ACCELERATED HELIUM PRODUCTION IN FE-54 DOPED ALLOYS – MEASUREMENTS AND CALCULATIONS FOR THE FIST (FERRITIC ISOTOPIC TAILORING) EXPERIMENT

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OBJECTIVE

To demonstrate and predict the production of helium in iron alloys enriched in Fe-54 to accelerate helium production for fusion irradiation simulations in mixed-spectrum reactors.

SUMMARY

Iron alloys enriched in Fe-54 were irradiated in the High Flux Isotopes Reactor (HFIR). Measurements and calculations have been performed to demonstrate and to predict the accelerated helium production due to higher fast and/or thermal neutron cross sections for helium-producing reactions on the iron isotopes of Fe-54 and Fe-55. A helium/dpa ratio of 2.3 was achieved for a 1.25-year irradiation.

PROGRESS AND STATUS

Helium analyses of natural iron samples irradiated in HFIR indicated a non-linear buildup of helium suggesting accelerated helium production due to isotopic shifts.[1] In order to study these isotopic differences in both helium and hydrogen production, iron alloys enriched up to 90% in Fe-54 were fabricated in Japan.[2] The F-82H alloy nominally consisted of 8Cr-2WVTa. Fe-54 in the alloy comprised 96% of the iron or 86% of the total alloy by weight. For comparison, similar alloys were produced both with natural iron and with natural iron doped with varying amounts of boron.

TEM (transmission electron microscopy) alloy specimens were irradiated in the JP17 and JP22 irradiations in HFIR.[3,4] Neutron dosimetry measurements and radiation damage calculations for the JP17 experiment were reported previously.[5] Dosimetry monitors have not yet been analyzed for the JP22 irradiation. The JP17 irradiation was conducted from December 31, 1991, to February 17, 1992 for an exposure of 43.5 EFPD leading to about 2.8 dpa and 1.0 appm helium in natural iron. The JP22 irradiation from December 1993 to January 1996 produced an exposure of 458.2 EFPD leading to about 29 dpa and 10.5 appm helium in natural iron. Calculations discussed in this report used the JP17 dosimetry data normalized to the JP22 exposure conditions.

Recently, four samples from these irradiations were analyzed for helium content at the Pacific Northwest National Laboratory. The composition of each specimen is listed in Table I. The helium content of each specimen was determined by isotope-dilution gas mass spectrometry following vaporization in a resistance-heated graphite crucible in one of the mass spectrometer system's high-temperature vacuum furnaces.[6] The absolute amount of ^{4}He released was measured relative to a known quantity of added ^{3}He "spike". The results of the helium analyses are given in Table II, and are listed as total atoms of ^{4}He released, and as ^{4}He concentrations in atomic parts per million (10^{-6} atom fraction). Conversion from total

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helium to helium concentration was based on an average value of 1.075 x 10^{22} atoms/g. The conversion values were calculated using the alloy compositions. It should be noted, however, that these conversion values, and the helium concentrations obtained using them, are not very sensitive to small changes in material composition. Absolute uncertainty (1σ) in the individual helium analysis results, determined from the cumulative uncertainties in the sample mass, the isotope ratio measurement, and the spike size, is estimated to be approximately 1%.

Table 1. Composition of Fe Alloys

Sample	Irradiation	Alloy*	Iron	Boron-10
FN51	JP-17	F-82H Fe54	96% Fe-54	-
C603	JP-22	F-82H Fe54	96% Fe-54	-
C103	JP-22	F-82H B-10	Natural Iron	0.0001 %
C203	JP-22	F-82H B-10	Natural Iron	0.0058 %

*F82-H Fe-54 alloy composition: 7.1Cr-1.8W-0.55Si-0.40Mn-0.17V-0.1C-0.04Ta

Table 2. Helium Concentrations in FIST Samples

Specimen	Mass[a] (mg)	Measured ^{4}He (10^{14} atoms)	Helium Concentration (appm)[b]	
			Measured	Average[c]
FN51-2-A	1.073	0.5278	4.550	4.52
-B	1.603	0.7765	4.481	0.05
C103-A	1.567	3.684	21.89	21.8
-B	1.892	4.407	21.69	0.1
C203-A	0.851	29.81	325.9	320
-B	1.864	63.05	314.7	8
C603-A	1.047	7.348	64.92	64.8
-B	1.711	11.96	64.66	0.2

[a]Mass of specimen for analysis. Mass uncertainty is ±0.002 mg.
[b]Helium concentration in atomic parts per million (10^{-6} atom fraction) with respect to the total number of atoms in the specimen.
[c]Mean and standard deviation (1σ) of duplicate analyses.

