Integrated Off-Gas System

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Materials Recovery and Waste Form Development

A Preconceptual Design of an Integrated Off-Gas Treatment System

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ABSTRACT

The U.S. has a vested interest in the advancement of nuclear energy to achieve aggressive net-zero goals, with reprocessing and recycling of used nuclear fuels (UNF) playing a vital role. It will not be possible to meet U.S. regulatory requirements without robust off-gas treatment, so it is crucial to advance treatment technologies to facilitate the design of future reprocessing facilities. For many years, teams of researchers across the U.S. Department of Energy (DOE) National Laboratory complex have been investigating off-gas treatment technologies for the capture and removal of volatile radionuclides (i.e., ^{85}Kr, Xe, ^{14}C, and ^{129}I) and oxides of nitrogen (NO_X) that are produced from reprocessing. These investigations have been focused on developing individual technologies for the capture of Kr, Xe, iodine, and CO_2. Capture technologies for each constituent were tested independently from one another by utilizing nonradioactive surrogates to simulate simplified off-gas streams. The tests have been relatively small, laboratory-scale experiments of up to approximately 1 L/minute total gas flow rate.

To increase the readiness of these technologies for deployment, an integrated test system with a larger-scale capacity is needed to bridge the gap between promising bench scale and fully scalable UNF reprocessing off-gas treatment. This document contains the goals, design basis, functional requirements, preconceptual design, and cost estimates for an integrated off-gas demonstration system for the capture and removal of NO_x, Kr, Xe, CO_2, and iodine at 10× higher throughput than earlier laboratory studies. The order-of-magnitude cost estimate for this system is approximately $886,000. Next phases include conceptual design, detailed design, fabrication, and commissioning.

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ACRONYMS

INL	Idaho National Laboratory
NOx	oxides of nitrogen
ORNL	Oak Ridge National Laboratory
P&ID	piping and instrumentation diagram (P&ID)
UNF	used nuclear fuel

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A Preconceptual Design of an Integrated Off-Gas Treatment System

1. INTRODUCTION

For many years teams of researchers across the U.S. DOE National Laboratory complex have been investigating off-gas treatment technologies for the capture and removal of volatile radionuclides (i.e., ^{85}Kr, Xe, ^{14}C, and ^{129}I) and NO_X that are produced from the reprocessing of UNF. These investigations have been focused on developing individual technologies for the capture of Kr, Xe, iodine, and CO_2. Capture technologies for each constituent were tested independently from one another by utilizing nonradioactive surrogates to simulate simplified off-gas streams. The tests have been relatively small laboratory-scale experiments of up to approximately 1 L/minute total gas flow rates. The next step to increase the readiness of these technologies for deployment is to incorporate them into an integrated test system with a larger-scale capacity. Constructing a modular, flexible test system for this purpose will allow testing of current state of the art technologies, and provide a pathway for advancing future promising technologies.

Previous efforts provided baseline data informing the preconceptual design proposed in this work and its integration into relevant systems. In 2009, Oak Ridge National Laboratory conducted a complete, coupled end-to-end demonstration of advanced nuclear fuel reprocessing in support of the Advanced Fuel Cycle Initiative.[1] This small-scale reprocessing operation provided a unique opportunity to test integrated off-gas treatment systems designed to recover the primary volatile fission and activation products (^{3}H, ^{14}C, ^{85}Kr, and ^{129}I) released from UNF. This, along with a 2016 engineering study, provided valuable information for a preconceptual design of an integrated off-gas treatment system for further testing.

This document contains the goals, design basis, functional requirements, preconceptual design, and cost estimates of an integrated off-gas demonstration system for the capture and removal of NO_x, Kr, Xe, CO_2, and iodine at 10× higher throughput than previous laboratory studies.

2. GOALS

A mobile, integrated off-gas system at 10× larger scale than previous laboratory testing will provide a flexible off-gas test bed that can demonstrate a wide range of off-gas capture technologies for target constituents from a UNF-reprocessing off-gas stream. This effort will define the functional requirements of each target technology, including gas conditioning, sizing (i.e., footprint), temperature control, materials of construction, peripheral equipment, process control, and analytical needs.

