THE TRIO-01 EXPERIMENT: IN-SITU TRITIUM RECOVERY RESULTS

R. G. Clemmer, P. A. Finn, M. C. Billone, B. Miara,
and L. R. Greenwood
Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois 60439
F. F. Dyer, I. T. Dudley, L. C. Bate, E. D. Clemmer,
P. W. Fisher, J. L. Scott, and J. S. Watson
Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, Tennessee 37830

CONF - 831203--35

CONF-831203--35

DE84 004003

Abstract

The TRIO-01 experiment was designed to test in-situ tritium recovery and heat transfer performance of a candidate solid breeder, γ-$LiAlO_2$. The results showed that nearly all the tritium generated was recovered. Only <0.1 wppm tritium remained in the solid after irradiation testing. The heat transfer performance showed that temperature profiles can be effectively controlled.

Introduction

The TRIO-01 experiment involves the irradiation of γ-$LiAlO_2$ in the Oak Ridge Research Reactor (ORR) under well-defined conditions of temperature, neutron flux, sweep gas flow, and configuration. Tritium, in its various chemical forms (and other gases evolved from the impurities in $LiAlO_2$ during irradiation) is moved by a sweep gas to an analytical train in which measurements on the composition of the effluent are performed. Previous reports have described the experimental configuration and some initial results.[1-4]

The primary purpose of the experiment was to test in-situ tritium recovery. To this end, the design made every attempt to provide conditions favorable for tritium release. $LiAlO_2$ was selected because of its chemical inertness, low hygroscopicity, good thermal stability, high melting point,[5] excellent stability under neutron irradiation,[6,7] and fabricability.[8] The breeder pellets were fabricated with an engineered microstructure, having a bimodal pore size distribution [5] with grains of 0.1 μm radius compacted into agglomerates ~50 μm in diameter. The overall density was 65% of theoretical. Fabrication and characterization of the pellets is described elsewhere.[1,2] The importance of obtaining quantitative values for both integral and real-time data on tritium production and tritium release was recognized. Wherever possible, a number of independent methods were used to determine such quantities.

The irradiation phase of the experiment was completed in June 1983 and post-irradiation examinations are underway. Significant results to date, and the implications thereof to blanket design, are presented.

Experimental

Full-power operation of the experiment began on March 12, 1983 and ended on June 13, 1983. During this period 33 tests, or runs (Table 1), were conducted. Observations made during testing have been discussed.[1] Presented below are recent results.

Neutron Flux

The thermal neutron flux incident upon the capsule was continuously recorded with three self-powered neutron detectors, as illustrated in Fig. 1.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

TABLE 1
Test Matrix for TRIO-01

Run No.	Time (Days)	Nominal[a] Temperature (°C)	Changes in Nominal[b] Sweep Gas Composition
1	2	600°	-
2	2	700°	-
3	1	700°	He/1.0% H_2
4	5	550°	-
5	2	500°	-
6	1	550°	-
7	3	600°	-
8	2	650°	-
9	6	650°	100% He
10	1	400°	100% He
11	1	500°	100% He
12	4	550°	100% He
13	5	600°	100% He
14	2	700°	100% He
15	1	700°	-
16	1	700°	300 cc/min
17	3	700°	30 cc/min
18	1	700°	-
19	1	650°	-
20	4	650°	He/0.2% O_2
21	2	650°	-
22	1	600°	-
23	3	550°	-
24	1	600°	-
25	2	560°	-
26	3	550°	-
27	7	525°	-
28	7	500°	-
29	3	480°	300 cc/min
30	4	480°	-
31	2	500°	-
32	2	550°	-
33	3	650°	-

[a]Average temperature on west side. Coldest temperature is ∿100° lower, hottest temperature is approximately 50° higher.

[b]Nominal sweep gas is 0.1% H_2 in He, at a flow rate of 100 cc/min.

The flux levels were about 30% lower than the levels in a core mockup test, and showed considerable variations. These variations in neutron flux were considerably greater than variations in the ORR power level. These flux data are being used to calculate tritium production rates during the experiment.

Capsule Temperatures

Temperatures were monitored continously with ten thermocouples, located on the inside and outside surfaces of the breeder ring. The ability to achieve temperature control was essential to

