RADIATION DAMAGE CALCULATIONS FOR THE FUBR AND BEATRIX IRRADIATIONS OF LITHIUM COMPOUNDS IN EBR-II AND FFTF - L. R. Greenwood (Pacific Northwest National Laboratory)*

OBJECTIVE

To calculate displacement damage for lithium compounds irradiated in the EBR-II and FFTF reactors.

SUMMARY

The Fusion Breeder Reactor (FUBR) and Breeder Exchange Matrix (BEATRIX) experiments were cooperative efforts by members of the International Energy Agency to investigate the irradiation behavior of solid breeder materials for tritium production to support future fusion reactors. Lithium ceramic materials including Li_2O, $LiAlO_2$, Li_4SiO_4, and Li_2ZrO_3 with varying ^6Li enrichments from 0 to 95% were irradiated in a series of experiments in the Experimental Breeder Reactor (EBR II) and in the Fast Flux Test Facility (FFTF) over a period of about 10 years from 1982 to 1992. These experiments were characterized in terms of the nominal fast neutron fluences and measured ^6Li burnup factors, as determined by either mass spectrometry or helium measurements. Displacement per atom (dpa) values have been calculated for each type of material and irradiation. Values up to 11% ^6Li-burnup and 130 dpa are predicted for the longest irradiations. Using these new calculations, previously measured radiation damage effects in these lithium compounds can be compared or correlated with other irradiation data on the basis of the dpa factor as well as ^6Li-burnup.

PROGRESS AND STATUS

The recoiling alpha and triton from the $^6Li(n,\alpha)^3H$ reaction create a cascade of secondary recoil atoms from their interactions with atoms in the compound material. Fast neutron interactions with atoms in the compounds can also cause primary recoil atoms that also lead to a cascade of secondary recoil atoms. Both of these processes lead to radiation damage characterized by the total number of displacements per atom (dpa). Although dpa is not in itself a measurable effect, it has proven useful as an exposure parameter for correlating material effects for different materials and radiation environments. The purpose of this report is to document the calculation of dpa for the Li breeder compounds in the FUBR and BEATRIX series of irradiations.

It should be noted, however, that other effects besides dpa will also be important for characterizing the irradiation performance of these materials, especially transmutation which leads to the creation of Li vacancies and the buildup of transmutant product interstitials. The absolute value of the calculated dpa is somewhat irrelevant due to the very high rate of atomic recombination in most materials. As an example, the longest exposures in this study lead to the prediction of about 11% burnup of ^6Li and 130 dpa. However, if 99% of the displaced atoms recombine, then burnup (transmutation) effects may be a factor of ten more important than stable product dpa in the evolution of material property changes. Irradiation experiments performed on these lithium ceramics do not indicate any significant property

*Pacific Northwest National Laboratory (PNNL) is operated by the U.S. Department of Energy by Battelle Memorial Institute under contract DE-AC06-76RLO-1830.

changes at these burnup or dpa levels that would seriously degrade the usefulness of these materials for tritium breeding in fusion reactor applications. Ultimately, it is the measured performance of lithium ceramic breeder materials as a function of irradiation exposure that determines their applicability in fusion reactor designs. Comparisons with dpa levels in structural materials such as stainless steel may thus be irrelevant.

The lithium containing ceramic materials were prepared by members of an International Energy Agency collaborative program, as documented in several publications.[1,2,3,4,5,6,7,8,9,10,11,12] In EBR-II, ceramic pellets were irradiated in canisters placed in several pins located in row 7 at different radial and axial locations. The irradiations in the FFTF MOTA were designed to measure tritium release. The lithium was enriched in ^{6}Li in varying amounts from 0 to 95%.

