Status of Contaminated Water Treatment and Tritium at Fukushima Daiichi Nuclear Power Station

Tokyo Electric Power Company, Inc.
Content

1. Reactor Cooling Status
2. Contaminated Water Status
3. Tritium Status
1. Reactor Cooling Status: Cooling by Recirculating Cooling Water

- Reactors are maintained in a low temperature stable state through continuous cooling which recirculates cooling water and injects it into the reactors.

- Groundwater inflow ~400m³/day

- Reactor cooling water injection ~400m³/day

- Cooling water injection pump

- Cooling water injection tank ~400m³/day

- Contaminated water inside building

- Multi-nuclide removal system (ALPS)

- Cesium removed

- Interim Storage Facility waste sludge, waste adsorbent, etc.

- Storage tanks

-/storage tanks.png
1. Reactor Cooling Status: Individual Unit Status

- Cold shutdown state continues to be maintained at each unit

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Unit 1</td>
<td>~16°C</td>
<td>Unit 1 ~16°C</td>
<td>Unit 1 ~20°C</td>
<td>Unit 1 Feed water system: 2.3 m³/h</td>
</tr>
<tr>
<td></td>
<td>~25°C</td>
<td>Unit 2 ~25°C</td>
<td>Unit 2 ~15°C</td>
<td>Core spray system: 2.0 m³/h</td>
</tr>
<tr>
<td>Unit 3</td>
<td>~22°C</td>
<td>Unit 3 ~20°C</td>
<td>Unit 3 ~15°C</td>
<td>Unit 2 Feed water system: 2.0 m³/h</td>
</tr>
<tr>
<td></td>
<td>–</td>
<td>Unit 4 ~19°C</td>
<td>Unit 4 ~19°C</td>
<td>Core spray system: 2.5 m³/h</td>
</tr>
</tbody>
</table>

*As of March 26, 2014 at 11:00AM*
2. Contaminated Water Status: Overview of Contaminated Water Treatment

① Cesium, which is a major radiation source (gamma ray), is reduced by cesium removal systems
② Saline matter is removed by the desalination system as the water will be used for cooling the reactors
③ The concentration of radioactive materials (excluding tritium) in water, which is retained in tanks, is reduced by the multi-nuclide removal system (ALPS)

Radioactivity Concentrations of Key Nuclides

- Retained water in buildings
- Cesium removal system outlet water
- Seawater desalination system outlet water (fresh water)
- Seawater desalination system outlet water (concentrated brine)
- ALPS outlet water

- Cs–134
- Cs–137
- Gross Beta
- H–3

※ Sampling date: Nov. 5, 2013 (Apr. 9–12, 2013 for ALPS outlet water)
※ Data for cesium removal systems outlet water is used for concentration of gross beta and H-3 in retained water in buildings
※ Sr–90 value is used for gross beta of ALPS outlet water
※ The detectable limit value is used for cases where concentration is below the detectable limit
2. Contaminated Water Status: Cesium Removal Systems

- Usage commenced: June 17, 2011 (Kurion) & Aug. 19, 2011 (SARRY)
- Treatment capacity: 1,200m3/day <respective rated treatment capacity (when one pump is operating)>

**Examples of SARRY (Simplified Active Water Retrieve and Recovery System)**

- Water input
- Pump
- Filtration filter (removes oil, etc.)
- Cesium adsorption tower
- Media filter (prevents outflow of adsorbent)
- Water outlet

Adsorption tower (Bessel)  Bessel loading  Bessel replacement
2. Contaminated Water Status: Desalination System (Reverse Osmosis (RO))

- Salt is removed to make fresh water by using the properties of reverse osmosis membranes, which do not allow ions, saline or other non-water impurities to pass through.
- The system is comprised of a receiving tank, filtration system, RO membranes, treated water tank and other components.
2. Contaminated Water Status: Multi-Nuclide Removal System (ALPS)

- Removes radioactive materials in contaminated water (excluding tritium)
- Testing underway using water containing radioactive materials

