LEAKAGE OF RADIOACTIVITY DURING TRANSPORTATION OF DEFECTIVE FUEL AND THE FEASIBILITY OF A DOUBLE-CONTAINMENT SYSTEM

*Analytical and experimental evaluation**

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Abstract

LEAKAGE OF RADIOACTIVITY DURING TRANSPORTATION OF DEFECTIVE FUEL AND THE FEASIBILITY OF A DOUBLE-CONTAINMENT SYSTEM: ANALYTICAL AND EXPERIMENTAL EVALUATION.

In order to study the issue of the safe transport of defective spent fuel, the following studies were carried out for LWR fuel contained in a wet-type container. (1) An analytical evaluation, using a scenario and model, of the leakage of radioactive materials from defective fuel and an experimental evaluation of dummy defective fuel with the aim of defining the source term. (2) A theoretical evaluation of the feasibility of a double-containment system in order to establish the transport container's leaktightness.

1. SCENARIO FOR LEAKAGE OF RADIOACTIVE MATERIALS FROM DEFECTIVE FUEL

The scenario used to evaluate the leaking of soluble nuclides (^{134}Cs, ^{137}Cs, ^{90}Sr, ^{125}Sb, ^{144}Ce, ^{106}Ru, $^{239}Pu/^{240}Pu$, etc.) from fuel pellets (important in the evaluation of the containment safety of a wet-type container) is shown in Fig. 1.

* Work partially supported by the Science and Technology Agency, Tokyo, Japan.
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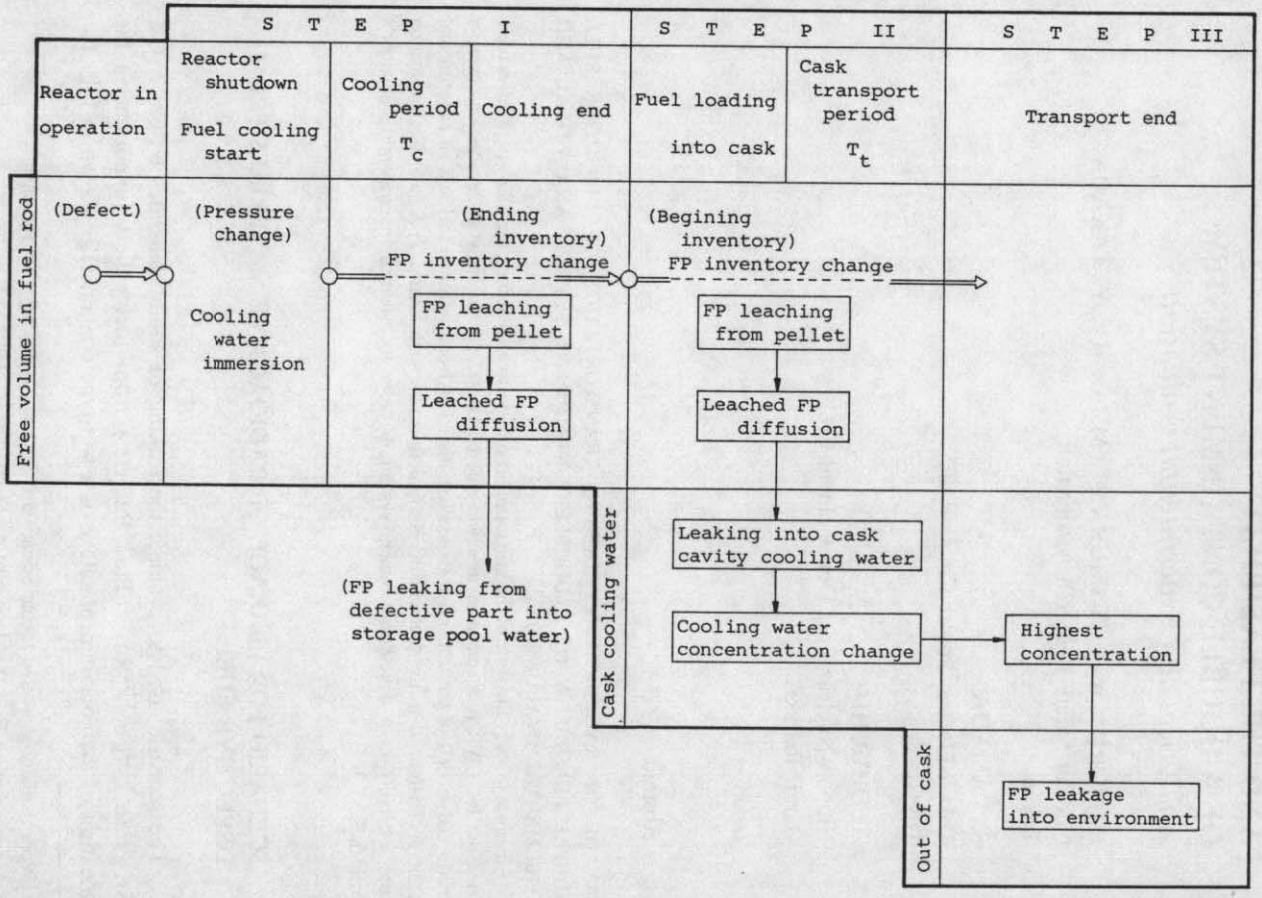

