Radwaste Reduction Technology for Spent Resins

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Abstract

Fuji Electric has developed a low pressure oxygen process (hereafter called LPOP) technology for mild decomposition and mineralization of an organic material such as ion exchange resin (here after called IXR). This method is suitable for radioactive spent resin volume/weight reduction, stabilization for final disposal and disposal cost reduction. In this process, the IXR is vaporized and decomposed into gas-phase with pyrolysis, and then are decomposed and oxidized with low-pressure plasma activity based on oxygen. This process is achieved under moderate conditions for radioactive waste.

・incinerate temperature: 752 - 1292 F (400 - 700 deg C)

・low-pressure plasma condition: 0.19 - 0.77PSI(1.3 - 5.3kPa)

From the result of this process, named LPOP(low pressure oxidation process) by the inductively coupled plasma, we have confirmed that the process is applicable for organic fireproof waste like IXR, and found that the used resin treatment performance is the same as cold test (using imitation spent resin) [1] [2] [3].

In this paper, the outline of the LPOP technology, and three research results on the possibility of LPOP residue for geological disposal are reported.

1. Study of the residue chemical form after LPOP process

2. Study of the geological disposal

3. Study of economic evaluation about LPOP technology using FRR

Introduction

In a nuclear power plant, spent IXR (hereafter called SR), low-level radioactive waste,is produced from water purification systems in association with the operation of the plant. These SR are different in a radioactivity concentration depending on purification systems, and SR which is relatively higher in the radioactivity concentration is stored in the nuclear power plant. Stored amounts continue to increase year after year.

In Japan, it is planned that SR will be buried in the ground in the future as a "waste solid" which is solidified in a specific metal container. In terms of securing long-term soundness of the waste solids, it is said the solidification of the SR is required after stabilization treatment is performed. Moreover, SR with high radioactivity concentration requires higher disposal expenses; therefore, the reduction of the disposal volume is required in terms of cost-cutting. To realize these requirements, processing technologies are required which satisfy both the volume reduction and stabilization of the SR at once.

Fuji Electric is continuing to develop LPOP technology and the equipment (FRR: Fuji Resin Reducer) as technologies to respond to these requirements. LPOP treatment is a technology enabling realization of both volume reduction and stabilization of SR which is low-level radioactive waste. The development of the technology has come at a performance evaluation by real scale testing equipment and at a survey testing the solid waste after the development of a principle testing model [1] and then the development by a hot test, in which actual resins were used in a functional testing machine [4][5].

This report describes the overview of LPOP technology and also prospects for application in the US.

1. Overview of LPOP technology

1-1 Basic principle

The LPOP processing is a method to conduct controlled burning and oxidation decomposition processing of flame-resistant substances by combining LPOP and heater-used heating. A basic concept of the LPOP equipment using high-frequency power and a work-coil is shown in Figure 1.

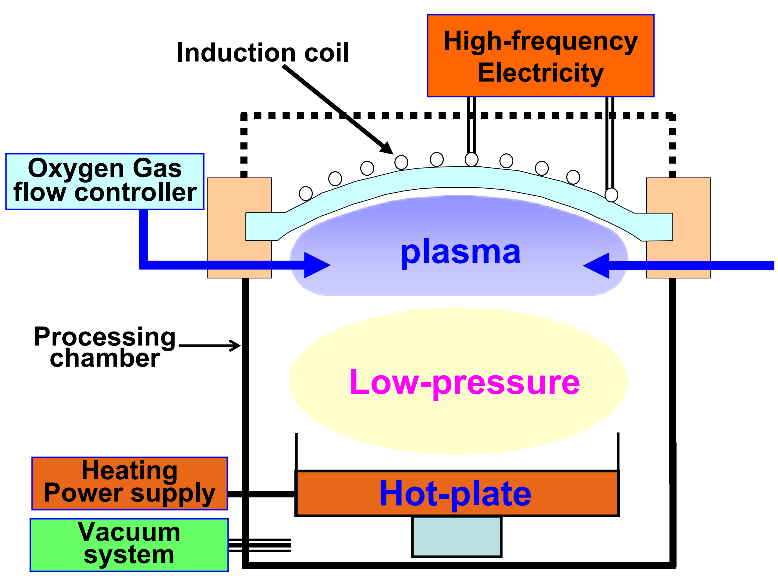

Figure 1
A basic concept of LPOP

1-1-1 Features of Low Pressure Oxygen Plasma (LPOP).

