The Effort for the nuclear material recovery from the reprocessing process and relevant MC&A in the TRP Decommissioning Project

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ABSTRACT

Tokai Reprocessing Plant (TRP) had reprocessed 1,140 tons spent fuel (SF) since 1977 to 2007, and the Post-Fukushima safety regulations triggered TRP to decide to shut the plant and shifted to decommissioning phase from 2018. The estimated entire duration of TRP decommissioning is expected to be 70 years.

The early stage of the TRP decommissioning includes the recovery of remaining nuclear material in the process and transferring those nuclear material between multiple facilities(“Flush-out”), vitrification activity of remaining high active liquid waste (HALW) and transferring collected nuclear material to the storage facility, and the management of radioactive waste.

The Flush-out is as the first step of TRP decommissioning and it is in progress since June 2022 for two years prior to the start of serious decommissioning activities such as the decontamination, dismantling/removing the process. Since these decommissioning activities are unexperienced challenges and they are not related to “spent fuel reprocessing”, therefore ensuring appropriate nuclear material control and accountancy (MC&A) and the transparency in terms of safeguards verification implemented by International Atomic Energy Agency (IAEA) under ensuring the safety measure are important role as the operator.

In this paper, we report about the residual nuclear material recovery process and relevant MC&A and safeguards measures in the Flush-out as the first stage of TRP decommissioning project.

1. Outline of TRP and verification activity during plant operation

1.1 TRP：Tokai reprocessing plant

Tokai reprocessing plant (TRP) had started the operation from 1977 as Japanese first reprocessing plant, total 1,140 tons of spent fuel (SF) from commercial nuclear power reactors reprocessed until 2007. TRP had contributed to the nuclear fuel cycle in Japan by separation of Uranium and Plutonium from spent fuel to recover them by solvent extraction of Plutonium Uranium Reduction Extraction (PUREX) method, making trioxide uranium oxide (UO_3) powder product and conversion of Uranyl nitrate (UNH) solution and Plutonium nitrate $Pu(NO_3)_4$ solution to MOX powder by direct microwave de-nitration method.

1.2 Material Control and Accountancy, and Safeguards for TRP during plant operation

Ensuring the transparency for management of nuclear material, TRP has kept suitable Material Control and Accountancy (MC&A) and received Safeguards measure consisted by Nuclear

Regulation Authority in Japan (NRA) and IAEA. The Material Balance Area (MBA) of TRP is consisted of three MBAs, which are batch-follow area (MBA1) from SF receipt and storage to input accountability tank (IAT), bulk handling area (MBA2) of main chemical process, and bulk area (MBA3) for product storage of $Pu(NO_3)_4$ solution and UO_3 powder.

TRP has conducted physical inventory taking (PIT) within 14 months after previous PIT, to confirm the amount of nuclear material stored in the facility and evaluated Shipment/Receipt Difference (SRD) between the nuclear power reactor and MBA1, and Material Unaccounted For (MUF) of the inventory change regarding MBA2 and MBA3. TRP has set up the several sub-MBAs and evaluated the inventory change for the transfer between sub-MBAs in order to investigate the cause of MUF. Regarding the MC&A in MBA1, SF receipted from other facility was sheared and dissolved, the dissolved solution was gathered into IAT. The amount of nuclear material in dissolved solution for the inventory change from MBA1 to MBA2 was determined by destructive analysis (DA) at IAT. The DA treatment for inventory change between MBA2 and MBA3 was also performed for $Pu(NO_3)_4$ product solution, and UO_3 powder product at MBA2 before storing product material at MBA3. At the timing of the transfer of $Pu(NO_3)_4$ product solution from MBA3 to Plutonium Conversion Development Facility (PCDF) which is another MBA, the inventory change was reported. High Active Liquid Waste (HALW) which is separated waste solution from extraction process in MBA2 has been transferred to storage area for retained waste (RW) after the evaporated concentration and DA for reporting the inventory change. The HALW transferred as RW is solidified into vitrified waste for stabilization at Tokai Vitrification Facility (TVF). This timing, HALW is transferred from RW area to TVF in MBA2, so HALW in TVF is back to the inventory of MBA2. The vitrified waste including HALW is stored as measured discards at TVF. The inventory change report (ICR) and interim inventory of Plutonium in main tanks in TRP has been periodically declared to NRA in Japan and IAEA.

