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Swelling of a V-5Fe Alloy After Irradiation in JOYO*

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Synopsis

Void swelling behavior of a vanadium iron binary alloy has been studied using JOYO*** as an irradiation facility. The swelling at a damage level of approximately 14dpa was as high as 30% for 600°C irradiation, which is qualitatively consistent with the large swelling observed after FFTF/MOTA**** irradiation about 30dpa. The evolution of dislocation microstructure is very fast and the sink strength ratio Q is much greater than unity except for the highest damage level studied, yet the swelling rate is more than 2%/dpa. Several possible mechanisms involving segregation effects have been discussed. It has been suggested that any successful model should be capable of predicting the observed swelling rate more than 2%/dpa under condition of Q value much greater than unity.

I. Introduction

In a previous study[1)] using FFTF/MOTA, it has been found that a V-5at.%Fe alloy exhibits a swelling close to 100% at 30dpa at 600°C. This level of swelling is unexpectedly large since body-centered cubic metals and alloys are generally known to have superior resistance to void swelling in comparison with FCC alloys, e.g. austenitic stainless steels [2)3)]. Low swelling rates observed in several ferritic steels and vanadium based alloys are well documented [4)5)] and are often considered to be inherent to the lattice structure [6)7)8)]. The large swelling observed after FFTF irradiation[1)] indicates that this is not the case. The full understanding of the

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[***] A liquid metal cooled experimental fast reactor of PNC (Power Reactor and Nuclear Fuel Development Corporation, Oarai, Japan).

[****] Fast Flux Test Facility/ Materials Open Test Assembly, at Westinghouse Hanford Company, Richland, Washington.

mechanism of the observed large swelling is necessary for succesfull development of swelling resistant alloys.

II. Experimental

The vanadium alloy was prepared from 99.9% pure vanadium and 5-nine iron by argon arc melting. The alloy was cold rolled to 0.25mm and was punched to 3mm disks for transmission electron microscopy. The specimens were doubly wrapped with tantalum foils inside and zirconium foils outside, and sealed in quartz capsules in a vacuum better than 2 x 10^{-5}Torr. They were annealed at 1100°C for two hours. The concentration of interstitial impurities after the above heat treatment was determined using the internal friction method. Nitrogen concentration was below the detection limit of the internal friction technique, i.e. approximately 25appm, while oxygen concentration was approximately 1100appm.

The annealed specimens were sealed with purified helium in stainless-steel capsules for irradiation. The irradiation was conducted in JOYO at 400, 500 and 600°C to four levels of displacement damage, i.e. 0.1, 0.59, 4.2 and 14dpa; the displacement damage was slightly dependent on irradiation temperature especially for the highest damage level of 14dpa, which represents the actual damage of 11.9, 12.6 and 14.1dpa at 400, 500 and 600°C, respectively.

After irradiation, the specimens were electrolytically polished with TENUPOL to prepare thin foils for TEM observation. The electrolyte used was a mixture of 4 part methanol and 1 part hydrosulphuric acid. The polishing was done at room temperature at 14V, 200mA. The thickness of the view area of the TEM foils was mainly determined using stereoscopic observation.

