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TRITIUM RELEASE FROM BERYLLIUM DISCS AND LITHIUM CERAMICS IRRADIATED IN THE SIBELIUS EXPERIMENT

C. E. Johnson,¹ D. L. Baldwin,² and J. P. Kopasz¹

¹Argonne National Laboratory, Chemical Technology Division,
9700 South Cass Avenue, Argonne, IL 60439 USA

²Pacific Northwest Laboratory, Chemical Sciences Department,
PO Box 999, Richland, WA 99352 USA

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The SIBELIUS experiment was designed to obtain information on the compatibility between beryllium and ceramics, as well as beryllium and steel, in a neutron environment. This experiment comprised irradiation of eight capsules, seven of which were independently purged with a He/0.1% H₂ gas mixture. Four capsules were used to examine beryllium/ceramic (Li₂O, LiAlO₂, Li₄SiO₄, and Li₂ZrO₃) and beryllium/steel (Types 316L and 1.4914) compacts. Isothermal anneal experiments have been run on representative beryllium and ceramic disks from each of the four capsules at 550°C to 850°C in steps of 100°C. The results indicate that tritium release from the beryllium did not exhibit burst release behavior, as previously reported, but rather a progressive release with increasing temperature. Generally, ~99 % of the tritium was released by 850°C. Tritium release from the ceramic discs was quite similar to the behavior shown in other dynamic tritium release experiments on lithium ceramics. The tritium content in beryllium discs adjacent to a steel sample was found to be significantly lower than that found in a beryllium disc adjacent to a ceramic sample. Recoil of tritium from the ceramic into the beryllium appears to be the source of tritium entering the beryllium, probably residing in the beryllium oxide layer.

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TRITIUM RELEASE FROM BERYLLIUM DISCS AND LITHIUM CERAMICS IRRADIATED IN THE SIBELIUS EXPERIMENT

C. E. Johnson,¹ D. L. Baldwin,² and J. P. Kopasz¹

¹Argonne National Laboratory, Chemical Technology Division,
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²Pacific Northwest Laboratory, Chemical Sciences Department,
PO Box 999, Richland, WA 99352 USA

1. Introduction

The SIBELIUS experiment is an EC/USA collaborative test [1] to assess the compatibility of beryllium/ceramic and beryllium/steel compacts in a neutron environment and to understand the potential for acceleration of beryllium oxidation caused by enhanced diffusion of oxygen or altered phase stability. The irradiation vehicle consisted of eight capsules, seven of which were independently purged with an He/0.1% H₂ gas mixture. The eighth capsule was not purged and contained beryllium pellets used to obtain data on lifetime void swelling. Seven capsules were operated at 550°C while the capsule containing the lithium orthosilicate pebbles was operated at 450°C. Four capsules were used to examine beryllium/ceramic (Li₂O, LiAlO₂, Li₄SiO₄, Li₂ZrO₃) and beryllium/steel (Types 316L and 1.4914) compacts. Two capsules contain only ceramic (Li₂O and LiAlO₂), one capsule contains beryllium pellets for lifetime void swelling tests, and one capsule contains 0.5-mm Li₄SiO₄ pebbles in contact with beryllium discs. These tests were designed to identify any performance-limiting issues associated with the use of beryllium as a neutron multiplier in ceramic breeder blankets.

Small quantities of tritium are generated in the beryllium via a ⁹Be neutron reaction producing tritium and helium. Because the buildup of the tritium could become a safety issue, there is strong interest in tritium removal. Prior tritium release studies on beryllium have been on material that has served as a reflector in a nuclear reactor. Generally, these reflector materials have remained in place for several years at low temperature, e.g., <100°C. When these reflector materials have been examined under isothermal anneal conditions, they have exhibited explosive release of tritium at high temperatures.

