

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

Noboru.Ishizawa

Project Planning Department

Fukushima Daiichi Decontamination and
Decommissioning Engineering Company

Tokyo Electric Power Company, Inc.



東京電力

Content

1

1. Reactor Cooling Status
2. Contaminated Water Status
3. Tritium Status

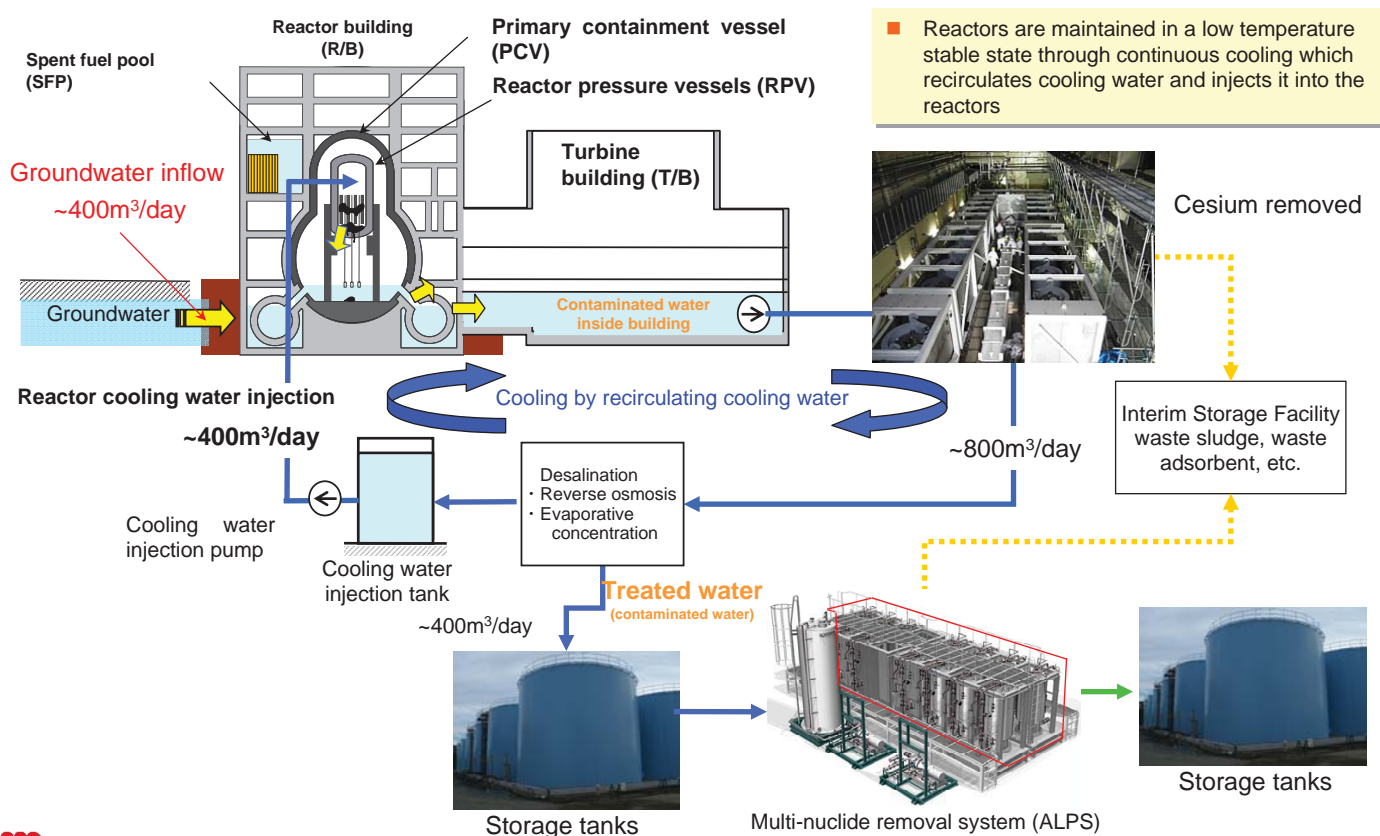
Layout of Fukushima Daiichi Nuclear Power Station

2



1. Reactor Cooling Status: Cooling by Recirculating Cooling Water

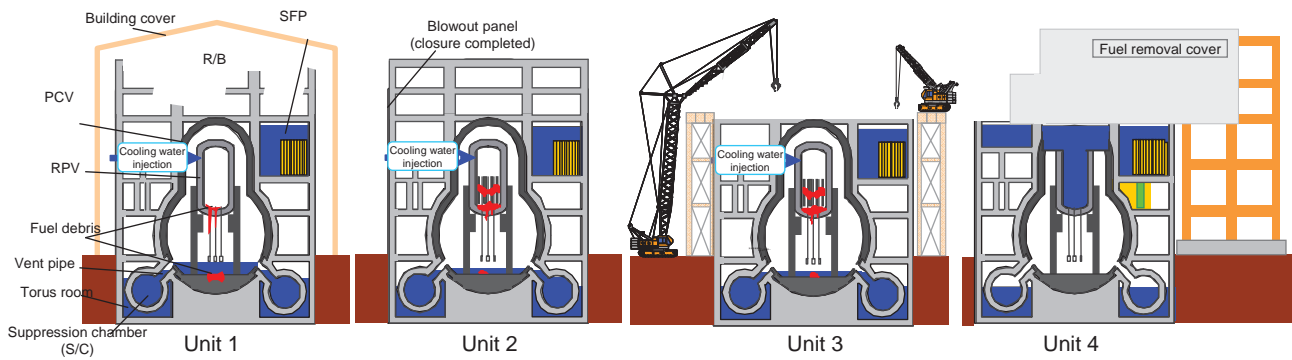
3



1. Reactor Cooling Status: Individual Unit Status

4

■ Cold shutdown state continues to be maintained at each unit



RPV bottom temp. [°C]		PCV internal temp. [°C]		Fuel pool temp. [°C]		Reactor cooling water vol.	
Unit 1	~16°C	Unit 1	~16°C	Unit 1	~20°C	Unit 1	Feed water system: 2.3m³/h Core spray system: 2.0m³/h
Unit 2	~25°C	Unit 2	~25°C	Unit 2	~15°C	Unit 2	Feed water system: 2.0m³/h Core spray system: 2.5m³/h
Unit 3	~22°C	Unit 3	~20°C	Unit 3	~15°C	Unit 3	Feed water system: 2.0m³/h Core spray system: 2.5m³/h
Unit 4	—	Unit 4	—	Unit 4	~19°C	Unit 4	—



