

HELIUM ANALYSES OF 1-MM BERYLLIUM MICROSPHERES FROM COBRA-1A2 – B. M. Oliver (Pacific Northwest National Laboratory)*

OBJECTIVE

The purpose of this work was to provide quantitative helium release response from prototypic irradiated beryllium pebbles. Such pebbles are under consideration as the neutron multiplier medium in the European Fusion Technology Program Helium Cooled Pebble Bed (HCPB) Blanket.

SUMMARY

Multiple helium analyses on four beryllium microspheres irradiated in the Experimental Breeder Reactor - II (EBR-II) at Argonne National Laboratory - West (ANL-W), are reported. The purpose of the analyses was to determine the total helium content of the beryllium, and to determine the helium release characteristics of the beryllium as a function of time and temperature. For the helium release measurements, sequential helium analyses were conducted on two of the samples over a temperature range from 500°C to 1100°C in 100°C increments. Total helium measurements were conducted separately using our normal analysis method of vaporizing the material in a single analysis run.

Observed helium release in the two beryllium samples was nonlinear with time at each temperature interval, with each step being characterized by a rather rapid initial release rate, followed by a gradual slowing of the rate over time. Sample Be-C03-1 released virtually all of its helium after approximately 30 minutes at 1000°C, reaching a final value of 2722 appm. Sample Be-D03-1, on the other hand, released only about 62% of its helium after about 1 hour at 1100°C, reaching a final value of 1519 appm. Combining these results with subsequent vaporization runs on the two samples, yielded total helium concentrations of 2724 and 2459 appm. Corresponding helium concentrations measured in the two other "C03" and "D03" samples, by vaporization alone, were 2941 and 2574 appm. Both sets of concentrations are in reasonable agreement with predicted values of 2723 and 2662 appm.

Helium-3 levels measured during the latter two vaporization runs were 2.80 appm for Be-C03-2, and 2.62 appm for Be-D03-2. Calculated ³He values are slightly lower at 2.55 and 2.50 appm, respectively, suggesting somewhat higher tritium levels in the beryllium than predicted.

PROGRESS AND STATUS

Introduction

Beryllium pebbles are being considered for the neutron multiplier medium in the European Fusion Technology Program Helium Cooled Pebble Bed (HCPB) Blanket. That design is

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also being considered for testing in ITER. The pebbles to be used are an inexpensive form of beryllium produced in an intermediate step of the production of higher purity beryllium. The opportunity to obtain helium release response from prototypic fast neutron irradiated pebbles became possible after pebbles were irradiated in the US/DOE COBRA-1A2 experiment in EBR-II. Experimental details are provided in a companion paper [1]. The helium release measurements were funded by Forschungszentrum Karlsruhe.

Experimental Procedure

Helium Analysis Samples

Four beryllium microspheres were analyzed in the present study. The materials had been irradiated in the Experimental Breeder Reactor - II (EBR-II) at Argonne National Laboratory - West (ANL-W), as part of the COBRA-1A2 experiment [1]. Two of the samples were from the "C03" assembly, and were fabricated by Brush Wellman. The other two were from the "D03" assembly, and were fabricated by NGK - Japan. One of each of the C03 and D03 spheres were analyzed to determine the helium release with time and temperature. These analyses were followed by separate vaporization runs to determine if any remaining helium was in the sample. The two other microspheres (one each for C03 and D03) were analyzed for total helium content by rapid vaporization in a single analysis.

Prior to analysis, the mass of each microsphere was determined using a microbalance with calibration traceable to National Institute of Standards and Technology (NIST). Mass uncertainty is estimated to be ± 0.001 mg.

Helium Analysis Procedure

The helium released from each microsphere was determined by isotope-dilution gas mass spectrometry during heating or vaporization in a resistance-heated graphite crucible in one of the mass spectrometer system's high-temperature vacuum furnaces [2]. The absolute amount of ^4He released was measured relative to a known quantity of added ^3He "spike". The ^3He spikes were obtained by expanding and partitioning a known quantity of gas through a succession of calibrated volumes [3]. The mass spectrometer was calibrated for mass sensitivity during each series of runs by analyzing known mixtures of ^3He and ^4He .

