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Monitoring of helium gas leakage from canister storing spent nuclear fuel: Radiological consequences and management

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Key Words: Spent nuclear fuel storage, Chloride-Induced Stress Corrosion Cracking, Helium gas leakage monitoring, Radiological consequence, Allowable leakage

ABSTRACT

Chloride-induced stress corrosion cracking (CISCC) of canisters has been identified as a potential degradation mechanism requiring ageing management — especially in a marine air environment. Methods for monitoring helium gas leakage from canisters have been developed on the basis of a phenomenon that the bottom temperature of the canister increases as the top temperature of the canister decreases when helium gas leaks during storage. In this paper, we consider the radiological consequences of CISCC based on two scenarios of helium gas leakage and propose effective management that includes a timeline for monitoring and mitigation. Monitoring helium gas leakage by measuring the difference in temperature between canister surfaces in the dry cask storage of spent fuel (SF) can detect leakage before SF undergoes oxidation as a result of air ingress and before country-specific regulatory limits of off-site doses are exceeded, thereby enabling mitigatory action.

1. INTRODUCTION

Spent nuclear fuel may be stored in dry concrete casks with welded stainless-steel canisters for extended periods. Chloride-induced stress corrosion cracking (CISCC) of canisters has been identified as a potential degradation mechanism, requiring ageing management — especially in a marine air environment. [1–3].

Chu has noted that investigations into the potential radiological consequences of a through-wall crack in a canister resulting from CISCC can help utilities assess ageing management actions for potential CISCC of welded stainless canisters [4]. Chu has also summarized and evaluated available references on the radiological consequences of CISCC under scenarios of air ingress, which causes oxidation of spent fuel (SF) in the canister and propagation of CISCC through the canister wall [5]. Various nondestructive evaluation (NDE) techniques to detect CISCC have been developed by using special devices, including a remote-controlled robot system for a single cask [6]. Such NDE techniques require preparation, special equipment, and risk management of radiation exposure for every inspection. Because accessibility to the welded surface inside some storage modules is limited, managing the effects of cracking may not be practical without additional efforts to retrieve the canister from the storage module or overpack. An alternative strategy to inspection relies on monitoring leakage to ensure timely detection of a potential breach of the canister confinement boundary to prevent degradation of the contents in the storage canisters.

Takeda et al. have developed methods to monitor helium gas leakage from vertical canisters based on a phenomenon in which the temperature at the canister bottom increases as the temperature at the top of the canister decreases when helium gas leaks during storage [7–9]. Once the monitoring system is set up with the dry concrete casks, the system self-monitors and detects temperature changes due to helium loss caused by CISCC during storage. However, one concern is that the rate at which helium gas leaks as a result of CISCC may be too low to be detected by using the temperature-difference monitoring method during extended storage. The practicality of the method to monitor the allowable leak rate of helium has not been well studied.

Researchers at the Central Research Institute of Electric Power Industry (CRIEPI) and Argonne National Laboratory (Argonne) began collaborating in 2019 on the technology development and demonstration of a canister gas-leakage detection system based on temperature information using Argonne National Laboratory's ARG-US Remote Area Modular Monitoring (RAMM) system [10]. CRIEPI is conducting a demonstration of RAMM for the detection of gas leakage from canisters by using a 1/4.5-scale model cask described in detail in reference [7]. The results obtained from the initial set of scale-model experiments showed that gas leakage can be detected within hours by using a RAMM device for canister surface temperature measurement (TM), or RAMM-TM [11].

In this paper, we consider the radiological consequences of CISCC based on two scenarios of helium gas leakage from canisters. The first scenario is monitoring of leakage against a threshold to prevent SF oxidation. If such a pressure drop could be detected soon after leakage begins, mitigatory action could be taken to avoid air ingress into the canister and SF oxidation. The second scenario is monitoring of leakage to ensure that the leakage does not reach the regulatory limit for an offsite dose. If a pressure drop could be detected after a leakage due to CISCC, then necessary action could be taken before the regulatory limit is exceeded. We also propose a timeline for monitoring and mitigation for each scenario.

2. PURPOSE

We discuss the practicality of the temperature-difference monitoring for detecting helium gas leakage due to CISCC to ensure that SF in the canister will not undergo oxidation by air ingress (scenario 1) or that the radiation dose at the site boundary, or fence line, will not exceed the regulatory limit (scenario 2), without the need to retrieve the canister from the storage module or overpack.

3. METHODS

The current work investigates the practicality of the temperature-difference monitoring method and is based on published studies of CISCC flaw growth with time and the radiological consequences of leakage due to CISCC from welded stainless-steel canisters used for dry storage.

