

Scaled Up Process Report – Apparatus and Model



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Nuclear Energy and Fuel Cycle Division

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ABBREVIATIONS

CETE	Coupled End-to-End [program]
CSTR	continuous stirred-tank reactor
DAC	data acquisition
DOE	US Department of Energy
DOG	dissolver off-gas
GUI	graphical user interface
MFC	mass flow controller
ORNL	Oak Ridge National Laboratory
Simfuel	simulated used nuclear fuel
SPS	spark plasma sintering
UNF	used nuclear fuel

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ABSTRACT

Advanced voloxidation with NO_2 is a proposed process for used nuclear fuel head-end reprocessing scheme that converts UO_2 to higher oxides, and it also converts partitioning volatile fission products into the gas phase, thus facilitating fuel dissolution and actinide recovery. NO_2 voloxidation is being studied on small batches of UO_2 Simfuel, but the real test of process feasibility will be when it is scaled up to work with >100 g of irradiated material. This report discusses the aspects of scale-up that must be considered for NO_2 voloxidation, including development of a stirred reactor to promote agitation of the mixture during processing, online process monitoring, and automation controls. Brief details on parallel efforts are also provided in this report, including (a) demonstration of iodine release from Simfuel made by Spark Plasma Sintering, and (b) development of an order-of-magnitude scale-up to react 100 g of UO_2 Simfuel in a metal reactor.

This report fulfills the work package Aqueous Separation Science & Novel Processes – ORNL, milestone M3FT-23OR0304020115, “Scaled Up Process Report – Apparatus and Model.”

1. BACKGROUND

1.1 VOLOXIDATION HISTORY AND PROJECT OBJECTIVES

Previous efforts with voloxidation used air/oxygen or ozone. This includes efforts during the Coupled End-to-End (CETE) processing program in which North Anna and Surrey-2 fuels were processed. A pilot-scale voloxidizer was designed and constructed for this use, and the used nuclear fuel (UNF) was processed in the Irradiated Fuels Examination Laboratory, a category 2 hot cell facility in Building 3525 at Oak Ridge National Laboratory (ORNL). This voloxidizer had issues with generating powder, e.g., entrainment, and total metal dissolution post-voloxidation proved to be time intensive. These issues were a result of the temperature-dependent particle size distributions and an increase in insoluble noble metals observed when using H. B. Robinson-2 fuel. Additionally, standard voloxidation liberated a significant quantity of volatile fission products (Spencer et al. 2007).



Figure 1. Pilot-scale voloxidizer for standard voloxidation use during ORNL’s Coupled-End-To-End program. Credit: Oak Ridge National Laboratory.

Enhanced voloxidation using ozone was investigated to address particle size issues, because the ozone was determined to generate a finer UO_3 powder. The fine powder alleviated diffusion-based evolution of volatiles when heated to higher temperatures, even with further oxidation of UO_3 to U_3O_8 because the fine powder morphology remained after oxidation. Coupling the use of ozone with steam allowed for removal of all volatile fission products, along with some of the semivolatiles (noble metals), thereby simplifying downstream separations processes for an aqueous acidic dissolution. One significant benefit of the enhanced voloxidation is the removal of technetium (Tc), which would simplify uranium separation by eliminating the need to co-extract it with Tc under low-acid conditions (Spencer et al. 2007).

Advanced voloxidation using NO_2 is being considered for reprocessing of used nuclear fuel (UNF) because it has the potential to simplify the overall process. One of the three main advantages is that voloxidation can be used to separate the fuel from the cladding before dissolution and recovery of actinides. The second recently investigated aspect is the simplification of the off-gas capture of volatile radionuclides by trapping them before fuel dissolution. This latter aspect has been the target of recent studies, with a focus on the fission product, iodine (Greaney et al. 2022). The third advantage is based on the fact that concentrated nitric acid is not needed for the downstream dissolution. Elimination of nitric acid reduces the requirement for HNO_3 and simplifies the dissolver off-gas system (DOG).

Early advanced voloxidation testing has been studied at Oak Ridge National Laboratory (ORNL) since 2012, and results show that U_3O_8 was easily oxidized to UO_3 at 225°C under 3:1 ratio of NO_2/O_2 , (Johnson et al., 2017), demonstrating the proof of concept (Del Cul et al. 2015, Johnson 2013, Johnson et al. 2012, Johnson et al. 2017, Rudisill et al. 2019). Recently, optimization studies of the voloxidation reaction at bench scale were pursued with two objectives: (1) to create and characterize realistic Simfuel, and (2) to quantify the capture of iodine and other semivolatile species during advanced voloxidation. Furthermore, optimization of the voloxidation reaction was pursued by studying the effects of iodine capture for CsI under different temperatures, reaction run time, purge time, solid-gas ratio of ($\text{NO}_2\text{-CsI}$), solid particle size, and multiple NO_2/O_2 charges, resulting in 0% to 95% iodine partitioning into the off-gas. The findings of these recent studies resulted in a process acumen with the highlights detailed below. This drove decisions in the development of a reactor for scaled up voloxidation experiments described herein.

