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Journal of Nuclear Materials 281 (2000) 71–79

journal of nuclear materials

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Performance of a Li_2TiO_3 pebble-bed in the CRITIC-III irradiation

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Received 17 December 1999; accepted 23 March 2000

Abstract

Lithium metatitanate (Li_2TiO_3) is a candidate material for tritium breeding in fusion reactor pebble-bed blankets. 173 g of Li_2TiO_3 pebbles were irradiated for 334 full power days (FPD) to a burnup of 0.9% ^6Li in the CRITIC-III experiment in AECL's NRU reactor. A key objective was to determine tritium release over a wide temperature band from 200°C to 900°C. On-line release and temperature measurements are reported in this paper. New analytical methods led to calculated inventories ranging from 15 wppm average at the lowest temperature of operation (200°C outer surface to 700°C inner surface) to less than 1.2 wppm average at 375°C outer-surface temperature and 875°C inner-surface temperature. The thermocouples indicated that the bed remained stable during the irradiation, which included thermal shocks from 90 reactor shutdowns. From this swept-capsule irradiation, Li_2TiO_3 appears to be a good candidate for fusion blanket pebble-beds. Crown Copyright © 2000 Published by Elsevier Science B.V. All rights reserved.

1. Introduction

Lithium metatitanate (Li_2TiO_3) is being studied as a potential material for ceramic tritium-breeding blankets. Its property database was summarized by Gierszewski [1]. As a relatively new candidate, there have been few in-reactor vented-capsule tests. Results from the EXOTIC-8 irradiation, begun after CRITIC-III, have been reported [2]. CRITIC-III used the same vented capsule and on-line measurement of temperature, tritium release, moisture and neutron flux, as was used in the CRITIC-II irradiation of Li_2ZrO_3 [3]. The Li_2TiO_3 was in the form of 1.2 mm pebbles in a packed bed. A major objective was to operate at low temperatures (200–400°C) in the periphery of the pebble-bed to determine tritium release capabilities at these low temperatures. The performance of Li_2TiO_3 during irradiation is described here – post-irradiation examination (PIE) was not done and is not planned.

2. Experiment

The test capsule (see Ref. [2]) was placed in an irradiation test-loop in the AECL Chalk River NRU reactor, a heavy-water moderated thermal reactor with a neutron flux of about 2×10^{18} nm^{-2} s^{-1} at the location of the capsule. Coolant in the test-loop was light water at about 30°C. NRU is re-fuelled with the reactor at full power (about 3 four-rod shifts per week); this, together with control-rod movements, results in a neutron flux at the capsule that undergoes both slow and rapid changes within a band of about ±8% of nominal full power. Since pebble-bed heating is mainly due to the $^6Li(n,\alpha)^3H$ reaction, the temperature of the pebble-bed undergoes many small fluctuations.

Li_2TiO_3 powder was made by solid-state reaction of TiO_2 and Li_2CO_3. Pebbles were fabricated [4] by a process of dry ball-milling, followed by extrusion/tumbling. This process had been previously developed for Li_2ZrO_3 [5]. The pebbles were XRD phase-pure, approximately 1.2 mm average diameter, with 18.5% porosity (36% of which was closed porosity, 64% open). Grain size of the pebbles was not measured, but fracture surface photos suggest that the grains were 10–100 μm,

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PII: S0022-3115(00)00057-X

Table 1
CRITIC-III irradiation test conditions

Time from start of test		He–Ar gap-gas composition[a] (%He)	Lowest temperature of pebble-bed (°C)
Calendar days	Full power days		
0	0	91–92	200
5	5	31–36	360
88	71	100	200
117	97	75–80	250
308	230	55–58	290
361	275	34	360
386	290	56–50	240
444	334	Irradiation terminated	

[a] Exact composition varied somewhat during each period. For example, gap gas drifted slowly from 31% to 36% He over the 5–68 FPD period, and then stayed at 32–33% for 68–71 FPD.

and that the closed porosity was in the form of uniformly distributed irregularly shaped bubbles of a few micrometers diameter. Surface area, measured by multipoint BET, was 0.069 m^2/g. The initial depleted Li_2CO_3, obtained from Martin Marietta Corporation, was quoted as 98.330 at.% Li-7. Measurements at Argonne National Laboratories [1] on the as-fabricated pebbles reported 98.154 ± 0.010 at.% Li-7, or, equivalently, 1.85 at.% Li-6. This value was used in calculations described below.

The pebble-bed was cylindrical, with dimensions 38 mm OD, 10 mm ID and 90 mm in height. It was loaded with 173 g of Li_2TiO_3, resulting in a smear density (overall density) of 53% of theoretical density (TD). Neutron flux depression, due to neutron absorption in the ^6Li, between the outer surface of the pebble-bed and the centre was calculated to be between 15% and 20% at startup.