The production cross sections for helium differ for each iron isotope, as shown in Figure 1. The cross sections shown in the figure were taken from the ENDF/B-VI libraries.[7] During the course of each irradiation, the iron isotope ratios change due to (n,γ) reactions, followed by the subsequent decay of the radioactive isotopes of Fe-55 (2.73 y) and Fe-59 (44.5 d). Reference 1 demonstrated these effects using mass spectrometry to measure the altered iron isotopic ratios. As the iron isotope ratios change, the helium production will increase or decrease according to the different cross sections shown in Figure 1.

These cross sections were integrated over the HFIR neutron energy spectrum to determine the helium production rates. The calculated ratios of fast neutron helium production relative to that of natural iron are 1.85 (Fe-54), 0.93 (Fe-56), 1.89 (Fe-57), and 0.16 (Fe-58). Helium production cross sections are not available for the radioactive isotopes Fe-55 and Fe-59. New theoretical calculations would be highly useful for predicting these effects. In reference 1, the semi-empirical computer code THRESH2 was used to calculate the helium production rate ratios. These calculations predicted a ratio of 5.95 for Fe-55, although the ratios differed somewhat from the ENDF/B-VI values for the other iron isotopes.

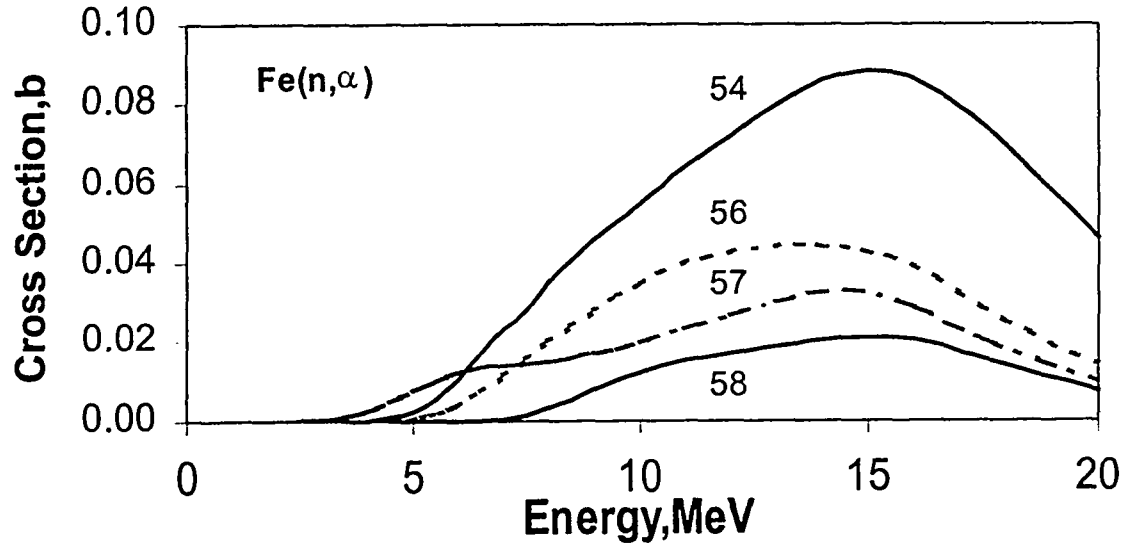

Figure 1. ENDF/B-VI neutron cross sections for the (n,α) reaction for various iron isotopes.

