

STEPPED-ANNEAL HELIUM RELEASE IN 1-MM BERYLLIUM PEBBLES FROM COBRA-1A2 - B. M. Oliver (Pacific Northwest National Laboratory)*

OBJECTIVE

The purpose of this work was to provide quantitative stepped 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

Stepped-anneal helium release measurements on two sets of fifteen beryllium pebbles irradiated in the Experimental Breeder Reactor - II (EBR-II) at Argonne National Laboratory - West (ANL-W), are reported. The purpose of the measurements was to determine the helium release characteristics of the beryllium using larger sample sizes and longer anneal times relative to earlier measurements. Sequential helium analyses were conducted over a narrower temperature range from approximately 800°C to 1100°C in 100°C increments, but with longer anneal time periods. To allow for overnight and unattended operation, a temperature controller and associated circuitry were added to the experimental setup.

Observed helium release was nonlinear with time at each temperature interval, with each step being generally characterized by an initial release rate followed by a slowing of the rate over time. Sample Be-C03 showed a leveling off in the helium release after approximately 3 hours at a temperature of 890°C. Sample Be-D03, on the other hand, showed a leveling off only after ~12 to 24 hours at a temperature of 1100°C. This trend is consistent with that observed in earlier measurements on single microspheres from the same two beryllium lots. None of the lower temperature steps showed any leveling off of the helium release.

Relative to the total helium concentrations measured earlier, the total helium releases observed here represent approximately 80% and 92% of the estimated total helium in the C03 and D03 samples, respectively.

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 also being considered for testing in ITER. The pebbles to be used are an inexpensive form

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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

A total of 30 beryllium pebbles 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. Fifteen of the samples were from the "C03" assembly, and were fabricated by Brush Wellman. The other fifteen were from the "D03" assembly, and were fabricated by NGK - Japan.

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

Helium Measurement Procedure

The helium released from the C03 and D03 pebbles was determined by isotope-dilution gas mass spectrometry during heating in resistance-heated graphite crucibles in one of the mass spectrometer system's high-temperature vacuum furnaces [2]. The absolute amount of ^4He released was measured relative to known quantities of added ^3He "spikes". 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 both groups, multiple sequential measurements were conducted while the temperature of the sample crucible was increased in approximately 100°C increments. 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. The crucible was then heated to the first temperature step, and additional aliquots taken for ^4He measurement. Each temperature was held for a minimum of ~ 24 hours, or until a leveling off of the ^4He was observed. The time period between each measurement was varied from a minimum of about 5 minutes to a maximum of about 2 hours depending on the observed ^4He release rate at each temperature.

The fifteen beryllium pebbles were loaded into the central section of one of the system's standard $3/16"$ (0.48 cm) diameter graphite crucibles. To accommodate the pebbles, the central coaxial hole in the crucible was enlarged to $0.094"$ (0.24 cm) diameter. Small graphite plugs were placed at each end of the hole to position the samples as close as

possible to the middle of the crucible. A diagram of the setup, showing a section of the high temperature furnace used, is shown in Figure 1. Crucible temperature was measured using a standard K-type (chromel-alumel) thermocouple inserted into the side of the crucible. As indicated in the figure, this thermocouple was also used in conjunction with a separate temperature readout and controller circuit to maintain a constant temperature in the crucible for each temperature step, and to allow for overnight and unattended operation.

Because of the relatively long periods of time the furnace remained isolated, it was decided to pump out the furnace at regular intervals (usually about 24 hours) in order to keep offgas pressure in the furnace to below about 500 microns (67 Pa). Immediately after pumpout, a new ^3He spike was added to the furnace, and the measurements continued. Pumpout times were chosen to coincide with periods of slower helium release, usually near the end of each anneal period. Small corrections were made to the total helium release data to account for helium release during the pumpout (about 3 minutes).

All measured helium levels were corrected for background helium buildup in the analysis furnace as a function of time. 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. Uncertainty in the ^4He background is estimated to be $\sim 10^{12}$ atoms. Background ^3He levels remained in the 10^{12} atom range, and were negligible compared to the added ^3He spike of $\sim 10^{16}$ atoms.