For two of the samples, multiple sequential analyses were conducted while the temperature of the sample crucible was increased in approximately 100°C increments, starting at 500°C and increasing to a maximum of 1100°C . For this procedure, a ^3He spike was first added to the furnace. While still at room temperature ($\sim 25^\circ\text{C}$), a known small fraction ($\sim 0.04\%$), or aliquot of the sample gas was then taken from the furnace to determine the absolute amount of ^4He initially present in the furnace. This analysis was done to determine the starting background ^4He level. The crucible was then heated to 500°C , and additional aliquots taken for ^4He analysis. Each temperature was held until some leveling off of the ^4He was observed, at which point the temperature was raised to the next temperature. The time period between each analysis was varied from a minimum of about 5 minutes to a maximum of about 30 minutes depending on the observed ^4He release rate at each temperature. Crucible

temperature was measured using a standard chromel-alumel thermocouple in direct contact with the crucible.

All measured helium levels were corrected for background helium buildup in the analysis furnace as a function of time. These background helium levels were determined by separate "control" analyses conducted immediately before or after the sample runs. Background ^4He levels generally rise linearly with time due largely to diffusion of helium from the atmosphere through the Pyrex furnace top. Background ^4He levels were in the range of 10^{12} to 10^{13} atoms over the 6 to 7 hour time period for each analysis series. Uncertainty in the ^4He background is estimated to be approximately 10^{12} atoms. Background ^3He levels remained in the 10^{12} atom range, and were negligible compared to the added ^3He spike ($\sim 10^{16}$ atoms).

Following the time/temperature measurements, the two microspheres were relocated, in their original crucibles, to different locations in the same furnace for subsequent vaporization analysis along with the other two beryllium samples analyzed as part of the present effort. For the vaporization runs, both ^3He and ^4He were determined in each sample. This was done by analyzing a first aliquot of the gas released during vaporization prior to the addition of the ^3He spike. Because only a small fraction of the gas was taken from the furnace for each analysis, this procedure did not adversely affect the accuracy of the final measured ^4He value.

Results

The results of the helium release measurements are shown graphically in Figures 1 and 2. Separate data tables can be found in the summary report [4]. Each data table gives the analysis time (in hours) from the start of the series, and the time at which each temperature level was started. Temperature increase for each interval took approximately 1 minute. Indicated uncertainties in the temperature data are the standard deviations in the observed temperature variations over the temperature interval. Helium-4 values are given as total atoms released, and as concentrations in atomic parts per million (appm, 10^{-6} atom fraction) with respect to the total number of beryllium atoms in each sample. Conversion from total helium to helium concentration was based on a calculated value of 6.682×10^{22} atoms per gram assuming pure beryllium.

Table 1 gives the total helium released after vaporization in the four beryllium microspheres. Results are listed as total atoms of ^3He and ^4He , and as concentrations, again in atomic parts per million. Also shown in Table 1 are the end results for Be-C03-1 and Be-D03-1 from the helium release measurements [4], and the combined helium release for these two samples from both measurements.

Absolute uncertainty (1σ) in the individual helium analysis results, determined from the cumulative uncertainties in the sample mass, isotope ratio measurement, spike size, and helium background subtractions, is estimated to be approximately 1% combined with 10^{12} atoms in quadrature. For the lowest measured helium levels, therefore, the uncertainty of 10^{12} atoms is the dominant term. For helium levels above about 10^{14} atoms, the uncertainty is approximately 1%.

Table 1. Total helium release from 1-mm beryllium microspheres

Sample	Sample Mass (mg)	Measured Helium (vaporized) ^a				Measured ⁴ He (total) (appm) ^b		
		³ He (atoms)	(appm) ^b	⁴ He (atoms)	(appm) ^b	Heated ^c	Total ^d	Predicted
Be-C03-1	1.419	3.05E13	0.322	1.698E15	1.791	2722	2724	2723
Be-C03-2	1.633	3.06E14	2.80	3.209E17	2941	--	2941	
						Mean	2833	
						±1σ	±153	
Be-D03-1	1.472	1.17E14	1.19	9.249E16	940.3	1519	2459	2662
Be-D03-2	1.206	2.11E14	2.62	2.074E17	2574	--	2574	
						Mean	2517	
						±1σ	±81	

^aMeasured helium from vaporization analysis.

