LABORATORY-SCALE UF_6 TEST BED SUPPORTING NONPROLIFERATION RESEARCH AND DEVELOPMENT*

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ABSTRACT

A laboratory-scale apparatus has been developed at Oak Ridge National Laboratory for the purpose of providing a UF_6 environment for testing of instruments and materials in simulated operating plant conditions. The system has been used for several diverse applications in support of nonproliferation and safeguards programs as well as other U.S. Department of Energy programs. Apparatus characteristics and capabilities are described and illustrated with brief descriptions of several recent projects.

I. INTRODUCTION

The International Atomic Energy Agency (IAEA) currently inspects and monitors uranium fuel cycle facilities such as enrichment, conversion, and blend-down facilities. The IAEA requires devices to monitor and sometimes to sample process streams containing UF_6. Alternate or improved monitoring devices must be tested prior to deployment in the field. While surrogate gases may be used for some of the development process, eventually it is necessary to demonstrate that the devices will work properly in a UF_6 environment. From a time and cost standpoint, it is generally advisable to perform initial UF_6 testing in a controlled laboratory setting prior to deployment in an operating plant.

Laboratory-scale systems capable of providing a UF_6 environment and simulating operating plant environments can be challenging to set up and maintain. This paper discusses a unique laboratory test-bed system at Oak Ridge National Laboratory (ORNL) that has been used for testing and demonstration of devices and materials intended for use in UF_6 or other fluorinating environments. Most recent and planned work has been in support of nonproliferation and safeguards programs, though other U.S. Department of Energy programs have also been supported, as have a few Work-for-Others activities.

II. APPARATUS

The laboratory test-bed consists of two independent gas manifolds (test loops) that are constructed of fluorine-compatible materials (largely Monel, nickel, and SS316). They contain a gas circulation loop driven by a metal bellows pump to produce the desired gas flow and to ensure mixing of gases. On-line instrumentation includes a mass flowmeter, pressure sensors, and a Fourier transform infrared (FTIR) spectrophotometer, which is capable of determining concentrations for most of the gaseous compounds of interest. FTIR spectra are displayed in real time and are also recorded on a computer for analysis later. Other environmental data (pressure, temperature, and flow) are monitored by and recorded on the same computer using a data acquisition (DAQ) system. Both the FTIR and the DAQ can be configured to take readings at fixed intervals, allowing unattended operation of the system for extended periods of time—runs of up to several weeks in duration have been done in this manner. Both test loops have the same general layout, and the schematic for Test Loop #2 is shown in Figure 1.

