Integrity Assessment of Dual-Purpose Metal CASK after Long Term Interim Storage – Seal Performance under Transport Conditions

Takeshi YOKOYAMA, Hiroto KAWAKAMI, Noriyuki UCHIYAMA[*1], Makoto YASUDA[*2] and Shinji MATSUOKA[*3]

Incorporated Administrative Agency Japan Nuclear Energy Safety Organization, Tokyo, Japan
*1 Present affiliation is Tokyo Electric Power Company, Tokyo, Japan
*2 Present affiliation is Toshiba Corporation, Yokohama-city, Japan
*3 Present affiliation is Kobe Steel Ltd, Takasago-city, Japan

Abstract

Spent fuels generated in nuclear power plants (NPPs) shall be stored until reprocessing as recyclable energy resources in Japan. The quantity of spent fuels stored at each NPP site is increasing, and early realization of the interim storage is expected. Dual-purpose metal cask will be used there and will not be reopened until it is delivered to a reprocessing plant in order to for example minimize personal exposure of radiation.

Japan Nuclear Energy Safety Organization (JNES) was established on October in 2003 with the mission to ensure the public safety from the potential hazard of nuclear energy. Study of “Metal Cask Storage Technology Verification” was originally initiated in 1999 at Nuclear Power Engineering Corporation with Japanese government funds, and transferred to JNES and conducted up to the end of FY2003. In the study, many tests were conducted to investigate material property change for main components relating to safety of cask due to degradation during interim storage, furthermore, to verify containment safety during the subsequent transport because there was a possibility of providing the cask with degraded metal gasket for the transport after interim storage and the such cask should be considered fragileness of lid containment system, especially for transport that cask would be provided external force.

This paper presents the results and consideration on seal performance during the subsequent transport.

1. Introduction

Interim storage in Japan will be applied by dry storage technology, and especially, it is expected that the dry cask will be realized earlier because of many experiments of cask for storage or transport, in both Japan and oversea. The dry storage system is proposed to use dual-purpose metal casks for storage and transport. In the system, the cask will fill with inert gas and be placed in the storage facility with removing decay heat from spent fuels by natural convection of air during normal storage. And then, the cask will maintain subcritical condition of spent fuels and temperature and stress of every component will not be severer.

However, the cask will be transported after the storage without reopening lids to replace spent fuels or metal gaskets of containment as mentioned above, and then, the influence will be important for transport safety, because transport conditions are severer than the passive storage condition.

The study of “Metal Cask Storage Technology Verification” was conducted on the influence of degradation of cask components on safety performance considering the above situation.

To find out issues from the possibility of influencing on safety point of view, simulated metal casks were established based on research on proposed design to cask, specifications of spent fuels and environment in storage facility etc., shown in Table 1, and temperature and cumulative irradiation dose of each component were estimated by analyses of the simulated cask for 60 years interim storage, shown in Table 2. And then, existing information relating to the dry storage technology and expected materials applied to the cask were reviewed. As a results of the review, 1) corrosion and SCC of cask body, lid and basket in corrosive environment including iodine released from postulated-failure spent fuels, 2) material properties change for basket by thermal aging, 3) degradation of shielding performance due to temperature and irradiation, 4) relaxation of metal gasket for lid containment system, 5) seal performance of lid containment system with relaxed metal gasket during transport were found out as issues that should be investigated, and tests were selected in Table 3, which were composed of “Material Property” tests and “Safety Performance” tests relating to the above issues.

Table 1 Basic Specification for Simulated Metal Cask

Material of Main Shielding Structure		Steel + PG Water, Steel + Resin, Lead + Resin
Total Weight (Max.)		120 ton
Spent fuel type & Min. Cooling Time		PWR 17*17(48GWd/t) , BWR(STEP3)(50GWd/t) 5 years
Type : Steel + PG Water		
	Max. Number of Assemblies	PWR 17 Assemblies per CASK, BWR 39 Assemblies per CASK
	Max. heat	PWR 22.0 kW per CASK, BWR 16.5 kW per CASK
Type : Steel or Lead + Resin		
	Max. Number of Assemblies	PWR 21 Assemblies per CASK, BWR 52 Assemblies per CASK
	Max. heat	PWR 27.1 kW per CASK, BWR 21.9 kW per CASK
Materials of Main Components		
	Body & lids	Carbon Steel, Stainless Steel
	Neutron Shielding Material	Resin Materials (Epoxy, Silicon), PG Water (Propylene Glycol)
	Basket	Borated Aluminum Alloy, Aluminum Alloy
	Seal for Lid Containment System	Metal Gasket

Table 2 Estimated Temperatures and Cumulative Irradiations during Storage (Max.)