The testing of individual components for UNF-reprocessing off-gas treatment is valuable and necessary. An important aspect of this work is quantifying the impact on capture efficiency caused by minor constituents present in the gas stream due to imperfect upstream components. To determine how those individual components interact with each other, assumptions and compromises must be made with varying degrees of proximity to reality. Keeping in mind that the goal of the off-gas research campaign is to continue to progress to higher technology readiness levels to support the eventual UNF reprocessing in the U.S., it becomes especially important to couple individual technologies in a modular system, ensuring that upcoming technologies can be compared in a meaningful way to the current state of the art. To that end, overarching goals of this mobile, modular system are as follows:

- Scale up testing between 4–10× for technologies with sufficient bench-scale data to support inclusion in larger tests.

- Incorporate a modular design as much as is practical to support the inclusion of new technologies as they arise and become competitive with current treatment methods.
- Utilize sufficient instrumentation for thorough measurement and control to ensure technology comparisons are meaningful.
- Maintain a system size small enough to fit inside a Conex box for ease of transport.

3. DESIGN BASIS

The design of an integrated off-gas system requires definition of the target off-gas composition, the desired throughput (i.e., flowrate), and the optimal footprint of the entire system. The off-gas composition will be an aqueous-based UNF-reprocessing scheme. Further details are provided in section 3.1. A throughput of 10 L/minute total off-gas flow serves as the target throughput and is used to determine column sizing, sorbent masses, and equipment requirements. The skid mounted system footprint is subject to the dimensions of a 20-ft-long cargo container which is ideal for shipping purposes. The target treatment technologies of the system will focus on NO_x, Kr, Xe, CO_2, and iodine (see section 3.2). Initially, it is envisioned that nonradioactive surrogates will be used to create the off-gas stream utilized for testing target capture technologies.

3.1. Representative Off-Gas Composition

A detailed material balance for UNF off-gas and the expected outcomes from treatment unit operations was developed in 2016. This report uses those material balance calculations as a basis, scaled down to meet the flow requirements of this smaller system. Table 1 provides the anticipated nominal composition of the dissolver off-gas to be processed by the integrated system.

Table 1. Estimated dissolver off-gas composition scaled to the integrated test-bed throughput.

Constituent	Concentrations to Off-Gas System (volume %)
Dry Air	95 – 98
NO	0.04 – 1.0
NO_2	0.08 – 2.0
CO_2	0.001 – 0.004
H_2O	2 – 3
Iodine	0.001 – 0.003
Xenon	0.08 – 0.15
Kr	0.005 – 0.02

3.2. Targeted Capture Technologies

Figure 1 presents the capture technologies that are to be included in the integrated off-gas system. The off-gas will enter the system's iodine beds first and continue through each of the other beds until finally exiting the system after the Kr capture beds. Water will have to be removed from the gas stream following the NO_x scrubber. In Figure 1, the options for capture techniques under consideration are located below the individual target constituents. Wet scrubbing and cryogenic distillation are considered the most mature capture techniques, but they are not a focus in this effort. An exception to this is the wet scrubbing of NO_x species. Another mature method to accomplish NO_x removal is with catalytic reduction, but such a system requires high temperatures (>600°C) and the use of hydrogen to operate, which would not be conducive to the size limitations of this entire system. This is why wet scrubbing of NO_x is the most viable option in the proposed preconceputal design.

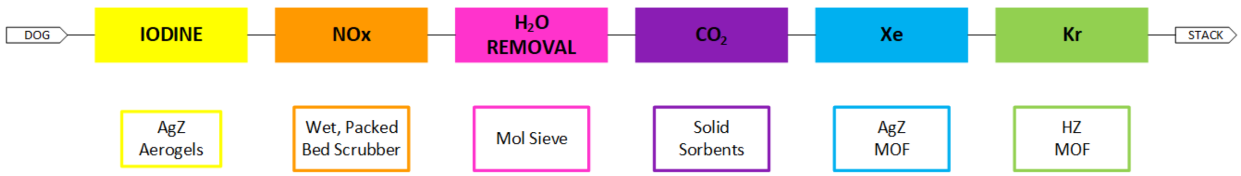

Figure 1. Targeted capture technologies and capture options (DOG for "dissolver off-gas," AgZ stands for "silver mordenite sorbent," MOF for "metal-organics framework" and HZ stands for "hydrogen mordenite sorbent").

4. FUNCTIONAL REQUIREMENTS

Each target capture technology will have its own unique functional requirements. These include operating temperature, gas preconditioning, liquid reservoirs, temperature control, and process monitoring/control capabilities. In addition, column sizing, scrubbing solution compositions, and flow control capabilities may also be required depending on the technology.