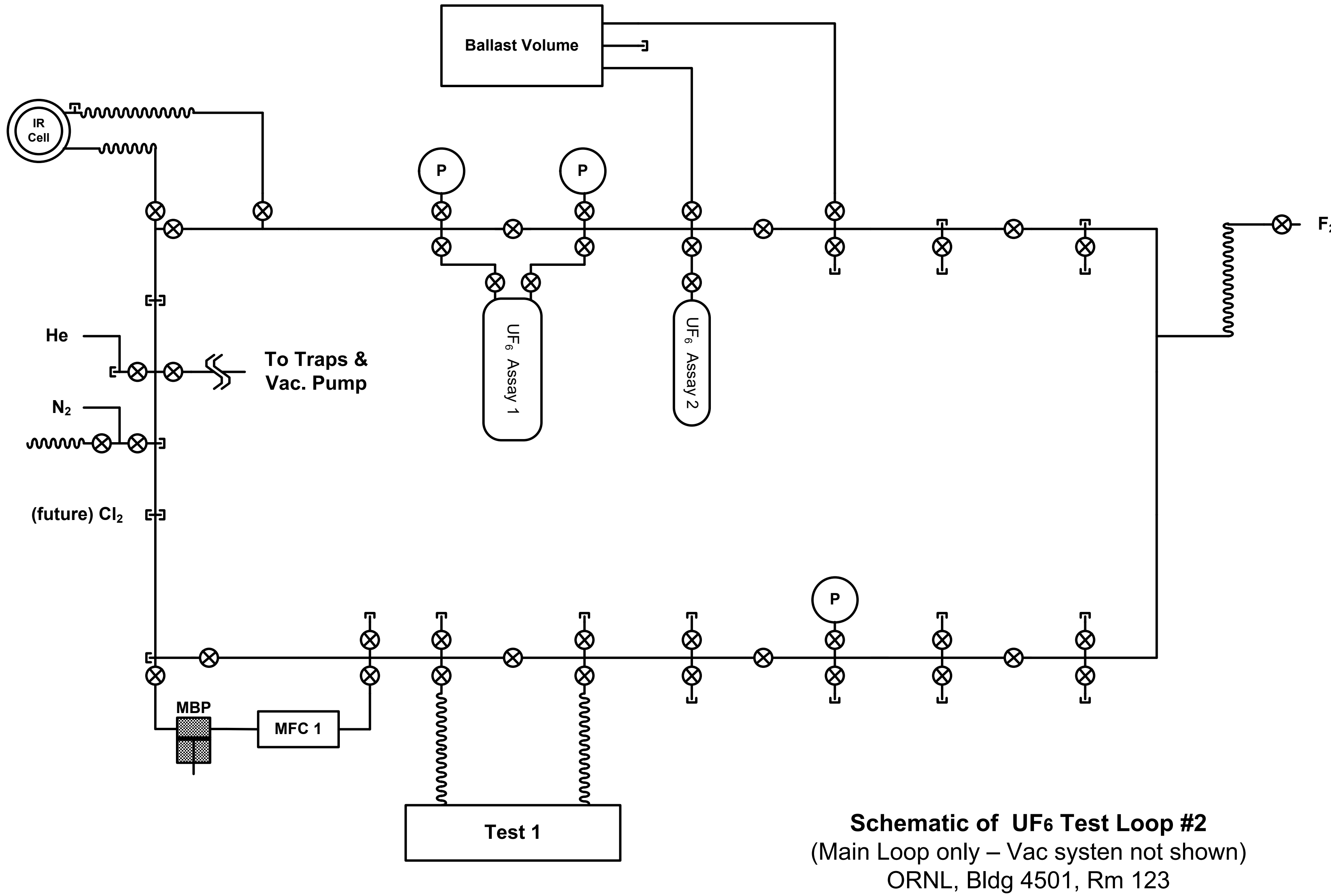

Figure 1. Schematic of UF_6 Test Loop #2 as configured for general compatibility testing.

Auxiliary systems include a dry vacuum pumping system protected by a soda-lime chemical trap, gas sources (He, N_2, and F_2), and sampling systems. Figure 1 shows a schematic of the apparatus in a configuration used for material compatibility testing of a device labeled "Test 1." Device or material compatibility testing is usually arranged by incorporating the device to be tested (or a chamber containing material samples) into the gas-flow loop. Figure 2 is a photograph of the apparatus in the same general configuration as shown in Figure 1, though in that case the "test device" was a chemical trap containing NaF pellets undergoing fluorine treatment. The experimental parameters (UF_6 concentration, uranium assay, flow/static conditions, etc.) can be easily changed as required by the particular investigation.

The test loops are located in a radiological and chemical fume hood, while the instrumentation controls and readouts, as well as the DAQ computers and FTIR, are located adjacent to the hood. Operational safety and radiological constraints vary with the gases used, the assay and quantity of uranium used, etc.

Figure 2. Test Loop configured for F_2 and heat treating a NaF chemical trap (the vertical cylinder in the lower left quadrant).

More exotic experiments have included systems for studying and containing small samples of UF_6 in the liquid state (Figure 3) and chambers for conducting contained releases of UF_6 into air (Figure 4).