The temperature of the pebble-bed was monitored by eight thermocouples. Two self-powered neutron flux detectors (SPNDs) were wound around the capsule exterior and used for on-line measurement of the local flux at the capsule. Flowing He or He–Ar gas mixture was used in a narrow space (0.18 mm) surrounding the pebble-bed to control the temperature.

He with 0.1% H_2 sweep gas (-80°C to -75°C H_2O dewpoint, i.e., <1.3 ppm H_2O at inlet) at atmospheric pressure flowed through the bed at 95 ml(STP)/min, carrying the released tritium to an analysis and removal system. The sweep gas line (6.35 mm stainless steel tube) from the top of the reactor to the tritium analysis equipment (about 50 m) was heated to 250°C; piping was unheated for about 2–3 m in the reactor core from the top of the capsule to the top of the reactor. Total tritium content in the sweep gas was measured using an ionization chamber (IC) with 120 ml active volume. Calibration of the IC was done prior to the irradiation using a known concentration of tritium in a carrier gas. The uncertainty in the calibration is estimated at $\pm 10\%$. The option of flowing the gas stream through ethylene glycol bubblers to collect and measure tritium was used rarely. Tritium buildup on the IC wall was minimized by adding humidified He (15 ml/min) to the sweep gas stream just before it flowed into the ion chamber (see Ref. [2]). The time constant for gas to sweep through the line from capsule to ion chamber was about 15 min.

The intent of the test matrix was to perform only a few temperature change tests in the 15 months of the irradiation, maintaining constant test conditions for long periods to observe long-term behaviour. The test program is summarized in Table 1, showing the changes to the He–Ar gap-gas concentration, and the resultant changes in the ceramic temperature.

The total irradiation time was about 334 full power days (FPD) between October 1996 and January 1998. Approximately 90 reactor shutdowns subjected the pebble-bed to many thermal shocks – many shutdowns required less than 5 s to go from full power to zero power.

At a nominal 20 Ci/m^3 tritium concentration in the sweep gas, the tritium partial pressure was about 2 Pa, the H:T ratio was about 100, and the tritium generation rate in the ceramic was 2×10^{-5} wppm/s.

3. Results and discussion

3.1. Temperatures

Temperature of the pebble-bed was monitored via eight thermocouples, four on the interior (hot) surface of the pebble-bed, and four around the exterior (cooler) surface. For convenience, the thermocouples on the inside of the pebble-bed annulus are referred to as 'hot' thermocouples, and those on outside are referred to as 'cold'. All thermocouples were tack-welded to the

[1] Courtesy of D.L. Bowers, Analytical Chemistry Laboratory, Chemical Technology Division, ANL.

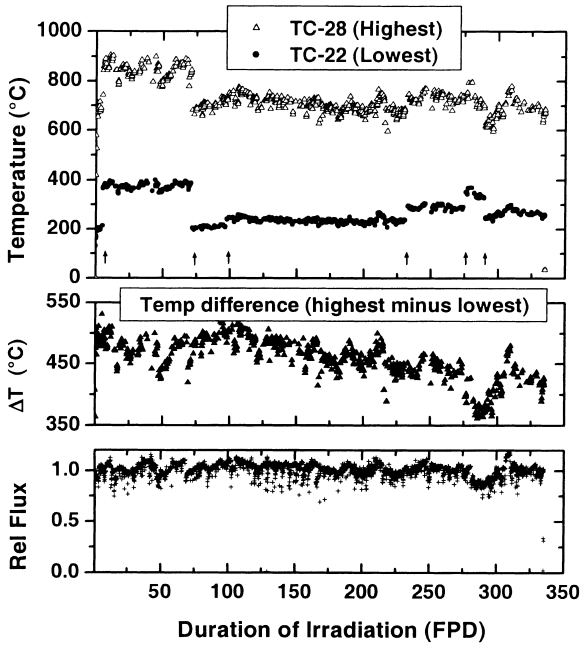

Fig. 1. Maximum and minimum breeder temperatures, temperature difference, and relative neutron flux at the CRITIC-III capsule. Arrows in top part show the intentional temperature changes induced by gap-gas concentration changes shown in Table 1; other temperature fluctuations are due to variations in the neutron flux.

outside of the metal cage holding the pebble-bed. The two hot thermocouples at the mid-plane of the pebble-bed annulus indicated the same temperature (within 2°C), while the other two hot thermocouples, located mid-way to the top and bottom were typically 60–80°C and 40–60°C lower, probably due to end effects. Three of the cold mid-plane thermocouples varied systematically from each other; one was always 20–30°C hotter and one 40–50°C hotter than the third. (Data from the fourth cold thermocouple was used as an alarm thermocouple and not logged digitally.) These differences are probably due to small azimuthal differences in the size of the gas gap. Fig. 1 shows the temperature for the whole experiment as measured by mid-plane thermocouples TC-28 (interior surface of pebble-bed annulus) and TC-22 (exterior surface), as well as their temperature difference (for bed stability and thermal modelling, Sections 3.5 and 3.6), and neutron flux.

