STORAGE AND MEASUREMENT DEMONSTRATION OF WASTES CONTAINING CONDITIONED TRITIUM AND RADIUM SOURCES

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ABSTRACT

For the final storage of wastes containing tritium (H-3) and radium sources, it is of particular importance to evaluate the containment of activity in order that it may be guaranteed for the service life of the repository. For this purpose, a storage demonstration was conducted at the KFA Nuclear Research Center in Julich using full-scale ductile cast-iron casks of the type MOSAIK. The test set-up was instrumented with a system for analyzing the solid, liquid and gaseous phases in the cask and for measuring the leak-tightness of the cask. The higher temperatures prevailing in a geological repository were simulated by performing the demonstration at a constant ambient temperature of 50°C.

DESCRIPTION OF TEST CASKS

Two casks of the type MOSAIK III were used for the storage demonstration, one loaded with H-3 wastes and one loaded with radium sources. The constructive design of these casks is especially suitable for performing leak tests.

The cask body is made of a ductile cast iron casting. The cross section is cylindrical, with a cylindrical inner cavity. The wall thickness is 80 mm (3.1 in) on the bottom and sides of the cask, and 120 mm on the top. In the center of the top area, there is a relatively small opening for loading.

The overall dimensions of the cask are:

Height	1,240 mm	48.8 in
Diameter	1,000 mm	39.4 in.

The dimensions of the cask cavity are:

Height	940 mm	37.0 in.
Diameter	840 mm	33.0 in

The containment system of the cask is constituted by the cask body and lids with seals. The lid, normally made of ductile cast iron, is secured to the cask with four bolts, with an elastomer seal in the area of the bolting to maintain leak-tightness. For the storage demonstration, a stainless steel lid was designed with penetrations for the test instrumentation, e.g. for taking gas samples from the cavity and for monitoring the pressure in the cask.

To simulate the temperatures in a geological repository, an insulated container was built for the cask, with heating, temperature regulation and monitoring. A schematic diagram of the cask and test set-up can be seen in Fig. 1.

CASK CONTENTS

H-3 Wastes

The wastes containing tritium were generated from the heavy water cycle of the research reactor DIDO. They consisted of bead resins, both wet and dried. Besides slight concentrations of fission and activation products, there were approximately 50 Ci of H-3, mainly in the form of tritiated water. Samples of the resins and of the sump water in the cask were analyzed, and yielded the following measurements:

Sample	Nuclide	Activity
Sump water	H-3	790 mCi/kg
Resins (Leaching)	H-3	9.9 mCi/kg
Resins (Incineration)	H-3	100 mCi/kg
Resins (Incineration)	C-14	1.3 mCi/kg

The resins were sucked into the cask by vacuum from 15 containers with approximately 30 l contents (smaller quantities in 200 l drums). This method proved to be a good way of loading both dry and wet resins, with loading in the latter case taking somewhat longer.

A total of approximately 450 liters of resins were filled into the cask, with approximately 50 liters of that being free water. The total tritium activity was approximately 50 Ci.

Radium Sources

The radium-containing wastes loaded into a similar cask consisted of various types, as are present at the collection center in Julich. These included:

