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SWELLING IN SIMPLE FERRITIC ALLOYS
IRRADIATED TO HIGH FLUENCE

CONTRIBUTION TO ADIP REPORT

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7.4 SWELLING IN SIMPLE FERRITIC ALLOYS IRRADIATED TO HIGH FLUENCE - D. S. Gelles and R. L. Meinecke (Westinghouse Hanford Company)

7.4.1 ADIP Task

The Department of Energy (DOE)/Office of Fusion Energy (OFE) has cited the need to investigate ferritic alloys under the ADIP program task, Ferritic Steels Development (Path E). The task involved is akin to task number 1.C.2, Microstructures and Swelling in Austenitic Alloys.

7.4.2 Objective

The objective of this work is to provide guidance on the applicability of martensitic stainless steels for fusion reactor structural components.

7.4.3 Summary

A series of Fe-Cr-C-Mo simple alloys has been measured for density change as a function of irradiation in EBR-II over the temperature range 400 to 650°C to fluences as high as 2.13×10^{23} n/cm² ($E > 0.1$ MeV) or 105 dpa. The highest swelling was found in a Fe-12Cr binary alloy, 4.72 percent, after 1.87×10^{23} n/cm² or 95 dpa at 425°C, which corresponds to a swelling rate of 0.06%/dpa. This peak swelling rate value can be used to define swelling predictions for commercial ferritic alloys to 40 MWy/m².

7.4.4 Progress and Status

7.4.4.1 Introduction

Ferritic and martensitic alloys appear to be very attractive for fusion reactor structural applications due to their high swelling resistance.¹⁻³ A primary objective of the ADIP program is to develop materials capable of operating in a fusion reactor up to a time-integrated neutron exposure of 40 MWy/m² or approximately 500 dpa. This corresponds to a fast reactor fluence on the order of 10^{24} n/cm² (all fluences given as $E > 0.1$ MeV). Irradiation experiments to neutron exposures of this magnitude require excessively long times and so procedures must be found which will allow extrapolation of materials properties beyond the available exposure range. An example can be provided by swelling measurements on martensitic alloys. The highest exposure thus far achieved is 125 dpa.¹ This exposure required 9 years of irradiation in EBR-II and the experiment has now been stopped. To reach goal fluence, an additional 36 years of irradiation time would have been required. Therefore, demonstration that ferritic and martensitic alloys will remain swelling resistant to a goal exposure of 40 MWy/m² will not be possible for some time to come. Extrapolation of available data must be considered.

Swelling as a function of fluence is generally modeled by a bilinear equation. The necessary parameters are R, the steady state swelling rate, τ , the fluence corresponding to the intersection of the linear extrapolation of steady state swelling with the zero swelling axis and α , a measure of the curvature of the swelling curve in the vicinity of τ . Modeling swelling in ferritic or martensitic alloys is difficult because a value for steady state swelling has not been established.

The present effort is intended to establish a steady state swelling estimate for ferritic and martensitic alloys. A series of simple ferritic alloys based on the martensitic stainless steel base composition Fe-12Cr-1Mo-0.1C and including binary Fe-Cr alloys covering the range 3 to 18 Cr has been irradiated as part of the National Cladding/Duct (NCD) Materials Development Program for LMFBR development. These specimens are no longer of interest to the NCD program so they have been made available to the MFE program. This report describes density change measurements on several of these specimens as a function of irradiation in EBR-II to fluences as high as 2.0×10^{23} n/cm².

7.4.4.2 Experimental Procedures

Specimens of the "simple ferritic alloy series" were included as ballast in very limited numbers to fill unused space in each of four capsules which were irradiated side-by-side in EBR-II. Details of the alloy series prior to irradiation and following irradiation to 4.3×10^{22} n/cm² and 1.38×10^{23} n/cm² have been reported previously.^{4,5} Each reactor discharge including the fluence obtained for the present study involved a separate set of specimens. In general, only one TEM dish for each condition of fluence and temperature was available and density measurements included at least three repeated determinations for each specimen. Following density measurements, the specimens irradiated to the highest fluence were reinserted into EBR-II in order to provide results to still higher fluences.

7.4.4.3 Results

It has already been noted⁵ and must be reemphasized here, that there is now reason to believe that the Fe-3Cr binary alloy contained a significant amount of impurities. A chemistry overcheck on this material indicates a comparison of Fe-3.27Cr-0.50Ni-0.005P-0.03Mn-0.0005C-0.00013N-0.0147O with no analysis for silicon. Also, precipitation following irradiation to 4.3×10^{23} n/cm² was characteristic of G-phase. Fig. 7.4.1 is provided to further emphasize this point. It shows the preirradiation density found for the Fe-Cr binary alloys (and the value for pure iron⁶) as a function of chromium content.

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Fig. 7.4.1. Density for Fe-Cr binary alloys as a function of chromium content. The value for pure iron is from reference (6).

