U.S. DEPARTMENT OF ENERGY
INTERNATIONAL NUCLEAR ENERGY RESEARCH INITIATIVE
DOE/ROK

ABSTRACT

Development of Voloxidation Process for Treatment of LWR Spent Fuel

Principal Investigator (U.S.): K. Howden, Argonne National Laboratory (ANL)

Principal Investigator (Republic of Korea): J.J. Park, Korea Atomic Energy Research Institute (KAERI)

Project Number: 2004-004-K

Project Start Date: June 2004

Project End Date: April 2007

The United States Department of Energy is developing a head-end fuel treatment process, known as the voloxidation process as part of the Advanced Fuel Cycle Initiative. Voloxidation, which involves oxidizing uranium oxide fuel using either air or oxygen, provides three important advantages when used as a head-end treatment process. First, it may be used to separate the fuel from the cladding. A decladding step is desirable in that it may simplify process flow sheets by excluding the cladding constituents from the fuel constituents. Segregation of cladding may also result in generation of less high level waste. In addition to decladding, voloxidation may be used to decrease the particle size of the fuel, which will in turn increase the kinetics of downstream treatment operations the efficiency of dissolution. Finally, voloxidation treatment may remove problematic constituents from the fuel prior to downstream treatment operations. Fission products such as cesium-137, krypton-85, xenon-133, technetium-99, carbon-14, and tritium (H-3) may be removed prior to fuel dissolution, simplifying the process flow sheets and yielding more flexible waste treatment operations. Head-end removal of gaseous fission products is particularly important in aqueous reprocessing where the presence of NO_x gases, generated during fuel dissolution operations, makes separation of these gases much more challenging. The head-end voloxidation process is equally applicable to aqueous or pyrochemical treatment processes.

KAERI is pursuing the development of a similar technology, known as the Oxidation and REduction of Oxide fuel or OREOX process. This process is being researched in association with development of the DUPIC process for recycle of PWR spent fuel to CANDU reactors. The objective of the OREOX process is to decrease the particle size of the fuel and to increase the sinterability of powder for being in the optimum range of surface area and crystallite size, etc. This will in turn give high density sintered pellets for CANDU fuel specification. During development of DUPIC process, KAERI scientists have gained considerable experience with oxidation and reduction processes of spent LWR fuel. The oxidation process is the same as the voloxidation process. KAERI has been measuring and analyzing the release rates of Kr-85, cesium, Xe, etc. Also, in order to trap these fission products, various concepts have been developed and applied to each off-gas treatment system of the OREOX system and the sintering furnaces in the DFDF (DUPIC Fuel Development Facility).

It is, therefore, proposed that the two parties conduct joint experiments in a U.S. DOE laboratory with actual LWR spent fuel for the purpose of measuring the efficiency of recovery of volatile fission products (Xe, Kr, I, H-3, and oxides of carbon). DUPIC process experience will provide an extremely useful baseline process to support the successful development of the voloxidation process. The two parties will conduct independent experiments on the development of various trapping concepts for immobilizing the volatiles released in the course of the voloxidation treatment, with results to be shared in periodic information exchange meetings.

2004-004-K (continued)

The work scope of the proposed work consists of three major tasks.

The first task in this collaboration involves development of analytical methods and equipment to measure volatile fission product (Cs, Mo, Ru, etc) release behavior from the voloxidation 3 process. Experiments will be carried out jointly by all organizations and hot testing will be performed in ANL facilities.

A second effort will focus on assessing the recovery and trapping of gaseous fission products (Kr, Xe, I, H-3, C-14, etc). In order to complete this work, literature will be reviewed and laboratory scale testing will be performed to evaluate treatment of gaseous fission products and to select the appropriate unit operations for each fission product. Next, an off-gas treatment system for a voloxidizer will be designed, manufactured, installed in the facilities at ANL. Using the off-gas treatment system, hot experiments will be performed to assess the recovery and trapping of gaseous fission products from spent fuel. For design and experiment of an off-gas treatment system, KAERI staff will collaborate with the ANL research team and with the ORNL team.

Thirdly, process cycles will be developed to optimize fuel cladding separation and fuel particle size. In this task, equipment will be designed to support process testing, and a series of experiments will be performed using irradiated test material. One first goal of this effort will be to achieve complete separation of the fuel from the cladding. The second goal is to create a particulate material that may be readily handled in subsequent hot cell operations. Tests will focus on developing process conditions that will allow both of these objectives to be achieved. ANL will direct this effort with input requested from both KAERI and ORNL.

In summary, we propose to conduct collaborative research on the advanced voloxidation system that has a high recovery efficiency of fuel material from cladding, and effectively recovers volatile fission products (specially, long-lived nuclides such as I-129, Tc-99, C-14). This system may be employed in both aqueous and pyrochemical based fuel treatment processes.