3.2. Moisture release

Lithium ceramics have a tendency to absorb moisture, and moisture can affect the rate and form of tritium release. Therefore, after loading the Li_2TiO_3 pebbles into the capsule in air, the assembly was pre-dried in flowing He at about 250°C. After the start of the irradiation, the moisture levels in the sweep gas were observed to increase from the background −67°C dew point (4 appm) to a peak of around −40°C (140 appm), and then return to below −60°C (14 appm) within one day and below −70°C (2.5 appm) within 20 days. Although humidity levels did vary during subsequent changes in capsule temperature or reactor operation, they were always low. No significant moisture peaks were observed along with the tritium release peaks during temperature change experiments. This low moisture release is similar to what was observed in Li_2ZrO_3 irradiation experiments [3,6].

The tritium production rate is equivalent to about 20 appm in the sweep gas. This is much larger than the measured moisture level (<2.5 appm at <70°C dew point), so we conclude that tritium was mostly released as HT. (It is possible that HTO could have been reduced to HT on the stainless steel gas line between the reactor and the measuring system, but probably only in trace amounts, given the low temperature, 250°C, and short transit time, 15 min.)

3.3. Tritium release

Fig. 2 shows the measured tritium release rate for the whole irradiation. The total tritium recovered from the capsule during irradiation was 830 ± 85 Ci as measured by the ion chamber. This gives a total lithium burnup of $0.9 \pm 0.1\%$.

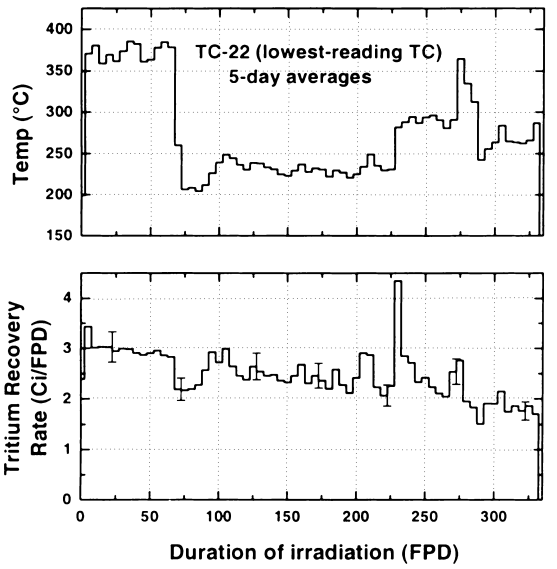

Fig. 2. Tritium recovery rate for the whole irradiation. The uncertainty is ±10% due to IC calibration uncertainty (a few error bars are shown). Temperature measured by the lowest reading thermocouple (located at mid-height on the outside of pebble-bed) is also shown. Data have been smoothed by using 5-day averages.

The release rate is shown in 5-day averages to show trends rather than details associated with the many small flux changes of the NRU reactor. The general decline in tritium release over the course of the experiment is due to burnup. The other variations in the tritium recovery rate in Fig. 2 are presumed to be due to buildup of tritium during operation at relatively low ceramic temperatures and subsequent release at higher temperatures.

3.4. Tritium inventory

A major reason for performing irradiations on candidate fusion blanket materials is to determine tritium inventory in the ceramic as a function of temperature and time. This is best determined by direct measurements of the inventory in small sections of the pebble-bed during PIE, as was done for the CRITIC-II Li_2ZrO_3 [7].

Since no PIE was done, the tritium generation rate was calculated and used with the measured recovery rate to provide an inventory as a function of time. This was done as follows (see Appendix A for details):

- a calculation was made of how the generation rate varied with time and burnup, taking into account neutron self-shielding by the ^6Li;
- the total tritium recovered (as measured by the IC) was added to a first estimate of the final inventory in the ceramic (i.e., at the end of the irradiation) to obtain the total tritium generated;
- the calculated generation rate was normalized (multiplied by a constant factor) so that the integral over the whole irradiation gave the same total tritium generated as that determined above;
- the inventory as a function of time was determined by integrating the difference between the generation rate and the recovery rate;
- the process was iterated with alternative estimates of the final inventory to obtain physically reasonable results – non-negative inventories at all times, and constant or decreasing inventories when operating at the highest temperatures;
- the whole process was repeated with different IC calibration factors (within the ±10% experimental uncertainty of the original value).