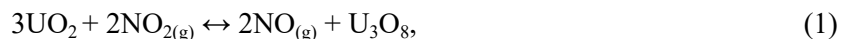
- Pellet development to produce idealized grain structure and more homogenous distribution of fission products is required to quantify voloxidation of iodine under conditions similar to those reported for UNF processing
- Solid-gas reaction at the reaction front is promoted by mechanical agitation to penetrate the diffusion barrier created by reacted material
- Multiple purges of the system with inert gas with subsequent recharges of fresh NO_2/O_2 may help volatilize iodine in near quantitative yield
- Recirculation through an iodine sorbent bed results in the consumption of the reagent needed for the voloxidation process (NO_2) because of the reaction with silver-based iodine sorbents
- Surface area / particle size, temperature, and volume of reactive gas are all necessary factors that must be incorporated when modeling process reactivity

Although voloxidation has also been considered for separating UNF from Zircaloy cladding, other research efforts are examining other reprocessing steps, such as advanced chlorination and off gas management, with great success. When considering a scaled up reactor design, it is advantageous to refocus this project's scope using this new body of knowledge to better inform design decisions and

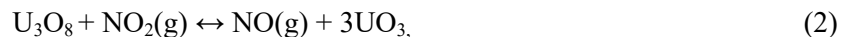
advanced voloxidation applicability within a larger multistep process. Therefore, this report is transitioning from an isolated or stand-alone method to an approach that will build a state-of-the-art system that is fully integrated into a multistep reprocessing flowsheet. This project will also include development of a comprehensive nuclear chemical process model for the voloxidation of UO_2 (uranium dioxide) to UO_3 (uranium trioxide) conversion in used nuclear fuels. The resulting model will provide valuable insights for process optimization and reactor design.

1.2 VOLOXIDATION CHEMISTRY

The NO_2 voloxidation process presented here comprises two oxidation steps to convert UO_2 to UO_3 , as shown in Eqs. [1] and [2]. First, UO_2 is oxidized to U_3O_8 using NO_2 at temperatures between 350°C and 500°C , as seen in Eq. (1).



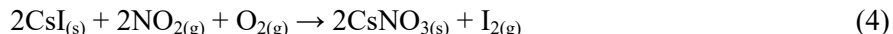
Then, the temperature of the system is lowered to within a range from approximately 200°C to 250°C , and the U_3O_8 is oxidized to UO_3 using NO_2 , as shown in Eq. (2).



During each reaction step, NO_2 is consumed in the reaction, and NO is left as a product in the gas phase. The NO_2 can be regenerated for continued reaction by adding O_2 into the system, as shown in Eq. (3).



If voloxidation is used for UNF, then the release of volatile fission products such as iodine can also be observed. The fraction of the iodine released may depend on temperature (favored under 700°C) and available oxidizing agent over other competitive reactions.



2. SCALED UP REACTOR

The DOE-NE Materials Recovery and Waste Form program develops methods to prepare UNF for final disposal, including separation of actinides and fission products. In voloxidation, the UO_2 is converted to UO_3 for direct dissolution. The process has been demonstrated on a small scale, but the program goal is to demonstrate voloxidation production capacity. Translation of the benchtop experiments to a semipilot scale enables identification of scaling issues that could be detrimental to pilot-scale experiments that were not properly planned. Therefore, scaling the reactor design to facilitate larger material quantities allows for higher throughput, which is critical to meet increasing production requirements efficiently, to verify that the process is scalable, and to overcome any unforeseen challenges not observed at the bench scale. It is anticipated that efficiency will be improved by optimizing reactor parameters, resulting in reduced energy consumption and operating costs, making the production process more sustainable.