FIG. 1. Scenario for radioactive materials leaking from defective fuel.

2. EXPERIMENTAL EVALUATION OF SOLUBLE NUCLIDE LEACH RATES

In order to determine the leach rates of soluble nuclides, leach tests in deionized water were carried out using fuel pellets prepared from spent BWR fuel rods.

2.1. Materials

Spent fuel fragments and cladding samples were prepared from the 8×8 lattice type of current BWR fuel rods which had been irradiated in the Fukushima Daiichi No. 3 reactor of the Tokyo Electric Power Co., Inc. The burnups of the fuel fragments were approximately 6 GW·d/t U for the series A, 17 GW·d/t U for the series B and 27 GW·d/t U for the series C leach tests. Fractional fission gas release was less than 1% for the fuel fragments of series A and series B tests and approximately 20% for the fuel fragments of the series C leach test. A selected batch prepared from the as-uncladded fuel fragments was used for the leach test in order to make it easy to evaluate surface areas of fuel fragments and amounts of radioactive nuclides involved in the leaching.

2.2. Equipment

Several kinds of equipment were used for the leach tests. Type I was designed to determine the leach rate of soluble nuclides from fuel fragments or claddings into deionized water below 100°C (40, 60 and 80°C). Type II was applied for leach-rate determination above 100°C (110 and 140°C), while type III was designed for a long term leach test (at 25°C for 240 d).

2.3. Analytical procedure

On each sample collection day, the basket holding fuel fragments or claddings was carefully removed and, after swirling the jar of leaching solution, a 10 cm^3 sample solution was withdrawn. This sample was then acidified to approximately pH1 using concentrated nitric acid to prevent the radioactive nuclides from adhering to the jar wall. Gamma ray emitters were measured by gamma-ray spectrometry, while the pure beta-ray emitter (^{90}Sr) was calculated from ^{90}Y ingrowth and the alpha-particle emitters $^{239}Pu/^{240}Pu$ were measured using alpha-ray spectrometry.

2.4. Results and discussions

The integrated release fraction of radioactive nuclides from fuel fragments over 15 days tended to decrease slightly with the increase in water temperature over the range of 80 to 140°C. However, this tendency might have been caused by adhesion of hydrolysed fission products (zirconium, rare earth elements, etc.) to the fuel fragment surface.

The average leach rates for the sampling interval in the long term leach test are shown in Table I. The equation used to calculate the incremental leach rate is:

$$R = a/(A \cdot S \cdot t) \quad (1)$$

where R is the incremental leach rate (g $UO_2 \cdot cm^{-2} \cdot d^{-1}$)
a is the activity of radioactive nuclide in the leachant (μCi)[1]
A is the specific activity of radioactive nuclide in fuel fragments (μCi/g UO_2)
S is the geometric surface area of fuel fragments (cm^2)
t is the incremental leaching period (d).

The incremental leach rates of non-volatile fission products tended to increase slightly with burnup. Those of volatile fission products (such as ^{134}Cs and ^{137}Cs) from fuel fragments and claddings prepared from a fuel rod that experienced high temperature during its irradiation period, however, show remarkably high values because of migration toward the fuel surfaces and cladding inner surfaces. The order of integrated release rate is:

$$^{134}Cs,\ ^{137}Cs > {}^{125}Sb > {}^{90}Sr > {}^{106}Ru > {}^{239}Pu/{}^{240}Pu,\ {}^{144}Ce$$

The leach rate profiles shown in Fig. 2 indicate the presence of two types of leach mechanisms (short term and long term leach mechanisms). The leach rate profile can be described empirically by the following equation:

$$R = R_s \cdot \exp(-B_s \cdot t) + R_l \cdot \exp(-B_l \cdot t) \quad (2)$$

where R is the leach rate (g $UO_2 \cdot cm^{-2} \cdot d^{-1}$)
s is the short term leach mechanism
l is the long term leach mechanism
R_s, R_l are the intercepts at t=0
B_s, B_l are the slopes (d^{-1})
t is the leaching period (d).