Earlier studies on tritium release from the SIBELIUS beryllium discs [2] reported that "excess" tritium was found in the beryllium discs, and that the source of the "excess" tritium was recoiled tritium from adjacent ceramic samples. How this tritium was supposed to collect in the beryllium was never fully explained. If truly

recoiled tritium, its location would be in the near surface region at a depth of ~40 microns; at this depth the tritium would be expected to release from the beryllium easily. The objective of the current studies was to critically examine the tritium release characteristics of the beryllium discs and to characterize any differences in tritium behavior from beryllium discs located adjacent to steel and beryllium discs located adjacent to ceramic.

2. Experimental

Stepped-temperature anneal experiments were run on the beryllium and ceramic discs from the SIBELIUS experiment. The anneals covered temperatures of 550, 650, 750, and 850°C for 24 h at each temperature. A He/0.1% H₂ gas mixture flowing at 100 cm³/min purged the sample contained in a quartz apparatus. Two independent release curves were obtained, one from an on-line ion-chamber measurement and one from a gas-collection/bubbler system followed by liquid scintillation counting. The gas-collection/bubbler data were considered to represent the principal data set, since this methodology is not subject to variation. This contrasts with the ion-chamber data, which are subject to variation because of the need to make time-dependent background corrections and the potential for calibration corrections due to changing characteristics in the electronics. Shifts in the ion-chamber background or shifts in system calibration need to be continually monitored to ensure high-quality data. For the current analysis, the ion-chamber release curve was background corrected to fit the bubbler curve.

3. Results and Discussion

Table I lists, in summary form, all the available data for tritium release from beryllium discs in the SIBELIUS experiment. The German data are taken from a recent publication by Dienst et al. [2]. The table is organized in a manner that closely corresponds to the location of a given sample in the SIBELIUS test vehicle [3]; the lithium orthosilicate capsule was at the bottom of the test vehicle, while the lithium metazirconate capsule was at the top. Each capsule contained a stack of 17 discs. The discs were arranged so that beryllium-steel and beryllium-ceramic discs alternated in the stack, with the first disc in each stack being a beryllium disc. The table lists the pellet weight, the amount of tritium released in MBq, the measured and calculated amount of tritium in 10⁸ Bq/g, and the measured-to-calculated ratio for retained tritium. Note that the beryllium discs within the body of the stack (adjacent to the ceramic) all show a higher tritium content than that expected from a transmutation reaction within the beryllium.

The last two columns in Table I enable a comparison between measured and calculated tritium concentrations for each of the different ceramic capsules. The importance of this ratio is its use to further investigate the concern expressed by Dienst et al. [2] that the original neutronic calculations had not computed the tritium generation rate correctly. The current data are not in good agreement with Dienst et al., especially with regard to the ratios of tritium measured to tritium calculated for the beryllium discs

Table I

TRITIUM CONTENT IN BERYLLIUM DISCS FROM THE SIBELIUS EXPERIMENT

Sample Number	Origin	Pellet Weight, g	MBq Tritium Released	10 ⁸ Bq/g		Meas./Calc. Ratio
				Meas.	Calc.	
1-1	Ger	0.1890	191	10.10	3.84	2.63
1-6	Ger	0.1771	719	40.60	3.84	10.57
2-1	USA	0.1829	112	6.12	5.57	1.10
2-7	USA	0.1891	190	10.04	5.57	1.80
2-9	Ger	0.1877	238	12.67	5.57	2.27
2-15	Ger	0.1761	244	13.85	5.57	2.49
3-1	USA	0.1882	136	7.23	7.71	0.94
3-7	USA	0.1874	236	12.59	7.71	1.63
5-1	USA	0.1865	130	6.97	8.12	0.86
5-7	USA	0.1861	218	11.71	8.12	1.44
5-9	Ger	0.1868	229	12.26	8.12	1.51
5-15	Ger	0.1720	271	15.75	8.12	1.94
6-1	USA	0.1858	105	5.67	6.67	0.85
6-7	USA	0.1846	187	10.15	6.67	1.52

First digit in sample number indicates the following: 1 = Li₄SiO₄ pebbles, 2 = Li₄SiO₄ pellets, 3 = LiAlO₂ pellets, 5 = Li₂O pellets, and 6 = Li₂ZrO₃ pellets.