Figs. 3 and 4 show the results. Fig. 3 shows the recovery rate, generation rate and estimated inventory assuming 10 Ci final inventory and an IC calibration factor 10% below the value originally measured. Fig. 4 shows alternative possibilities. The one chosen for use in Fig. 3 is one that does not exhibit a negative inventory at any time and has a constant or decreasing value for the period of high-temperature operation between 5 and 75

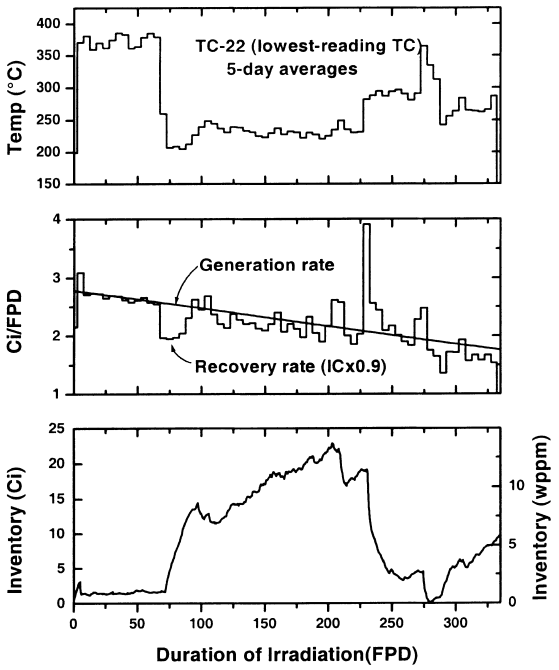

Fig. 3. Estimated tritium inventory versus time (bottom). The top figure shows the temperature as measured by the lowest reading thermocouple (placed at the outside of the pebble-bed at mid-height). The middle figure shows measured tritium recovery rate and the generation rate under the assumptions of 10 Ci final inventory and IC calibration 10% lower than that determined in the laboratory prior to the start of the irradiation. This is within the uncertainty for the IC calibration.

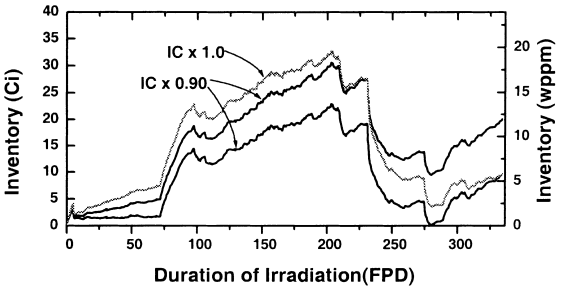

Fig. 4. Estimated tritium inventory versus irradiation time, showing the effects of altering the assumed final inventory (10 or 20 Ci) and of IC calibration coefficients equal to, and 10% below that determined prior to the irradiation. IC calibration coefficient 10% larger would lead to negative inventories.

FPD. This is consistent with the behaviour of Li_2ZrO_3 measured by PIE in CRITIC-II [7], which showed very low inventories (< 0.2 wppm below 300–400°C). However, Fig. 4 shows that other possibilities within experimental uncertainty are possible.

Data in Fig. 3 are summarized in Table 2. (Note that most of the tritium inventory at any time is expected to be in the outer cooler portions of the pebble-bed.) The data between 97 and 230 FPD and from 290 to 330 FPD imply that at about 250°C, small variations in temperature lead to rather large changes in inventory. At temperatures of over 350°C, the estimated inventory was small – about 1.5 Ci (or about 0.8 wppm average for the whole bed).

Table 2
Estimated tritium inventory versus time and temperature; summary of Fig. 3

FPD	TC-22 (°C)	Comments on tritium inventory
0–5	200	Total inventory rose rapidly to 3 Ci before temperature increase
5–75	360–380	Inventory reasonably steady at about 1.5 Ci
75–97	~200	Rapid inventory rise – terminated by temperature increase
97–205	250–230	Inventory decreased initially (at 250°C), then continued a slow steady rise
205–230	250, 230	Period at 250°C led to decline in inventory; resumption at 230°C resumed the inventory rise
230–275	290–300	Inventory dropped from about 18 to 5 Ci, then rose to 6 Ci
275–290	360 decreasing to 310	Inventory dropped to 0–1.5 Ci
290–330	~260	Inventory rose slowly to 10 Ci

Fig. 3 indicates that after an initial brief rise at low temperature in the first 5 FPD, the inventory decreased and remained constant at 0–2 Ci (0–1.2 wppm average for the whole pebble-bed) best estimate for about 75 FPD during high-temperature operation (375°C at the outside surface to 875°C centre temperature). When the temperature was dropped to 200–225°C, after 125 FPD, the estimated inventory rose to its maximum of about 20 Ci (12 wppm average for the whole bed). The estimated inventory was still rising at this temperature when the temperature was again raised to about 370°C for a period of about 20 FPD. The estimated inventory fell to near zero, before rising again as the temperature was decreased and held approximately constant to the end of the irradiation.

