Development of the Practical Technology for Thermal-Chemical Treatment of Low-Level Spent Resin

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1. Introduction

About 620,000L of low-level spent resin was generated during NPP operations in 2018, and it is now temporarily stored in 200 L drums. The incineration or vitrification treatment technology may be considered for treatment of this large quantity of low-level spent resin, but they have problems, i.e. generation of large amounts of harmful gases and discharge of radioactive nuclides. Also, as the cement and polymer solidification technology has a low volume reduction effect, it is regarded as inappropriate for treatment of large quantities of spent resin generated by a secondary system.

Meanwhile, among the technologies that can be used for treatment of low-level spent resin of NPPs, the technology for thermo-chemical treatment of spent resin, developed by the Korea Atomic Energy Research Institute, is a technology for coalification of spent resin whose main component is carbon. It is an outstanding volume-reducing technology that evaporates the radioactive nuclides in the spent resin at a high temperature. The principles of the spent resin thermo-chemical treatment process are as follows:

1) Removing and recovering the moisture containing tritium (H-3) included in the spent resin
2) Analyzing the functional group by means of pyrolysis
3) Coalification of spent resin containing inorganic nuclides whose functional group is removed
4) Chlorination of nuclides in spent resin carbide, such as Cs and Co, and other inorganic components at a high temperature
5) High-temperature evaporation, separation, and condensation recovery of chlorinated metal nuclides and inorganic matters

2. Process Description

In this study, the authors will try to use the original technology, developed by the Korea Atomic Energy Research Institute, to make a pilot device for thermo-chemical treatment of low-level spent resin generated in NPPs and use mock spent resin to derive practical applicability and a treatment technology.

2.1 Spent Resin Thermo-Chemical Treatment Process

The technology for thermo-chemical treatment of spent resin gasifies only the hydrogen (H), oxygen (O), nitrogen (N) and sulfur (S) component among the organic components (C-H-O-N-S) in the spent resin, and coalifies the carbon (C) component. This technology halogenates, evaporates and separates a very small amount of inorganic matters, including the radioactive nuclides remaining in carbide, at a high temperature. The principles of the spent resin thermo-chemical treatment process are as follows:

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[Step 1] is the process of recovering and treating the tritium (H-3) in the spent resin by drying the spent resin at a temperature ranging between 100℃ and 120℃. [Step 2-1] causes the pyrolysis reaction by maintaining the temperature between 150℃ and 500℃. In this pyrolysis reaction temperature range, as styrene divinyl benzene copolymer and the
functional group are separated, SO₂ and NH₂ gas are generated in the spent resin. [Step 2-2 / 2-3] analyzes the styrene divinyl benzene copolymer and coalifies it. To separate the inorganic nuclides in the coalified spent resin, it reacts with Cl₂ of a certain concentration, and evaporated into such chlorides as CsCl(g) and CoCl₂(g) at a temperature exceeding 1,000 °C.

2.2 Making a Device for Thermo-Chemical Treatment of Spent Resin

Based on the spent resin thermo-chemical treatment process, the authors made a pilot device for batch-type thermo-chemical treatment of spent resin with a capacity of 20 kg that can treat spent resin once. The device is comprised of the high-vacuum thermo-chemical treatment reactor for treating the radioactive nuclides, contained in spent resin, at a high temperature, and the plasma incinerator and wet scrubber for treating the harmful gases and volatile radioactive nuclides, generated in the high-vacuum thermo-chemical treatment device.

Fig. 2. Device for thermo-chemical treatment of spent resin.

2.3 Mock Spent Resin Thermo-Chemical Treatment Experiment

The same numbers as used in NPPs were used to conduct the mock thermo-chemical treatment experiment. To treat mock spent resin thermo-chemically, the holding time for the optimal temperature range of each stage, i.e. intermediate temperature, high temperature and ultra-high temperature, must be derived. In the mock spent resin in the intermediate temperature range, styrene divinyl benzene copolymer and the functional group are separated, and in the high temperature and ultra-high temperature range, it reacts with Cl₂ of a certain concentration, and the mock radioactive nuclides, contained in the mock spent resin, are evaporated. The plasma incineration used to remove the harmful gases and volatile radioactive nuclides generated in the spent resin thermo-chemical treatment reactor essentially generates NOx. As for the N₂ and O₂ doses to minimize NOx generation, optimal conditions must be derived in consideration of the sample dose.

3. Experimental & Result

In this study, the authors used the technology for thermo-chemical treatment of spent resin, developed by the Korea Atomic Energy Research Institute, to make the pilot device for thermo-chemical treatment of spent resin. The mock spent resin was used to conduct the device performance verification experiment for deriving the nuclides separation and recovery process. To treat the mock spent resin thermo-chemically, the temperature ranges were set up, i.e. the intermediate temperature (300 ~ 350 °C), high temperature (500 ~ 1,000 °C) and ultra-high temperature (1,000 °C or higher), and the pyrolysis reaction in each temperature range was verified. Also, to minimize NOx generated essentially in the plasma incineration method, the optimal N₂ and O₂ doses were derived in consideration of the sample dose.

4. Conclusion

The technology for thermo-chemical treatment of spent resin can address the problems of the existing incineration or vitrification technology, i.e. discharge of harmful gases and low volume reduction. Also, coalified spent resin can be recycled industrially, or self-disposed. Accordingly, based on the result of the experiment using mock spent resin, the authors will verify the practical use of the technology by demonstrating treatment of the spent resin of NPP sites and secure the spent resin treatment technology.

REFERENCES