3. FISSION PRODUCT INVENTORY CHANGE AND LEAKAGE QUANTITY FROM A SMALL OPENING IN FUEL CLADDING

A small opening in the defective fuel rod is modelled as shown in Fig. 3. Assuming that the leached nuclides move in the immersing water by means of a diffusion mechanism, the change in the fission product inventory (given in the scenario) and the quantity of leakage from the small opening have been calculated.

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[1] 1 Ci = 3.70×10^{10} Bq.

TABLE I. INCREMENTAL LEACH RATE FOR RADIOACTIVE NUCLIDES IN DEIONIZED WATER AT 25°C

		Incremental leach rate (g $UO_2 \cdot cm^{-2} \cdot d^{-1}$)						
Nuclide	Fuel	0-1	2-4	5-15	16-30	31-60	61-120	121-240
Cs-137	A	4.93E-4	2.85E-5	6.86E-6	4.24E-6	7.27E-7	1.89E-6	7.57E-8
	B	3.62E-4	2.23E-4	6.15E-5	5.33E-5	2.96E-6	1.54E-6	1.42E-7
	C	3.46E-2	1.40E-3	2.42E-4	1,18E-4	1.48E-5	7.39E-6	7.39E-6
Cs-134	A	5.55E-4	3.45E-5	7.70E-6	4.39E-6	9.41E-7	2.19E-6	7.84E-8
	B	4.11E-4	2.53E-4	6.88E-5	5.97E-5	4.59E-6	2.10E-6	2.87E-7
	C	4.03E-2	1.46E-3	3.10E-4	1.42E-4	1.42E-5	7.10E-6	7.10E-6
Sr-90	A	4.79E-5	1.94E-5	2.08E-6	6.94E-7	9.02E-7	3.12E-7	1.13E-7
	B	9.63E-5	2.46E-5	8.19E-6	1.53E-6	8.02E-7	1.36E-7	1.33E-7
	C	1.08E-4	8.98E-5	1.03E-5	8.40E-7	5.88E-7	3.36E-7	1.05E-7
Pu	A	—	—	—	—	5.64E-6	—	2.35E-8
	B	—	—	9.32E-7	—	8.10E-8	—	1.80E-8
	C	—	—	2.93E-6	2.38E-7	5.96E-8	—	2.98E-8
Ce-144	A	—	—	—	—	—	—	—
	B	—	—	6.73E-7	—	6.58E-8	—	—
	C	—	—	1.99E-6	5.08E-7	—	—	1.38E-9
Ru-106	A	9.83E-6	4.03E-6	6.36E-7	8.48E-8	5.30E-8	—	5.30E-8
	B	1.35E-5	4.50E-6	6.38E-7	7.20E-8	3.60E-8	6.30E-8	2.70E-8
	C	2.29E-5	1.05E-5	2.08E-7	5.94E-8	7.21E-8	2.12E-8	2.12E-8
Sb-125	A	1.12E-3	5.89E-5	5.36E-5	7.85E-6	9.82E-6	1.96E-6	1.47E-6
	B	1.36E-3	2.59E-4	4.15E-5	8.23E-6	4.12E-6	1.85E-6	1.13E-6
	C	1.64E-3	1.78E-4	7.03E-5	6.83E-6	4.27E-6	4.27E-7	6.40E-7