Electric discharge is very easy method of generating plasma that consists of electron, ion and un-ionized neutral molecules. The Melting process, using arc-discharge in atmospheric pressure, it generate a high temperature field of several thousand degrees. However, LPOP is using a glow discharge under low-pressure atmosphere. This plasma is a partially ionized gas consisting of neutrals and activates. And it generate a low temperature (as a plasma) field of less than thousand degrees. This type of plasma is called low-temperature plasma.

Oxygen in the low-temperature plasma is produced as very-high atomic oxygen with chemically-activity by passing processes shown in the following formulas.

In general;

Collision of nearly 5eV electron energy;

$$O_2 + e^- \rightarrow O_2^* + e^- \rightarrow O_2 + e^- + h\nu \rightarrow O + O + e^-$$

Collision of 12 eV or more electron energy;

$$O_2 + e^- \rightarrow O_2^+ + 2e^- \rightarrow O + O^+ + 2e^-$$

Figure 2. shows the result of emission spectro-chemical analyses of the LPOP in FRR. The 777nm spectrum, the emission of atomic oxygen, was clearly measured. In addition, the dependency of 777nm emission strength upon the high-frequency power. With the high-frequency power increasing, 777nm emission strength increases, and generation of the atomic oxygen is - promoted. Based on this result, degradation capability of organic materials increased with the increased amount of the high-frequency power. That is, the atomic oxygen has high degradation capability of organic materials and the LPOP method uses the atomic oxygen to decompose organic materials.

1-1-2 Processes.

Typical IXR is synthetic resins having a chemical structure in which a functional group such as sulphonate - ($-SO_3$), 4-grade ammonium group ($-NH_3$), etc. is introduced in a cross-linked three dimensional highpolymer base substrate. The high-polymer substrate used in most IXR is a copolymer of styrene and di-vinyl benzene (DVB). Furthermore, it is flame-resistant organic substances. The SR used in water treatment systems of a nuclear power plant accompany ions such as Fe, Ni, Co, Cs and solid components consisting of them.

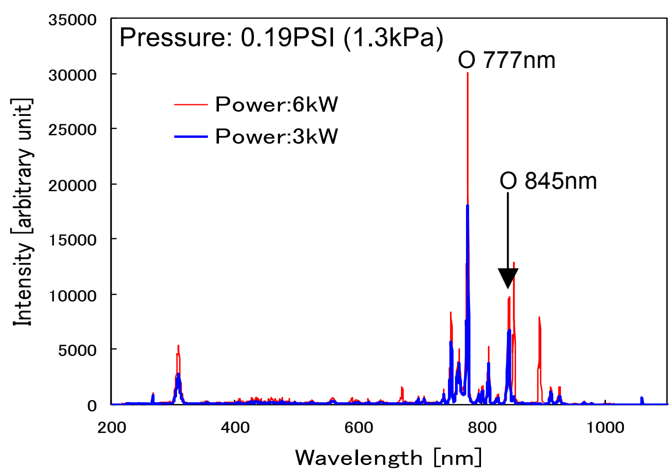

Figure 2
Emission spectro-chemical analyses of FRR.

The LPOP treatment performing the volume reduction and stabilization of SR consists of two processing steps. LPOP treatment performs controlled combustion of flame retardant materials and their oxidative degradation by means of combination of plasma activity in a low pressure oxygen atmosphere not developing to thermal plasma and; temperature control by heater heating. The concept of the LPOP treatment using IC plasma is shown in Figure 3.