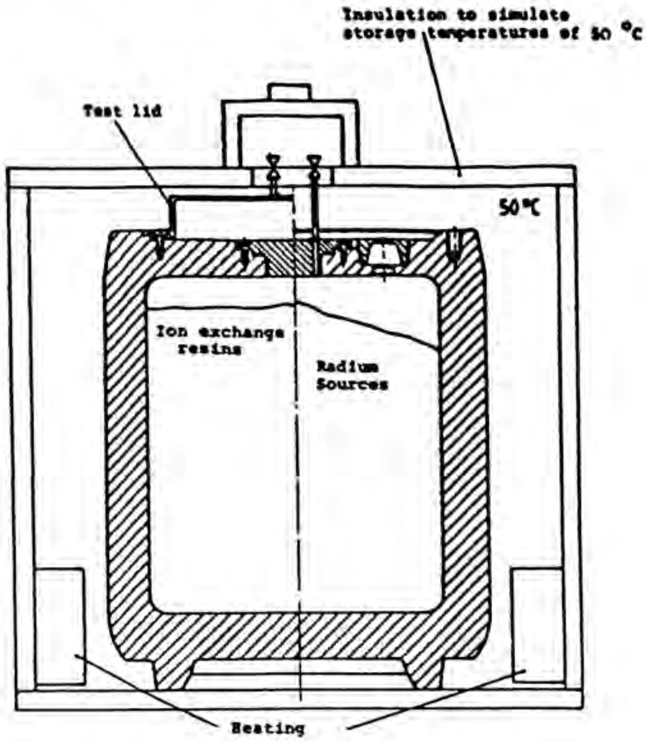

Fig. 1. Schematic Diagram of the Test Set-Up.

- Radium needles from the medical field
- Watch faces
- Radium tubes
- Fire detectors
- Luminous paint
- Radium-containing solutions.

Part of these were sealed air-tight in soldered cans with active coal. It can be assumed that a representative mixture of wastes was stored in the cask, just as is present at the collection center.

Due to varying nature of the types of waste, the exact radium activity of the sources could not be determined. The amount of activity in the loaded cask was estimated at approximately 100 mCi of Ra-226.

For loading the individual sources, a grab was used to insert the objects through the opening in the cask. The cask was thus filled as much as was possible.

ANALYSIS ON THE CASK WITH H-3 WASTES

The following analyses and tests were performed on the MOSAIK III cask with ion-exchange resins:

- Gas sampling and analyses
- Analysis of liquid contents
- Analysis of solid contents

The procedures used in these analyses are described below.

Gas Sampling and Analyses

After loading the cask with tritium-containing ion exchange resins, the cask was loaded with a test lid, through which it was possible to take gas samples from the cask cavity for analysis. Composition was determined by gas chromatography.

To determine the H-3 and C-14 activity, gas samples of five liters each were treated with the apparatus shown in Fig. 2. The H-3 determinations were done on distilled aliquots from the wash solutions. The C-14 activity of the gas in the cask was determined with $BaCO_3$ precipitated from the NaOH wash solution. The activity concentration of the non-absorbable active gas residue was measured with a proportional gas counter. For this, a measuring chamber containing a counter foil on the front side was filled with the gas. To detect the gamma particles penetrating the counter foil, a proportional counter was used, which had the same base area as the measuring chamber. In the transmission measurements, aluminum foils of known thickness were set between the chamber and the counter, for aimed absorption of the gamma radiation.

The unknown activity was identified as Krypton-85. For the measurement, a pressure vessel was used (volume = 200 ml). In order to increase the detection limit, the gas was compressed down to 50 bars.

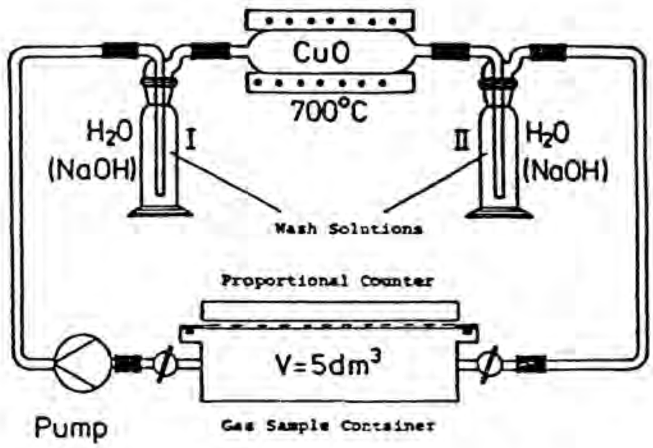

Fig. 2. Apparatus for Analysis of Gases.

To quantify the results, the same vessel was filled with Kr-85 gas of a known activity concentration.

Analysis of the Liquid Contents of the Cask

The cask contained an unknown quantity of water. Through the tube passing down to the bottom of the resins, samples of sump water were taken.