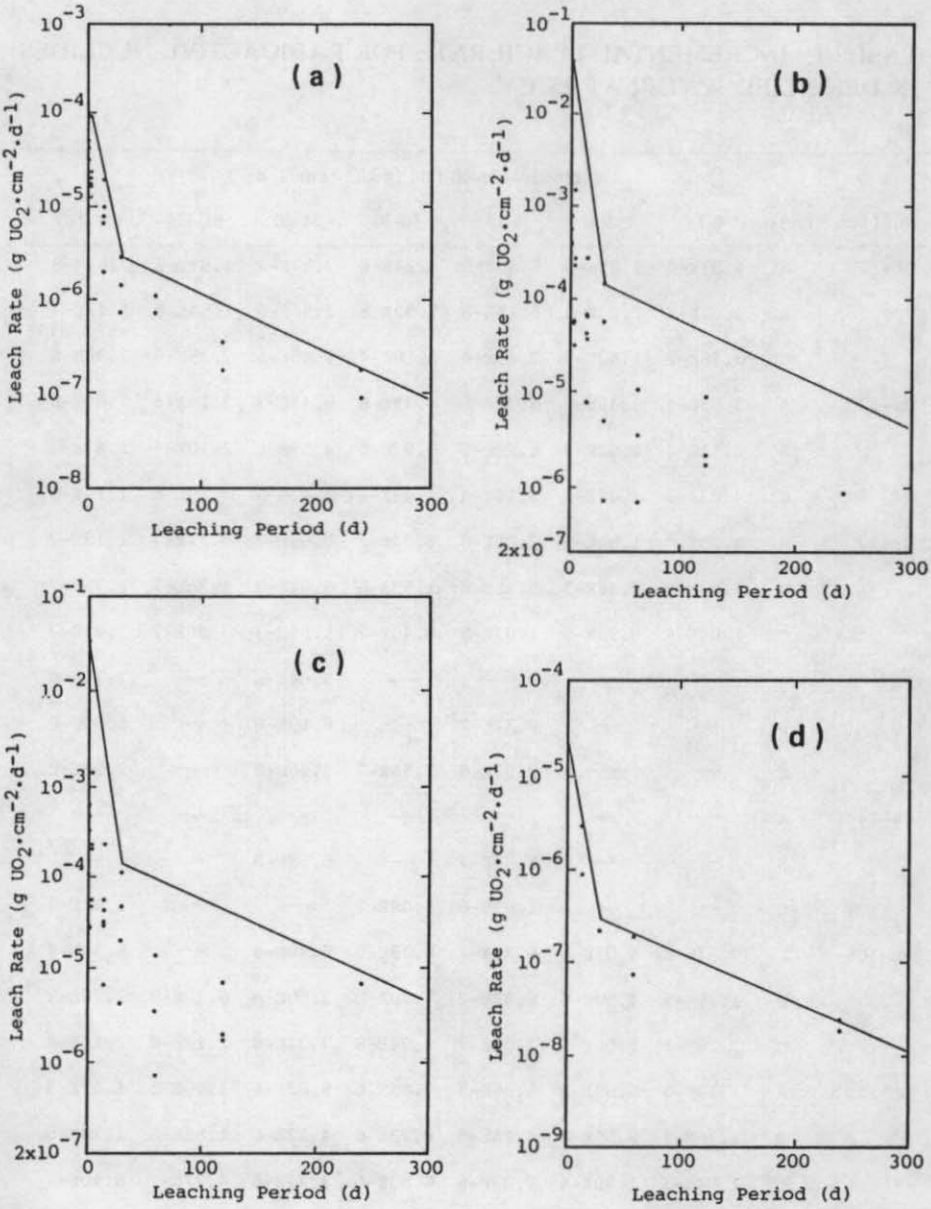

FIG. 2. Leach rate profiles for important radioactive nuclides (a) ^{90}Sr; (b) ^{134}Cs; (c) ^{137}Cs; (d) Pu.

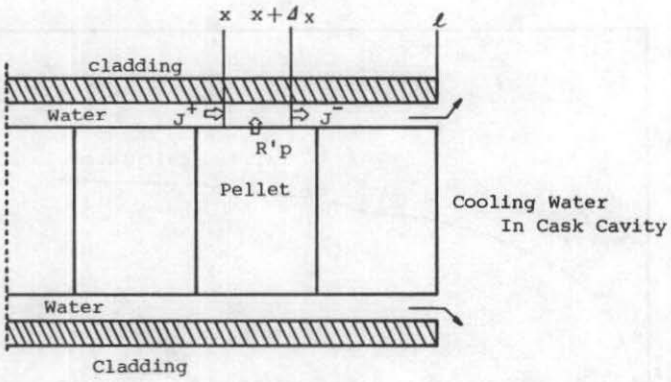

FIG. 3. Model for transfer of radioactive materials from defective fuel rod.

4. EXPERIMENTAL EVALUATION OF LEAKAGE QUANTITY FROM DUMMY DEFECTIVE FUEL ROD

Using dummy defective fuel rods prepared from BWR fuel rods, leach tests in deionized water were carried out for 30 days at 25°C and for the following 30 days at 75°C. Type IV equipment was applied for the leach test of the dummy defective fuel rods.