Several options are available for scaling up the reactor design for voloxidation. Selection of the best options depends on factors such as budget, space availability, and process requirements. Some potential options are detailed below:

1. **Batch reactors:** scaling up existing batch reactor designs to increase production capacity while maintaining the current process setup

2. **Continuous flow reactors:** transitioning to continuous flow reactors for a more efficient and consistent production process
3. **Parallel reactors:** implementing multiple reactors in parallel to accommodate higher production demands without significantly altering existing reactor design

The choice of the scaled up reactor design should be made after a comprehensive feasibility study is conducted that considers the project's specific needs and constraints. Additional considerations include coupling of advanced voloxidation to alternate chlorination and direct dissolution. Given the desire to demonstrate all three tasks sequentially, the voloxidation reactor design can be informed by needs for alternate chlorination—another corrosive process—and direct dissolution. The alternate in which the other programs are informed by needs for advanced voloxidation to ensure compatibility is also preferable, whether a single system is used for all three processes, or the other systems are incorporated around the advanced voloxidation system.

2.1 ENGINEERING DRAWINGS AND GLASS REACTOR FABRICATION

Significant progress has been made in the project's engineering phase. Detailed engineering drawings have been successfully developed for the scaled up reactor that incorporate the key aspects discussed in this report. These drawings serve as a foundation for the construction phase. The reactor has been accurately measured, with specifications of a 1,000 mL flask, a cylindrical shape, and a 100 mm Duran flange. Furthermore, substantial progress has been made in the fabrication of small glass reactors using the facilities available at ORNL's glassblowing shop. These small glass reactors represent a critical step in testing and validating reactor design concepts. Figure 1 and Figure 2 depict the first step in reactor scale-up, the considered intermediate scale reactor, and the final metal reactor, along with the components of the first-step reactor.

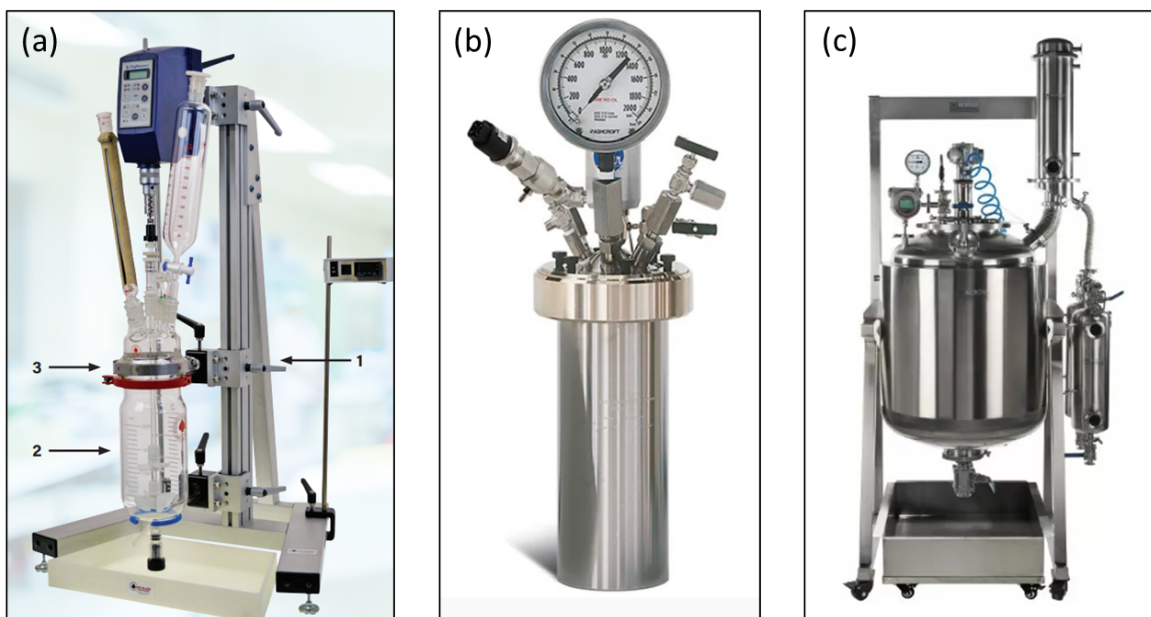


Figure 2. (a) The current reactor, excluding the metallic upper section below the reactor, (b) the proposed intermediate reactor design under consideration for process optimization, and (c) the metallic reactor selected for the final stage of development.



Figure 3. (a) Various sections of the first step scaled up reactor, (b) motorized stirrer, pressure transducer, and data collection system, and (c) the stand, clamp, and rotary feedthrough.