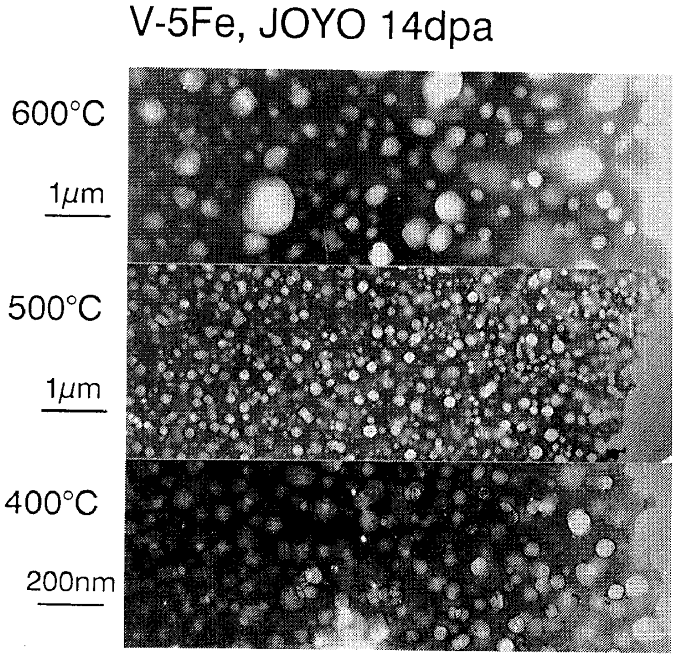

Figure 1 Cavity microstructure of V-5Fe alloy after irradiation in JOYO to 14dpa. Note the different magnification of the 400°C micrograph.

III. Results

Cavity Microstructure

The cavity microstructures of the V-5Fe irradiated to 14dpa are shown in Fig. 1. The size of the cavities becomes larger with temperature. At 600°C, the diameter of some of the cavities becomes close to 1μm. Figure 2 is a plot of cavity number density and their average diameter against irradiation temperature for two dpa levels, i.e. 0.59 and 13dpa. The somewhat irregular trend observed in the number density for 0.59dpa may be caused by insufficient statistics due to the low density.

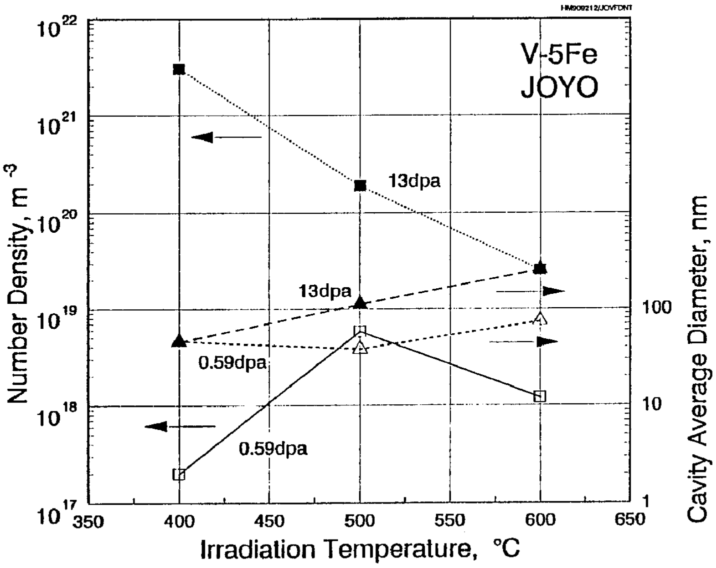

Figure 2 Number density and average size of cavities in V-5Fe after irradiation to 0.59 and 13dpa.

The distribution of cavities was often inhomogeneous at 500°C irradiation. Figure 3 shows three examples of cavity distribution. In the top figure, cavities are localized within a band-like zone about 6μm wide along a grain boundary. The foil is thicker outside of this zone and the visibility of the cavities is reduced, but the difference in the cavity density is obvious. To the contrary, in the micrograph in the middle, the region along grain boundaries is depleted of cavities. These different cavity distributions may reflect the difference in the solute segregation behavior due to the different grain boundary structures. Cavities are also observed just like as threaded beads as shown in the micrograph at the bottom. These strings of cavities never have an end within the crystal, they often form triple junction as shown in this micrograph. Apparently these cavities have been nucleated along preexisting dislocation networks.

Dislocation Microstructure

Figures 4-6 show dislocation microstructures for 400, 500 and 600°C irradiations. At 400°C, dislocations are clustered in fairly dense tangles for low fluences. Density of dislocations increases rapidly with fluence, and above 4.2dpa, the dislocation clusters merge each other so that individual clusters are not discernible any more. Tendency for tangling of dislocations are also visible at 500°C as shown in Fig. 5. Cavity formation in the middle of such tangles is typically observed in this alloy throughout the temperature range studied. At 600°C, the tangling is still seen for low fluence range, while for 14.1dpa specimen, dislocation tangles have disappeared and the dislocation density has decreased dramatically.