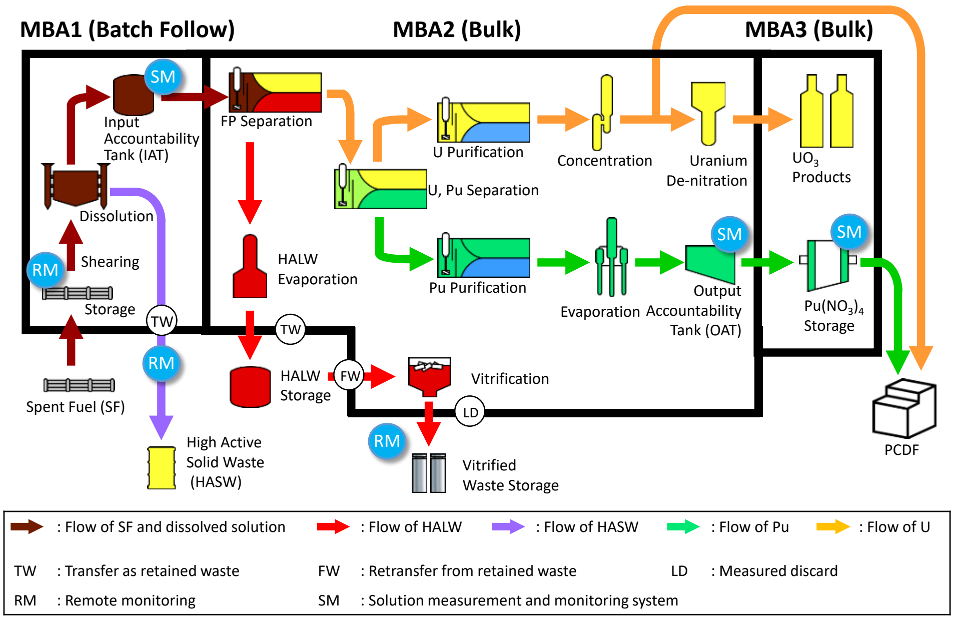

Figure 1 The main flow of nuclear material in operating TRP.

During the reprocessing operation in TRP, Safeguards measure by NRA and IAEA has been subjected to the inventory change between MBAs, inventory declaration and operational status in TRP. Interim inventory verification on fixed day had been implemented to in-operating TRP [1]. Since IAEA concluded "Broader Conclusion" for Japan in June 2004, introduction of integrated safeguards was started in September 2004, so the frequency of safeguards measures to direct use material has been changed to be random basis. Main tanks and storage area in TRP are under the containment/surveillance(C/S) monitoring of NRA and IAEA such as Solution Measurement and Monitoring System (SMMS). TRP provides the information about the operation in the storage area under the C/S monitoring or the volume change in tank through SMMS.

2. TRP decommissioning project [2]

TRP suspended the reprocessing operation in 2007 to start construction work for earthquake resistance improvement of the facilities based on regulatory standards considering Niigata-ken Chuetsu-oki Earthquake. TRP had planned the re-start of reprocessing operation after that construction work. However, TRP decided to shut the plant and proceed the decommissioning phase in 2014 due to the enforcement of the Post-Fukushima safety regulations in 2013, the status of TRP has been shifted to decommissioning phase applying "TRP decommissioning project" to NRA in Japan. For the decommissioning of reprocessing plant, a considerable period of long-term effort is estimated for the decontamination prior to dismantle facility building since variable radioisotope materials are widely remained or adhered in the equipment and pipelines of the process. On the other hand, additional construction work to the facilities in TRP is required in order to ensure the new safety regulation and proceed the decommissioning safely. From the viewpoint of above condition, reducing the risk by radioactive waste is as the top priority in TRP

