

Integrated System for Spent Ion Exchange Resin Processing in Nuclear Power Stations – 11164

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ABSTRACT

The Tsuruga nuclear power station Unit 1 owned by the Japan Atomic Power Company (JAPC) will terminate its commercial operation in 2016. For a safe decommissioning of the station, JAPC and JGC Corporation have investigated the processing of stored radioactive wastes such as spent ion exchange resin (IEX) and filter sludge (FS). A unique wet-oxidation process operating at 100 °C under atmospheric pressure will be applied for decomposing organic substances in the waste. The treated waste containing radioactive species will undergo cementation using our Super Cement (SC) solidification process. For the treatment of high activity waste, the combined process of wet oxidation and SC solidification provides advantages over conventional incineration and direct solidification methods; for example, it can be operated environmentally safely under milder operating conditions with simple equipment and has the capability to reduce the volume of the waste material. It is expected that the combined process can be applied for the treatment of FS as well as spent IEX in other nuclear power stations.

INTRODUCTION

In recent years, the interest in the management of radioactive wastes such as spent ion exchange resin (IEX) and filter sludge (FS) generated at nuclear power stations has increased rapidly from the viewpoint of the safe decommissioning of the stations. In order to dispose of these wastes environmentally safely, cement solidification technique can be applied. However, conventional direct cementation techniques produce a large quantity of solidified wastes, which results in high costs for their disposal and transportation [1]. Moreover, organic compounds packed in cement products could cause the mobilization of radioactive nuclides as well as the generation of unfavorable gases by reacting with alkaline substances [2-4].

Decomposition of organic compounds contained in IEX and FS is considered effective in reducing waste volume and achieving savings in disposal capacity and costs. In addition, the potential impact of organic compounds in cement products can be eliminated [5].

JGC Corporation has been developing a unique wet-oxidation process and Super Cement (SC) solidification process for the treatment of radioactive wastes. The combined process can accomplish the effective decomposition of the organic compounds, followed by the cement solidification. It is noteworthy that the combined process has only a small footprint which enables the equipment to be installed in a restricted space. Moreover, the solidified product can be made homogenous and hard enough to meet the desired level for radioactive waste disposal. The Japan Atomic Power Company (JAPC) and JGC aim to apply this process for the treatment of IEX and FS stored in the Tsuruga nuclear power station Unit 1.

PROJECT SCHEDULE

The Tsuruga nuclear power station Unit 1, which began operations in 1969 as the first commercial LWR plant in Japan, will be taken out of service in 2016. Thus, the waste is scheduled for processing based on the scenario as described in Fig. 1.