For scenario 1, we need to understand the ageing degradation mechanism of SF, obtain the pressure drop curve with time from literature, set up a threshold value of canister pressure before air ingress into the canister, and determine a time frame for mitigatory action. For scenario 2, we need to understand the ageing degradation mechanism of the canister due to CISCC and determine the leakage rate and the radiological consequences (total effective dose equivalent [TEDE]). Using that knowledge and information from literature, we can calculate an allowable leakage rate for the country-specific (Japan and the United States, in this example) regulatory limit for a SF storage facility, as well as the allowable pressure drop, and determine a time frame for mitigatory action.

These calculations will confirm that early detection and mitigatory action are possible before SF oxidation occurs, or a regulatory radiation dose limit is exceeded.

4. LEAKAGE EVALUATION

Table 1 provides details of the two scenarios for canister leakage monitoring.

Table 1. Leakage Monitoring Scenarios

Parameter	Scenario 1	Scenario 2
Objective	Evaluate time for oxidation of SF rods by air ingress from CISCC of the canister	Evaluate radiological consequences due to helium gas leakage from canister due to CISCC
Description	CISCC begins on the canister outer wall → CISCC penetrates the canister wall → He gas leaks out → Air ingresses → SF rods oxidize	CISCC begins on the canister outer wall → CISCC penetrates the canister wall → He gas with radioactive materials leaks to environment → Radiation exposure to the public
Assumptions	• SF rods are intact • Leakage rate: 28.7 cm^3/s • COA: 4E-5 cm^2 • Backfill gas temp.: 423 K	• 1% failure of SF rods • Leakage rate: 9.5E-6 cm^3/s • COA: unknown • Backfill gas temp.: 673 K for fuel cladding of long-term, normal condition design temperature limits
Reference	EPRI report [12]	NRC's best estimate [13]

4.1. Scenario 1: Monitoring of leakage limited by SF oxidation

If the CISCC penetrates the canister wall, the helium gas leakage would begin because of the difference between the internal and external pressures of the canister. The leakage continues until the pressure difference becomes zero. The example calculation for a HI-STORM MPC (multipurpose canister) showed this duration was 52 days, assuming a crack opening area (COA) of 4E-05 cm^2 [12]. After the canister interior reaches atmospheric pressure, the helium gas will continue to leak and be gradually replaced with air. This process is driven by the temperature variation of the surrounding atmosphere. In some cases, 90% of the helium gas in the canister is replaced with air within a year [12].

The air ingress into the canister will affect heat removal, resulting in a temperature increase in the SF. Oxidation of SF rods cladding by air at elevated temperature should be avoided. Therefore, pressure drops in the canister caused by leakage due to CISCC should be detected early by monitoring before the internal pressure reaches atmospheric pressure.

The temperature-difference monitoring method can be used to detect this pressure drop. Figure 1 shows canister pressure drop with time due to CISCC (reproduced with reference to the results of leakage analysis [12]). If such a pressure drop can be detected in, for example, 10 days after leakage begins, action could be taken in the next 40 days to avoid air ingress into the canister and prevent SF oxidation. Scenario 1 focused on the effect of air ingress on SF oxidation, assuming the SF assemblies are intact [12]. Therefore, no radiological consequences due to helium leakage from the canister were considered until air ingress.

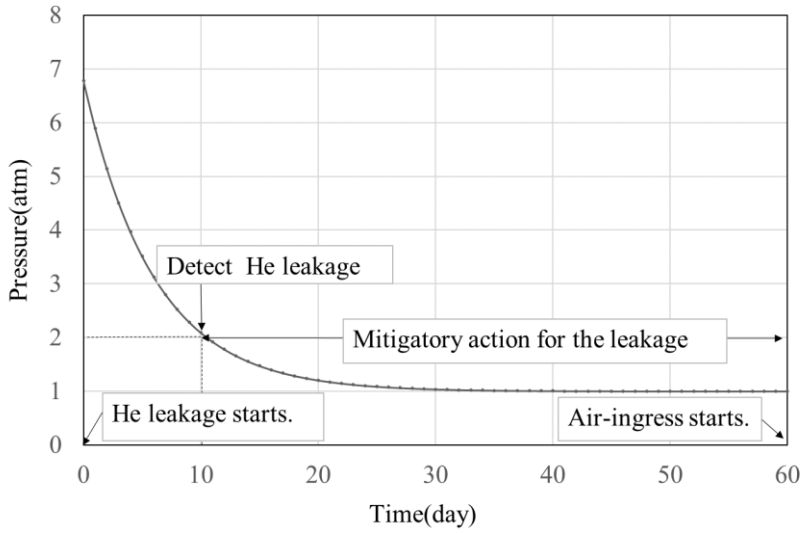

Figure 1. Example of Canister Pressure Drop with Time due to CISCC (Scenario 1)

4.2. Scenario 2: Monitoring of leakage limited by offsite dose

The United States Nuclear Regulatory Commission (NRC), Office of Nuclear Regulatory Research, published "Best-Estimate Offsite Dose from Dry Storage Cask Leakage." [13] This report evaluated the offsite radiological consequence from dry storage cask storage for a Private Fuel Storage (PFS) facility by using the RADTRAD

reactor accident analysis code. Although this report did not explicitly indicate leakage word was due to CISCC, the result could be applied to the radiological consequence due to CISCC [5]. Chu's analysis assumed 1% failure of the SF rods under normal condition in the canister, which would be a conservative source term for the radiological consequence to people at the site boundary. Figure 2 shows the steps for calculating leakage rate due to CISCC limited by offsite dose, including allowable leakage rate under normal condition of storage.