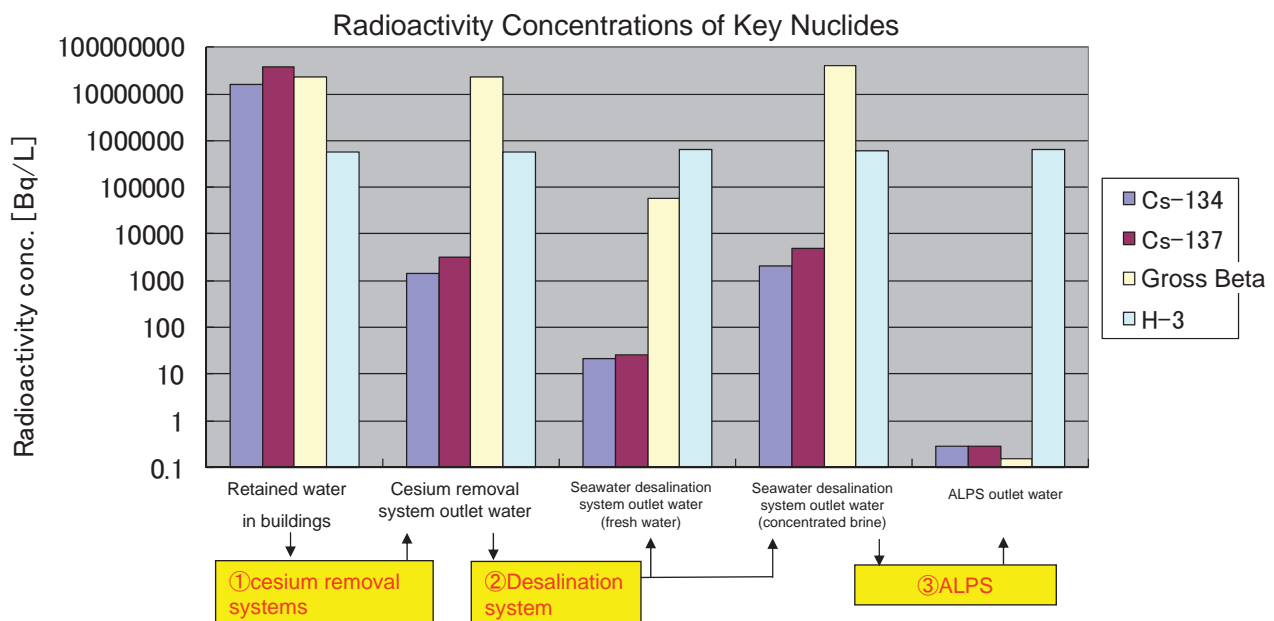
Unauthorized duplication / reproduction prohibited. Tokyo Electric Power Company

As of March 26, 2014 at 11:00AM

2. Contaminated Water Status: Overview of Contaminated Water Treatment

5

- 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)

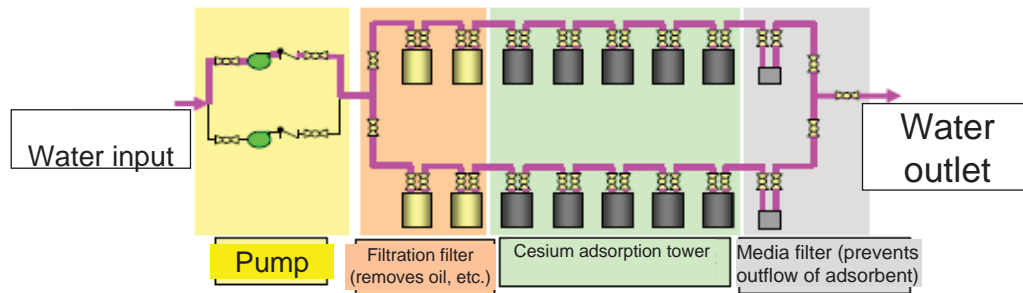


※Sampling date: Nov. 5, 2013 (Apr. 9-12, 2013 for ALPS outlet water)
 ※Data for cesium removal systems outlet water is used for concentration of gross beta and H-3 in retained water in buildings
 ※Sr-90 value is used for gross beta of ALPS outlet water
 ※The detectable limit value is used for cases where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



Unauthorized duplication / reproduction prohibited. Tokyo Electric Power Company

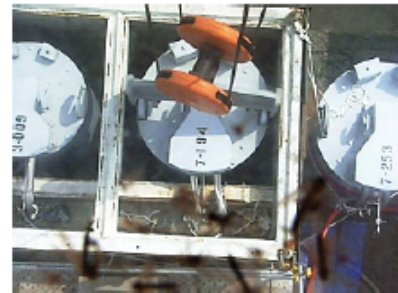
- Usage commenced: June 17, 2011 (Kurion) & Aug. 19, 2011 (SARRY)
- Treatment capacity: 1,200m³/day <respective rated treatment capacity (when one pump is operating)>



Adsorption tower (Bessel)



Vessel loading



Bessel replacement

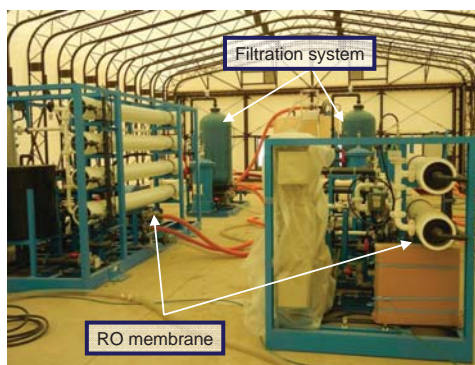
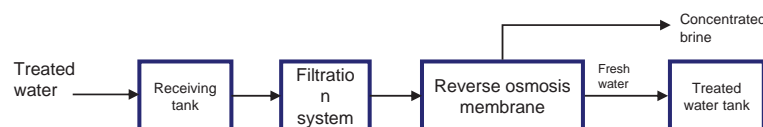
【Examples of SARRY (Simplified Active Water Retrieve and Recovery System)】



Unauthorized duplication / reproduction prohibited. Tokyo Electric Power Company

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.