Main Parts	Temperatures (degree C)		Cumulative Irradiation Dose (for 60 years)	
	Initial Storage	After 60 years	Neutron (n/cm^2)	Gamma (Gy)
Body & lids *1	182	84	$2.12*10^{15}$	$1.04*10^{8}$
Neutron Shielding Material (Resin)	163	79	$4.13*10^{14}$	$6.34*10^{2}$
(PG Water)	147	78	$1.52*10^{14}$	$1.23*10^{3}$
Basket	265	100	$4.87*10^{15}$	$3.73*10^{8}$
Metal Gasket	119	69	(Less than *1)	(Less than *1)

Table 3 Tests in "Metal Cask Storage Technology Verification" Study

Tests		Objectives
Material Property	Corrosion & SCC characteristics	To investigate corrosion & SCC characteristics for cask body, lid and basket during storage under assumed failure of spent fuels
	Borated Aluminum Alloy	To investigate property change due to thermal ageing during storage
	Neutron Shielding Materials	To investigate the influence on degradation of shielding performance due to thermal ageing and/or irradiation during storage
	Metal Gasket for Lid Containment	To investigate relaxation during storage and the influence on seal performance during the subsequent transport
Safety Performance	Lid Containment System under Fire Condition	To verify seal performance considering relaxed metal gasket under fire condition during the subsequent transport
	Lid Containment System under Drop Condition	To verify seal performance considering relaxed metal gasket under drop condition during the subsequent transport

2 Material Properties

2.1 Corrosion & SCC Characteristics

It had not been reported yet that original intact spent fuels were damaged and released FP gas to the cask cavity during the present dry storage of actual spent fuels that the cavity maintained inert gas atmosphere and components maintained moderate temperature, however, the storage period was not considered enough long to refuse the possibility of failure of spent fuels during the dry storage of 60 years. Furthermore, there was no quantitative data for failure frequency of spent fuels during the long term dry storage. Therefore, it was assumed that spent fuels would be damaged and release FP gases in the cavity, and the influence of corrosive gas, iodine, was experimentally investigated for cask body, lid and basket.

Materials tested were borated aluminum alloy, aluminum alloy and borated SUS304 for basket, and ASTM A350 LF5 and SUSF304 for cask body and lids. The tests were conducted in atmosphere including iodine gas of which density was equivalent to 1% and more failure of stored spent fuels.

The test specimen was installed in a furnace which the temperature and the pressure were kept at 270 degree C, that was expected as higher than actual, and 0.2 MPa during the test, respectively. The maximum period of exposure was 6 months.
As results of the tests, general corrosion occurred in each material, without pitting or local corrosion, and decrease of thickness of each material was estimated to be very small for 60 years storage based on weight loss coefficient obtained from test results. Furthermore, no SCC indication was inspected in each material.
Therefore, if failure fraction of spent fuels is assumed to be less than 1%, it was expected that general corrosion should be only considered although the decrease of thickness would be very small for the storage.

2.2 Material Property of Borated Aluminum Alloy for Basket

The basket would be exposed to high temperature and radiation during the storage. For the metal cask, basket composed from borated aluminum alloy would have functions of absorbing neutron, removing the decay heat and maintaining spent fuels arrangement for subcriticality. From review of existing data, however almost of published information was for aluminum alloy not including boron, it was evaluated that cumulative irradiation dose shown in Table 3 would be less than a value causing significant mechanical property change of aluminum alloy. However, aluminum alloy was likely to be influenced by such temperature as 265 degree C shown in Table 3, and annealing and creeping were found out as factors that might influence on properties of borated aluminum alloy during the storage.
Therefore, for annealing, the influence on material properties, including tensile and proof strengths, absorbing energy of impact tests, micro structure, modulus, thermal conductivity specific heat and thermal expansion was investigated, and for creeping, the influence on tensile and proof strengths was investigated when the material showed a little creep deformation. JIS H4080 A5052 H34 (No boron), 5wt%B4C Borated Aluminum Alloy (Base: JIS H4100 A6N01), 1wt% over Borated Aluminum Alloy (Base: ASTM A6351-T5), and 1wt% Borated Aluminum Alloy (Base: ASTM A3004-H112) were tested.
For annealing, material properties were measured at various temperatures up to 250 C using the specimen provided annealing at constant temperature of 200 or 250 degree C for 1000, 3000 or 10000 hours.
For creeping, tensile and proof strengths were measured at 250 degree C using the specimen provided annealing at constant temperature of 250 degree C for 1000 hours and creep deformation from 0.1% to 1.0% in addition.
And then, as features of borated aluminum alloy influenced due to high temperature, it was confirmed that tensile and proof strengths were made lower by annealing, however, there was no additional influence if creep deformation was provided to annealed metal additionally, as shown in Fig. 1. On the other hand, there was no significant change for the other properties. Therefore, it was considered that lowering of strengths would be important for the long term storage.