3.5. Bed stability

The measured temperatures can be used to infer information about the bed physical stability and thermal conductivity. Cracking, settling or sintering would affect bed thermal conductivity and, therefore, bed temperatures.

The best experimental test of stability is the temperature difference between the outer and the inner surfaces at the in-breeder mid-plane since this parameter (rather than, say, breeder maximum temperature) is not directly dependent on variations or uncertainties in gas-gap dimensions and composition. This breeder temperature difference (see Fig. 1) shows an overall decrease of about 20% over the course of the irradiation, with short-term variations superimposed. The overall drop is that expected due to burnout of ^6Li (i.e., reduction in ^6Li transmutation rate), while the short-term variations are largely due to daily variations in the neutron flux. These results qualitatively indicate the absence of crumbling, settling or sintering of the pebble-bed.

Furthermore, changes in bed structure would likely affect different parts of the bed differently, and lead to changes in the temperature differences between the mid-plane hot thermocouples and the hot thermocouples mid-way to the ends of the pebble-bed stack. This was not observed. Further evidence of bed stability is provided in Section 3.6.

3.6. Thermal modelling

Based on the thermocouple readings and thermal modelling of CRITIC capsules, we expect that the heat transfer is largely radial at mid-plane, with cooling due to end effects becoming significant in the top and bottom quarters of the capsule. Therefore, we compared the mid-plane breeder temperature difference with a 1-D

radial model. This 1-D model used a single average bed thermal conductivity and assumed uniform heating across the ceramic. Heating in the ceramic is mainly due to two sources – gamma-photon absorption, called gamma heating, and neutron absorption by ^{6}Li to produce ^{3}H and ^{4}He.

Accurate prediction of the minimum breeder temperature is complicated by the inevitable small azimuthal variations in the gas-gap dimensions and uncertainties in heat transfer coefficients between pebble-bed and capsule wall. But the 1-D model is expected to reasonably predict the breeder temperature difference if the heating rates and pebble-bed thermal conductivities are reasonably accurate, and the bed is stable.

In fact, the 1-D model was able to match the measured mid-plane temperature difference over the course of the irradiation within 10% with the following assumptions:

- gamma heating rate of 1 W/g average (within the range of uncertainty of values estimated from other irradiations in NRU);
- gamma heating is proportional to the neutron flux;
- breeder-capsule heat transfer coefficient of 2000 W/m^2 K;
- bed thermal conductivity increased uniformly by a factor of 1.15 from Ref. [1] prediction;
- the tritium generation rate (i.e., one of the heat sources) was affected by self-shielding and followed the form as indicated in Fig. 3 (i.e., using an IC calibration 10% lower than the pre-irradiation determination).

Therefore, we conclude first that the bed thermal conductivity predicted in Ref. [1] underestimated that calculated from the data of this irradiation by 15%, and secondly that the thermal conductivity varied by less than 10% during the irradiation. The second conclusion also provides additional evidence of the stability of the bed during irradiation.

3.7. Temperature step-increase tests

Three temperature step-increase tests were performed by changing the He–Ar gap-gas proportions. Each temperature increase initiated a step-increase in the tritium release, followed by a slow decrease. Figs. 5 and 6 show two of these tests (background noise due to NRU power changes interfered with the third and is not shown). Due to subsequent neutron flux changes in NRU, and hence tritium-production changes, correlation of the tritium release rate to the temperature change test was not possible after 2–3 days. Moreover, different regions of the lithium titanate pebble-bed operated at very different temperatures, so it is not clear precisely what the physical meaning of these time constants is. Nevertheless, estimates of time constants have been made by fitting the initial decay of the tritium release peak to a decreasing exponential (Table 3). There may also be a longer time constant associated with the coolest regions of the blanket, or due to slower release mechanisms that are masked by the subsequent neutron flux changes.