The integrated leakage quantity of radioactive nuclides from the dummy defective fuel rods with one open end is less than two-sevenths of their release rates from fuel fragments, while that from a pin-hole defect in the dummy fuel rod is less than one-fourteenth of that from the open-ended dummy fuel rod. Finally, the integrated leakage quantity from the well-washed dummy defective fuel rod decreased by less than one-tenth of that for the unwashed dummy defective fuel rod.

Figure 4 shows the leach test results from one open-ended dummy defective fuel rod and the benchmark calculated values. Some conclusions:

(1) The calculated result has a large margin (approximately 15- to 50-fold greater) in comparison with the experimental results.
(2) The difference may be caused by the analytical treatment of the nuclide transfer phenomenon as a simple diffusion mechanism in the very narrow gap between the fuel cladding and the pellets.
(3) The difference can also be explained by the effective solubility reduction of radioactive nuclides from the fuel fragment surface (adhesion of hydrolysed fission products) or by the introduction of the smaller diffusion coefficient value for narrow gap water with hydrolysed fission products.
(4) In a practical evaluation of the quantity of radioactive material that leaks from a defective fuel rod, an approximately tenfold correction factor should be expected.

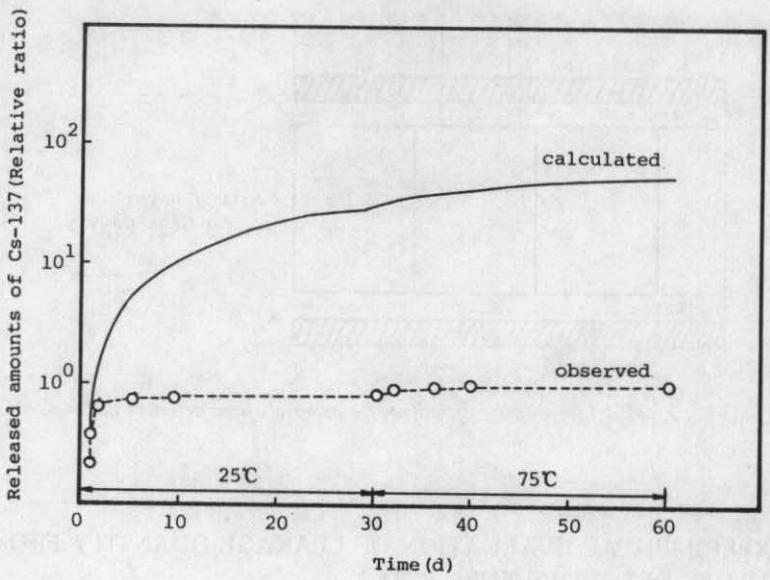

FIG. 4. Time dependency of integrated release quantities of ^{137}Cs from a dummy defective fuel rod.

5. LEAK RATES OF RADIOACTIVE MATERIALS FROM THE TRANSPORT CONTAINER

The leak rates of radioactive materials from the transport container have been evaluated using the following assumptions:

(1) The radioactive nuclides leak from a small opening in the fuel rod and distribute uniformly into the cooling water of the transport container.
(2) The radioactive nuclides leak from the seal boundary at a liquid leak rate, which is peculiar to the transport container.

The equation used to calculate the leak rate of radioactive materials from the transport container is:

$$M_{out}(j) = \frac{M_{cask}(j)}{V_c} \cdot L_R \tag{3}$$

where $M_{out}(j)$ is the leak rate of radioactive material j from the container (Ci/h).
$M_{cask}(j)$ is the quantity of radioactive material j that leaks from a defective fuel rod into the cooling water (Ci).
V_c is the volume of cooling water in the transport container (cm^3).
L_R is the liquid leak rate at the seal boundary (cm^3/h).

The relative ratio of the radioactive material leak rate from the Excellox-3 type container to the requirements for a Type B cask ($=10^{-6}$ A_2) has been calculated to be approximately 2.5×10^{-3} using the following assumptions:

— Burnup : 25 GW·d/t U
— Defects : 10 fuel rods with an opening as shown in Fig. 4
— Cooling : for 500 d in pool (25°C)
— Transportation: for 100 d in container (60°C).

