ASSESSMENT OF RECENT VANADIUM ALLOY IRRADIATION EXPERIMENTS - D.L. Smith (Argonne National Laboratory)

OBJECTIVE

The objective of this task is to review and assess the status of the database obtained from recent vanadium alloy irradiation experiments.

SUMARY

A brief assessment of the results of irradiation tests of vanadium alloys conducted in the HFIR-11J, -12J, 13J, and 10J, indicate that significant uncertainties in the data exist which affect the reliability of the data. This assessment concludes that vanadium specimens from the 12J experiment were severely oxidized, and that the irradiation data are unreliable. There is a significant uncertainty in the irradiation temperature for specimens in the 11J, 12J and 13J experiment due partially to gap conductance in the experiment. The very high irradiation creep rates obtained for the vanadium alloys in the 12J experiment are attributed to a large thermal creep component. A detailed thermal analysis of each specimen is required to provide a reliable temperature specification for the data obtained in these experiments. Similar temperature uncertainties may also exist for other materials irradiated in similar test assemblies.

PROGRESS AND STATUS

Background

The purpose of this document is to provide a brief assessment of the status and results obtained from recent irradiation experiments on vanadium alloys in order to provide a basis for developing priorities for future irradiation testing of vanadium alloys. Early irradiation testing of vanadium alloys conducted primarily in FFTF focused on higher damage levels and temperatures projected for fusion power applications, viz., 15 - 100 dpa and 400 - 600°C. These experiments, which demonstrated that vanadium alloys containing titanium were highly resistant to irradiation-induced swelling and alloys with less than ~ 10% (Cr + Ti) exhibited favorable mechanical properties after irradiation, provided the basis for proposing V-4Cr-4Ti as a reference composition for Li-cooled fusion power systems. In the early phases of ITER, the possibility of using the vanadium/lithium system was also explored, leading to the conduct of low temperature irradiation experiments (T < 400°C) to identify candidate compositions for this application. Since the ITER application was rejected, the primary interest for vanadium alloys is for high performance lithium-cooled systems at higher temperatures (400 - 750°C). The primary uncertainty for this application involves the effects of fusion-relevant helium generation rates with displacement damage on the properties of vanadium alloys.

This assessment is limited to the irradiation experiments on vanadium alloys that have been conducted during the last five years at temperatures in the range of primary interest for fusion power applications, viz., > 400°C. Primary issues associated with these recent irradiation experiments relate to oxygen contamination of the specimens during irradiation, uncertainties in the specimen temperatures during irradiation, and limited damage levels attained. Severe oxidation of vanadium alloy specimens was observed in the HFIR-12J experiment, thermal analyses indicate that the specimen temperatures exceeded the reported temperatures in HFIR 11J, 12J and 13J, and the displacement damage in HFIR-10 J was only a few tenths of a dpa, since the experiment was terminated prematurely.

Vanadium irradiation experiments

HFIR-12J experiment

The HFIR-12J experiment was inserted into the HFIR on February 7, 1997 and was removed on July 17, 1998 for a total of 224 days of irradiation, which corresponds to a damage level of ~7 dpa. The specimen temperature for this experiment was reported to be ~ 500°C. This experiment contained irradiation creep specimens in addition to tensile and TEM specimens. In reviewing various aspects of this experiment, there are two issues which indicate that much of the data obtained from this experiment is not reliable. The two issues relate to severe oxidation of the vanadium alloy specimens exposed to the helium purge gas during irradiation and uncertainties in the actual specimen temperature during the irradiation. Both issues can have a significant effect on the results. In this experiment, the tensile specimens were exposed to low-pressure helium gas during the irradiation. The creep specimens and the TEM specimens were contained in sealed capsules.

Vanadium alloy tensile specimens examined during PIE indicated severe oxidation of the alloy. The surfaces of specimens from this irradiation were covered with a heavy black scale that appears to be similar to the VO_2 oxide observed on vanadium alloy specimens obtained from oxidation studies in high purity helium, which contained low oxygen concentrations. Although the oxygen content of the helium purge in this experiment was not measured, it is expected to be similar to the oxygen levels used for some of the oxidation experiments. A few parts-per-million of oxygen in helium is sufficient to oxidize the vanadium alloys. Although the tensile specimens were wrapped with zirconium foil, the gettering capacity for this material is limited at ~500°C. Based on thermodynamic and kinetic data on oxygen interactions with vanadium and vanadium alloys, large amounts of oxygen are predicted to diffuse into the thin (typically 0.25 mm thick) tensile specimens during the relatively long irradiation time (~ 5000 hr). The observations from this experiment certainly confirm this. A major fraction of the reduction in ductility observed in these specimens after irradiation in the HFIR-12J experiment can be attributed to the oxidation rather than the irradiation.