All of the irradiation histories are summarized in Table 1. Details of the EBR-II irradiations were provided by D. G. Porter (Argonne National Laboratory – West, 1999.) Details of the FFTF-MOTA irradiations are documented in references 9 -12. Neutron dosimetry measurements have been published for the EBR-II reactor[13,14] and for the FFTF irradiations.[15,16]

Table 1. Summary of the FUBR and BEATRIX Irradiation Histories

Experiment	Subassembly	Subcapsules	Runs	MWd	EFPD
	EBR-II				**@62.5 MW**
FUBR-1A	X370	1,2,3,4	119-120	6041	96.7
	X370A	5,6,7,8	123-126	11101	177.6
	In both runs	9,10,11	119-126	17142	274.3
FUBR-1B	X415	B1,B3,B6,B7	135A-141C[a]	21344	341.5
BEATRIX-I	X415A	B8,B9,B10,B1 1	144A-151	37455	599.3
	In both runs	B2,B4,B5	135A-151	58799	940.8
FUBR-1B	X416	S1,S2	135A-141C[a]	21104	337.7
BEATRIX-I	X416A	S4,S5	144A-151	37455	599.3
	In both runs	S3	135A-151	58559	936.9
	FFTF				**@291 MW**
BEATRIX-II	MOTA-2A	1B,1C,1D,1E, 5E,8B	11	87213	299.7
BEATRIX-II	MOTA-2B	1B,2C,5F,8B	12	59160	203.3

[a] X415 was removed for runs 137E and 138B; X416 was removed for 137E, 138B, and 138C.

Radiation Damage Calculations

The ^{6}Li(n,α)^{3}H reaction releases 4.782 MeV of energy (Q value) resulting in energetic tritium (2.733 MeV) and alpha (2.050 MeV) particle recoils. These high-energy recoils lose energy and are stopped within a short distance in the lithium compounds. A small fraction of the time, the recoiling particles hit a nucleus of one of the atoms in the compound leading to either an elastic scattering event or a nuclear reaction. These nuclear events cause secondary atoms to be displaced from their lattice sites. If we use the Lindhard model of energy loss and a modified Kinchin-Pease model for secondary displacements, then we can calculate the total number of displaced atoms for each $^{6}Li(n,\alpha)^{3}H$ reaction. This leads to a fixed relationship between burnup of ^{6}Li atoms and dpa (for this reaction only). Each compound will have a different ratio of burnup to dpa due to the differences in lithium content and energy loss characteristics. In the modified Kinchin-Pease model, constant displacement energy is assigned to each atom in a given compound. Unfortunately, these values are not readily available for these compounds. Efforts are underway to use a better model for these compounds; however, these results are not currently available. If we assign values of 10 eV for Li, 30 eV for O, 27 eV for Al, 25 eV for Si, and 40 eV both for Ti and Zr, then the calculated conversion of ^{6}Li burnup to dpa is given in Table 2, assuming 100% enrichment of ^{6}Li. For a measured level of ^{6}Li burnup and enrichment, the dpa can be calculated by multiplying the values in Table 2 times these two factors. For example, if we start with 56% enriched Li_2O and the ^{6}Li burnup is measured to be 8%, then the dpa due to this reaction will be 117 x 0.56 x 0.08 = 5.2. Once the ^{6}Li is fully burned out, this process will stop and no further dpa can be generated from this reaction. It should be noted that the conversion factors in Table 2 are technically only correct at low neutron energies since at higher energies the energy of the incident neutron will raise the recoil energies slightly. However, since this reaction mainly occurs with neutrons well below 1 MeV, this correction for the neutron spectra of interest is negligible. Due to the burnup of ^{6}Li as well as the variations in the ^{6}Li enrichment, dpa cross sections for these compounds depend on both of these factors and change with irradiation as the ^{6}Li burns up. Both of these effects were taken into account in these calculations. Due to the high cross section of ^{6}Li at lower neutron energies, neutron self-shielding can also be an important time-dependent effect. Simple approximations to the neutron self-shielding effects for the materials and neutron spectra used in these experiments indicate that the corrections would typically be on the order of 5%. Due to the complicated geometries of these experimental assemblies and the relatively small order of these corrections, neutron self-shielding corrections were not included. The calculated ^{6}Li burnup values presented below agree favorably with measurements made for these materials, as documented in various references in this report.