![Diagram showing the Multi-Nuclide Removal System (ALPS)](image)

- **Subsystem-A (50% flow rate):** 250 m³/day
  - 3 subsystems A, B & C: 750 m³/day treated by 3 subsystems
  - 2 towers (column type)
  - 14 towers (adsorbent replacement type)

- **High integrity container (HIC):**
  - Removes radioactive materials in contaminated water (excluding tritium)
  - Testing underway using water containing radioactive materials

- Slurry
- Spent adsorbent
- Treated water (stored in tanks, etc.)
2. Contaminated Water Status: Evaluation of Removal Performance During Hot Testing

**Evaluation of Removal Performance During Hot Testing**

ALPS hot tests were conducted using contaminated water (RO concentrated brine). Removal performance was assessed for the 62 nuclides※ targeted for removal. Analysis results of treated water during hot tests of subsystems A, B and C have confirmed the following.

Radiation concentration of Sr-90, a major nuclide, was reduced to 1/100 millionth~1/billionth

Co-60, Ru-106(Rh-106), Sb-125(Te-125m) and I-129 were detected at comparatively high levels

Nuclides in parentheses indicate radioactive equilibrium

※Selection of Nuclides for Removal (Excerpt of items indicated in Implementation Plan)

<table>
<thead>
<tr>
<th>Nuclides Reviewed</th>
</tr>
</thead>
<tbody>
<tr>
<td>・Radioactive materials having their origin in fuel inside the reactors of Units 1~3 (FP nuclides).</td>
</tr>
<tr>
<td>・Radioactive materials having their origin in corrosion products contained in water retained during plant operation (CP nuclides).</td>
</tr>
</tbody>
</table>

**Estimating concentration**

・FP nuclides: Nuclides, which are assumed to be present at significant concentrations based on the results of core inventory assessments, are selected and their concentrations estimated based on the results of accumulated water measurements (2011/3) and core inventory assessments.

・CP nuclides: Selection is made of nuclides, which are contained in water retained in reactors during plant operation, and nuclides, which are present in water retained in concentrated liquid waste tanks and mixed with accumulated water when it was transferred to the high temperature incinerator building, and the results of measurements of such retained waters are used to estimate the concentrations in accumulated water.

**Selecting nuclides for removal**

・Decay during the period from accident occurrence to ALPS operation (approx. 1 year) is assessed to estimate the concentrations.

・Nuclides whose estimated concentration exceeds 1/100 of the notification concentration limit are selected as nuclides for removal which are present at significant concentrations.

・However, as tritium is difficult to remove, it is excluded from the nuclides subject to removal.
2. Contaminated Water Status: Evaluation of Removal Performance in Hot Tests

Ratio to Notification Concentration of ALPS Treated Water

※Sampling dates: Sept. 30~Oct. 4, 2013 (time of subsystem C hot test)
However, for H-3, the value of desalination system outlet water on sampling date Nov. 15, 2013 was used for both values before and after treatment.
The value of cesium removal systems outlet water on sampling date Feb. 14, 2013 was used for C-14 value before treatment and the value of water treated by subsystem C on sampling date Jan. 15, 2014 was used for the value after treatment.
※The detectable limit is used for cases where concentration is below the detectable limit

Notification concentration: Public notice prescribing the dose limit in accordance with the provisions of the Rule for the Installation, Operation, etc. of Commercial Nuclear Power Reactors

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2. Contaminated Water Status: Review of Removal Performance Improvement Measures (Overview of In-plant Flow Passing Testing)

**Review of Performance Improvement Measures**

- There are prospects for improving removal performance by passing nuclides having relatively high radiation concentrations through activated carbon-type adsorbent.
- In laboratory tests, the maintenance of removal performance over a long period of time when being passed by large amounts of water could not be verified.
- Accordingly, test devices, which are filled with an activated carbon-type adsorbent and other materials, are connected to the actual system to conduct flow passing tests (in-plant flow passing tests) to verify the maintenance of removal performance.
- In the in-plant flow passing tests, verification will also be conducted of alternative adsorbents, which are expected to improve removal performance, in addition to activated carbon-type adsorbent.

