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Long-term Environmental Assessment of Waste from PyroGreen System

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Abstract

We have conducted a long-term environmental assessment of a geological repository for Intermediate Level Wastes (ILW) arising from PyroGreen processes that has been developed to decontaminate all HLW from the pyrochemical partitioning of spent nuclear fuels (SNF). PyroGreen process has been designed so that final ILW can meet conservative acceptance criteria such as one established for the Waste Isolation Pilot Plant (WIPP) in U.S.A. The nuclide inventory of final vitrified PyroGreen waste is calculated using ORIGEN 2.1 based on the design decontamination factor of PyroGreen processes applied to 18,171 metric tons of PWR SNF with 45 GWD/MTU burnup. Using GoldSim model, the environmental impact of ILW upon geological disposal at an intermediate depth. Among radioactive nuclides, Ra226, Rn222 and Sn126 are identified as key contributors to radiological dose for general public. The environmental impact of PyroGreen wastes satisfies the Korean dose limit of 0.1 mSv/year with sufficiently high margin. Sensitivity studies have shown that the predicted dose can vary significantly by distribution coefficient of Ra226 and Rn222, solubility limit of Se79.

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Keywords: envrionmental assessment; pyrochemical process; long-tem safety; geologic repository

1. Introduction

The accumulation of spent nuclear fuels at most storage pools in the Republic of Korea(ROK) has been raised questions for sustainable supply of nuclear power energy. Despite the situation, ROK energy policy recommended

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nuclear energy portion keep 29% for national energy security, economy and CO2 sequestration[1]. According to this plan SNF of 18,171 tons at PWR and 12,658 tons at CANDU are expected to be accumulated by year 2035 and existing plant storage facilities will start to be saturated from 2019 year[2]. To solve the problem, both direct disposal and recycling based on pyro-chemical process are being pursued as national programs.

A conceptual design of direct disposal system in deep geologic environment in ROK has been developed by the Korea Atomic Energy Research Institute(KAERI) as the Korea Reference repository System(KRS)[3]. Long-term safety assessment results for KRS revealed important uncertainties that need to be reduced to assure the safety[4,5] like other similar studies including one for the Yucca Mountain Project[6]. Although the uncertainty problem could be solved by scientific verification based on sufficient understanding on features, events and processes(FEPs) in geologic repository system like Finnish approach, it would be long-term research project and needs wide repository area with low population density which is not appropriate in ROK situation.

Considering this situation, Advanced Korean Reference disposal System(A-KRS) for pyro-chemical process waste with reduced uncertainty and waste volume has also been developed in ROK. The pyro-chemical process design in ROK has been being developed by two organizations: KIEP-21 of KAERI and PyroGreen of Nuclear Transmutation Research Center of Korea(NUTRECK) in Seoul National University(SNU)[7]. The environmental impact assessment of KIEP-21, however, pointed out that the impact of actinide elements is highly sensitive to the release rate of waste which leaves high uncertainty[8]. To eliminate all uncertainties associated with HLW, PyroGreen process has been being designed to significantly increase the decontamination factor (DF) both on actinides, Tc, I, Cs and Sr[7,9] such that the characteristics of WIPP waste can be satisfied.

PyroGreen wastes based on database produced at NUTRECK at SNU in collaboration with KAERI, Central Research Institute of Korea Hydro & Nuclear Power Co. (KHNP-CRI), Chungnam National University (CNU), Yonsei University(YSU) and Soonchunhyang University(SCHU) in Korea. By applying DF goal of PyroGreen process, the final waste inventory is calculated assuming the PWR SNF with 45GWD/MTU after 50 years cooling time. CANDU SNF is not considered in this study because the application of pyro-chemical process to CANDU required analysis in economic benefits due to their low burnup. Also long-term environmental impact of the PyroGreen waste disposal in A-KRS is assessed in case of PWR SNF accumulation by 2035 year and the results are discussed in view point of current regulatory criteria to reduce uncertainties in future.

2. Environmental Impact Analysis Approach for PyroGreen Wastes

2.1. PyroGreen Process and Recovery Factor

The basic process design of PyroGreen is the same as KIEP-21 except for three additional processes: Zr hull electrorefining, transmutation for eliminating Tc/I and PyroRedSox[7]. The DF goal and final product of PyroGreen is given in Table 1. Because zircaloy is the second largest weight and volume in SNF[10] recycling option is more appropriate than disposal option in ROK, while in KIEP-21, zircaloy remains as waste to be disposed. Thus, in PyroGreen process, using LiCl-KCl eutectic chloride salt-based electrorefining process, i.e. Zr hull electrorefining process, almost all Zircaloy is recovered for potential reuses for nuclear industry[11].

Tc99 and I129 are long-living fission products radionuclides with high solubility in groundwater and small sorption coefficient in bentonite-filled Engineering Barrier System(EBS) and rock. Thus, significant amounts of Tc99 and I129 in waste can be released and cause serious radiological impact on biosphere unless isolation of these radionuclides from groundwater is assured for over millions of years. In KIEP-21, 98 wt% of Tc and 98 wt% of I is separated from SNF in voloxidation process[12] and vitrified to ILW which is to be disposed. In this case, long-term waste performance is extremely dependent on degradation rate of glass waste, yielding high radiological uncertainty[8]. For this reason, I and Tc in PyroGreen process are fabricated as transmutation target and eliminated in transmutation reactor. Additionally, Cs and Sr are separated with a recovery of up to 99.66%[9] and are to be kept at interim storage for 300 years because they have relatively short half-life (Cs-137: 30yr, Sr-90: 29yr). The biggest advantage of separating Cs and Sr from waste is significant retardation of waste form degradation by heat: Cs and Sr account for about 33% of heat production in PWR SNF with 45GWD/MTU.

As pointed out earlier, the release rate of TRU from vitrified waste of KIEP-21 is ridden with high uncertainty in environmental assessment results[8]. To decontaminate TRU from the wastes of pyro-chemical process of KIEP-21,