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HELIUM/SOLID POWDER O-RING LEAKAGE CORRELATION EXPERIMENTS USING A RADIOTRACER*

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INTRODUCTION

A cooperative program to assess possible special nuclear material (SNM) leakage from transport packages is being conducted by the U. S. Department of Energy (DOE) and the Power Reactor and Nuclear Fuel Development Corporation of Tokyo, Japan (PNC). Previous experiments to determine the correlation between He and particulate UO_2 leakage through typical unlubricated face-type O-ring seals were reported at the 7th International Symposium on Packaging and Transportation of Radioactive Materials (PATRAM-83)[1]. Quantities (10-250 ng) of uranium were found in some tests, but the source was questionable because of high blanks and the possibility of airborne contamination. We have redesigned these experiments to use neutron activated UO_2 powder as a radioactive tracer. This method greatly simplifies the tests and eliminates the possibility of contamination.

EXPERIMENTAL

Fixture

The experimental method used for this work duplicates the experiments reported at PATRAM-83 except irradiated UO_2 tracer and gamma ray counting are substituted for unirradiated UO_2 tracer and laser fluorescence uranium

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DISCLAIMER

detection. Figure 1 shows a typical test fixture, further described in Reference 1. The primary O-ring seal (supplied by PNC) is the only barrier between the central chamber and the test cavity. The valves, filters and other plumbing connect with either the central chamber or test cavity and are used for measuring pretest He leak rates, controlling pressure, introduction of tracer to the central chamber and for post-test sampling of the test cavity.

Tracer

The previous experiments used depleted UO_2 powder, > 95 % by count 0.2 to 2.0 μm. diameter, as a surrogate for the PNC mixed oxide fuel powder (nominal size 2 to 3 μm). The experiments reported here use the same UO_2 powder neutron activated by reactor irradiation to convert a small fraction of the ^{238}U atoms to ^{239}Np ($t_{1/2}$ = 2.35 days) by the reaction

$$^{238}U(n,\gamma)\ ^{239}U \xrightarrow[t_{1/2} = 23.5\ \text{min.}]{\beta^-} {}^{239}Np \xrightarrow[t_{1/2} = 2.35\ \text{days}]{\beta^-} {}^{239}Pu\ldots$$

The ^{239}Np produced is trapped in the UO_2 particles and is a radioactive tag for the UO_2. Each test used 1 gram of UO_2 activated to a ^{239}Np activity of 1×10^8 to 2×10^8 Bq.

Test Routine

Each fixture has its He leak rate measured and is then taken to the test temperature. The UO_2 holder, isolated by valves V7 and V8, is removed, filled with tracer and reinstalled. The test is started by opening valve V8 and then V7 to introduce tracer and He (at test pressure) to the previously evacuated central chamber. A vibrator agitates the fixture during the test and thermocouples monitor temperature.

Post-test Sampling

At the completion of a test, all plumbing above valves V3, V8 and V4 is removed. Three washes of approximately 8N HNO_3 are circulated through the test cavity to dissolve any UO_2 tracer present. All washes are combined and gamma ray counted on a Ge(Li) detector to determine the amount of tracer that passed the O-ring seal.

RESULTS

Table 1 shows the experimental conditions for the tests and the results. UO_2 was detected in three tests; upper limits are reported for the others.

CONCLUSIONS

UO_2 definitely leaked past the O-ring in three of the tests confirming the major results of the previous work.[1] Continuous leakage at these levels may require additional precautions under present regulatory policies. The mechanism and the time and particle size dependence for the leakage are not known, but there is some indication leakage is more likely at low temperatures. It is possible leakage is due to movement of the O-ring during temperature or pressure cycling at the beginning or end of a test. The radiotracer method involves less labor and is much less susceptible to contamination than the previous method. Future work will investigate leakage past lubricated O-rings and time dependence of leakage.

REFERENCE

1. W. B. Leisher, S. H. Weissman, D. R. Tallant, M. Kubo. Helium/Solid Powder O-ring Leakage Correlation Experiments. Proc. 7th International Symposium on Packaging and Transportation of Radioactive Materials, May 15-20, 1983, New Orleans, LA. ORNL CONF. -830528, 1983.

Table 1. Results of analyses

Test	Duration (Hr)	Temp[a] (°C)	Press[a,f] (atm)	Initial He Leak Rate (mL/s)	Final He Leak Rate (mL/s)	UO_2 Found (ng)
1	168	+130	7	$<1\times10^{-10}$	$\sim 5.5\times10^{-4}$	<38.
2	3	+ 26	0	$<1\times10^{-10}$	not meas[b]	65. ±14.
3	3	+ 26	1	$<5\times10^{-8}$ [c]	1.5×10^{-5}	< 6.
4	3	- 40	1	1×10^{-10}	$< 1.0\times10^{-8}$	<18.
5	3	- 40	5	$<5.0\times10^{-10}$	6.0×10^{-9}	23. ± 4.
6	3	- 40	7	$<5\times10^{-8}$	2.9×10^{-8}	74. ±10
Blank[e]	3	+ 26	0	3×10^{-7} [d]	1.7×10^{-5}	<27.

[a] Nominal
[b] No vacuum used on test cavity after UO_2 introduction.
[c] Correcting valve leak took ~ 30 min., permeation had begun.
[d] O-ring had previously been saturated with He.
[e] Run like other tests except UO_2 used not neutron activated.
[f] Measured across primary seal O-ring; "O" differential means both sides at local ambient pressure, for all others test cavity was under vacuum.

Figure Caption

Figure 1. Test Fixture

STANDARD LEAK: 3.31×10^{-8} atm cm^3/sec (HELIUM)
V1-V5: BELLOWS SEALED VACUUM VALVES, 1/4 NPT
V6-V8: BALL VALVES, 1/8 NPT
F1, F4, & F5: FILTER, 0.2 μm SILVER ELEMENT
F2 & F3: FILTER, 0.2 μm FIBER GLASS ELEMENT