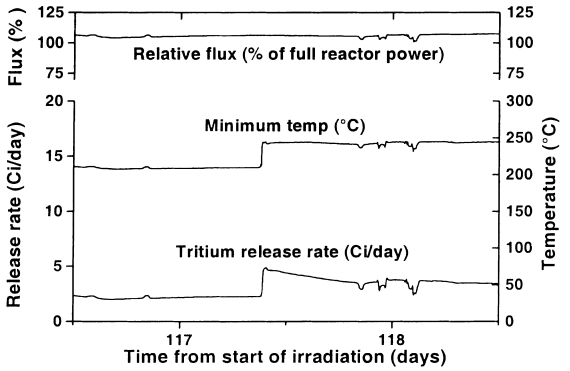

Fig. 5. Temperature increase test at constant flux after 117.4 days (97 FPD). Time shown is calendar days from experiment startup. Temperature shown is breeder outer mid-plane temperature (lowest reading thermocouple). Tritium release rate incorporates IC calibration factor 10% lower than that determined in the laboratory prior to start of the irradiation.

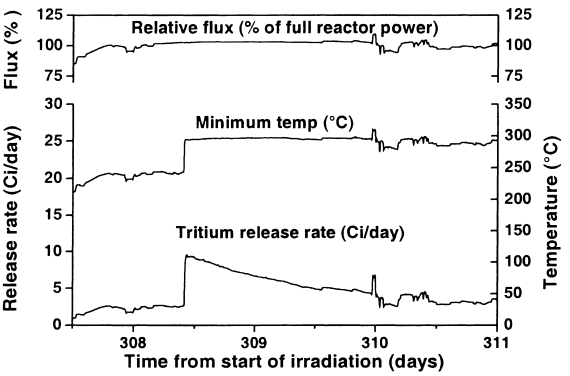

Fig. 6. Temperature increase test at approximately constant neutron flux after 308.4 days (227 FPD). See Fig. 5 caption for further details.

Table 3
Approximate time constants for three temperature step-increase tests

Temperature change (°C)	Date (FPD)	Time constant (FPD)
210–244	97	0.6–1.4
240–290	227	1.0–1.1
280–360	270	0.5–0.9

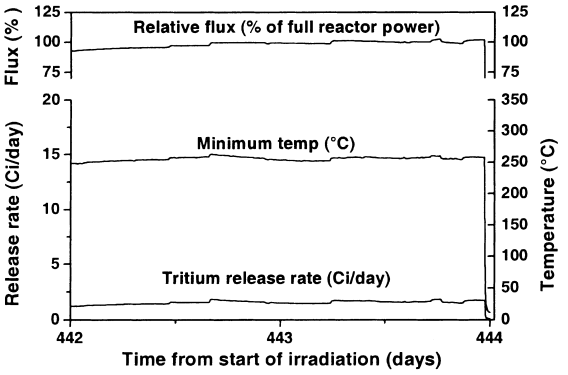

Fig. 7. Conditions just before final shutdown. See Fig. 5 caption for further details.

4. Shutdown

The irradiation was terminated in January 1998. Fig. 7 shows that the final two days were at relatively steady conditions and that the shutdown was fast (5 min to half reactor power, 10 min to 1/10). After shutdown, the sweep gas flow continued through the capsule for 2 days, after which the capsule was isolated, sealing the lithium titanate inside under He atmosphere, and removed to a storage block. Purging of the piping (exclusive of the capsule) continued for more than a week, including a period with increased H_2 concentration in the gas. Tritium was collected in bubblers during this time; total quantity was about 0.5 Ci.

5. Summary

CRITIC-III, the first irradiation of lithium titanate in a swept capsule, successfully demonstrated removal of tritium from a pebble-bed at temperatures as low as 200°C for a period of 334 FPD to a burnup of 0.9% of total Li (about 50% of the initial ^{6}Li). Temperature, tritium release, neutron flux, and moisture levels were recorded throughout the irradiation. Temperatures ranged from a low of about 200°C to a high of about 380°C at the outside surface of the pebble-bed; centre temperatures were about 500°C higher than outside surface. Due to program re-direction, this work has been concluded; PIE will not be performed.

Calculations of tritium inventory suggest the best estimate of 0–2 Ci (0–1.2 wppm average for the whole pebble-bed) when operating at the highest temperatures – 375°C at the outside surface to 875°C centre temperature; 5 Ci (3 wppm average) at 290°C outside surface temperature; and over 20 Ci (12 wppm average) after prolonged operation at the lowest outer temperature of about 225°C. Most of the tritium inventory at any time is expected to be in the outer cooler portions of the pebble-bed.

No evidence of pebble-bed disintegration could be seen from the temperature records. Temperatures are consistent with predictions for bed thermal conductivity. Thus, lithium titanate appears to be an excellent tritium-producing blanket material, capable of long-term operation with low tritium buildup at relatively low temperatures.

Acknowledgements

We thank H.B. Hamilton and P.M. Ryan for fabrication of the lithium titanate pebbles; L.R. Bourque for assembly of the capsule; L.K. McMillan for day-to-day management and operation of the irradiation and on-line tritium facility, and the AECL machine shops and reactor operators and many other personnel required to assemble and operate such a major experiment.