Step-1 aims to process volatile components in the SR. Step-1 controls the heating temperature of subject materials by an electric heater and decomposes thermally the SR by heating them to approximately 752 F (400 degrees C). In addition to the treatment, pyrolysis gases are decomposed by oxidation through using the oxygen heated and activated by the low pressure oxygen plasma. Because the pyrolysis components are produced in ample amounts in Step-1, swirling gas flow is applied. The swirling flow controls - the thermal flow generated by the low pressure oxygen plasma flowing into the subject materials. By these processes, Fuji Electric achieves “mild treatment” controlling the progression of burning even in an oxygen atmosphere. In addition, as pyrolysis gases contact frequently with the oxygen due to the swirling flow, the produced pyrolysis gases burn well in a condition of less soot and tar.

Step-2 aims to achieve high volume reduction by driving the combustion of carbon components. At Step-2, carbonized residues remaining in Step-1 are further heated to approximately 1292 F (700 degrees C). In addition to the treatment, it is aimed to promote the oxidation decomposition of the carbon components by using the heated and activated oxygen produced by the low-temperature plasma and to make the combustion of carbonized SR progress, and achieve the high volume reduction. It is important at Step-2 to make sufficient oxygen to be taken into the SR. A downward gas flow is applied as a gas flow by which the oxygen is taken effectively into the SR. Through the above processes, the mineralization of the SR is progressed immediately with volume reduction.

In each treatment of these steps, carbon (C), a main component of the SR, is converted into CO_2, and hydrogen (H) into gas by oxidation in H_2O. Moreover, the components of the functional group such as $-SO_3$ or $-NH_3$, etc., performing ion-exchange, react to SOx or NOx - gases and are emitted as exhaust gases. The LPOP does not use air in treatment, In addition, the components absorbed in the SR remain as oxidized substances and salt [4].

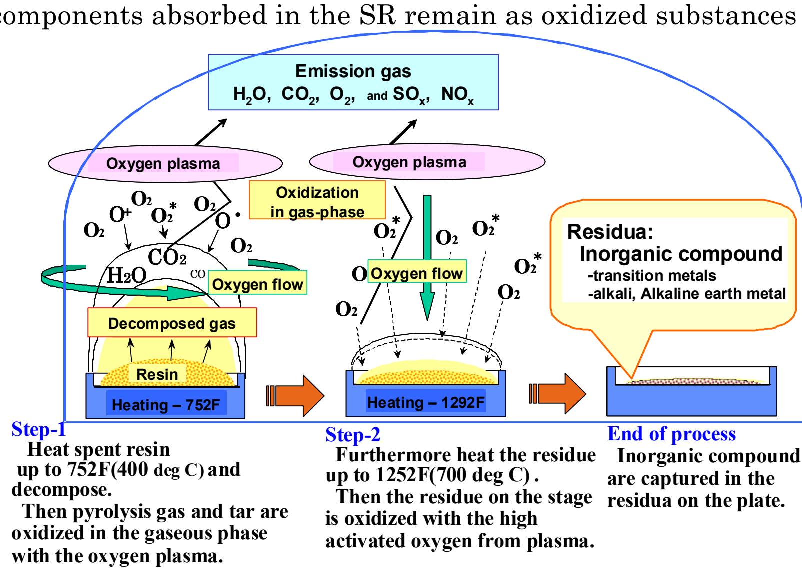

Figure 3
Conceptual diagram of the LPOP method

1-2 Actual scale testing equipment

As shown in Figure 4, The real scale testing equipment which has a low pressure processing chamber with a bore diameter of 2.6 ft (800mm) and a height of 3.3 ft (1000mm) includes a stage where resins with a diameter of about 2.1 ft (650mm) are heated, and has the processing capacity of 10.6 gals (40 liters) per day. The footprint is 127.4 ft^2 (mm^2).

The top side of the processing chamber is covered with a window made of quartz, and an induction coil to produce plasma is placed on it. The, processing chamber is connected with a vacuum pump to make the pressure in the processing chamber low and an oxygen gas supplying system. Moreover, it is connected with resin feeding equipment enabling the supply of - resins at low pressure and with resin collecting equipment by which resin ashes processed at atmospheric pressure is collected by suction [6].