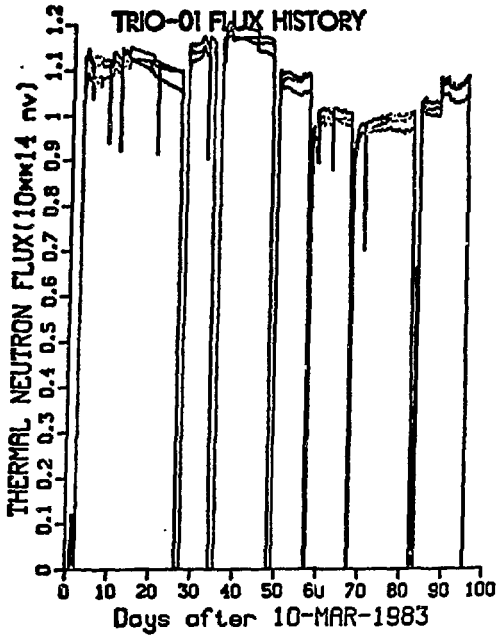

Fig. 1. TRIO-01 Flux History

successfully conducting the 33 runs (Table 1). The temperature history is shown in Fig. 2. From these data heat transfer analysis was performed and the key results were:[4]

1) The capsule performed well throughout the course of the experiment, and the design models accurately predicted temperature profiles.

2) One side was about 50°C colder, presumably owing to gap asymmetry. PIE results confirmed this.

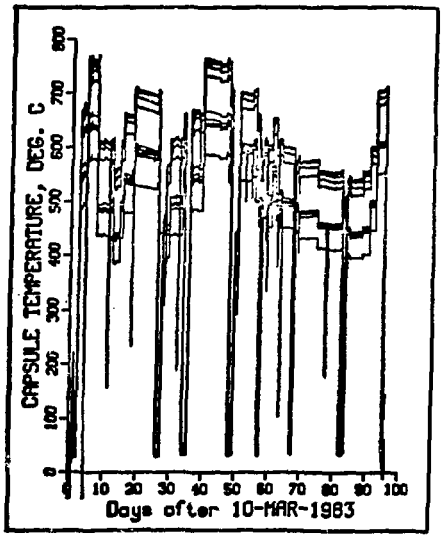

Fig. 2. TRIO-01 Capsule Temperatures

3) Irradiation to a fluence of 2×10^{21} nvt - equivalent to about 0.6 MW/y in STARFIRE [5] had no detectable effect upon either heat transfer coefficients or thermal conductivity. It is noted that thermal conductivity changes were expected to be significant, possibly as much as a 50% decrease, at these dose levels. The fact that the thermal conductivity did not change is a significant and positive result for solid breeder blankets.

4) The radial temperature gradients systematically decreased, as nuclear heating decreased, owing to burnup of ^6Li. The ^6Li burnup was calculated to be 0.18%.

Tritium Recovery

During irradiation, tritium in its various chemical forms was collected. The results are summarized in Table 2. The predominent form, in the sweep gas being about 94% of the total, was the noncondensable or HT form. The condensable (HTO) form accounted for about 4% of the total and most of the HTO was evolved after Run #20, when 0.2% O_2 was added to the sweep gas. A total of 0.6 Ci or about 2% of the tritium was collected in the gap gas. The total tritium recovered was about 35 Ci. This would correspond to a ^6Li burnup of 0.18%.

TABLE 2
Total Tritium Collected

Form	Curies	Percent of Total
Molecular (HT,T_2) (noncondensible)	32.6	94%
Oxide (HTO,T_2O) (condensible)	1.5	4%
Permeation into gap	0.6	2%
TOTAL	34.7 Recovered	

Two samples of $LiAlO_2$ breeder specimens were analysed for tritium. The residual content was 0.02 and 0.05 wppm tritium in the two samples. From these preliminary results, the residual content is less than 0.1 wppm following run number 33, which had a nominal temperature of 650°C. This result shows that nearly all the tritium produced during irradiation was released. More specimens of $LiAlO_2$ are being analyzed for tritium in order to obtain more accurate values.

Post-Irradiation Examinations

Samples of the irradiation pellets of $LiAlO_2$ are being investigated by scanning electron microscopy. A preliminary survey indicate that the microstructure has not significantly changed as a result of test conditions. Grain size appears to be 0.1 μm in radius in the irradiated specimens.

Integral Results and Dosimetry

In addition to tritium accounting and thermal analysis, two more methods were used to determine integral quantities such as total tritium production and lithium burnup, namely Li-6 assay and dosimetry. Li-6 assay was determined by mass spectrometry. The irradiated material showed a residual

Li-6 content of 0.38% and 0.38% in two samples. Since the initial content was 0.55% lithium-6, the burnup as determined by mass spectrometry is 0.17%. The accuracy of that number is approximately 10% of the nominal value.

Neutron doximetry measurements were conducted in the ORR in order to measure the flux and energy spectrum as well as ^{6}Li burnup reaction rates. Measurements were also made during the TRIO irradiation in order to determine the neutron exposure, ^{6}Li burnup, and radiation damage. The spectral measurement was performed in the A2 position for about nine hours on April 26, 1982. Twenty-one different neutron activation rates were measured, including ^{6}Li monitors, fission detectors, and thermal covers. The STAYSL computer code was then used to adjust calculated neutron spectra to fit the activation measurements. Selected reaction rates were measured over the vertical height of about 60 cm at eight different radial positions, four outside and four inside of a dummy TRIO sub-assembly. The ^{6}Li burnup rate was measured by ORNL and RIES and calculated from the dosimetry results. The measurements and calculations agreed with $\pm$4%. These results are reported in [9].