Table 2. Conversion of ^{6}Li Burnup to dpa
(Multiply times ^{6}Li enrichment)

Material	Dpa / % ^{6}Li Burnup
Li_2O	117.0
$LiAlO_2$	33.7
Li_2ZrO_3	44.9
Li_4SiO_4	67.8

Of course, all other possible nuclear reactions will also cause dpa. The SPECOMP[17] computer code was used to calculate these more conventional dpa cross sections as a function of neutron energy for each type of lithium compound. These dpa cross sections were then added to the SPECTER[18] computer code for integration over the neutron flux spectra in each series of irradiations. The calculated dpa values for each series of irradiations are discussed below. Specific comments describe each experiment. In all of these irradiations, the reactor power and hence dpa rates are essentially constant (<10% variation over the run cycles) except during reactor downtimes. For all of the irradiations, the Li compounds were encapsulated in 316 stainless steel containers. The dpa values for the 316 SS cladding are also presented below along with the total neutron fluences. The composition of 316 SS was taken as Ni(0.13), Cr(0.18), Mn(0.019), Mo(0.026), and Fe(balance).

FUBR-1A Irradiation in EBR-II

Eleven pins were fabricated for irradiation designated PIN-1 through PIN-11. Solid pellets were irradiated at four different elevations in each pin. Canisters were placed symmetrically about the reactor centerline at 1.2 to 7.9 cm and at 15.2 to 21.6 cm for irradiations of 96.7, 177.6, and 274.3 EFPD at a nominal power of 62.5 MW. The dpa calculations for the highest exposure in these positions and exposure parameters are given in Table 3. The dpa values in the table are average values for the assemblies and radial variations are <10%. Axial flux variations are also relatively small (<10%) near core midplane.

Table 3. dpa Calculations for FUBR-1A – Capsules 9, 10, 11 – 274.3 EFPD

Fluence ($x10^{22}$ n/cm^2)		3.94	4.11	3.79
Dpa, 316 SS		15.9	16.8	15.2
^{6}Li Burnup,% :		3.38%	3.46%	3.14%
Height,cm:		-20	0	+20
Material	^{6}Li Enrichment,%	dpa	dpa	dpa
Li_2O	56	35.4	37.3	34.0
$LiAlO_2$	95	37.1	39.1	35.6
Li_2ZrO_3	95	37.6	39.5	36.0
Li_4SiO_4	95	34.6	36.4	33.1

FUBR-1B/BEATRIX-I Irradiations in EBR-II

Two different types of pins were fabricated for these irradiations. B7A pins labeled B1 through B11 contained canisters with Li ceramic pellets at three different elevations, -15.5 to –21.6 cm, -7.6 to –13.2 cm, and +16.8 to +22.4 cm. S pins labeled S1, S2, and S3 contained canisters with Li ceramic pellets at two elevations symmetrically around midplane at 1.3 to 7.9 cm. The B7A pins were irradiated for 341.5, 599.3, and 940.8 EFPD at 62.5 MW. Table 4 lists the dpa values at the maximum exposure. Calculations for capsules S1-S3 differed slightly due to small differences in the irradiation exposures of 337.7, 599.3, and 946.9 EFPD. Variations in dpa are not very large for all of the different positions. The dpa values in the table are average values for the assemblies and radial variations are <10%. Axial flux variations are also relatively small (<10%) near core midplane.

Table 4. dpa Calculations for FUBR-1B/BEATRIX-I – Capsules 2, 4, 5 – 940.8 EFPD

Fluence ($x10^{22}$ n/cm^2)		13.5	14.1	13.0
Dpa, 316 SS		54.5	57.6	52.2
^{6}Li Burnup,% :		11.11%	11.36%	10.38%
Height,cm:		-20	0	+20
Material	^{6}Li Enrichment,%	dpa	dpa	dpa
Li_2O	56	121.0	127.3	116.1
Li_2O	7.5	114.6	120.8	110.0
Li_2O	0.07	113.7	119.9	109.1
$LiAlO_2$	95	127.1	134.0	122.0
Li_2ZrO_3	95	128.6	135.2	123.2
Li_4SiO_4	95	118.1	124.5	113.2