Table 2 shows the key dose assessment values of a concrete cask system (HI-STORM 100 cask system [14]) and the calculated results on allowable leakage rate and pressure drop. The helium gas temperature was assumed to be the fuel cladding temperature for a long-term normal condition design temperature limit. Assuming that the radiological dose is proportional to the helium leakage rate, the allowable leakage rate for the regulatory offsite dose limit is given by equation 1.

Equation 1: $L_{allow} = L_m \times$ TEDE of regulatory limit/TEDE at L_m
where L_{allow} = allowable leakage rate, std atm/y

The helium leakage rate from canister is given by equation 2 [15].

Equation 2: $L_m = VT_s/3{,}600HP_s \times (P_1/T_1 - P_2/T_2)$ std•cm^3/s
where L_m = measured leakage rate, std•cm^3/s
V = gas volume in test item, cm^3
T_s = standard temperature, 298 K
H = test duration, hours
P_s = standard pressure, 1 atm abs
P_1 = gas pressure in test item at start of test, atm abs
P_2 = gas pressure in test item at end of test, atm abs
T_1 = average gas temperature at start of test, K
T_2 = average gas temperature at end of test, K
It is assumed that $T_1 = T_2$.

Equation 2 can be rewritten as equation 3.

Equation 3: $\Delta P = P_1 - P_2 = 365 \times 24 \times L_m \times T_1 \times 3{,}600P_s / VT_s$ atm abs

The allowable pressure drop ΔP_{allow} is obtained by replacing the L_m with L_{allow} in eq. 3.