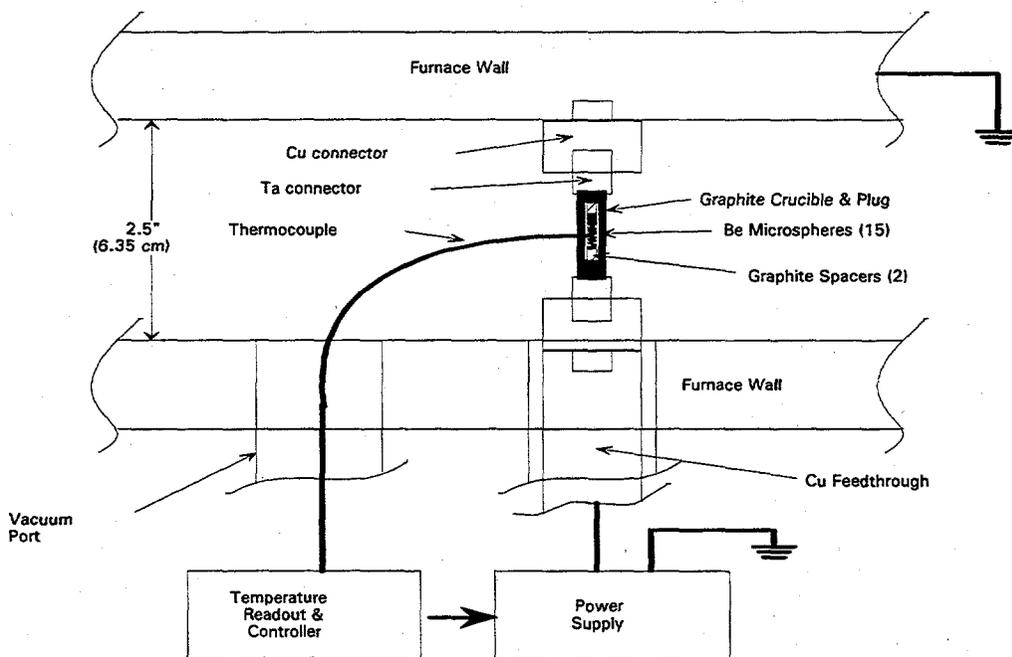


Figure 1. Diagram of sample crucible and temperature control

Results

The results of the helium release measurements are shown graphically in Figures 2 and 3. Separate data tables can be found in the summary report [4]. Each data table gives the anneal 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. 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 at 100% of theoretical density. In Figures 2 and 3, the top figure shows the cumulative helium release in atoms, and the bottom figure shows the calculated helium release rate in atoms/second.

After initiation of the measurement series for Sample Be-C03, it was observed that the temperature of the thermocouple was lower than that of the graphite crucible. As a result, additional temperature readings were taken at each temperature step using a separate analog optical pyrometer (the pyrometer data suggested actual crucible temperatures about 100°C higher than indicated by the thermocouple). Later examination verified that the thermocouple had only been pressing against the surface of the crucible, rather than being fully inserted into the interior through the small side hole (see Figure 1). At the conclusion of the Be-C03 run, therefore, this thermocouple was replaced with a second smaller diameter unit which fit completely into the crucible side hole. This second thermocouple was then used to correct the earlier pyrometer readings using identical furnace conditions, and also was used for the subsequent Be-D03 measurements. As a result of the above, the temperature range for the Be-C03 sample was approximately 800 to 1100°C rather than the desired range of 700 to 1000°C.

Absolute uncertainty (1σ) in the individual helium measurement results [4], determined from the cumulative uncertainties in the sample mass, isotope ratio measurement, spike size, and helium background subtractions, is estimated to be approximately 1% to 2% combined with 10^{12} atoms (in quadrature). Indicated uncertainties in the temperature data are estimated based on uncertainties in the measuring equipment, thermocouple calibration, and pyrometer readings (for Be-C03).

Discussion

Consistent with the results obtained earlier for single Be-C03 and Be-D03 pebbles [5], helium release from both sets of pebbles was nonlinear with time at each temperature level. Both samples showed definite "steps" in the helium release at each temperature, with each step being generally characterized by an initial release rate, followed by a slowing of the rate over time. The one exception to the above behavior was the initial helium release in Sample Be-C03 at the first temperature step of 774°C. Here, the release rate initially rises, then falls, then rises again and continues to rise slowly over the remaining anneal period.