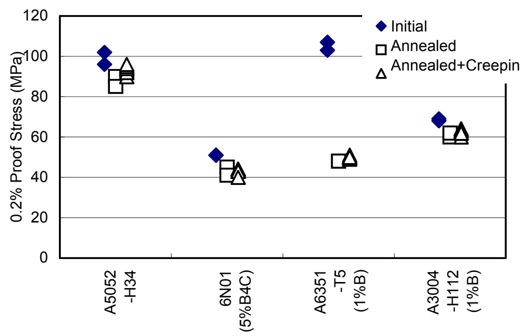

Fig. 1 Proof Strength of Aluminum Alloy

2.3 Degradation for Neutron Shielding Materials

Epoxy resin, silicon resin, or PG water would be applied to metal cask as neutron shielding material. Degradation of the shielding due to thermal aging and irradiation were experimentally investigated and a prediction method of shielding performance was discussed.
For epoxy resin and silicon resin, irradiation tests of neutron or gamma ray, heating tests in/ after/ without irradiation were conducted in closed system with forced ventilation, with constant pressure controlled, or without ventilation. The maximum temperature was estimated 163 degree C at the beginning of the storage and decreased year by year depending on decay heat attenuating. However, to confirm the feature of degradation, parameters of the tests were temperature and period of heating and cumulative irradiation dose. Temperature of heating was from 130 to 170 degree C, and the maximum period was 15,000 hours. Cumulative irradiation dose of neutron and gamma ray were $1.5*10^{15}$ n/cm^2 and $3.9*10^4$ Gy at the maximum, respectively.
After heating or irradiation, weight loss, appearance, chemical components of remainder and gas were inspected. The following features were found out.

- Weight loss was estimated to occur by release of oxide products of low molecular weight from base materials and of H_2O due to dehydrate reaction of tri-hydrate-alumina.
- The influence of heating on weight loss was dominant, and that of neutron and gamma ray irradiations was small. In addition, there was no synergistic effect of heating and irradiation.
- Relation between weight loss and environment of the tests was,
  Closed system with forced ventilation > with content pressure controlled > without ventilation

Weight loss was selected as an index of the degradation of shielding performance, and it was plotted for LMP (Larson·Muller·Parameter) shown in Fig. 2, it was example of epoxy resin, and prediction of weight loss was discussed instead of degradation of shielding performance.

LMP was defined by the following expression.

LMP=T (C + log t)

T: absolute temperature of heating (K)

C: constant, t: heating time (hour)

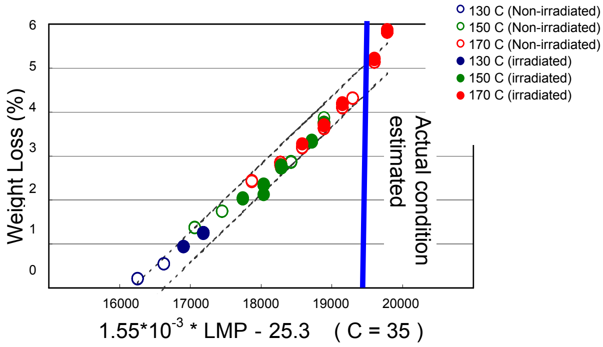

Fig. 2 Degradation of Epoxy Resin
(for closed system with forced ventilation)

As shown in Fig. 2, it was confirmed that a linear relation between weight loss and LMP existed, and it was considered that weight loss during the storage could be predicted using the relation and LMP, however, the relation to use prediction should be selected considering the actual environment that neutron shielding material placed in, because it was found out from the test results that the weight loss depending on the environment, as mentioned above feature.