adjacent to steel, namely, samples 2-1, 3-1, 5-1, and 6-1, where the measured-to-calculated ratios are 1.10, 0.94, 0.86, and 0.85, respectively. These values are close to unity and thus suggest that the neutronic calculations for tritium generation are satisfactory. These results also indicate that very little tritium has left the beryllium at 550°C, at least for this fluence level. However, beryllium sample 1-1, from the capsule containing the orthosilicate pebbles operated at 450°C, shows an abnormally high tritium content for a beryllium disc adjacent to steel. It is unlikely that the high tritium content is due solely to the lower temperature operation.

For the remainder of the beryllium discs within the body of the stack, where beryllium was in contact with a ceramic pellet, the measured-to-calculated tritium ratio is significantly greater than unity, ranging from 1.44 to 2.49. In fact, the ratio reaches 10.57 if one includes the capsule with lithium orthosilicate pebbles at 450°C. In examining the data further, we identified two numerical groupings for the measured-to-calculated tritium ratio. The lithium orthosilicate samples form one group, where the ratio ranges from 1.80 to 2.49 (10.57 if the pebbles are included), while the other ceramics form another group, where the ratio ranges from 1.44 to 1.94. There are also some differences in the measured-to-calculated ratio between laboratories and between materials. For the orthosilicate, the USA value (1.80 for disc 2-7) is somewhat lower than that of the German value (2.27 for disc 2-9). The comparison for lithium oxide is mixed; the measured-to-calculated ratios for discs 5-7 and 5-9 are quite similar, while the value for disc 5-15 is high at 1.94. This latter ratio is the only outlier in this data set.

As a first approximation, assume that the "excess" tritium came from recoil of tritium from the higher tritium-generating ceramic disc. This assumption may have some merit. However, the relative loading of tritium in the adjacent beryllium discs does not rigorously correspond to the relative tritium generation rates for each ceramic. The tritium generation rates in Becquerels per day are 7.10×10^9 for lithium orthosilicate, 5.48×10^9 for lithium zirconate, 4.22×10^9 for lithium oxide, and 4.00×10^9 for lithium aluminate. Thus, the orthosilicate has the highest generation rate, the zirconate is in the mid-range, and the oxide and the aluminate have the lowest tritium generation rate. There is certainly correspondence between the high tritium content in the beryllium disc adjacent to the orthosilicate and its position with the highest tritium generation rate, but the correspondence is only marginal for the other three ceramics. The three ceramics differ by some 45% in tritium generation capability, but the beryllium discs adjacent to each of the other ceramics appear to have retained about the same amount of "excess" tritium. The tritium generation rate for lithium zirconate is some 35% lower than that of lithium orthosilicate. Does this suggest that one needs a minimum flux of tritium to ensure retention in the beryllium, and that 5.48×10^9 Bq/day for the zirconate is below the threshold needed for retention? Maybe tritium retention improves at a lower temperature, as shown by the capsule at 450°C with orthosilicate pebbles (disc 1-6). Thus, it does not appear to be simply tritium recoil that sets the tritium concentration in the beryllium disc, but rather tritium recoil with additional material characteristics that ensure tritium retention.

The ceramic in each of the capsules is as follows: Li_4SiO_4 in capsule 2, LiAlO_2 in capsule 3, Li_2O in capsule 5, and Li_2ZrO_3 in capsule 6. A typical tritium release curve for the beryllium discs was found to be similar to that of previous isothermal anneal experiments on lithium ceramics [4]. That is, the tritium release generally showed a sharp increase in tritium release with each increasing temperature increment. This increased release reached a maximum, which quickly settled down with time. There did not appear to be any "burst" release with increasing temperature; indeed, the amount of tritium available for release decreased significantly once the temperature had been increased above 650°C .

