number density is much greater in temperature cycled specimens than in steady temperature irradiation. Keeping the upper temperature constant, cavity number density is greater for smaller difference between the upper and the lower temperature. It follows that relatively small temperature excursions may have rather significant effects on the fusion material performance in service.

# **1. INTRODUCTION**

Materials in fusion reactors are exposed to neutron irradiation under conditions that are often not steady. During normal start-up and shutdown, partial load operations, or disruption events, temperature, stress, neutron flux, etc. change with time in very complex ways. Temperature of irradiation, for instance, has decisive effects on the microstructural evolution in materials<sup>[1]</sup>. Defect clusters nucleate from elemental defects more frequently at low temperatures, while at high temperatures, the growth of defect clusters predominates. If the material is irradiated at a temperature that is lower than the design temperature, defect cluster density resulting from the irradiation at the design temperature will be much higher because of the high cluster nucleation rate during the low temperature irradiation. High defect density usually result in large irradiation hardening, leading to DBTT increase. Since material performance has been evaluated based on irradiation data obtained under steady irradiation conditions, non-steady irradiation conditions anticipated in fusion reactors may significantly alter the performance of materials. In the present paper, effects of temperature history on the microstructural evolution in fusion materials are presented. Particular emphasis is placed on the temperature cycling effects in vanadium alloys.

#### 2. EXPERIMENTAL

A series of vanadium based binary alloys have been prepared to cover a wide range of solute atomic size factor, i.e. V-5%Fe, -1%Si, -5%Cr, -5%Ti, -5%Mo and -5%Nb in addition to unalloyed vanadium. Irradiation was done in JMTR using specially designed temperature control rig with a set of electric heater. With this rig, precise temperature control is possible without being interfered by the change in gamma heating rate caused by the reactor power change. Temperature was changed in an alternate fashion between two temperatures, i.e. T<sub>high</sub> and T<sub>low</sub>, for six cycles up to 0.15dpa (see Fig. 1). Here, the temperature combinations in Centigrade are 200/400, 350/400 and 350/450. Transmission



Fig. 1 Temperature and reactor power history.

<sup>&</sup>lt;sup>1</sup>. M. Kiritani, T. Yoshiie, S. Kojima, Y. Satoh and K. Hamada, J. Nucl. Mater. 174(1990)327.

electron microscopy and positron annihilation lifetime measurements were conducted after the irradiation.

## **3. RESULTS**

### 3.1. Positron annihilation

Fig. 2 shows the raw data of positron annihilation lifetime measurements on V-1%Si specimens after irradiation at different irradiation conditions. Count of positron annihilation events is plotted as a function of time after positron is injected to the specimen. The curvature after the maximum is characteristic of the lifetime of the positron. The result of two-component analysis is given in the two diagrams in the middle (intensity) and at the bottom (lifetime). Lifetime component longer than 400ps is observed in specimens irradiated at 200°C-constant and 200/400 and 300/400 temperature cycling. This long lifetime component is characteristic to microvoids. The absence of microvoids in other specimens does not necessarily mean that there are no cavities at all, which will be discussed later. Important point to note here is that the intensity of the microvoid component is rather significant in specimens irradiated in 200/400 and 300/400 cyclic temperature condition. Other alloys show similar trends except for V-Ti alloy, where there is no lifetime component longer than 200ps in all the specimens after irradiation under a variety of conditions. Mono-vacancies in pure vanadium has Fig. 2 Positron annihilation lifetime measurement. positron lifetime of about 185ps and the second lifetime components in all of the V-5Ti specimens



are very close to this value, with an exception of 200°C constant temperature irradiation, where the lifetime is somewhat longer than 185ps indicative of some small vacancy cluster formation.

#### **3.2. Electron microscopy**

Fig. 3 shows a set of electron micrographs of V-1%Si alloy irradiated under different temperature conditions. The 400°C constant temperature irradiation, cavities are several tens nm in diameter and the density is low. On the other hand in the two micrographs on the left showing specimens corresponding to 200/400 and 300/400°C temperature cycling irradiation, cavity density is



Fig. 3 Cavity microstructure of V-1%Si alloy after irradiation under various temperature conditions.

much higher and the size is smaller than the 400°C constant temperature irradiation. In the previous section of positron annihilation, absence of microvoids has been noted in specimens irradiated under constant 400°C and 300/450°C temperature cycling condition. Comparing with the relatively large voids in these specimens shown in Fig. 3, it is apparent that microvoids have grown resulting in the density too low to be detected by positrons.

In V-5%Mo alloy, similar tendency is seen. In this case, however, no cavities are seen in the 200/400 temperature cycled specimen, which is in contrast with V-1%Si in Fig. 3. By positron annihilation lifetime measurement (not shown here), microvoids are detected in 200°C constant temperature irradiation and also in 300/400°C temperature cycling irradiation but not in other conditions. These results suggest that microvoids formed at 200°C in this alloy cannot act as stable nuclei during 400°C irradiation but dissociate into mono-vacancies which migrate quickly to sinks.

Titanium is one of the most important alloying elements in vanadium based alloys for fusion structural applications. No cavities are observed in V-5% Ti alloy in all irradiation conditions and only feature observed is precipitates which is presumed to be titanium oxide. The absence of cavities in this alloy is in good accordance with the positron annihilation data indicating existence of monovacancies or small clusters only.