^bHelium concentration in atomic parts per million (10^{-6} atom fraction) with respect to the total number of beryllium atoms in the sample.

^cFinal ⁴He from helium release measurements [4].

^dTotal ⁴He [4].

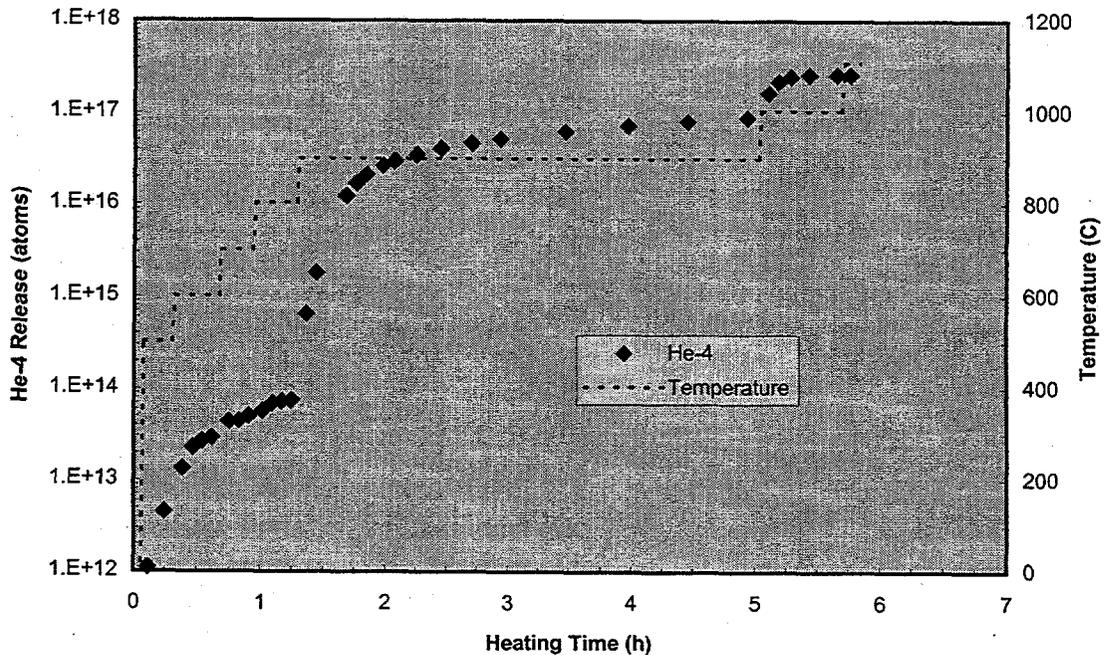


Figure 1. Helium release from sample Be-C03-1

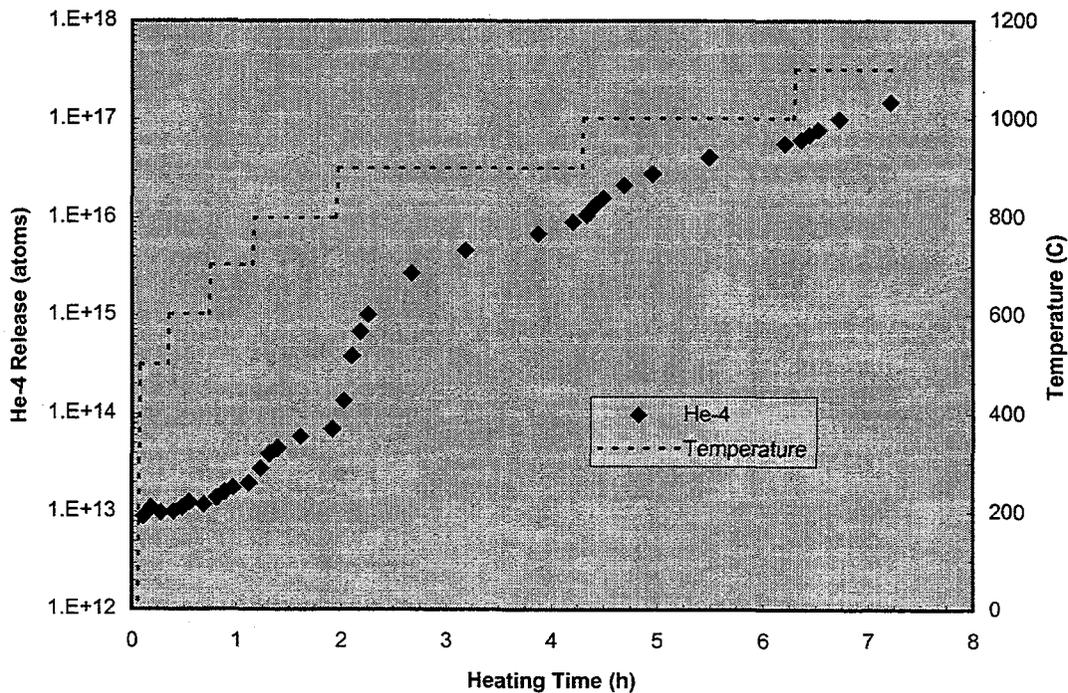


Figure 2. Helium release from sample Be-D03-1

Discussion

Observed helium release from both microspheres was nonlinear with time at each temperature level. As is evident in Figures 1 and 2, both microspheres showed definite "steps" in the helium release at each temperature, with each step being characterized by a rather rapid initial release rate, followed by a gradual slowing of the rate over time. Except for Sample Be-C03-1 at 1000 °C, none of the release steps showed saturation over the 1 to 2 hour time periods the temperatures were held.

Helium release as a fraction of the total helium in the sample was markedly different for the "C03" and "D03" material. Sample Be-C03-1 released virtually all of its helium after approximately 30 minutes at 1000 °C. Sample Be-D03-1, on the other hand, released only about 62% of its helium even after about 1 hour at 1100 °C. Absolute helium release was correspondingly lower for Be-D03-1 at each temperature level. As indicated earlier, the C03 beryllium material was fabricated by Brush Wellman, whereas the D03 material was fabricated by NGK - Japan. The different helium release characteristics suggests some material differences in the two beryllium lots. However, additional analyses would be required to verify any material effects.

