# Small Gas Leakage Rate Measuring and Monitoring System

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#### INTRODUCTION

A sealing function of transport and/or storage casks of radioactive materials such as spent fuels is essential to prevent radioactive materials from being released excessively into the environment. The sealing function of a transport cask is inspected at shipment, and the release rate of radioactive materials into environment is evaluated based on the correlation specified in ANSI N 14.5 1987 (ANSI 1987). The inspection and evaluation methods on the leakage rate of gas from a cask is being standardized by the International Organization for Standardization.

Reprocessing the spent fuels and usage of plutonium are the national policy of nuclear energy in Japan. However, the interim storage of the spent fuels may be an alternative option for continuously smooth operation of light water reactors because of world-wide public opinion against the utilization of plutonium, and for a dry storage system such as a storage cask, monitoring the sealing function is essential from safety point of view.

Release of radioactive materials from a transport cask is evaluated from a diameter and length of the leak path obtained from gas leakage rate measured in a pre-shipment test. Gas pressure decrease or increase method is usually applied for the test. In this test, a pressure change rate in a space between double O-rings is measured by a pressure transducer. If the pre-shipment test is carried out under choked flow conditions specified in ANSI N14.5 1987, the leak path diameter evaluated for a choked flow is one order less than that evaluated for a laminar flow. However, the evaluation method of gas leakage discussed at the ISO has deleted the concept of choked flow. Aritomi et al. (1993,1994) also indicated that a consideration for a choked flow is not necessary for the gas leak-age rate of 10<sup>-4</sup> to 10<sup>-2</sup> cm<sup>3</sup>/s through the leak path whose length is 3 mm to 10 mm, corresponding to an O-ring thickness. In addition, since the weight of cask is mostly about 100 tons and its thermal capacity is so large, it takes a significantly long time to reach the thermal equilibrium. And the temperature of the cask is rising during the test. Moreover, the change in an ambient temperature, which varies in a day or in a year, affects the temperature of the cask. For these reasons, the leakage rate is often overestimated. even for a high leaktight cask

Provided that the concept of choked flow is not adopted to the leakage rate evaluation, the criteria of pre-shipment test should be severer, because the leak path diameter which is evaluated for a

laminar flow is larger than that evaluated for a choked flow for a same leakage rate. As a result, it is required for the pre-shipment test to measure much smaller leakage rate. The conventional measuring system used for the pre-shipment test can measure the required leakage rate but is not practical for the actual transport because the period required for the measurement is too long. To cope with the revised evaluation method described above, it is required to develop a new leakage rate measuring system with high accuracy and reasonable measuring time.

The shipping casks are often stored in the temporary cask-storage facility before shipment. When the pre-shipment test is carried out at the facility, the shortening the measuring time is not always required but measuring at many points simultaneously is usually required. In the interim storage facility, multiple casks are monitored their containment functions for a long storage period, and the method for monitoring many points for a long period is required for such facility.

The Research Laboratory for Nuclear Reactors of Tokyo Institute of Technology is developing various kinds of small gas leakage rate measuring systems in cooperation with Hitachi Zosen Corporation and Tsukasa Sokken Corporation. One of them is to measure gas leakage rates directly and is called "flow measuring system" in this work. The other is to measure the pressure decreasing rate induced by the leakage and is called " pressure decreasing rate measuring system". The former is developed for the pre-shipment test for transport casks which can measure gas leakage rate of  $10^{-4}$  to  $10^{-2}$  cm<sup>3</sup>/s with high accuracy. The latter, which can monitor the pressure change at many points, is developed to apply to the pre-shipment test for several casks or monitor pressure changes in the containment boundaries of storage casks.

In this study, concepts and results of calibration for both "flow measuring system" and "pressure decreasing rate measuring system" are reported. The measuring range of both systems is  $10^4$  to  $10^{-2}$  cm<sup>3</sup>/s.

# REQUIREMENTS FOR SMALL GAS LEAKAGE MEASURING SYSTEM

If the choked flow is not applied to the gas leakage evaluation method, the criteria for the preshipment test will be one order smaller ,and, in a severer case, the leakage rate in the order of  $10^{-4}$  cm<sup>3</sup>/s will be required.

The volume of the space between double O-rings of the cask with measuring line usually will be several hundred cubic cm. The leakage during a reasonable measuring period is so small that the pressure in the measuring volume changes slightly. Suppose that the criteria of leakage rate at shipment test is  $5 \times 10^{-4}$  cm<sup>3</sup>/s, the measuring volume is 300 cm<sup>3</sup> in standard condition, and the measuring time is 15 minutes. It is calculated from these requirements that the resolution of the transducer should be less than 150 Pa, which requires an accuracy of  $\pm 0.015\%$  taking into consideration a reading error in practical measurement.

