Treatment of contaminated water stored in Fukushima Dai-ichi Nuclear Power Plant

As a result of the delay in primary containment vessel (PCV) restoration, injected water for cooling of reactor pressure vessels (RPVs) of No.1 through 3 Units must still be recirculated through large areas from the RPVs to the turbine buildings (T/Bs) and approximately 100,000 ton of recirculated water should be cleaned up by removing radioactivity and salt (**Figure 1**). Additionally, underground water leaking into the recirculation water has resulted in an increasing total amount of contaminated water stored in the site, which might possibly be released to the environment accidentally. Total inventory of the contaminated water is reaching about 500,000 ton (Aug. 2013). The latest subjects related to the contaminated water are as follows;

(1) Suppression of increase in the inventory of the contaminated water

Any intrusion should be sealed to prepare for a smaller recirculation restricted only in PCVs but the leakage locations are not detected yet. Mixing of the underground water into the contaminated water is going to be reduced by bypassing flow and by its shielding with frozen water walls or water glass walls.

(2) Control of its leakage to the environment

Underground water should be isolated by frozen water walls or water glass walls to prevent contamination. The leaked water should be pumped and then radioactive materials in the water should be removed.

(3) Long-term storage of surplus contaminated water

Long-term integrity of the storage tanks should be monitored. Radioactive nuclei except tritium should be removed by multi-nuclei removal facility, ALPS.

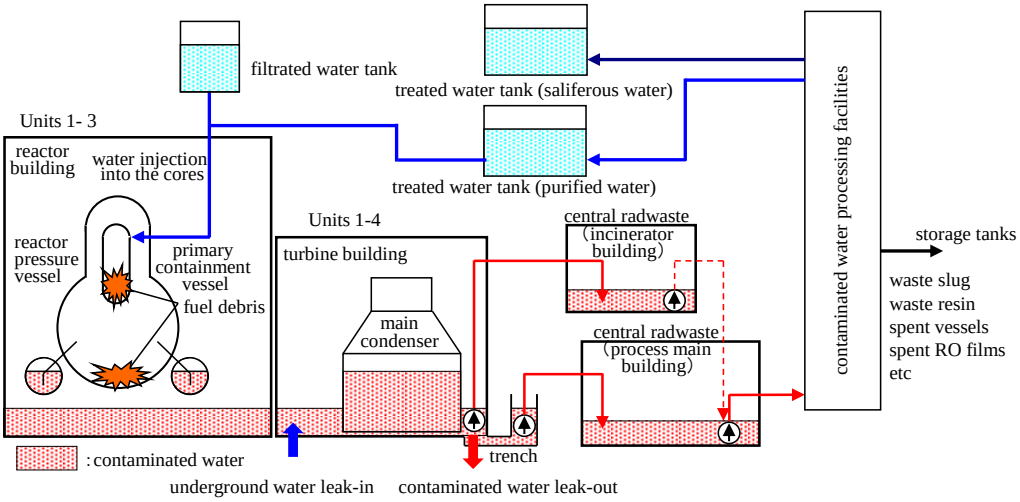

Figure 1 Recirculation system of cooling water in Fukushima Daiichi NPP and its treatment system

As a result of continuous operation of waste water treatment system, ^{137}Cs radioactivity in the contaminated water has been reduced and then it has leveled off at about 20,000 Bq/g (**Figure 2** a)). Tritium is not removed by the treatment system but it is diluted by the mixing with underground water, its radioactivity has also reduced and leveled off at around 1,000 Bq/g (Figure 2 b)).