The glass reactor will be equipped with continuous stirrers and other necessary components to replicate the key features of the scaled up design. This intermediate step will include initial testing and optimization under controlled conditions. The implementation of a septum option for sample collection and sampling allows for periodic sampling of reaction products without interrupting reactor operation. LabView sensors will be incorporated as the automation proceeds as described in the following section. Automation and a sample port will allow for periodic reaction modeling of the process without disassembling the reactor or stopping the process. Following successful testing with the stirred glass

reactors, the process will transition to development of a metal reactor to provide the durability and reliability required for sustained UO_3 production in a shielded facility.

A 5 L reactor has been purchased for analysis of the alternate chlorination process. This reactor may be viable for use as a continuous batch reactor for testing voloxidation. The schematic of the system is shown in Appendix A. Details of this reactor are included in the Alternate Chlorination project data; however, the 5 L reactor is a continuous stirred tank reactor with a Teflon stir vane, a bottom-draining valve to remove liquids or powders, and a multiport glass lid to facilitate the multiple inlet/outlet connections required for the process. Although it is not directly applicable at this time, the base unit will be available for voloxidation when it is not in use for alternate chlorination. Future development of alternate chlorination reactors will be informed by the advanced voloxidation systems.

3. AUTOMATION

Automation supports the scaled up process by streamlining and expediting critical tasks. It enhances the efficiency and accuracy of a process, reduces human error and intervention, and allows for consistent, controlled experimentation. This approach not only accelerates advanced voloxidation into the state-of-the-art reactor designs for online monitoring, but it also enables the collection of larger volumes of data and experiments, ultimately facilitating faster and more cost-effective research and development, thus streamlining the route to commercialization.

3.1 SENSOR AND MONITORING DEVICE REQUIREMENTS

Monitoring devices employed in the manifold in past studies (Greaney et al. 2023) include pressure transducers (MKS model #722B53TGA2FJ) and thermocouples (Omega model #SA3-K-SRTC) to monitor reaction pressure and temperature respectively. The existing manifold was also connected to a glass reactor, and decisions on reaction progression and completion were largely based on powder/pellet color. However, stainless-steel reactors are ultimately favorable in process scale-up, thus resulting in the need to equip reactors with an optical transmission sensor such as the ColorMax probe.¹

3.2 LABVIEW PROGRAM

Implementation of the LabVIEW Data Acquisition System (DAC) supports the desired scale-up work with its flexible, user-friendly platform and its capability to support design and automation of laboratory experiments and measurements. The software's programming interface is graphical, allowing for easy integration with various instruments and devices, automation of data acquisition, and analysis of results in real time. LabVIEW's scalability and modular design allow for expansion to accommodate the growing, complex needs associated with process scale-up. It is a valuable tool for efficiently managing and accelerating advanced voloxidation into process scale. Furthermore, remote monitoring and safety interlocks for enhanced efficiency and safety are included.