#### CONCEPT OF SMALL GAS LEAKAGE MEASURING SYSTEM.

#### Flow Measuring System.

A concept of a flow measuring system is shown in Figure 1. The system consists of a measurement part, a constant pressure supply system, thermo-couples for measuring temperature in the space between the double O-rings, and the personal computer for data acquisition and processing. The measurement part is composed of a sensor made of a capillary tube, a digital quartz differential

pressure transducer with high accuracy (full scale: 0.2 MPa, resolution: 1Pa, accuracy:  $\pm 0.005\%$ ) to measure the differential pressure between the inlet and outlet of the capillary, a pressure transducer with high accuracy (full scale: 0.7 MPa, resolution: 10Pa, accuracy:  $\pm 0.005\%$ ) to measure pressure of the upper stream of the capillary, and thermo-couples to measure the temperatures of both the upstream and downstream of the capillary. There is a bypass line which is parallel to the measuring part to equalize the pressure in the space between the double O-rings with the back pressure of the measurement part.

After equalizing the pressures of the space with the back pressure of the leakage measurement part, the bypass line is closed, and the differential pressure of capillary, pressure of the upstream and temperatures of both the upstream and downstream are measured. If the gas leakage rate of O-rings exceeds  $10^{-2}$  cm<sup>3</sup>/s, the upstream pressure decreases, and the sealing function is judged to be unacceptable. For the leakage rates of  $10^{-4}$  to  $10^{-2}$  cm<sup>3</sup>/s, it can be measured through the capillary tube. On the other hand, for the leakage rate lower than  $10^{-4}$  cm<sup>3</sup>/s, the measurement accuracy might exceed 0.5%, but the sealing function can be ensured.

It was concluded from our previous works that the gas leakage rate of  $10^4$  to  $10^2$  cm<sup>3</sup>/s can be evaluated by the friction loss of laminar flow with gas expansion as the following correlation:

$$L_{u} = \frac{\pi (P_{u}^{2} - P_{d}^{2})}{256\mu P_{u}} \left(\frac{D^{4}}{a}\right), \tag{1}$$

where  $L_u$  is volumetric leakage rate,  $P_u$  and  $P_d$  are the upstream and downstream pressures,  $\mu$  is viscosity, and D and a are a capillary diameter and length. (D<sup>4</sup>/a) is the sensor coefficient and is calibrated before measurement as mentioned later. Since the change in temperature in the space between double O-rings induces a pressure change, the pressure is compensated by the personal computer using the temperatures measured simultaneously.

# Pressure Decreasing Rate Measuring System

Sometimes several transport casks are temporally stored in a cask storage facility before their shipment, where the system which can measure the leakage rates of casks simultaneously is required. Moreover, storage casks are requested during a interim storage period of 20 to 40 years to monitor their containment functions. In these cases, a monitoring system, which can measure the leakage rate of multiple points of the casks simultaneously, is required.

In this work, a system as shown in Figure 2 is proposed to measure the pressure at multiple points simultaneously. The system has an A/D converter with a scanner to accumulate signals from sensors by scanning. Generally speaking, the zero point and gain of the pressure transducer is drifted by the change of temperature. To avoid the drift, a diffusion type pressure transducer with a polysilicone strain gauge is adopted for the measuring system. To remove the drift of a DC amplifier, no amplifier is used, but the signal is directly accumulated to the personal computer through an A/D converter. The pressure transducer achieved a 600,000-hour MTBF (Mean Time Between Failure) in the continuous usage at 85°C.

The change in the pressure induced by the change in the temperature is compensated by the following formulas.

$$G = \frac{V(\rho_1 - \rho_2)}{\delta t}$$
(2)  
=  $\frac{V(P_1 / T_1 - P_2 / T_2)}{R \delta t}$ , (3)

where G is a mass leakage rate, V is a volume of tank for calibration, P means pressure, T means temperature,  $\rho$  is density, R means the gas constant,  $\delta$  t is measuring period, and subscripts 1 and 2 mean beginning and end of measurement. The volumetric gas leakage rate for the upstream condition, L<sub>u</sub> is:

$$L_{u} = \frac{2V(P_{1} / T_{1} - P_{2} / T_{2})}{\delta t(P_{1} / T_{1} + P_{2} / T_{2})},$$
(4)

In this system, the estimated total accuracy is  $\pm 0.25\%$  including linearity of the sensor and the drift with a change in temperature of -20 °C to 80°C.

#### CALIBRATION AND ADAPTABILITY FOR FLOW MEASURING SYSTEM

### **Apparatus and Method for Calibration**

The characteristic value of the capillary tube  $(D^4/a)$ , which is the coefficient of the sensor of the measuring system, was calibrated using the experimental apparatus shown in Figure 3 which was composed of a test tank, a pressure measuring part, a temperature control part, a vacuum pump and a gas feeder. The detailed information of the test tank is shown in Figure 4. A pressure measurement line, a test gas feeder and vacuum lines , and a flange to install the capillary tube were attached on the upper plate of the test tank. O-rings made of Viton were used for sealing of the flange. The volume of the tank including pressure measurement lines was 459.6cm<sup>3</sup> with error of  $\pm 0.5\%$ . The gas leakage rate was calculated from the pressure change in the tank during measuring period whose measuring error was less than  $\pm 0.5\%$ .