It is noted in the previous micrographs, i.e. Fig. 4-6, that some of the dislocations are obviously decorated with some precipitates. The splinter-like image along dislocations in the

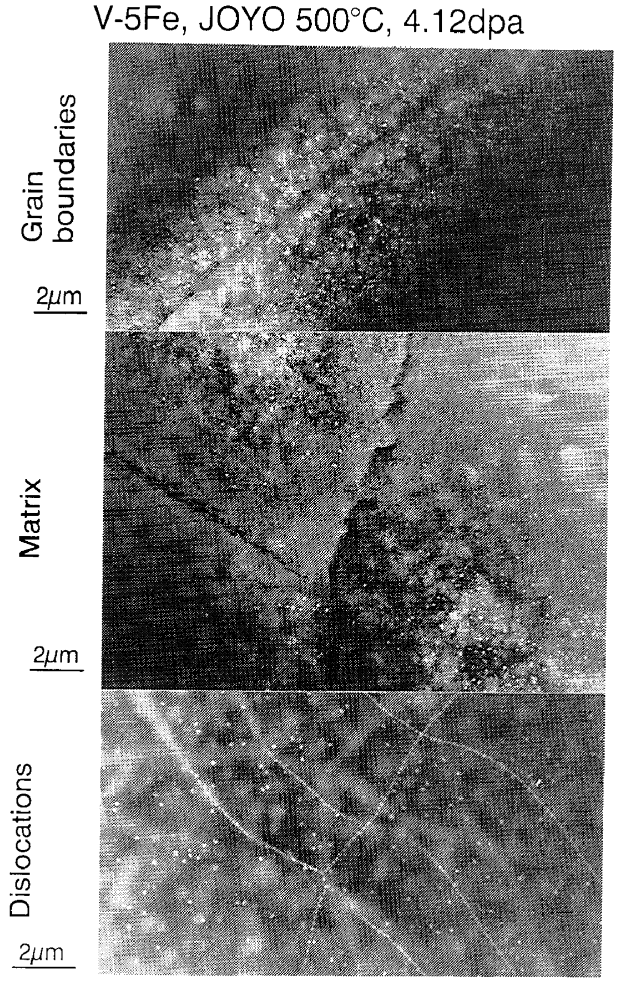

Figure 3 Inhomogeneous distribution of cavities in V-5Fe irradiated at 500°C. Cavity formation along grain boundaries (top micrograph), in the matrix (middle) and along dislocations (bottom).

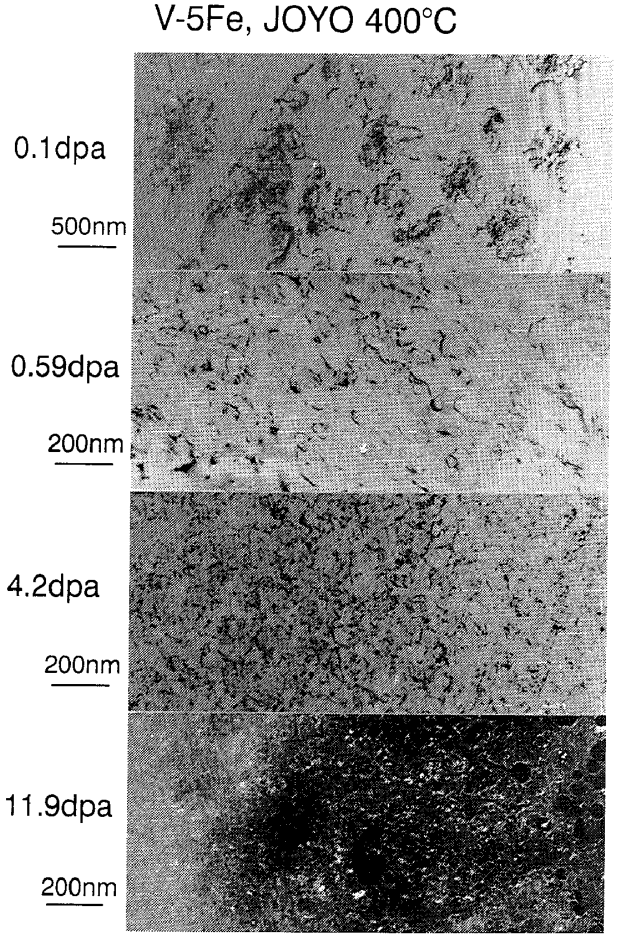

Figure 4 Dislocation images of V-5Fe irradiated at 400°C.Inhomogeneous distribution of cavities in V-5Fe irradiated at 500°C.