¹ <https://www.emxindustrialsensors.com/product/colormax-1000-color-sensors/>

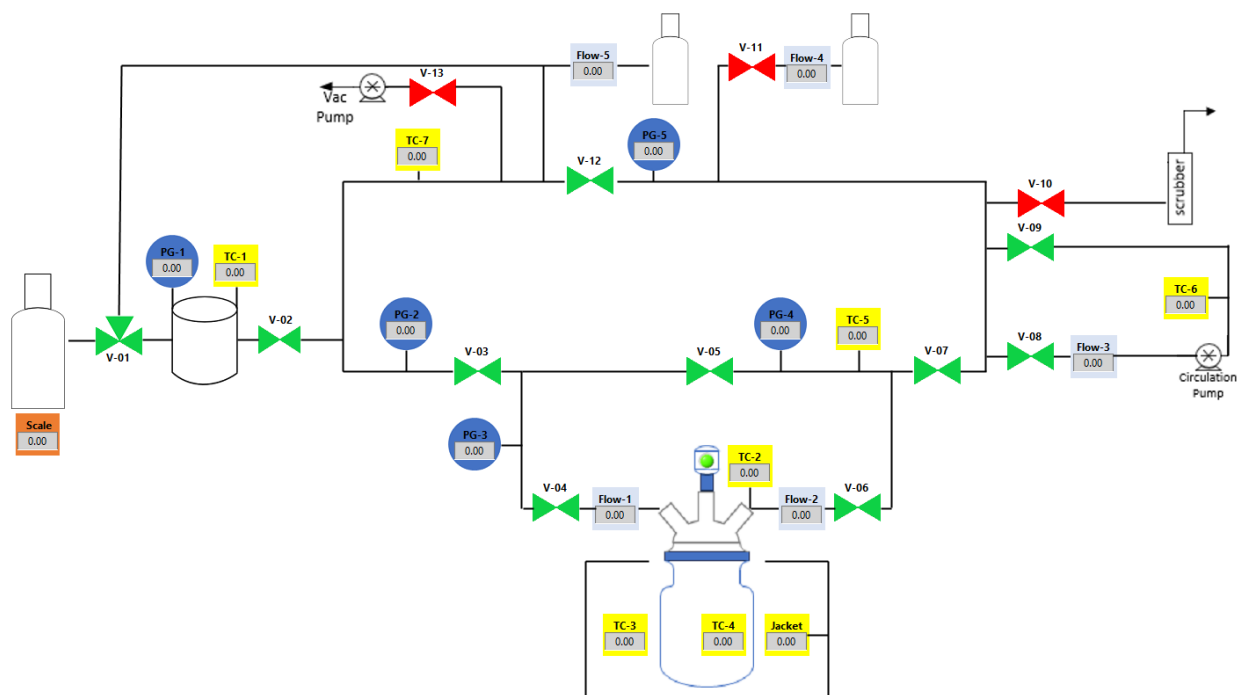


Figure 4. LabVIEW diagram showing automated valves and detectors added to the voloxidation scaled up reactor design and manifold.

LabVIEW DAC will be further developed and fine-tuned for the metal reactor. It will play a central role in gathering and processing data from the scaled up reactor, facilitating real-time adjustments, and ensuring the safety and efficiency of the production process. Sensor integration will continue as part of the transition to a metal reactor, including online monitoring of reactor progress, as well as pressure sensors, color monitoring sensors, flowmeter sensors, and temperature sensors. These sensors will be integrated into the reactor's design to ensure precise control and monitoring of the voloxidation process.

4. PROCESS MODELING

DWSIM, an open-source chemical process simulator that is designed to adhere to the CAPE-OPEN standards. It is compatible with Windows, Linux, and macOS operating systems. Powered by Microsoft .NET and Mono Platforms, DWSIM offers a user-friendly graphical user interface (GUI) and encompasses advanced capabilities, including precise thermodynamic calculations, support for chemical reactions, and hypothetical component generation.

DWSIM can simulate a wide range of processes, including a solid–vapor reaction such as voloxidation. It accomplishes this by including models for species thermodynamics and unit operations. This project includes investigation of existing tools, as well as extension of DWSIM to study the UO_2 -to- UO_3 voloxidation process. This project also includes development of custom programs for oxidation reactions, as well as adjustment of reactants and products based on the best-available literature on relevant properties in this field.

4.1 MODELING PARAMETERS

Physical and chemical properties of reactants (UO_2 , NO_2 , O_2 , and CsI) and products (UO_3 , CsNO_3 , NO , and I_2) are required for accurate modeling. Some properties are readily accessible in the DWSIM program

library, such as CsI, NO₂, NO, CsNO₃, and I₂. However, the data on UO₂ and UO₃ shown in Table 1 were added to the library.

Table 1. Physical and chemical properties used as inputs for UO₂ and UO₃ into DWSIM (Cordfunke and Konings 1990)

Species	Molecular weight (kg/kmol)	X-ray density (g/cm ³)	S° (J/mol*K)	DH _f ° (kJ/mol)	Cp (J/mol*K)	DG _f ° (kJ/mol)
UO ₂	0.27002	10.95	77.03 ± 0.20	-1,085.0 ± 1.0	63.597	-1,031.868
U ₃ O ₈	0.8421	8.396	282.55 ± 0.50	-3,574.8 ± 2.5	238.957	-3,369.616
g-UO ₃	0.28629	7.99	96.11 ± 0.40	-1,223.8 ± 1.0	81.670	-1,144.791
e-UO ₃	0.28629	n.f.	n.f.	-1217.2 ± 1.3*	n.f.	n.f.

*(Grenthe et al. 2020); n.f. = not found

An example of implementation of process reactions in DWSIM is shown in Figure 5, illustrating successful implementation of Eq. (4) presented above in Section 1.2.