The decreasing rate of the pressure in the tank was measured by a digital quartz pressure transducer with very high accuracy (full range of 0.7MPa and resolution of 1Pa), and the back pressure, which was atmospheric, was measured with another digital quartz pressure transducer (full range of 1.4MPa and resolution of 10Pa). The measurement error was within 0.005% as long as they were calibrated once a year. The accuracy of this measurement method was confirmed for the actual volume measuring system with gas for plutonium nitrate solution. From such a countermeasure, total measurement error of within  $\pm 2\%$  was obtained.

In the case of the leakage rate measurement for the range of  $10^{-4}$  to  $10^{-2}$  cm<sup>3</sup>/s, the change in temperature induces a significant measuring error as follows:

PV = nRT,	(5)
$\delta PV = nR\delta T,$	(6)
$\frac{\delta P}{\delta T} = \frac{\delta T}{\delta T}$	(7)
P T	(,)

As the measurement is carried out at 300K, temperature change of 1K induces pressure change of 1/300. Therefore, the test tank was installed in a isothermal bath filled with water, and the whole experimental apparatus was set in a room at a constant temperature. The temperature change in the

test tank could be controlled within 0.1K. The measurements were carried out several times, and the averaged value was adopted as the leakage rate.

Pressure boundaries of the experimental apparatus consists of weld, fittings, and valves. Background leakage rate of the system should be two order less than the measured value. Especially for the leakage rate of 10<sup>-4</sup> cm<sup>3</sup>/s, it should be less than 10<sup>-6</sup> cm<sup>3</sup>/s. In a pressure measurement line, both a bellows seal valve and a stop valve were installed to minimize the leakage from the experimental apparatus. After pressurized gas was filled in the tank with all valves closed except the valves in measuring line, it was confirmed that the leakage rate is less than 10<sup>-6</sup> cm<sup>3</sup>/s after 72 hours (259,200 second).

After attaching the capillary to the measurement tank, gas was filled into the tank, and its pressure was regulated to a desired value. Leaving the system until the temperature became constant over several hours, a leakage rate from a capillary (sensor) was obtained by measuring pressure decreasing rate in the tank. When gas except air was used as working fluid, the system was replaced with working gas three times.

### **Results of Calibration**

The coefficient of the sensor was measured using air, helium, argon, and nitrogen in rather high pressure difference condition; that is, at the high flow rate to obtain the high accuracy value. Measured results are shown in Figure 5. It is seen from the figure that the sensor coefficient is  $3.050 \times 10^{-17} \text{ m}^3$ . It should be clearly noted that one working gas is enough to determine the coefficient. The adaptability of the sensor was examined using air as shown in Figure 6. It is confirmed from the figure that gas leakage rates of  $10^{-4}$  to  $10^{-2} \text{ cm}^3$ /s can be measured with the accuracy of  $\pm 0.2\%$ . The wider measurement range of flow rates can be achieved, provided that lower accuracy is permitted.

# CALIBRATION AND ADAPTABILITY FOR PRESSURE DECREASING RATE MEASURING SYSTEM

A diffusion type pressure sensor with a polysilicone strain gauge was used which has superior temperature characteristics up to 85°C and has accuracy and stability for long measuring period. The temperature characteristic and measuring accuracy were examined in this work. The results of calibration of the transducer as the relative error to the full scale of pressure are shown in Figure 7.

Generally speaking, zero point and gain of pressure transducer vary with the change in an ambient temperature. The transducer has a function to ensure this temperature drift. To ensure this function, the temperature dependency was measured in the range of  $-20^{\circ}$ C to  $80^{\circ}$ C at 600 kPa abs. The results are shown in Figure 8, which indicates that the function of the transducer is satisfactory. The temperature drift of the pressure transducer against the several working pressures were also measured, and the results are shown in Figure 9. It can be seen from the figure that the measurement errors were within  $\pm 0.25\%$ , which is within the target. From these results, reproducibility, linearity, and temperature characteristic of the sensor were extensively evaluated, and it was confirmed that the sensor has sufficient accuracy at the temperature range of  $-20^{\circ}$ C to  $80^{\circ}$ C.

## CONCLUSIONS

The flow measuring systems were developed to measure a small gas leakage rate of 10<sup>-4</sup> to 10<sup>-2</sup>



Figure 2 Concept of pressure decreasing rate measuring system



Figure 3 Measuring apparatus for calibration





cm<sup>3</sup>/s, which is required for the sealing performance of the casks for a pre-shipment test. It was clarified from this work that the leakage rate could be measured for the desired period with the required accuracy. In addition to the flow measuring systems, the pressure decreasing rate measuring system was developed to measure gas leakage rate simultaneously at several points of a transport cask at the pre-shipment test and to monitor the containment functions at multiple points of storage casks. It was clarified from this work that the pressure decreasing could be detected with an accuracy of  $\pm 0.25\%$  at the temperature range of  $-20^{\circ}$ C to  $80^{\circ}$ C.

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Figure 1 Concept of flow measuring system