LEAKAGE TESTS ON A CONTAINMENT SYSTEM FOR A SOLUTION CONTAINING MOLYBDENUM-99

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Abstract

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According to the 1979 edition of the IAEA Regulations, the calculated Type B(U) permissible leakage for the particular molybdenum-99 liquid solution discussed here is 8.85×10^{-7} mL·h^{-1} and 8.85×10^{-4} mL in one week for normal and accident transport conditions, respectively. In order to demonstrate compliance with containment requirements, the methods specified in ANSI N14.5 to establish leakage test procedures for containment system design, fabrication, assembly and periodic verification were used. However, the authors developed a unique procedure for measuring solution leakage *during* and *after* the thermal test. The paper discusses how the permissible leakage values and the procedures used to demonstrate compliance were established.

1. INTRODUCTION

A packaging for Type B quantities of molybdenum-99 (Mo-99) in a liquid solution has been designed, certified and used for over 2 years between Atomic Energy of Canada's Chalk River Nuclear Laboratories (CRNL) and its Radiochemical Company (RCC) in Ottawa, a distance of about 200 km. Mo-99 is produced at CRNL and bulk quantities are shipped to the RCC where the product is refined and distributed to medical centres.

CRNL has its own Drop Test and Fire Test Facilities and although we completed an assessment for packaging and shipping certification purposes, this paper describes only the containment assessment program. In particular, it describes the methods we used to establish the degree of leaktightness of the containment system both during and after the thermal test. It also describes how the containment system has been designed for remote handling procedures.

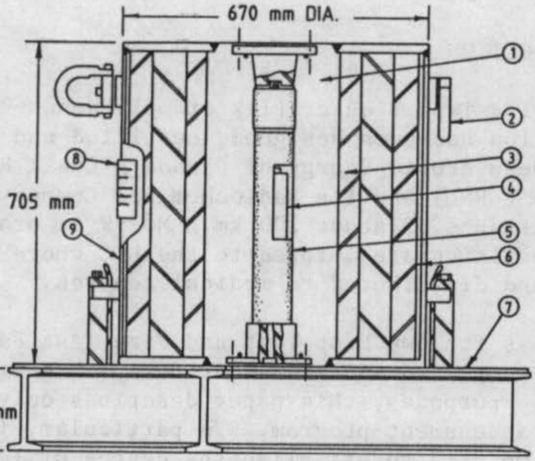
Because leakage test procedures are involved, the requirements and methods specified in ANSI N14.5 [1] played a significant role in our program. Consequently, the term "leaktight", as used in this paper, means a leakage rate equal to or less than 10^{-8} Pa.m^3.s^{-1} for a pressure differential of 100 kPa across the containment boundary.

2. PACKAGING AND CONTAINMENT SYSTEM DESIGN

Fig. 1 shows the essential features of the gamma shielding and containment system packaging components which have a commbined mass of 2265 kg.

The inner stainless steel forging, the 3 concentric carbon steel pipes and the stainless steel shell provide the 300 mm of gamma shielding that are required. This component may be loaded vertically or horizontally and in a storage pool or at the face of a hot cell.

The containment systems (i.e. the product bottles) are made of AISI Type 316 stainless steel and incorporate two closure plugs each with a silicone elastomeric seal. Some steel shielding is provided between the seals and the radioactive material to ensure that the accumulated dose does not exceed the seals' rating of 10^5 Gy. New seals are used for each shipment. For each closure, numerous helium leakage tests were completed to verify that the design, fabrication and assembly procedures would be leaktight for both routine and normal transport conditions.



① ST. ST. FORGING
② SAFETY HOIST RING
③ CARBON STEEL PIPES
④ PRODUCT BOTTLE - 300 mL, 73 mm OD
⑤ DRAWER, 91 mm DIA
⑥ REFRACTORY INSULATION
⑦ PALLET
⑧ NAMEPLATE
⑨ ST. ST. SHELL

FIG. 1. Molybdenum-99 packaging (mass: 2265 kg).