<Desalination system (RO module)>



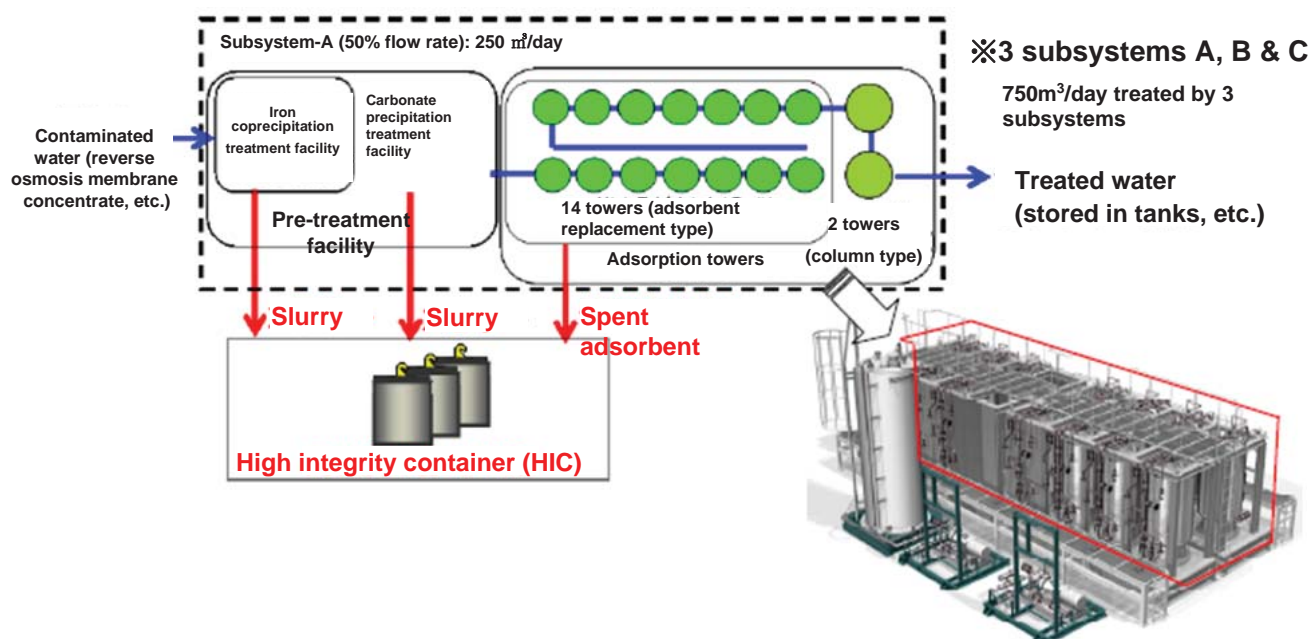
<Desalination system (filtration system)>



Unauthorized duplication / reproduction prohibited. Tokyo Electric Power Company

2. Contaminated Water Status: Multi-Nuclide Removal System (ALPS) 8

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



2. Contaminated Water Status: Evaluation of Removal Performance During Hot Testing 9

■ 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 1/100millionth~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)

【Nuclides Reviewed】

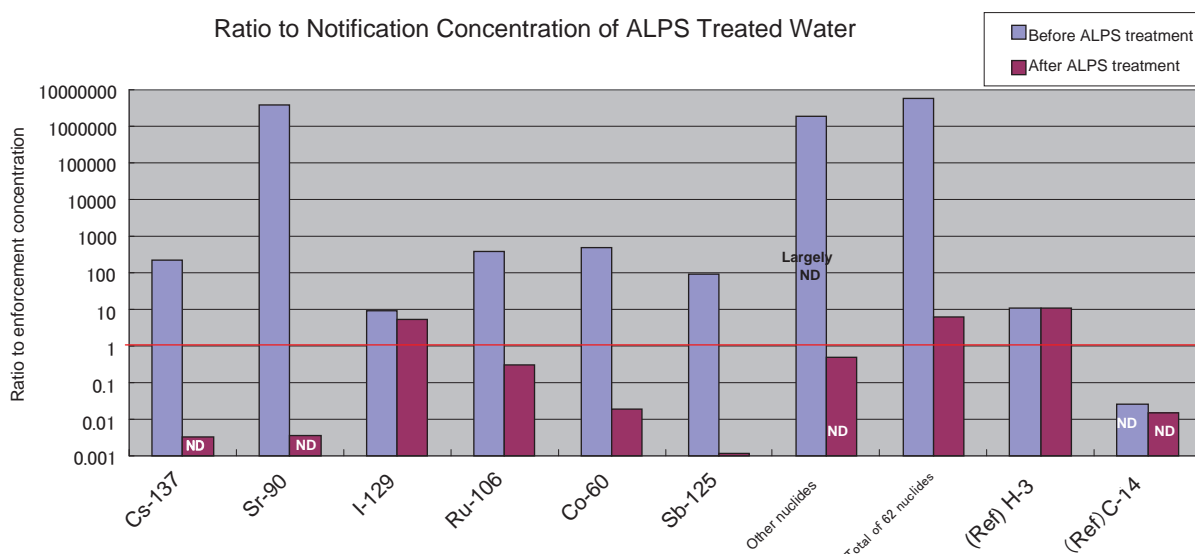
- Radioactive materials having their origin in fuel inside the reactors of Units 1~3 (FP nuclides).
- Radioactive materials having their origin in corrosion products contained in water retained during plant operation (CP nuclides).

【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.



※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



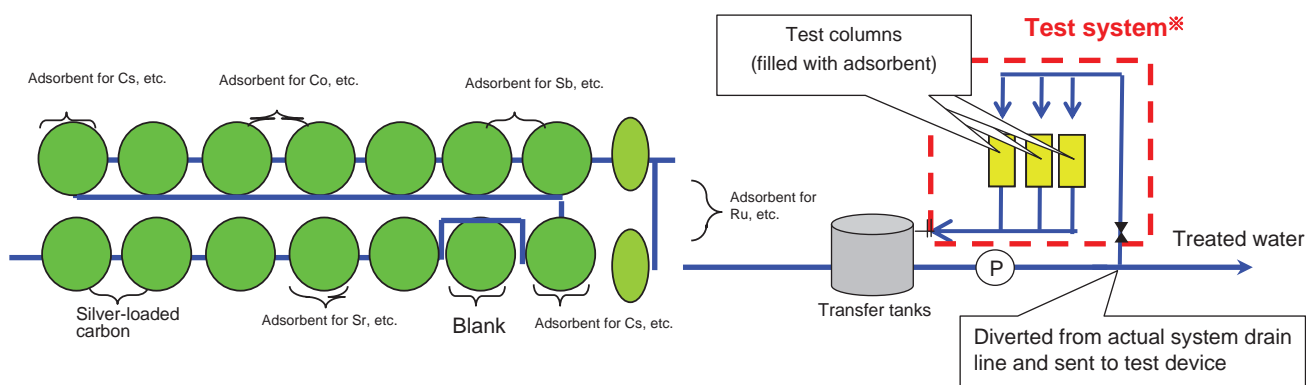
Unauthorized duplication / reproduction prohibited. Tokyo Electric Power Company

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

11

■ 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.