※Test device installed on subsystem A. Flow passing test conducted 1/24~3/18.
Co-60
There are prospects for removal using activated carbon (confirmation of presence of colloidal-form radioactive materials).
If there is an increase of two columns of activated carbon, higher removal performance can be expected to be obtained.

Sb-125
It is estimated that there is insufficient adsorption capacity with the current two towers of activated carbon and two towers of adsorbent for Sb, etc.
If there is an increase to four towers of activated carbon and four towers of adsorbent for Sb, etc., then higher removal performance can be expected to be obtained.

I-129
In addition to the formation of iodine ions and colloids, it is estimated that there are iodate ions present.
The results of test simulating “iodate ion adsorbent” + “silver-loaded adsorbent” + “activated carbon” confirmed a higher removal performance when water was passed through for approximately 10 days.
In-plant tests are scheduled to be continued to verify lifespan.

Ru-106
Based on cold tests, media capable of removing Ru was selected (verification scheduled to be conducted using in-plant tests).

Based on test results, improved removal performance (sum of notification concentration limits from the current approximately 6 to 0.5~0.6 after improvements) can be expected to be obtained by adding an additional two adsorption towers and modifying the tower configuration as in the diagram below.
2. Contaminated Water Status: Storage of Contaminated Water

- Total storage capacity*: approx. 490,000 m³
- Total storage volume*: approx. 460,000 m³
- Plan to increase capacity to 800,000 m³ (completion target: end of FY2014)

【Storage capacity by tank type*】
- Steel square tanks: approx. 3,000 m³
- Steel cylindrical tanks (flange): approx. 300,000 m³
- Steel cylindrical tanks (welded): approx. 140,000 m³
- Steel horizontal tanks: approx. 40,000 m³

* As of April 22, 2013
2. Contaminated Water Status:

Map of Contaminated Water

- **Trenches** [approx. 11,000m³]
- **Units 3 & 4 buildings** [approx. 39,700m³]
- **HTI building** [approx. 4,100m³]
- **Processing building** [approx. 14,040m³]
- **Units 1 & 2 buildings** [approx. 35,300m³]

Contamination sources

- Retained water
- High level: Contaminated water with Cs-137 10^3 - 10^8 Bq/cm³ concentration as well as adsorption towers, high integrity containers, etc.
- Medium/low level: Contaminated water under Cs-137 10^2 Bq/cm³ concentration

- **Fresh water receiving tanks** [approx. 29,000m³]
- **Horizontal tanks** (concentrated brine) [approx. 30,000m³]
- **Flange tanks** (concentrated brine) [approx. 250,000m³]
- **Welded tanks** (concentrated brine) [approx. 30,000m³]
- **Units 1 & 2 buildings** [approx. 35,300m³]
- **HTI building** [approx. 4,100m³]
- **Processing building** [approx. 14,040m³]
- **Units 3 & 4 buildings** [approx. 39,700m³]
- **Trenches** [approx. 11,000m³]
- **Horizontal tanks** (concentrated brine) [approx. 30,000m³]
- **Flange tanks** (concentrated brine) [approx. 250,000m³]
- **Welded tanks** (concentrated brine) [approx. 30,000m³]

- **Adsorption tower**
- **Drain ditch**
- **Welded tanks** (concentrated brine) [approx. 30,000m³]
- **Flange tanks** (concentrated brine) [approx. 250,000m³]
- **Treated water storage tanks** (ALPS treated water) [approx. 23,000m³]

※Source: December 2013 meeting of Committee on Countermeasures for Contaminated Water Treatment “Preventive and Multi-tiered Contaminated Water Treatment Measures at TEPCO’s Fukushima Daiichi Nuclear Power Station”

<As of October 15, 2013>
2. Contaminated Water Status: Volume of Treated Water (Water Stored in Tanks)