Because the Mo-99 product is loaded in a hot cell, the product bottle closures have to be assembled and leakage tested remotely before each shipment. High temperature elastomeric O-ring seals were selected because simple, reproducible leaktight joints could be attained. Further, calculations and tests showed that the seal area would be exposed to less than 250°C as a result of the thermal test. Both closure plugs are matched to their respective product bottles and they are marked to give a visual indication of proper torque. An extensive series of tests showed that both seals performed satisfactorily under such adverse conditions as dirty seal surfaces and small nicks or cuts in the seals themselves.

One of the items on the check-list for preparation of the product bottle for shipment is a vacuum leakage test. Before each shipment a pressure of 15 kPa absolute is created above the external surface of the inner closure plug and this pressure must not increase by more than 10 kPa in a 5 minute period. Experience shows that the increase does not exceed 1 kPa. An external vacuum leakage test procedure was selected because, during transport conditions, the pressure within the containment system is always positive. The extremely small void volume between the inner and outer closure plugs invalidates a similar vacuum leakage test for the outer seal.

The time required to load the product bottle with Mo-99 solution and complete all shipment preparation procedures is about 45 minutes.

3. CONTAINMENT REQUIREMENTS

The specified payload for the package is 555 TBq of Mo-99. This radionuclide has a half-life of 66.7 h and decays by β^- to Technitium-99m (87.5%) which has a half-life of 6 h and Technitium-99 (12.5%) which is stable. In addition, germanium gamma spectroscopy measurements showed that the payload could contain up to 35 TBq of Iodine-132, which has a half-life of 2.3 h. Fig. 2, which gives the time-activity history for this payload, shows that the radioactive contents change significantly with respect to time. Therefore, before the containment requirements could be defined it was necessary to determine the A_2 value of the mixture and the activity of the contents as a function of time.

The equivalent A_2 value for the mixture, A_{2eq}, is given by

$$A_{2eq} = \frac{\Sigma A_i}{\Sigma (A/A_2)_i} \text{ TBq} \qquad (1)$$

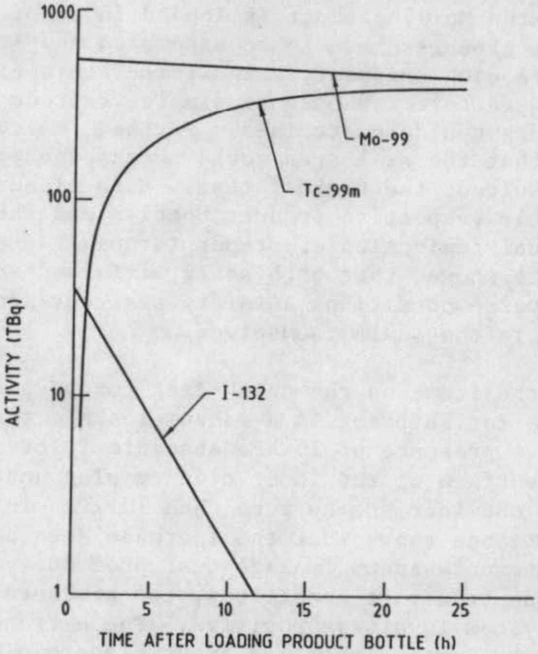


FIG. 2. Time activity histories.

TABLE 1. SOLUTION MIXTURE DATA

Time h	A_2eq TBq	A_S TBq.mL^{-1}	L_N mL.wk^{-1}	Q W
0	2.07	2.81	7.4×10^{-7}	63.7
1	2.39	2.99	8.0×10^{-7}	60.5
3	2.92	3.30	8.85×10^{-7}	56.6
5	3.25	3.53	9.2×10^{-7}	54.5
7	3.45	3.68	9.4×10^{-7}	53.4
10	3.60	3.88	9.3×10^{-7}	52.3
30	3.70	3.71	10.0×10^{-7}	45.0
50	3.70	3.08	12.0×10^{-7}	36.7

where i refers to the ith radionuclide of the mixture, A refers to the activity of a single radionuclide and A_2 refers to the Regulatory value of that single radionuclide.