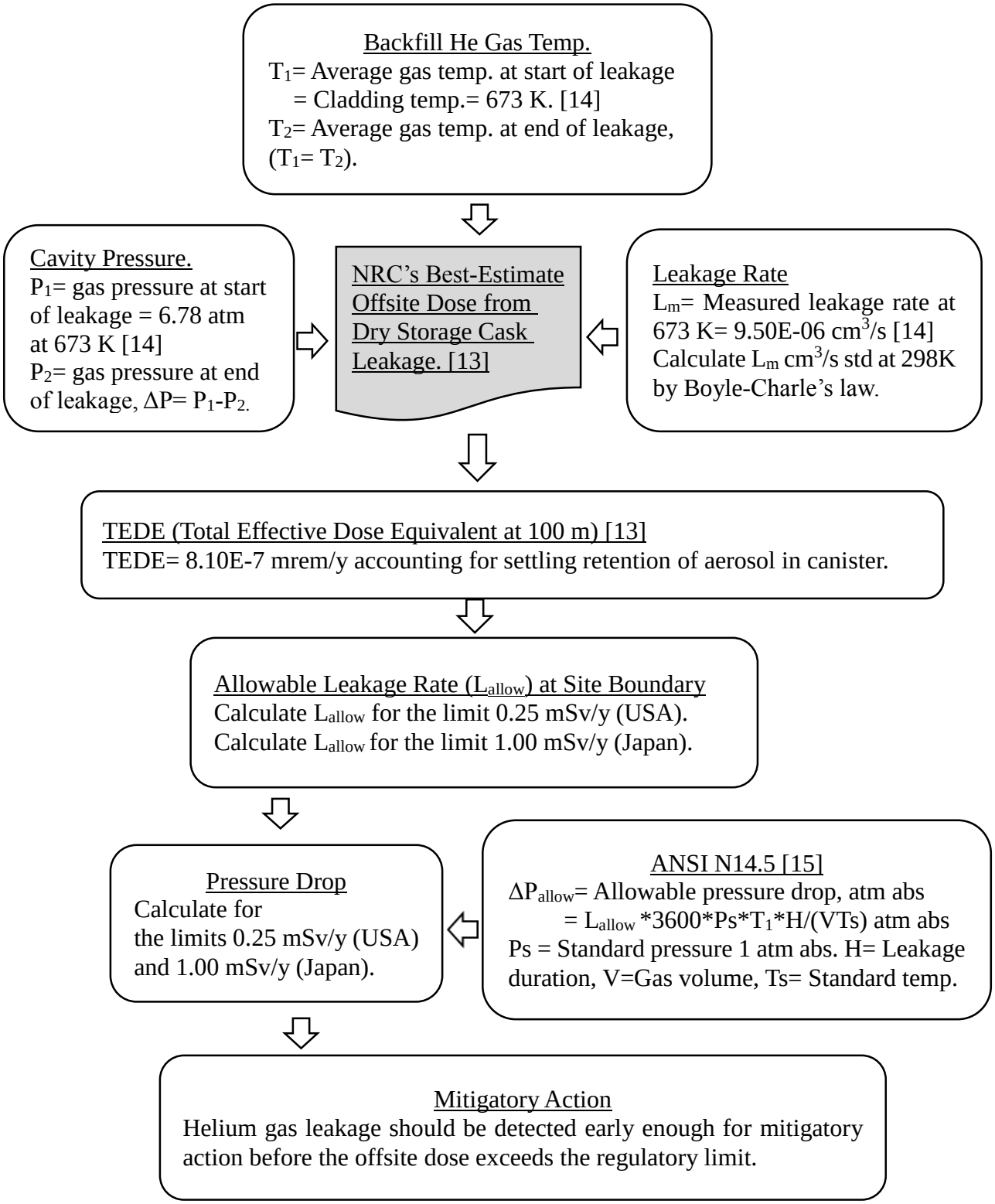

Figure 2. Calculation Steps for Allowable Leakage and Mitigatory Action due to CISCC Limited by Offsite Dose (Scenario 2)

Table 2 shows the calculated results of allowable leakage rate for the regulatory limits of 0.25 mSv/y (USA) and 1.0 mSv/y (Japan) at the site boundary, which is 100 m from the storage casks. The NRC report [13] included cases with and without the deposition of aerosol particles inside the confinement (canister). The NRC concluded that including a realistic treatment of aerosol deposition in a dry storage cask would significantly ease regulatory burden. A total effective dose equivalent (TEDE) value at 100 m, TEDE = 8.10E-7 mrem/y, that accounted for settling retention of aerosol has been used to calculate the allowable leakage rate (L_{allow}) that would satisfy the limits at site boundaries in Japan and the United States. The values of the corresponding allowable pressure drops (ΔP_{allow}) are then calculated by using eq. 3.

For the offsite dose limit of 1.0 mSv/y (Japan), the ratio of the allowable pressure drop in 12 months to the complete depressurization to 1 atm is 0.621/(6.78-1) = 0.11 atm/y, which is about 10% of the potential pressure drop. Such a drop would be linear with time, as shown in Figure 3, assuming the pressure drop behaves as that shown in Figure 1. The canister pressure may drop from 6.78 to 6.46 atm in 6 months when He leakage is detected, as shown in Figure 3, which would allow mitigatory action to be taken during the following 6-month period, before the Japanese regulatory offsite dose limit of 1 mSv/y is violated.

For the offsite dose limit of 0.25 mSv/y (United States), the ratio of the allowable pressure drop in 12 months to the complete depressurization to 1 atm is 0.155/(6.78-1) = 0.027 atm/y, which is about 3% of the potential pressure drop. If a pressure drop due to CISCC to about 6.7 atm (97% of 6.78 atm) in 6 months could be detected, it would be possible to take mitigatory action in the next 6-month period, before the U.S. regulatory limit of 0.25 mSv/y is violated. This calculation also assumed the distance from the storage casks to the site boundary is 100 m. If the actual distance were greater than 100 m, the allowable leakage and the allowable pressure drop would be larger, thus providing even more time to implement mitigatory action before the regulatory limit is violated.

Table 2. Allowable Leakage Limited by Offsite Dose under Normal Operating Condition (HI-STORM Cask System)

Parameter	Value	Remark
Pay load	32 PWR	-
V = Net free cavity volume (cm^3)	6.00E+06	-
T_1 = Backfill gas temperature in storage (K)	673	400℃ cladding temp. long-term normal cond.
P_1 = Normal cavity pressure (atm abs with 1% rods rupture)	6.78	99.7 psig at 400℃
L_m = Leakage rate at 673 K (cm^3/s)	9.50E-06	-
L_m = Measured leakage rate at 298 K (cm^3/s std)	2.85E-05	-
TEDE accounting for settling retention (mSv/y)	8.10E-07	-
L_{allow} = Allowable leakage rate for TEDE (USA) (cm^3/s)	8.80E+00	Offsite dose limit = 0.25 mSv/y (USA)
L_{allow} = Allowable leakage rate for TEDE (Japan) (cm^3/s)	3.52E+01	Offsite dose limit = 1.0 mSv/y (Japan)
ΔP_{allow} = Allowable pressure drop for TEDE (USA) (atm abs)	1.55E-01	For offsite dose limit 0.25 mSv/y (USA)
ΔP_{allow} = Allowable pressure drop for TEDE (Japan) (atm abs)	6.21E-01	For offsite dose limit 1.0 mSv/y (Japan)