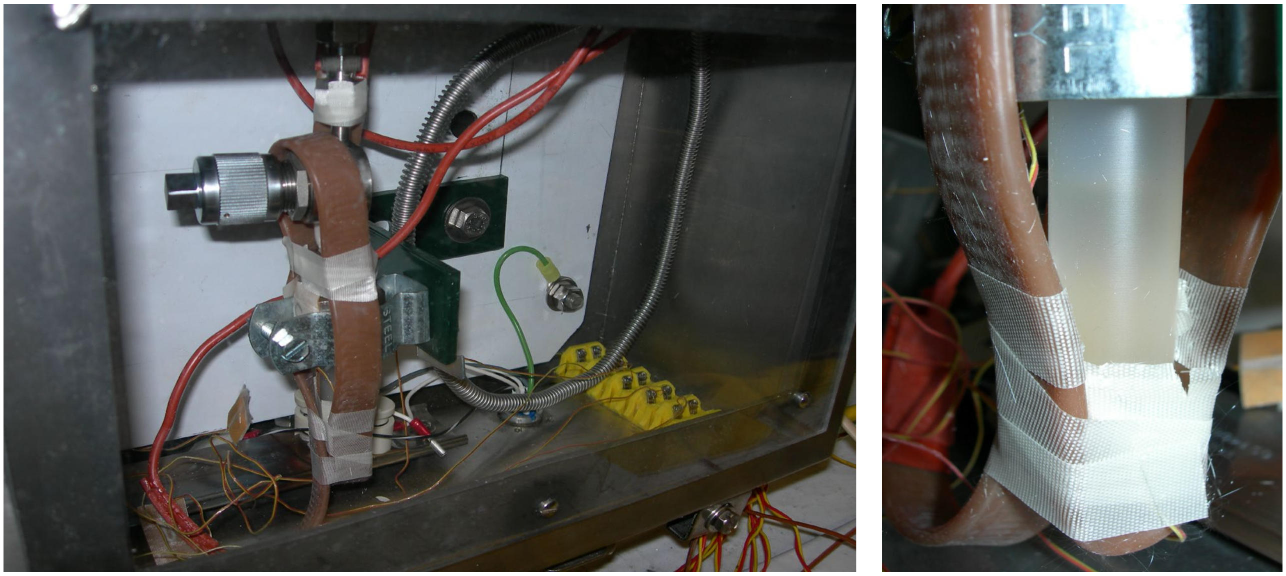

Figure 3. Auxiliary containment for UF_6 liquid studies at left; close-up of Kel-F container with liquid UF_6 at right.

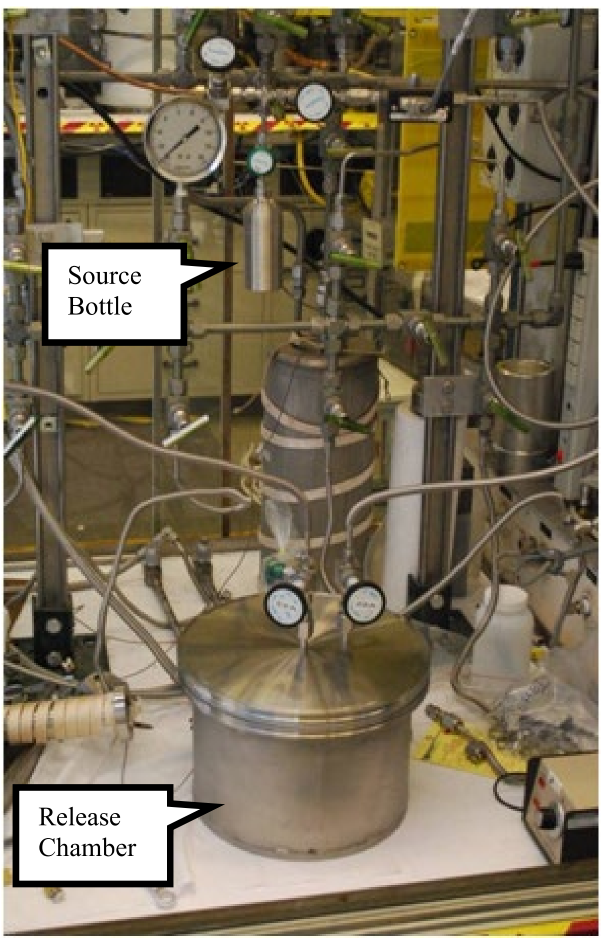

Figure 4. Auxiliary chamber for conducting UF_6 releases into (contained) air.

