

**RCUT: A Non-Invasive Method for Detection, Location, and Quantification of Radiological Contaminants in Pipes and Ducts - 12514**

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**ABSTRACT**

Radiological Characterization Using Tracers (RCUT) is a minimally invasive method for detection and location of residual radiological contamination in pipes and ducts. The RCUT technology utilizes reactive gaseous tracers that dissociate when exposed to gamma and/or beta radiation emitting from a radiological contaminant in a pipe or duct. Sulfur hexafluoride ( $\text{SF}_6$ ) was selected as a tracer for this radiological application, because it is a chemically inert gas that is both nonflammable, nontoxic, and breaks down when exposed to gamma radiation. Laboratory tests demonstrated that the tracer pair of  $\text{SF}_6$  and  $\text{O}_2$  formed  $\text{SO}_2\text{F}_2$  when exposed to a gamma or beta radioactive field, which indicated the presence of radiological contamination. Field application of RCUT involves first injecting the reactive tracers into the pipe to fill the pipe being inspected and allowing sufficient time for the tracer to interact with any contaminants present. This is followed by the injection of an inert gas at one end of the pipe to push the reactive tracer at a known or constant flow velocity along the pipe and then out the exit and sampling port at the end of the pipeline where its concentration is measured by a gas chromatograph. If a radiological contaminant is present in the pipe being tested, the presence of  $\text{SO}_2\text{F}_2$  will be detected. The time of arrival of the  $\text{SO}_2\text{F}_2$  can be used to locate the contaminant. If the pipe is free of radiological contamination, no  $\text{SO}_2\text{F}_2$  will be detected.

**INTRODUCTION**

The United States Department of Energy (DOE) continually seeks safer and more cost-effective remediation technologies for use in the decontamination and decommissioning (D&D) of nuclear facilities. One of the major efforts within the DOE complex is the closure of the tanks farms and their associated ancillary systems. Ancillary systems consist of those components used to both transfer waste (e.g., transfer lines, pump tanks and pits, diversion boxes and valve boxes) and reduce waste volume through evaporation (e.g., the evaporator systems) [1]. The ancillary systems and the tanks must have the residual radiological inventories accounted for as part of facility closure.

Safely closing ancillary systems, as with waste tanks, involves an intricate set of steps that includes removing as much of the residual waste as possible through various

technologies and techniques. After completing ancillary equipment cleaning operations, a small amount of residual radioactive waste may still remain. As with the tanks, these residuals will need characterization to confirm that radionuclide and hazardous constituent concentrations meet performance objectives to ensure protection of the public and the environment.

For all the sites, the piping that was used to transport process materials represents one of the larger challenges. As the demolition of these systems occur, disposal of this piping has become a costly issue. Currently, all process piping is cut into 3-meter or-less sections, and the ends of the piping are wrapped and taped to prevent the release of any potential contaminants into the air. The piping is then placed in roll-off boxes for eventual repackaging and final disposal. Alternatives that allow for the onsite disposal of process piping are greatly desired due to the potential for dramatic savings in current offsite disposal costs.

