Reaction System Design for NO_2 Oxidation of Used Nuclear Fuel for the FY17 Hot Test

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SUMMARY

A dry pretreatment process based on the oxidation of used nuclear fuel to convert it to a fine powder is being studied for the removal and capture of tritium and iodine before subsequent processing. The process converts oxide fuel into a fine powder at low temperature using NO_2/O_2 mixtures. The form of the powder product can be selected to be U_3O_8, UO_3, or a nitrate by adjusting the processing conditions.

All the fundamental tenets of the process have been successfully demonstrated as a proof of principle, and many aspects have been corroborated multiple times at laboratory scale. The present thrust is to develop the process to a technology-readiness level sufficient to evaluate and estimate the cost of an engineering-scale implementation. A previous roadmap analysis of the implementation determined that the most desirable approach would be based on kilogram-scale experiments using real fuel in parallel with multi-Kg testing of prototype systems using surrogate material.

Following the planned approach, kilogram-scale experiments will be conducted in FY 2017. This report describes the primary reaction system designs and the equipment that has been fabricated. Details related to instrumentation, data acquisition, and controls have been previously reported.

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FIGURES

ACRONYMS

B.P.	boiling point
ORNL	Oak Ridge National Laboratory

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1. Introduction

A dry head-end process for removal of tritium and iodine by NO_2 oxidation before dissolution has been studied at Oak Ridge National Laboratory (ORNL) for a number of years. The results of these studies have demonstrated the ability to convert the mostly monolithic UO_2 to U_3O_8, UO_3, or uranyl nitrate, all at temperatures below 350°C.[1] The rates of conversion to each of these products have been previously studied and reported.[1,2] Additionally, the complete release of iodine has been demonstrated with both unirradiated UO_2 pellets and gram quantities of irradiated commercial UO_2 fuel.[3] While the studies to date have shown significant potential for the process at laboratory scale, there remain a number of questions that must be addressed to more thoroughly assess the process for implementation at industrial scale.

Kilogram-scale experiments will be performed at ORNL in FY 2017 to resolve questions related to scaled performance and cost. The experimental system will utilize NO and O_2 to produce NO_2 reactant gas, and the system will regenerate NO_2 in situ from byproduct NO and excess O_2. The system will be operated in Building 3525 where O_2/air voloxidation demonstrations were performed for the Coupled End-to-End Demonstration Campaign. Testing will be performed with declad fuel to minimize variability. Initial steps previously reported include preliminary design of instrumentation, data acquisition, and controls[4]. This report details the completed equipment design and fabrication progress.

2. System and Equipment Design

The NO_2 reaction system with gas recirculation is depicted in Fig. 1. This schematic shows three primary in-cell reaction components, the reaction vessel, an intermediate reaction vessel for NO_2 generation, and recirculation pump.

The reaction vessel, shown in Fig.2, consists of a 15-in. long 4-in. diameter stainless steel tube with a conflat flange connection on the feed end to mate to a powder mixer, which is also shown in Fig 2. The vessel exit has a sintered 1.5-in. diameter stainless steel 20-micron frit to prevent powder entrainment. The vessel has a perpendicular port at approximately half the length to allow for loading of pellets and unloading of the product powder. The port tapers from 3 in. diameter at the vessel face to 1.5 in. The vessel can be rotated 180° after reaction for unloading into product bottles that will be threaded onto the port. During the reaction, the port will be capped. The mixer consists of helical ribbons attached to a 1-in. diameter stainless steel rod. The spacing of the ribbons to the outer wall was chosen to allow for powder mixing without binding of unreacted fuel pellets. The mixer drive has a controller and is operated at 0-100 rpm. The vessel is being altered to have thinner walls to reduce weight and the port funnel opening is being enlarged to increase the ease of product removal. Additionally, a thermocouple well is being welded to the surface that will extend ~75% of the length of the unit.

The recirculating pump is a bellows pump with gas compartments constructed of stainless steel with Teflon seals. The pump is rated to operate up to 150°C and will be heated to prevent NO_2 condensation in the N_2O_4 (B.P. 21.69°C). The "intermediate vessel" in Fig. 1 is used to mix the NO reaction byproduct and feed with the O_2 feed to make the NO_2 reactant and is to be a 10-in. diameter, 8-in. tall stainless steel heated container with a dip leg for the gas entrance.

