EVALUATION METHOD OF GAS LEAKAGE FROM CASKS

M. Aritomi,[1] *N. Li,*[1] *K. Tanaka,*[2] *T. Hayashi,*[2] *T. Noura,*[3] *M. Yokozeki,*[3]
R. Asano[4] *and N. Niyomura*[5]

1 Tokyo Institute of Technology, 2-12-1 Ohokayama, Meguro-ku, Tokyo, 152 Japan
2 Tokyo Electric Power Co., 1-1-3 Uchisaiwai-cho, Chiyoda-ku, Tokyo, 100 Japan
3 Transnuclear Ltd., 1-5-3 Yaesu, Chuo-ku, Tokyo, 103 Japan
4 Hitachi Zosen Co., 1-3-22 Sakurajima, Konohana-ku, Osaka, 554 Japan
5 Ocean Cask Lease Ltd., 2-5-11 Nishishinbashi, Minato-ku, Tokyo, 105 Japan

INTRODUCTION

A sealing function is essential for transportation casks of radioactive materials under both normal and accidental conditions in transportation in order to prevent radioactive materials from being released into the environment. In the safety analysis report, the releasing rate of radioactive materials from the casks into the environment is evaluated using the correlations specified in ANSI N14.5 1977 and 1987. Furthermore, an evaluation method on the leakage rate from the casks is being standardized in the International Organization for Standardization (ISO).

In ANSI N14.5 1987, laminar, molecular and choked flows are taken into account for the evaluation of the gas leakage rate. However, molecular flow is not necessary to be considered for the leakage rate of 10^{-4} to 10^{-2}std·cm^3/s which is related to the sealing performance of transportation casks for radioactive materials. There are still problems of the evaluation method specified in ANSI N14.5 1987 as follows:

1. Since flow patterns in a leak path are classified into laminar and choked flows on the basis of the ratio of a downstream pressure to an upstream one, the evaluated leakage rate becomes discontinuous at the critical pressure ratio with changing the pressure ratio.
2. The compressibility of gas is not considered under laminar flow conditions, so the evaluated leakage rate is proportional to differential pressure between the upstream and the downstream.
3. The units of leakage rates, cm^3/s and atm·cm^3/s, are not universal.

Higson, Vallepin and Kowalewsky pointed out the discontinuity of the leakage rate evaluated according to ANSI N14.5 1987. It is thought that this problem can be solved by considering the exit loss in viscous tube flow as proposed by Santeler. It does not always get a conservative leakage rate to neglect the gas compressibility for laminar flow, as described later in detail. Helium leak tests are carried out on various components and in plants under conditions of atmospheric pressure in the inside and vacuum on the outside, and atm·cm^3/s or Pa·cm^3/s is used as an unit of leakage rate. However, they are converted as the SI unit from torr·l/s which was adopted for indication of vacuum pump performance but are not universal for various pressure conditions and for various kinds of testing gas. These problems cannot be solved for lack of data on the gas leakage with high accuracy.

The purpose of this work is to establish an evaluation method for leakage rate of 10^{-4} to 10^{-2}std·cm^3/s which is related to the sealing performance of transportation casks for radioactive materials. In this paper, leakage rates from very narrow capillary tubes and orifices are investigated experimentally using helium gas to obtain fundamental data for choked flow, non-choked free expansion flow and laminar flow with the gas expansion. Next, an evaluation method of leakage rates of 10^{-4} to 10std·cm^3/s from a very narrow leak path is established. A simplified evaluation method for the leakage rates of 10^{-4} to 10^{-2}std·cm^3/s is proposed. Finally, the points to which attention should be paid for using the evaluation method specified in ANSI N14.5 1987 are discussed.

EXPERIMENTAL APPARATUS

The experimental apparatus was composed of a test tank, a pressure measurement system, a temperature control system, a vacuum pump and a helium gas feeder. Figure 1 shows the test tank. A flange to attach a leak path, a pressure measurement line and a helium gas feeder and vacuum line were attached on the upper plate of the tank. The leak path is simulated by very narrow capillary tubes and orifices. Both a bellow seal valve and a stop valve were installed in each line connected with the tank to minimize the leakage from the experimental apparatus.