Exterior view of the actual scale testing equipment is shown in Figure 5. An interesting aspect of this system is that there is not a 2nd combustion chamber.

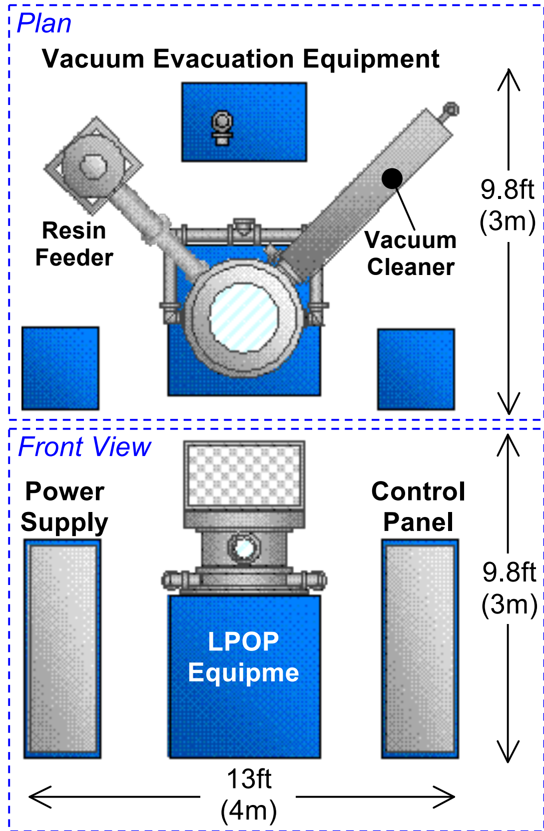

Figure 4
Configuration of Fuji Resin Reducer (FRR)
* Horizontal Projection

1-3 Features

1-3-1 Volume reduction and mineralization performances.

The IXR contains carbon by about 50 %. The LPOP treatment is capable of changing carbon components into carbon dioxide (CO_2) through using the chemical activity of LPOP. As a result, it is possible to perform high volume reduction and high weight reduction of the SR.

The result of the performance tests for the volume reduction and weight reduction after the LPOP treatment has been implemented in two steps shown in item 1-1-2 is shown in Figure 3. For the tests, IXR which absorbed cold components, were used. In this matter, main ion components in the water from the nuclear power plant are considered. Through the LPOP treatment, we attained the result that the ratio of the volume reduction was not less than 90% (1/10) and the ratio of the weight reduction was not less than 95% (1/20).

Figure 5
Exterior view of the actual scale testing equipment

Actual picture of this condition is shown in Figure 6.

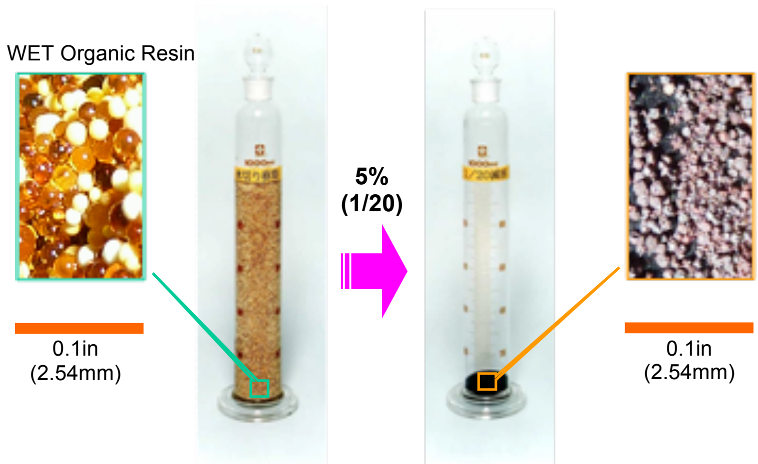

Figure 6
Actual Picture of before LPOP treatment and after treatment resin

The ratios of the volume reduction and weight reduction were calculated by the next equations with reference to aqueous resin after additive adjustment.