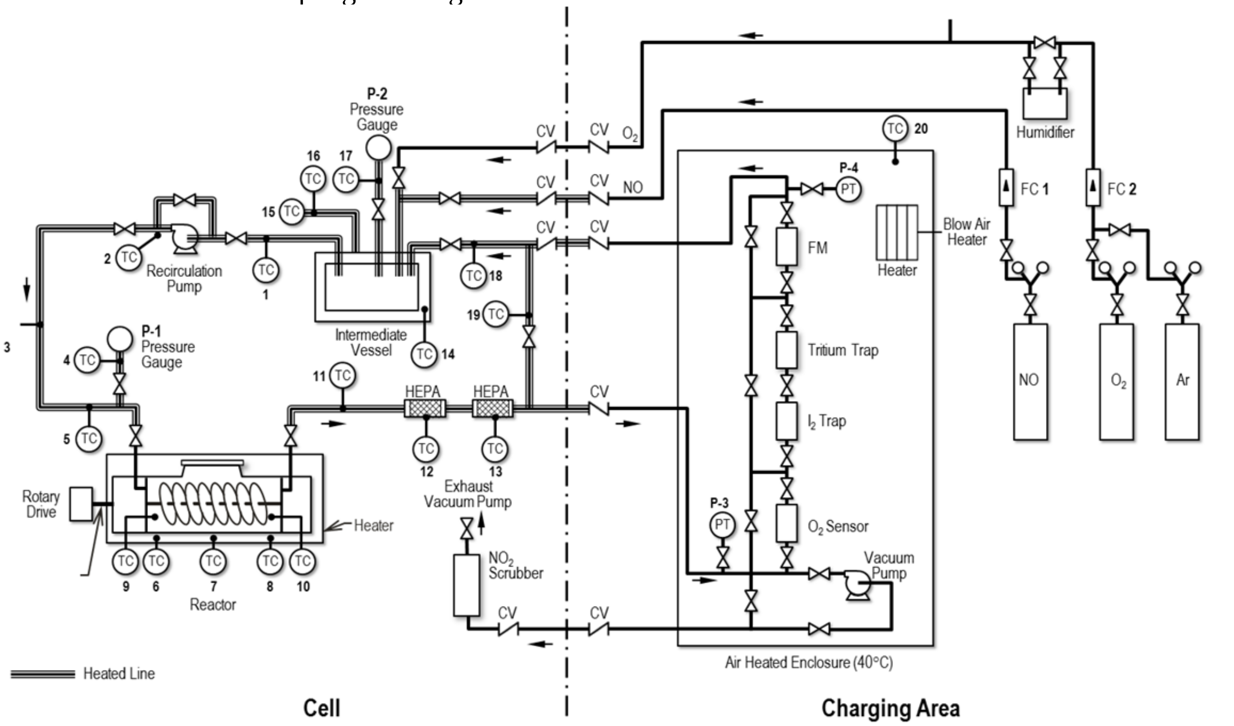

Fig. 1. Flowsheet for NO_2 treatment at ORNL's Building 3525.

Fig. 2. Primary reaction vessel and mixer.

To enable installation into the hot cells in Building 3525, the system has been designed to be compact, which required the housing for the reaction equipment to be designed and revised several times. The planned stand for the equipment is illustrated in Fig. 3. The primary reaction vessel runs perpendicular to the recirculation pump which is located below the reactor vessel on a tongue and groove removable sheet of aluminum. The primary reaction vessel and mixer rest above the stand to allow for easy manipulation. The intermediate vessel is housed on the opposite side of the base. The vessel will be removable, and the stand has a cylindrical lip to secure the vessel in place. The spacing of the recirculation pump and the intermediate vessel allows for the rotation of the reactor to empty product powder after reaction. The entire stand will be fabricated of aluminum for reduced weight for easier remote manipulation.

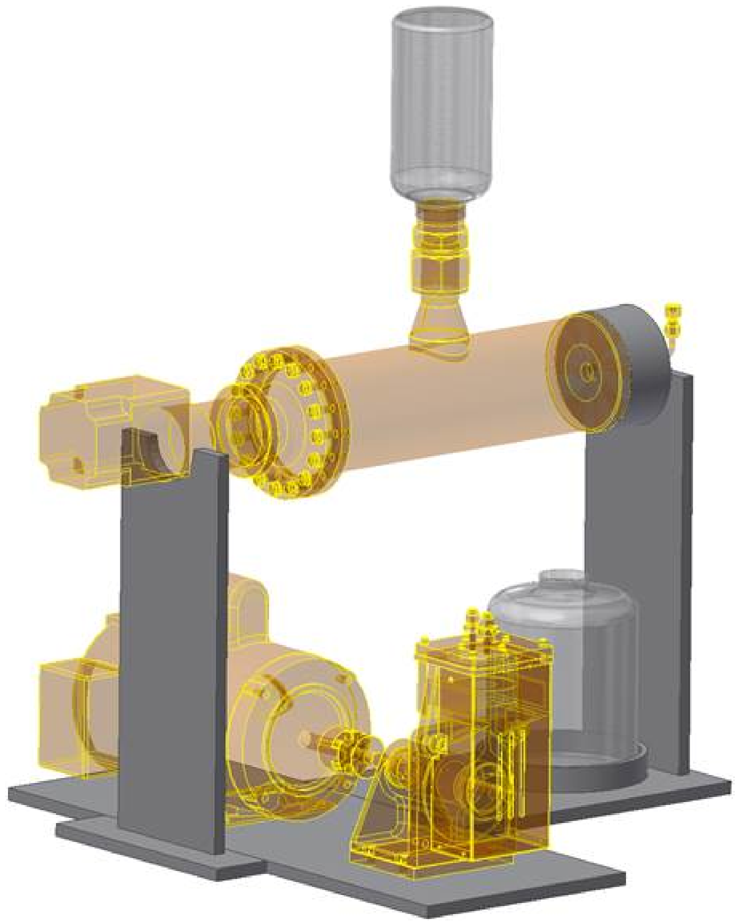

Fig. 3. Stand design with primary components present.

The next steps likely to be performed in FY17 are to include fabrication of the frame; procurement of heating elements, instrumentation, and control equipment; and cold testing of the system before installation. As a result of the modularity, simplicity, and maneuverability of the system, it will serve as a versatile and durable reaction unit.

References

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