CORRECTIONS AND CLARIFICATIONS ON THE EVALUATION OF THE DHCE EXPERIMENT - D. L. Smith (Argonne National Laboratory)

OBJECTIVE

The objective of this task is to provide a viable simulation for investigating the effects of neutron irradiation damage with fusion-relevant helium generation rates on the properties of vanadium-base alloys.

SUMMARY

A critical issue in the development of structural materials for the fusion application involves the effects of high helium generation rates on the performance limits of neutron irradiated materials. Since we do not have a high flux neutron source with fusion-relevant energies, we must rely on simulation techniques to obtain experimental information on these effects. The Dynamic Helium Charging Experiment (DHCE) provides a unique approach for simulating the helium production rates in vanadium alloys in fission reactor irradiations. An assessment of the DHCE-1 proof-of-principle experiment and a subsequent evaluation of the DHCE concept have been presented in the last two semiannual reports. This report attempt to correct and clarify several misinterpretations, incorrect statements and misleading conclusions from the evaluation, which contributed to the decision not to conduct a second DHCE experiment in the early phases of the JUPITER-II collaboration. Specific responses to statements and conclusions presented in the evaluation are presented in this report.

PROGRESS AND STATUS

Background

The Dynamic Helium Charging Experiment (DHCE) was developed to simulate the effects of fusion-relevant helium generation rates in vanadium alloys on the properties of neutron irradiated alloys during fission-reactor irradiations [1]. A proof-of-principle experiment (DHCE-1) was conducted in the FFTF reactor and the results reported in several publications (see ICFRM conference proceedings 1996-date). A detailed assessment of the DHCE concept was distributed in June 1999 and updated condensed versions were reported in the Fusion Materials Semiannual report [2] and presented as a selected oral presentation at the ICFRM-10 in Baden Baden, Germany in October 2001. The results of these assessments suggest that the DHCE provides a viable simulation for simultaneously producing fusion-relevant helium generation rates during fission-reactor irradiations of vanadium-base alloys.

An evaluation of the results from the first Dynamic Helium Charging Experiment (DHCE-1) and an assessment of the readiness to conduct a future DHCE (referred to as the evaluation) was also conducted by L.R. Greenwood, D.L. Baldwin, G.W. Hollenberg, and R.J. Kurtz and reported in the Fusion Materials Semiannual report [3]. This evaluation contains several misinterpretations, incorrect statements, and misleading conclusions, which contributed to the decision not to conduct a second DHCE experiment in the early phases of the JUPITER-II collaboration. The purpose of this report is to correct and clarify the major inaccuracies presented in the evaluation by Greenwood, et al., in order to provide a technically correct assessment of the status of the DHCE technique. For convenience, the specific issues are presented below in the order they are contained in the evaluation report and are not necessarily in the order of importance.

Unfortunately, we did not take advantage of an opportunity to conduct a DHCE as part of the JUPITER-II collaboration. A future experiment of this type will be much more expensive since the ANL program on vanadium alloy research has been terminated and the unique facilities developed for disassembly of the tritium-containing capsules will be dismantled. Replacement of

the ANL facilities used for tritium charging and capsule disassembly for the DHCE-1 would cost $500-1000 K, which now will make a DHCE-type experiment significantly more expensive.