For degradation of PG water, it was considered that corrosion of casing due to decomposed products and rising of freezing point due to property change were important from for availability of the long term storage.
And, irradiation tests of neutron or gamma ray, heating tests after/ without irradiation were conducted, and chemical components of remaining water and released gas, and freezing point were inspected.
To confirm the feature of degradation, parameters in the tests were temperature and period of heating, and cumulative irradiation dose of neutron and gamma ray. Temperatures of heating specimen were from 100 C to 180 C, and the maximum period was 10,000 hours. Cumulative irradiation dose of neutron and gamma ray were $5.0*10^{14}$ n/cm^2 and $2.0*10^4$ Gy at the maximum, respectively.
The following features were found out, and availability of PG water for the storage was confirmed for the condition estimated here in Table 2.

- PG density was decreased by a factor of 3% due to degradation in the test of 160 degree C with 10,000h.
- Acetone, aldehyde that is a precursor of organic acid, oxygen, and hydrogen were detected as major decomposed products. However, organic acid was little detected.
- A little general corrosion and no SCC occurred on inner surface of casings of SUS.
- Freezing point was slightly changed, for example, -46.4 degree C increased to -42.9 degree C in the test of 160 C with 10,000h.

2.4 Degradation for Metal Gasket

Metal gasket applied to the lid containment system would be exposed to high temperature and radiation during the storage. From review of existing information, it was estimated that cumulative irradiation dose during the storage in Table 2 would be less than a value causing significant mechanical property change of component materials, that were aluminum alloy or silver of outer shell and high Ni alloy of inner shell and coil spring. However, the previous study showed that the relaxation due to high temperature occurred but the seal performance of relaxed metal gasket was kept under normal storage condition.
Generally, the metal gasket was assembled from a coil spring, inner and outer shells, and the leak tight would be achieved by the outer shell adhered to flange surface. Therefore, the relaxation of metal gasket was considered to influence seal performance during the subsequent transport significantly, because of external force under the condition. The following tests were conducted in order to simulate the relaxation and to verify the effects.

a. Heating tests to measure the relaxation of metal gasket
b. Seal performance tests against quasi-static or dynamic displacement for the metal gasket simulated relaxation

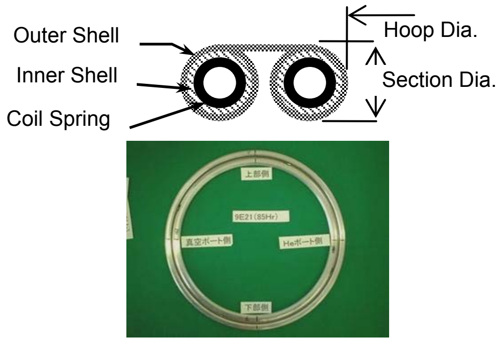

Fig. 3 Metal Gasket (Double Type)

The following metal gaskets were used in the tests, however, the metal gasket, which was double type structure, outer shell of aluminum alloy and section diameter of 10 mm, was intensively investigated. Each size of the tested metal gaskets was about 1/10 of actual one for hoop diameter and the same as actual one for section diameter, and small size flanges fitting to compress the gasket were used.

Type of structure: Single type or Double type

Materials: (Coil Spring) High Ni Alloy
(Inner Shell) High Ni Alloy
(Outer Shell) Al or Ag *Ag: for heating tests only

The followings were results for the above metal gasket intensively investigated.

- Compressed metal gaskets with flanges were heated in thermostatic chamber and reduction of spring back forces (SBFs) was measured. Relation between reduction ratio and LMP was evaluated. Reduction ratio was defined by the following expression.

  RF = (Fi – F) / Fi

  RF: Reduction Ratio of SBF
  Fi: Initial SBF
  F: Reduced SBF after Heating

- Heating was conducted up to LMP=7800 and leak rates were verified to change little for steady state condition. Incidentally, the LMP equivalent to 60 years storage here was 7232.