BEATRIX-II Irradiations in FFTF

The BEATRIX-II experiments were conducted in the MOTA-2A and MOTA-2B irradiations in FFTF. These experiments were primarily designed to evaluate the rate of recovery of tritium during irradiation. The experimental assemblies consisted of solid pellets or tubes (rings) of lithium ceramics with flowing gas in the center or sphere beds. In MOTA-2A, the Li_2O ring canister was in position 1E and solid pellets and single crystals were in positions 1B, 1C, 1D, and 1E. In MOTA-2B, the Li_2O ring canister was located in position 1B and a Li_2ZrO_3 sphere bed was located in position 2C. $LiAlO_2$, Li_2ZrO_3 and Li_4SiO_4 disc samples were also irradiated in both experiments. The dpa calculations listed in Tables 5 and 6 are quoted for positions where the neutron flux spectra were determined from neutron dosimetry measurements. Radial flux gradients are relatively small (<10%). Axial flux gradients are relatively small near midplane where the main testing was done, but become quite steep at the top of the core 8B position. However, the dpa values are so low at these out-of-core positions that the materials damage effects are probably negligible.

Table 5. dpa Calculations for BEATRIX-II – MOTA-2A – 299.7 EFPD

Fluence ($\times 10^{22}$ n/cm^2)		8.89	7.44	0.24
Dpa, 316 SS		24.9	21.8	0.25
^{6}Li Burnup,% :		9.96%	7.60%	1.68%
Reactor Position:		1-B,C,D,E	5E	8B
Height,cm:		-42.7	+37.7	+122.4
Material	^{6}Li Enrichment,%	dpa	dpa	dpa
Li_2O	61	68.1	56.6	1.93
Li_2O	0.07	60.8	51.2	0.72
$LiAlO_2$	95	68.3	57.5	1.27
Li_2ZrO_3	95	63.9	53.7	1.39
Li_4SiO_4	95	70.4	58.8	1.81

Table 6. dpa Calculations for BEATRIX-II – MOTA-2B – 203.3 EFPD

Fluence ($\times 10^{22}$ n/cm^2)		5.63	7.91	4.77	0.20
Dpa, 316 SS		14.2	21.1	12.9	0.23
^{6}Li Burnup,% :		6.68%	7.92%	5.29%	1.27%
Reactor Position:		1B	2C	5F	8B
Height,cm:		-41.2	-27.3	+44.3	+121.3
Material	^{6}Li Enrichment,%	dpa	dpa	dpa	dpa
Li_2O	95	41.5	59.9	36.9	2.07
Li_2ZrO_3	85	35.5	52.4	32.3	1.08
Li_2ZrO_3	0.2	32.9	49.4	30.3	0.59
Li_4SiO_4	0.2	35.3	53.1	32.4	0.64

CONCLUSIONS

The calculations clearly show that most of the dpa damage in these materials arises from fast neutron reactions such as elastic and inelastic scattering rather than from the ^{6}Li(n,α)t reaction. This effect can be seen most clearly in the tables that include materials that were both enriched and depleted. For example, in Table 4 at midplane the dpa for Li_2O with 56% enrichment is 127.3 compared to 119.9 for depleted Li_2O. This means that with 56% enrichment, only 6% of the dpa is coming from the ^{6}Li(n,α)t reaction. Using the dpa to burnup conversion factors in Table 2, it is easy to estimate the contribution to the dpa from the $^{6}Li(n,\alpha)t$ reaction for any entry in the dpa tables. The dpa values in this report were calculated using the conventional dpa model. Newer calculations are in progress using improved models. When these cross sections become available, it would be straightforward to recalculate the dpa factors for these experiments. However, the current dpa values are compatible with the conventional dpa calculations that are widely used and quoted in the literature. For example, the dpa values for the 316 SS cladding is also given for each of the irradiations. Transmutation effects due both to the $^{6}Li(n,\alpha)t$ reaction as well as other nuclear reactions may be very important in influencing the degradation of materials since these effects produce a permanent change in the irradiated material. In the comparison of these fast neutron experiments on lithium ceramics to similar experiments in thermal or mixed-spectrum reactors, it is likely that the fast neutron dpa rates are much lower in the mixed-spectrum reactors. Fusion reactors will also exhibit differences from fast reactors in the neutron spectra due to both the 14 MeV neutrons and increased lower-energy neutron flux due to Be blankets and other moderating materials. A more comprehensive technical report of this work is being prepared.[19]

ACKNOWLEDGMENTS

G. W. Hollenberg (Pacific Northwest National Laboratory) and C. E. Johnson (Argonne National Laboratory - East) provided guidance and technical assistance with assembling the information concerning these experiments. D. G. Porter (Argonne National Laboratory – West) was very helpful in providing information regarding the irradiation history of these experiments in the EBR II reactor.