Unauthorized duplication / reproduction prohibited. Tokyo Electric Power Company

2. Contaminated Water Status: In-plant Test Results (Interim) & Future Policy 12

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

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

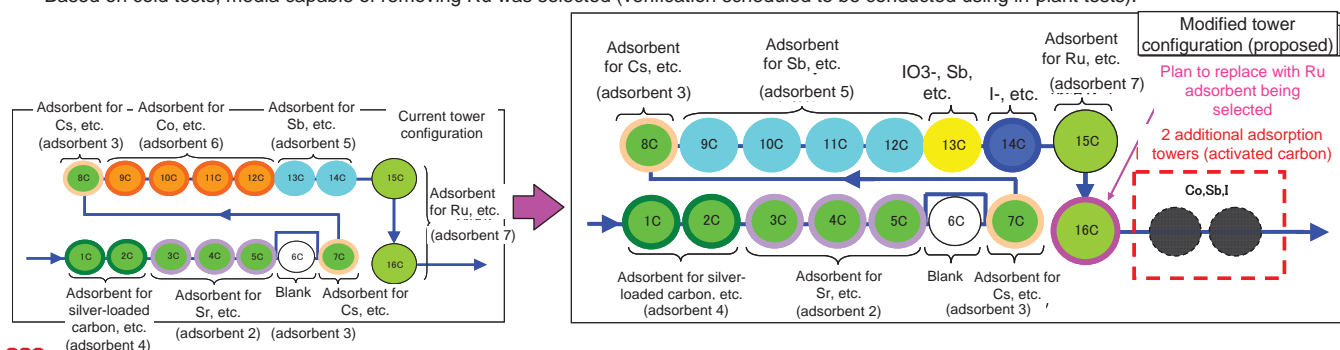
In addition to the formation of iodine ions and colloids, it is estimated that there are iodate ions present.

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).



Unauthorized duplication / reproduction prohibited. Tokyo Electric Power Company

2. Contaminated Water Status: Storage of Contaminated Water 13

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



【Storage capacity by tank type*】

Steel square tanks	: approx. 3,000m ³
Steel cylindrical tanks (flange)	: approx. 300,000m ³
Steel cylindrical tanks (welded)	: approx. 140,000m ³
Steel horizontal tanks	: approx. 40,000m ³

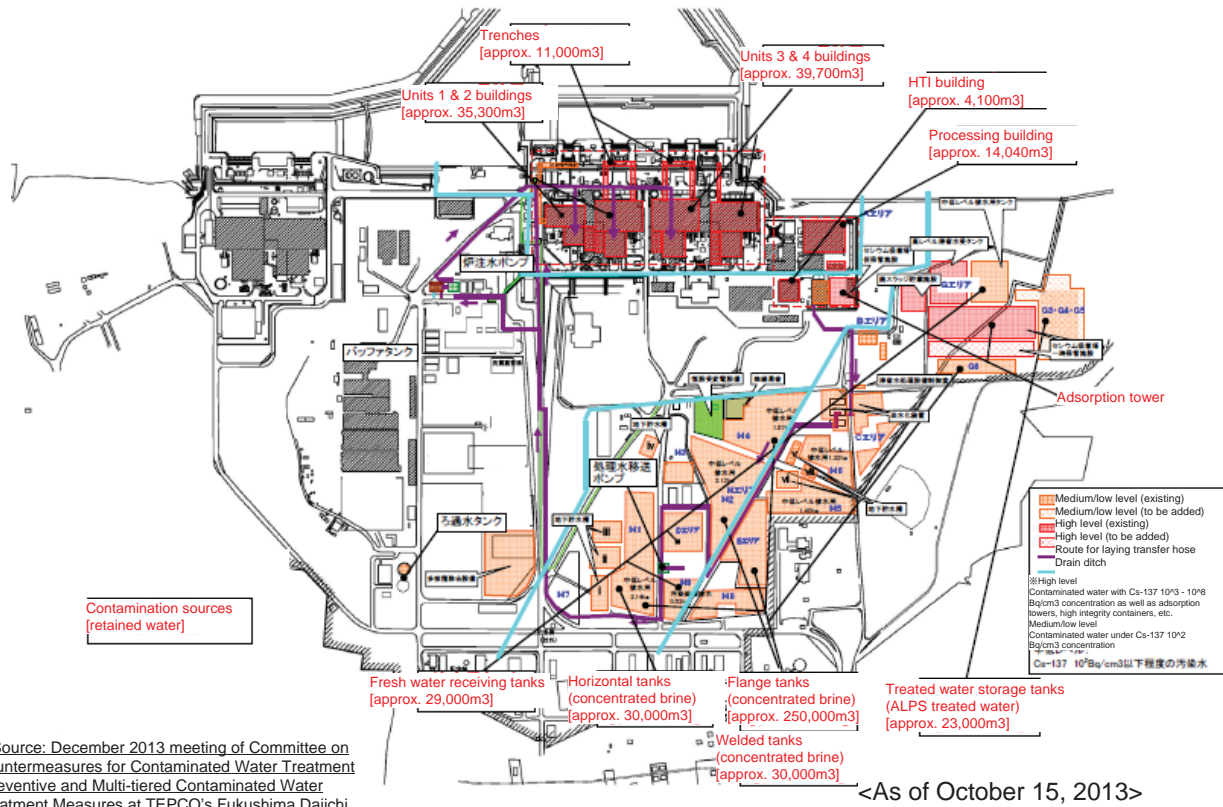
* As of April 22, 2013



Unauthorized duplication / reproduction prohibited. Tokyo Electric Power Company

2. Contaminated Water Status: Map of Contaminated Water

14



※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"



