Monitoring of Radioactive Gaseous and Liquid Wastes at Rokkasho Reprocessing Plant

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ABSTRACT:
Rokkasho Reprocessing Plant (RRP) started its active test with spent fuels at the end of March 2006. In order to limit the public dose as low as reasonably achievable, RRP removes radioactive materials, and then releases them to atmosphere and to sea. Concerning the radioactive gaseous and liquid wastes to be released, the target values of the amount of radioactive materials to be released annually have been defined. RRP controls the amount of radioactive materials not to exceed these target values by monitoring those in exhaust and effluent.

From the start-up of active test up to June 26th, an amount of about 30 Upr fuel assemblies has been treated. The monitoring results of $^{85}$Kr release have faithfully reflected operation patterns of shearing and dissolution and have been in accordance with the calculated results on the basis of burn-up, cooling period and so on.

KEYWORDS: radioactive waste, monitoring, krypton-85

1. Introduction

RRP is a commercial reprocessing facility of spent fuels built at Rokkasho-mura, Aomori Prefecture. Purex method is adopted to RRP and its annual reprocessing quantity is 800 Upr in maximum, while its daily maximum reprocessing quantity is 4.8 Upr. The average burn-up of spent fuel to be reprocessed per day is below 45,000 MWd/tUpr.

RRP started active test with spent fuels at the end of March 2006. The active test consist of 5 steps and Step 1 ended on 26th June, 2006. When spent fuel assemblies are sheared and dissolved, radioactive gaseous waste and radioactive liquid waste such as $^{85}$Kr and $^3$H are released to atmosphere and to sea.

In order to limit the public dose as low as reasonably achievable, RRP removes radioactive materials by evaporation, rinsing, filtering, etc, and then releases them through the main stack and the maritime release pipe that make dispersion and dilution efficiently.

Also, concerning radioactive gaseous and liquid waste to be released to the environment, the target values of annual release have been defined in the Safety Rules of RRP based on the annual release evaluated at the safety review of RRP. By monitoring the radioactive nuclides in gaseous and liquid wastes RRP controls them not to exceed the target values.

2. Outline of Radioactive Gaseous and Liquid Waste Treatment

2.1 Radioactive materials removal equipment

It is our basic policy to process properly radioactive gaseous and liquid wastes to be released from RRP to atmosphere and to sea, and to reduce their concentration.
and quantity as low as reasonably achievable. RRP is designed for radioactive materials in gaseous waste to be removed with HEPA filters and iodine filters, etc., as much as possible. RRP is also designed for radioactive materials in liquid waste to be removed by evaporation, filtering, etc., as much as possible, according to the nuclides and property of liquid waste.

2.2 Release of radioactive gaseous and liquid waste

Radioactive gaseous waste is released to atmosphere from the main stack (150m high from the ground level and its ventilation volume is more than 1,500,000m³/h) with sufficient dispersion and dilution effects. While radioactive gaseous waste is released, radioactive nuclides concentration in the exhaust gas is continuously monitored.

Radioactive liquid waste is released to sea through the maritime release pipe (3km offshore and 44m in depth) with sufficient dispersion and dilution effects, after radioactive concentration and amount are checked at a storage vessel in every release. While radioactive liquid waste is released, the effluent is monitored on the pipe after the storage vessel to ensure that no unexpected release is occurred.

2.3 Dose evaluation for members of the public in the surrounding areas of RRP

The result of dose evaluation for members of the public from radioactive gaseous and liquid wastes released by RRP shows a dose of about 1.9x10⁻²mSv/y for radioactive gaseous waste, a dose of about 3.2x10⁻³mSv/y for radioactive liquid waste and a total of about 2.2x10⁻²mSv/y, which means that the dose is controlled as low as reasonably achievable.

3. Monitoring of Radioactive Gaseous and Liquid Wastes

3.1 Radioactive nuclides subject to monitoring and target values for release control

To maintain the public dose at very low level as mentioned in the above 2.3, the target values for release control are determined according to the estimated annual release quantities which are the bases of the evaluation of the public dose. By limiting wastes below the target values, it is possible to maintain the public dose very low. Table 1 indicates the target values for radioactive gaseous and liquid wastes.

At RRP, the releases of radioactive nuclides in exhaust and effluents listed in Table 1 are controlled during active test as well as the commercial operation. In addition, soil, sea water, agricultural products, dairy products, fish and shellfish, etc. in the environment are sampled and monitored together with the environmental dose rate around RRP in order to verify that there is no impact from wastes to the environment.

<table>
<thead>
<tr>
<th>Radioactive nuclides in gaseous waste (Bq/year)</th>
<th>Radioactive nuclides in liquid waste (Bq/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>^85Kr</td>
<td>3.3x10⁻⁷</td>
</tr>
<tr>
<td>^3H</td>
<td>1.9x10⁻¹⁵</td>
</tr>
<tr>
<td>^14C</td>
<td>5.2x10⁻¹³</td>
</tr>
<tr>
<td>^129I</td>
<td>1.1x10⁻¹⁰</td>
</tr>
<tr>
<td>^131I</td>
<td>1.7x10⁻¹⁰</td>
</tr>
<tr>
<td>Total alpha</td>
<td>3.5x10⁸</td>
</tr>
<tr>
<td>Total beta (gamma)</td>
<td>9.4x10⁻¹⁰</td>
</tr>
<tr>
<td>^3H</td>
<td>1.8x10⁻¹⁶</td>
</tr>
<tr>
<td>^129I</td>
<td>4.3x10⁻¹⁰</td>
</tr>
<tr>
<td>^131I</td>
<td>1.7x10⁻¹¹</td>
</tr>
<tr>
<td>Total alpha</td>
<td>3.8x10⁹</td>
</tr>
<tr>
<td>Total beta (gamma)</td>
<td>2.1x10⁻¹¹</td>
</tr>
</tbody>
</table>

3.2 Monitoring equipment for radioactive gaseous wastes

The composition of monitoring equipment for the main stack is shown in Fig. 2. The monitoring of radioactive gaseous waste released from the main stack is carried out with on-line monitors (gas monitors, dust monitors, iodine monitors) and sampling equipment of ^3H, ^14C, gaseous ^10⁶Ru, iodine and aerosols. There are two sets of on-line monitors and sampling equipment.
The measurement values of on-line monitors are monitored at the control room and are stored in the electronic data system at the same time, so that it is possible to access the monitoring data at the terminal. The on-line monitors have their alarm to detect an unexpected release. The samples taken by sampling equipment are periodically analysed.