Iodine, CO_2, H_2O, Xe, and Kr capture technologies are all based on solid-phase sorbents packed into adsorption columns. For each constituent, three columns in parallel are required to maintain continuous operation. While one column is adsorbing, the next column is ready to adsorb, and the third column is being regenerated or replaced.

The NO_X capture technology is based on a wet, packed bed scrubber design. The column is packed with pall rings or another suitable packing material compatible with nitric acid. The scrubbing solution, consisting of aqueous nitric acid at concentrations up to 3 mole/L, is held in a tank at the bottom of the packed column and recirculated through the packed column continuously with a pump. For improved scrubbing efficiency, the temperature of the scrubbing solution must be maintained below 20°C. Periodically, slipstreams of the scrubbing solution are removed from the system and replaced with fresh H_2O to control the acid concentration.

Each of the capture technologies requires process control equipment that can maintain the required operating temperatures during adsorption and desorption operations (i.e., tube and shell heat exchangers, electrical heaters, thermocouples, and electrical coolers). Required off-gas and liquid flows will be controlled by flowmeters, pumps, and blowers. All relevant operating parameters will be collected utilizing a data acquisition system. The effluent from each capture technology will be monitored by a residual gas analyzer with a multiport switching valve to determine the outlet concentrations of the target analytes leaving the columns. Using one analytical instrument with a switching valve will create some lag between analyses but will simplify data gathering while minimizing system footprint. Table 2 presents a summary of the key operating parameters for each capture technology.

Table 2. Key operating parameters for each capture technology.

	Capture Technology	Superficial Velocity (m/min)	Length-to-Diameter Ratio (L/D)	Column Diameter (m)	Column Height (m)	Adsorption Temperature (°C)	Desorption Temperature (°C)
Iodine	Chemisorption	10.0	7.63	0.036	0.273	150	N/A
NO_x	Aqueous scrubber	15.5	5.2	0.27	1.5	20	N/A
H_2O	Mol sieve	5.0	2.0	0.054	0.108	3	260
CO_2	Physisorption	10.0	7.63	0.036	0.273	25	N/A
Xe	Physisorption	4.29	8.67	0.054	0.472	25	150
Kr	Physisorption	4.29	8.67	0.054	0.472	-80	150

5. PRECONCEPTUAL DESIGN

Figure 2 shows a preconceptual piping and instrumentation diagram (P&ID) of the off-gas system. Each required target analyte technology is represented, as well as some of the associated auxiliary equipment.