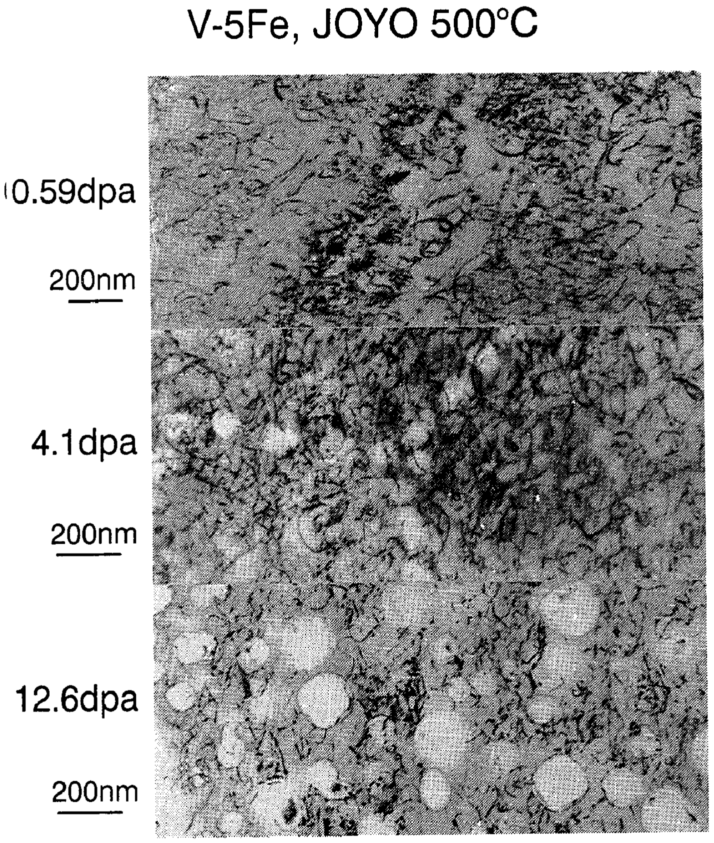

Figure 5 Dislocation images of V-5Fe irradiated at 500°C.

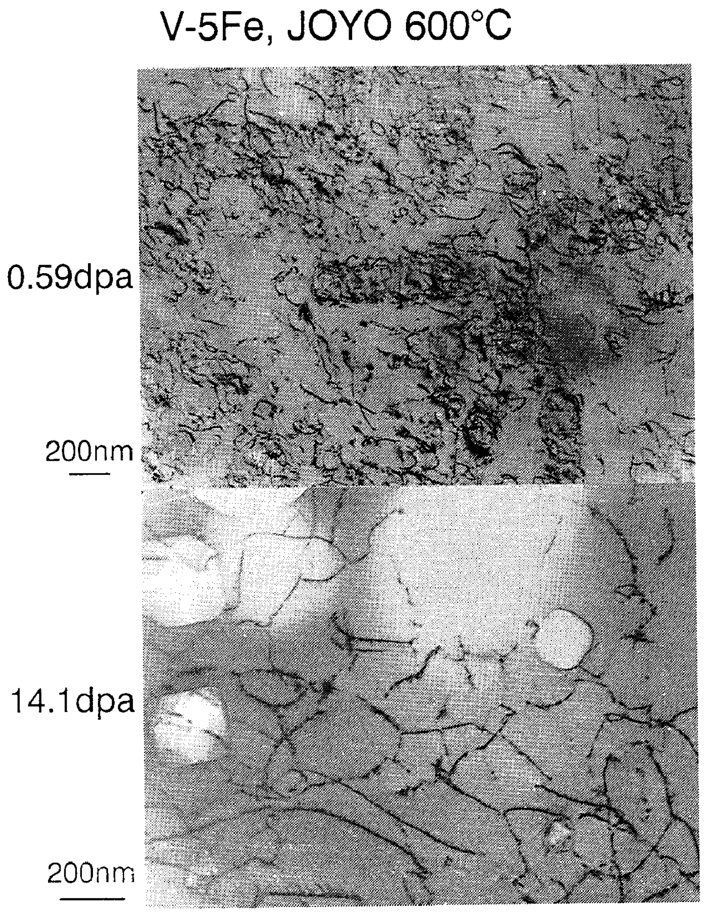

Figure 6 Dislocation images of V-5Fe irradiated at 600°C.