Appendix A. Tritium generation and inventory

This appendix calculates the tritium generation rate for an in-reactor irradiation of a lithium-containing ceramic based on the total tritium recovered, and demonstrates that this can be used to obtain a reasonable value of inventory in the ceramic as a function of time.

A.1. Case A: negligible self-shielding

When there is negligible neutron self-shielding from the ^{6}Li in the lithium ceramic, the neutron flux everywhere in the ceramic is constant in time (full power time) and, therefore, the tritium-generation rate is always proportional to the current amount of ^{6}Li present in the pebble-bed. The generation rate (per FPD) is

$$G(t) = kL(t), \tag{A.1}$$

where G is the tritium generation rate, t is time measured in FPD, $L(t)$ is the amount of ^{6}Li present in the bed at time t, and k is the proportionality constant.

The tritium generation rate is also equal to the rate of decrease of ^{6}Li (each atom of ^{6}Li that reacts with a neutron produces a tritium atom), i.e.,

$$G(t) = -\frac{\mathrm{d}}{\mathrm{d}t}L(t). \tag{A.2}$$

These two formulae mean that the amount of ^{6}Li decreases exponentially with (full power) time:

$$L = L_0 \mathrm{e}^{-kt}, \tag{A.3}$$

where L_0 is the initial amount of ^{6}Li in the ceramic.

If the amount of ^{6}Li is L_F at the end of the irradiation (time t_F), then the previous relation gives

$$k = \frac{1}{t_F} \ln\left(\frac{L_0}{L_F}\right) \tag{A.4}$$

and

$$G(t) = -\frac{dL}{dt} = kL_0 e^{-kt} = kL \tag{A.5}$$

showing how the generation rate varies with (full power) time.

A.1.1. Dependence on burnup

The fractional burnup of ^{6}Li, β, is defined as the amount of ^{6}Li consumed, divided by the initial amount:

$$\beta = \frac{L_0 - L(t)}{L_0}. \tag{A.6}$$

Rearranging,

$$L(t) = L_0(1 - \beta) \tag{A.7}$$

Eqs. (A.5) and (A.7) give

$$G = kL(t) = kL_0(1 - \beta) \tag{A.8}$$

or

$$G = G_0(1 - \beta), \tag{A.9}$$

where $G_0 = kL_0$ is the generation rate at the beginning of the irradiation.

A.1.2. Inventory

The tritium inventory in the ceramic as a function of time is given by the difference between the generation rate $G(t')$ and the release rate $R(t')$, shown in Fig. 2, integrated from the start of the irradiation to the time t:

$$I(t) = \int_0^t (G(t') - R(t'))\,dt' = L_0\left(1 - e^{-kt}\right) - \int_0^t R(t')\,dt'. \tag{A.10}$$

The unknown parameter in Eqs. (A.5) and (A.7) for $G(t)$ and $I(t)$ is k. If the final amount of ^{6}Li in the ceramic, L_F, were known, then k could be determined from Eq. (A.4). The amount of ^{6}Li at the end of the irradiation is equal to the initial amount minus that converted to tritium during the course of the irradiation:

$$L_F = L_0 - I_F - R_T, \tag{A.11}$$

where I_F is the final inventory of tritium in the ceramic, and R_T is the total tritium recovered during the irradiation (units are moles of tritium and moles of ^{6}Li). The final tritium inventory I_F is not presently known without PIE. However, because the final inventory, I_F, will likely be small compared to the total recovered ($\sim$ 830 Ci), it can be neglected for a first approximation; a more realistic value would then be determined from this first approximation, and the process iterated. These are the methods that would be followed for CRITIC-III except that the premise of Case A, negligible self-shielding, does not apply.

A.2. Case B: non-negligible self-shielding

When neutron self-shielding in the lithium ceramic is not negligible, the relation (A.1) in Case A is no longer true. A neutronics calculation is required to determine how the generation rate varies with time or burnup. Only the relative generation rate is required, $G(t)/G_0$, since it is best to normalize so that the total tritium generated in the irradiation equals the amount recovered plus the (postulated) final inventory. This ensures that the calculated final inventory agrees with the postulated final inventory.