# 4. DISCUSSION

The intensity of the second lifetime component roughly correspond to the number density of the defect clusters responsible for this component, i.e. microvoids, in the present case. The density of microvoids may be obtained from the lifetime and intensity data using the trapping theory<sup>[2]</sup>. The void number density determined from positron data along with those obtained from electron microscopy are plotted in Fig. 4 for all the alloys studied in this research.

The behavior looks rather complicated while there is a certain rule especially in those alloys in the top row including pure vanadium. The graphs are arranged in the order of atomic size of the solute in the alloys. Iron is the smallest and chromium is slightly smaller than the matrix vanadium lattice. The bottom row is for oversized solutes. According to Fig. 4, the void number density in V-5% Cr alloy specimen, for instance, after the constant temperature irradiation at 200°C and at 400°C is



Fig. 4 Summary of void density data in all the alloys studied. Void size is also plotted in the inset above each bar-graph. Data are derived either from positron lifetime or TEM.

<sup>&</sup>lt;sup>2</sup>. W. Brandt and A. Dupasquier (ed.) *Positron Solid-State Physics*, North-Holland, Amsterdam, (1983).

 $3.7 \times 10^{23} \text{m}^{-3}$  and  $6.1 \times 10^{20} \text{m}^{-3}$ , respectively. Thus, cavity number density is highest for 200°C constant temperature irradiation and is lowest for 400°C constant temperature irradiation. By temperature alternation between 300 and 400°C, the void number density becomes by a factor of 30 greater than the value obtained after 400°C constant temperature irradiation. It is generally observed that the temperature cycling between 300 and 400°C, rather than that between 200 and 400°C, yields higher cavity density. This is somewhat strange since the number density of cavity nuclei is expected to be higher for lower irradiation temperatures.

The stability of vacancy clusters depends on the size of the cluster, i.e., the larger the cluster size, the more stable they are. This is because the binding energy of a vacancy to a cluster increases with cluster size. During the low temperature period of the irradiation at  $T_{Low}$ , numerous microvoids or vacancy clusters are formed. Upon heating to  $T_{High}$ , most of these small clusters dissociate by emitting vacancies or by absorbing interstitials. When  $T_{Low}$  is a little higher, the size of the clusters are larger and thus more stable and have better chances to survive during the irradiation at  $T_{High}$ . More quantitative and detailed analysis is clearly needed, while this crude analysis suggests qualitatively why cavity density is higher when  $T_{High} - T_{Low}$  is smaller. Obviously, there will be a temperature difference for which this effect becomes maximum, since very small temperature fluctuation will not cause any significant effect. Anyhow, this effect has a very important consequence since small fluctuations in temperature of some structural component of the reactor may result in rather significant impact on the microstructural evolution, and further, on the radiation resistance of the material.

In V-5% Ti alloy, the situation is entirely different: due to the strong trapping of vacancies by titanium atoms, only mono-vacancy component or small clusters was detected by positron lifetime measurements in all irradiation conditions. This is true up to at least 500°C in V-Ti binary alloys[<sup>3</sup>]. In V-Cr-Ti ternary alloys, which is more important from engineering view point, vacancy mobility appears to be much higher[<sup>4</sup>]. At still higher temperatures expected in fusion reactor applications, vacancies are certainly very much mobile, and even titanium-containing alloys will behave similarly to other alloys.

In another oversize-type alloy, i.e. V-5%Nb, a tendency similar to undersized alloys was observed, but with much lower void density. Molybdenum solute is slightly oversize in vanadium matrix, and although the behavior of V-5%Mo was more complex, the general trend was similar to those observed in undersize-type alloys.

Detailed numerical analysis is being undertaken in order to understand the temperature alternation effects. There are a number of different cases depending on the combination of temperatures, material composition, stability of defect clusters, mobility of elemental lattice defects, etc. Limited analysis shows that majority of vacancy clusters formed at  $T_{Low}$  become unstable by an upward temperature step, while the fraction of surviving defect clusters at  $T_{High}$  is greater if the temperature difference is small. This analysis explains the higher void density observed in 300/400°C temperature combination than in 200/400°C combination.

There are cases where much more complicated behavior is observed. For the assessment of non-steady irradiation effects on fusion reactor materials, it is further necessary to explore the mechanism of microstructural evolution under non-steady conditions both experimentally and theoretically. Effects of non-steady conditions other than temperature also need to be studied. In any case, desirable materials for fusion applications should be those materials rather insensitive to the temperature variation during irradiation. This will be a guiding principle for material development and there may be a clue in V-Ti alloy where no significant difference is seen in specimens irradiated in a variety of conditions.

## CONCLUSION

Temperature cycling during neutron irradiation results in significant change in the microstructural evolution of vanadium based alloys. It has been found that relatively small temperature excursions may result in significant change in material performance. More thorough study covering not only temperature change but also stress, and other environmental parameters is strongly needed.

<sup>&</sup>lt;sup>3</sup>. H. Matsui, K. Kuji, M. Hasegawa and A. Kimura, J. Nucl. Mater. 212-215(1994)784-789.

<sup>&</sup>lt;sup>4</sup>. H. Matsui, et al. unpublished work.