ON HYDROGEN AND HELIUM EMBRITTLEMENT IN ISOTOPIC TAILORING EXPERIMENTS - D. S. Gelles, M. L. Hamilton, B. M. Oliver and L. R. Greenwood (Pacific Northwest National Laboratory)*

OBJECTIVE

The objective of this effort is to quantify irradiation hardening response in isotopically tailored ferritic alloys following irradiation at 300°C in order to better understand behavior in this alloy class at low irradiation temperatures.

SUMMARY

The results of shear punch testing performed on irradiated isotopically tailored alloys are considered in terms of hydrogen and helium embrittlement in order to quantify the observed behavior. The results indicate that hydrogen embrittlement may be more significant than helium embrittlement.

PROGRESS AND STATUS

Introduction

Post-irradiation deformation response using shear punch testing has been reported for two series of alloys based on isotopic tailoring in order to study hydrogen and helium embrittlement in single variable experiments. Helium embrittlement using this approach was first studied in a series of Fe-12%Cr steels with intentional additions of 1.5% Ni either as natural Ni, ^{60}Ni or ^{59}Ni.[1,2] Following irradiation in HFIR at 300°C to 6.5 dpa, helium levels were estimated at 2.1 appm for both Fe-12Cr and Fe-12Cr-1.5^{60}Ni, at 41 appm for Fe-12Cr-1.5^{Nat}Ni and 70 appm for Fe-12Cr-1.5^{59}Ni, whereas hydrogen production was expected to be low. In the second experiment known as FIST, isotopic tailoring based on ^{54}Fe addition to the F82H composition was used.[3] In that case, following irradiation in HFIR at 300°C to 34 dpa, helium and hydrogen levels for the ^{54}Fe-containing alloy were measured at 65 appm and 400 appm, respectively and following irradiation at 250°C to 2.3 dpa they were 4.5 appm and 185 appm, respectively.[3] This data base provides an opportunity to separate behavior due to hydrogen and helium embrittlement. However, first, hydrogen and helium levels for the Ni doping experiment are reexamined and then the data are plotted and quantified (with the limitations of such an analysis well understood).

Results and Discussion

Helium concentrations have been measured for the Ni doped specimens irradiated at 300□C and hydrogen concentrations have been estimated for those irradiation conditions, assuming that all hydrogen is retained. The results are provided in Table 1 along with results obtained previously for FIST specimens but including both a revised prediction of the He levels for FIST and a revised measurement of H in FN51-2. The origin of higher H levels in specimen

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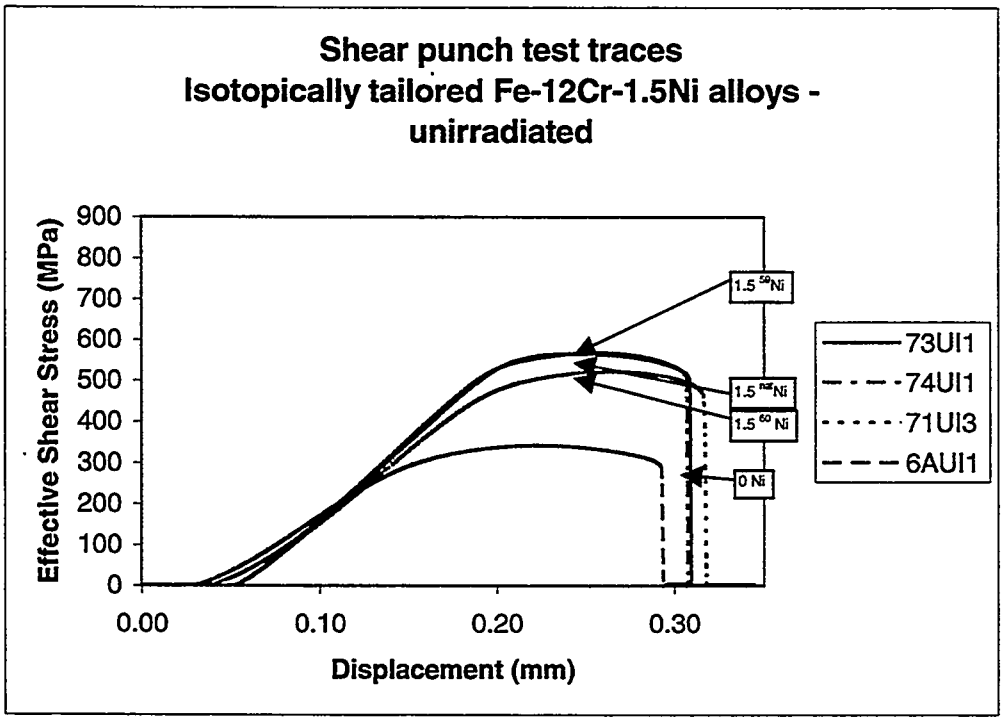
FN51-2 is not yet understood. From Table 1, it can be shown that the estimates for He in Ni doped specimens given above and in Reference 2 are very close to the measured values. The H levels anticipated in the Ni doped specimens are low and not very different as a function of composition. Most of the hydrogen in these specimens arises from the Fe-12Cr base composition because the Ni content is low, and at these doses, the ^{59}Ni effects are small relative to the fast neutron reactions. Therefore, the results of the Ni doped alloy series can be straightforwardly considered as an effect of He, and the consequences of an effect of H initially ignored. It should be noted that H levels measured for FIST irradiated specimens are closer than expected based on calculation.[3] This has been explained as a difference in behavior due to temperature, 250°C being low enough to retain H and include absorption from other causes and 300°C being high enough to allow release of most of the H produced.[3]