micrograph at the bottom of Fig. 6 is a typical example. Another example is shown in Fig. 7, where pairs of dislocations observed in a specimen irradiated to 0.1dpa at 400°C are shown. The fringe image accompanying the dislocations is of particular interest. The three micrographs in Fig. 7 were taken using different diffraction conditions from the same area. The micrograph at the top was taken using $g=110$, and $g=\bar{1}\bar{1}0$ was used to take the second micrograph. The micrograph at the bottom is a weak-beam dark field micrograph showing the fine structure of the dislocations. The remarkable asymmetry of the dislocation images observed when operating g is reversed suggests that these dislocations are accompanied by thin layer of another phase. By contrast analysis and stereo microscopy, it has been deduced that these dislocations have b=a[100] and the fringed regions between the pair of dislocations correspond to the extra-half plane. At 400°C, a significant fraction of dislocations have such fringe contrast. Unfortunately, no information on the local chemistry is available.

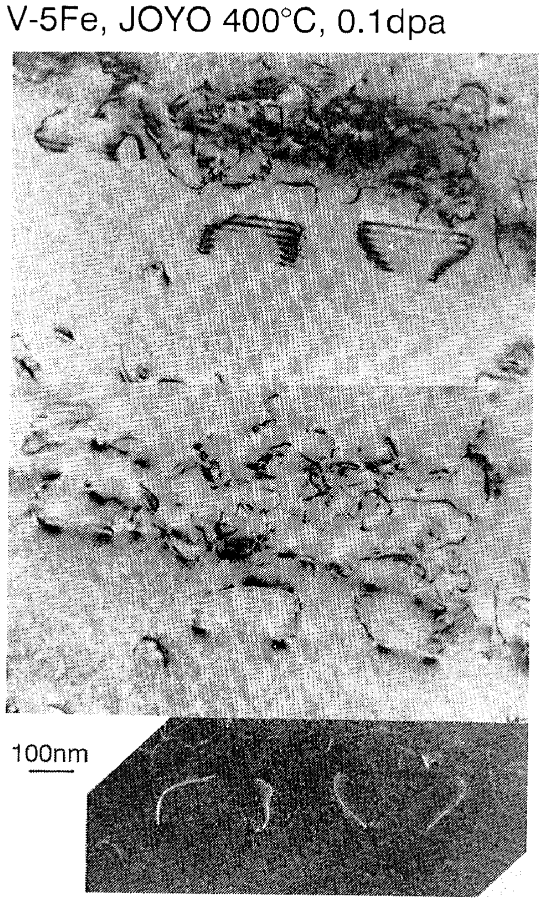

Figure 7 Contrast analysis of dislocation loops with fringe contrast. The image at the bottom is an weak beam dark field micrograph using g=110.