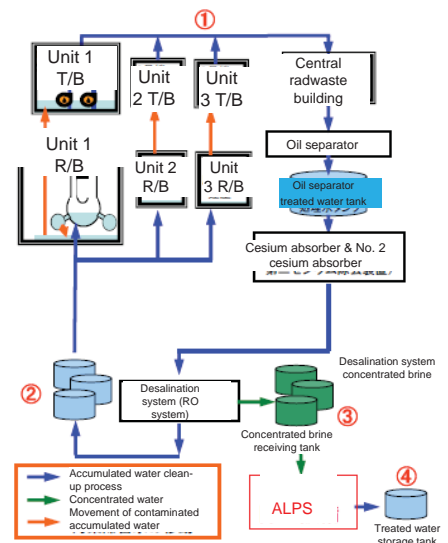
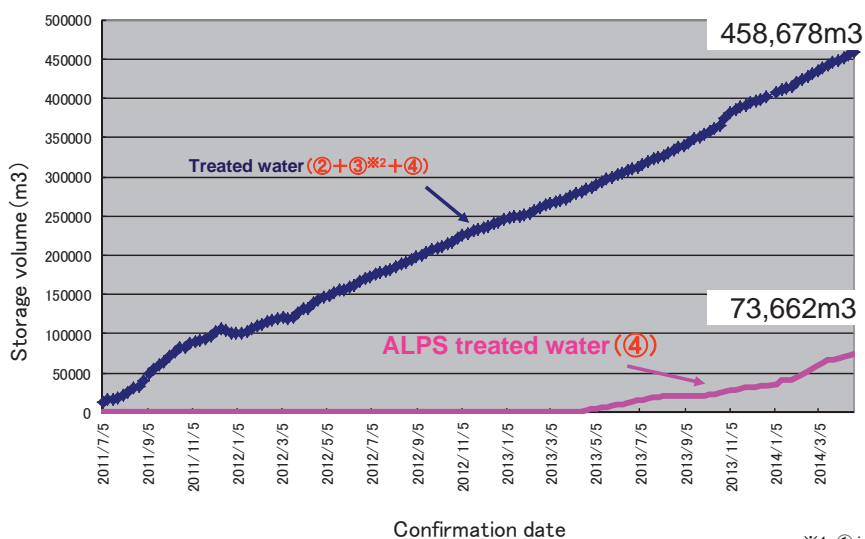
Unauthorized duplication / reproduction prohibited. Tokyo Electric Power Company

2. Contaminated Water Status: Volume of Treated Water (Water Stored in Tanks)

15

- 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



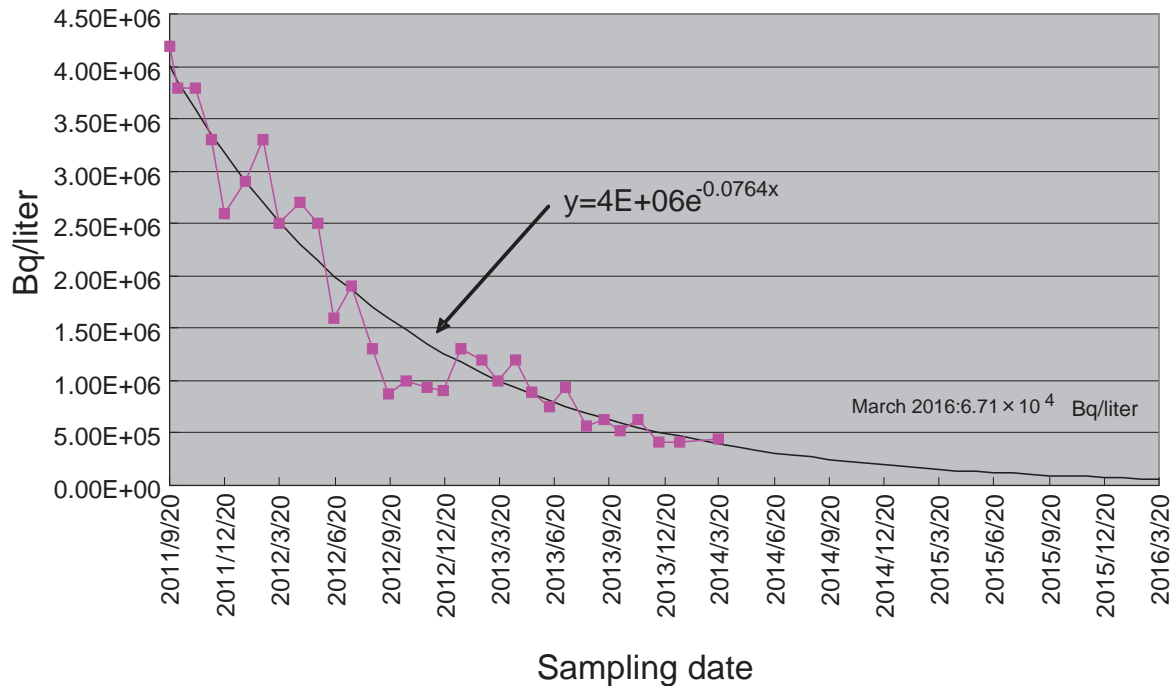
※1: ① is volume of building accumulated water stored: approx. 94,640m³
※2: ③ is total for concentrated brine and concentrated liquid waste