Weight reduction rate = ((1- residue weight) / aqueous resin weight) x 100%

Here, aqueous resin weight = weight of resin processed by drying - (1-moisture percentage of aqueous resin/100)

Volume reduction rate = ((1- residue volume)/aqueous resin volume) x 100%

Furthermore, we evaluated the reduction effects of the LPOP treatment for components such as Carbon (C), Hydrogen (H), Nitrogen (N) and Sulfur (S) which are the main components in IXR.

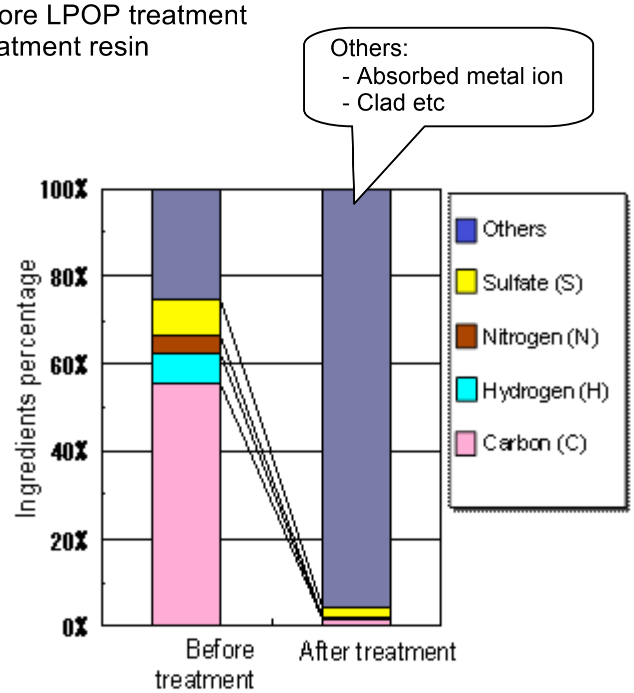

Figure 7
The result of mineralization by LPOP treatment

As a result, the 99wt percent of C, H, N, S components in the SR were removed by the LPOP treatment. Because the components C, H, N and S are organic components of the resins, to remove these components is to mineralize resins. Through the results, we could see that the SR has been mineralized by 99 percent [4]. Figure 7 shows the result of mineralization.

1-3-2 Retention performance of nuclear species.

The temperature in the LPOP treatment is set at an appropriate low value as a feature of IXR. This setting aims at the control of unnecessary steam spread of radioactive substances. We implemented the LPOP treatment, and evaluated the ratio of the amount (w) of nuclides contained in residues collected from the stage after the treatment to the amount (W) of a nuclides absorbed in resins before the treatment, and the ratio of the amount (m) of nuclides collected from gas emissions. As a simulation of SR, we evaluated the retention of absorbed nuclides to emission systems by using resins in which cold cobalt (non-radioactivity) and cesium had been absorbed.

As the LPOP treatment equipment, we used the real scale testing equipment. Table 1. shows the evaluation result. We could obtain the result that the transitivity amounts of both cobalt and cesium were under detective limits (approximately 100 percent was collected.)We considered that the fluctuation of collected amounts is due to the impact of the fluctuation of absorbed amounts and analytical precision.

Table 1
Results of the transitivity amount of cobalt and cesium (Example)

Nuclide	Retention (Inside-Chamber) (w)/(W) x 100 (wt%)	Transitivity (Vacuum-pump) (m)/(W) x 100 Exhaust (wt%)
Cobalt	90 – 100	No-Detect ($<10^{-4}$)
Cesium	80 – 100	No-Detect ($<10^{-4}$)

Furthermore, we implemented the LPOP treatment of the real SR produced at ATR (Advanced Thermal Reactor) "Fugen" and evaluated the retention capacity of γ species and H-3 and C-14. As the LPOP treatment equipment, we used compact testing equipment which can be taken into the testing field of power generation plants. The testing results after actual SR processing by LPOP are shown in Figure 8. The result of the test shows that the transitivity rate of Co-60 into the exhaust systems was low, and we could collect it on the stage. In addition, we could take the results that almost H-3 and C-14 transferred into exhaust gases, however, H-3 remained in the resin ashes at the ratio of about 10^{-2} and C-14 at the ratio of about 10^{-3}. We consider these results to be valuable data for the planning of waste solids [5].