Table 1. Revision of measured (Exp.) and calculated helium and hydrogen concentrations for JP23 alloys irradiated at 300°C and FIST alloys irradiated at 250 and 300°C

Specimen	Material	Condition	He Conc. (appm)		H Conc. (appm)	
			Exp.	Calc.	Exp.	Calc.
6A5M-A	Fe-12Cr	300°C, 6.5 dpa	2.4	2.0	-	32
715M-A	Fe-12Cr-1.5^{60}Ni	300°C, 6.4 dpa	3.0	2.2	-	32
735M-A	Fe-12Cr-1.5^{59}Ni	300°C, 6.6 dpa	64.7	63	-	48
745M-A	Fe-12Cr-1.5^{Nat}Ni	300°C, 6.5 dpa	36.4	37	-	43
FN91-3	F82H ^{54}Fe	unirradiated	-	0	44±7	0
FN51-1&2	F82H ^{54}Fe	250°C, 2.3 dpa	4.5	4.6	340±199	182
C603	F82H ^{54}Fe	300°C, 34 dpa	65	65	396±21	2119

The shear punch results are provided both as test traces in Figure 1 and in tabulated form in Table 2. From Table 2, changes in shear strength values due to irradiation can be determined by comparison of unirradiated and irradiated response, and the results are plotted in Figure 2 as a function of He for the Ni doped specimen series and as a function of H for the FIST specimens. This choice for abscissa values is considered reasonable because the Ni doped alloys show greater variation in He production and the FIST specimens show greater variation in H production. From Figure 2, it can be shown that a trend appears evident in the FIST data as a linear function of H production but with large uncertainties in He level shown using labels in the plot. (Note that this is not likely to be a dose effect because hardening generally saturates quickly.) However, for the Ni doped alloys, the scatter is too large to demonstrate trends clearly. It is possible that there is no effect of He content on yield response whereas a linear response can be identified for the maximum strength response. But in both cases the scatter in the low He data is as large as the trends indicated.

It is well understood that quantitative definition of He and H embrittlement based on very limited data may be questioned; more data is needed. Nonetheless, this analysis indicates that H may play a larger role in embrittlement than He, depending on circumstances such as hydrogen release at higher temperatures. At 10 MWY/m^2 or approximately goal dose, He production is expected to be on the order of 1000 appm and H production on the order of