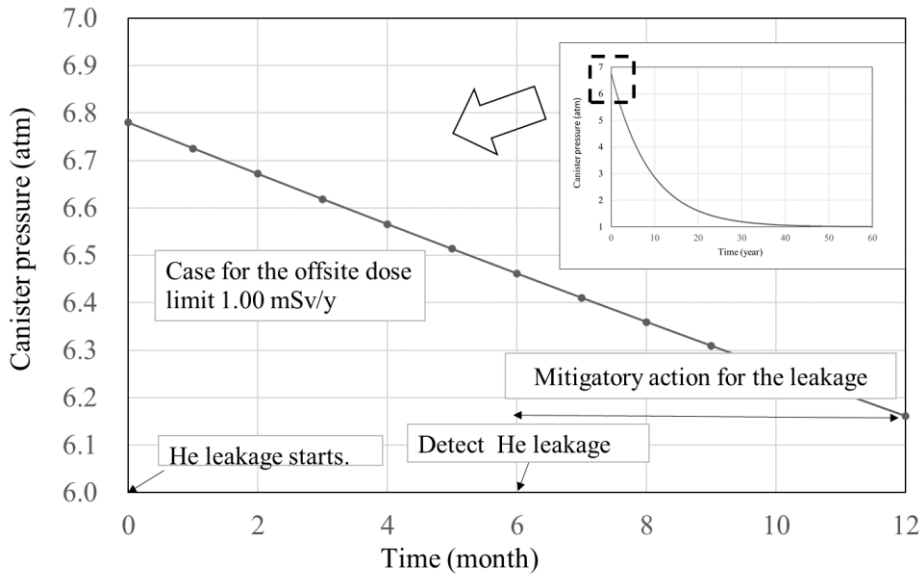

Figure 3. Illustration of Canister Pressure Drop with Time due to CISCC (Scenario 2)

5. DISCUSSION

For Scenario 1 (described in Table 1), the COA has a strong influence on the calculated leakage rate. The COA, 4E-05 cm^2, was obtained by assuming an idealized planar through-wall crack, initiated by residual stress at the welded canister [12]. This COA value is conservatively large since the actual CISCC crack is highly branched and tighter through the wall. Therefore, the pressure drop with time would be much smaller, thus giving more time for mitigatory action after leakage detection.

For Scenario 2 (described in Figure 2 and Table 2), the calculated allowable leakage rates (L_{allow}) are relatively slow, and the allowable pressure drops (ΔP_{allow}) are small. However, the time interval of monitoring could be as long as several months before pressure drop or leakage is detected. This period should not be an issue, as demonstrated in the scale-model gas leakage experiments conducted to date by using RAMM) [16], based on the canister surface temperature-difference monitoring methods developed by Takeda et al. [7]. Additional scale-model gas leakage experiments are being conducted to establish the technical basis for applying the methodology to the detection of helium leakage from canisters in actual cask systems containing SF during long-term storage.

6. SUMMARY AND CONCLUSION

In this paper, we considered the radiological consequences of two scenarios of helium gas leakage from a canister storing SF due to CISCC. The first scenario is monitoring of helium gas leakage against the limit by SF oxidation. If such a pressure drop can be detected within 10 days after the start of a leakage, the necessary actions to avoid air ingress into the canister and SF oxidation could be taken within the next 40 days. The second scenario is monitoring of helium gas leakage against the regulatory limits of site boundary doses. If the allowable pressure drop can be detected within six months after the start of a leakage, the necessary actions can be taken within the next six-month period before Japanese and U.S. regulatory off-site dose limits are exceeded.

We also proposed ageing management strategies that include time to enable leakage detection and implementation of mitigation measures by applying canister temperature-difference monitoring methods using RAMM. Early detection of gas leakage has been demonstrated in the scale-model gas leakage experiments conducted at CRIEPI. Testing will continue, leading to a demonstration of the canister surface temperature-difference monitoring methodology by using RAMM for detecting canister gas leakage in actual dry cask systems.

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1. INTRODUCTION

Spent nuclear fuel may be stored in dry concrete casks with welded stainless-steel canisters for extended periods. Chloride-induced stress corrosion cracking (CISCC) of canisters has been identified as a potential degradation mechanism, requiring ageing management — especially in a marine air environment. [1–3].