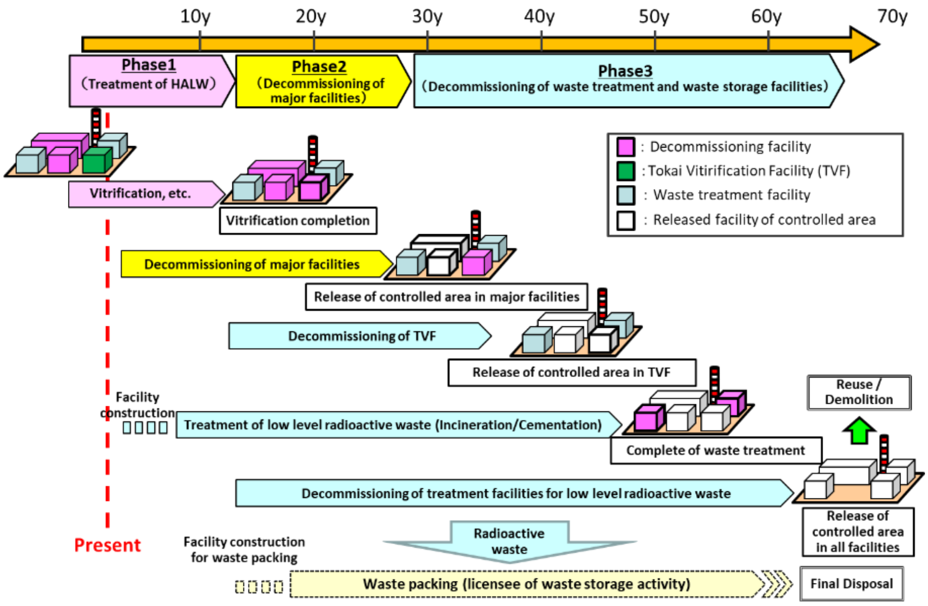

Figure 2 Load map for decommissioning of TRP.

decommissioning project. Therefore, ensuring safety through earthquake-resistant construction of HALW storage facility, and vitrification of HALW at TVF are conducted at first, improvement of storage condition of High Active Solid Waste storage (HASWS) and solidification of Low Active Liquid Waste (LALW) at the Low-level radioactive Waste Treatment Facility (LWTF) will be proceeded in order. Furthermore, removal and clean-up of the nuclear material which is remained in the main facilities since the suspension of reprocessing operation in 2007 (Flush-out plan for decommissioning) and shipment of unprocessed SF to the oversea will be performed in parallel with the above construction work, treatment of waste items, de-contamination and dismantling of TRP will be implemented in the future. The duration for the completion of TRP decommissioning project is estimated to be 70 years. TRP currently proceeds the safety measures of the facilities, vitrification of HALW and Flush-out plan based on the decommissioning plan.

2.2 Flush-out plan for decommissioning

After TRP suspended its operation since 2007, the nuclear materials have been remained in main process, which are sheared powder generated from chopped spent fuel assemblies remained around the shearing machine during the past operation, purified $Pu(NO_3)_4$ solution remained after the rising of Plutonium product storage tanks (Pu storage tanks) at the timing of last MOX powder production campaign, purified UNH solution remained from the last reprocessing operation and so on. Those nuclear materials should be removed from the main process before the start of decontamination and dismantling of main facilities, so TRP planned the Flush-out to collect the recoverable nuclear materials and transfer to out of main process.

TRP decommissioning project describes that Flush out plan is graded as the top priority with early and reliable implementation in terms of safety. TRP proceeds Flush-out plan with following policy considering the safety measure for aging management; a) implementation without "Reprocessing operation"(extraction is not performed), b) use limited process and equipment, c) no new equipment will be installed/modified, d) maintain the safety considering the influence of