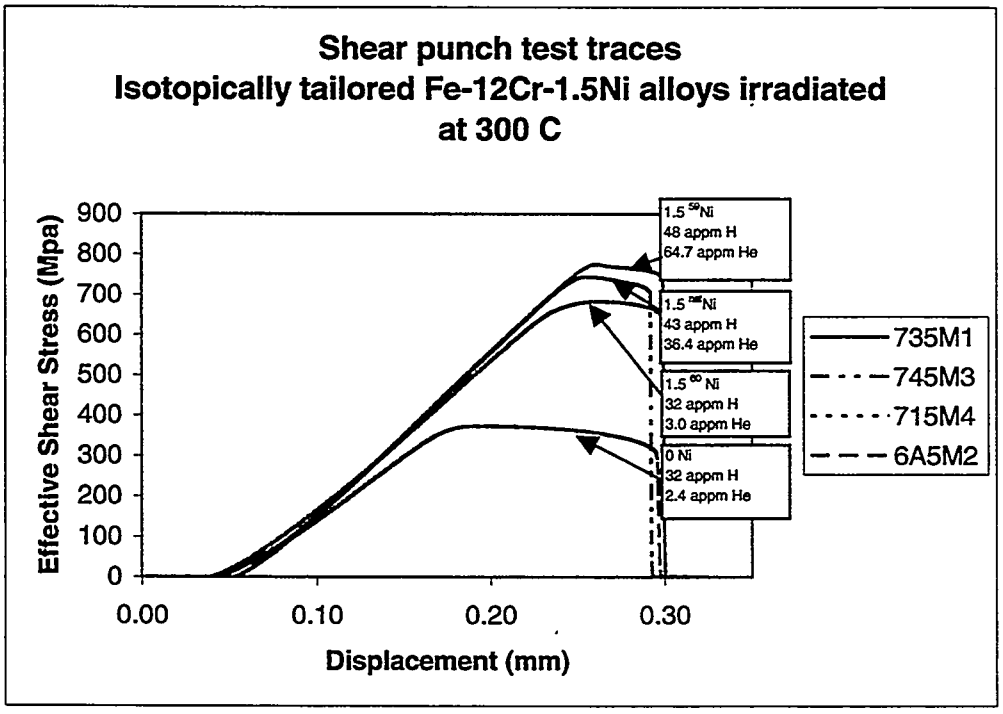


Figure 1 Shear punch test traces for isotopically tailored Fe-12Cr-1.5Ni a) unirradiated and b) irradiated at 300°C and c) for Fe F82H irradiated at 250 and 300°C.

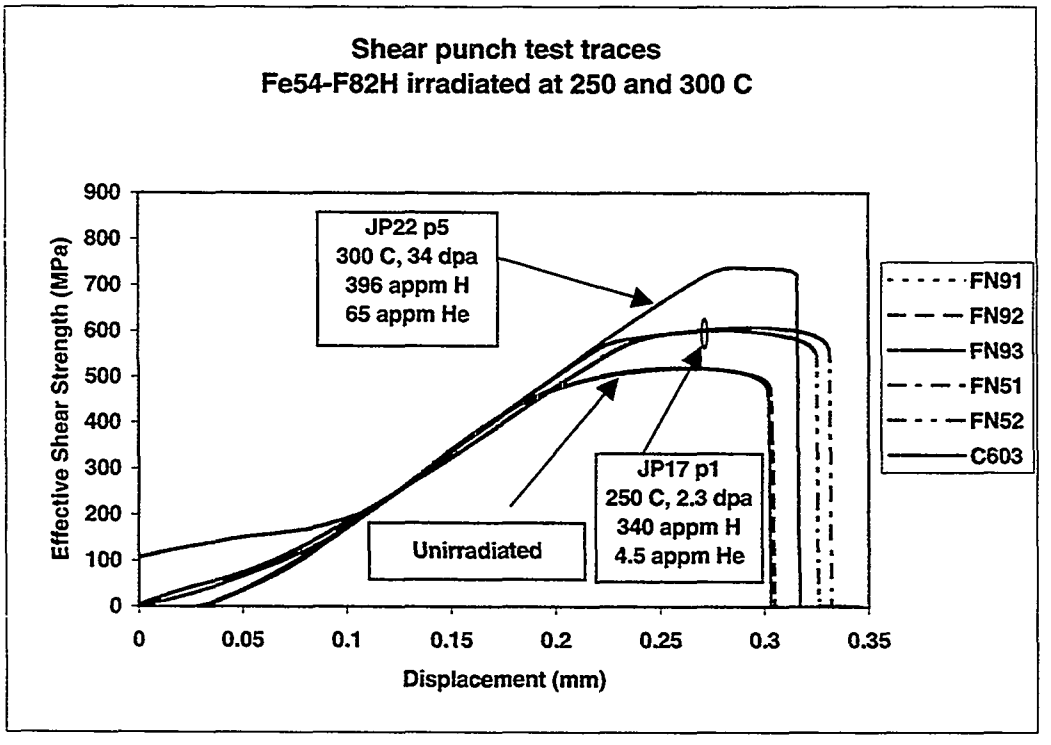


Figure 1 cont. **Shear punch test traces for isotopically tailored Fe-12Cr-1.5Ni a) unirradiated and b) irradiated at 300°C and c) for Fe F82H irradiated at 250 and 300°C.**

Table 2. Results of shear punch testing for Ni doped and FIST specimens.