In order to calculate the helium production for the materials used in the present study, the measured neutron energy spectrum was used to calculate the transmutation rates for the various iron isotopes. Helium production was then integrated over short time intervals over the course of each irradiation. For the samples FN-51 and C603, which are enriched in Fe-54, the calculations using the above cross section ratios underpredict the measured helium values by about 50%. In order to fit the JP-17 helium measurement for sample FN-51, it is necessary to increase the fast neutron helium production in Fe-54 by about 86%. This cross section is not very well known at lower neutron energies, which are the most important in the HFIR neutron energy spectrum. This new measurement would thus suggest that the Fe-54 (n,α) cross section in ENDF/B-VI is too low. The higher helium level seen in the JP-22 irradiation for sample C603 further requires some adjustment of the helium production cross section in Fe-55. Mathematically, it is possible to fit the data with either a greatly elevated rate of fast neutron production in Fe-55 or a weak thermal neutron effect in Fe-55, as originally suggested in reference 1. If the effect is solely due to fast neutrons, then the ratio of helium production in Fe-55 to that of natural iron would have to be quite high at about 37:1. Looking at Figure 1, this would require a neutron cross section peaking at about 1.8 barns, which seems rather unlikely. The Q-value for the Fe-55(n,α)Cr-52 reaction is quite high at +3.584 MeV. This would suggest the possibility of a thermal neutron cross section. If the Fe-55/natural iron helium production ratio is held at the THRESH2 predicted value of 5.95, then the helium data can be very well fit using a small thermal neutron helium production cross section in Fe-55 of about 0.011 barns,

well within the range of the suggested value given in reference 1. Since we do not know either the fast or thermal helium cross sections for Fe-55, we can only give a range of values that would fit the data. The most likely answer is that there is a thermal neutron cross section in Fe-55 of about 0.01 barns and that the fast helium cross section ratio to natural iron is about 5-10. The data and calculations are compared in Figure 2. The dotted line is calculated without considering any possible helium production from Fe-55. The curve starts to bend over at high dpa due to the burnup of Fe-54. The solid line includes helium production from Fe-55 using a thermal neutron cross section of 0.01 barns and a fast helium cross section of 6 times the natural iron helium production. The calculations were extended to higher exposures to predict achievable helium/dpa ratios in HFIR for these alloys, as discussed below.

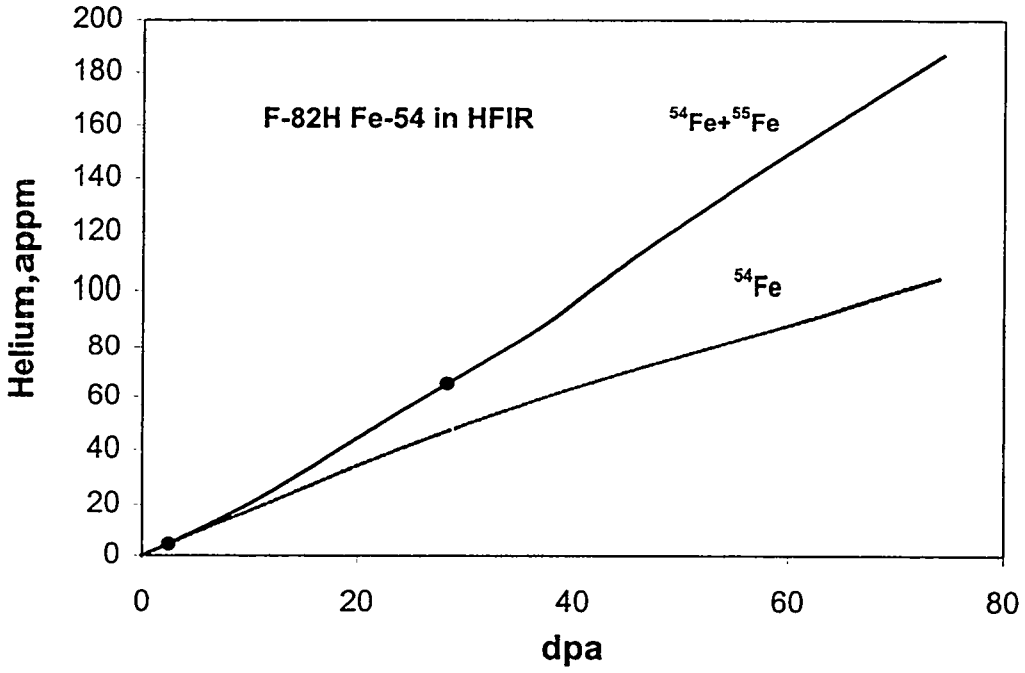

Figure 2. Measured helium production (solid circles) compared to calculations with only the modified Fe-54 cross sections (dotted line) and with both Fe-54 and Fe-55 modified cross sections (solid line).