6. THEORETICAL ANALYSIS AND EVALUATION OF THE FEASIBILITY OF A DOUBLE-CONTAINMENT SYSTEM

6.1. Leak rate of double containment

The problem of leakage in a double-containment system is shown in Fig. 5. In its most simple form, the leak rate can be expressed as follows:

$$Q_1 = C_1(P_1^{n1} - P_2^{n1})^{m1}, \quad Q_2 = C_2(P_2^{n2} - P_0^{n2})^{m2} \tag{4}$$

where n and m are indices determined so that flow conductance C might be independent of the pressure. The equations governing leak phenomena are written in the form

$$Q_1dt = -V_1dP_1, \quad -Q_1dt + Q_2dt = -V_2dP_2 \tag{5}$$

Equations (4) and (5) for the given n's and m's may be solved to obtain pressures and leak rates for each boundary. When the flow conductances are prescribed through vacuum testing, and the leak rate is so small that the pressure decrease in the containment cell can be neglected, the standard leak rate of the double-containment system can be expressed as follows:

(a) Q_1, Q_2: molecular flow ($n_1=n_2=1$, $m_1=m_2=1$)

$$Q_2 = \frac{Q_{10}\cdot Q_{20}}{Q_{10}+Q_{20}}\left[1 - \exp\left(-\frac{Q_{10}+Q_{20}}{P_AV_2}\cdot t\right)\right] \tag{6}$$

(b) Q_1, Q_2: laminar flow ($n_1=n_2=2$, $m_1=m_2=1$)

$$Q_2 = \frac{Q_{10}\cdot Q_{20}}{Q_{10}+Q_{20}}\tanh^2\left(\frac{[Q_{10}(Q_{10}+Q_{20})]^{1/2}}{P_AV_2}\cdot t\right) \tag{7}$$

where Q_{10} and Q_{20} denote the standard leak rates of each boundary.

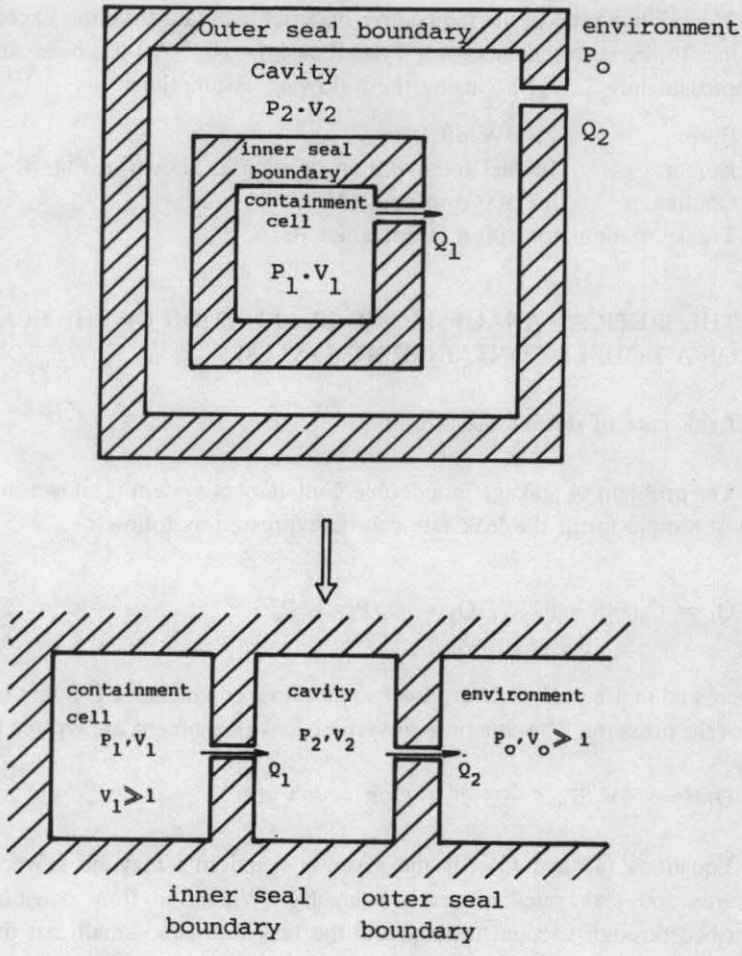

FIG. 5. Model of a double-containment system.