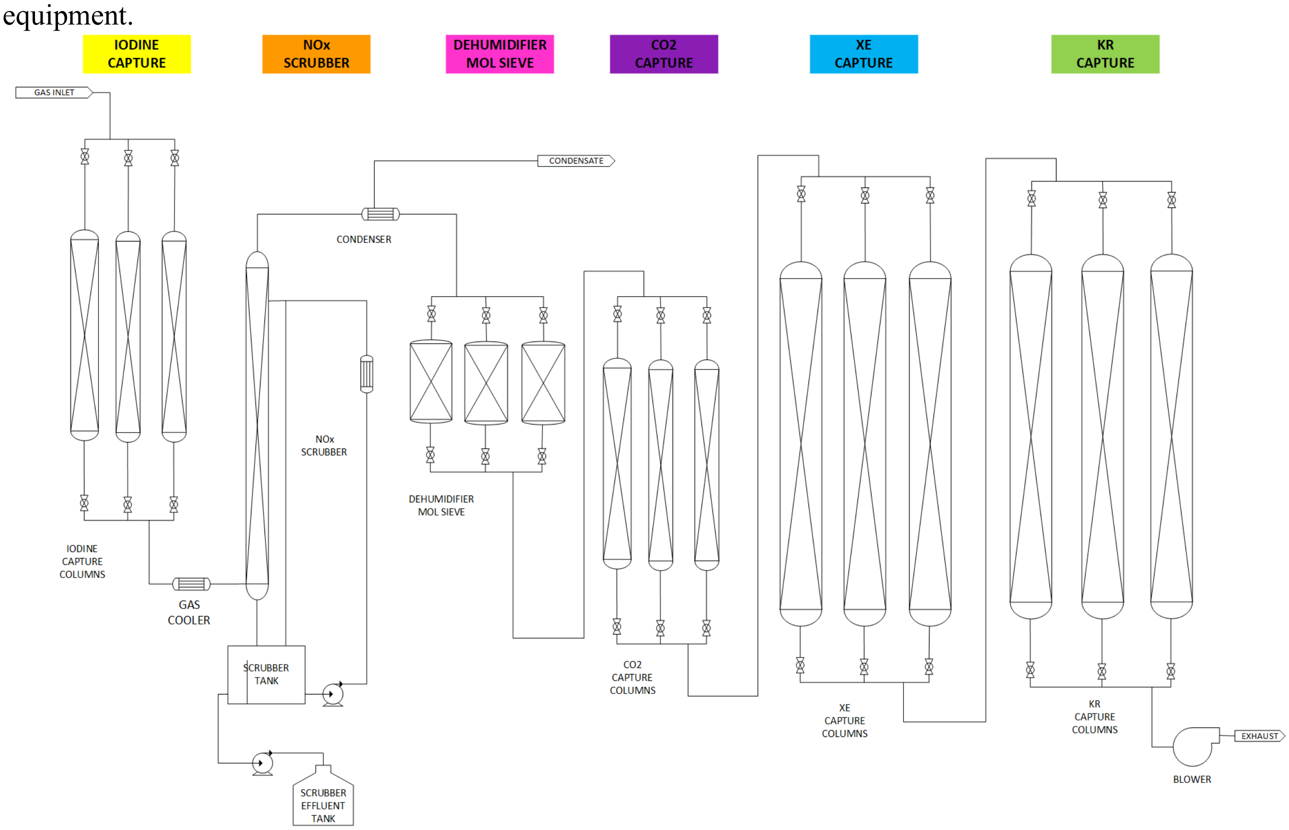

Figure 2. Simplified P&ID of the proposed integrated off-gas system. Columns are depicted at relative scale.

Notional system layouts for shipping and operation of major equipment are provided in Figures 3 and 4, respectively. Only one cargo container is depicted, but the system is expected to be housed in three containers, one for the main process equipment, one for auxiliary equipment for heating and cooling, and a third for control systems, data acquisition and storage, and human-machine interface.