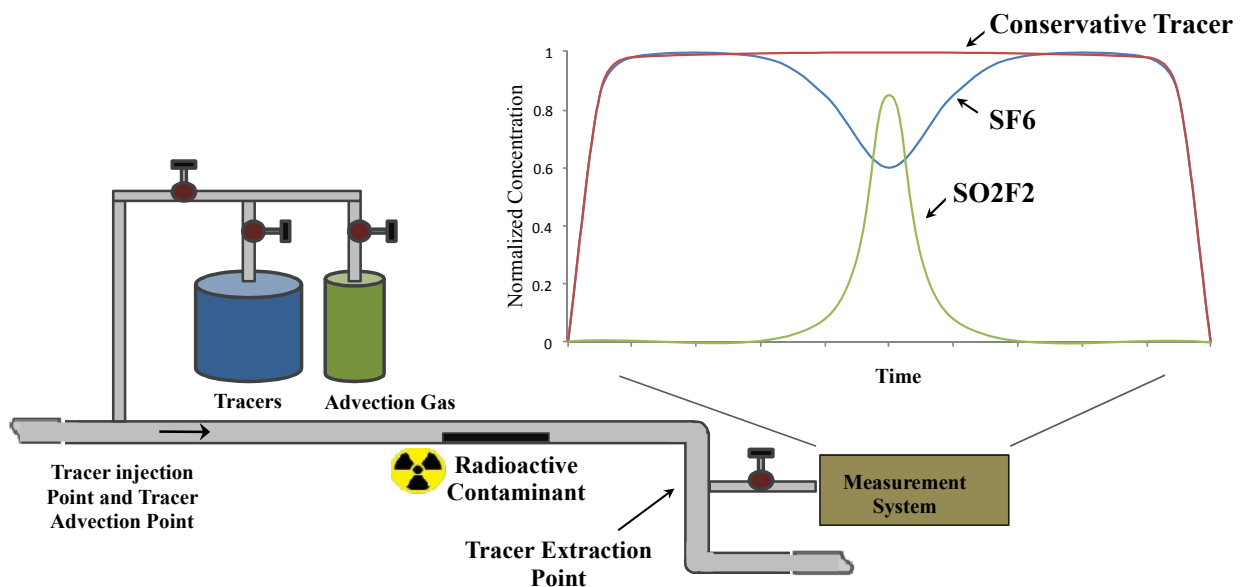
## **TECHNICAL APPROACH**

To address this need for an *in situ* determination of radiological contamination, a minimally invasive method for *detection, location, and quantification of residual beta and gamma radiological contamination in pipes and ducts* using gaseous tracers has been developed. The method is called RCUT, which is a pseudonym for Radiological Characterization Using Tracers [2,3]. RCUT is very similar to an existing method called PCUT (Pipeline Characterization Using Tracers), which was developed for the purpose of detecting, locating, and quantifying non-radiological contaminants in pipes and ducts. PCUT was demonstrated under a Phase I and II SBIR for the US Department of Energy (DOE) [4-6]. PCUT has been demonstrated for piping of various lengths ranging from 3 m to 45 m that have been contaminated with chlorinated solvents including TCE, PCE and CCl<sub>4</sub>, various petroleum products, and heavy metals such as mercury. If appropriate tracers are available, the PCUT approach is able to address nearly any pipeline characterization need and may be used to characterize other man-made environments like ducts, containers, rooms, buildings, and other enclosures. The location and quantification measurements have been demonstrated to within 5% and 10%, respectively, during previous PCUT studies [4-6].

### **Gaseous Tracer Technology – PCUT and RCUT**

There are a variety of ways to implement RCUT and PCUT. The method that is used depends on the tracers being used and how they interact with the contaminant. For some of the PCUT methods, a conservative tracer, i.e., a tracer that does not react with the contaminant, is utilized as a reference or control. *For RCUT, only the reactive tracer*

is required. RCUT (and PCUT) are best implemented by injecting enough reactive tracer into the pipe to completely fill the entire pipe being inspected and allowing sufficient time (minutes to tens of minutes) for the tracer to interact with any contaminant that may be present. Then, an inert gas is injected into the pipe at one end of the pipe to push the reactive tracer at a known or constant flow velocity along the pipe and out the exit port and sampling location at another section or at the end of the pipe. The concentration of the tracer gases can be measured *online* with a gas chromatograph (GC) as they flow out the pipeline; alternatively, the concentration of the tracer gases can be sampled at set time intervals and measured with a gas chromatography/mass spectroscopy (GC/MS), or some other analytical system. If a radiological contaminant (beta or gamma) is present, the presence of  $\text{SO}_2\text{F}_2$  will be detected; there may also be a slight change in the  $\text{SF}_6$  concentration. The time of arrival of the  $\text{SO}_2\text{F}_2$  can be used to locate the contaminant, because the advection velocity of the inert gas is known. If the pipe is free of radiological contamination, no  $\text{SO}_2\text{F}_2$  will be detected. Figure 1 illustrates the basic components of the system and the expected results from a region of radiological contamination within the pipeline. A conservative tracer can be used to monitor whether the pipeline leaks and the presence of other additives within the pipeline.