REFERENCES

1. R. C. Knight and G. W. Hollenberg, HEDL-TC-2147, Fabrication Data Package for HEDL FUBR Solid Breeder Irradiation Experiment in EBR-II, February 1982.

2. G. W. Hollenberg, Journal of Nucl. Mater. 122&123, 896-900 (1984).

3. G. W. Hollenberg and D. L. Baldwin, Journal of Nucl. Mater. 133&134, 242-245 (1985).

4. D. L. Baldwin and G. W. Hollenberg, Journal of Nucl. Mater. 141-143, 305-310 (1986).

5. G. W. Hollenberg and R. C. Knight, HEDL-TC-2682, Revised Data Package for FUBR-1B Solid Breeder Experiment in EBR-II – First Insertion, April 1985.

6. L. S. Williams and G. W. Hollenberg, HEDL-TC-2704, Fabrication Data Package for FUBR-1B Solid Breeder Experiment in EBR-II, June 1985 and Addendum A-Second Insertion, March 1987.

7. G. W. Hollenberg, R. C. Knight, P. J. Densley, L. A. Pember, C. E. Johnson, R. B. Poeppel, and Y. Ya G. W. Hollenberg, C. Alvani, B. Rasneur, H. Elbel, H. Wedemeyer, D. E. Walker, T. Takahashi, K. Noda, and H. Akiyama, Journal of Nucl. Mater. 155-157, 563-567 (1988).

8. O. D. Slagle, G. W. Hollenberg, and D. L. Baldwin, Tritium Release and Physical

Stablility of Solid Breeder Materials, Fusion Reactor Materials Semiannual Progress Report, DOE/ER-0313,8, pp. 281-285, August 1990.

9. M. L. Hamilton and R. M. Ermi, Preparation and Irradiation of the Fusion MOTA-2A, Fusion Reactor Materials Semiannual Progress Report, DOE/ER-0313/8, pp. 3-16, August 1990.

10. G. W. Hollenberg, H. Watanabe, I. J. Hastings, and S. E. Berk, Journal of Nucl. Mater. 191-194, 23-29 (1992).

11. R. A. Verrall, O. D. Slagle, G. W. Hollenberg, T. Kurasawa, and J. D. Sullivan, Journal of Nucl. Mater. 212-215, 902-907 (1994).

12. M. L. Hamilton, R. M. Ermi, and A. M. Ermi, Preparation and Irradiation of Fusion MOTA 2B, Fusion Reactor Materials Semiannual Progress Report, DOE/ER-0313/12, pp. 3-14, July 1992.

13. L. R. Greenwood, Fission Reactor Dosimetry – EBR-II X287, Damage Analysis and Fundamental Studies Quarterly Progress Report, DOE/ER-0046/7, pp. 8-16, November 1981.

14. F. C. Franklin, E. R. Ebersole, and R. R. Heinrich, Analysis of EBRII Low-Power Dosimetry Run 78C, ANL-77-76, 1977.

15. L. R. Greenwood and L. S. Kellogg, Neutron Dosimetry for the MOTA-2A Experiment in FFTF, Fusion Reactor Materials Semiannual Progress Report, DOE/ER-0313/12, pp. 49-52, July 1992.

16. L.R. Greenwood and L. S. Kellogg, Neutron Dosimetry for the MOTA-2B Experiment in FFTF, Fusion Reactor Materials Semiannual Progress Report, DOE/ER-0313/16,pp. 21-27, September 1994.

17. L. R. Greenwood, ASTM STP 1001, 598-602 (1989).

18. L. R. Greenwood and R. K. Smither, ANL/FPP/TM-197, Argonne National Laboratory (1985).

19. L. R. Greenwood, PNNL-12200 (1999).