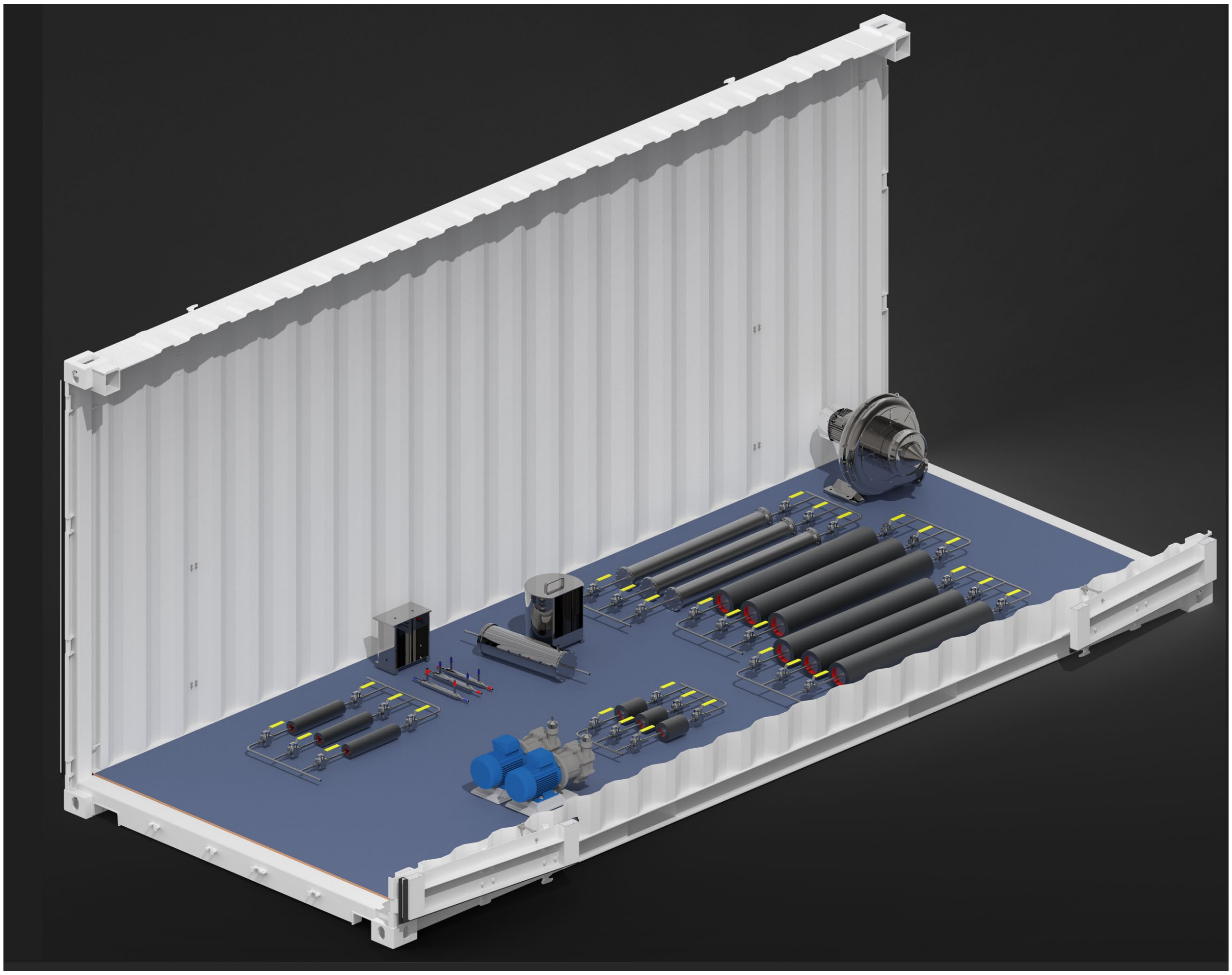

Figure 3. Cutaway view of potential configuration inside a ConEx box for safe shipping.

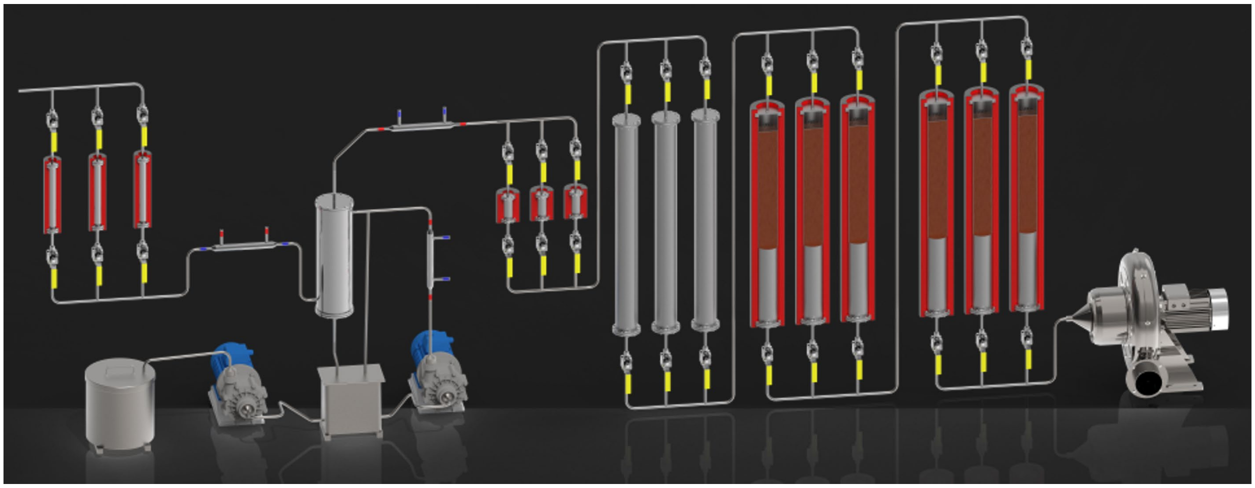

Figure 4. Operational system layout at relative scale.

6. COST ESTIMATES

Table 3 presents estimates of the main equipment costs broken down by each capture technology's operational requirements. Table 4 presents the estimated costs of the required auxiliary equipment in the context of the entire system. These estimates are based on previous experience with purchasing this type of equipment. Table 5 presents labor costs, and Table 6 the total estimated cost of the system: $885,800.

Table 3. Main equipment cost estimates, broken down by system.