**Figure 1.** Schematic of reactive  $\text{SF}_6$  tracer deployed for pipeline characterization of radiologically contaminated pipes. The presence of  $\text{SO}_2\text{F}_2$  and the time of arrival of  $\text{SO}_2\text{F}_2$  indicate the presence and location of the radioactive contaminant along the pipeline. Multi-peak concentration curves will occur if there is more than one region of contamination. If the entire pipe or large sections of the pipe are contaminated, then the concentration curve will be broad.

RCUT and PCUT can be used in support of deactivation and decommissioning (D&D) of piping and ducts that may be contaminated with radioactive or hazardous materials.

These methods can be used in the following three ways:

- Demonstrate that the pipe or duct is not contaminated and can be removed without special equipment or special safety precautions.
- Determine the location of any contamination that might be detected.
- Determine the amount of the detected contaminant.

The potential amount of cost savings and schedule reduction related to the first bullet is immense. Knowing that a pipe or duct is free of contamination, especially radioactive contamination is key. Since many of the DOE pipelines are buried, the ability to characterize the pipe in place and possibly avoid excavation represents a significant cost savings.

RCUT and PCUT methods of detecting radiological and non-radiological contamination of pipes offer significant advantages over conventional pipe inspection techniques, including:

- Tracer movement is not impacted by pipe diameter or configuration, and therefore, can be used on small diameter piping.
- No *a priori* information about the pipe diameter or configuration is required for the method to work (although configuration information is needed if a contaminant is detected and its location is desired).
- There are no moving parts or equipment that must be introduced into the pipe.
- There is no sparking potential or ignition source with gaseous tracers.
- There are no decontamination requirements.

In addition, the RCUT technology has immediate application for the D&D of piping exposed to radiological contamination at the DOE's Hanford Site and Savannah River Site (SRS). Many of these piping systems are either buried underground or are otherwise inaccessible, where external inspection techniques requiring direct or safe access to the outside wall of the pipe cannot be used.

### **RCUT Measurement Approach for Pipelines**

The tracers identified for use with RCUT dissociate<sup>1</sup> into other compounds when they interact or pass by radiologically contaminated sections of the pipe (or duct). As illustrated below, our laboratory tests indicate that the tracer pair of SF<sub>6</sub> and O<sub>2</sub> will form SO<sub>2</sub>F<sub>2</sub> when exposed to a gamma or beta radioactive field of sufficient strength. Thus, we know that the pipe contains radioactive contaminants if SO<sub>2</sub>F<sub>2</sub> is detected at the

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<sup>1</sup> Dissociation is the process by which the action of a solvent or a change in physical condition, as in pressure or temperature, causes a molecule to split into simpler groups of atoms, single atoms, or ions. It also may involve the separation of an electrolyte into ions of opposite charge.

measurement point. If only SF<sub>6</sub> is detected, then the pipe is free of radioactive contamination.

To date our testing has focused on rather high exposure levels as noted below to develop a tracer strategy. Additional efforts will evaluate the time/exposure relationship to determine the detection limits and the strategies to reach regulatory levels for disposal in place.

A gas chromatograph/mass spectrometer (GC/MS) is used to monitor the concentrations of the analytes (gaseous compounds) in real time; other analytical techniques that can detect SO<sub>2</sub>F<sub>2</sub> concentrations in the presence of high SF<sub>6</sub> concentrations can also be used. RCUT uses tracer gases that are predominantly dissociated (i.e., the tracer gases split into simpler groups of atoms, single atoms, or ions) when exposed to gamma and/or beta radiation emitting from a radiological contaminant in a pipe or duct. For RCUT, the tracer gas is carefully selected to insure that (1) the tracer will react with a radiological contaminant, (2) the tracer does not break down if radioactive sources are not present, (3) the decomposition products are not rapidly recombined to form the original compound after exposure to a radioactive contaminant or when no longer in the presence of the radioactive contaminant, and (4) the tracers are safe (i.e., nonflammable and nontoxic). All of these criteria were validated for the chosen RCUT tracer sulfur hexafluoride (SF<sub>6</sub>).

