Advanced Fuel Cycle Initiative Coupled End-to-End Research, Development, and Demonstration Project: Integrated Off-Gas Treatment System Design and Initial Performance-9226

R. T. Jubin, G. D. DelCul, B. D. Patton, R. S. Owens, D. W. Ramey, B. B. Spencer
Oak Ridge National Laboratory
P.O. Box 2008, Oak Ridge, TN 37831

ABSTRACT

Oak Ridge National Laboratory is conducting a complete, coupled end-to-end (CETE) demonstration of advanced nuclear fuel reprocessing to support the Advanced Fuel Cycle Initiative. This small-scale reprocessing operation provides a unique opportunity to test integrated off-gas treatment systems designed to recover the primary volatile fission and activation products (H-3, C-14, Kr-85, and I-139) released from the spent nuclear fuel (SNF). The CETE project will demonstrate an advanced head-end process, referred to as voloxidation, designed to condition the SNF, separate the SNF from the cladding, and release tritium contained in the fuel matrix.

The off-gas from the dry voloxidation process as well as from the more traditional fuel dissolution process will be treated separately and the volatile components recovered. This paper provides descriptions of the off-gas treatment systems for both the voloxidation process and for the fuel dissolution process and provides preliminary results from the initial CETE processing runs. Impacts of processing parameters on the relative quantities of volatile components released and recovery efficiencies are evaluated.

INTRODUCTION

Equipment for a coupled end-to-end (CETE) research and development (R&D) capability has been installed at the Oak Ridge National Laboratory (ORNL) to provide all primary processing operations, ranging from spent fuel receipt to production of products and waste forms. The head-end portion includes a voloxidation process step to (1) oxidize the fuel; (2) convert it to a free flowing powder; (3) release a major portion of the tritium (99+%) and significant fractions of the volatile fission products, iodine, C-14 (as CO_2), krypton, and xenon; and (4) separate the fuel powder from the cladding. When this powder is dissolved, much if not all of the remaining volatile components are released. A schematic of the process is shown in Figure 1.

Based on current U.S. regulations, I-129 and C-14 must be sequestered essentially indefinitely, but H-3, xenon, and krypton (e.g. Kr-85) can be managed in decay storage. Over the past two to three decades a number of capture technologies were developed to various stages of maturity. The CETE off-gas treatment study evaluates capture methods using parameters such as selectivity, efficiency, regeneration of sorbent, and conversion to final waste forms. Silver-exchanged zeolite for iodine, molecular sieve for tritium, caustic scrub for C-14, and zeolite (mordenite, faujasite) for xenon/krypton are the current

Prepared by Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831-6285, managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract DE-AC05-00OR22725.

baseline technologies for the Advanced Fuel Cycle Initiative. Whether these isotopes should be stripped and stored as compressed gases, stabilized in grout (H-3, C-14), or stabilized in place (grouted or collapse of zeolite structure) is still to be determined. A large body of historical data was evaluated before a particular method was selected for demonstration and testing. The following sections of this paper summarize the source terms applicable to the CETE demonstration, the regulatory drivers, the processing options considered for testing, and the CETE demonstration system and results.