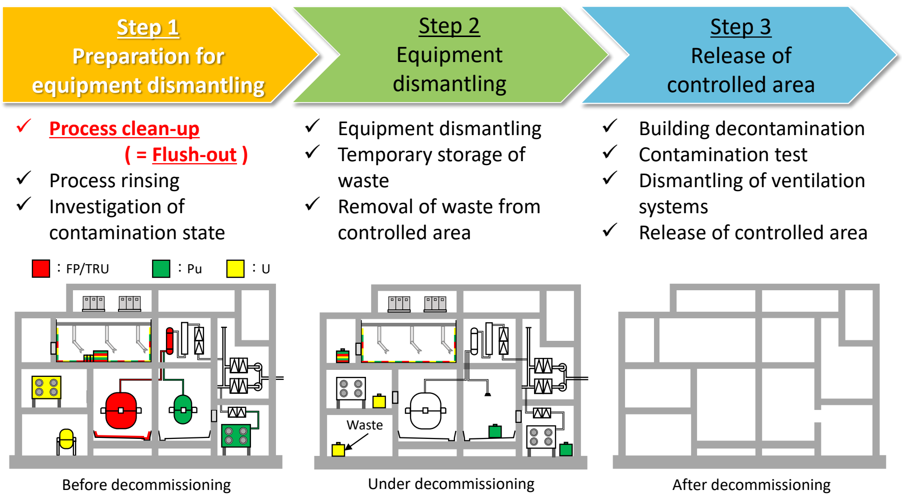

Figure 3 Step for the dismantle of main facilities.

nuclear criticality by wrong operation during the solution transfer.

During the Flush-out, the activities for removal of recoverable nuclear material are performed as Stage A: Dissolution of the shared powder and transfer to HALWS as retained waste, Stage B: Collection of $Pu(NO_3)_4$ solution and transfer to HALWS as retained waste, and Stage C: Storing UO_3 powder in stepwise, not simultaneously.

2.2.1 Stage A: Dissolution of shared powder and transfer to HALWS as retained waste

The sheared powder which was generated during shearing process of SF was remained around shearing machine and distributors [3]. Since the composition of sheared powder is basically same as SF, the amount of nuclear material in sheared powder can be determined after dissolution in the dissolver. The determination of transferred volume and sampling for ICR is conducted at IAT (see Figure 1). After the inventory change from MBA1 to MBA2, the dissolved solution is transferred to HALW storage as RW via extractor and evaporator without their operation. The used equipment for the transfer of dissolved solution of sheared powder is rinsed by nitric acid and water to achieve the practically lower concentration which is the goal of Stage A. The analysis method during Stage A is changed to other appropriate technique as concentration decreases by rinsing.

2.2.2 Stage B: Collection of $Pu(NO_3)_4$ solution and transfer as retained waste

TRP had the purified $Pu(NO_3)_4$ solution in Pu storage tanks in MBA3 at main plant since the suspension of the reprocessing operation, TRP and PCDF conducted the campaign to fabricate MOX powder for the stabilization by 2016. Since the last TRP campaign, the amount of the heel level of $Pu(NO_3)_4$solution has been remained in Pu storage tanks, therefore, the subject of Stage B is to transfer those low concentration $Pu(NO_3)_4$ solution in MBA3 to HALW storage tanks as RW via MBA2 instead of the originally designed material flow which is the conversion to MOX powder securing safety at an early stage of decommissioning plan. After the transfer of $Pu(NO_3)_4$ solution, rinsing of equipment used for the transfer and Pu purified process is performed by nitric acid and water to achieve the target concentration similar to the Stage A, rinsing solution is transferred to RW area treated as a part of HALW. Therefore, during the solution transfer in Stage B, a part of the material flow was reversed from that of originally designed. Even under those circumstances, it is required to perform the suitable accounting measures and maintain the transparency for safeguards, the new accountancy tank was applied on the material flow of the solution transfer during Stage B, after the technical discussion between TRP, NRA and IAEA.

2.2.3 Stage C: Storing the UO_3 powder

There is a lot of purified UNH solution in main process of TRP after the last reprocessing operation. Since UNH solution (Depleted Uranium) has a low safety risk compared to Plutonium due to the low calorific value and specific radioactivity, the stabilization of the remained UNH solution to convert to UO_3 powder by de-nitration and stored in TRP will be implemented applying originally designed reprocessing operation. The generated solution including uranium by rinsing during Stage C is also converted to UO_3 product powder. Those solution with low uranium concentration that is not suitable for fabricating UO_3 powder, is transferred to RW area as LALW (Low Active Liquid Waste). Since the flow of transfer of nuclear material during Stage C is almost same as usual operation, TRP plans to proceed that activity applying the conventional accounting and safeguards measure along with ensuring current safeguards requirement of IAEA.