- Total volume of treated water (concentrated brine, concentrated liquid waste, ALPS treated water, and fresh water): **approx. 460,000m³**.
- Of this, total volume of ALPS treated water: **approx. 73,000m³**. ※As of April 22, 2014 (By end of FY2014, ALPS treatment of all tank water is scheduled to be completed)

### Volume of Treated Water Stored※1

<table>
<thead>
<tr>
<th>Year</th>
<th>Storage Volume (m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2011/7/5</td>
<td>50,000</td>
</tr>
<tr>
<td>2011/9/5</td>
<td>55,000</td>
</tr>
<tr>
<td>2011/11/5</td>
<td>60,000</td>
</tr>
<tr>
<td>2012/1/5</td>
<td>65,000</td>
</tr>
<tr>
<td>2012/3/5</td>
<td>70,000</td>
</tr>
<tr>
<td>2012/5/5</td>
<td>75,000</td>
</tr>
<tr>
<td>2012/7/5</td>
<td>80,000</td>
</tr>
<tr>
<td>2012/9/5</td>
<td>85,000</td>
</tr>
<tr>
<td>2012/11/5</td>
<td>90,000</td>
</tr>
<tr>
<td>2013/1/5</td>
<td>95,000</td>
</tr>
<tr>
<td>2013/3/5</td>
<td>100,000</td>
</tr>
<tr>
<td>2013/5/5</td>
<td>105,000</td>
</tr>
<tr>
<td>2013/7/5</td>
<td>110,000</td>
</tr>
<tr>
<td>2013/9/5</td>
<td>115,000</td>
</tr>
<tr>
<td>2013/11/5</td>
<td>120,000</td>
</tr>
<tr>
<td>2014/1/5</td>
<td>125,000</td>
</tr>
<tr>
<td>2014/3/5</td>
<td>130,000</td>
</tr>
</tbody>
</table>

**Source:** Decommissioning Promotion Council data (status of accumulated water treatment)

※1: ① is volume of building accumulated water stored: approx. 94,640m³
※2: ③ is total for concentrated brine and concentrated liquid waste

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### 3. Tritium Status: Total Tritium Amount

Amount of tritium at Fukushima Daiichi Nuclear Power Station (Units 1~4) is estimated to be as follows.

<table>
<thead>
<tr>
<th>Points estimated</th>
<th>Tritium amount</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>[Bq]</td>
<td>[g]※1</td>
</tr>
<tr>
<td>Total amount</td>
<td>~3.4 × 10(^{15})</td>
<td>T: ~9.5</td>
</tr>
<tr>
<td>[Break down]</td>
<td></td>
<td></td>
</tr>
<tr>
<td>• Water accumulated in tanks</td>
<td>~8.3 × 10(^{14})</td>
<td>T: ~2.3 (THO: ~15.5)</td>
</tr>
<tr>
<td>• Water accumulated in buildings</td>
<td>~5.0 × 10(^{13})</td>
<td>T: ~0.14 (THO: ~0.9)</td>
</tr>
<tr>
<td>• Water in seawater pipe trenches</td>
<td>~4.6 × 10(^{13})</td>
<td>T: ~0.14 (THO: ~0.9)</td>
</tr>
<tr>
<td>• Other</td>
<td>~2.5 × 10(^{15})</td>
<td>T: ~6.9</td>
</tr>
</tbody>
</table>

※1: Weight of tritium atoms (figure in parentheses shows the weight corresponding to the THO form)
※2: ORIGEN 2 is used to assess the in-core tritium inventory at the time of the accident (see pp. 10~12)
※3: Estimated based on desalination system outlet concentration data and volume of water stored in tanks (see pp. 13~14)
※4: Estimated based on desalination system outlet concentration data (March 2014) and volume of water accumulated in buildings (approx. 92,000m\(^3\))
※5: Estimated based on desalination system outlet concentration data (September 2011) and volume of water accumulated in trenches (approx. 11,000m\(^3\))
※6: Calculated by subtracting amount of tritium in water stored in tanks, water accumulated in buildings and trenches from total amount (It is estimated that tritium other than that in tanks, buildings and trenches is mainly present in fuel debris and other such matter)
### 3. Tritium Status: Chemical Properties of ALPS Treated Water