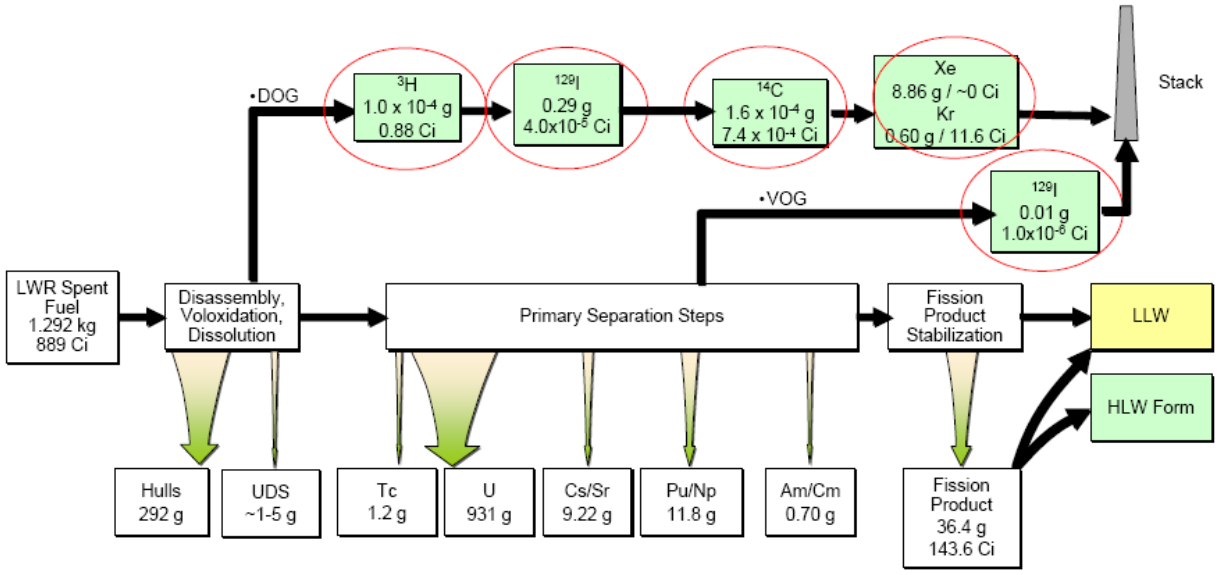

Fig. 1. Volatile fission products from processing 1 kg of spent nuclear fuel at 55 GWd/MTIHM with 5 years of cooling.

SOURCE TERM ESTIMATES

A recent report by Gombert et al. [1] provides estimated head-end (voloxidizer off-gas [VoxOG]) and dissolver off-gas (DOG) concentrations. These were based on ORIGEN data for light-water reactor (LWR) fuel at a burnup of 51 GWd/MTIHM and processing after a 20-year decay period from reactor discharge. Off-gas concentrations for tritium were made assuming that the tritium was released into the off-gas stream using the voloxidation process. Voloxidation is a dry, thermal head-end process designed to further oxidize the fuel, resulting in a fine U_3O_8 powder, and release the tritium from spent uranium (or mixed-oxide) reactor fuel before aqueous processing, thus potentially avoiding the need for subsequent tritium recovery from the aqueous streams [2]. Data from Goode and Stacy [3] indicated near total release of the tritium contained in the fuel oxide matrix during voloxidation, but 40–50% of the tritium may be contained in the cladding and not released.

Minor but radiologically significant quantities of other fission products are also released during the standard voloxidation process. These include C-14, iodine, and krypton (C-14 ~50%, iodine ~1%, and krypton ~5%). The fraction of xenon released is similar to that of krypton but is not radiologically significant due to the short xenon half-lives. Small fractions of the semivolatiles would also be expected under standard voloxidation conditions.

DOG and VoxOG compositions are shown in Table I. These compositions were estimated using data from a large engineer-scale reprocessing equipment test facility located at ORNL [4]. The estimated concentrations shown in the table assume leakage into the equipment from a hot cell with an air atmosphere and the use of air (with CO_2 removed for voloxidation feed gas). Key points to note are that more than 99.97% of the CO_2 that must be treated is not from the fuel, that tritiated water content in the

VoxOG is extremely small [i.e., has a very low dew point (<-50°C)], and that even in the DOG the concentrations of the volatile species are very low.

Table I. Volatile Gas Source Terms for a Notional Facility[a] [2]

	VoxOG (gm/MTIHM)	DOG (gm/MTIHM)	VoxOG (mol/l)	DOG (mol/l)
Tritium (as HTO)	0.3218	--	2.18×10^{-8}	--
H_2O (from leakage into the equipment or air sparges/purges)	5.60	58,100	4.00×10^{-7}	1.12×10^{-3}
$^{14}CO_2$	0.0828	0.0828	Combined with DOG	9.85×10^{-10}
$^{12}CO_2$ (from leakage into the equipment or air sparges/purges)	---	1311	Combined with DOG	1.05×10^{-5}
Iodine	---	359.2	Combined with DOG	3.81×10^{-7}
Krypton	26.57	504.7	Combined with DOG	1.73×10^{-6}
Xenon	402.2	7641	Combined with DOG	1.67×10^{-5}

[a]Basis: VoxOG rate 540 L/min for 1 MTIHM
DOG rate 2,000 L/min for 1 MTIHM
Gas to Voloxidizer has -60°C dew point and is CO_2 free
Air cell at 15°C dew point
DOG cooled to 25°C leaving dissolver
5% krypton/xenon release in Voloxidizer
50% CO_2 release in Voloxidizer

REGULATORY REQUIREMENTS/DRIVERS

Volatile gas emissions from a nuclear fuel recycle facility are addressed in several regulatory documents. The U.S. Environmental Protection Agency (EPA) has established through 40 CFR 190 annual dose limits resulting from nuclear fuel cycle facilities in the commercial sector. 40 CFR 190.10 provides dose limits for specific organs and for the whole body. Specific release limits for Kr-85, I-129 and Pu-239 in terms of curies released per unit of power produced are also defined in 40 CFR 190. 10 CFR 20 provides the dose limits for both workers and individual members of the public. The release limits and required decontamination factors (DFs) to meet these regulations are shown in Tables II and III. 40 CFR 61.92 provides additional limits of 10 mrem/yr dose equivalent to the public.