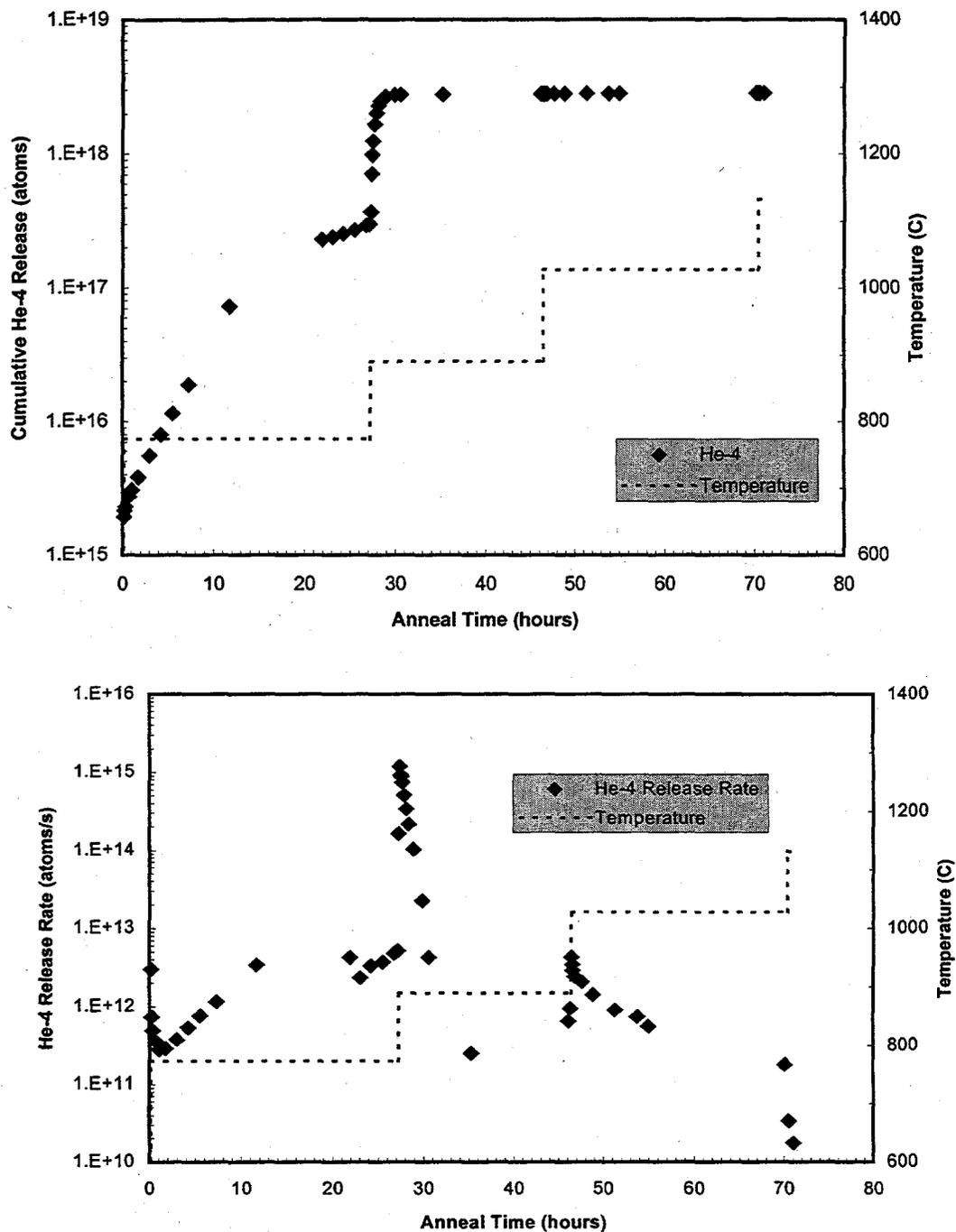


Figure 2. Cumulative helium release and release rate from sample Be-C03

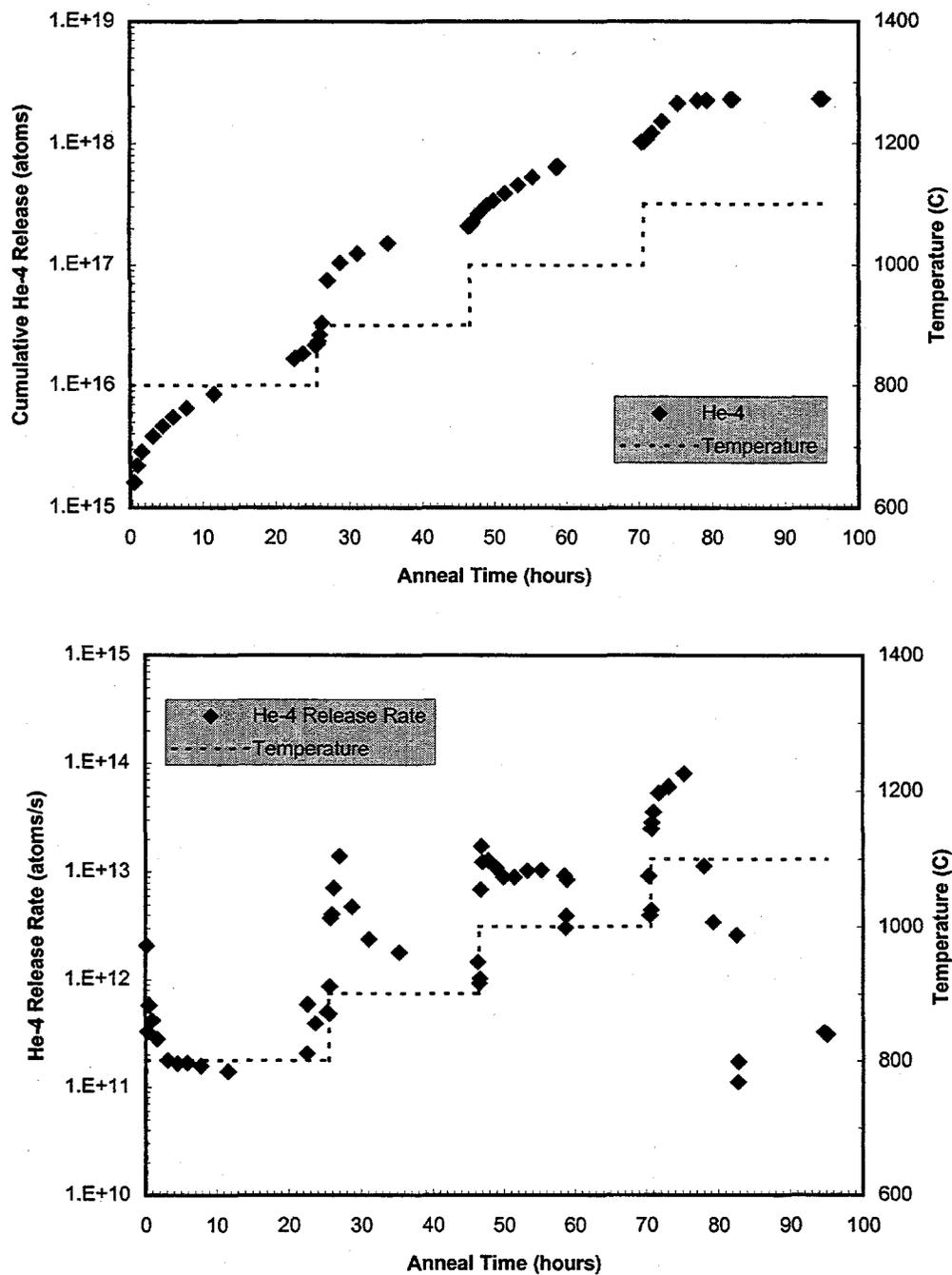


Figure 3. Cumulative helium release and release rate from sample Be-D03

Observed helium release from Sample Be-C03 showed a leveling off at the 890°C temperature step. Additional increases to 1028°C and 1132°C, showed very little extra helium release (<2%). Sample Be-D03, on the other hand, showed a leveling off only at the highest step of 1100°C. Final measured ⁴He concentrations were 2276 appm for Be-C03 and 2312 appm for Be-D03.

The above trend of lower temperature helium release in the C03 material is consistent with that observed earlier for the single pebble [6], and further supports the conclusion of differences in the helium release characteristics of the two material lots. None of the lower temperature steps showed any leveling off of the helium release, even with increased anneal times of at least 24 hours. The earlier measurements had anneal times of only 1 to 2 hours.

Based on the average helium concentrations measured earlier for the C03 and D03 beryllium (2833 appm for Be-C03-1, and 2517 appm for Be-D03-1), the values observed here represent ~80% and ~92% of the total "expected" helium