The volume of the tank including pressure measurement lines was 459.6cm^3 whose error was within 0.5%. After filling pressurized helium gas in the tank, the leakage rate was obtained by measuring the pressure decreasing rate in the tank through a digital quartz pressure transducer with very high accuracy (full range of 1.4MPa and resolution of 10Pa), and atmospheric pressure was measured with another digital quartz pressure transducer. In this measurement, a temperature change in the tank causes an error of leakage rate. Therefore, the test tank was installed in a isothermal bath filled with water and the whole experimental apparatus was set in an air conditioning room. The change in the temperature in the test tank could be controlled within 0.1K. Consequently, total measurement error of the leakage rate was within 2%.

Mass leakage rate is obtained by

$$\mathrm{G} = (\rho_1 \mathrm{V} - \rho_2 \mathrm{V}) / \delta \mathrm{t} = \rho_1 \mathrm{V} (1 - \mathrm{P}_2 / \mathrm{P}_1) / \delta \mathrm{t}, \tag{1}$$

where V is the volume of the test tank, δt is measuring period, ρ is density and subscripts 1 and 2 mean the beginning and the end of measurement. Mass leakage flux is given by

$$\dot{\mathrm{G}} = \mathrm{G} / \mathrm{A}, \tag{2}$$

where A is the cross section of a leak path. Volumetric leakage rate evaluated at the upstream pressure is defined by

$$\mathrm{L_u} = \mathrm{G} / \rho_{12}, \tag{3}$$

where $\rho_{12} = (\rho_1 + \rho_2) / 2$. Standard volumetric leakage rate is defined by

$$\mathrm{L_{std}} = \mathrm{G} / \rho_{\mathrm{std}}, \tag{4}$$

where ρ_{std} is the density at atmospheric pressure and 298K. Mass-like leakage rate is given by

$$\mathrm{Q} = \mathrm{L_u} \cdot \mathrm{P_a}, \tag{5}$$

where $P_a = (P_u + P_d) / 2$ and P_u and P_d are the upstream and downstream pressures.

Orifices and capillary tubes used for this work are tabulated in Table 1 together with experimental conditions. Their diameters were obtained from electron micrographs.

EXPERIMENTAL RESULTS

Leak from Orifice

The leakage rates from orifices were measured for various upstream pressures. The results are shown in Fig.2. The ordinate in the figure means volumetric leakage rate defined by Eq. (3) and the abscissa does the pressure ratio (P_u/P_d). The volumetric leakage rate increases greatly with an increase in orifice diameter. In the region of the pressure ratio higher than 2.053 where choked flow appears, the volumetric leakage rate is almost constant even for the change in the pressure ratio. **Figure 3** shows the results converted to standard volumetric leakage rates defined by Eq. (4). The standard volumetric leakage rate is proportional to the pressure ratio.

Assuming the release from an orifice as an adiabatic process, mass leakage flux is expressed by

$$\dot{G} = \alpha \left[\frac{2\kappa}{\kappa-1} P_u \rho_u \left\{\left(\frac{P_d}{P_u}\right)^{2/\kappa} - \left(\frac{P_d}{P_u}\right)^{(\kappa+1)/\kappa}\right\}\right]^{1/2}, \tag{6}$$

where κ is the ratio of specific heat and α is the contraction coefficient. Mass choked flux is obtained by

$$\dot{G} = \alpha \left[\kappa \left(\frac{2}{\kappa+1}\right)^{(\kappa+1)/(\kappa-1)} p_u \rho_u\right]^{1/2}. \tag{7}$$

Since the fluid temperature in the upstream is constant in this work, the volumetric leakage rates shown in **Fig.2** are constant even for the change in upstream pressure.

All data under choked flow conditions are replotted as shown in **Fig.4** and an empirical correlation of the contraction coefficient is obtained as follows;

$$\begin{aligned} &\alpha = 0.83P^* \text{ for } 1 < P^* < 22.3 \\ &\alpha = 1 \text{ for } P^* > 22.3, \end{aligned} \tag{8}$$

where $P^* = P_u/P_{cu}$ and P_{cu} is the critical pressure against the downstream pressure (P_d).

Next, **Fig.5** shows a comparison of mass leak fluxes measured for non-choked free expansion flow with Eq.(6). The experimental results of 49.3μm diameter are correlated well with Eq. (6). On the other hand, the experimental results become smaller with narrowing orifice diameter because friction loss becomes significant for 50μm thickness of the orifice as orifice diameter decreases.