Table II. Decontamination Factors (DFs) Required to Meet 40 CFR 190

Isotope	Ci/MTIHM	Ci/GW(e)-yr	Minimum Required DFs
I-129		0.83	167

	0.04149		
Kr-85 (5-year cooled)	11,620	234,000	4.67
Kr-85 (10-year cooled)	8,413	169,000	2.38
Kr-85 (30-year cooled)	2,309	46,000	0.93

Table III. Key Release and Exposure Limits

	10 CFR 20			40 CFR 190		40 CFR 61
	Air (Ci/m^3) at site boundary	Water (Ci/m^3)	Dose to member of the public (mrem/yr)	Discharge ($Ci/gW\text{-}yr_e$)	Annual dose (mrem)	Effective dose to any member of the public (mrem/yr)
^{3}H-3	1.0×10^{-7}	1.0×10^{-3}				
C-14 (as CO_2)	3.0×10^{-5}	--				
K-85r	7.0×10^{-7}	N/A		50,000		
I129	4.0×10^{-11}	2.0×10^{-7}		0.005		
Whole body			100		25	10
Thyroid					75	
Other organs					25	

POTENTIAL RECOVERY PROCESSES

Tritium

In the traditional chop-leach process used in LWR fuel reprocessing plants, a small fraction of the tritium inventory would be found in the DOG as water vapor and the remainder would ultimately end up in the high-level liquid waste, primarily from the first cycle solvent extraction system. As noted above, the voloxidation process has been proposed to promote the release of the tritium from the fuel to the off-gas system and thus significantly reduce the fraction entering the aqueous systems. To ensure that the tritium released from the fuel is converted to water, which facilitates its recovery, the proposed design includes passing the off-gas stream through a heated catalytic combiner. It has also been proposed that the uranium oxides processed in the voloxidizer may also act as a catalyst for oxidizing tritium, and thus there is the potential for eliminating the need for a separate catalytic combiner [2]. Tests under conditions typical of that in a voloxidizer, including flow behavior, are still needed to determine whether the combiner can be eliminated.

Tritium recovery from the off-gas stream can be accomplished using desiccants or molecular sieves. Anhydrous $CaSO_4$ and Type 3A molecular sieves are potential candidates.

Iodine

A variety of technologies have been developed for the recovery of I-129 from the off-gas streams. These technologies include scrubbing with caustic or acidic solutions and chemisorption on silver containing materials or adsorbents.

Studies of the distribution of I-129 from the spent nuclear fuel (SNF) being processed into the gas and liquid process streams indicate that about 94% to 99% of the I-129 ends up in the DOG [5, 6]. As the DOG contains the highest fraction of the volatile iodine, the primary iodine recovery technology is applied to this stream, but if DFs greater than ~100 are required, then treatment of the vessel off-gas (VOG) and other off-gas streams may also be required to recover additional I-129.

While the recovery processes were designed to capture iodine and iodine compounds, they will also recover the other halogen elements (e.g., chlorine and bromine).

Silver-exchanged solid sorbents

Numerous studies have been conducted using natural or synthetic sorbent materials. These include zeolites, (e.g. mordenite and faujasite), alumina, and silica gels. To improve the iodine retention capacity of these materials, they are often loaded or ion-exchanged with metals (e.g., Ag, Cd, Pb) and/or a metal nitrate (e.g., $AgNO_3$). Several of these materials are commercially available, including silver-exchanged faujasite (AgX), silver-exchanged mordenite (AgZ), and silver-impregnated silicic acid (AC-6120). The development of silver-exchanged AgX and AgZ was conducted primarily at the laboratory scale in the United States for I-129 recovery. The high acid resistance of the AgZ sorbent is a desirable property when used in the DOG stream. Loadings of 170 mg of iodine per gram of AgZ and 140 to 180 mg CH_3I per gram of AgZ substrate have been reported from simulated DOG streams [7–10]. Extensive development work has also been conducted in Germany on AC-1620 [6].

Charcoal/Activated Carbon

Although activated carbon has been used successfully in nuclear power plants, it has several serious drawbacks such as combustibility that have eliminated the use of this material in reprocessing plant off-gas systems in the past.

Liquid Scrubbing

A variety of liquid scrubbers have been developed for radioiodine control. The principal ones are caustic scrubbing, IODOX, and Mercurex.

Iodine recovery DFs of 50 have been reported for caustic scrubbing at the Windscale Fuel Reprocessing Plant [6, 11]. One disadvantage of caustic scrubbing is that the organic iodides pass through the scrubber essentially unreacted, and the CO_2 and NO_x in the off-gas stream deplete the caustic and form carbonate and nitrates. Methods for immobilizing the waste have not been developed. Because C-14 will be simultaneously removed with the iodine by caustic scrubbing, an independent C-14 trapping technology could not be applied. This could complicate subsequent waste management operations.