Dosimetry measurements were also performed during the actual TRIO irradiation in the A2 position of ORR from March 10 to June 13, 1983 for an exposure of 2558 MWD. Fe, Ni, and Ti monitor wires were coirradiated in separate dosimetry tubes on the west side, east side, and center of the TRIO capsule. The 10 mil wires were cut into six pieces, each about 1.4 cm, and gamma counted by Ge(Li) spectroscopy. The reaction rates were corrected for decay during the irradiation using the flux history measured by the self-powered neutron detectors.

The measured reaction rates are listed in Table 3. As can be seen, the thermal flux (indicated by the 58Fe(n,g) reaction) is about the same on the east and west sides, but about 32% lower in the center of the assembly due to self-shielding effects caused by the stainless steel and Li compounds, as discussed in detail below. It is interesting to note that the activity and flux levels are significantly lower (30-40%) than in the previous mockup measurements. The measured reaction rates were used to adjust the neutron spectrum measured in the mockup tests with the STAYSL computer code. The resulting fluence values are listed in Table 4. The fast flux gradients are rather steep (100% from west to east); however, this has little effect on the tritium rates, only the fast damage rates.

TABLE 3
Measured Activation Rates Results are Normalized to 30 MW. Values are averages of six samples.

Position	58Fe(n,g)	54Fe(n,p)	46Ti(n,p)
West	1.24E-10	5.34E-12	7.47E-13
Center	8.31E-11	3.42E-12	5.26E-13
East	1.22E-10	2.67E-12	3.82E-13

TABLE 4
Adjusted Neutron Fluence Values

Energy, MeV	Neutron Fluence, E20 n/cm^2 Center	West	East
Total	19.1	28.7	19.7
Thermal (>.5 eV)	6.93	10.30	9.52
(2200 m/s)	6.14	9.13	8.44
0.5 eV - 0.1 MeV	6.01	8.98	5.13
Fast, >.1 MeV	6.15	9.05	5.00

The lithium burnup rates were then computed using the adjusted neutron spectrum. The outer rate was found to be 9.7E-08 atoms/atom-s while the inner rate was 6.5E-08 atoms/atom-s. These two values were used to determine the total burnup using an iterative procedure to calculate the time-dependent burnup and resultant decrease in self-shielding. The stainless steel sleeve was about 0.5 cm thick resulting in a calculated neutron absorption loss factor of about 0.69 using an analytical approximation for self-shielding. This effect was estimated to be about 0.74 using our previous flux measurements in the mockup experiment. The initial lithium self-shielding is about 0.952 decreasing to about 0.968 by the end of the irradiation. These two factors would predict an average lithium burnup rate of 5.87E-08 atom/atom-s. Over the 97 days of the run this would result in a net burnup of 37.7% of the initial ^{6}Li atoms and a net tritium activity of 39.2 Ci. Due to the uncertainties in the fluence measurements and self-shielding approximations, the calculated tritium values have an estimated accuracy of 10-15%. If we use the measured Li-to-iron activity ratios from the mockup experiment, the estimated tritium level would be lowered to about 37.5 Ci.

On the basis of lithium burnup, the radiation damage is equivalent to 0.6 MW-yr, or about two months in STARFIRE.[5] The neutron dose was calculated to be 4.9 x 10^{12} Rads. The gamma dose was calculated to be 3.5 x 10^{12} Rads, based on a gamma heating rate of 4.8 W/g. The total dose is about 8.4 x 10^{12} Rads - equivalent to about four months in STARFIRE.[5]

The integral data, as determined by four independent methods, is shown in Table 5. The agreement between the various methods is quite good - all are within experimental error. The error in the mean values is thus reduced about 5%. These results demonstrate that the tritium is accounted for.

Work in Progress

A number of tasks are in progress or planned, including:

1) Calculations of tritium inventory (or content) in the $LiAlO_2$ for the 33 test runs. This data will then be correlated to test conditions, notably temperature.

2) Evaluation of the dynamic data on tritium release to provide information on transport mechanisms.

TABLE 5
Summary of Results - Integral Tritium Data

Method	Tritium, Ci	% Burnup (^{6}Li)	% Burnup (Li)	Estimated Error
Tritium Collected	[35]	33	0.18	5%
^{6}Li Assay	32	31	[0.17]	10%
Thermal Gradient	35	[33]	0.18	10%
Dosimetry	39	[38]	0.21	10-15%
Average	35	33	0.18	5%

[ ] - Quantity directly measured or determined by the method.