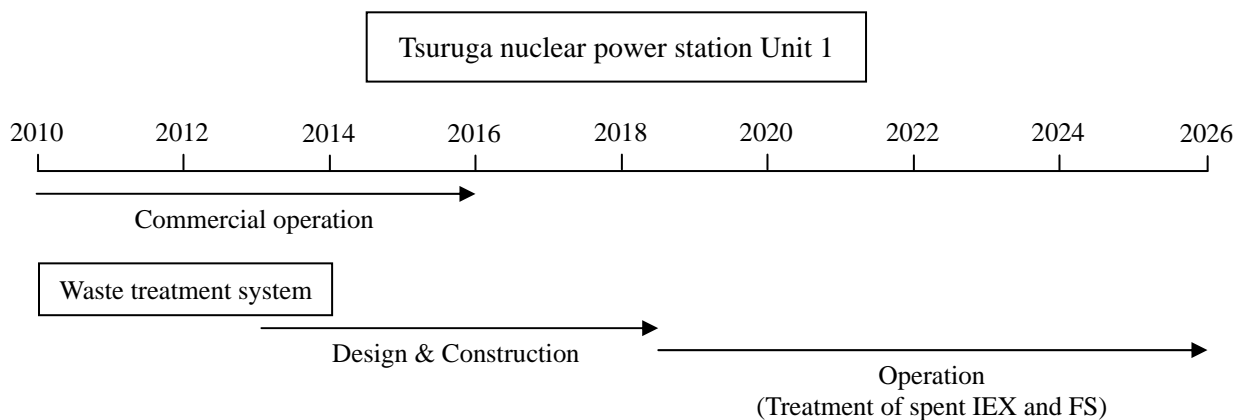


Figure 1. Decommissioning Schedule of the Tsuruga Nuclear Power Station Unit 1

As previously reported, JAPC has undertaken an investigation on these difficult to treat wet radioactive wastes stored at the station [6]. The wastes containing spent IEX and FS are stored in four spent IEX storage tanks and five FS storage tanks, respectively. Their contents, volumes, and activity concentrations are shown in Table 1.

Table 1. Contents of Waste Storage Tanks in the Tsuruga Nuclear Power Station [6]

	Spent IEX storage tanks	FS storage tanks
Contents	Spent IEX Crud	Filter aids – cellulose base – diatomaceous earth base, etc. Spent IEX Crud
Total volume of waste (mass)	280 m ³ (205 ton-wet)	260 m ³ (270 ton-wet)
Co-60 activity concentration	1 × 10 ¹¹ Bq/ton-wet	1 × 10 ⁹ –10 ¹¹ Bq/ton-wet

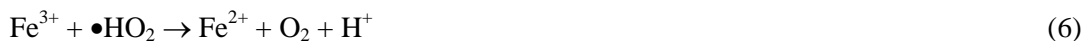
The volumes of the stored spent IEX and FS are 280 m³ (205 ton-wet) and 260 m³ (270 ton-wet), respectively. The dose rates at the surfaces of the storage tanks range from a few to dozens of mSv hr⁻¹. It is planned that the treatment of these difficult to treat wet wastes will continue until 2026.

TECHNIQUE

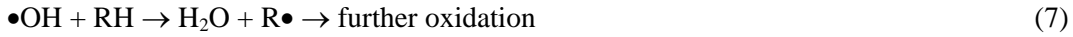
Wet-oxidation process

Since the early 1980s, JGC has been developing the wet-oxidation process using Fenton's reagents such as hydrogen peroxide and iron ion catalyst for the decomposition of organic compounds in radioactive wastes and the reduction of the waste volume [2,6-8].

In this process, hydroxyl radical (•OH) can be continuously produced *via* several sequential reactions described as follows [9-11]:



Since the hydroxyl radical is a strong oxidant, organic compounds in radioactive wastes will be easily decomposed as shown in the following equation:



Although various techniques including incineration [12,13], steam reforming [14,15], plasma assisted combustion [16,17], and supercritical water oxidation [18-20] have been developed, the wet-oxidation process provides advantages over other alternatives. For example, wet oxidation can be operated at lower temperatures with simple equipment and avoid the emission of polluting gases such as NO_x and SO_x during the operation [13, 21,22].

A schematic flow of the JGC wet-oxidation process is represented in Fig.2. Waste is mixed with water to adjust its concentration in the waste feed tank in advance of the transfer to the reactor tank. The pH of the wastewater should be maintained below 4 by the addition of sulfuric acid or sodium hydroxide during the operation. The decomposition reaction begins with the addition of a catalyst and 35% hydrogen peroxide after heating up to 100 °C. Both the waste and hydrogen peroxide can be added to the reactor tank, if necessary, until the complete mineralization of the preplanned amount of waste. Finally, the wastewater is concentrated to accomplish the high volume reduction.

The secondary products are off-gas and the concentrated wastewater. The off-gas is passed through the catalytic combustor by the off-gas blower to decompose the small amount of volatile organics before exhaustion to the environment. The vapor component in the off-gas is condensed and received in the distillate tank. The concentrated wastewater is transferred to the SC solidification system as described below.