Corrections and Clarifications on the DHCE Evaluation

Post test prediction of helium levels

Based on results from analyses of specimens from two of the seven test capsules (4D1 and 4D2) in DHCE-1, the evaluation states [3](pg. 16, par 2) that "up to 35% of the ^{3}He in the samples in these positions may have been generated from tritium decay after the irradiation". This statement is highly misleading and inaccurate. The evaluation fails to point out that analyses obtained from the other five capsules indicate that much smaller fractions of the ^{3}He was generated after termination of the irradiation. For example, specimens from the other capsule (5E2) irradiated at the same temperature as the two capsules mentioned above actually indicated more tritium in the later analysis than in the earlier analysis, which would indicate a negative contribution to the ^{3}He content. This certainly cannot be attributed to tritium decay. Specimens from the 500°C capsules indicated only a variation of 3.8 and 3.9% increase in the ^{3}He content. Specimens from one of the 600°C capsules indicated an additional contribution to the ^{3}He of only ~10%; however, this was based on a very high analysis, most likely due to a mix-up of specimens as discussed in previous assessments [2]. Results from the other 600°C capsule indicated so much variation in the helium analyses that a meaningful correlation is not possible. Helium measurements on this specimen were made at three different times. Analyses of two of these time periods indicate a large depletion of ^{3}He which would indicate a large negative effect (impossible), while analysis of the third condition would indicate an unreasonably large (~180 %) increase in ^{3}He. The average calculated tritium from this capsule was listed as 20 ± 66 appm, which is not very meaningful. The only plausible explanation for these results is an anomaly in the chemical analyses since they were all conducted on samples from the same specimen. The contribution of ^{3}He after termination of the irradiation for the 4D1 and 4D2 capsules are also significantly lower than the values reported in the evaluation, ~23% for 4D2. One might also point out that the differences in the average analyses of the ^{3}He for the different times is only ~1.4% which is quite small. In any case, the results indicate that the contribution to the ^{3}He after termination of the irradiation is very small in most of the specimens analyzed. Based on calculations of the hydrogen distribution in the V/Li system using recent data, one also predicts very low contributions to the ^{3}He concentration after termination of the irradiation. It is grossly misleading to convey that the ^{3}He generated after termination of the irradiation is as high as stated in the evaluation.

The evaluation also states that "most of the samples were degassed sometime in 1994 prior to the ^{3}He measurements in 1995". All specimens included in the analysis discussed above were not degassed as indicated by the specimen ID numbers, which contain "ND" in the ID. Furthermore, the He and tritium analyses relevant to this discussion were analyzed during the period June-August, 1994, and not 1995, and mechanical properties data on the V-4Cr-4Ti alloys from this experiment were conducted and reported in 1994.

The high uncertainty in the ^{3}He generation after termination of the irradiation, which is stated in the review, is very misleading.

Tritium trapping and correlation with stainless steel data

The suggestion that the high ^{3}He content of the V-1Si and V-5Fe specimens is possibly due to tritium trapping similar to that observed in irradiated stainless steels (pg 18, par 2) is not well founded. Extensive experience and experimental results indicate that this is not the case. There are major differences in the solubility and mobility of hydrogen in vanadium and hydrogen in steel. One only needs to compare the vast differences associated with hydrogen or tritium charging and hydrogen permeation results for the two metal systems to conclude that hydrogen trapping is very different. Several simple examples follow. Hydrogen solubility measurements for vanadium (and

alloys) are difficult because the hydrogen migrates into or out of the alloy so fast that quenching in the hydrogen is almost impossible (see H-solubility data). This also shows up in the hydrogen adsorption/desorption experiments. In the early irradiation experiments, specimens were normally degassed to assure that hydrogen did not affect the measurements of the radiation effects. Experience showed that degassing of irradiated specimens was typically easier than non-irradiated specimens. This was not attributed to the irradiation effects, but demonstrates that tritium trapping has not been observed in a large number of alloy compositions tested. Results from tests in sodium also confirm this conclusion. In contrast, it is well documented that hydrogen trapping can occur in steels and that hydrogen charging is very difficult. Correlations of hydrogen effects observed in steels with those in vanadium are not supported by extensive data.

High helium generation in some alloys

The suggested correlation of tritium trapping with the high helium content (pg 20, par 1) does not hold up even for the V-5Cr alloy discussed. Although Nakajima's data for V-5Cr might suggest this, other data on V-5Cr, including Garner, et al. in same proceedings, indicate high swelling at both 500 and 600°C. Swelling data for V-5Fe also do not correlate with the helium concentrations. Very high swelling of the V-Fe alloys are observed both with and without helium. In some cases the swelling is lower in this alloy with higher helium concentrations. As discussed in the previous paragraph, a substantial database indicates that significant trapping of tritium in vanadium alloys does not occur at the temperatures of interest.

Tritium permeation losses through TZM capsules

The suggestion that tritium leakage from the TZM capsules at 600°C appears to be lower than the ideal leakage rate based on pure molybdenum data (pg 20, par 2) is consistent with the previous assessments provided for the DHCE. Recent data presented in the assessments indicate that tritium permeation through TZM is significantly lower than that for pure molybdenum. It has also been suggested that some surface contamination, e.g., oxide, on the capsule walls may also reduce the tritium permeation rates. Calculated values for both Mo and TZM have been presented previously [2].