For CRITIC-I [8], with 0.7 g ^{6}Li in Li_2O, a neutronics calculation using the WIMS-CRL code with the ENDF/B-V data base was done (R.E. Donders, private communication, 15 April 1988). The decrease in tritium generation rate with burnup is shown in Fig. 8. For CRITIC-III, with 0.34 g ^{6}Li in Li_2TiO_3, the dependence was derived by one of us (PG) using a 2-D model. In this model, the ceramic was divided into rings, and the incident neutron flux (appropriately weighted for each incident angle) was tracked to determine how many interactions occurred in each ring, based on the local ^{6}Li atom density. Account was made for neutron attenuation along the track. The change in ^{6}Li atom density profile was computed in time steps of 30 FPDs. As a

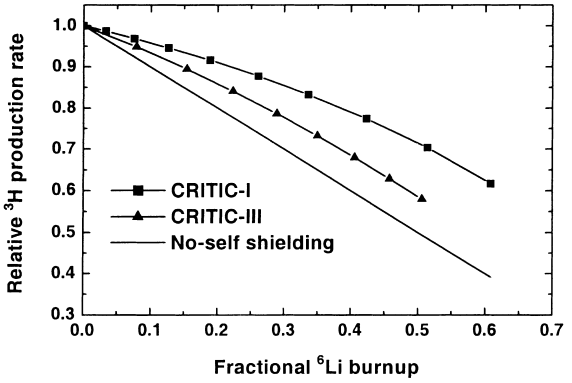

Fig. 8. Computed tritium generation rate, normalized to the initial generation rate, for CRITIC-I and for this irradiation, CRITIC-III. Note the deviation from generation with no self-shielding.

check, the method was applied to CRITIC-I and results similar to those of Donders were obtained. The results for CRITIC-III are also shown in Fig. 8. The effects of self-shielding are seen in the figure.

The generation rate for CRITIC-III can be reasonably fitted to a straight line:

$$G(t)/G_0 = 1 - 0.820\beta. \tag{A.12}$$

A second-order polynomial provides a slightly more accurate fit and was used, with the requisite changes to the math below, for the figures and calculations given in the main body of this paper. The linear fit described here is itself quite accurate, simpler and shows the methods.

We also have from Eq. (A.2), $G(t) = \mathrm{d}L/\mathrm{d}t = L_0\mathrm{d}\beta/\mathrm{d}t$. Therefore, the above formula can be rearranged to

$$\frac{G_0}{L_0}\mathrm{d}t = \frac{\mathrm{d}\beta}{1 - 0.820\beta} \tag{A.13}$$

and integrated to get:

$$t = \frac{-L_0}{0.820G_0}\ln(1 - 0.820\beta), \tag{A.14}$$

or

$$1 - 0.820\beta = \exp\left(-\frac{0.820G_0}{L_0}t\right). \tag{A.15}$$

Putting this in Eq. (A.12) gives

$$G(t) = G_0\exp\left(-\frac{0.820G_0}{L_0}t\right). \tag{A.16}$$

In Eq. (A.14), G_0, the initial generation rate, can be determined from the value of burnup, β_F, at the end of the irradiation, t_F,

$$G_0 = \frac{-L_0}{0.820t_F}\ln(1 - 0.820\beta_F). \tag{A.17}$$

These last two equations give the generation rate, linked to the total tritium produced, as a function of time.

A.2.1. Inventory

As with Case A, a value is assumed for the final inventory in the ceramic, to be added to the total measured release during the irradiation to get L_F (Eq. (A.11)) and thus, β_F. Fig. 4 in the main text shows the results.

References

[1] P. Gierszewski, in: Proceedings of the Fourth International Symposium on Fusion Nuclear Technology, Tokyo, Japan, 6–11 April 1997, Fusion Eng. Design 39&40 (1998) 739.
[2] J.G. van der Laan, R. Conrad, K. Bakker, N. Roux, M.P. Stijkel, in: B. Beaumont, P. Libeyre, B. de Gentile, G. Tonon (Eds.), Association EURATOM-CEA, Proceedings of the 20th Symposium on Fusion Technology, Département de Recherces sur la Fusion Contrôlée, CEA/Cadarache report, 1999, p. 1239.
[3] J.M. Miller, R.A. Verrall, in: Proceedings of the Sixth International Conference on Fusion Reactor Materials 1993, Stresa, Italy, J. Nucl. Mater. 212–215 (1994) 897.
[4] J.M. Miller, H.B. Hamilton, J.D. Sullivan, J. Nucl. Mater. 212–215 (1994) 877.
[5] J.D. Sullivan, et al., Fusion Eng. Design 17 (1991) 79.
[6] R.A. Verrall, O.D. Slagle, G.W. Hollenberg, T. Kurasawa, J.D. Sullivan, J. Nucl. Mater. 212–215 (1994) 902.
[7] R.A. Verrall, J.M. Miller, L.K. Jones, in: S. Casadio (Ed.), Proceedings of the Fifth International Workshop on Ceramic Breeder Blanket Interactions 23–25 September 1996, Italian ENEA Report, Rome, 1997, p. 117.
[8] R.A. Verrall, D.H. Rose, J.M. Miller, I.J. Hastings, D.S. MacDonald, J. Nucl. Mater. 179–181 (1991) 855.