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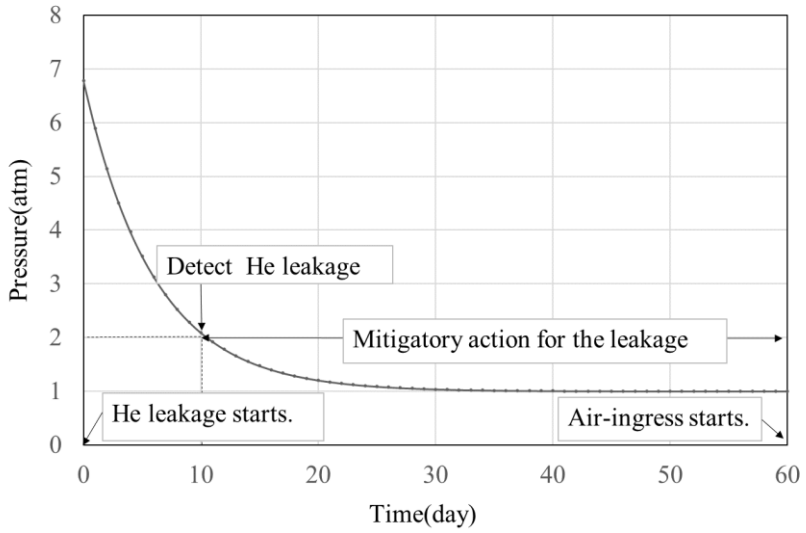

Figure 1. Example of Canister Pressure Drop with Time due to CISCC (Scenario 1)

4.2. Scenario 2: Monitoring of leakage limited by offsite dose

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The helium leakage rate from canister is given by equation 2 [15].

Equation 2: $L_m = VT_s/3{,}600HP_s \times (P_1/T_1 - P_2/T_2)$ std•cm^3/s
where L_m = measured leakage rate, std•cm^3/s
V = gas volume in test item, cm^3
T_s = standard temperature, 298 K
H = test duration, hours
P_s = standard pressure, 1 atm abs
P_1 = gas pressure in test item at start of test, atm abs
P_2 = gas pressure in test item at end of test, atm abs
T_1 = average gas temperature at start of test, K
T_2 = average gas temperature at end of test, K
It is assumed that $T_1 = T_2$.

Equation 2 can be rewritten as equation 3.

Equation 3: $\Delta P = P_1 - P_2 = 365 \times 24 \times L_m \times T_1 \times 3{,}600P_s / VT_s$ atm abs

The allowable pressure drop ΔP_{allow} is obtained by replacing the L_m with L_{allow} in eq. 3.