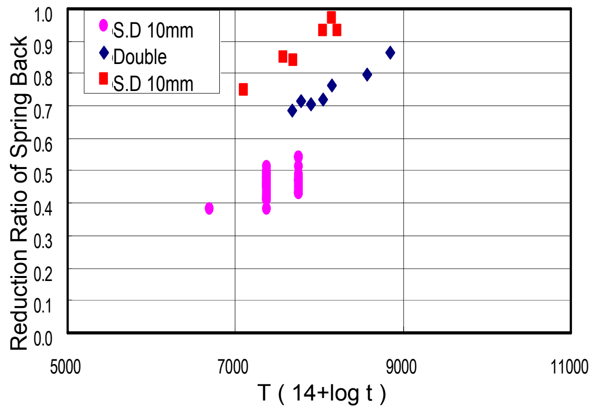

Fig. 4 Reduction Ratio of Spring Back Force vs. LMP

- On providing displacement to flanges with relaxed metal gasket at RT, leak rates were measured. The followings were results for the degradation of LMP=7375 that was severer than actual condition estimated and shown in Table 2. For the above main gasket, SBFUL (SBF per Unit hoop Length) was estimated to be more than 1.6kN/cm using the relation of reduction ratio and LMP for LMP=7375.

  The following leak rates were measured in tests and they should be 10 times larger considering the difference of hoop diameter when they were compared to actual.

  - If a gap was made temporally between the metal gasket and seal surface of flange quasi-statically and SBFUL was more than 1kn/cm, on recompressing metal gasket, leak rate was lower than 10^{-5} Pa·m^3/s.
  - On providing quasi-static radial displacement of 6 mm, which was a sum of go and return motion of 3 mm, leak rate was lower than 10^{-5} Pa·m^3/s.
  - If dynamic axial displacement was provided, on recompressing metal gasket, leak rate was lower than 10^{-5} Pa·m^3/s. The maximum displacement was about 0.35 mm and it was estimated that a gap between the metal gasket and the seal surface of flange was made at a moment when such displacement occurred.
  - Leak rate was measured in a few minutes after impact. The maximum leak rate was about 10^{-5} Pa·m^3/s and the maximum residue radial displacement was about 5 mm. It was observed that leak rate increased depending on residual displacement.

3. Safety Performance Tests during subsequent transport

3.1 Degraded Lid Containment under Fire Condition

To verify seal performance for the lid containment system with the relaxed metal gasket under fire condition of the subsequent transport, thermal tests were conducted using partial cask lid model that had an actual size of diameter but 1/5 body length from the top.

Cask lid model was heated up to over 7375 of the LMP of metal gasket with the heaters of inside and outside before the tests.

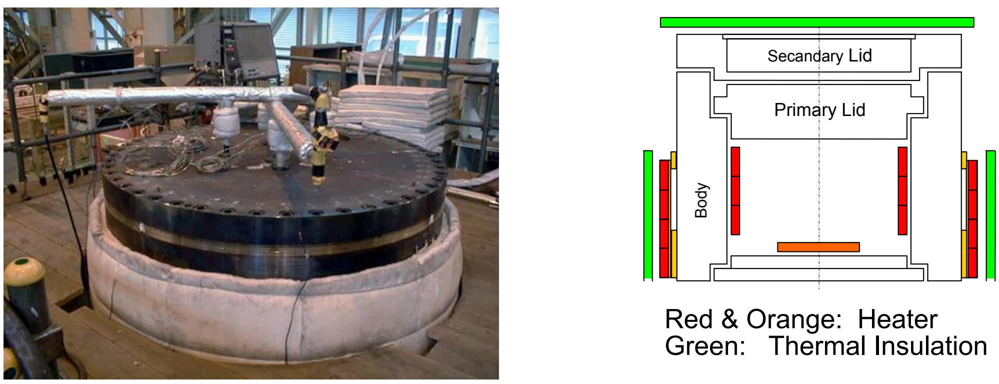

Fig.5 CASK Lid Model for Thermal Tests

The following tests were conducted.

a. Thermal test equivalent to fire condition
b. Symmetrical thermal parametric test
   Parameter: Heater Power, Secondary Lid Bolt Torque (Minimum torque was about a quarter of design value)
c. Non-symmetrical thermal parametric test
   Parameter: Heater Power, Secondary Lid Bolt Torque (Minimum torque was about a quarter of design value)

To refer to the tests, pre-analysis for fire condition was conducted. For the thermal test equivalent to fire condition, control of heating power was established according to the analysis results. For each parametric test, heating power and lid bolt torque were established to make the lid displacement more than analytical results for fire condition.