Total helium release after vaporization for all four microspheres are given in Table 3. In terms of helium concentration, the totals for Be-C03-1 and -2 are 2724 and 2941 appm, with an average value of 2833 appm. The corresponding totals for Be-D03-1 and -2 are 2459 and 2574 appm, with an average value of 2517 appm. In both cases, the total ^4He for the combined heating+vaporization runs is lower than that for the separate vaporization runs alone. This variation amounts to 8% for Be-C03-1, and 5% for Be-D03-1. Some of this variation may be due to radial gradients in the two irradiation assemblies, although this was not anticipated to be larger than perhaps 5%. Predicted helium concentrations in the C03 and D03 beryllium are 2723 and 2662 appm, respectively [5]. Mean calculated to experimental ratios (C/E's) are 1.04 for Be-C03 and 0.95 for Be-D03.

Helium-3 levels measured during the vaporization runs were 2.80 appm for Be-C03-2, and 2.62 appm for Be-C03-1. Predicted ^3He levels, assuming no tritium loss during or after irradiation, are slightly lower at 2.55 and 2.50 appm [3], suggesting somewhat higher levels of tritium in the beryllium than calculated.

FUTURE WORK

The data presented here raise some interesting questions which could be answered by additional work. However, funding has not yet been identified. Suggestions for such additional work are:

1. Perform analyses on replicate samples of the C03 and D03 material to verify any differences related to the helium release characteristics with time and temperature, and to verify any variations in total helium content within the two material lots.
2. Perform more sensitive analyses at lower temperatures to accurately determine the low-temperature release characteristics of the C03 and D03 material.
3. Increase the temperature hold times to determine if the release curves eventually "saturate" at any given temperature.
4. Determine total helium and helium release characteristics of replicate samples of the 3-mm and 5-mm microspheres as outlined above.

ACKNOWLEDGEMENTS

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