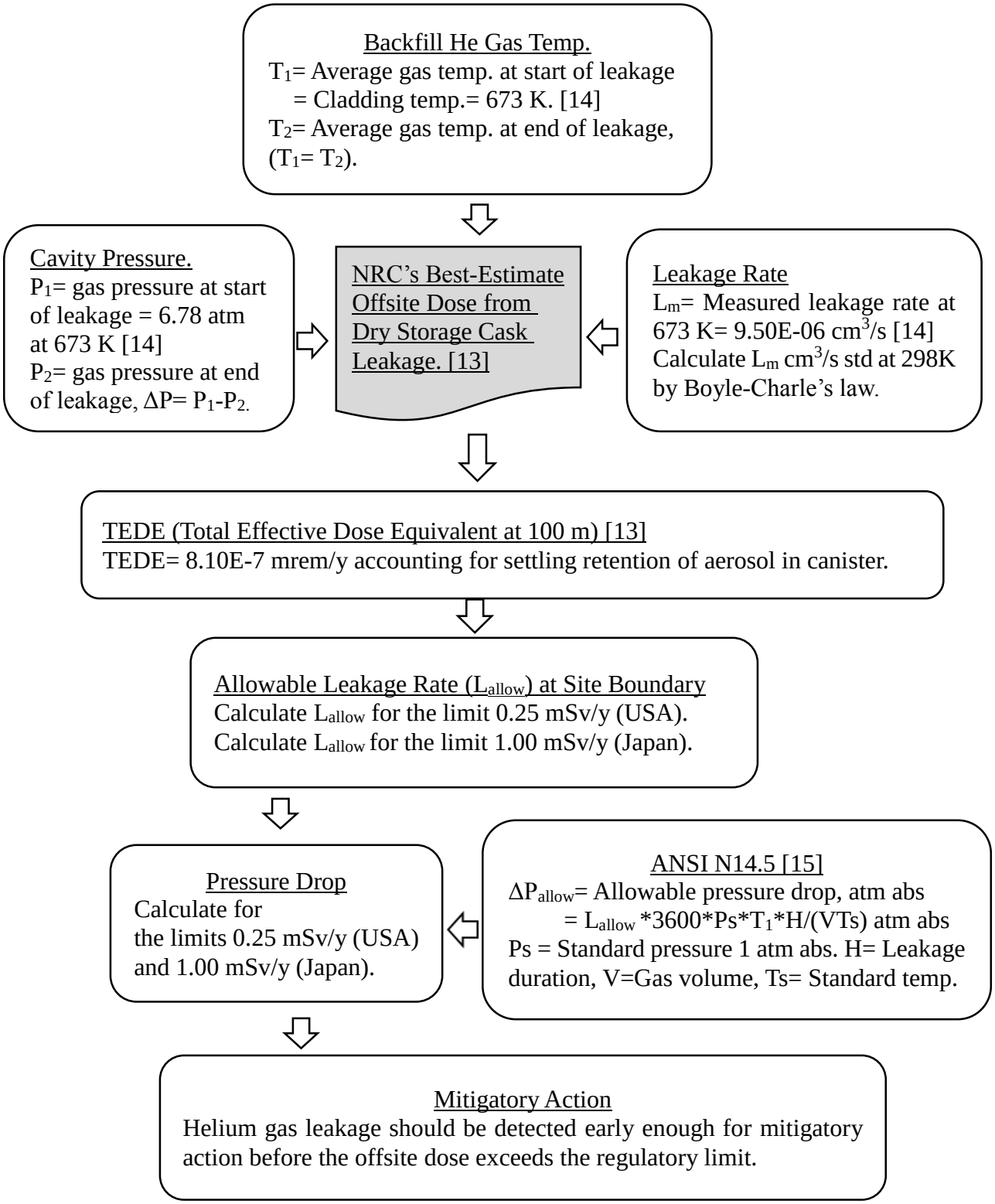

Figure 2. Calculation Steps for Allowable Leakage and Mitigatory Action due to CISCC Limited by Offsite Dose (Scenario 2)

Table 2 shows the calculated results of allowable leakage rate for the regulatory limits of 0.25 mSv/y (USA) and 1.0 mSv/y (Japan) at the site boundary, which is 100 m from the storage casks. The NRC report [13] included cases with and without the deposition of aerosol particles inside the confinement (canister). The NRC concluded that including a realistic treatment of aerosol deposition in a dry storage cask would significantly ease regulatory burden. A total effective dose equivalent (TEDE) value at 100 m, TEDE = 8.10E-7 mrem/y, that accounted for settling retention of aerosol has been used to calculate the allowable leakage rate (L_{allow}) that would satisfy the limits at site boundaries in Japan and the United States. The values of the corresponding allowable pressure drops (ΔP_{allow}) are then calculated by using eq. 3.

For the offsite dose limit of 1.0 mSv/y (Japan), the ratio of the allowable pressure drop in 12 months to the complete depressurization to 1 atm is 0.621/(6.78-1) = 0.11 atm/y, which is about 10% of the potential pressure drop. Such a drop would be linear with time, as shown in Figure 3, assuming the pressure drop behaves as that shown in Figure 1. The canister pressure may drop from 6.78 to 6.46 atm in 6 months when He leakage is detected, as shown in Figure 3, which would allow mitigatory action to be taken during the following 6-month period, before the Japanese regulatory offsite dose limit of 1 mSv/y is violated.

For the offsite dose limit of 0.25 mSv/y (United States), the ratio of the allowable pressure drop in 12 months to the complete depressurization to 1 atm is 0.155/(6.78-1) = 0.027 atm/y, which is about 3% of the potential pressure drop. If a pressure drop due to CISCC to about 6.7 atm (97% of 6.78 atm) in 6 months could be detected, it would be possible to take mitigatory action in the next 6-month period, before the U.S. regulatory limit of 0.25 mSv/y is violated. This calculation also assumed the distance from the storage casks to the site boundary is 100 m. If the actual distance were greater than 100 m, the allowable leakage and the allowable pressure drop would be larger, thus providing even more time to implement mitigatory action before the regulatory limit is violated.

Table 2. Allowable Leakage Limited by Offsite Dose under Normal Operating Condition (HI-STORM Cask System)

Parameter	Value	Remark
Pay load	32 PWR	-
V = Net free cavity volume (cm^3)	6.00E+06	-
T_1 = Backfill gas temperature in storage (K)	673	400℃ cladding temp. long-term normal cond.
P_1 = Normal cavity pressure (atm abs with 1% rods rupture)	6.78	99.7 psig at 400℃
L_m = Leakage rate at 673 K (cm^3/s)	9.50E-06	-
L_m = Measured leakage rate at 298 K (cm^3/s std)	2.85E-05	-
TEDE accounting for settling retention (mSv/y)	8.10E-07	-
L_{allow} = Allowable leakage rate for TEDE (USA) (cm^3/s)	8.80E+00	Offsite dose limit = 0.25 mSv/y (USA)
L_{allow} = Allowable leakage rate for TEDE (Japan) (cm^3/s)	3.52E+01	Offsite dose limit = 1.0 mSv/y (Japan)
ΔP_{allow} = Allowable pressure drop for TEDE (USA) (atm abs)	1.55E-01	For offsite dose limit 0.25 mSv/y (USA)
ΔP_{allow} = Allowable pressure drop for TEDE (Japan) (atm abs)	6.21E-01	For offsite dose limit 1.0 mSv/y (Japan)