Target Analyte	Required Equipment	Quantity	Cost ($/each)	Total Cost ($)
Iodine	SS columns	3	$ 600	$ 1,800
	Thermocouples	6	$ 100	$ 600
	Heaters	4	$ 100	$ 400
	Controlling valves	7	$ 1,000	$ 7,000
	Heater controllers	4	$ 600	$ 2,400
	Heat exchangers	2	$ 1,500	$ 3,000
	Pressure transducers	6	$ 500	$ 3,000
NO_x	Scrubbing column	1	$ 5,000	$ 5,000
	Packing material	1	$ 3,000	$ 3,000
	Pumps	2	$ 5,000	$ 10,000
	Thermocouples	3	$ 100	$ 300
	Controlling valves	2	$ 1,000	$ 2,000
	Temperature controller	1	$ 600	$ 600
	Flow controller	1	$ 600	$ 600
	Pressure transducers	4	$ 500	$ 2,000
	Tanks	2	$ 5,000	$ 10,000
H_2O	SS columns	3	$ 600	$ 1,800
	Thermocouples	7	$ 100	$ 700
	Cooler	1	$ 8,000	$ 8,000
	Heaters	3	$ 100	$ 300
	Controlling valves	7	$ 1,000	$ 7,000
	Temperature controllers	3	$ 600	$ 1,800
	Heat exchanger	1	$ 1,500	$ 1,500
	Pressure transducers	6	$ 500	$ 3,000
CO_2	SS columns	3	$ 600	$ 1,800
	Thermocouples	6	$ 100	$ 600
	Controlling valves	7	$ 1,000	$ 7,000
	Heater controllers	4	$ 600	$ 2,400
	Pressure transducers	6	$ 500	$ 3,000
Xe	SS columns	3	$ 600	$ 1,800
	Heaters	3	$ 100	$ 300
	Thermocouples	3	$ 100	$ 300
	Controlling valves	7	$ 1,000	$ 7,000
	Temperature controllers	3	$ 600	$ 1,800
	Pressure transducers	6	$ 500	$ 3,000
Kr	SS columns	3	$ 600	$ 1,800
	Thermocouples	7	$ 100	$ 700
	Coolers	3	$ 8,000	$ 24,000
	Heaters	3	$ 100	$ 300
	Controlling valves	7	$ 1,000	$ 7,000
	Temperature controllers	3	$ 600	$ 1,800
	Pressure transducers	6	$ 500	$ 3,000
			Total	$ 143,400

Table 4. Cost estimates for required auxiliary equipment.

Auxiliary Equipment	Quantity	Cost ($)	Total Cost ($)
Connective tubing, ft	300	$ 25	$ 7,500
Misc. Fittings	50	$ 200	$ 10,000
Chiller	1	$ 18,000	$ 18,000
Blower	1	$ 7,000	$ 7,000
DACS	1	$ 150,000	$ 150,000
Gas Flowmeter	1	$ 2,500	$ 2,500
Cargo Container	2	$ 5,500	$ 11,000
Electrical	1	$ 50,000	$ 50,000
Residual Gas Analyzer	1	$ 120,000	$ 120,000
Control software	1	$ 50,000	$ 50,000
Computer hardware	2	$ 12,500	$ 25,000
Controls wiring/cable, 100 ft	10	$ 50	$ 500
Electrical wiring/cable,100 ft	6	$ 150	$ 900
Exhaust stack	1	$ 3,000	$ 3,000
		Auxiliary Total	$ 462,400

Table 5. Labor cost estimates.

Labor	Hrs	$/hr	Total Cost ($)
Engineering	250	$ 400	$ 100,000
Support services	100	$ 200	$ 20,000
Skilled crafts	640	$ 250	$ 160,000
		Labor Total	$ 280,000

Table 6. Cost estimate summary.

Main process equipment	$ 143,400
Auxiliary equipment	$ 462,400
Labor	$ 280,000
TOTAL	$ 885,800

7. CONCLUSIONS AND RECOMMENDATIONS

The cost estimates herein do not include the development of a conceptual design, a detailed design package, siting, or commissioning as these items are outside the scope of a preconceptual design. Rather, this effort is intended to provide an order-of-magnitude estimate for future planning efforts and provide a roadmap to aid in the development of an integrated test bed. To continue advancing the technology readiness levels of UNF off-gas treatment, it is recommended that the integrated off-gas test bed be moved to the conceptual design phase, and eventually through detailed design and construction. Given that the plan is to provide a flexible, modular system, construction could be done in phases.

8. REFERENCES

1. Jubin, R. T., G. D. DelCul, B. D. Patton, R. S. Owens, D. W. Ramey, and B. B. Spencer. "Advanced Fuel Cycle Initiative Coupled End-to-End Research, Development, and Demonstration, Project: Integrated Off-Gas Treatment System Design and Initial Performance-9226." WM 2009 Conference, March 1–5, 2009, Phoenix, AZ.