Many of the experimental applications to date have involved exposing materials or instruments to a fluorinating gas environment. To maintain a reasonably stable gas composition for the duration of an experiment, several techniques are utilized. A thorough leak test is performed prior to any experiment, and the apparatus is passivated with F_2 after any contact of an interior surface with the atmosphere. The purpose of the passivation is to stabilize newly exposed surfaces to the fluorinating environment and, more importantly, to consume any adsorbed moisture. Passivation conditions vary depending on the prospective physical conditions in the experiment and the particular study being undertaken. Typically F_2 will be introduced to the evacuated system at a low partial pressure (~40 mbar) and then is diluted with dry He or N_2 to ~300 mbar. The mixture is circulated through the flow loop by the metal bellows pump for a period of time (20 min or more). Infrared (IR) spectra are taken in order to monitor buildup of HF or other reaction products, which are removed via the soda-lime trap. Successively more concentrated F_2 mixtures are introduced to the system, and the process is repeated until the reaction products are removed. Passivation is carried out at or above the temperature that will exist in the planned experiment.

UF_6 is a condensable gas, which has a saturation vapor pressure of 105 mbar at 20°C. Most applications have required UF_6 partial pressures below this value, allowing the apparatus to operate at ambient laboratory temperatures. For applications requiring UF_6 partial pressures up to 800 mbar, heat tracing and temperature controlling the active portion of the system were needed.

During a typical experiment, IR spectra are taken periodically to monitor gas composition. Reaction of UF_6 with contaminants, materials of construction, or test materials will produce solid reaction products (e.g., UF_5 or UO_2F_2) and possibly gases, which generally are IR active (e.g., HF, CF_4,or SiF_4). Figure 5 is a micrograph of a typical reaction product from UF_6.

After an experiment, much of the UF_6 can be recovered by condensing it in the in-line cold trap. Residual and noncondensable gases are evacuated via the soda-lime chemical trap, which captures the fluorinating gases.

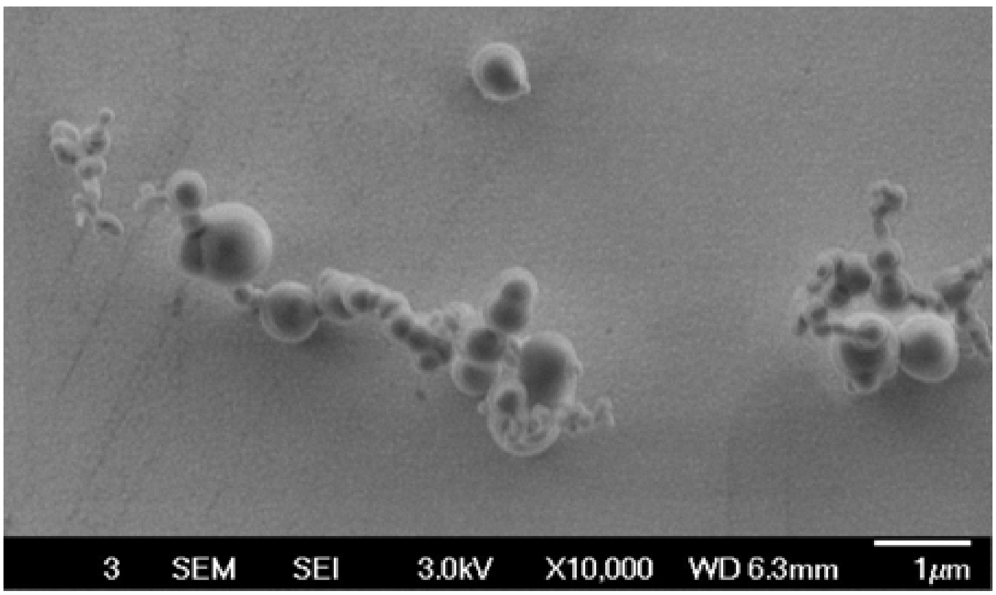

Figure 5. Micrograph of typical product of UF_6 release into humid air.