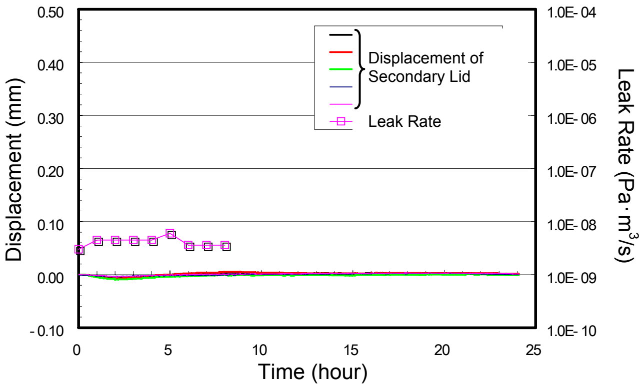

Fig.6 Displacement & Leak Rate (Thermal Test Equivalent to Fire Condition)

Degradation conditions to relax the gasket tested achieved to about 7400 of LMP.

Major results of the tests were as follows;

- ☐ Secondary lid hardly moved and leak rate changed little even though the metal gasket was relaxed.
- ☐ In the parameter tests, displacement of the secondary lid was more than that in fire condition test "a" (max. axial displacement: 0.07 mm, max. radial displacement: 0.45 mm). Leak rates indicated increase up to about 10^{-5} Pa·m^3/s temporary but they were decreased presently soon, and were stabilized at lower than 10^{-6} Pa·m^3/s.

3.2 Degraded Lid Containment under Drop Condition

To confirm seal performance for the lid containment system with the relaxed metal gasket under drop condition of the subsequent transport, furthermore to investigate the relation between leak rate and lid displacement, drop tests were conducted using full size cask model.

Drop tests were conducted for the following positions. For the horizontal position (1), metal gasket was relaxed by heating from inside and out side of the model. The degradation condition was achieved to about 7400 of LMP.

Drop Position Horizontal Drop (1) & (2) / Vertical Drop with Lid Down / Corner Drop with Lid Down

Fig. 7 Horizontal Drop with full size cask model

Accelerations of main components, strains around body flange and of lid bolts, and relative displacements of the lid to body the flange were recorded in the tests. Leak rate of primary and secondary lids, and pressure between lids were inspected before and after each test.

Major results of the tests were as follows;

- ☐ Leak rate of the secondary lid with relaxed metal gasket was estimated lower than10^{-4} Pa·m^3/s on the drop of any position.
- ☐ Leak rate on each drop was estimated considering relaxation of gasket and required time to prepare leak rate inspection, based on the inspected value, and the estimated value was confirmed to agree with the results of "Degradation tests for metal gasket". Here, relative dis-

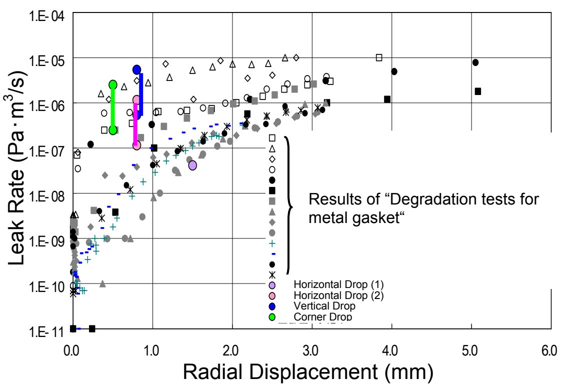

Fig. 8 Comparison of leak rate between full size drop test vs. Elementary test

placement between the lid and the body flange of each drop test was the sum of go and back displacements in drop, because time history of relative radial displacement was go and return motion especially in the horizontal drop that the maximum displacement occurred.

- It was verified that the sum of relative displacement was important to estimate leak rate at drop accident.
- Pressure between both lids decreased but it was more than atmospheric pressure after the drop in each test.

Post-analyses were conducted using DYNA-3D of general FEM code for dynamic response analysis for each drop, and the analysis results could simulate the time histories of relative displacement between the lid and the body flange considering the initial relative position between lid and body flange.