<table>
<thead>
<tr>
<th>Sample date/time</th>
<th>April 16, 2013 10:30</th>
<th>April 18, 2013 11:30</th>
<th>[Ref]* Guidelines for General Drainage Channels and Discharged Water</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>General properties</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Conductivity (μS/cm)</td>
<td>6220</td>
<td>6200</td>
<td></td>
</tr>
<tr>
<td>Chlorine (ppm)</td>
<td>2100</td>
<td>1900</td>
<td></td>
</tr>
<tr>
<td><strong>Items with possible impact on environment</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>pH</td>
<td>7.6</td>
<td>7.4</td>
<td>Water area: 5.8～8.6 Sea area: 5.0～9.0</td>
</tr>
<tr>
<td>Suspended solids (mg/L)</td>
<td>&lt;1</td>
<td>&lt;1</td>
<td>Max. 70 or below Max. 40 or below</td>
</tr>
<tr>
<td>Chemical oxygen demand (COD) (mg/L)</td>
<td>&lt;1</td>
<td>1</td>
<td>Mean 50 or below Mean 30 or below</td>
</tr>
<tr>
<td><strong>Items with possible impact on tritium separation, etc.</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Concentration of ionic species (ppm)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Na⁺</td>
<td>1480</td>
<td>1590</td>
<td></td>
</tr>
<tr>
<td>NH₄⁺</td>
<td>30</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>NO₂⁻</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>NO₃⁻</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>SO₄²⁻</td>
<td>760</td>
<td>610</td>
<td></td>
</tr>
</tbody>
</table>

※According to Guidelines for General Drainage Channels and Discharged Water prescribed by in Appendix 5 of Article 25 of the Ordinance for Enforcement Concerning Conservation, etc. of Living Environment in Fukushima Prefecture
Sources of Tritium Production at Nuclear Power Stations

① Production by ternary fission of fuel (reaction in which nuclear fission breaks uranium into three fragments)
② Production by neutron irradiation of boron-10 contained in boron carbide control rods
③ Activation of reactor water (production by neutron irradiation with lithium, etc. as heavy water and impurities)

Numeral ① above is a major source, but there is no additional production as Fukushima Daiichi NPS is currently subcritical

① Production by ternary fission of fuel

➢ Tritium is produced as a fission product of $^{235}\text{U}$ and $^{239}\text{Pu}$ during “burning (fission)”. The amount is 0.013% of $^{235}\text{U}$ and 0.023% of $^{239}\text{Pu}$, and $^{239}\text{Pu}$ in fuel becomes predominant as the fuel burns further. The amount of tritium generated per 1MW with assuming 0.018% as the average is as follows.

$$N_F \times 1.8 \times 10^{-4} \times \lambda = 1.01 \times 10^4 \text{ Bq/s} \cdot \text{MWt}$$

NF: No. of fissions per 1MWt in 1 second = $3.15 \times 10^{16} / \text{s} \cdot \text{MWt}$

➢ The amount of tritium generated inside a reactor is approximately that given below. If the fuel rods are not damaged, then almost none is released (percentage permeating the cladding is $\sim 10^{-4}$).

<table>
<thead>
<tr>
<th>Amount of tritium produced</th>
<th>500,000kWe</th>
<th>800,000kWe</th>
<th>1,100,000kWe</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bq/month</td>
<td>4.07E+13</td>
<td>6.29E+13</td>
<td>8.51E+13</td>
</tr>
</tbody>
</table>
② Production by neutron irradiation of boron-10 contained in boron carbide control rods

- Tritium is produced by the following reaction inside the poison rods of control rods using \( B_4C \) (boron carbide) for neutron absorption
  \[
  ^{10}B + n \rightarrow ^3H + 2^4He
  \]
  \[
  ^{10}B + n \rightarrow ^7Li + ^4He
  \]
  \[
  ^7Li + n \rightarrow ^3H + ^4He + n
  \]

- In 1 g of \( B_4C \) irradiated until \(^{10}B\) decreases approximately 50%, a conservative assessment puts the amount of tritium produced at \(1.48 \times 10^9\)Bq/g\( B_4C \), but, even if the control rod is damaged, the tritium will not be released all at once.