In the case of the natural iron alloys doped with boron (C103 and C203) the high thermal neutron fluences in these HFIR experiments totally burned out the B-10 to helium. Converting from weight percent to atom percent this would predict 4 appm helium from B-10 in sample C103 and 321 appm helium from B-10 in sample C203. Using the same iron isotopic cross sections derived for the Fe-54 doped alloys, we would predict 20 appm helium from the natural iron alloys. Adding this to the B-10 values would predict 24 appm helium in sample C103 compared to the experimental value of 22 appm helium (C/E = 1.10). Similarly, the calculated value for sample C203 is 344 appm helium compared to the measured value of 321 appm helium (C/E = 1.07). This level of agreement is well within the uncertainties on the boron concentrations. Hence, the Fe-54 and Fe-55 cross sections also give good agreement for natural iron, similar to that seen previously in Reference 1.

RECOMMENDATIONS FOR FUTURE IRRADIATIONS

The present work demonstrates that elevated helium production can be achieved in iron by enriching with Fe-54 or Fe-55. Irradiations with natural iron produce a nearly constant ratio of helium (appm) to dpa of about 0.54 using the ENDF/B-VI calculated rate. It should be noted that this rate is somewhat higher than the ENDF/B-V Gas Production File value of 0.37, as quoted in reference 5, for example. The present experiment achieved a helium/dpa ratio of 2.3, an increase of a factor of 4.3 over natural iron. In order to simulate fusion reactor conditions, we would like to achieve a helium/dpa ratio of about 10. Using the present alloys, which are nearly fully-enriched in Fe-54, it would only be possible to achieve a helium/dpa ratio of about 3:1 with a similar irradiation in HFIR lasting about 4 years, as shown in Figure 2. HFIR produces about 22.7 dpa in iron per full power year of operation. If alloys were produced with an initial doping with Fe-55, then fusion-like ratios of helium/dpa could be easily achieved. Regardless of whether the Fe-55 effect is due to thermal or fast neutrons, Fe-55 will produce about 37 times more helium than natural iron in HFIR. Pure Fe-55 would thus produce a helium/dpa ratio of about 20:1. The desired fusion helium/dpa ratio of 10:1 could be achieved by doping natural iron with 50% Fe-55 or with a mix of Fe-54 and Fe-55. This production rate would, however, decrease with time as Fe-55 either decays (2.73 y) or is transmuted to Fe-56. It would be possible to offset these losses by using a mix of Fe-54 and Fe-55 so that Fe-54 would be transmuted to Fe-55 to replace these losses. Total replacement is possible if we match the initial Fe-54/Fe-55 ratio to the ratio of the decay rate of Fe-55 to the transmutation rate of Fe-54 of about 3.5:1. In this case, we could achieve a nearly constant helium/dpa ratio of about 5.9:1. Of course, one would have to allow for losses of Fe-55 due to decay from the time of production and including possible reactor down times.

Fe-55 is a low-energy x-ray emitter. Hence, it could be safely handled in relatively large quantities for production of doped alloys, although cost considerations might be a more serious problem. An alternative would be to first irradiate Fe-54 to breed Fe-55, then use this transmuted material to make the alloy for the fusion simulation experiment. For example, a two-year irradiation of Fe-54 in HFIR would breed about 8% Fe-55. The Fe-54 will decrease about 15%, the difference mostly going to Fe-56. The decay of Fe-55 produces Mn-55 which also rapidly burnsup to Fe-56. This transmuted iron would then produce an initial helium/dpa ratio of about 3.3:1 increasing to about 4:1 after a year of irradiation. In order to achieve higher helium/dpa ratios, it would be necessary to isotopically separate the Fe-55 produced from the Fe-54 irradiation (or purchased commercially) and then use the separated Fe-55 for further alloy preparation. In this way, one could produce alloys with any desired helium/dpa ratio including 20:1 (pure Fe-55), 10:1 (50:50 mix of Fe and Fe-55), or a steady-state production at 6:1 (3.5:1 ratio of Fe-54 to Fe-55).

FUTURE WORK

Work is in progress to measure and calculate hydrogen production in these samples. Additional helium measurements at higher exposure levels would be highly useful in establishing the helium production cross sections in the iron isotopes.

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