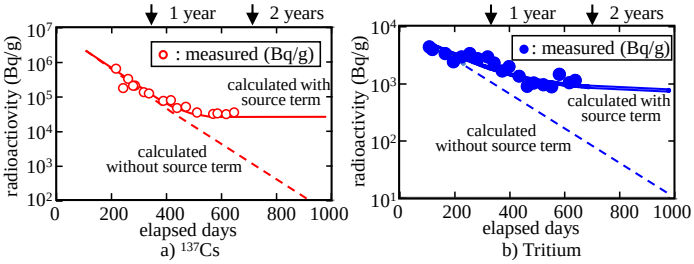

Figure 2 Cl^- ion and radioactivity in the storage water

In 2013, multi-nuclei removal facility, ALPS, with average decontamination factor, DF, of 600,000 has started operation to reduce the present radioactivity level of the contaminated water to lower than the permissible

level for release.

Tritium is one of the naturally abundant radioisotopes, which is generated by nuclear reactions of air with cosmic rays, e.g., ^{14}N (n,^{3}H) ^{12}C, and its natural background is around 0.01 Bq/g in water. Its half-life time is 12.3 years, while its biological half-life time is only 12 days.

Table 1 Major features of tritium

Half-life time:	12.3 years
Biological half-life time:	12 days (small effects of concentration in living bodies)
Major reaction for tritium generation:	^{14}N (n,^{3}H) ^{12}C
Generation rate on the earth:	1 EBq/y (10^{18} Bq/y)
Natural background:	0.01 Bq/g

In BWR, tritium is generated mainly as a consequence of three bodies nuclear fission reaction of ^{235}U (probability of three bodies fission: 10^{-4} of total fission reactions). Tritium inventory in the fuel can be calculated by applying total burn-up of the fuel (total fission of the fuel). At present time total tritium inventory in the contaminated water is estimated to be a third of the total generation amounts in the three reactors. This tritium will be released continuously into the water for a long period.

Tritium is one of the hydrogen isotopes, which can be removed only by isotopic separation methods but not by co-precipitation methods and ion exchange methods. Multi-nuclei removal facility, ALPS, can be applied to reduce radioactivity of major radioactive species, e.g., fission products and corrosion products, to below the permissible level for release. But the decontamination factor of tritium by industrial scale of isotopic separation methods is expected to be about 10, which means difficulty of application of the isotopic separation methods for tritium removal. Latest possible options for tritium treatment applied at Fukushima Dai-ichi NPP are listed in **Table 2**.

Table 2 Tritium treatment - Contingency plans

Procedures	Outline	Subjects	Reliability for application	Risks for environment
1. Stored in the site	storage without release	possible contamination of underground water	high	high
2. Tritium removal or concentration	isotopic exchanger	engineering difficulty [reasonable DF: <10]		
2.1 release of diluted water	accompanied by dilution process		low	low
2.2 storage of highly ^{3}H enriched water	decrease of tritium inventory in the release water		low	high
3. Dilute and release	remove of other nuclei	agreement with		
3.1 release into the ocean	^{3}H is diluted for release in the ocean	local community	high	low
3.2 release into the air	tritiated water is evaporated to release ^{3}H in the air (same as TMI)	neighboring contamination due to rain	high	middle

*1 Reactor Regulation (Operational safety provision) for previous Fukushima Dai-ichi NPP: concentration; <20 Bq/g

It is possible to release tritium at considerably lower than the permissible level but the latest highly sensitive monitoring device such as liquid scintillator can detect the natural BG level tritium. When considering public acceptance of radioactivity release and rumors of radioactive contamination, it is important to explain carefully and circumspectly not only to people living in the local areas but also to neighbor countries that release of diluted tritium cannot be concentrated in living bodies in the food chain and then it is expected to reduce its radioactivity to the natural BG level rapidly by isotopic dilution.

The committee concludes that the most realistic approaches are as follows;

1) to decontaminate the surplus contaminated water stored in the Fukushima Dai-ich NPP with the multi-nuclei removal facility, ALPS,
2) to dilute the residual tritium in the water with large amount of sea water to much lower than the permissible level, and
3) to release it into the ocean with continuous monitoring for reaching the natural BG level.

This is considered to minimize the risks of radiation exposures and environmental contamination due to accidental radioactivity leakage. [Division of Water Chemistry, Fusion Engineering Division]