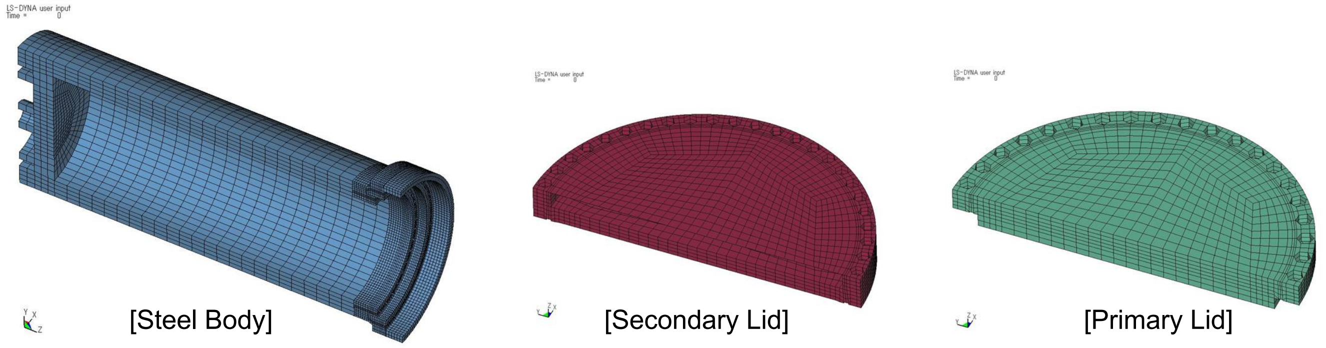

Fig.9 DYNA-3D Model (Main Components)

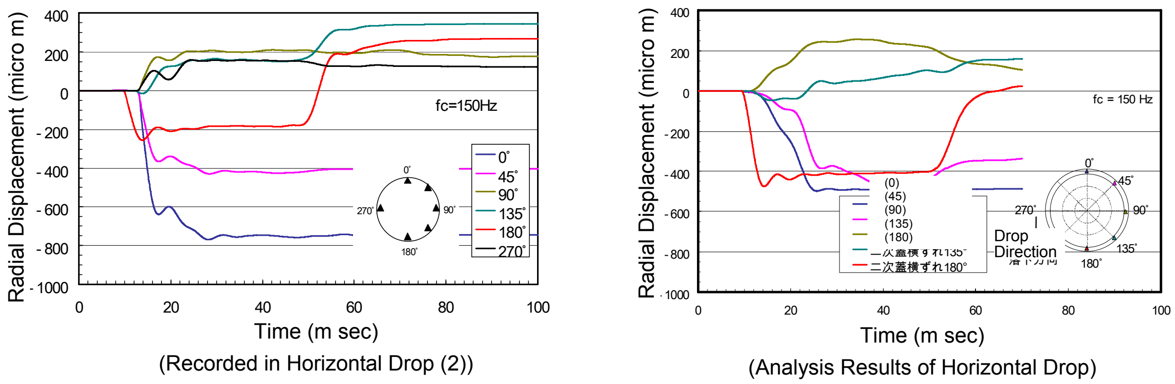

Fig.10 Test & Analysis Results for Displacement of Secondary Lid

4. Consideration for Seal Performance of Lid Containment during Subsequent Transport

For the interim storage for spent fuels in Japan, the lid will not be reopened until it is delivered to a reprocessing plant. Therefore, there is a possibility that lid containment system with relaxed metal gasket is required to secure the transport safety.

To discuss seal performance during transport, in this study, elementary tests for the relaxed metal gasket and thermal and drop tests using the relaxed metal gasket were conducted. Here, the seal performance of the lid containment system with such relaxed metal gasket under transport conditions was considered concerning to the following metal gasket.

Section Diameter: 10 mm, Structure: Double Type,
Materials: coil spring and inner shell of High Ni Alloy, outer shell of aluminum alloy

LMP of metal gasket equivalent to 60 years interim storage was 7232 based on the estimated temperature in Table 2 considering decay heat.

Criteria for seal performance should be decided according to A2 value rightfully. However, it was not convenient way here and standard leak rate 10^{-4} Pa·m^3/s was established as a temporary criteria, that was 1/10 of a design leak rate for domestic transport cask.

However, for the elementary test results, the standard leak rate should be 10^{-5} Pa·m^3/sec that was 1/10 of the above because of difference of hoop diameter.
If LMP was calculated from temperature of metal gasket and storage period, reduced SBFUL could be estimated using the relation between reduction ratio of SBF and LMP of the elementary test results. As the mentioned above, reduced SBFUL was estimated to be over 1.6kN/cm for LMP=7375 even though it was more sever condition than the interim storage condition as the above.
The condition on leak rate to be lower than standard value was SBFUL > 1 kN/cm when a gap was made temporally between the metal gasket and the seal surface of flange in the worst and metal gasket was recompressed afterward. Therefore, the leak rate for the above metal gasket would be lower than the standard value.