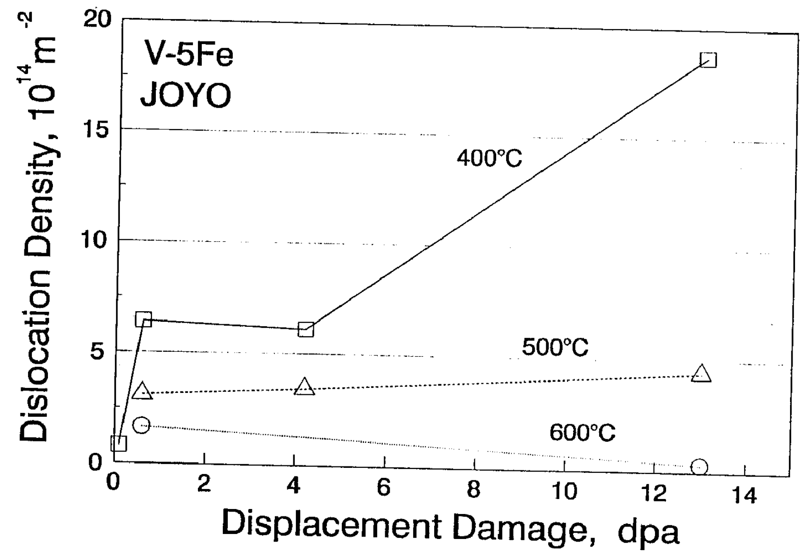

Figure 8 Dislocation density as a function of displacement damage.

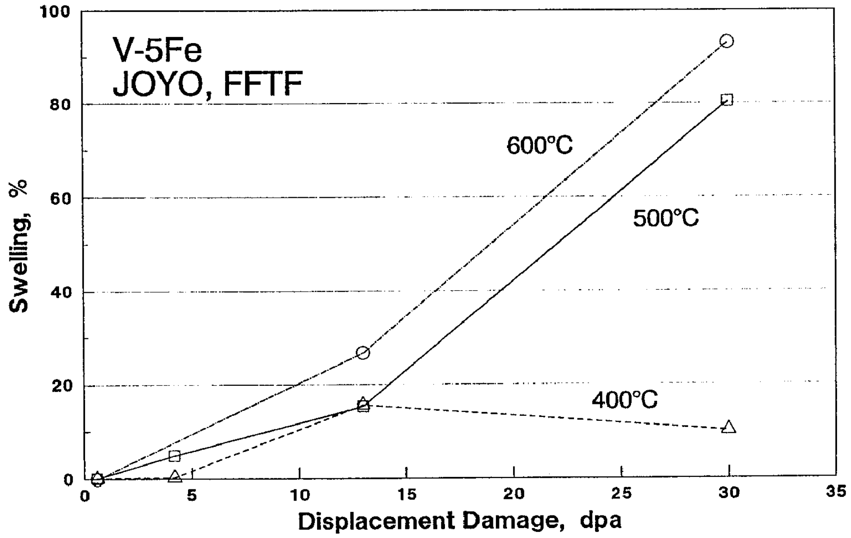

Figure 9 Swelling as a function of damage. Previous data of FFTF irradiation to 30dpa are also included for comparison.

Dislocation density has been measured from micrographs taken with g=110. It is to be noted that for dislocations with b=a/2<111>, one half of the dislocations are out of contrast, and for b=a<100>, one third are out of contrast. By contrast analysis, it has been found that majority of dialocations have b=a<100>. The result of measurement is given in Fig. 8. The rate of accumulation of dislocation is very steep at 400°C, while at 600°C, dislocation density decreases at a high fluence level.

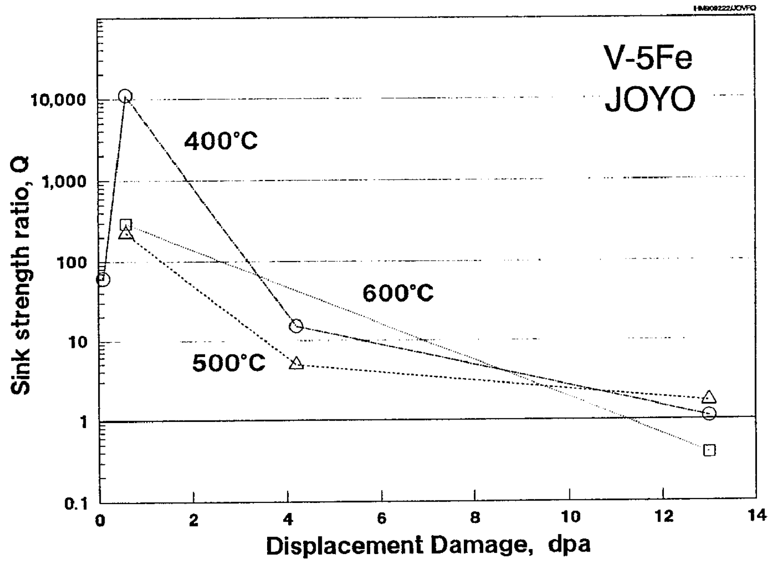

Figure 10 Sink strength ratio Q plotted against displacement damage.

