The New Technologies for the Immobilization of Radioactive Iodine and Its Implication to the Deep Waste Disposal

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1. Introduction

Radioactive iodine 129 is the dangerous radioactive substance affecting the safety of a proposed deep geologic repository significantly. Many studies indicate that it is the most hazardous isotope against the long-term post closure safety of a repository. Even though it is not either a gammy or an alpha emitter, a week beta emitter due to the following characteristics, its contribution to the annual individual dose is significant for the following reasons;

(1) It exists in the void volume of a gap between a uranium matrix and cladding materials and inside uranium matrix in used nuclear fuels[1]. When the intruding groundwater penetrates into a solid waste form after corrosion of a waste container, the iodine residing in a gap shall be instantaneously dissolved into the groundwater. The solubility limit of volatile iodine is almost infinite. The remaining iodine embedded into a uranium dioxide matrix is congruently released with the uranium dioxide matrix. The contribution of the iodine from a gap is far greater than that from a uranium matrix[1]:

(2) Iodine is not absorbed on the bentonite blocks adjacent to the waste form and the bedrock surface and migrate along the fractures and pores with the speed of the local ground water without significant dilution.

(3) The half-life of the iodine is around 16 M years quite longer than Tc-99, Pu-239 so that the effect of the concentration drop by the decay cannot be seen for the time of the interest.

(4) And iodine can be transferred into agricultural as well as marine products such as seaweeds popular in the ROK potentially enhancing the thyroid cancer in a certain case.

The feasible solutions to diminish the effect of radioactive iodine to ensure the safety of the repository, the so-called P&T measures have been tried over many decades for transmutation or spallation of iodine using either the sodium fast reactors and the accelerator driven hybrid systems. In reality, none of the approaches brought the good success to limit the contribution from this hazardous radioactive substance. The only practical approach is to dispose of it under a tighter geologic medium using the so called KBS-3 repository for the direct disposal of spent nuclear fuels or the vitrified wastes from the PUREX reprocessing.

The authors propose the new approach to safe management of this dangerous radioactive substance by applying the well-known technologies of the solidification using the HIP process in combination with the new disposal technologies based on the industrially well proven shale gas extraction et al.

2. Immobilization

In this section some of the results from studies over the immobilization of radioactive iodine in the ROK[2] and the overseas are summarized[3,4,5,6.]

2.1. Capture Processes and Size Determination

In this study, the technologies developed through the pyro-processing research[2, 7] are analyzed for the iodine captures and solid waste forms. Pyro-processing can be categorized into (1) the head-end, (2) the electrolytic reduction, and (3) the final electro-refining and winning. All iodine is captured throughout the first process, head-end while cesium and technetium are captured throughout the entire processes by the tailored filters. Therefore, it shall be good enough to identify all spent filters capturing all iodine in the head-end and begin to immobilize them with the available technologies to be discussed in the next section.

The size of the final solid waste form is a key factor to implement innovative deep geologic disposal technologies. The current proposed diameter and the height of a waste form to be used for the so called KRS+ option, the disposal option of radioactive wastes, byproducts of the pyro-processing are 20 and 10 cm respectively. When the immobilized solid waste forms are commercially manufactured, the corresponding wastes deposited into waste containers made of stainless steel or other materials shall be disposed into a deeper geologic medium.

The KRS+ option proposes to use the Deep Borehole Disposal(DBD) recently developed by Sandia National Laboratories. The other approach is to implement the Deep Isolation System(DIS) which relies on the inclined boreholes and the horizontal deposition boreholes at the depth of the final disposal. Both options aim at dispose of the hazardous radioactive wastes at the depth of a few to several kilometers.

2.2. Immobilization Technology Review

The captured iodine from the head-end high temperature treatment process shall be in the form of AgI. It is the well-known process of the Silver II to capture the volatile iodine in the traditional PUREX process. There are many excellent review papers to compare the advantages and disadvantages of the iodine immobilization technologies. The Japanese approach[3] compared three competitive approaches for the immobilization;

(i) Syn-rock using HIP Process with Al at 175 MPa and 1200 ^{o}C conditions for 3 hours
(ii) High Performance Cementation, and
(iii) BPI Glass Vitrification.

All three solid forms are known for high performance satisfying the low dissolution rates assuring the leach time of more than 100,000 years. The syn-rock forms are reported to perform well under high alkaline environment, which can be achieved with the high alkaline cement such as OPC as an engineered barrier. The long-term dissolution experiments up to 600 days were performed to validate the low dissolution characteristics of the syn-rock solids.

The integrity of the high-performance cement turns out to be excellent also. However, under the condition of high salinity, the leach rate becomes higher. It might create the burden to emplace this cement solid waste form in a repository near the marine environment and the very deep geologic media for the DBD and DIS disposal.

The BPI glass also demonstrates the excellent performance except for the potential complexation with cement.

All three candidate materials are good for the immobilization. Many studies were done to materialize the syn-rock technology with Hot Isotropic Pressing technique for final product of the solid waste form.
The independent study by Masuda et al[5] developed the stepwise approaches

(1) to eliminate the $AgIO_3$ and $AgNO_3$ throughout the pre-heating process so that all iodine is in the form of AgI,
(2) to eliminate excessive water content throughout the vacuum dry to avoid the creation of void volumes or inclusions, and finally
(3) to manufacture the solid waste form without allowing the escape of gaseous iodine throughout the 3 hour long HIP process at the temperature of 1200 ^{O}C and the 175 MPa conditions.

In addition, the appropriate scale-up studies were done without any significant problem.

3. Innovative Disposal Approaches

The long-term radiological safety of a proposed deep repository significantly depends upon the iodine-129. The safety of the traditional Swedish KBS-3 disposal concept at the depth of around 500 meters can be significantly enhanced if the disposal depth goes down to the level of a few to several kilometers. The new secure excavation technologies developed in the area of the petroleum and shale gas extraction, such as DBD and DIS can be applied for this action. However, the actual large size of the fuel assembly hinders the application of the new innovative excavation technologies.

The application of the pyro-processing, such as the head-end, can reduce the size of the waste forms for the DBD and the DIS. The corresponding immobilization technologies shall get rid of the significant role of the volatile fission products such as iodine resided in a void gap between a uranium matrix and a cladding.

These are the key advantages of the new approach to combine the technologies developed through the pyro-processing research and the new excavation technologies. Detailed analysis to illustrate the effect of the combined efforts shall be available in the future.

4. Conclusions

In this paper, the new approach to safely dispose of radioactive iodine in the form of solid is proposed to maximally enhance the long-term post closure radiological safety of a repository. The innovative disposal technologies suitable for the permanent management of radioactive wastes from the fuel cycle activities such as the pyro-processing can be a good pragmatic option to further isolate the hazardous materials away from the human environment.

When the isolation function is breached after the long time by events such as corrosion and other natural events, the release of iodine into the engineered and the natural barriers shall be limited by the performance of the immobilized waste forms. In addition, the longer migration pathways from a proposed repository to the biosphere enhances the dilution of groundwater contaminated by the advent of the iodine plume. It shall significantly decrease the annual individual dose rate of a proposed future repository.

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