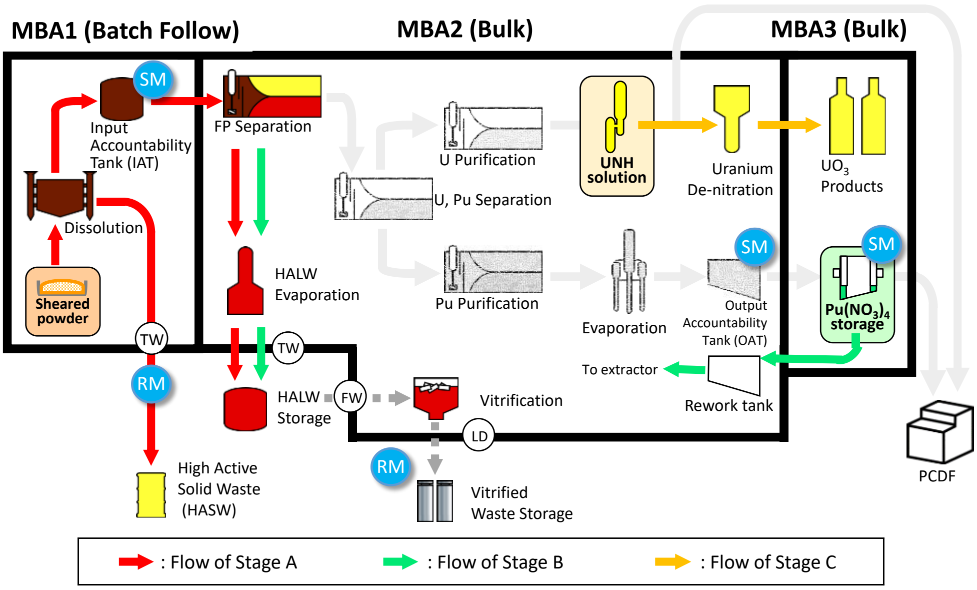

Figure 4 The flow diagram for recoverable nuclear material.

2.2.4 Evaluation of S/RD and MUF

TRP plans to evaluate a material balance of S/RD and MUF to the all of inventory change between MBAs for Flush-out. When the implementation of decommissioning including Flush-out, using the different flow from the usual operation, replacement of storage area of nuclear materials, and the limitation of used facilities will be occurred. It is difficult that the application of the regular accounting and safeguards measurement across into the different transfer of nuclear material from usual operation, TRP must explain the activity about the process flow and accounting method for the facility side to NRA and IAEA, implement the Flush-out plan with their comprehension. The duration and fluent communication are necessary for the enough consideration of appropriate safeguard measure by NRA and IAEA based on the information from the facility. Although decommissioning activities are different from reprocessing operations, these activities are paid attention as equivalent activities in terms of safeguards, so it is important to keep to reliably prove that nuclear materials are under the surveillance of the NRA and IAEA and that there is no concern of diversion. Based on the above items, TRP certainly implements following items, reliable observance of agreements, provision of decommissioning plans and related information, assessment of influence on safeguards activities, response to issues to be discussed (early consultation, rational proposals, implementation of technology development), realization of achievement and harmonization of requirements related to 3S (nuclear safety, nuclear security, safeguards)[4].

3. Nuclear material accountancy report regarding flush-out activities

At present, Stage A was implemented from June 2022 to September 2022 as TRP scheduled. The activity of Stage B and related accounting and safeguards measures based on the result of discussion in advance with NRA and IAEA via the annual or technical meeting, are progressing from March 2023 for about four months. Prior to the start of flush out, the vessel calibration checks to confirm the uncertainty within the criteria was performed for the accountability tanks for Stage A, and newly assigned accountability tank for Stage B, and the result was shared with NRA and IAEA. The end-of-day inventory of tanks are confirmed on the flow of solution transfer based on the latest analysis result and calculation value during each stage of the Flush-out activity. TRP declared the interim inventory of additional tanks voluntarily as well as subject tanks. The interim MUF of MBA2 during Stage A was evaluated based on the inventory change report and interim inventory, and they informed the result to NRA and IAEA every month.