The Fe-3Cr specimen does not fit the trend, being 0.05 gm/cm³ too dense. The results for this material should therefore be interpreted as an example of a more complicated alloy in the Fe-3Cr composition range.

Table 7.4.1 is provided to summarize all results of swelling and density change measurements on the simple ferritic alloy series. This includes preirradiation density values, estimates for void swelling at low fluence based on transmission electron microscopy (TEM),⁴ results of density measurements at intermediate fluences⁵ and the results of the present effort. Examination of the information in this table reveals that the simple ferritic alloys remain swelling resistant even at fluences on the order of 2.0×10^{23} n/cm². The highest swelling observed was 4.72% for Fe-12Cr at 425°C for a fluence of 1.9×10^{23} n/cm². Somewhat lower swelling was found for Fe-6Cr and 9Cr at 425°C for this fluence level. The peak swelling temperature is in the vicinity of 425°C, and is a slight function of chromium content; higher chromium contents result in slightly higher peak swelling temperature. Additions of carbon and molybdenum (and in the case of Fe-3Cr, various impurity additions) have the effect of reducing the peak swelling temperature to 400°C or below. Finally, examples can be found where swelling saturation may have occurred. See for example, Fe-3Cr at 400°C, Fe-6Cr at 400°C, and Fe-12Cr-0.1C-2Mo at 400°C.

Table 7.4.1. Results of swelling measurements of simple ferritic alloy specimens contained in the AA-VII test^{4,5}

Alloy	I.D. Code	Cold Density	Temp.°C: Flu.°C:	400 3.4	400 10.3	400 16.3	425 4.3	425 18.7	450 2.8	450 10.7	450 11.5	482 ^b 11.8	510 20.4	540 20.0	650 21.3
Fe-3Cr	65	7.8826	---		1.76	0.70	0.09 ^b	1.65	0.0 ^b	0.62	0.36	0.83			
Fe-6Cr	66	7.8185		0.28 ^b	1.91	1.43	0.63 ^b	3.51	0.10 ^b		1.91	1.32		0.55	0.11
Fe-9Cr	67	7.7933			1.82	2.22	0.68 ^b	4.14	0.24 ^b	1.79	2.11	1.29		0.42	0.03
Fe-12Cr	6A	7.7596		0.25 ^b	1.05	1.36	0.63 ^b	4.72	0.29 ^b	2.18	2.27	1.17		-0.21	-0.08
Fe-12Cr-0.1C	6B	7.7774			-0.46			1.60							
Fe-12Cr-0.1C ^a	6C	7.7952			-0.07										
Fe-12Cr-0.2C	6E	7.7680			0.28						-0.19				
Fe-12Cr-0.1C-0.5Mo	6F	7.7933			1.65			1.12	0.10 ^b						
Fe-12Cr-1Mo	6H	7.7911			2.41			0.01	0.05 ^b		-0.09				
Fe-12Cr-0.1C-1Mo	6J	7.7953			0.75	1.08		0.54							
Fe-12Cr-0.1C-2Mo	6K	7.8046			2.88	2.62		0.63	0.06 ^b		0.16				
Fe-12Cr-0.1C-3Mo	6L	7.8231			1.40	1.59		0.94	0.07 ^b		0.10				
Fe-15Cr	6N	7.7461			0.17	0.53		0.83	0.0 ^b	0.66	0.06	0.18	0.14	0.80	0.01
Fe-18Cr	6R	7.7233								0.32	0.0 ^b	-0.23		-0.01	

^aHeat treatment was 1038°C/1 hour/air cooled whereas all other conditions received 1038°C/1 hour/air cool + 760°C/2 hours/air cool.

^bTEM measurement whereas all other values are by density measurement.

^cTemp. - Irradiation temperature (°C), Flu - Irradiation fluence (10^{22} n/cm²).

These swelling measurements can be used to estimate a peak steady state swelling rate for ferritic alloys. Fig. 7.4.2 shows the results for swelling in Fe-6, 9 and 12Cr specimens at 400, 425 and 450°C. The 400°C points are connected by dashed lines, the 425°C points by solid lines and the 450°C points by short dashed lines. The peak swelling rate occurs for the 12Cr specimen for which a rate of 0.057%/dpa is found between 4.3×10^{22} n/cm² and 1.87×10^{23} n/cm². A rate of 0.06%/dpa appears to approximate the swelling rate observed at 400 and 450°C as can be perceived by comparison with the line for 0.06%/dpa inset in the figure. Therefore, a value of 0.06%/dpa is recommended as an estimate for the peak steady state swelling rate to be expected in ferritic alloys. This rate appears to apply over the temperature range 400 to 450°C and the temperature dependency of swelling in ferritic alloys is likely a consequence of differences in swelling incubation response. Effects of swelling saturation may also play a role at lower temperatures.