$^{85}$Kr is continuously measured by the gas monitors. The gas monitors that are made to introduce sampling gas into each chamber measure $^{85}$Kr with plastic scintillation detectors. In routine monitoring, two types of monitors are used for a wider range of measurement: low range and middle range monitors. A high range monitor is equipped with an ionization chamber detector for the case of incident.

Iodine is collected in a charcoal cartridge in the iodine sampler. The charcoal cartridge is periodically recovered and $^{129}$I and $^{131}$I are measured with gamma spectrometer.

For $^3$H, hydrogen gas and methane gas are oxidized with oxidation furnace in the tritium sampler, then water including $^3$H is frozen and trapped with cooling trap. The collected water is periodically collected and $^3$H in the water is measured with liquid scintillation counter.

For $^{14}$C, carbon monoxide and methane gas are oxidized with oxidation furnace in the carbon sampler, then $^{14}$C is trapped by organic solvent in the form of CO$_2$. The organic solvent is periodically recovered and $^{14}$C is measured with liquid scintillation counter.

Materials in the form of aerosols are collected on a paper filter in the dust sampler. The samples are periodically recovered and $^{106}$Ru/$^{106}$Rh, $^{137}$Cs/$^{137}$mBa are measured with total alpha detector, total beta detector and gamma spectrometer. Moreover, Pu (alpha) and $^{90}$Sr/$^{90}$Y are measured by composite samples in a month or 3 month (in principle).

Furthermore, the iodine monitor and the dust monitor are installed for the purpose of grasping the release status. In order to avoid the obstruction of $^{85}$Kr gas during shearing and dissolving of spent fuels, the sampling part and the detecting part are separated from each other and the charcoal cartridge or the paper filter paper cartridge for sampling can be moved around by a turn table: cf.: Fig.3.
4. Release Control Results

4.1 Radioactive release quantities during Step 1 of the active test

The radioactive quantities released during Step 1 (From 31st March through 26th June, 2006) of active test are shown in Table 2. 67 assemblies of about 30tUpr of PWR (17x17) spent fuels have been reprocessed during this period. $^3$H, $^{14}$C, $^{85}$Kr and $^{129}$I have been detected as radioactive gaseous wastes, while $^3$H has been detected as radioactive liquid waste. They were all much below the target values. The nuclides that are subject to control, but not listed in the Table 2 were below the detection lower limits.

Table 2 Radioactive release quantities during Step 1 of the active test

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Release qty (A)[Bq]</th>
<th>Release est* (B)[Bq]</th>
<th>$(A)/(B)\times100$ [%]</th>
<th>Target value [Bq/year]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radioactive gaseous waste</td>
<td>$^{85}$Kr</td>
<td>$2.9\times10^{15}$</td>
<td>$2.9\times10^{15}$</td>
<td>100</td>
</tr>
<tr>
<td></td>
<td>$^3$H</td>
<td>$1.3\times10^{12}$</td>
<td>$1.8\times10^{13}$</td>
<td>6.9</td>
</tr>
<tr>
<td></td>
<td>$^{14}$C</td>
<td>$1.9\times10^{11}$</td>
<td>$1.3\times10^{12}$</td>
<td>14</td>
</tr>
<tr>
<td></td>
<td>$^{129}$I</td>
<td>$3.9\times10^{7}$</td>
<td>$2.3\times10^{8}$</td>
<td>17</td>
</tr>
<tr>
<td>Radioactive liquid waste</td>
<td>$^3$H</td>
<td>$1.3\times10^{13}$</td>
<td>$1.7\times10^{14}$</td>
<td>7.7</td>
</tr>
</tbody>
</table>

*The release estimate is calculated using ORIGEN 2 and the evaluated DFs at the safety review, etc.

The released quantity of $^{85}$Kr from the main stack measured with gas monitor has almost corresponded to the estimated quantity of $^{85}$Kr, which was calculated by ORIGEN 2 based on the specification of burn-up, etc., of the spent fuels that has been actually reprocessed during Step 1. $^3$H, $^{14}$C and $^{129}$I in exhaust and effluent were much below the expected quantities.

4.2 Release of $^{85}$Kr during shearing and dissolution of spent fuels

The release of $^{85}$Kr that has been monitored during shearing and dissolution of PWR spent fuels is shown in Figure 4. The dissolver of spent fuels has a rotary wheel with 12 buckets. Sheared spent fuels drop into the bucket and are dissolved by nitric acid solution. One third of an assembly of PWR spent fuels can be dissolved per one bucket, so that one assembly is sheared and dissolved in 3 segments. The concentration of $^{85}$Kr in exhaust varies according to shearing and dissolution stage.

Figure 4 Release of $^{85}$Kr during shearing and dissolution

5. Conclusion

Since 31st March 2006 active test have been carried out at RRP. The radioactive gaseous waste and liquid wastes have properly been processed as initially designed and the wastes have been released to atmosphere and to sea under the proper release control.