The tritium release data from the isothermal anneal experiments are plotted in Figs. 1 and 2 for beryllium discs adjacent to steel and beryllium discs adjacent to ceramic, respectively. The general characteristics of tritium release from the beryllium discs adjacent to a ceramic are easily seen in Figure 2 where the percent of tritium release from the beryllium is plotted against temperature. For these discs, the tritium release increased in the order lithium orthosilicate, lithium zirconate, lithium aluminate, and lithium oxide and is similar to the order for increased tritium generation rate, though we are not sure why. The differing tritium generation rates mean that the beryllium discs in each capsule were exposed to differing amounts of tritium in the purge gas flowing by the disc. For the four capsules investigated, the tritium generation rate in the ceramic increases in the order lithium aluminate, lithium oxide, lithium zirconate, and lithium orthosilicate. The largest amount of tritium was retained by the disc in the orthosilicate capsule followed by the aluminate, the oxide, and the zirconate. Tritium release from beryllium in the Li_4SiO_4 capsule was similar for both discs (no effect of ceramic or steel or fraction of tritium released at an anneal temperature), while for the other ceramics, a higher fraction of the tritium was released at 750°C for the beryllium adjacent to the ceramic.

Figure 3 presents data on tritium release from four different investigations covering four different beryllium samples. As arranged in the figure, these materials are a low density material (80.9% TD), high density arc-cast material analyzed by Jones and Gibson [5], a high density arc-cast disc containing <300 ppm BeO from the SIBELIUS experiment, and a high density (99.6% TD) beryllium sample analyzed by Billone and Baldwin [6]. The 80.9 % TD material exhibits the best release characteristics, with significant tritium being released at 400°C . The tritium release from the SIBELIUS beryllium discs is not as high as the Jones and Gibson data, at least at temperatures below 800°C , but is higher than the 99.6% TD data at temperatures above 600°C . Because of the many variables covered in these four experiments, it is difficult to develop a firm picture of the optimum material. What does appear to be attractive is to select a lower density beryllium and ensure that blanket designs include beryllium temperatures of 500°C or above. Optimizing the density and temperature parameters is certain to provide conditions that facilitate ease of tritium release.

A significant characteristic shown by the SIBELIUS beryllium discs was the lack of "burst" tritium release at higher temperatures as compared with earlier studies on high density material. This release behavior may be the result of irradiating

at 550°C rather than the lower temperatures experienced by the reflector beryllium. Irradiating at this higher temperature may be sufficient for the diffusion and collection of tritium atoms into a more easily released molecular form. Also, the beryllium discs did not contain similar amounts of tritium, the amount retained depending on whether a disc was adjacent to a steel disc or adjacent to a ceramic disc. Those beryllium discs adjacent to a ceramic contained significantly more tritium, probably due to recoil from the ceramic. The beryllium oxide layer appears to influence the amount of tritium retained by the beryllium discs. The tritium release from the ceramic discs is consistent with other investigations of tritium release from ceramic materials [4], and there appears to be little or no effect from the adjacent beryllium. Further, all the ceramics exhibited little retention of tritium, the amount being similar to that shown in other experiments at 550°C with a He/0.1% H₂ purge gas.

4. Conclusions

Investigations were carried out to determine the amount of retained tritium and the rates of tritium release from beryllium discs in the SIBELIUS experiment. These studies have shown that the bulk of the generated tritium was retained in the beryllium, and when the discs were heated to 650°C and above, the tritium was readily released. None of the samples examined exhibited the burst release characteristics observed earlier but rather an orderly release dependent solely upon temperature. In comparison with literature information on tritium release from beryllium, the SIBELIUS data have a steeper slope for the temperature coefficient. Also, the literature information shows that for lower density beryllium (80.9 % TD), tritium release is considerably enhanced.

Acknowledgments

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Figure 1 Tritium Release from SIBELIUS Beryllium Discs Adjacent to Steel.

Figure 2 Tritium Release from SIBELIUS Beryllium Discs Adjacent to Ceramic.

Figure 3 Tritium Release from Several Different Beryllium Samples.

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