There also appears to be a substantial uncertainty in the temperature of the vanadium alloy specimens in this experiment. This is particularly important since the temperature uncertainty would also exist in other similarly designed experiments. The primary source of the temperature uncertainty relates to the temperature rise across multiple gas gaps in the experiment design. This explains the unexpectedly high radiation creep measurements reported for the specimens irradiated in this experiment. In this case there is a significant gas gap between the pressurized creep tube (PCT) specimen and the zirconium capsule. This gap was necessary to avoid any constraint in the creep of the specimen, but the gap is sufficient to contribute a significant temperature increase in the specimen. In addition, differential thermal expansion between the zirconium capsule and the aluminum holder plus the required assembly tolerances creates a second gap that would provide additional temperature rise. A thermal analysis based on the specified dimensions indicates that the temperatures of the PCT's were significantly higher than the design reported temperature of 500°C. Most of the measured creep in this experiment can be attributed to thermal creep and not to irradiation creep. This conclusion regarding temperature uncertainties is further supported by the recent results obtained from the flux monitors reported by L. Greenwood [1]. Although the explanation provided by Greenwood for the mechanism of the melting process is questionable since some melting would provide a good contact and thus lower the temperature, the flux monitors were obviously at a significantly higher temperature than the design temperature. This can be partially attributed to the gap outside the flux monitor capsule. A similar conclusion is reached for the vanadium tensile specimens in this experiment. These specimens also involved a double gap design with the zirconium foil and the differential expansion with the aluminum capsule. The tensile specimens were not only severely oxidized, but the actual temperature is uncertain. This temperature uncertainty also exists for the steel specimens irradiated in this experiment, although to a lessor degree since they were not wrapped

with zirconium. As a result of these considerations, the results obtained on the mechanical properties of vanadium alloys irradiated in this experiment are not considered reliable.

HFIR-11J experiment

The HFIR-11J experiment was conducted simultaneously with the HFIR-12J experiment, except the reported specimen temperatures for the HFIR-11J experiment are ~300°C. This temperature is below the range of primary interest for fusion power applications. However, similar to the case for the HFIR-12J experiment discussed above, the temperatures are uncertain but appear to be significantly above the reported temperatures. The oxidation problem associated with the HFIR-12J experiment should not be severe at the temperatures of the HFIR-11J experiment. Consistent with the oxidation experiments, specimens from this experiment did not exhibit the heavy surface oxide that was observed on the HFIR-12J specimens.

HFIR-10J experiment

The vanadium alloy specimens from the HFIR-10J experiment were contained in lithium- bonded capsules, which eliminates any oxidation problem. The measured specimen temperatures for this experiment should also be more reliable. However, the damage level achieved was extremely low because of leaks encountered in the gas system during irradiation. Since details of the second insertion of this experiment have not been reported in the semiannual reports, exact values cannot be given. However, since the damage levels are so low, results from this experiment are of limited value.

HFIR-13J experiment

The HFIR-13J experiment was inserted into the reactor on July 22, 1998 and was removed on May 16, 1999 after an irradiation of 195 effective full power days, which corresponds to a predicted damage level of ~ 5.4 dpa for vanadium specimens. In this experiment the regions of the vanadium alloy test specimens were sealed with static helium so oxidation, even at the higher temperatures of this experiment, should not be an issue. We have not observed the specimens to verify this conclusion. However, as in the previous experiments, there is a significant uncertainty in the temperature of the specimens during irradiation. The design temperatures for the HFIR-13J experiment ranged from 200 - 500°C. However, as indicated in reference [2], the thermocouple temperatures in many cases varied from the design temperatures by as much as 50°C. The temperatures also varied significantly from cycle-to-cycle because of the rotation of the subassembly, which was rotated 180 degrees each cycle to obtain a more uniform average damage level. This capsule also contained multiple gas gaps between the specimens and the bulk specimen holder where the temperature was measured. In addition, tensile specimens were contained in stacks of ~ 15 which were loosely held in the specimen holder. Significant temperature gradients are predicted through the stack of loosely packed specimens, which would result in significantly higher temperatures for specimens located in the center of the stacks. For the HFIR-13J irradiation, the measured temperatures were generally higher than the design temperatures, significant variations in the recorded temperatures were obtained during the irradiation, and significant temperature differences between the actual specimen temperatures and the measured temperatures are predicted because of gaps/contact resistances. This leads to a conclusion that there are significant uncertainties in the actual specimen temperatures for this irradiation experiment. For any case where the effects of irradiation are sensitive to the irradiation temperature, a careful review of the thermal history of the test specimens is required to provide a reliable evaluation of the data.

CONCLUSIONS

A review of the experimental conditions for vanadium alloy irradiations in three of the four recent HFIR irradiation experiments (11J, 12J and 13J) indicate that much of the reported mechanical property data, particularly tensile and irradiation creep data, are questionable. Vanadium alloys

irradiated in HFIR-12J were severely oxidized, which would have a major impact on the properties of the thin specimens. The effects on the thicker specimens such as Charpy specimens and bend bars is less certain. Actual temperatures of the vanadium specimens in all three experiments are subject to significant uncertainties produced by multiple gap thermal-conductance considerations. This temperature uncertainty also extends to other materials irradiated in the same experiments. The damage level accrued in the fourth experiment, HFIR-10J, was so low as to be of limited value. A detailed evaluation of these problems should be conducted before additional irradiation experiments using similar capsule designs are initiated.

REFERENCES

1. L.R. Greenwood and C.A. Baldwin, Fusion Materials Semiannual report DOE/ER-0313/30, p. 214, October 2001.
2. A.L. Qualls and T. Muroga, Fusion Materials Semiannual report DOE/ER-0313/28, p. 266, September 2000.