The Iodox technology was developed to treat the DOG from the reprocessing of liquid metal fast breeder reactor fuel. In this application, the spent fuel was to have been processed within 180 days of leaving the reactor. Such short cooling times would have required very high iodine DFs ($>10^4$). Cold engineering scale tests of the IODOX process have shown iodine DFs of greater than 10^5. The process uses a counter-current stream of 20–22 M HNO_3 in a bubble cap column to recover the iodine as HI_3O_8 [6, 12]. The major advantages of the process are that no unusual chemicals are added to the reprocessing plant (i.e., nitric acid is ubiquitous in the plant), high DFs are achievable for both elemental and organic iodine compounds, and the iodine products are suitable for conversion to a waste form without significant added volume. The major disadvantage is the capital cost associated with the materials of construction that are

required by the highly corrosive nature of the hyperazeotropic nitric acid scrub solution and the need to add or produce the hyperazeotropic nitric acid used in the scrubbing process.

The Mercurex process uses a less concentrated nitric acid scrub solution (6–14 M) but adds mercuric nitrate as a catalyst to recover the iodine as HgI_2. The mercuric iodide is subsequently oxidized to the iodate, which can be recovered by filtration. Achievable DFs for elemental iodine and methyl iodide are 1,000 to 5,000 and 100 respectively. The scrubber column has an operating temperature of ~50°C [6, 11]. The major advantages of the process are that no unusual materials of construction are required, no special processes are required to produce the hyperazeotropic nitric acid, and high DFs are achievable. The major disadvantage is the toxic nature of the mercury involved and the probable need to convert the mercury-iodine compounds into a more suitable form for disposal.

Silver Reactors

The Hanford Purex Plant and the Savannah River Plant have used silver reactors to remove I-131 from off-gas streams [13]. The reactors consist of a bed of heated ceramic saddles glazed with silver nitrate through which the DOG stream is passed. The iodine is chemisorbed as silver iodide and iodate. DFs of 10 to 10^4 for I-131 recovery have been reported. The reactor efficiency can be restored by using a basic sodium hyposulfite wash solution or 5 M $AgNO_3$. A method to convert the retained iodine from the silver reactors for long-term storage does not appear to have been specifically included as part of the process development.

Krypton

Because krypton is chemically inert, recovery processes for krypton are based on physical separation from the off-gas stream. The primary Kr-85 recovery technologies are cryogenic distillation, fluorocarbon adsorption, and sorption on molecular sieves or charcoal. Xenon is also present at about 10 times the krypton concentration in the gas stream. All xenon radioisotopes have either half-lives less than 30 days or are stable; thus in fuel cooled more than 1 year, Kr-85 is essentially the only radioactive noble gas. Due to its similar properties, xenon is also recovered by these processes.

Cryogenic distillation has been used to commercially recover rare gases for many years. While the technology has been used successfully at the Idaho Chemical Processing Plant to recover krypton from other applications, the process was not optimized for high krypton recovery. Additional development work has been conducted in Belgium, France, Germany and Japan. DFs of 100 to 1,000 have been reported [14]. In Japan, a hot pilot plant cryogenic unit for Kr-85 recovery went active in 1988 [15]. When using the cryogenic distillation process on the dissolver off-gas stream, certain pretreatment steps are needed to remove interfering constituents, thus ensuring system safety and operability. This included the removal of all gases that would condense at liquid nitrogen temperatures. Oxygen is also removed to avoid the formation and accumulation of ozone.

Fluorocarbon absorption technology which uses an organic solvent (CCl_2F_2 called R-12) to selectively absorb noble gases from the DOG stream has been demonstrated in both the United States and Germany. The krypton is thermally stripped from the solvent, which is reused. Krypton concentration factors of greater than 1,000 and krypton recoveries greater than 99% have been demonstrated. The typical R-12 free product stream composition is as follows: CO_2—78%, Xe—13%, N_2—5.5%, Kr—2.0%, O_2—1.4% and Ar—0.1% [16–19].

Solid sorbent separation processes

Both activated carbon and zeolites to recover krypton from the DOG stream have been studied. One proposed system uses a bed of synthetic silver mordenite (AgZ) to recover xenon at ambient temperatures. This is followed by a second bed of hydrogen mordenite (HZ) at ~-80°C that absorbs the krypton and any xenon that passes through the AgZ bed. Krypton is recovered from this second bed and concentrated on a third HZ column. The krypton is recovered from this third column in a cold trap. This process uses temperature swing to effect the transfer from the loaded beds. Laboratory tests have shown DFs of 400 for krypton and 4,000 for xenon [20].