The following points are taken into account if the contraction coefficient is assumed as unit: So long as a leakage test is carried out at the highest pressure ratio supposed during the operation of a transportation cask and a leak path diameter is evaluated, the leakage rate evaluated for a lower pressure ratio is conservative. In contrast with this, if the pressure ratio at the testing is less than that for the evaluation, the leakage rate is underestimated.

Leak from Capillary

The leakage rates from capillary tubes were measured for various upstream pressures. The effects of capillary diameter and length on volumetric leakage rate are shown in **Fig.6**. The abscissa in the figure means the differential pressure between the upstream and downstream (P_u-P_d). The volumetric leakage rate increases greatly with an increase in capillary diameter and slightly with a decrease in capillary length. The volumetric leakage rate is proportional to the differential pressure if the gas compressibility is negligibly small for laminar in a leak path flow as specified in ANSI N14.5 1977 and 1987. However, it is seen from **Fig.6** that the volumetric leakage rate is not proportional to the differential pressure. This fact is caused by the gas compressibility and becomes more visible when the leakage rates are replotted with the standard volumetric leakage rate defined by Eq. (4) as shown in **Fig.7**. Since the effect of gas expansion on friction loss becomes lower with a decrease in the differential pressure, the power of the leakage rate to the differential pressure gets to one at low differential pressure. In the contrast with this, it but becomes larger with an increase in differential pressure.

Figure 8 shows the results converted to mass-like leakage rate defined by Eq. (5). It is made clear from figs.2,3,6,7 and 8 that cm^3/s, std·cm^3/s and atm·cm^3/s are not universal as the units indicating leakage rates because these values divided by the differential pressure are not constant for the change in testing condition. Therefore, it is necessary for another universal unit of leakage rate to be set up. We recommend a standard condition indicating the leakage rate universally as follows:

(1) Evaluation fluid is helium gas.
(2) The upstream conditions are 202.6kPa.
(3) 298K and the downstream conditions are 101.3kPa.

EVALUATION METHOD OF LEAKAGE RATE

The pressure drop in viscous tube flow is composed of (1) acceleration loss at the inlet, (2) friction loss and (3) exit loss due to choked flow or non-choked free expansion flow. For flows in a very narrow capillary, acceleration loss can be evaluated by $\triangle P_{acc} = G_2 / \rho$, exit loss can be evaluated by Eq.(6) or Eq.(7) and the friction loss for laminar flow with volumetric expansion can be evaluated by

$$\dot{G} = \frac{(P_u^2 - P_d^2)\rho_u D^2}{64\mu P_u a}, \tag{9}$$

where μ is viscosity, D and a are leak path diameter and length. Equations (6) and (7) include acceleration and exit loss and acceleration loss in capillary flow is much smaller than friction loss and exit loss for choked flow. Therefore, we propose an evaluation method based on Santeler's work. The pressure drop in the capillary is expressed by

$$\dot{G} = \frac{(P_u^2 - P_t^2)\rho_u D^2}{64\mu P_u a}, \tag{10}$$

where P_t means the exit pressure of a capillary. The exit loss for choked flow is presented by

$$\dot{G} = \alpha \left[\kappa\left(\frac{2}{\kappa+1}\right)^{(\kappa+1)/(\kappa-1)} P_t \rho_t\right]^{1/2}, \tag{11}$$

where the contraction coefficient is unit because of choked flow from a tube. The exit loss for non-choked free expansion flow is given by

$$\dot{G} = \alpha \left[\frac{2\kappa}{\kappa-1} P_t \rho_t \left\{\left(\frac{P_d}{P_t}\right)^{2/\kappa} - \left(\frac{P_d}{P_t}\right)^{(\kappa+1)/\kappa}\right\}\right]^{1/2}. \tag{12}$$

If P_t is higher than the critical pressure (P_{cd}),

$$P_{cd} = P_d / \left(\frac{2}{\kappa+1}\right)^{\kappa/(\kappa-1)}, \tag{13}$$

the combination of Eqs. (10) and (11) is applicable to analyze the leakage rate. Otherwise, the combination of Eqs. (10) and (12) is applicable. P_t is obtained only by calculating repeatedly Eqs. (10) and (11) or Eqs. (10) and (12) until the mass leakage flux converges in both equations.