III. EXPERIMENTAL

A wide variety of experimental campaigns have been conducted using this system. A select number of examples will illustrate the capabilities of the test loops.

III.A. Enrichment Monitor Testing

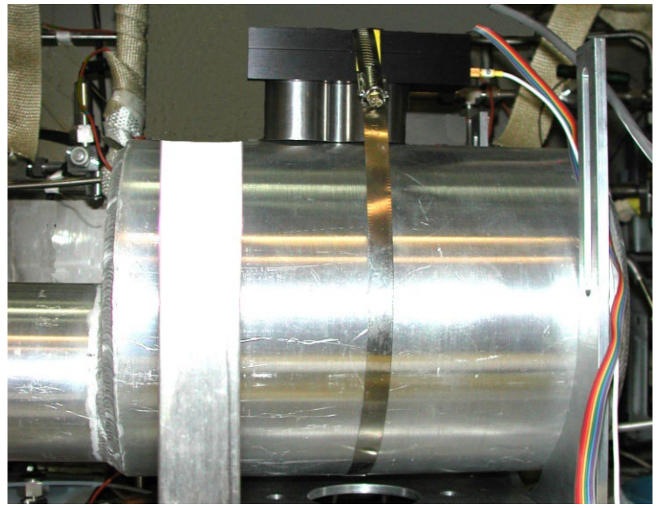

Figure 6. CdZnTe sensors mounted in shielded holder at top of 8-in. aluminum pipe containing UF_6 at various enrichments and pressures.

Several campaigns have been conducted for users needing to test various technologies for nonintrusively monitoring the enrichment of uranium inside process pipes. Figure 6 illustrates the testing of one such device, a CdZnTe detector developed by Drs. Gary Gardner and Bill Murray of Los Alamos National Laboratory. In the figure, the sensors are mounted to the exterior of an 8-in.-diameter aluminum pipe mounted to the test loop. During this series of tests, UF_6 at various pressures and assays was flowed through this pipe, with the EM sensor attempting to detect those changes [1]. Similar tests were conducted with pipe sections simulating 4- and 2-in.-diameter process piping that can be found in uranium enrichment facilities.

III.B. Ion Trap Mass Spectrometer Testing

Dr. William Whitten and Cyril Thompson of ORNL have developed a portable ion trap mass spectrometer for in-field UF_6 assay determination and purity. A recent and ongoing activity has tested this instrument's effectiveness and component longevity when analyzing gas streams that contain UF_6. The test loop has been used to provide a low-pressure continuous sample (~10^{-5} mbar) to a field demonstration system to determine if critical components of the system could function without adverse effects. The low-flow, low-pressure sample was metered to the high-vacuum mass spectrometer chamber via a leak valve and capillary. The high-vacuum pumping system, pressure monitoring instrumentation, ionizer, and the mass spectrometer's ion multiplier were then subjected to the UF_6 sample at pressures slightly in excess of those expected to be experienced in actual operation.

III.C. Materials Compatibility Tests

A variety of simple chemical compatibility tests have been performed for several sponsors. For such tests, a sample exposure chamber was connected in the "Test 1" position as designated in Figure 1. The exposure chamber was heated as required by the experiment in Figure 7. In some cases, passivation measures had to be modified to avoid compromising the intent of the test. At a minimum, the sample chamber was purged with dry inert gas for an extended period of time prior to the test.

