DESIGN AND OPTIMIZATION OF SCREW CONVEYOR REACTOR SYSTEM FOR THE PYROLYSIS OF SPENT ION-EXCHANGE RESINS

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Abstract
A screw conveyor reactor system was developed for the thermal decomposition of radioactive spent ion-exchange resins. Kinetic analyses of the reaction steps involved in the thermal decomposition process of spent resins doped with radioactive metal surrogates were performed and the results were used to make kinetic predictions of the desirable and undesirable reaction steps. Optimum thermal treatment conditions were established based on the kinetic predictions and the thermodynamic radioactive metal volatility. The established optimum condition, which requires multistep heating and control of residence time under the specified gaseous atmosphere and temperature, was implemented and demonstrated using a lab-scale screw conveyor reactor system.

Keywords
Screw conveyor reactor, pyrolysis, spent resins, kinetic analysis, radionuclide volatility

Introduction
Commercial nuclear industry utilizes ion exchange resins to clean their process and process water. Over time these resins have to be disposed of. Owing to the high disposal cost and the shortage of available radwaste repository, developing a process the volumes of radioactive spent resins prior to disposal is an attractive goal from an economic perspective. Pyrolysis is considered a promising, safe approach for volume reduction of spent resins, one that does not release of semi-volatile radioactive elements such as cesium.

The volatilization of radioactive elements, as well as the thermal destruction of organics, is thermally stimulated process. Therefore, the temperature and residence time of the spent resins in the pyrolysis reactor should be properly controlled to retain semi-volatile radioactive metal species such as cesium while organics should be substantially destroyed. A novel reactor system, a screw conveyor reactor system with multiple temperature zones, was proposed for the implementation of this idea. To design and to optimize the screw conveyor reactor system, detailed kinetic analyses were performed and kinetic predictions were made for the important reaction steps involved in the pyrolysis process for spent resins.

Methods
Non-isothermal TG and FTIR analyses of spent resins doped with radioactive metal surrogates were performed to establish reaction steps involved in the pyrolysis process. Non-isothermal TG analyses with five different heating rates were then performed for the kinetic analysis and prediction of each reaction step. Thermodynamic equilibrium model analyses were performed to predict the volatility of cesium species during pyrolysis process of spent resins. The results of the kinetic prediction of the destruction of organics and the thermodynamic model

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analysis of the cesium volatility were used to design and optimize a screw conveyor reactor system for the safe pyrolysis of spent resins without volatilization of cesium species.

**Results and Discussions**

Dehydration, dissociation of functional groups and carbonization of remaining polymer were identified as the three major reaction steps involved in the pyrolysis of spent cation-exchange resins, as shown in Figure 1. Thermodynamic analysis reveals that cesium sulfate (Cs$_2$SO$_4$) is the only non-volatile cesium species at high temperatures (Yang et al., 2016). Therefore, all cesium species should be converted into Cs$_2$SO$_4$ prior to vaporization. The scheme of the desirable pyrolysis reaction process is shown in Figure 2. The major components in the spent resin pyrolysis system are C, H, O, N, and S. The fates of traceable amounts of radioactive elements are dependent on the compositions of major gaseous species such as O$_2$, CO$_2$, N$_2$, H$_2$O, SO$_2$, and NOx.

Since all the cesium species, including Cs$_2$O, Cs$_2$CO$_3$, CsOH and CsNO$_3$ but excepting Cs$_2$SO$_4$, are volatile at pyrolysis temperatures, all the cesium species should be converted into its sulfate before they reach the high-temperature region for the pyrolysis of organics. In a conventional incinerator or pyrolysis system, H$_2$O and NOx exist at high temperature region, where waste are burning or gasified; the step-wise processing of thermal treatment impossible. A screw conveyor reactor system, of which a schematic diagram is shown in Figure 3, was designed for the step-wise thermal processing of spent resins. This has three different heating zones.

**References**