	ID	Material	Condition	τ_{ys} (MPa)	τ_{ms} (MPa)
Ni doped	6A	Fe-12Cr	unirradiated	160	310
	71	Fe-12Cr-1.5^{60}Ni	unirradiated	360	530
	73	Fe-12Cr-1.5^{59}Ni	unirradiated	440	550
	74	Fe-12Cr-1.5^{Nat}Ni	unirradiated	410	580
	6A5M-A	Fe-12Cr	300°C, 6.5 dpa	300	400
	715M-A	Fe-12Cr-1.5^{60}Ni	300°C, 6.4 dpa	560	680
	735M-A	Fe-12Cr-1.5^{59}Ni	300°C, 6.6 dpa	610	780
	745M-A	Fe-12Cr-1.5^{Nat}Ni	300°C, 6.5 dpa	600	760
FIST	FN91-3	F82H ^{54}Fe	unirradiated	400	520
	FN51-2	F82H ^{54}Fe	250°C, 2.3 dpa	510	605
	C603	F82H ^{54}Fe	300°C, 34 dpa	600	738

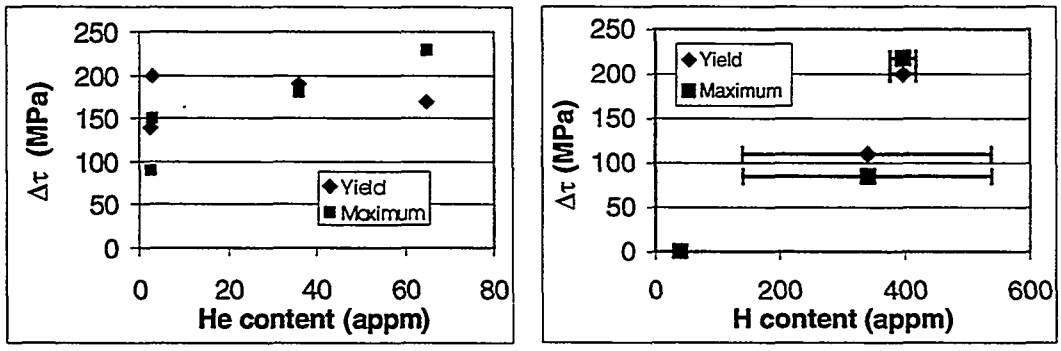


Figure 2 Change in τ_{ys} and τ_{ms} due to irradiation as a function of a) He content for Ni doped alloys and b) H content for FIST alloys.

10,000 appm. If our interpretation of H measurements in FIST specimens is correct, irradiation at temperatures above 250°C mitigates the problem because the H is not retained. The same is not expected for He. Also, synergistic effects can be anticipated at higher doses such that He bubbles may trap H at higher operating temperatures.

CONCLUSIONS

He levels obtained in Ni doped isotopically tailored alloys have been measured and the measurements agree with predictions.

H levels obtained in Ni doped isotopically tailored alloys have been estimated and it is found that a significant fraction of the H is obtained from transmutation of the base alloy for these low doses.

Analysis of limited shear punch data indicates a well-defined increase in strength with increasing H content whereas the data scatter as a function of He is as large as any trend indicated. However, more data is needed.

FUTURE WORK

This work will be continued when more specimens are available for testing.

REFERENCES

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2. D. S. Gelles, G. L. Hankin, and M. L. Hamilton, J. Nucl. Mater. 258-63 (1998) 1216.
3. M. L. Hamilton, D. S. Gelles, S. Ohnuki, K. Shiba, Y. Kohno, and A. Kohyama, DOE/ER-0313/25 (1999) 136.
4. L. R. Greenwood, B. M. Oliver, S. Ohnuki, K. Shiba, Y. Kohno, A. Kohyama, J. P. Robertson, J. W. Meadows and D. S. Gelles, submitted for publication in the proceedings of ICFRM-9 to be published in J. Nucl. Mater.