concentrations. These percentages are in contrast to the previous results which showed virtually complete helium release from Be-C03-1 at 1000°C, but only about 62% release in Be-D03-1 at 1100°C. Additional vaporization helium analyses on the two beryllium sets analyzed here would be required to determine the actual release percentages.

FUTURE WORK

This work is completed.

ACKNOWLEDGEMENTS

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REFERENCES

1. D. S. Gelles, "US/DOE OFES Neutron Irradiation Experiments containing Beryllium," in this semiannual report.
2. H. Farrar and B. M. Oliver, "A Mass Spectrometer System to Determine Very Low Levels of Helium in Small Solid and Liquid Samples," J. Vac. Sci. Technol. **A4**, 1740 (1986).
3. B. M. Oliver, J. G. Bradley, and H. Farrar, "Helium Concentration in the Earth's Lower Atmosphere," Geochim. Cosmochim. Acta **48**, 1759 (1984).

4. D. S. Gelles, L. R. Greenwood, B. M. Oliver, D. L. Baldwin, R. M. Ermi, and H. Tsai, "Post-Irradiation Examination of Beryllium - Project 26929", PNNL-29550 (December 1997)
5. L. R. Greenwood, "Production of ^4He and Tritium from Be in the COBRA-1A2 Irradiation", PNNL Report, May 1997.
6. B. M. Oliver, "Helium Analyses of 1-mm Beryllium Microspheres from COBRA-1A2", PNNL Report, May 6, 1997.