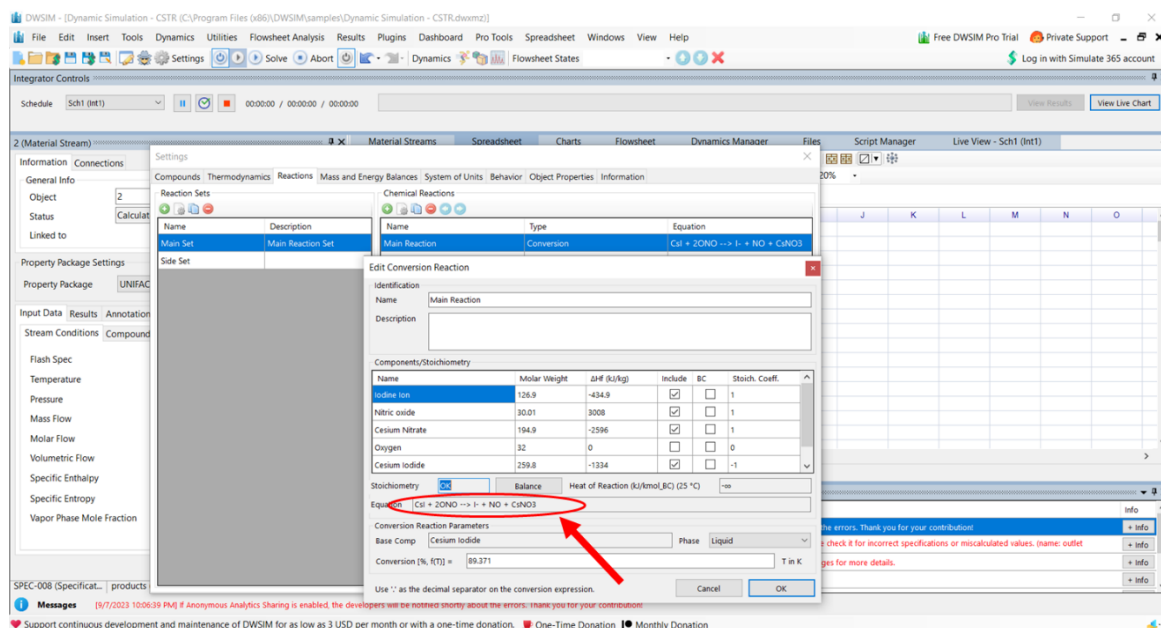


Figure 5. Screenshot showcasing the configuration of CsI-to-I₂ conversion reactions within the DWSIM interface.

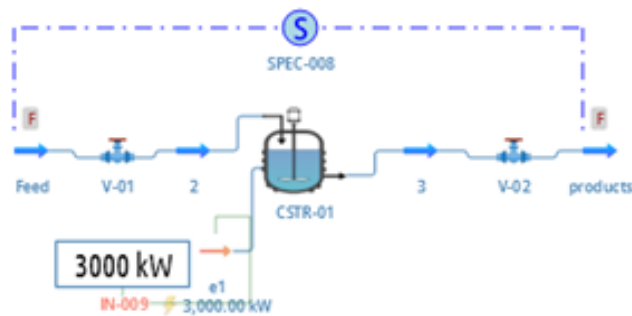


Figure 6. Schematic outlining the selected continuous stirred-tank reactor (CSTR) for conducting reaction process modeling.