Adsorptive chromatographic separation of krypton on activated charcoal at low temperatures in an industrial scale demonstration has been reported [21]. Krypton DFs of 1,600 have been obtained and krypton purities greater than 99% in the final product.

Carbon-14

The C-14 in the used nuclear fuel is assumed to be evolved primarily into the DOG as CO_2 during the dissolution process. When dissolution is preceded by standard voloxidation, about 50% of the C-14 will be released into the voloxidizer off-gas.

Here again, a variety of technologies have been developed to recover CO_2. These processes include caustic scrubbing, molecular sieve adsorption, and adsorbent bed fixation.

Caustic Scrubbing

CO_2 scrubbing into a caustic solution is a common industrial process [22]. This results in the formation of carbonates. While caustic scrubbing had not been applied to C-14 recovery from the nuclear fuel cycle, EPA determined in 1977 that it was most probable candidate for this application [23]. Limited studies on the use of a caustic slurry scrubber have been conducted. This process uses an alkaline earth hydroxide slurry to react with CO_2 in a stirred tank reactor [24].

Molecular Sieve Adsorption

Another common industrial process for the recovery of CO_2 is adsorption on molecular sieves (e.g., Type 4A). Typical DFs are greater than 100, and a laboratory-scale system has been shown to reduce the CO_2 rich stream from more than 90% CO_2 down to the level of detection (10 ppm). The loaded beds can be regenerated by heating.

Adsorbent Bed Fixation

Researchers at Ontario Hydro have conducted pilot scale studies on a gas solid reaction process to remove $^{14}CO_2$ using beds of either $Ca(OH)_2$ or $Ba(OH)_2 \cdot 8H_2O$. DFs greater than 3,000 have been reported for $Ba(OH)_2 \cdot 8H_2O$.

DEMONSTRATION TEST SYSTEM

Equipment for the CETE demonstration provides all primary processing operations, ranging from spent fuel receipt to production of products and waste forms. The head-end portion will include a voloxidation process step to (1) oxidize the fuel; (2) convert it to a free flowing powder; (3) release a major portion of the tritium (99+%) and significant fractions of the volatile fission products, iodine, C-14 (as CO_2), krypton, and xenon; and (4) separate the fuel powder from the cladding.

The CETE project is also developing and demonstrating an advanced voloxidation process using ozone, to not only remove all tritium but potentially all iodine, xenon, krypton, C-14, and possibly other semi volatiles such as technetium, molybdenum, ruthenium, selenium, tellurinum, and cesium. As indicated previously, current U.S. regulations require that I-129 and C-14 must be sequestered essentially indefinitely, but H-3, xenon, and krypton (e.g., Kr-85) can be managed in decay storage. The treatment of the airborne waste cannot be viewed as simply "add-on" technology to a plant design but must be carefully integrated into the process design.

The 5 to 20 kg scale of the CETE reprocessing operations provide a unique opportunity to test integrated off-gas treatment systems designed to recover the primary volatile fission and activation products released from the SNF. Based on the results of the review of potential processing technologies described above, an initial suite of processes was selected for testing as part of the CETE. The proposed capture methods were selected using parameters such as selectivity, efficiency, regeneration of sorbent, and conversion to final waste forms. The initial suite includes silver-exchanged mordenite (AgZ) for iodine, molecular sieves for tritium, caustic scrub for C-14, and a zeolite (mordenite) for absorption of xenon/krypton. Whether these sorbed elements are stripped and stored as compressed gases, stabilized in grout (H-3, C-14), or stabilized in place (grouted or collapse of zeolite structure) is still to be determined.

A schematic of the head-end off-gas treatment system for the voloxidizer is shown in Figure 2. A photograph of the voloxidizer off-gas test rack is shown in Figure 3.