The specific activity of the contents, A_s, is given by

$$A_s = \frac{\Sigma A_i}{V} \quad \text{TBq.mL}^{-1} \qquad (2)$$

where v is the solution volume, in this case, 210 mL.

Typically, regulatory containment requirements are specified as sub-multiples of A_{2eq} for mixtures but, if the specific activity of the solution mixture can be determined, these requirements can be expressed in volumetric terms. For example, for normal transport conditions,

$$L_N = \frac{A_{2eq}}{A_s} \times 10^{-6} \quad \text{mL.h}^{-1} \qquad (3)$$

where L_N is the normal transport condition permissible volumetric leakage rate. Table 1 gives A_{2eq}, A_s, L_N and Q, the decay heat, as functions of time. The values of L_N show that the permissible leakage rate is a "moving target". For simplification, we assumed a constant rate at time = 3h; thus $L_N = 8.85 \times 10^{-6}$ mL.h^{-1}. A time equal to 3 hours was chosen because it takes about 3 hours for the radioactive decay heat to raise the temperature of the liquid contents to a quasi-equilibrium condition.

Similarly, as per Ref. [2] the following values were assumed for accident transport conditions,

$L_A = 8.85 \times 10^{-4}$ mL.wk^{-1} for a Type B(U) package and

$L_A = 8.85 \times 10^{-1}$ mL.wk^{-1} for a Type B(M) package

4. THERMAL ANALYSES

The temperature and pressure of the liquid contents could affect the leaktightness of the containment system significantly. These conditions of state depend on the radioactive decay heat and the effects of the thermal test. Consequently, we completed tests and calculations to establish time-temperature-pressure relationships for both normal and accident transport conditions. For normal transport conditions, we simulated the radioactive contents with water, installed an immersion heater in the containment system, placed the containment system in the packaging and measured liquid temperatures for various heat inputs. A quasi-equilibrium

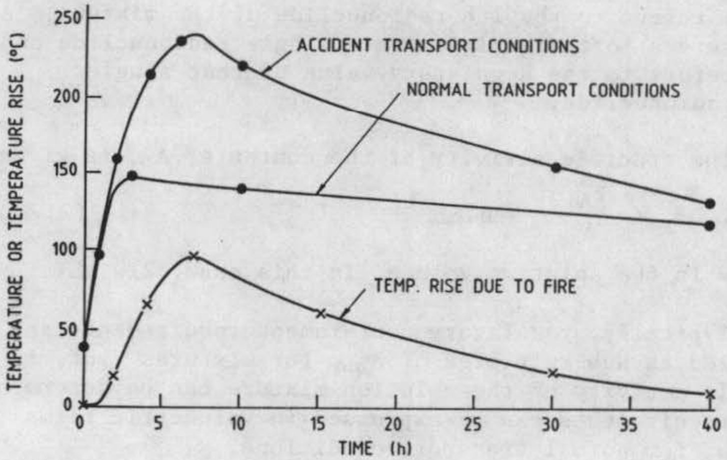


FIG. 3. Measured time–temperature histories.

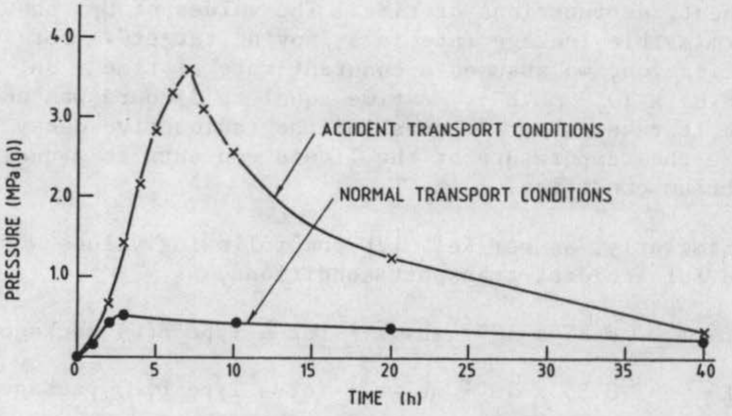


FIG. 4. Calculated time–pressure histories.