All data of leakage rates from capillary tubes were analyzed by the proposed method and the results are shown in Fig.9. The proposed evaluation method is well correlated with experimental ones. Since a capillary tube is made of glass, its diameter cannot be measured from electron micrographs taken directly. Hence, after gold plating is performed on the inner wall of capillary tubes, electron micrographs were taken. The diameters were underestimated about 5%. Consequently, measured mass leakage fluxes are somewhat greater than calculated ones.

It is seen in the comparison of Fig.2 with Fig.6 that the flow rates in capillary tubes are much smaller than the critical flow rates. Let us examine how much exit loss amounts to the whole pressure loss. All data of leakage rates from capillary tubes were analyzed only by Eq. (9) and the results are shown in Fig.10. It is made clear in comparison between Figs.10 and 11 that the effect of the exit loss on leakage rates is negligibly small for mass leakage fluxes less than $100 kg/m^2 s$

which corresponds to data except 48μm diameter. Namely, it is not necessary for the exit loss to be considered for evaluating the leakage rates less than 10^{-2}std·cm^3/s.

APPLICATION OF ANSI N14.5

In order to clarify the points to which attention should be paid for using correlations specified in ANSI N14.5, the following five cases of evaluation methods were investigated:

(1) Case A is the proposed evaluation method of leakage rate.
(2) Case B is the method based on Hargen-Posseiulle flow for the upstream conditions specified in ANSI N14.5.
(3) Case C is the method based on Hargen-Posseiulle flow for the downstream conditions specified in ANSI N14.5.
(4) Case D is the method based on choked flow specified in ANSI N.14.5.
(5) Case E is the method based on laminar flow considering expansion effect defined by Eq.(9).

Considering the gas expansion effect for laminar flow, the mass leakage rate is calculated by

$$G = \frac{(P_u - P_d)\pi\rho_u}{128\mu} \; \frac{P_u + P_d}{2P_u} \; \frac{D^4}{a}. \tag{14}$$

For Case B, the mass leakage rate is calculated by

$$G = \frac{(Pu - Pd)\pi\rho_u}{128\mu} \; \frac{D^4}{a}. \tag{15}$$

For Case C, the mass leakage rate is calculated by

$$G = \frac{(P_u - P_d)\pi\rho_u}{128\mu} \; \frac{P_d}{P_u} \; \frac{D^4}{a}. \tag{16}$$

Although air is usually used for leakage tests on packages for the shipment, the relationship between leakage rates and leak path diameters was investigated for the following conditions; a working gas is helium, leak path length is 3mm that is widely used in the safety analysis reports and the upstream pressure is 810.4kPa. Calculated results are shown in Fig.11. The following insights are made clear:

1. The choked flow model, Case D, greatly overestimates the leakage rate for a narrow leak path.
2. Concerning Hargen-Posseiulle flow neglecting the expansion effect and specified in ANSI N14.5, Case B always overestimates the leakage rate but Case C always underestimates it.
3. The laminar flow model, Case E, considering the expansion effect, is identical to the proposed model for leakage rates less than 0.5std·cm^3/s.

The expansion coefficient is defined by

$$\beta = 2P_u / (P_u + P_d). \tag{17}$$

For the test conditions,

$$\left(\frac{P_u + P_d}{2P_u} \; \frac{D^4}{a}\right)_{At} = \left(\frac{D^4}{a}\right)_{Bt} \tag{18}$$

where *A*and *B* mean Case A and Case B ,and *t* means testing condition.

If the expansion coefficient for the testing condition is equal to that for the evaluation one, the leakage rates are identical among three cases. If β for the evaluation condition is greater than that for the testing one,

$$\left(\frac{P_u + P_d}{2P_u}\right)_e \left(\frac{D^4}{a}\right)_{At} < \left(\frac{P_u + P_d}{2P_u} \; \frac{D^4}{a}\right)_{At} = \left(\frac{D^4}{a}\right)_{Bt}, \tag{19}$$

where subscript *e* means evaluation condition. Case B evaluates the leakage rate conservatively. On the other hand, if β for the evaluation condition is smaller than that for the testing one,

$$\left(\frac{P_u+P_d}{2P_u}\right)_e\left(\frac{D^4}{a}\right)_{At} > \left(\frac{P_u+P_d}{2P_u}\ \frac{D^4}{a}\right)_{At} = \left(\frac{D^4}{a}\right)_{Bt}, \tag{20}$$

Case B does not evaluate the leakage rate conservatively.