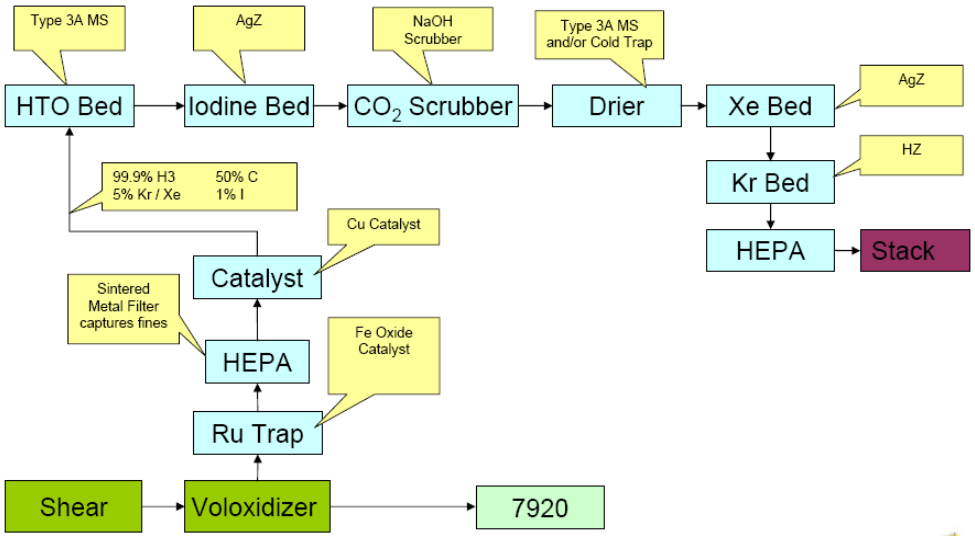

Fig. 2. Voloxidizer off-gas capture system.

Fig. 3. Voloxidizer off-gas rack.

RESULTS AND IMPLICATIONS

The first batch of material tested under CETE used Surry-2 fuel with an initial enrichment of 3.11% and a burnup of 36 GWd/MT heavy metal. The cooling time was 27 years (the fuel was discharged from the reactor in 1981). The amount of material tested was 1,704 g (316 g hulls and 1,388 g of fuel).

The fuel was oxidized in air at about 500°C. The run was prematurely terminated due to apparent cessation of oxygen consumption that occurred after 4 hours of operation at temperature. Weighing of product powder and hulls showed that the fuel was only 70% oxidized. There was also the potential for a cold zone near the closure flange, causing slow kinetics.

The complete batch of fuel was rerun to finish the oxidation. Operation at temperature for 4 hours was done to ensure complete oxidation, and 1,542 g fuel powder was recovered (indicating depleted uranium carryover from shakedown runs).

The voloxidizer was periodically tilted from the horizontal to cause fuel to migrate from the cold zone into the hot furnace zone. Both oxygen consumption and Kr-85 evolution was used to monitor progress and reaction endpoint.

Results from the initial voloxidation are shown in Figure 4. Figure 4(B) shows the temperature of the voloxidizer chamber during the entire run. This shows that it required about 2 hours for the voloxidizer to reach the 500°C operating temperature and that this was followed by a 3-hour period of relatively stable operating temperatures before the initiation of shutdown procedures. Figure 4(A) shows the corresponding responses of the oxygen concentration in the inlet gas and the exit gas from the voloxidizer, the Kr-85 in the gas stream entering the off-gas treatment rack, and the Kr-85 leaving the rack. Figure 4(C) shows the feed gas rates over the same time frames.

The dark blue track in Figure 4(A) shows the inlet oxygen concentration, with 100% corresponding to the oxygen in ambient air, which is expected to remain relatively stable. Some minor drift in the O_2 monitor has been observed. The purple track (exhaust O_2) clearly shows a depletion in the oxygen, as would be expected from the oxidation of the UO_2 fuel. Oxygen consumption was observed to be closely correlated with the Kr-85 concentration in the inlet to the off-gas treatment rack. The apparent sharp increase in the Kr-85 rack inlet concentration at roughly 22:00 hours directly corresponds to the decrease in feed air to the voloxidizer from 3 liters/min to 1.5 liters/min. Finally, the Kr-85 track on exit from the treatment rack indicates that high trapping efficiency for the krypton was achieved.

During the temperature ramp-up there is a rapid release of krypton that is being attributed to the initial rapid oxidation of the fuel fines that are in the voloxidizer.

When the fuel was removed from the voloxidizer for examination it was observed that roughly 30% of the fuel remained “unburned” in the cladding. Again, looking at the oxygen consumption and the krypton release track on Figure 4(A), this physical observation is consistent with the fact that the oxygen in the exit stream is still showing some depletion and the krypton release has not stopped.

During the course of each voloxidation run and off-gas recovery operation gas samples were taken between each unit operation at about 1-hour intervals. At the time this paper was prepared, these samples were still being analyzed. Following each run the trapping beds are removed and sampled, the scrubber solution sampled and replaced, the krypton/xenon traps regenerated, and the recovered gas sampled. Data from these samples are pending.

As of the preparation of this paper, the systems are in place. The voloxidation system is now in active use and the dissolution system is awaiting final approval for hot operations. Approval is expected in January 2009. The data obtained from these systems will serve multiple purposes. We anticipate these systems will provide data to close the material balance on the volatile fission products release during head-end operations. This work will also provide performance data on the recovery processes themselves and on the performance of the processes generating the off-gas streams. Information on the impact of fuel type and voloxidation conditions on the overall volatile gas release and impacts on the dissolution will also be obtained.