Consideration of hydrogen solubility limits in lithium.

The evaluation states that when the original DHCE calculations were performed, the solubility limits for H in lithium were not considered (pg 20, par 3). This is not the case. Based on the predicted hydrogen distribution coefficients at that time, the predicted tritium concentrations attainable in the vanadium at ~425°C was close to the level needed for the desired He generation rate in vanadium. The original distribution coefficients were biased because of experimental measurements, which were based on measured H concentrations in V at levels at or below the limits of the analysis.

Uncertainties in the solubility of H in Li

The evaluation concludes that there are still relatively large uncertainties in the data for the solubility of H in lithium and the LiH equilibrium (pg 21, par 2). This is true in the original calculations for the DHCE and in the references cited in the evaluation. However, these uncertainties have been largely resolved by the references we have used in the recent assessment. Veleckis, et al. [4] have published in 1986 a summary report that includes an updated evaluation of the hydrogen solubility data. This appears to be the most reliable assessment of all of the relevant data and is recommended. This assessment has significantly reduced the uncertainties in the hydrogen solubility in lithium. The points made in the evaluation regarding the equilibrium related to the saturation pressure of hydrogen in lithium, and hence in the capsule, are completely correct. An earlier concern expressed by others suggesting that the formation of LiT could lead to higher tritium partial pressures is not valid.

Limits of He generation in a future experiment

Values presented in the evaluation of the He generation limits (pg 23, par 1 and Table 2) are underestimated since the most recent hydrogen solubility data were not used. The proposed temperatures for the next vanadium alloy irradiation test have been either 450/600/750°C or 500/600/700°C. Calculations based on the recent data indicate a maximum He generation rate of 22 and 45 appm ^{3}He/yr in V4Cr4Ti in tritium saturated lithium at 450 and 500°C, respectively. This corresponds to about half the desired rate at 450°C and approximately the desired rate at 500°C, assuming a damage rate of ~12 dpa/fpy, which is typical of the ATR peak damage rate. Although the spectrum in a fusion device varies some with the blanket composition, the more recent neutronic analyses indicate a He/dpa rate for V in a Li/V blanket is ~4.5 appm He/dpa. The ORNL data indicate a value of 4 appm/He/dpa for V-15Cr-5Ti; however, I do not know what blanket system they assumed. Since we have carried V-5Ti alloys in most irradiation tests, these alloys, which have a higher tritium solubility, would generate somewhat higher helium concentrations in the same lithium/tritium exposure. Certain other alloys exhibit even higher H solubilities and would provide higher He concentrations. Since our primary objective is to develop an improved understanding of He effects in irradiated materials, a judicious selection of vanadium alloys would provide range of He concentrations for investigation. At temperatures above 500°C, He concentrations higher than values corresponding to projected He/dpa ratios for vanadium alloys in a fusion spectrum can be obtained in most alloys.

Leakage of tritium from capsules at 600°C and higher

As stated in the fourth item above, recent data that we referenced in the recent assessment [2] indicate that the hydrogen permeation rate through TZM is lower than that of pure vanadium. We also suggest that surface films may also contribute to lower tritium permeation rates. However, at higher temperatures of interest, viz., ~750°C, tritium permeation losses would be excessive for a single-walled capsule. The proposed solution is the use of a double-walled capsule, with the outer capsule at a temperature low enough (~ 400°C) to contain the tritium. This appears to be a straightforward solution since one also needs a significant thermal gap to attain the higher test temperatures in water-cooled reactors. One would not need the high ^{6}Li suggested in the evaluation in this case (pg 24, par 1 and Table 2) and the ^{4}He generation rate would be much smaller and not a serious problem.