It is questionable whether or not a leak path length less than 50μm exists in transportation casks for radioactive materials. It is thought that a special defect generated during welding might form such a leak path. If the test at the fabrication is carried out under conditions of two different upstream pressures higher than the critical pressure, it is easy to grasp its existence. So long as there is not such a leak path at the fabrication, it may not be necessary to consider the choked flow at the test before the shipment.

CONCLUSIONS

From the present work, the following insights are obtained and clarified:

1. The leakage rates of 10^{-4} to 10 std·cm^3/s can be evaluated by a laminar flow model considering the gas compressibility and choked flow with adiabatic expansion or non-choked free expansion flow. If the pressure at the exit of the leak path is superior to the critical pressure ratio against the back pressure, the former combination is applicable to evaluating the leakage rate. Otherwise, the latter combination is applicable.
2. An empirical correlation of contraction coefficient is obtained for choked flow.
3. For the leakage rates of 10^{-4} to 10^{-2} std·cm^3/s which is related to the sealing performance of transportation casks for radioactive materials, choked flow, non-choked flow with adiabatic expansion and molecular flow are not necessary to be considered.
4. For an application of the Hargen-Posseiulle correlation specified in ANSI N14.5, the following points should be considered: (1) If the expansion coefficient for an evaluation condition is identical to that for the testing condition, the leakage rate agrees perfectly with the value calculated by the correlation based on the upstream or downstream condition. (2) If the expansion coefficient under evaluation condition is greater than that under the testing one, the leak rate evaluated on the basis of the upstream condition is always conservative. Otherwise, the leak rate evaluated on the basis of the upstream condition is not conservative.

This work has been performed at Tokyo Institute of Technology in collaboration with Tokyo Electric Power Co., Transnuclear Ltd. and Hitachi Zosen Co.

REFERENCE

American National Standards Institute (ANSI), Standard for Radioactive Materials - Leakage Tests on Packages for Shipment, ANSI N14.5 1977.

American National Standards Institute (ANSI), Standard for Radioactive Materials - Leakage Tests on Packages for Shipment, ANSI N14.5 1987.

Higson, J. et al. (1989), A Review of Information on Flow Equations for the Assessment of Leaks in Radioactive Transport Containers, Proc. 9th PATRAM, 1989, pp.195-205.

International Organization for Standardization (ISO), Leakage Testing on Pakages for the Safe Transportation of Radioactive Materials, Draft K, 1992.

Santeler, D.J. (1986), Exit loss in viscous tube flow, J. Vac. Sci. Technol. A, 4, 1986, pp.348-352.

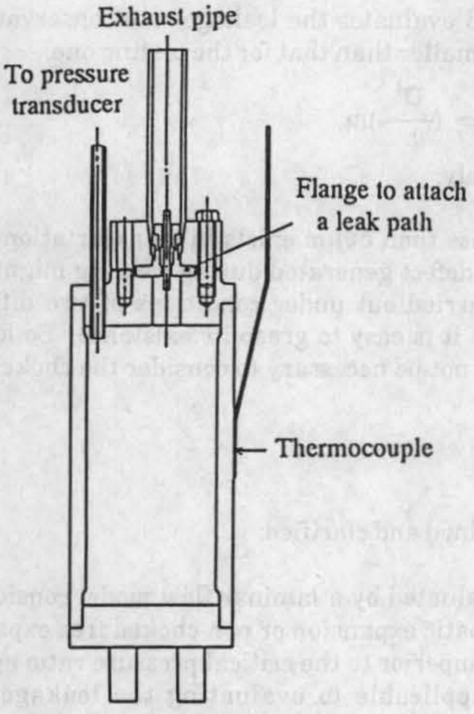

Fig.1 A test tank

Table1 Experimental conditions

Working fluid	Helium gas
Pressure in the test tank	0.11 – 1.1 MPa
Back pressure	Atmospheric pressure
Volume of the test tank	459.6 cm^3
Orifice : Diameter	3.3, 6.0, 10.3, 19.3, 49.3 μm
Thickness	50 μm
Capillary tube : Diameter	11.4, 20.8, 48.0 μm
Length	10, 20, 30 mm