Displacement at lid would be considered under drop and fire condition during transport.
For drop condition, relative total radial displacement between lid and body flange as sum of go and return motion was an important factor and it could be evaluated by DYNA-3D analysis or drop test. Leak rate could be evaluated by comparing between the evaluated displacement and relation to dynamic radial displacement of the results of elementary tests. If the evaluated radial displacement was lower than 5 mm, leak rate would be about the standard value and the containment safety could be evaluated for A2 value.
For fire condition, relative displacement was very little and leak rate was verified to change hardly in the test. However, it was important that the function of lid system to recompress the metal gasket was maintained in both drop and thermal test. Therefore, to evaluate leak rate, it was as important as dynamic radial displacement that the components of lid system was not plastic deformation to recompress metal gasket after the phenomena and torque for lid bolts was established moderately to occur little displacement under fire condition.
Therefore, for seal performance for the above relaxed metal gasket, leak rate would be about the standard value at least on condition that CASK design satisfied the following, and then, the lid system would satisfy the requirement in transport regulation.

a. SBFUL to be more than 1kN/cm after interim storage
b. Total dynamic relative displacement to be lower than 5 mm in drop during transport
c. Lid system not to make a plastic deformation during transport and to consider it in structural design of CASK
d. Torque for lid bolts to be established moderately

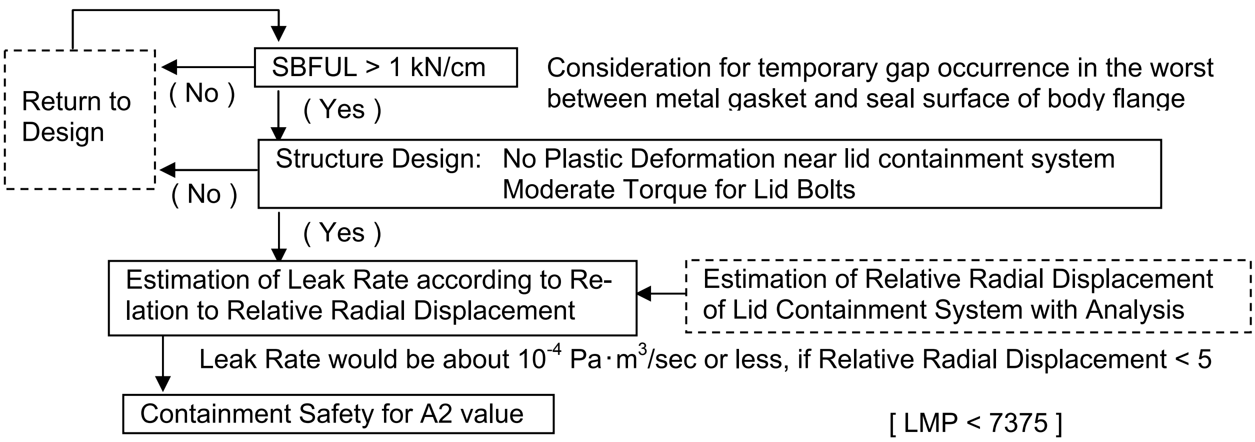

Fig.11. Containment Safety Evaluation during Transport after Interim Storage

5. Conclusion

In this study, property change of component materials during the interim storage were investigated in "material property tests", and seal performance of lid containment system with the relaxed gasket due to degradation during transport was verified and consideration for design was found out.

Reference

[1] "Long-term Program for Research, Development and Utilization of Nuclear Energy", Atomic Energy Commission, Japan, Nov 2000.
[2] O.Kato and T.Ito, "Study on long-term Sealability of Gaskets for Spent Fuel Storage Cask", U92009, Central Research Institute of Electric Power Industry
[3] K.Shirai, K.Fujisawa, "Current Status of R&D Program of Spent Fuel Storage Technology and Long-Term Storage", TCM on "Requirements for Extremely Long-Term Spent Fuel Storage Facilities", IAEA, October 16-20, 2000
[4] T.Yamamoto, "Dual-Purpose Metal Cask Integrity After Long-Term Interim Storage (Activities for the verification test of a dual-purpose metal cask)", PATRAM 2001, September 3-7, 2001, Chicago, IL USA
[5] S.Matuoka, etc. "Verification of Dual-Purpose Metal CASK Integrity -Verification Tests-", IAEA, June 2-6, 2003