The samples were purified of any residual nuclides by distillation. The H-3 activity of the distillate was determined by a liquid scintillation technique. In addition, the H-3 activity in the humidity of the cask atmosphere was determined.

Analysis of the Solid Contents of the Cask

Various methods were used to analyze the ion exchange resins, as described in the following.

Water Exchange

Approximately 50% of the weight of the ion exchange resins is source water. To determine the H-3 activity concentration of this water, it was exchanged with excess H_2O by boiling 10 g of resins in 100 ml of H_2O for a period of 24 hours.

The H-3 activity was then measured on the distilled samples by liquid scintillation.

The incineration of resin samples in O_2 yielded a gas from which both H-3 and C-14 activities could be determined.

Two grams of resins were incinerated in an oven under O_2 at 1000°C. The gas was then led through solutions of H2O, concentrated H_2SO_4, $KMnO_4$, and 2 M NaOH.

The H-3 activity was determined with a distilled sample from the first solution. The C-14 activity remaining in the last solution was precipitated as $BaCO_3$, suspended in a scintillation gel and measured against a $BaC^{14}O_3$ standard.

Regeneration of the Ion Exchange Resins

The C-14 activity of the ion exchange resins is present in anionic form in the ion exchanger. Thus, it must go into solution as Na_2CO_3 when the exchanger is regenerated with NaOH. Approximately 10 g of resin were mixed with 100 ml of 2 M NaOH, the base acidified, the Co_2 released was led into a $Ba(OH)_2$ solution and the $BaCO_3$ measured as described above.

Measurement of the Leak Tightness of the Cask

To measure the leak-tightness of the casks used, integral leak tests were performed on all cask connection points.

For this, the cask was filled with hy designed hood (see Fig. 1). The leak test measurements were made with a He-mass spectrometer. A control measurement was made from the gas chromatographic evaluation of a gas sample.

ANALYSES ON THE CASK WITH RADIUM SOURCES

Due to the varying nature of the sources contained in the cask and since an analysis of the contents is not very informative, the tests performed with this cask were limited to an analysis of the cask atmosphere and of the leak tightness.

Gas Sampling and Analyses

A special test lid was placed on the cask after loading, with which it was possible to take gas samples from the cask cavity. For the analysis of such samples for radium activity, some methods were used, in part new, as described below:

Ra-226 decays to (stable) Pb-206 through a series of α β and decays. An intermediate product along the way is the noble gas isotope Rn-222 with a half-life of $t_{1/2}$ = 3.824 days. Thus, according to the condition of the waste and the storage temperature, a fraction of Rn-222 atoms get into the gas atmosphere in the cask cavity. The aim of the analyses is to determine the concentration of Rn-222 activity in the cask atmosphere.

Rn-222 cannot be determined through gamma spectroscopy. Instead, a gas sample was pumped out of the MOSAIK IIIb cask via a polyethylene gas sack. After several hours, equilibrium was reached between the Rn-222 and its daughter elements Pb-Bi-214. The sack was then empties, and with the aid of an Rn emanation source with a known emanation rate, it was shown that when the sack is emptied, Rn-222 goes out but the non-gaseous products Pb-Bi-214 remain.

Thus, the highly charged sack apparently acts like an electrofilter and retains the products. After emptying, the sack was folded together to the smallest possible volume. The Pb-Bi-2214 activity was immediately determined.

Leak Tightness of the Cask

(Same as in earlier section entitled "Measurement of the Leak Tightness of the Cask.)

ANALYSES ON ADDITIONAL CASK WITH TRITIUM-CONTAINING WASTES

Since the MOSAIK III cask contained tritium wastes of varying composition, the sampling performed later could not assure that the specimens taken were representative for the entire contents. To check the results, additional analyses were performed on unmixed ion exchangers.

For this, a mixed bed filter was examined in a transport cask owned by the KFA.