Swelling

Volumetric swelling has been obtained from TEM micrographs as given in Fig. 9. In this figure are also included the swelling data obtained in the previous study using FFTF/MOTA[1]. Note that the swelling values in the previous study were obtained by density measurements, and the values may be under-estimation as will be discussed in the following section.

IV. Discussion

Swelling

Body-centered cubic metals have been claimed to have good swelling resistance in comparison with fcc metals and alloys[8)]. In fact many of the vanadium alloys show less than 1% of swelling at a damage level of 84dpa[9)]. Therefore, the large swelling observed in V-5Fe after FFTF irradiation was first considered to be exceptional[1)]. However, in a recent study[10)] on V-Cr and V-Si, as well as in the study by Loomis et. al[9)], large values of swelling have been also obtained; several tens per cent of swelling have been reported for some vanadium-chromium alloys with damage levels of 77 to 84 dpa at 600°C [9)]. These findings suggest that bcc metals and alloys, including probably ferritic steels, are <u>not</u> inherently immune to swelling, and certain precautions must be followed in order to develop swelling resistant alloys.

In the past, the swelling resistance of bcc alloys have been discussed in terms of several mechanisms, while most of them are focused on ferritic/martensitic steels. Mechanisms which may be relevant to the present study [4,6,7)] are:

(1) Low dislocation bias for interstitials due to their smaller relaxation volume than in fcc structure [6)],

(2) Effect of segregation to void surfaces [11)17)] in changing bias factors.

In order to explain the large swelling observed in V-5Fe alloy, the meaning of the above mechanisms must be reversed. That is, interstitial bias factors in V-5Fe alloy must be enhanced by some mechanism. The enhanced bias may promote both cavity nucleation process and their growth process. The cavity number density at 400°C shown in Fig. 2 is comparable to those of pure vanadium or vanadium based binary alloys showing only modest swelling reported in the literature [12)13)]. The very inhomogeneous cavity formation observed at 500°C as shown in Fig. 3 also suggests that cavity nucleation is not particularly promoted in this alloy. The large swelling observed in V-5Fe must be related to some mechanism causing efficient cavity growth.

Segregation and Swelling Mechanism

The fringe contrast accompanying dislocations as shown in Fig. 7 may either be stacking fault or a thin layer of another phase with different chemistry than the matrix. The stacking fault energy in vanadium is known to be too high to measure, so that it is more likely that the fringe is caused by some chemically different phase. Since iron is undersized and expected to segregate to interstitial sinks[14)], it is probable that the fringe is due to a thinly segregated layer of iron along the extra-half plane of the dislocations. In the absence of any information about the local chemistry, the assumption here is speculative.

The segregation layer observed in Fig. 7 may play a very important role in changing bias factors of dislocations in three ways:

i) The effective Burgers vector may be smaller than that in the bulk if the segregated layer consists of large fraction of under-sized solutes. This effect may reduce the interstitial bias.

ii) Segregation may hinder the climb motion of dislocation by pinning effect. If this occurs in a symmetrical fashion, i.e. climb up and climb down is equally suppressed, this effect will also decrease bias.

iii) Weertman and Green$^{15)}$ have proposed that if a Cottrell cloud is made up of undersized solute, the dislocation line will become an even better sink for interstitials and even worse sink for vacancies.

The first of these three effects tends to suppress swelling. The second effect also tends to reduce bias. If, however, the pinning effect is asymmetrical as has been proposed in the previous paper$^{1)}$, this effect also may enhance swelling. The third mechanism by Weertman and Green appears to be most likely, while as shown in Fig. 7, the segregation is not like a Cottrell atmosphere but a thin layer. It is not clear at the moment if Weertman and Green mechanism is also valid for the layer type segregation. Moreover, as the authors themselves admit, their theory may not be valid because of the Thomson effect$^{16)}$.