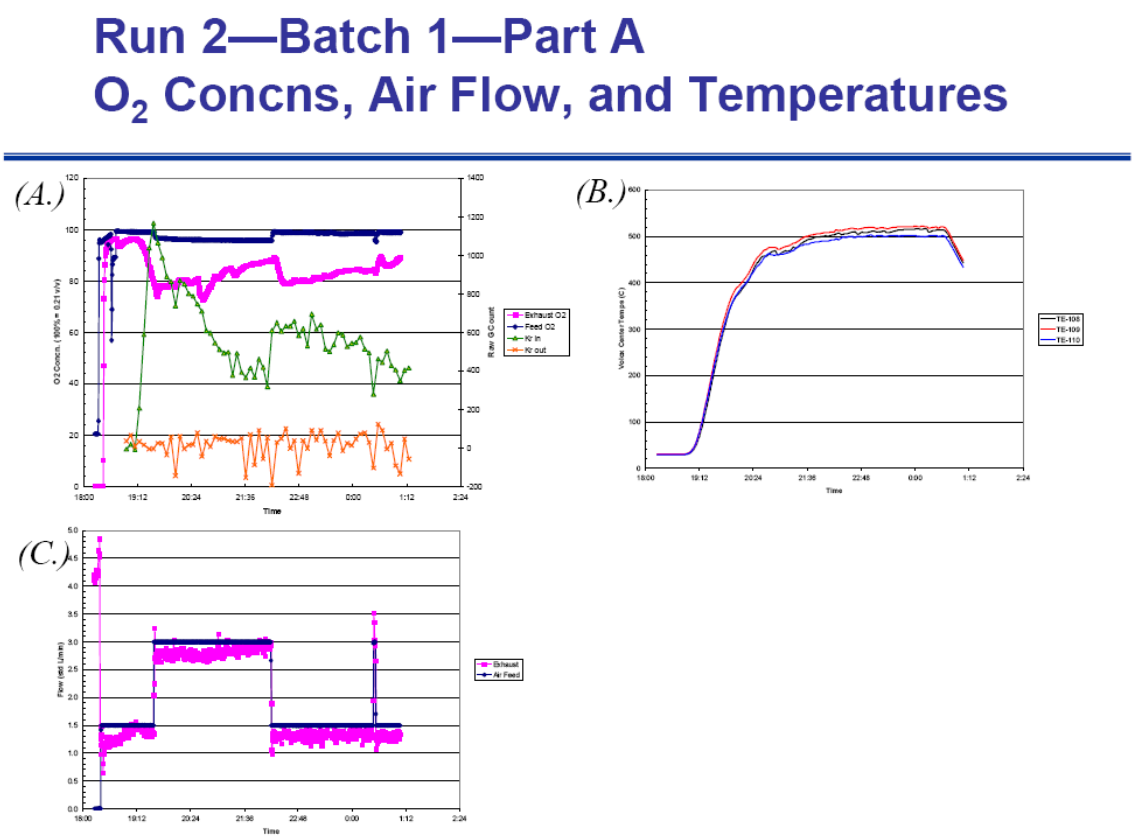

Fig. 4. Oxygen concentrations, air flow, and temperatures from CETE demonstration run.

SUMMARY

Equipment for a CETE R&D capability has been installed at ORNL to provide all primary processing operations, ranging from spent fuel receipt to production of products and waste forms. The system is anticipated to provide data to close the material balance on the volatile fission products release during head-end operations. The data obtained from these technologies will serve multiple purposes, including providing performance data on the processes themselves and on the performance of the processes generating the off-gas streams.

Excellent correlation between the observed reactions in the voloxidizer and the release of krypton to the off-gas system has been noted.

REFERENCES

1. D. GOMBERT II, et al., *Global Nuclear Energy Partnership Integrated Waste Management Strategy Waste Treatment Baseline Study*, Vol 1, GNEP-WAST-AI-RT-2007-000324 (September 2007).

2. B. B. SPENCER, personal communications, 2006.

3. J. H. GOODE and R. G. STACY, "Head-End Reprocessing Studies with H. B. Robinson-2 Fuel," ORNL/TM-6037 (Oak Ridge National Laboratory, June 1978).

4. J. F. BIRDWELL, "Iodine and NOx Behavior in the Dissolver Off-Gas and Iodox Systems in the Oak Ridge National Laboratory Integrated Equipment Test Facility," Proceedings of the 21st DOE/NRC Nuclear Air Cleaning Conference, August 13–16, 1990, Vol 1, CONF-900813, pp. 299–314 (February 1991).

5. F. J. HERRMANN, et al., "Retention and Measurement of Iodine-129 and of Organoiodine in the Offgas Streams of the Karlsruhe Reprocessing Plant WAK," Proceedings of the 21st DOE/NRC Nuclear Air Cleaning Conference August 13–16, 1990, Vol 1, CONF-900813, pp. 222–233 (February 1991).