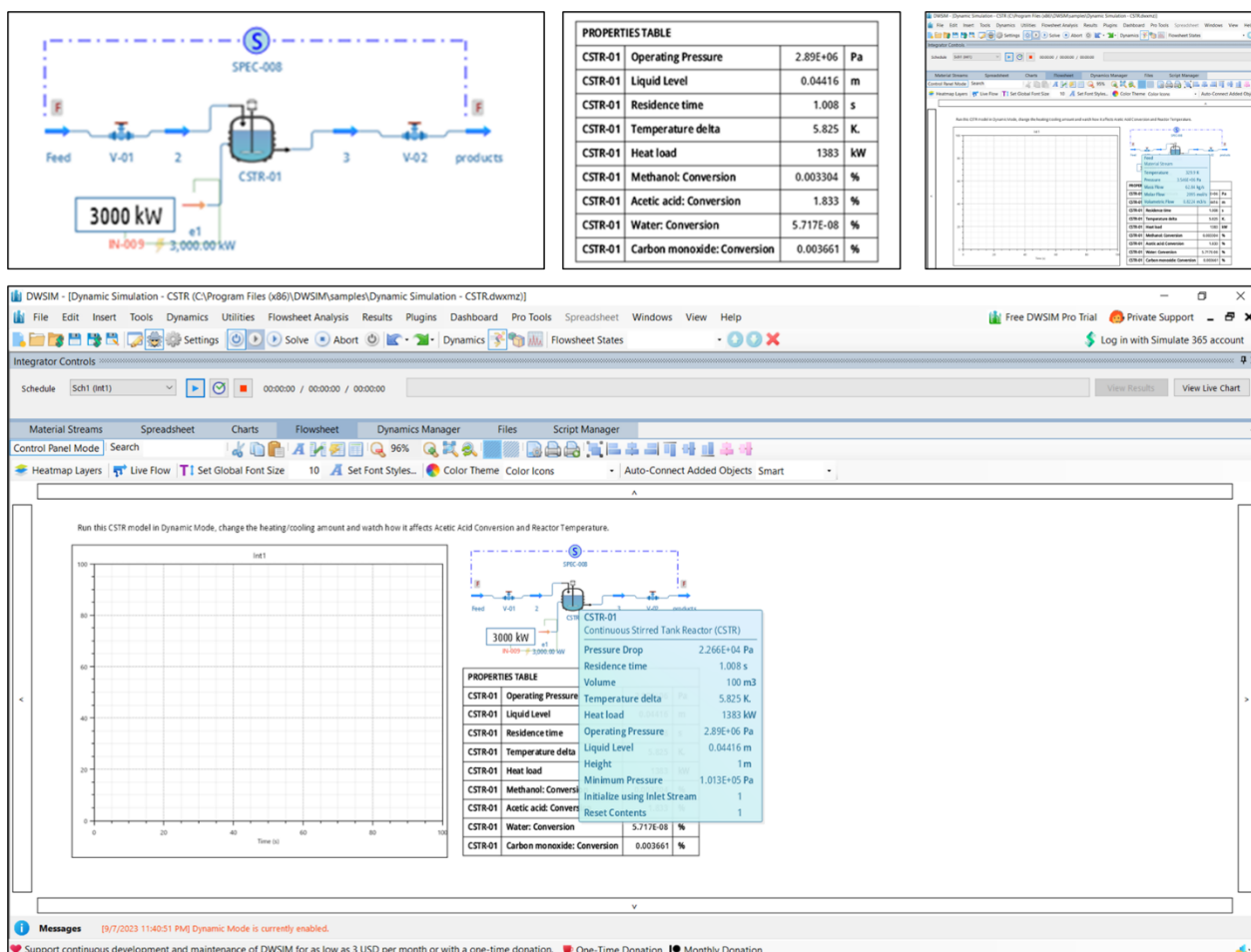


Figure 7. CSTR reactor schematic, control parameters, reactor feed parameters, and condition descriptions—all essential information for reaction process modeling.

4.2 NEXT STEPS

The next phase in developing a suitable model for advanced voloxidation focuses on refining kinetic equations, extending the model validation, and exploring integration with reactor design workflows as outlined below:

1. **Refinement of Kinetic Equations:** Further refine the kinetic equations by incorporating additional experimental data and exploring alternative reaction mechanisms. Experimental parameters such as particle size will be included.
2. **Complex Feed Stream Validation:** Extend the model validation to complex feed streams, simulating real-world scenarios encountered in nuclear fuel reprocessing, such as the impacts of water vapor and adsorbed water.
3. **Reactor Design Integration:** Integrate the voloxidation model and reactor design to process optimization workflows. Other upstream and downstream processes will be included.
4. **Multiscale Modeling:** Explore multiscale modeling approaches to connect microscopic particle-level kinetics with macroscopic process behavior.

The test matrix provided below (Table 2) includes future tests focusing on Eqs. (2) and (4) provided in Section 1.2. Each test can be further customized by adjusting parameters and criteria based on project development.

Table 2. Test matrix for UO₂-to-UO₃ voloxidation and CsI partitioning reactions

Test ID	Test description	Objectives	Parameters	Expected results	Success criteria
1	Kinetic equation refinement - UO ₂ -to-UO ₃	Refine kinetic equations for the UO ₂ -to-UO ₃ conversion reaction based on additional experimental data	Reaction rates, temperature, pressure, species concentrations	Improved model accuracy and agreement with experimental data for the UO ₂ -to-UO ₃ conversion	Model predictions within 10% of experimental values
2	Kinetic equation refinement - CsI-to-I	Refine kinetic equations for the CsI-to-I conversion (fission) reaction based on additional experimental data	Reaction rates, temperature, pressure, species concentrations	Improved model accuracy and agreement with experimental data for the CsI-to-I conversion	Model predictions within 10% of experimental values
3	Complex feed stream validation	Extend model validation to complex feed streams with varying compositions for both UO ₂ -to-UO ₃ and CsI-to-I reactions	Feed composition, temperature, pressure	Accurate model prediction of UO ₂ -to-UO ₃ conversion, CsI-to-I conversion, and oxygen consumption for complex feed streams	Model predictions within 10% of experimental values for both reactions
4	Reactor integration	Collaborate with reactor design experts to integrate the voloxidation model into reactor design and optimization workflows	Integration with reactor design software, process optimization	Successful integration and compatibility with reactor design tools for both UO ₂ -to-UO ₃ and CsI-to-I reactions	Seamless interaction and consistent results between the voloxidation model and reactor design software for both reactions
5	Multiscale modeling - UO ₂ -to-UO ₃	Explore multiscale modeling to connect microscopic particle-level kinetics with macroscopic process behavior for the UO ₂ -to-UO ₃ conversion	Particle-level kinetic parameters, reactor conditions	Development of a multiscale modeling approach that provides insights into UO ₂ -to-UO ₃ conversion behavior at different scales	Successful linkage between microscopic and macroscopic modeling results for the UO ₂ -to-UO ₃ conversion
6	Multiscale modeling - CsI-to-I	Explore multiscale modeling to connect microscopic particle-level kinetics with macroscopic process behavior for the CsI-to-I conversion	Particle-level kinetic parameters, reactor conditions	Development of a multiscale modeling approach that provides insights into CsI-to-I conversion behavior at different scales	Successful linkage between microscopic and macroscopic modeling results for the CsI-to-I conversion