3) Evalution of permeation rates as a function of test conditions.

4) Thermal analysis to provide more precise values of thermal conductivity and heat transfer coefficients.

Discussion

The experiment sought to test the feasibility of in-situ tritium recovery under conditions which simulate a fusion reactor blanket. The results, although for rather short irradiation times and burnup levels, are very positive, showing that inventories could be very low, <0.1 wppm, in a solid breeder blanket. This would be an inventory of less than 100 g in STARFIRE [5]. It is noted that inventories were apparently higher at lower temperatures. However, concerns raised during the STARFIRE study, which indicated that radiation damage could possibly substantially increase tritium inventories, appear not to be serious. Heat transfer results demonstrate that design codes work well [4] and that thermal conductivity is not significantly changed under the test conditions. Thus, for conditions of limited radiation damage, it appears that $LiAlO_2$ performs well with respect to tritium recovery and heat transfer.

The results for $LiAlO_2$ are believed to give some confidence that chemically similar materials will also perform well. Such materials are the ternary lithium oxides: lithium zirconates, lithium silicates, etc. Li_2O is considered to be somewhat different chemically because it is more reactive. Preliminary results from sealed-capsule tests of Li_2O show tritium retention values of ~10 wppm at about 900°C and ~100 wppm at about 500°C [7] for neutron doses similar to TRIO-01. In the Tulip experiment [10], sealed-capsule tests of Li_2O showed tritium retention values of about 100 ppm at 600°C. It is likely that tritium release would be better in an open capsule. A TRIO-type test with Li_2O would help to better quantify tritium inventories for this breeder material.

Conclusions

Although much analysis work remains, some significant conclusions can be made at this time:

1) It was demonstrated that nearly all the tritium was released and recovered.

2) The residual content of tritium in the $LiAlO_2$ was <0.1 wppm, following a test run at a nominal temperature of 650° with 0.1% H_2 in the sweep gas.

3) Irradiation to a ^6Li burnup of 0.18% (equivalent to 0.6 MW-y/m^2) did not appear to change thermal conductivity or microstructure of the $LiAlO_2$.

Acknowledgments

The authors wish to express their appreciation for the support of the Office of Fusion Energy, U.S. DOE. Many people have contributed to the technical success of this project. We wish to thank V. A. Maroni, C. C. Baker, M. J. Steindler, and J. R. Conlin. We wish to thank L. J. Jensen and R. T. Riel for preparation of this manuscript.

References

[1] R. G. Clemmer, P. A. Finn, M. C. Billone, B. Misra, R. M. Arons, R. B. Peoppel, F. F. Dyer, I. T. Dudley, L. C. Bate, E. D. Clemmer, J. L. Scott, J. S. Watson, and P. W. Fisher, "The TRIO-01 experiment: In-situ tritium recovery results," presented at the Third Topical meeting on Fusion Reactor Materials, Albuquerque, New Mexico, September 19-22, 1983. To be published in the Journal of Nuclear Materials.

[2] R. G. Clemmer, R. F. Malecha, and I. T. Dudley, "The TRIO-01 experiment," Nuclear Technology/ Fusion, vol. 4, p. 83, September 1983.

[3] P. A. Finn, G. A. Reedy, M. I. Homa, R. G. Clemmer, G. Pappas, M. A. Slawecki, D. G. Graczyk, D. L. Bowers, and E. D. Clemmer, "The design, fabrication, and testing of the gas analysis system for the tritium recovery experiment TRIO-01," to be presented, Philadelphia, Pennsylvania.

[4] B. Misra, R. G. Clemmer, and P. A. Finn, "Heat transfer performance of a solid breeder blanket comparison of TRIO-01 experimental data with model," ANS Transactions, 45, 176 (1933).

[5] C. C. Baker, et al., " STARFIRE—A commercial tokamak fusion power plant study," Argonne National Laboratory Report No. ANL/FPP-80-1, 1980.

[6] L. Yang, et al., "Irradiation study of lithium compound samples for tritium breeding applications," GA Technologies Report, GA-A16047, July 1981.

[7] G. W. Hollenberg, et al., "A fusion solid breeder experiment in a fast neutron fission reactor," in Proceeding of the Conference on Fast, Thermal, and Fusion Reactor Experiments, April 12-15, 1982, American Nuclear Society, La Grange Park, Illinois.

[8] R. B. Poeppel, Argonne National Laboratory, private communication.

[9] L. R. Greenwood, "Dosimetry results for the TRIO test in ORR," Damage Analysis and Fundamental Studies Quarterly Progress Report, DOE/ER-0046/10, pp. 9-18, August 1982.

[10] D. L. Porter, et al., "Irradiation and compatibility testing of Li_2O materials at EBR-II," Argonne National Laboratory Report No. ANL/FPP/TM-167, December 1982.