6. W. HEBEL and G. COTTONE, editors, McKay, H. A. C. , et al., "Management Modes for Iodine-129," Proceedings of a meeting organized by the Commission of the European Communities and held in Brussels on September 25, 1981 (February 1982).

7. B. A. STAPLES, et al., "Airborne Elemental Iodine Loading Capacities of Metal Zeolites and a Dry Method for Recycling Silver Zeolite," Proceedings of 14th ERDA Air Cleaning Conference, Vol 1, CONF-760822, p. 363 (1977).

8. T. R. THOMAS, et al., "The Development of Ag°Z for Bulk ^{129}I Removal from Nuclear Fuel Reprocessing Plants and PbX for ^{129}I Storage," Proceedings of the 15th DOE Nuclear Air Cleaning Conference, Vol 1, CONF-780819, p. 394 (1978).

9. R. T. JUBIN, "Organic Iodine Removal from Simulated Dissolver Off-Gas Systems Using Partially Exchanged Silver Mordenite," Proceedings of the 17th DOE Nuclear Air Cleaning Conference, CONF-820833, pp. 183–197 (1983).

10. R. D. SCHEELE, L. L. BURGER, and C. L. MATSUZAKI, *Methyl Iodide Sorption by Reduced Silver Mordenite,* PNL-4489, (Pacific Northwest Laboratory, Richland, Washington, July 1983).

11. INTERNATIONAL ATOMIC ENERGY AGENCY (IAEA), *Treatment, Conditioning and Disposal of Iodine-129,* IAEA Technical Report Series No 276, (International Atomic Energy Agency, Vienna, 1987).

12. U.S. Energy Research and Development Administration (ERDA), *Alternatives for Managing Wastes from Reactors and Post-Fission Operations of the LWR Fuel Cycle,* Vol 2 of 5, ERDA-76-43, Chap 13 and 14, (U. S. Department of Energy, 1976).

13. W. A. RODGERS, and S. L. REESE, "The Removal of Iodine from Reprocessing Plant Effluents," *Reactor and Fuel-Processing Technol.,* **12**, p. 173 (1969).

14. W. R. A. GOOSSENS, G. G. EICHHOLZ, and D. W. TEDDER, editors, "Treatment of Gaseous Effluents at Nuclear Facilities," *Radioactive Waste Management Handbook,* Volume 2 (Harwood Academic Publishers, Newark, New Jersey, 1991).

15. I. W. LEIGH, *International Nuclear Fuel Cycle Fact Book,* PNL-9450-1 (Pacific Northwest Laboratory, Richland, Washington, 1994).

16. D. K. LITTLE, et al., "Noble Gas Removal and Concentration by Combining Fluorocarbon Absorption and Adsorption Techniques," Proceedings of the 17th DOE Nuclear Air Cleaning Conference, CONF-820833, pp. 694–717 (1983).

17. IAEA, *Separation, Storage and Disposal of Krypton-85,* Technical Report Series No 199 (International Atomic Energy Agency, Vienna, 1980).

18. E. HENRICH, et al., "Selective Absorption of Noble Gases in Freon-12 at Low Temperatures and Atmospheric Pressure," Proceedings of the 18th DOE Nuclear Air Cleaning Conference, CONF-820833, pp. 959–981 (1985).

19. W. HEBEL, and G. COTTONE, editors, Leudet, A., et al., "Methods of Krypton-85 Management," Proceedings of a meeting organized by the Commission of the European Communities and held in Brussels, 29 June 1982 (February 1983).

20. D. T. PENCE, "Critical Review of Noble Gas Recovery and Treatment Systems," *Nucl. Saf.* **22**(6), pp. 751–765 (1981).

21. H. D. RINGEL, et al., "Development of a Process for Adsorptive Separation of Kr-85 from Off-Gas of Nuclear Facilities," *Proceedings of the 21st DOE/NRC Nuclear Air Cleaning Conference,* Vol 1, CONF-900813, pp. 299–314 (1990).

22. L. E. TREVORROW, et al., *Compatibility of Technologies with Regulations in the Waste Management of H-3, I-129, C-14, and Kr-85, Part I, Initial Information Base,* ANL-83-57 Part I (Argonne National Laboratory, August 1983).

23. R. A. BROWN, J. D. CHRISTIAN, and T. R. THOMAS, *Airborne Radionuclide Waste Management Reference Document,* ENICO-1133 (Exxon Nuclear Idaho Company, Inc., Idaho Falls, 1983).

24. K. J. NOTZ, et al., "Processes for the Control $^{14}CO_2$ During Reprocessing," Proceedings of the International Symposium on Management of Gaseous Wastes from Nuclear Facilities, Vienna, February 1980, IAEA-SM-245129, pp. 191 (1980).