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

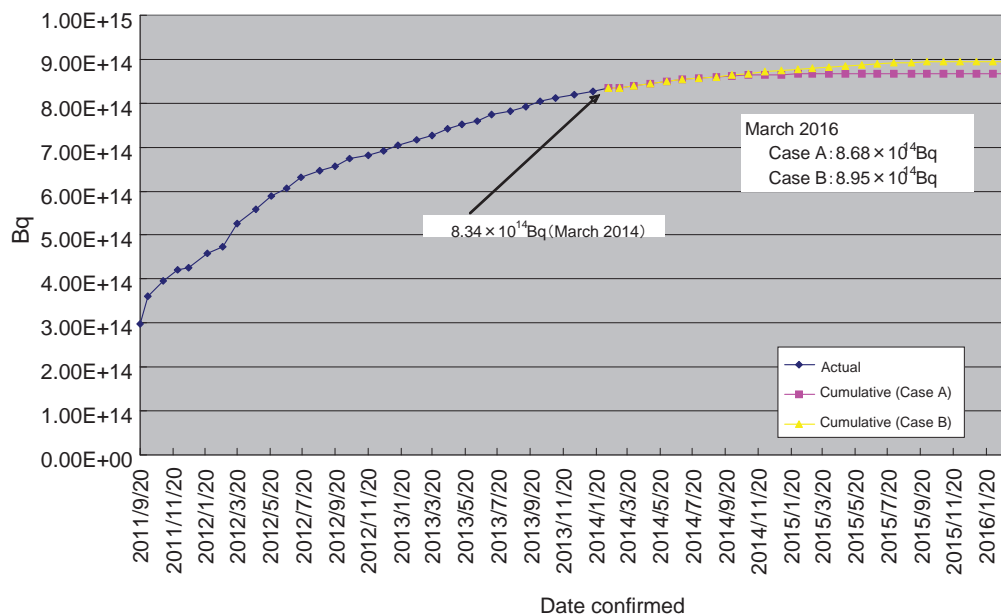


Unauthorized duplication / reproduction prohibited. Tokyo Electric Power Company

Concentration of Tritium in Newly RO Treated Water



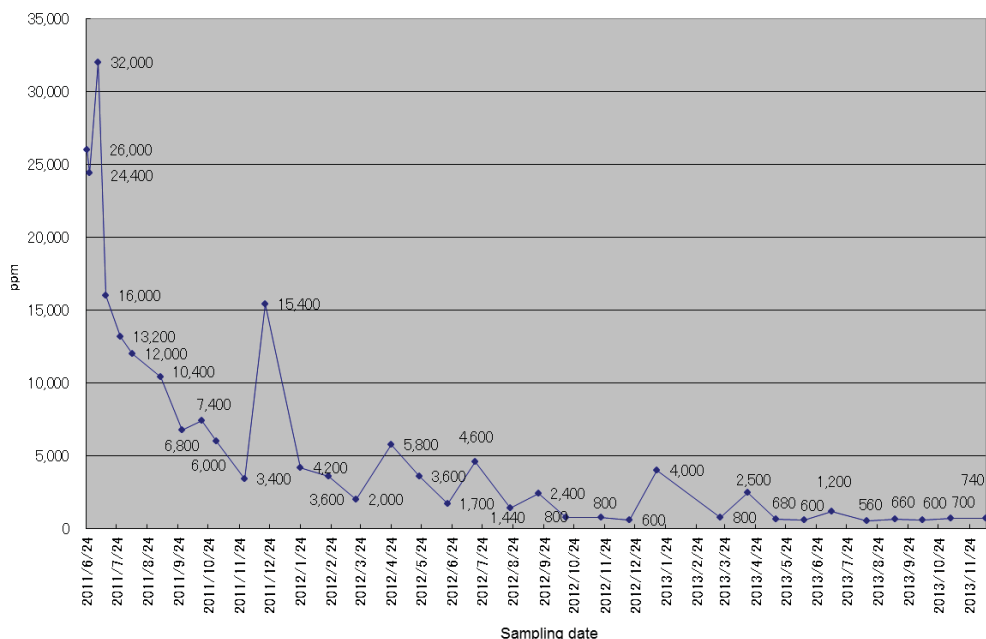
Cumulative Amount of Tritium



- 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 6.08×10^{12} Bq in the R/B, T/B, Central RW and HIT buildings by March 2016.

Concentration of Saline in Water after Reverse Osmosis Treatment



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.

3. Tritium Status: Total Tritium Amount

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

As of March 25, 2014

Points estimated		Tritium amount		Notes
		[Bq]	[g]※1	
Total amount		$\sim 3.4 \times 10^{15}$	T: ~ 9.5	※2
[Break down]	•Water accumulated in tanks	$\sim 8.3 \times 10^{14}$	T: ~ 2.3 (THO: ~ 15.5)	※3
	•Water accumulated in buildings	$\sim 5.0 \times 10^{13}$	T: ~ 0.14 (THO: ~ 0.9)	※4
	•Water in seawater pipe trenches	$\sim 4.6 \times 10^{13}$	T: ~ 0.14 (THO: ~ 0.9)	※5
	•Other	$\sim 2.5 \times 10^{15}$	T: ~ 6.9	※6

※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. 92,000m³)

※5: Estimated based on desalination system outlet concentration data (September 2011) and volume of water accumulated in trenches (approx. 11,000m³)

※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

20

			ALPS Treated Water		【Ref】* Guidelines for General Drainage Channels and Discharged Water
	Sample date/time		April 16, 2013 10:30	April 18, 2013 11:30	
General properties	Conductivity (μ S/cm)		6220	6200	
	Chlorine (ppm)		2100	1900	
Items with possible impact on environment	pH		7.6	7.4	Water area:5.8~8.6 Sea area:5.0~9.0
	Suspended solids (mg/L)		<1	<1	Max. 70 or below Mean 50 or below
	Chemical oxygen demand (COD) (mg/L)		<1	1	Max. 40 or below Mean 30 or below
Items with possible impact on tritium separation, etc.	Concentrat ion of ionic species (ppm)	Na ⁺	1480	1590	
		NH4 ⁺	30	0	
		NO2 ⁻	0	0	
		NO3 ⁻	0	0	
		SO4 ²⁻	760	610	

※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

3. Tritium Status: Research of Remove Technology

21

Research and development of handling are performed by the atomic power (Heavy Water reactor etc.) which needs to separate tritium as impurities, and the nuclear fusion which treats tritium as a fuel substance.

Heavy Water Reactor (CANDU type, Advanced Thermal Reactor "Fugen" , etc.)

Deuterium will generate tritium, if a neutron is absorbed. In order to separate tritium from heavy water, the isotope separation of DTO-D2O is required.

Nuclear Fusion Reactor

Technology of fuel treatment

Unreacted fuel is processed and it uses efficiently.

Recovery of waste water and waste gaseous

Recovery from tritium contamination in Groundwater

Hanford site (DOE)

Study in EPRI

Such technology is investigated focusing on literature documentation and an interview.

Literature documentation

The technology adopted the past research and in and outside is investigated for a paper etc.

Interview

Directly, the talk is heard from a domestic researcher and an engineer.

Example: Researcher, Expert, Institution of research (JAEA, NIRS, etc.)

The thing applicable by the present is not found.

As main remove technology, some were following as a result of investigation so far.

- ① Water Distillation
Example····Tokai Reprocess (JAEA), Manhattan project (USA), etc.
- ② Cryogenic Distillation
Example····JRR-3M (JAEA), Institute Laue-Langevin (France), etc.
- ③ Vapor Phase Catalytic Exchange (VPCE)
Example····Darlington NPS (Canada), Institute Laue-Langevin (France), etc.
- ④ Liquid Phase Catalytic Exchange (LPCE)
Example····Chalk River Laboratories (Canada), Wolsong NPS (Korea), etc.
- ⑤ Electrolysis
Example····Can't find
- ⑥ Combined Electrolysis Catalytic Exchange (CECE)
Example····"Fugen" Upgrader II (JAEA), etc.
- ⑦ Bithermal Hydrogen-Water Process
Example····Can't find

Besides method :Isotope Effect (Girdler Sulfide Process, Ammonia Process, Amine Process, etc), Adsorption remove under deep low temperature, etc. Anything method is troublesome and small throughput.