state was attained in about 3 hours for each setting. For the thermal test, we disconnected the immersion heater, conducted a 35 minute furnace test as per ASTM E-119 [3] and measured the temperature rise of the liquid contents. Fig. 3 shows the results for the two tests. We established the simultaneous temperature effects of decay heat and the thermal test on the liquid contents by simply superimposing the results of one test upon the other. This result is also shown on Fig. 3. Fig. 4 shows the time-pressure histories that we calculated.

5. DEMONSTRATION OF CONTAINMENT INTEGRITY

For normal transport conditions, we simply completed helium leakage tests and showed that the containment system was leaktight.

For accident transport conditions, we assumed $L_A = 8.85 \times 10^{-4}\ \mathrm{mL.wk^{-1}}$ for a Type B(U) package. The coefficient, 8.85, is the same as that for normal transport conditions and was assumed to be the same for convenience only. If Fig. 3 and Table 1 are reviewed simultaneously, one can deduce that a larger coefficient could be justified. However, any refinement of the coefficient would be of academic interest rather than practical value. Incidentally, 8.85×10^{-4} mL is equivalent to about the volume occupied by the head of a pin.

We then loaded the containment system with 210 mL of heavy water (D_2O) and installed an extraneous cap around the outer closure plug. As shown in Fig. 5, this assembly was placed in an oven and the oven heat was controlled so that the accident transport conditions were reproduced on the liquid contents for periods up to 48 hours. After the test, 3 mL of ordinary water were injected into the extraneous cap, the containment system was thermally cycled and a sample was analyzed for heavy water content. Two tests were completed with a single set of seals; the first with the liquid phase of the heavy water adjacent to the seals and the second with the

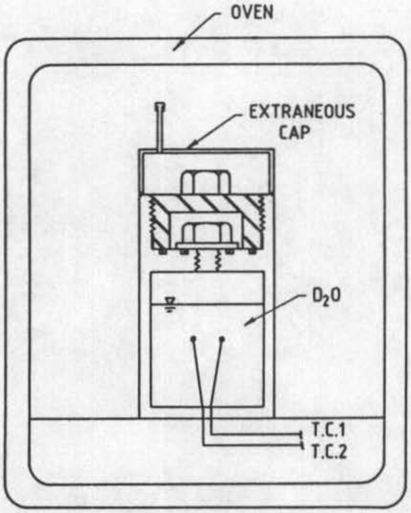


FIG. 5. D_2O leak-test method.

vapour phase adjacent to the seals. Heavy water losses measured 2.48 x 10^{-2} mL and 2.75 x 10^{-2} mL, respectively. Hence, we demonstrated that the containment system complies with Type B(M) requirements, but not Type B(U).

There were many advantages to this test procedure:

a) Containment integrity could be assessed during thermal test conditions

b) The test procedure was simple and reproducible

c) The mass of the packaging did not interfere with the leakage tests.

REFERENCES

[1] AMERICAN NATIONAL STANDARDS INSTITUTE, Leakage Tests on Packages for Shipment of Radioactive Materials, ANSI N14.5, New York (1977)

[2] INTERNATIONAL ATOMIC ENERGY AGENCY, Regulations for the Safe Transport of Radioactive Materials, 1973 Revised Edition (As Amended), Safety Series No. 6, IAEA, Vienna, 1979

[3] AMERICAN SOCIETY FOR TESTING AND MATERIALS, E-119-83, Fire Tests on Building Construction and Materials, ASTM, Philadelphia, (1983)