RESULTS OF THE ANALYSES

Results for the Cask with Tritium-Containing Wastes

Results were obtained for the following analyses:

- Pressure buildup in the cask
- Analyses of samples of resins
- Analyses of water samples
- Analyses of gas samples
- Determination of the empty cask volume
- Determination of the leak tightness of the cask.

The results of these analyses are as follows.

Pressure Buildup

During the time period in which the demonstration was performed, no pressure buildup due to reactions of the cask contents was detected. Pressure variations were due to temperature differences (e.g. after heating the system up to 50°C) and to changes of location.

Analyses of Samples of Resins

Co-60	= 1.04 E07 Bq/kg	0.28 mCi/kg
Cs-134	= 1.11 E05 Bq/kg	0.003 mCi/kg
Cs-137	= 7.77 E06 Bq/kg	0.21 mCi/kg

Analyses of Water Samples

Analysis of the water samples resulted in the following activities:

H-3 in water	= 2.92 E10 Bq/kg	790 mCi/kg of water
H-3 in resins	= 3.7 E08 Bq/kg	10 mCi/kg of resins
C-14 in resins	= 7.4 E-7 Bq/kg	2 mCi/kg of resins

Analyses of Gas Samples

Analysis of the gas from the cask cavity yielded the following:

H-3 as HTO	= 5.92 E04 Bq/cask	1.6 Ci/cask
C-14 as CO2	= 3.37 E05 Bq/cask	9.1 Ci/cask

Determination of the Empty Volume of Cask

The empty volume of the cask was approximately 165 liters.

Determination of the Leak Tightness of Cask

The integral helium leak rate for all cask closures was: Determined with a He-mass spectrometer, 4.1 E-05 mbar 1/sec, determined with gas chromatography of He, 3.9 E-05 mbar 1/sec.

Results for Cask with Radium Containing Wastes

Results are given for the following analyses of the cask with wastes containing radium sources:

- Pressure buildup
- Analysis of gas samples
- Determination of the empty volume of cask
- Determination of the leak tightness of cask.

Pressure Buildup

During the time period in which the demonstration was performed, no pressure buildup due to reactions of the cask contents was detected. Pressure variations were due to temperature differences (e.g. after heating the system up to 50°C) and to changes of location.

Analysis of Gas Samples

From the analysis of the gas in the cask, the activity of Rn-222 and its daughter was measured at Rn-222 + daughters = 6.36 E06 Bq/cask, 172 μCi/cask.

Determination of the Empty Volume of the Cask

The empty volume of the cask was determined to be approximately 470 liters.

Determination of the Leak Tightness of the Cask

The integral helium leak rate for all cask closures was:

Determined with He-mass spectrometer,	8.1 E-5 mbar 1/sec,
Determined with gas chromatography,	3.8 E-5 mbar /1 sec.

CONCLUSION OF THE STORAGE AND MEASUREMENT PROGRAM

After completion of the measurements, the test lid was taken off of the casks and the original lid placed on. The casks are currently stored at the KFA Nuclear Research Center Julich, DE-Dept. It is planned to place the cask in the KONRAD final repository when it is finished.

CONCLUSIONS

The storage and measurement program for the conditioning of wastes containing tritium and radium sources for final storage in MOSAIK III casks showed no reason preventing the use of such casks in a repository.

Besides trying a new method of loading, a series of analyses were performed on the cask, which were in part new and proved themselves to be successful. The analyses were performed both at room temperature and at 50°C, as is anticipate in geological repository. No changes pertinent to final storage were observed due to this elevated temperature.

In the activity measurements of the radioactive wastes, safety factors were found which were far beyond those used in accident analyses, thus, confirming the conservatism there.

Sufficient leak tightness was demonstrated for the closure system of the casks. In the process, the helium leak rate determined on a cask loaded with actual waste with mass spectroscopy was confirmed with gas chromatographic measurements.

Unexpected reactions in the loaded casks and biological transformations, etc. were not observed.

The loaded cask are currently stored at the KFA Nuclear Research Center and are ready for final storage at the KONRAD repository.