- ① Water Distillation
 - ✓ The method of separating by the difference in the steam pressure of H_2O , HTO , T_2O .
 - ✓ Although dissociating to an environmental level theoretically is possible, since relative volatility is close to about 1, the separation performance per unit number of stages is small, and becomes very large-scale equipment including a building.
 - ✓ Sufficient consideration for that energy consumption is large and the measure at the time of failure is required.
- ② Cryogenic Distillation
 - ✓ How to separate by the difference in the boiling point of hydrogen gas (H_2 , HT , T_2)
 - ✓ The energy consumption for using cryogenic temperature is large, and a throughput's is small.
 - ✓ Sufficient consideration for the measure against gas disclosure at the time of coolant loss is required.
- ③ Vapor Phase Catalytic Exchange (VPCE)
 - ✓ How to perform the displacement reaction of a hydrogen atom under high temperature using a catalyst.
 - ✓ With the technology for high concentration tritium, since it is a reaction of gas and gas, the multi-stage effect cannot be acquired.
- ④ Liquid Phase Catalytic Exchange (LPCE)
 - ✓ How to perform the displacement reaction of a hydrogen atom at low temperature using a catalyst.
 - ✓ With the technology for high concentration tritium, since the internal structure for distributing liquid uniformly in a tower is complicated, a processing flow has a maximum.

⑤ Electrolysis

- ✓ It uses that a heavy constituent decreases compared with the corresponding contrast in an electrolysis solution for the hydrogen gas emitted with cathode.
- ✓ Energy consumed is large, and if a multi-stage cascade is built, the amount of consumption will become serious. Furthermore, it is easy to be subject to the influence of impurities, and independent use is disadvantageous.

⑥ Combined Electrolysis Catalytic Exchange (CECE)

- ✓ What combined two kinds of technology.
- ✓ With the technology for high concentration tritium, a throughput has a maximum.
- ✓ The process of removing alkali is required for the alkali electrolysis vessel adopted by "Fugen."

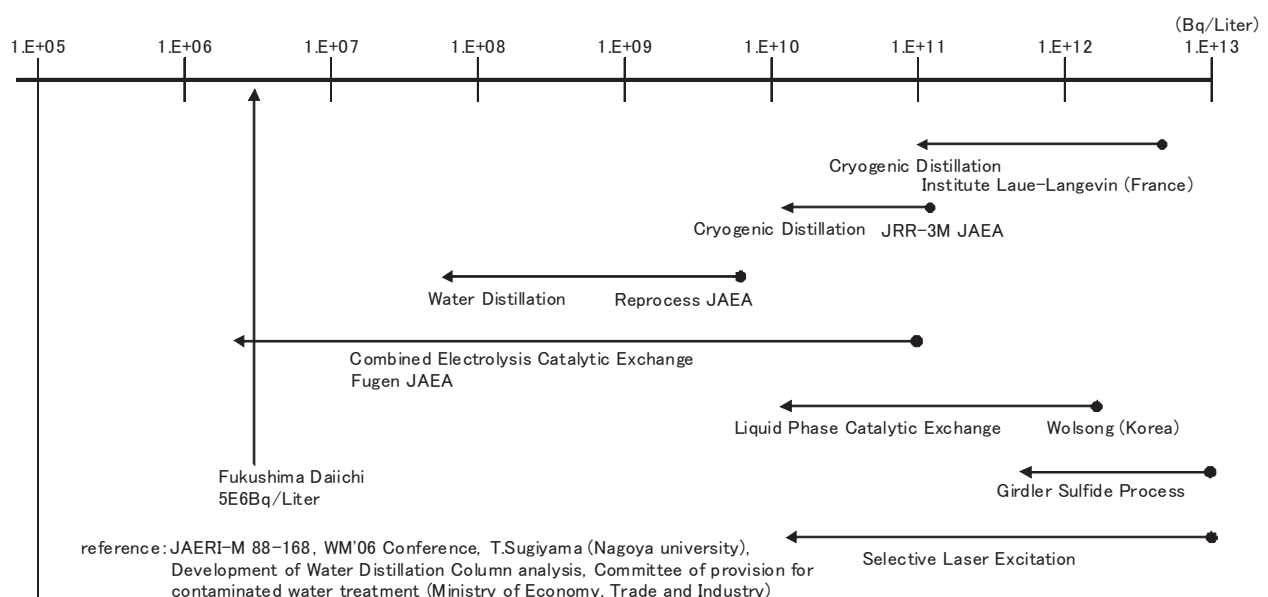
⑦ Bithermal Hydrogen-Water Process

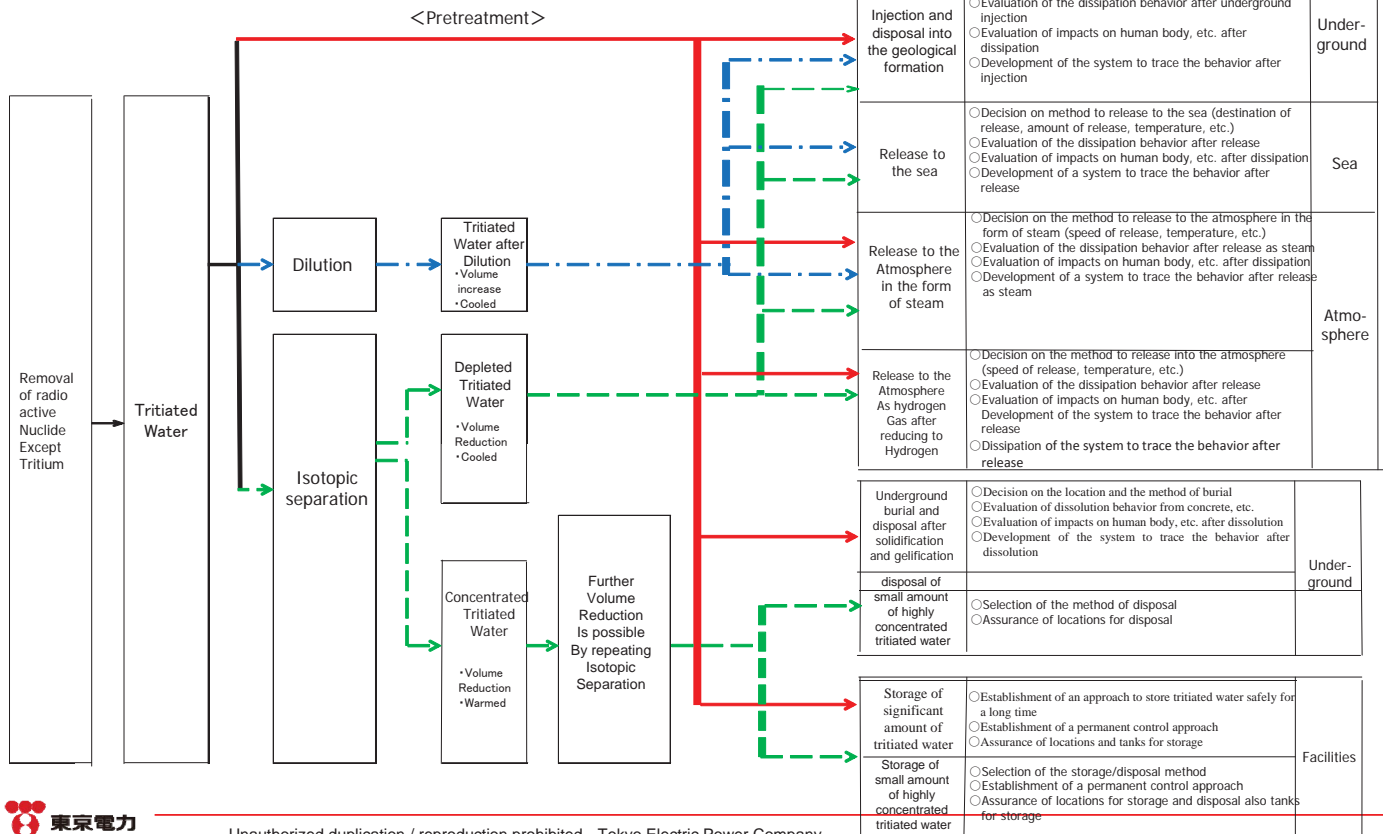
- ✓ The method using the isotopic chemistry balance shift of a high temperature state and a cold condition.
- ✓ It is a thing aiming at heavy water manufacture, and when applying to tritium, it has a difficulty in control and the operativity of concentration.