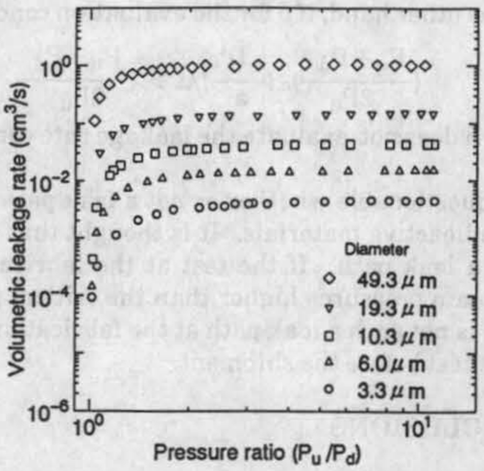

Fig.2 Volumetric leakage rates from orifices

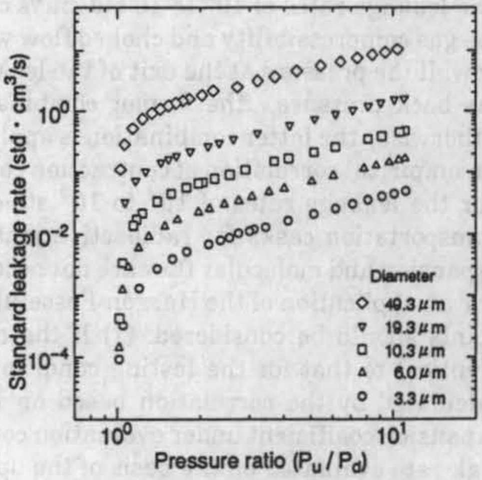

Fig.3 Standard volumetric leakage rates from orifices

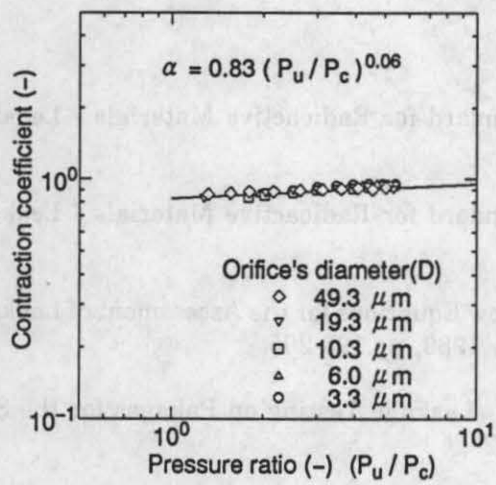

Fig.4 Contraction coefficient for choked flow

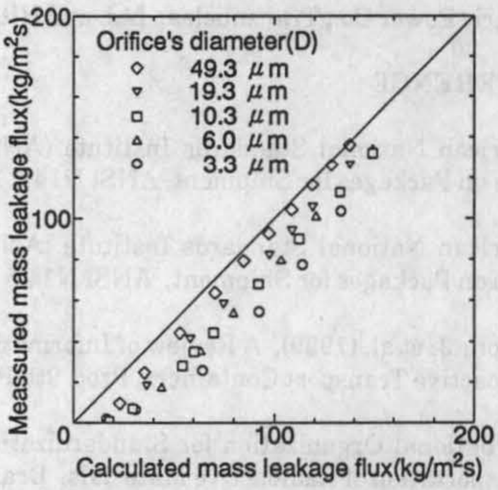

Fig.5 Comparison of mass leakage fluxes of non-choked free expansion flow

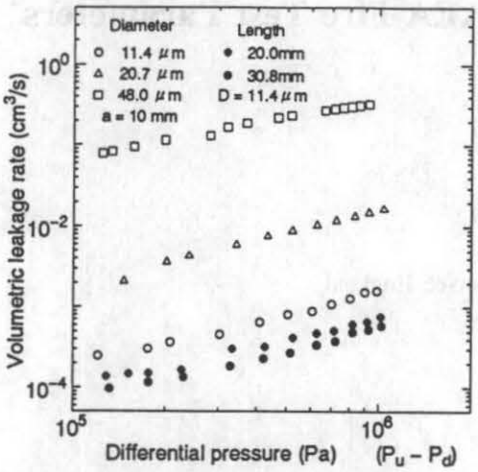

Fig.6 Volumetric leakage rates from capillaries

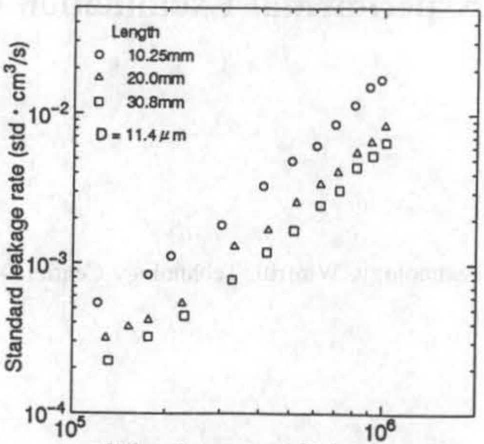