6.2. Results and discussion

Figure 6 shows the effects of the leak rates of the inner and/or outer boundaries to the total leak rate at t = 1 year. For a molecular flow mode, each boundary leak rate contributes equally to the total leak rate. For a laminar flow mode, on the other hand, the leak rate of the inner boundary has a greater influence on the total leak rate. Consequently, it is important to ensure the integrity of the inner seal boundary in order to improve the overall leaktightness of the double-containment system. As can be seen from Eqs (6) or (7), the cavity volume V_2 controls the total leak rate. This

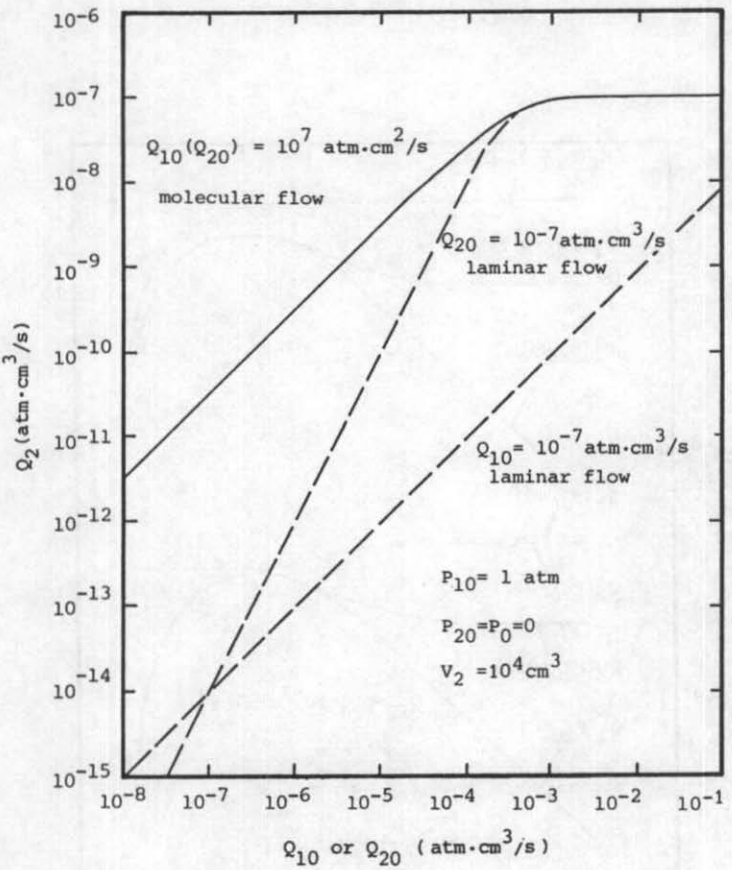

FIG. 6. Contributions of the inner and outer sealing boundaries to the total leak rate (t = 1 year) (1 atm = 1.013 25×10^5 Pa).

means that the cavity volume, which is large enough not to cause a significant pressure increase due to the leak from the inner boundary to the cavity, improves the safety margin.

Furthermore, the initial pressure of the cavity volume (P_{20}) can be considered to dominate the leak rate of the double-containment system. In order to examine the effect of P_{20}, the volume of the containment cell should be treated as being finite. Examples of the numerical results for a molecular flow mode, which actually leads to conservative results, are shown in Figs 7 and 8. In the case shown in Fig. 7, leaked material from the containment cell might be diluted and might then leak into the environment according to Q_2. This means that the substantial leak rate might be