Issue of testing alloys that produce high He concentrations

The comment is made that the future DHCE experiment is more complicated since separate capsules would be required to test the alloys that generate higher helium concentrations (pg 24, par 3). This does not appear to be any more complicated than the requirement to have different capsules to test at different temperatures. In the DHCE-I we used multiple capsules at each temperature, typically with different tritium concentrations in order to investigate the effects of different He concentrations. In this case one only needs to decide on priorities for the test as all conditions, whether they be temperature, fluence, alloy composition or helium content, cannot be accommodated in a single experiment. The current objective of the program is to develop an improved understanding of helium effects on irradiated alloys. It is not clear that one must accommodate all alloys of interest in this particular part of an experiment. It might be more important to emphasize different variables in one or only a few alloys to provide a better understanding of helium effects..

Correlation of post-test tritium measurements in test capsules

A question is raised regarding the poor correlation of the post-test tritium measurement obtained during opening of the capsules with the amount of tritium in the precharge (pg 25, par 2). It should be pointed out that only the tritium released in the gas phase during specimen retrieval was measured. It is certainly assumed, and partially indicated that a significant fraction of the tritium in the capsule remained in the condensed phases after dissolution of the lithium to remove

the specimens. As indicated in the report on the specimen retrieval, the ratio of tritium measured in the off-gas to the tritium precharge was relatively constant for all capsules, typically 40 - 60 %. The variations observed appear to be fairly consistent with the inconsistencies in the He measurements, which have been attributed partially to observed tritium losses during capsule filling and welding. This conclusion is considered a much more likely explanation than the suggestion that tritium leaked from the 430°C capsule. The alternate explanation suggested in the evaluation relates to He produced in the specimens after termination of the irradiation. As stated above, this is not a likely explanation and is not consistent with results from the other capsules.

Tritium transport in Na or NaK which have low solubilities

The evaluation states that it is essential that the Sieverts' constant for tritium solubility in the liquid metal must exceed that of vanadium (pg 26, par 1). This is completely incorrect. Extensive data exist that demonstrate that mass transport of trace elements in liquid metals occurs even though the solubility of these trace elements have a very low solubility in the liquid metal. Extensive experience in the sodium systems shows that hydrogen transport occurs effectively in sodium at much lower concentrations than are proposed here. Even carbon transfer in sodium/steel systems, which has been the subject of extensive investigations because of its importance, readily occurs even though the solubility of carbon in sodium is orders of magnitude lower than even that of hydrogen. Tritium transport from a mother alloy in the capsule through the sodium to the test specimens is not a constraint for the conditions of interest. The primary reason this alternative has not been recommended, even though we suggested it as a possibility, relates to the redistribution of tritium at the end of the irradiation. In this case, significant helium would be generated in the vanadium after termination of the irradiation unless the specimens were removed and degassed in a short period of time. The comment on this issue in the evaluation is not correct. A concern is also raised that the He/dpa ratio for this case would decline during the irradiation. This is true but the magnitude is very small since the tritium decay is only ~5%/yr. This is not considered to be a significant issue.

Required work to conduct a future DHCE experiment

The evaluation concludes that although a future DHCE experiment is feasible, a great deal more effort is required to design such an experiment (pg 27, par 5). I believe all of the critical technical issues associated with a DHCE experiment in the ATR reactor have been resolved to the extent that an effective irradiation experiment could be conducted on the most critical issue associated with the viability of structural materials for fusion applications. A detailed design is still required, but the critical issues have been addressed. The most critical issue relates to the burnout of ^{3}He from thermal neutrons. This requires a highly effective thermal neutron shield and a reactor with a higher fast/thermal neutron ratio is much preferred. This is the main reason that use of the HFIR reactor for a DHCE experiment is questionable. If alloys with high hydrogen solubility such as those that contain yttrium are to be included in the experiment, they should be contained in separate capsules.

CONCLUSIONS

The greatest uncertainty related to the performance limitations of candidate structural materials in a fusion environment involves the effects of the high-helium generation rate characteristic of a fusion neutron spectrum on the properties of neutron-irradiated materials. The Dynamic Helium Charging Experiment (DHCE) with vanadium-base alloys provides the most viable simulation for investigating these effects without a high-flux, high-energy neutron source. A detailed review of the proof-of-principle DHCE-1 indicates that, with lessons learned from DHCE-1, this approach provides a validated method for obtaining essentially constant values of He/dpa ratios in vanadium alloys during fission-reactor irradiations for a wide range of fusion-relevant parameters.

REFERENCES

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