3. Tritium Status: Parallel of remover concentration

Although some are remove to an environmental level (several Bq/Liter) theoretically, when performing processing on an industrial scale, equipment and consumption energy become huge.

Only a thing deeper than concentration of Fukushima Daiichi can remove by the remove equipment put in practical use on a scale of industry. Remove can't at comparatively low concentration of Fukushima Daiichi.





Demonstration Project Verification Tests of Tritium Separation Technologies

27

Request for Proposal for entities to implement with subsidies the "Verification of technologies for contaminated water management (Demonstration Project for Verification Tests of Tritium Separation Technologies)" project in the FY2013 Supplementary Budget (Updated on June 2)

Mitsubishi Research Institute, Inc.
MRI News
June 2, 2014

The Mitsubishi Research Institute, Inc., having been selected by the Agency for Natural Resources and Energy (ANRE, METI, Japan), is now executing the tasks of the Project Management Office for the "Project of Decommissioning and Contaminated Water Management". In this project, based on the "Grant Policy for Subsidy for the Project of Decommissioning and Contaminated Water Management", we are assisting activities in support of the development of technologies that will be useful in decommissioning and contaminated water management. Through this, we aim to improve the level of science and technology and to advance decommissioning and contaminated water management smoothly in Japan.

At this time we are proceeding with the RFP (Request for Proposal) for entities to receive subsidies and implement the "Verification of technologies for contaminated water management (Demonstration Project for Verification Tests of Tritium Separation Technologies)" as the fifth RFP. If you are interested in responding to this RFP, please take into consideration the information provided in this document as well as the detailed information provided in the separate document "Guidelines for applying to the "Verification of technologies for contaminated water management project" in the FY2013 Supplementary Budget".

Duration of the project : From the date of the grant decision to March 31st, 2016

Separation performance of tritium : Separation factor higher than 100

Adoption entrepreneur (August 26, 2014)	Separation method
Kurion, Inc. 【USA】	Combined Electrolysis Catalytic Exchange (CECE)
GE Hitachi Nuclear Energy Canada Inc. 【Canada】	Water Distillation
Federal State Unitary Enterprise "Radioactive Waste Management Enterprise "RosRAD"" 【Russia】	Combination of CECE and Water Distillation

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}U and ^{239}Pu during “burning (fission)”. The amount is 0.013% of ^{235}U and 0.023% of ^{239}Pu , and ^{239}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.

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

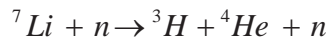
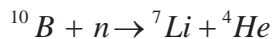
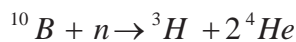
$$\text{NF: No. of fissions per 1MWt in 1 second} = 3.15 \times 10^{16} / \text{s} \cdot \text{MWt}$$

- 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 $\sim 10^{-4}$).

Amount of tritium produced	500,000kWe	800,000kWe	1,100,000kWe
Bq/month	4.07E+13	6.29E+13	8.51E+13

② Production by neutron irradiation of boron-10 contained in boron carbide control rods

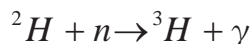
- Tritium is produced by the following reaction inside the poison rods of control rods using B_4C (boron carbide) for neutron absorption



- In 1 g of B_4C irradiated until ^{10}B decreases approximately 50%, a conservative assessment puts the amount of tritium produced at $1.48 \times 10^9 \text{ Bq/gB}_4\text{C}$, 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.



$$N^3\text{H} = \Sigma(D_2\text{O}) \times \phi \times V \times \rho \times (D/H) \times t$$

$N^3\text{H}$: Amt. of tritium produced V : Vol. of reactor water
 $\Sigma D_2\text{O}$: Reaction cross section of heavy water ρ : Density correction for water
 ϕ : Thermal neutron flux D/H : Heavy water abundance ratio

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

Amount of tritium produced	500,000kWe	800,000kWe	1,100,000kWe
Bq/month	4.44E+09	6.29E+09	1.07E+10

- 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.

(Bq)

	Unit 1	Unit 2	Unit 3	Total
TEPCO	1.0E+15	1.2E+15	1.2E+15	3.4E+15
<Ref> JAEA*	9.4E+14	1.2E+15	1.2E+15	3.3E+15

• Atomic Energy Society of Japan, "Radionuclide Release to Stagnant Water in Fukushima-1 Nuclear Power Plant" (Kenji NISHIHARA et al.) (in Japanese)

- 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 8.34×10^{14} Bq. By the end of February 2016, this amount is estimated to be approximately 9×10^{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 6.7×10^4 Bq/liter.
- 3.4×10^{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.)

※ 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.