③ Activation of reactor water (production by neutron irradiation into heavy water, etc.)

- Tritium is produced by neutron irradiation into heavy water in the reactor water. The amount of tritium generated into heavy water is calculated using the following equation.

\[
^2H + n \rightarrow ^3H + \gamma
\]

\[
N^3H = \Sigma(D_2O) \times \phi \times V \times \rho \times (D/H) \times t
\]

- The amount of tritium produced inside a reactor is approximately as given below.

<table>
<thead>
<tr>
<th>Amount of tritium produced</th>
<th>500,000kWe</th>
<th>800,000kWe</th>
<th>1,100,000kWe</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bq/month</td>
<td>4.44E+09</td>
<td>6.29E+09</td>
<td>1.07E+10</td>
</tr>
</tbody>
</table>
The in-core tritium inventory is mainly that which is produced by ternary fission. Thus, ORIGEN 2 (nuclear fuel burn-up calculation code) was used to perform a detailed assessment of the time of the accident, which resulted in the following.

- Of the tritium found as the result of assessment prior to the accident, approximately 60% is believed to be occluded in fuel rod cladding.
- Currently (end of March 2014), the total amount of tritium contained in water stored in tanks is estimated to be \(8.34 \times 10^{14}\)Bq. By the end of February 2016, this amount is estimated to be approximately \(9 \times 10^{14}\)Bq.
- The concentration of tritium contained in water which will be newly treated at that time (end of February 2016) is estimated to be \(6.7 \times 10^4\)Bq/liter.
- \(3.4 \times 10^{15}\)Bq, which is the total amount of tritium in Units 1~3 in the above table, is an amount equivalent to a tritium atoms’ weight of 9.5g. (If tritium is present in “THO” form, then the atoms’s weight is equivalent to 63.3g.)

<table>
<thead>
<tr>
<th></th>
<th>Unit 1</th>
<th>Unit 2</th>
<th>Unit 3</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>TEPCO</td>
<td>1.0E+15</td>
<td>1.2E+15</td>
<td>1.2E+15</td>
<td>3.4E+15</td>
</tr>
<tr>
<td>&lt;Ref&gt; JAEA</td>
<td>9.4E+14</td>
<td>1.2E+15</td>
<td>1.2E+15</td>
<td>3.3E+15</td>
</tr>
</tbody>
</table>

* Toshiba Corporation “Compaction Tests of Irradiated Hulls” (report for work performed under contract with JAEA (formerly, the Power Reactor and Nuclear Fuel Development Corporation) September 1996.
Concentration of Tritium in Newly RO Treated Water

![Graph showing concentration of tritium over time with a curve fitting equation and a specific value for March 2016]

- Equation: $y = 4 \times 10^6 e^{-0.0764x}$
- March 2016: $6.71 \times 10^4$ Bq/liter
【Reference】4. Cumulative Amount of Tritium Stored in Tanks at Fukushima Daiichi NPS

- Case A: groundwater bypass implemented, sub-drains pumped out, rainwater drained, and groundwater drains drained
- Case B: groundwater bypass not implemented, sub-drains not pumped out, rainwater drained, and storage of groundwater in drains

In addition, it is estimated that there will be $6.08 \times 10^{12}$ Bq in the R/B, T/B, Central RW and HIT buildings by March 2016.
Source: Decommissioning Promotion Council data (status of accumulated water treatment): concentrated brine after RO treatment has twice the saline matter as that prior to treatment, therefore the values noted in the data were doubled and then plotted.