A mechanism was also proposed in the previous paper$^{1)}$ where migration energy of a self interstitial atom may become much larger as it approaches the dislocation core. There should be a wide variety of situations depending on the dislocation Burgers vectors, dislocation line direction, size factor of the partner atom of the mixed dumbbells, elastic anisotropy of the host alloy lattice, etc. This effect is very general and should be a subject of intense study in the future.

Segregation of solute atoms to void surface is another possible cause of the swelling enhancement. Effects on the void bias of segregation to void surfaces have been studied by Wolfer and co-workers $^{6)17)18)}$. When voids are covered with a "shell" of solute rich material with a larger shear modulus than the matrix, the bias factor of the voids will change in such a way that interstitials encounter a barrier before jumping into the void. Without knowing the composition of the void shell material, it is difficult to estimate the shear modulus of the shell in the present study. If the shell is iron rich, it is likely that the shell has a larger shear modulus than the matrix, since iron has a greater shear modulus than vanadium. If this is the case, the cause of the large swelling observed in the present study may be explained in terms of the cavity bias factor in favor of absorbing vacancies.

Comparison with FFTF Irradiation

The swelling data obtained in the present study are compared in Fig. 9 with those for FFTF irradiation. Before discussing if there is incubation period, the swelling value obtained in the previous study must be re-examined. The swelling value of FFTF irradiation at 600°C given in Fig. 9 does not differ significantly from the one for 520°C irradiation. To the contrary, the external size of the TEM disks after irradiation is significantly different from each other. Indeed, the swelling obtained from the change of the TEM disk size is more than 160% for 600°C irradiation and is 76% for 500°C irradiation, the latter being in fair agreement with the value obtained by density measurements. A possible reason for the large discrepancy for 600°C data lies in the void microstructure at the specimen surface. For 600°C irradiation, the voids have grown to such an extent that they are interconnected to become continuous channels. During the density measurement, it was experienced that the measured density value increased during repeated measurements. It is quite likely that the measuring liquid wets the specimen surface and even penetrate into the specimen giving apparently higher density value. Thus, the FFTF point of 600°C must be located much higher than it is plotted here.

The swelling rate for 600°C irradiation in JOYO is more than 2%/dpa. One might feel hesitation to call this high rate to be in the incubation period. In Fig. 10 is plotted the sink strength ratio Q[19)] as a function of displacement damage. Here, Q is simply given as dislocation density divided by 4π times cavity number density times their radius, and no sink efficiency factor has been incorporated. In all cases of JOYO irradiation, Q is much greater than unity, which reflects the high dislocation density observed. The Q value approaches unity when the damage level becomes greater than about 10dpa. Usually, the steady state swelling is characterized by the Q value close to unity. The behavior of Q in this study indicates that the swelling is still not in the steady state region below 10dpa, where the swelling rate is as high as 2%/dpa. The possible mechanism listed in the preceding section should be examined quantitatively if they can reproduce this high swelling rate under the condition of Q value much larger than unity, and this will be a subject of future studies.

The Q value for 600°C irradiation becomes less than unity at the highest dpa level studied (Fig. 10). This implies that the swelling curve as a function of dpa should tend to saturate, while if the data point of FFTF is to be connected with JOYO data, the curve becomes even steeper. It is likely that the swelling curves may be different in the two irradiation conditions. There are differences in e.g. dpa rate, atmosphere, and possibly, the irradiation temperature and its history. The irradiation in FFTF was done in purified lithium, while in JOYO, in purified helium. In the former, oxygen concentration may be reduced during irradiation while nitrogen may be supplied from lithium. The thermal contact in a helium atmosphere may be worse than in lithium. All of these differences may combine to give rather unpredictable difference in the swelling behavior. Nevertheless, it is appropriate to conclude that basically the same phenomenon of the large swelling occurs in both of the irradiation fields.

V. Conclusions

(1) A V-5at.%Fe alloy shows large swelling after JOYO irradiation to approx. 14dpa at 500°C and 600°C, in agreement with FFTF irradiation.

(2) Several possible mechanisms involving segregation effects have been discussed.

(3) Any successful model should be capable of predicting the observed swelling rate more than 2%/dpa under condition of Q value much greater than unity.

Acknowledgement

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