5. CONCLUSION

The scaling up of the reactor design for an advanced voloxidation process is a critical step to meet increased production demands efficiently and safely. This report has outlined the reasons for scaling up, the available options for reactor design, and the automation aspects, including sensor requirements and LabVIEW implementation. Successful implementation of these elements will contribute to a robust and effective scaled up process for UO_3 production. Ongoing efforts will demonstrate iodine release from Simfuel made by Spark Plasma Sintering (SPS) as preliminary results indicate that the NO_2 voloxidation kinetics are dependent on the morphology of the starting material and grain size of the powders. These pellets are also kept in a desiccator before reaction because adsorbed H_2O can affect the kinetics of the voloxidation process. The next steps in this process are to use these pellets in an order-of-magnitude scale-up to react 100 g of UO_2 Simfuel in a metal reactor.

6. REFERENCES

- Cordfunke, E. H. P., & Konings, R. J. M. (1990). *Thermochemical Data for Reactor Materials and Fission Products*. Elsevier Science Publishers.
- Del Cul, G. D., Hunt, R. D., Spencer, B. B., & Jubin, R. T. (2015). *The Effects of Process Variables on the Nitration of UO_3* . (ORNL-SPR-2015-221).
- Greaney, A., McFarlane, J., Johnson, K. R., Peruski, K., Hunt, R. D., Vick, M., & Cobble, C. (2023). *Quantify Mass Balances of Semi-Volatile Species during Advanced Voloxidation Conditions* (ORNL/TM-2023/3019).
- Greaney, A., Walker, T., McFarlane, J., Johnson, K. R., Hunt, R. D., Vick, M., & Cobble, C. (2022). *Surrogate Testing for NO_2 Voloxidation and Direct Dissolution* (ORNL-TM-2022-2685).
- Grenthe, I., Gaona, X., Plyasunov, A. P., Rao, L., Konings, R. J. M., Smith, A. L., & Moore, E. E. (2020). *Second Update on the Chemical Thermodynamics of U, No, Pu, Am, and Tc*.
- Johnson, J. A. (2013). *Studies of Reation Processes for Voloxidation Methods*, University of Tennessee, Knoxville.
- Johnson, J. A., Del Cul, G. D., Spencer, B. B., Collins, E. D., & Hunt, R. D. (2012). *Advanced Voloxidation with NO_2 and Tests with Used Nuclear Fuel* (FCRD-SWF-2012-000105).
- Johnson, J. A., Rawn, C. J., Spencer, B. B., Meisner, R. A., & Del Cul, G. D. (2017). "Oxidation Kinetics For Conversion of U_3O_8 to $\epsilon\text{-UO}_3$ with NO_2 ." *Journal of Nuclear Materials*, 490, 211–215.
- Rudisill, T. S., Shehee, T. C., J, ones, D. H., & Del Cul, G. D. (2019). "Dissolution of Used Nuclear Fuel Using a Tributyl Phosphate/N-Paraffin Solvent." *Separation Science and Technology*, 54(12), 1904–1911.
- Spencer, B. B., Del Cul, G. D., & Collins, E. D. (2007). *Standard Voloxidation and Potential Benefits of Enhanced Voloxidation* (not published).

**APPENDIX A. ENGINEERING DRAWING OF THE 5L
ALTERNATE CHLORINATION REACTOR**

APPENDIX A. ENGINEERING DRAWING OF THE 5L ALTERNATE CHLORINATION REACTOR

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