7.4.4.4 Discussion

Comparison with other materials

It is constructive to compare the present results with those of other materials of high technological impact, i.e., Path A. Such a comparison allows one to extend the present results so as to provide swelling predictions for commercial Path E alloys. Therefore, swelling values at 425°C for simple Fe-Cr-Ni ternary austenitic alloys,⁷ commercial austenitic alloys,¹ Fe-12Cr and commercial ferritic alloys^{1,2} are presented in Fig. 7.4.3. From Fig. 7.4.3 it can be shown that swelling in simple alloys is generally higher than in the commercial counterpart of the given alloy, that swelling in austenitic alloys is higher than in ferritic alloys and that a wide-range of commercial ferritic alloys show only a small variation in swelling response whereas a wide range of swelling response is found for a relatively narrow range of commercial austenitic alloys.

Swelling equation development

An approach to predict swelling in austenitic stainless steels has been proposed by Garner based on results for simple Fe-Ni-Cr ternary alloys.⁷ The ternary alloy results can be interpreted to establish that the steady state swelling rate is 1%/dpa and that swelling differences among alloys and conditions arise from differences in the onset of swelling. Swelling incubation is increased with increases in nickel content or temperature and with decreases in chromium content. This behavior is expected to apply for commercial austenitic alloys in various heat treatment conditions, as well.⁸ Fig. 7.4.3 has been prepared in this manner. Swelling for the commercial austenitic alloys has been drawn to predict a steady state swelling rate of 1%/dpa.

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Fig. 7.4.2. Swelling in Fe-6Cr, 9Cr and 12Cr binary alloys at 400, 425 and 450°C demonstrating a peak swelling rate of 0.06%/dpa.

Swelling response in ferritic alloys may be analyzed in the same way. For example, the peak swelling rate could be set at 0.06%/dpa based on the present results. This swelling rate appears to be satisfactory over the temperature range 400 to 450°C. Swelling differences from alloy to alloy can be ascribed to differences in the onset of swelling. (An exception must be made for the 400°C results at fluences above 50 dpa and this will be discussed presently.) Such an approach has been used in an accompanying report on 2-1/4Cr-1Mo.⁹ In order to predict swelling in commercial ferritic alloys such as EM12 and AISI 416, one must set a value for the incubation parameter and Fig. 7.4.3 indicates that a value of 100 dpa is probably conservative. A conservative estimate for swelling in commercial ferritic alloys at 500 dpa is then predicted to be 24 percent.

The appearance of void swelling in ferritic alloys does not necessarily mean that steady state swelling will immediately follow. Voids were found in both 2-1/4Cr-1Mo and EM12 after irradiation at 425°C to 1.6×10^{23} n/cm².¹⁰ However, comparison of density change results for these materials at 425°C to 1.6×10^{23} and to 2.1×10^{23} (Refs. 1-9) reveals only moderate increases in swelling, 0.06 percent for 2-1/4Cr-1Mo and 0.24 percent for EM12. A swelling rate of 0.06%/dpa yields a value of 1.5 percent for swelling. This means either that the transition to steady state swelling for commercial ferritic alloys is very gradual or that steady state swelling rates for simple ferritic alloys do not apply to commercial ferritic alloys. In either case, the prediction of 24 percent swelling in commercial ferritic alloys at 500 dpa is an overestimate.

Saturation

The results in Fig. 7.4.2 which are most difficult to explain are those for the 400°C irradiation temperature. The deviation from steady state swelling which is observed for the 400°C condition might be a consequence of specimen to specimen variations or experimental difficulties such as temperature control problems. However, the most likely explanation is that swelling saturation may have occurred. The condition required for saturation is that the void structure becomes the dominate sink for point defects. Such a situation is most likely to occur when the void density is very high (as is the case at lower temperature) and when the material is unable to maintain a high dislocation density (as is the case for soft, pure materials). Therefore, it is concluded that the results for the 400°C irradiation condition do not discredit the approach recommended for swelling equation development for commercial ferritic alloys.

Fig. 7.4.3. Comparison of swelling at 425°C in austenitic^{1,7} and ferritic alloys.^{1,2}

7.4.5 Conclusions

Swelling is moderate in Fe-Cr-C-Mo simple alloys irradiated to fluences on the order of 100 dpa. The peak swelling observed was 4.72 percent in an Fe-12Cr specimen irradiated at 425°C to 95 dpa. This corresponds to a swelling rate of 0.06%/dpa.

It is shown that this peak swelling rate of 0.06%/dpa may be used as a basis for developing swelling design equations for commercial ferritic alloys. Such equations, which are considered conservative estimate, that swelling in commercial ferritic alloys will be less than 24 percent after neutron exposures of 40 Mwy/m².

7.4.6 Future Work

An MHFES swelling equation for HT-9 will be developed in the next reporting period.

7.4.7 References

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