1-3-3 Processing time.

As shown in Figure 9, the LPOP treatment by FRR, in treatment of 1 batch, can reduce the weigh of dry resin to 80% in 250 minutes and to 90% in 300 minutes.

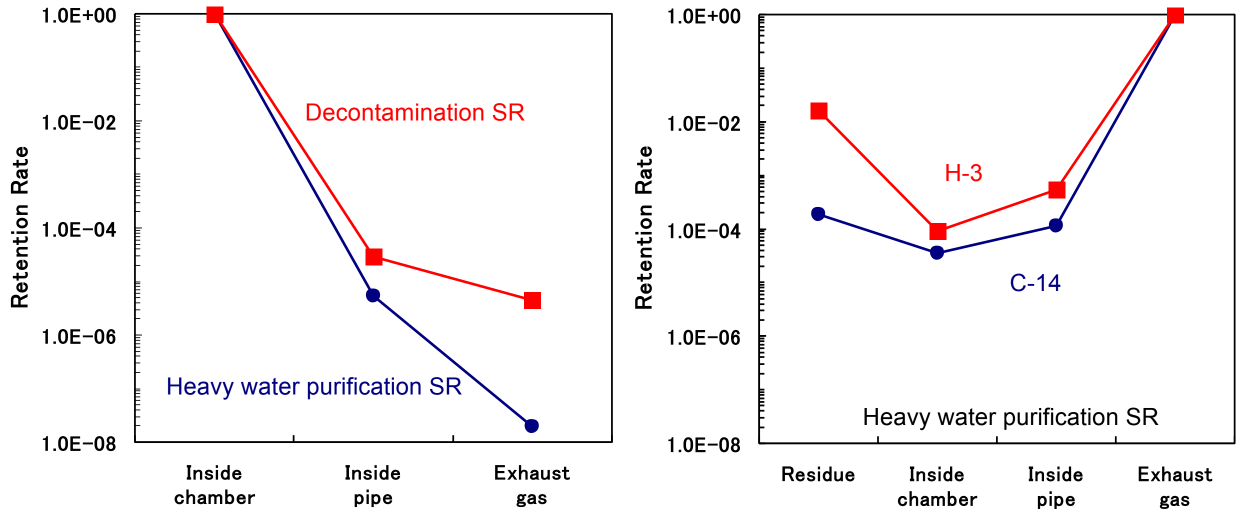

Figure 8
Co-60, H-3 and C-14 distribution

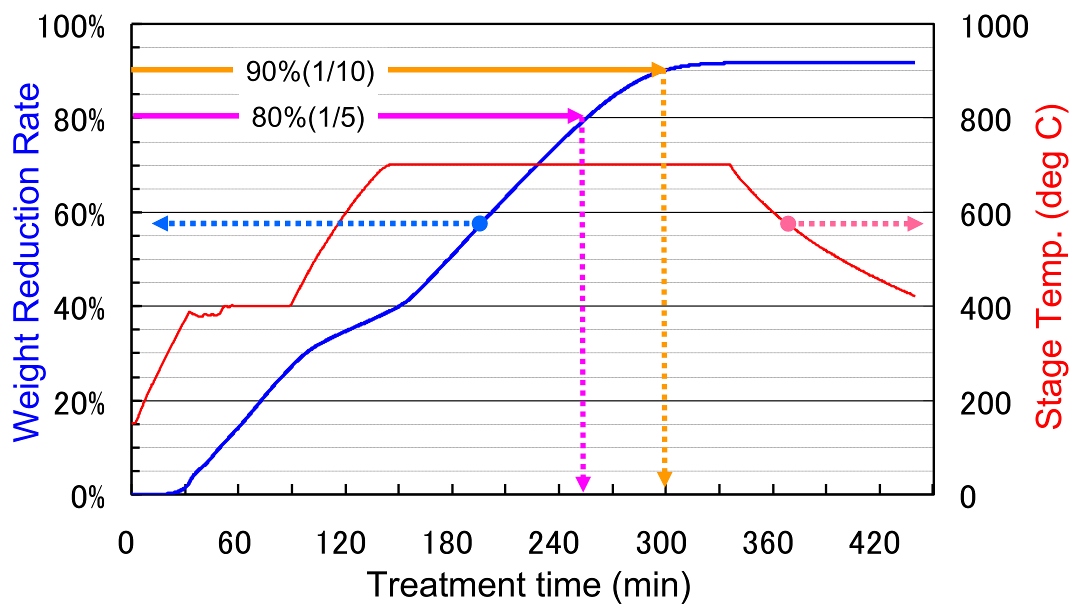

Figure 9
Example of progress of weight reduction
(Ion exchange resin)

1-3-4 Low secondary waste.

To produce plasma in the FRR, high-frequency discharge is used. Through electric discharge, the high-frequency current is supplied to the induction coil installed at the outside of the processing chamber, consequently, generated inside the processing chamber.

FRR has the following features.

- As the induction coil and insulators are separated from the plasma, they do not suffer from degradation and damages.

- As the electrodes and coil are not worn, no exchange is required.
  Because low temperature plasma is used, it has the following features.

- The processing chamber has a simple structure without refractory materials, frequency to maintain and repair - the inside processing chamber is decreased.

- As fire-resistant bricks are not used, those which contain radioactive substances are not produced as waste.

The material needed to process is merely oxygen, and the materials to make SOx and NOx produce are merely components in resins. Thus, the following features.

- Lesser amounts of the exhaust gas systems more compact, and the amount of the secondary waste such as filters is less.

- The low temperature plasma makes the generation of NOx less (no thermal NOx is generated.)

2. Prospects of applications of LPOP/FRR

2-1 Maintainability and expansibility

The basic configurations of the FRR mounted with LPOP technology have very small sizes and compact systems. To utilize these features, the main configurations of the FRR are unitized when considering the handling of high dose SR. According to compact configurations, it is easy to disassemble and exchange parts, and to maintain the body.