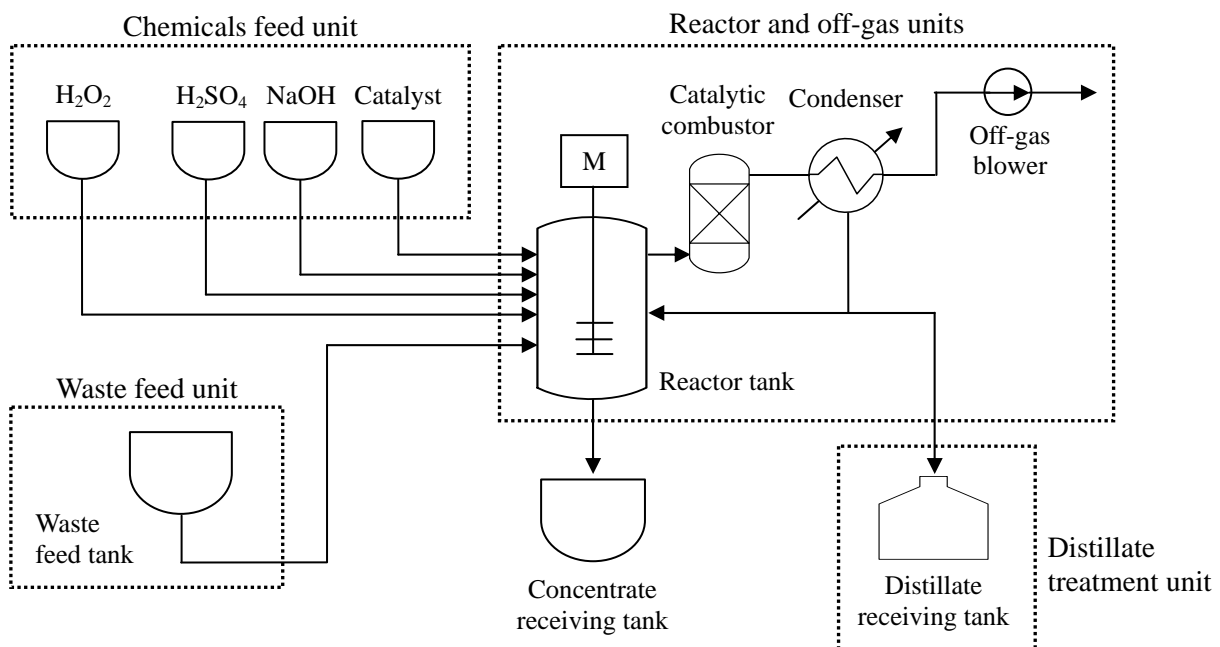


Figure 2. Schematic Flow of the JGC Wet-oxidation Process

SC solidification process

JGC has been developing the SC solidification process since the mid-1990s for the immobilization of various wastes at nuclear power stations; for example, concentrates containing sodium nitrate, sodium borate, and sodium sulfate, incineration ash, and HEPA filters [23-28].

A schematic flow of the JGC SC solidification process is represented in Fig. 3. Waste, Super Cement, and some additives are mixed in the reactor tank. The component and operation parameters should be optimized for each kind of waste.

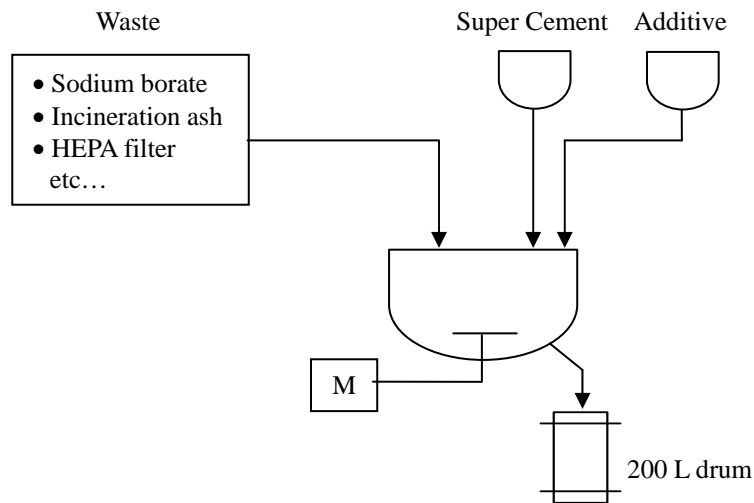


Figure 3. Schematic Flow of JGC SC Solidification Process

The advantages of this process are listed below:

1. High strength after solidification
2. High fluidity of the cement paste
3. High waste loading in the cement product

A large quantity of radioactive waste can be encapsulated if the cement matrix has high strength. Super Cement, which is categorized as alkaline activated slag cement, has higher compressive strength and bending strength as compared with ordinary Portland cement [28]. The high fluidity of the cement paste can improve the ease in handling when it is discharged from a cement mixer and the inside of the mixer is rinsed out for the maintenance work. JGC has measured the fluidity of Super Cement by the P-funnel method.

These excellent properties enable the high waste loading in the solidified product. Table 2 shows the

expected amount of waste loading in 200 L drums with the compressive strength and density of the solidified products. The treatment of HEPA filters was accomplished by the use of an alkaline activator followed by the SC solidification process [27]. Each solidified product shows a high strength with high waste loading.

Table 2. Physical Characteristics of Solidified Products [26,28]

Waste	Compressive strength [MPa]	Density [g cm ⁻³]	Waste loading in 200 L drum [kg]
Sodium borate	20	1.8	200
Sodium sulfate	35	1.8	220
Incineration ash	20	1.8	130
HEPA filter	10	1.8	8 modules

SYSTEM

The combination of the wet-oxidation process and the SC solidification process was investigated for the treatment of spent IEX and FS. The processing scheme of the integrated system is shown in Fig. 4. The waste sample is diluted with water to 5–10 wt. % of solid concentration. Then it is treated by the wet-oxidation process to decompose and concentrate the waste. The obtained concentrate undergoes the SC solidification process to form the solidified waste.

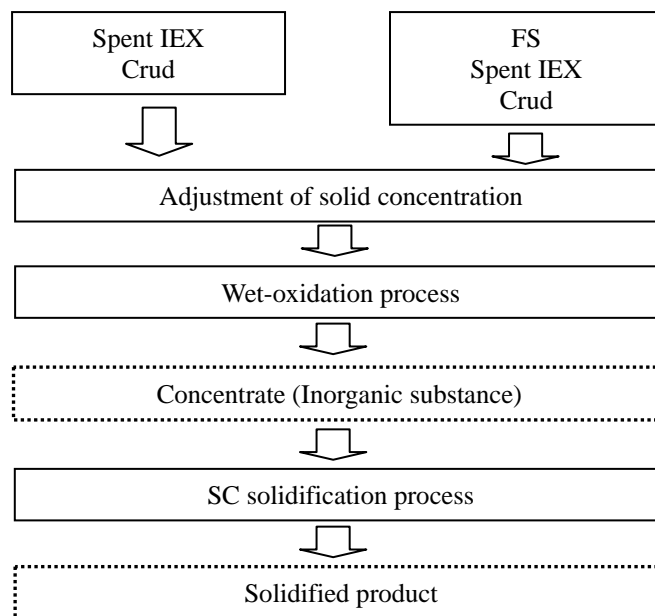


Figure 4. Schematic Flow of the Waste Processing

In order to investigate the degradation of IEX and FS and the physical properties of the solidified products, both lab-scale and pilot plant experiments were carried out in the JGC R and D center. In these experiments, simulated wastes of IEX and FS were used.