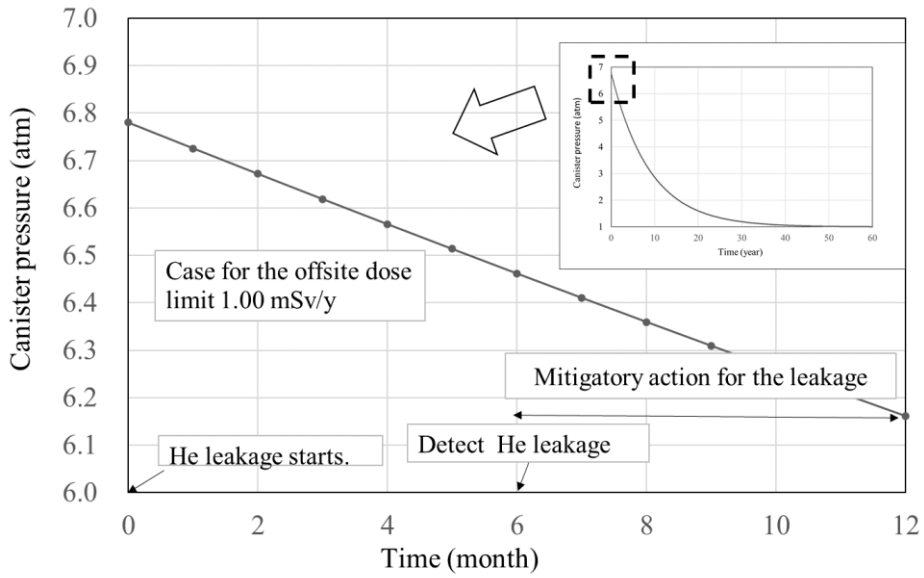

Figure 3. Illustration of Canister Pressure Drop with Time due to CISCC (Scenario 2)

5. DISCUSSION

For Scenario 1 (described in Table 1), the COA has a strong influence on the calculated leakage rate. The COA, 4E-05 cm^2, was obtained by assuming an idealized planar through-wall crack, initiated by residual stress at the welded canister [12]. This COA value is conservatively large since the actual CISCC crack is highly branched and tighter through the wall. Therefore, the pressure drop with time would be much smaller, thus giving more time for mitigatory action after leakage detection.

For Scenario 2 (described in Figure 2 and Table 2), the calculated allowable leakage rates (L_{allow}) are relatively slow, and the allowable pressure drops (ΔP_{allow}) are small. However, the time interval of monitoring could be as long as several months before pressure drop or leakage is detected. This period should not be an issue, as demonstrated in the scale-model gas leakage experiments conducted to date by using RAMM) [16], based on the canister surface temperature-difference monitoring methods developed by Takeda et al. [7]. Additional scale-model gas leakage experiments are being conducted to establish the technical basis for applying the methodology to the detection of helium leakage from canisters in actual cask systems containing SF during long-term storage.

6. SUMMARY AND CONCLUSION

In this paper, we considered the radiological consequences of two scenarios of helium gas leakage from a canister storing SF due to CISCC. The first scenario is monitoring of helium gas leakage against the limit by SF oxidation. If such a pressure drop can be detected within 10 days after the start of a leakage, the necessary actions to avoid air ingress into the canister and SF oxidation could be taken within the next 40 days. The second scenario is monitoring of helium gas leakage against the regulatory limits of site boundary doses. If the allowable pressure drop can be detected within six months after the start of a leakage, the necessary actions can be taken within the next six-month period before Japanese and U.S. regulatory off-site dose limits are exceeded.

We also proposed ageing management strategies that include time to enable leakage detection and implementation of mitigation measures by applying canister temperature-difference monitoring methods using RAMM. Early detection of gas leakage has been demonstrated in the scale-model gas leakage experiments conducted at CRIEPI. Testing will continue, leading to a demonstration of the canister surface temperature-difference monitoring methodology by using RAMM for detecting canister gas leakage in actual dry cask systems.

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Declaration of interests

☐ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☒The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

The authors are research partners and have known each other's work and professional activities since 2014. The relationships are complementary, and does not involve any potential competing interests.

Author contributions - Credit roles of co-authors-

Toshiari Saegusa: Conceptualization, Methodology, Formal analysis, Investigation, Writing - Original Draft, Supervision

Hirofumi Takeda: Conceptualization, Methodology, Data curation, Writing- Original draft preparation, Writing- Reviewing and Editing

Yung Liu: Conceptualization, Methodology, Reviewing and Editing

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