Utilizing the characteristic that the configurations are compact, it is considered possible to put on a trailer as a transportable module.

By this, the transfer and installation of FRR becomes easy, and will reduce installation time and expenses. FRR has the potential to be mobile though there are some challenges such as decontamination when relocated. Additionally, if there is a requirement for high volume treatment, FRR can adapt to the requirement to increase the throughput by increasing the number of units by sharing the basic utilities. In this case, by sharing the basic utilities, it is possible to reduce the cost per unit as well.

2-2 Advantages of introducing FRR

Fuji Electric demonstrates the following advantages such as economic efficiency, long term storage, etc., by introducing FRR using LPOP technology.

1. Disposal amount is reduced by the volume reduction.

· companies who outsource- disposal can reduce outsourcing costs.

· companies implementing the disposal can make life of the disposal field longer.

2. Mineralization can be attained at once as well as volume reduction.
   Long-term soundness is improved by lightening the impact on the vessel for waste.

3. Conclusion

Fuji Electric continues to develop POP and the (FRR)equipment The LPOP treatment is technology achieving volume reduction processing and stabilization processing of low radioactive waste together. By substantially reducing the volume, it makes possible disposal cost savings and storage space reduction. In addition, because the equipment uses no refractory materials and is compact, it is possible to deliver the equipment as flexible equipment matching the processing volume and installation location.

Fuji Electric expects to apply the treatment of high-dose SR in the future. We hope to contribute to reduction of disposal cost and effective utilization of storage space for SR with FRR in US.

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