TRP investigated the evaluation method of interim inventory. Since TRP was operated 24 hours a day in the past reprocessing operation, and the solution flowed constantly and homogeneously in the process, so it was relatively easy to estimate the inventory in the process. However, in the case of Stage A, a concentration gradient was expected to occur between upstream and downstream in the process because of the transfer of dissolved solution into the process with nitric acid at the very early of Stage A activity as well as the transfer of rinsing solution into the process with dissolved solution of sheared powder at the end of stage A. This concentration gradient between upstream side and downstream side will make the difference of the estimated value and actual value of solution concentration for inventory management. So the application of conventional evaluation method for a homogeneous state which might cause the irrelevant material control. Therefore, TRP set additional sampling point within the interim inventory that optimized material balance evaluation. Similar phenomenon is expected to be observed as decommissioning progresses after flush out such as process rinsing for decontamination, and optimization of the evaluation of material balance according to the situation is required.

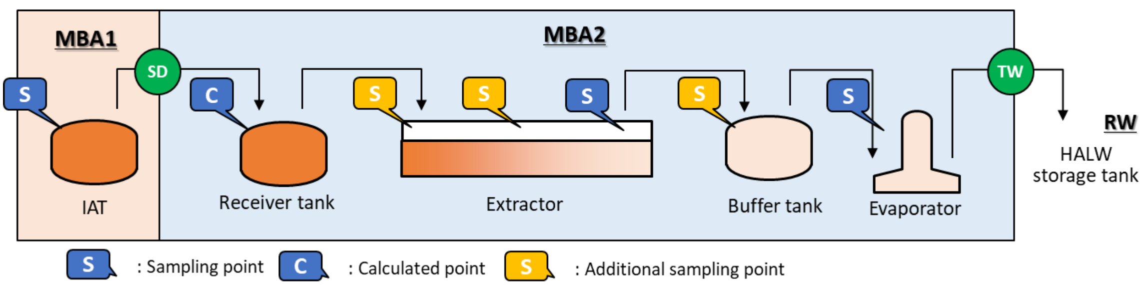

Figure 5 Optimization of evaluation method of interim inventory.

In addition, TRP reported the cumulative S/RD and cumulative MUF from the start of reprocessing operation at TRP because Flush-out plan is the large-scale inventory change since the suspension of the operation, and rinsing which might cause the washing out of remained nuclear material is performed. As the result of MC&A during Stage A, the amount of nuclear material of sheared powder which was measured at IAT was reported as negative S/RD, it contributed to the decreasing the cumulative S/RD.

For safeguard measures to Flush-out activities, TRP communicated with NRA and IAEA in order to ensure the safeguards requirement. Since solution transfer is performed on day-shift basis not 24 hours operation, TRP provided the entire schedule for the transfer between main tanks before the start of each stage in order to support the smooth verification by NRA and IAEA without stationed inspector. When the postponement of transfer or schedule change were happened by the operation trouble, updated schedule was informed urgently to NRA and IAEA. TRP tried to reschedule the operational activity for flush-out in order to minimize the influence to dispatch of inspector and human resource toward a coexistence of Safety and Safeguards. Furthermore, random basis inspection by short-notice has been maintained based on the result of discussion with NRA and IAEA. The priority of activities was discussed in advance assuming various cases such as random inventory inspection is selected on the day when the solution transfer to main tanks subjected to RII is scheduled, so TRP made effort to response the safeguard approach without discrepancy of recognition and delay.

4. Future plan

Flush-out plan Stage B for the collection of $Pu(NO_3)_4$ solution and transfer to HALW storage as RW is started from March 2023, and implementation of Stage C for fabricating UO_3 powder from remaining uranium in the process is planned from the end of 2023 to complete all of activity for Flush-out by March 2024. After the Flush-out plan, TRP is planning the shipment of unprocessed SF to other facility and process rinsing by alkaline agent as the decontamination for the equipment dismantling. The zero confirmation for no inventory tanks after process rinsing will be required that is, the concluding the MC&A of TRP. TRP continues the cooperation and discussion with NRA and IAEA for conducting the appropriate accounting and safeguards measures toward decommissioning.

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