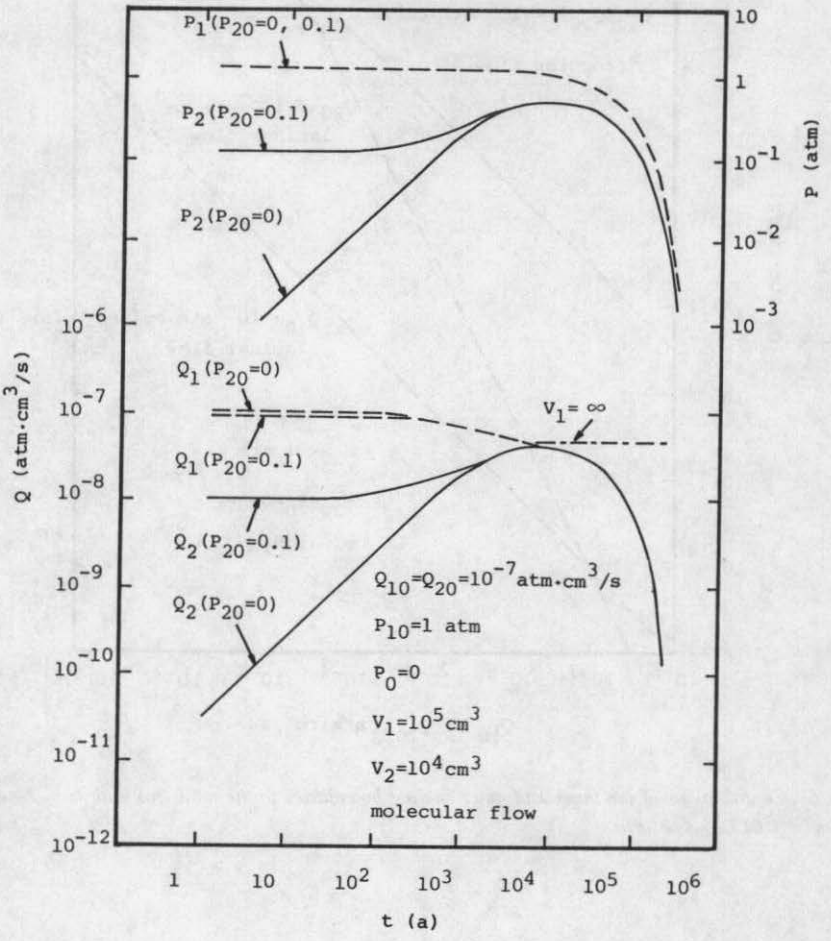

FIG. 7. Cavity pressure and total leak rate ($P_{10} > P_{20}$).

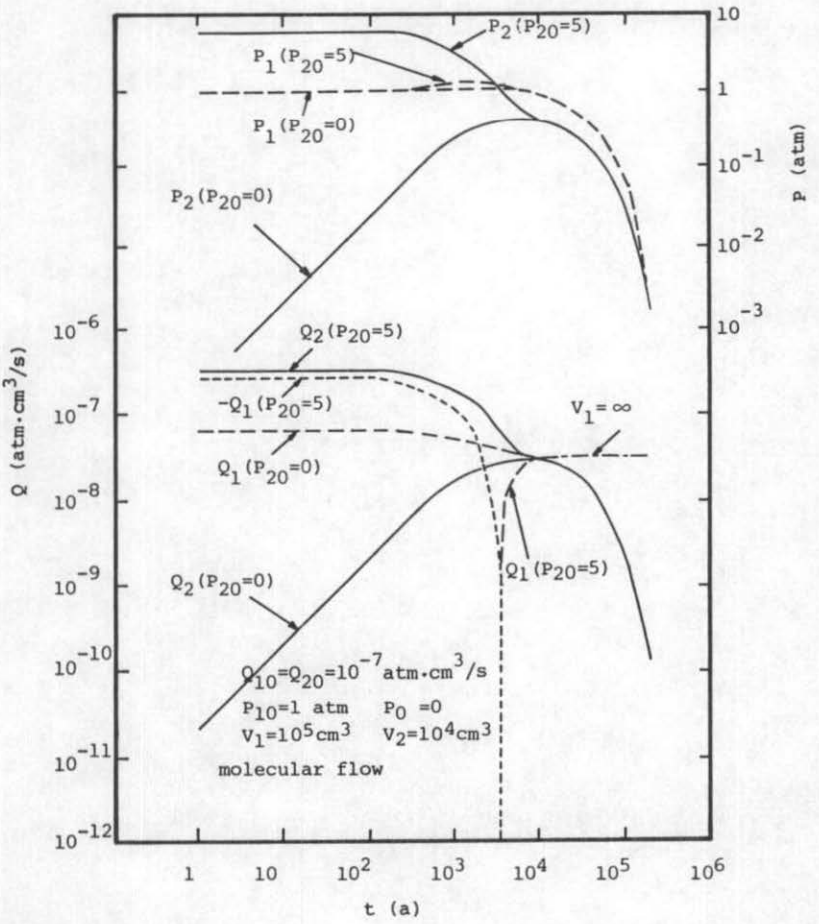

FIG. 8. Cavity pressure and total leak rate ($P_{10} < P_{20}$).

significantly smaller than Q_2. Figure 8 shows the results for $P_{10} < P_{20}$. It should be noted that when t is smaller than about 3500 years, the pressure in the cavity still remains higher than in the inner cell, which means that a leak flow inward exists. It is clear that the leak flow to the environment does not involve confined material.

Although the above discussions deal with somewhat simple cases, the cavity volume of the double-containment system is considered to be useful for controlling the total leak rate. The feasibility of the double-containment system as a positive measure to improve the leaktightness should be investigated in more detail.