Based on the results of the safety assessment of waste disposal site, the K_d value (the ratio of nuclide concentrations between the solid and water) should be decided upon according to the desired levels. The lab-scale wet-oxidation experiments suggested that the K_d value became acceptable when the TOC (Total Organic Carbon) concentration was decreased to a few dozen ppm [6]. Figure 5 shows the change in TOC concentration of the wastewater containing a simulated IEX during the wet-oxidation process using a pilot plant with a reactor capacity of 150 L. Since the IEX in the wastewater added into the reactor tank is constantly decomposed by the oxidant, and the temperature condition causes evaporation, a large amount of wastewater can be treated. In the first 24 hours of the experiment, the wastewater was continually added into the reactor tank; however, the TOC concentration remained largely unchanged as it ranged from a few hundreds to a thousand ppm. This means the added waste was decomposed effectively. The sharp decrease in the TOC concentration after the cessation of the wastewater supply indicates almost all of the wastes were decomposed successfully. Likewise, the TOC concentration of the wastewater containing FS decreased to below 10 ppm. This suggests most of the FS can be decomposed in the same way as that of the IEX.

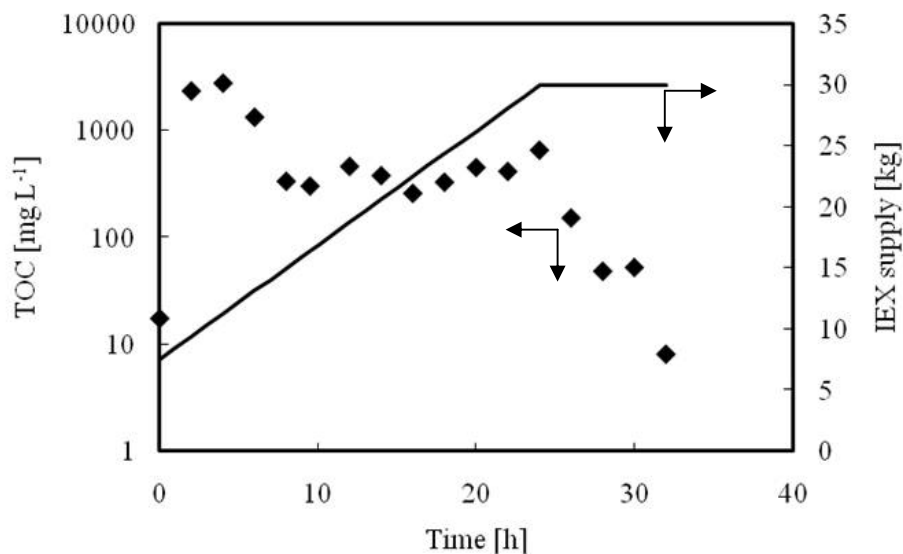


Figure 5. Change in TOC Concentration with the Additive Amount of the IEX during the Wet-oxidation Process

Solidified products are required to be homogenous and have compressive strength higher than 8 MPa.

JGC conducted the SC solidification experiment on a laboratory scale. In this experiment, the concentrate obtained by the wet-oxidation pilot scale equipment was solidified with Super Cement. Table 3 summarizes the physical parameters of the solidified products. Both products showed high compressive strength. Moreover, their waste loadings in 200 L drums reached as much as 300–400 kg while conventional direct solidification achieves only a few dozen kg. This means that the waste volume of spent IEX and FS could be reduced to one-tenth or less.

Table 3. Physical Characteristics of Solidified Products

Waste	Compressive strength [MPa]	Density [g cm ⁻³]	Waste loading in 200 L drum [kg]	
			JGC process	Conventional*
Spent IEX	13	1.5	300	30
FS	13	1.6	400	30

*Direct solidification by ordinary Portland cement

CONCLUSION

JAPC and JGC have been intensively conducting research activities to develop a method for processing spent IEX and FS to support decommissioning of the Tsuruga nuclear power station Unit 1. The wet-oxidation process, which requires only simple and compact equipment, was able to completely decompose the organic portion of the simulated waste into CO₂ and H₂O, and decrease the waste volume dramatically. At the same time, the residue could be successfully immobilized by the SC solidification process, which will satisfy the safety requirements necessary for its final disposal in Japan. As a conclusion, a combination of wet-oxidation and SC solidification is best suited for processing IEX and FS satisfactorily, especially as its compactness allows use in the space restrictions imposed by the scale of existing facilities.

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