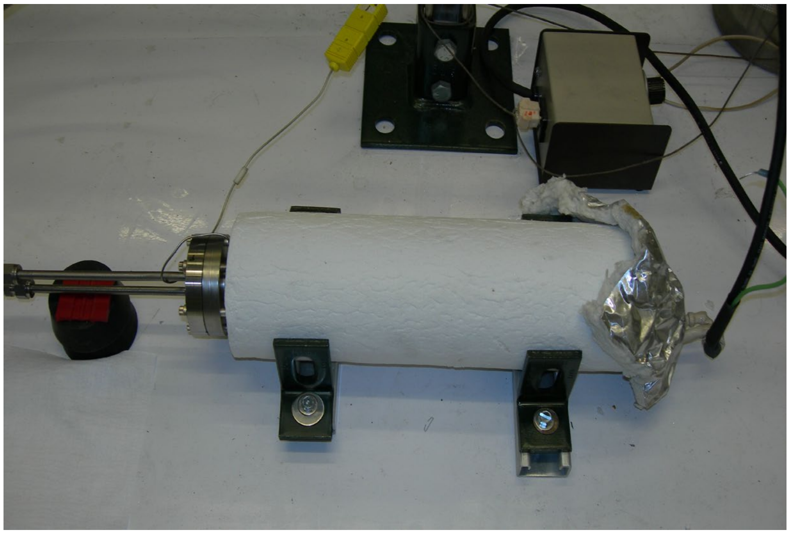

Figure 7. Typical chamber and heating element used for UF_6 materials compatibility testing.

Test samples have ranged from metal coupons to sintered metal filters to elastomers to composite materials. Gases used in the testing have included HF, F_2, UF_6, and ClF_3. The measure of performance varied with the application. In many cases, weight gain or loss has served as an indication of corrosion or reaction product deposition. In some cases, electron micrographs and surface elemental analysis were performed to map morphology changes to better characterize corrosion and deposition.

Figure 7 shows a typical compatibility test chamber enclosed in a commercial heating element to allow testing at elevated temperatures.

III.D. Chemical Trapping Agent Testing

Another short-term R&D campaign involved testing the HF chemical trapping performance of a new brand of NaF pellets. As part of ORNL's Molten Salt Reactor Experiment (MSRE) Remediation Program (a project operated at the time by Bechtel Jacobs Company, LLC), anhydrous HF gas was to be trapped using NaF pellets during remediation. While it is well known that NaF will react with HF to form $NaHF_2$, no specific information was available on the practical loading that could be achieved on the specific material purchased from Morita, Inc., nor were quantitative pellet-bed kinetics found in the literature that could allow the trap capacity and operating characteristics to be confidently designed. A series of tests was conducted with the goals of qualifying and characterizing the performance of the new trapping material and providing operating parameter guidance to the trap designer.

Figure 8. Miniature NaF-containing chemical trap used for determining loading behavior of HF on Morita NaF pellets.

These tests were performed by operating miniature traps, which were shorter and smaller in diameter than the full-scale trap (Figure 8). In a given run, the trap would be subjected to an inert gas stream with a fixed HF concentration and constant flow rate. The gas was passed through the trap, and its exit HF concentration was monitored by the FTIR. As the trap became loaded, the exit concentration eventually increased from its initial very low HF level to a level near that of the inlet concentration. This breakthrough profile and the total effective capacity of the material for HF at breakthrough were the critical parameters for the design of the full-size trap.

IV. CONCLUSIONS

The UF_6 testing system is a flexible and useful facility in supporting several fairly diverse applications and programs. There appears to be increasing interest in availability of this system, largely for its (difficult-to-duplicate) capability of conducting laboratory studies with UF_6 in a relatively cost-effective manner. The ability to handle chemically similar corrosive gases has also allowed the system to be used for non-uranium tests when availability permitted.

1. G. Gardner, W. S. Murray, L. D. Trowbridge, and D. W. Simmons, "A Miniature Self-contained CdZnTe Gamma Spectrometer for Use in Unattended Nuclear Material Monitoring," 2005 IEEE Nuclear Science Symposium Conference Record, p. 545 (23 October 2005).