Fig.7 Standard volumetric leakage rates from capillaries

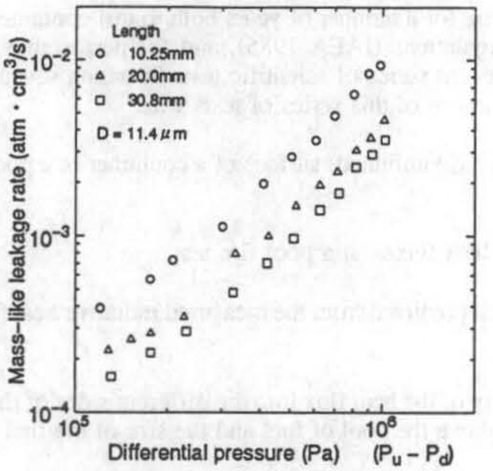

Fig.8 Mass-like leakage rates from capillaries

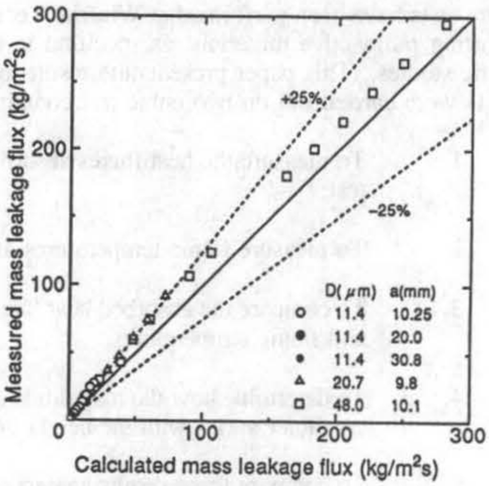

Fig.9 Comparison of the proposed evaluation method with experiments

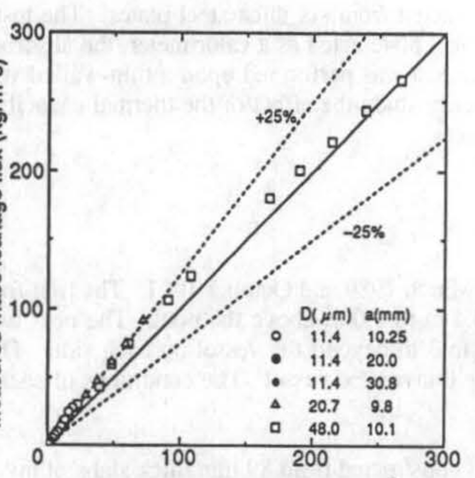

Fig.10 Comparison of laminar flow considering the expansion effect with experiments

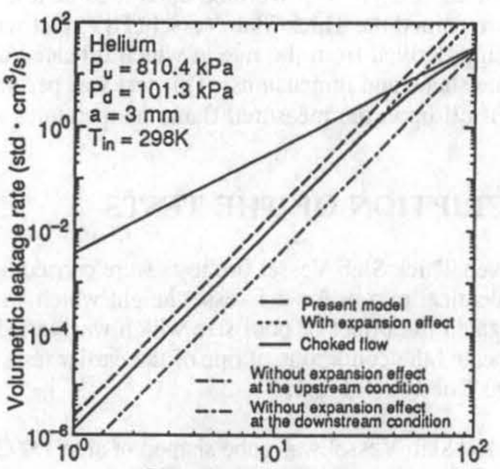

Fig.11 Comparison of analytical results of leakage rates among evaluation methods