## RESULTS

The initial tracer gas chosen for RCUT was sulfur hexafluoride (SF<sub>6</sub>). SF<sub>6</sub> is a chemically inert gas that is both nonflammable, nontoxic, and breaks down when exposed to gamma radiation. The feasibility of using SF<sub>6</sub> as an RCUT tracer was tested by subjecting samples of SF<sub>6</sub> combined with O<sub>2</sub> and H<sub>2</sub>O (liquid and vapor forms) to various levels of gamma irradiation at the PNNL High Exposure Laboratory located in the Radiological Calibration and Standards Facility. Samples of various SF<sub>6</sub> concentrations both with and without O<sub>2</sub> or H<sub>2</sub>O additives, were prepared and then exposed to uniform <sup>60</sup>Co radiation at three levels of exposure (0.0258 C/kg, 0.258 C/kg and 2.58 C/kg)<sup>2</sup>. The exposed samples were compared to non-exposed samples. The measurements were made with a GC/MS.

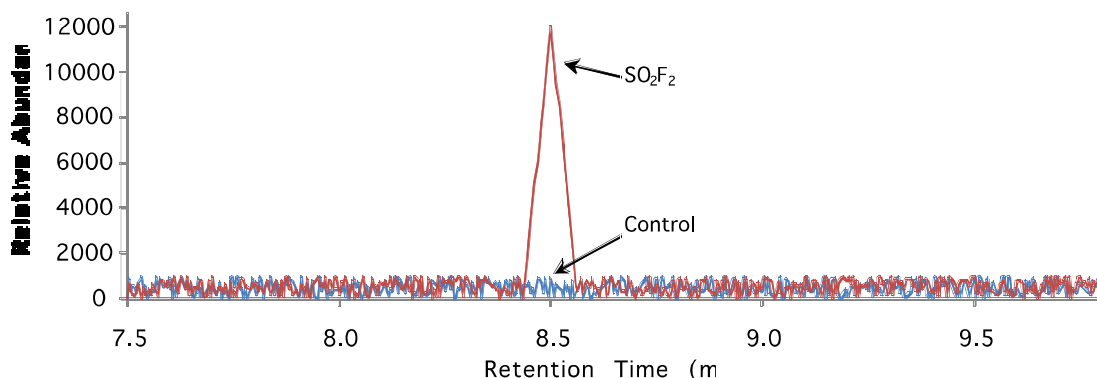
For operational implementation, the preferred gaseous tracers are comprised of SF<sub>6</sub> and O<sub>2</sub>, which forms SO<sub>2</sub>F<sub>2</sub> when exposed to radioactive contaminants. The method also works equally well when SF<sub>6</sub> and water vapor are used. When SF<sub>6</sub> is irradiated,

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<sup>2</sup> 1 R = 2.58 × 10<sup>-4</sup> C/kg of air.

chromatographs of the exposed samples indicate the presence of  $\text{SO}_2\text{F}_2$  as determined by a peak at a retention time of 8.5 minutes (Figure 2). The mass spec results [2] showed that the peak seen at 8.5 minutes has mass to charge ( $m/z$ ) ratios of 102 and 83 and are the results of the presence  $\text{SO}_2\text{F}_2$ . The peak at 102  $m/z$  represents the intact  $\text{SO}_2\text{F}_2$  molecule (the molecular weight of  $\text{SO}_2\text{F}_2$  is 102), and the peak at 83  $m/z$  represents the primary ionic breakdown compound of  $\text{SO}_2\text{F}_2$ . This was confirmed with an analysis of pure  $\text{SO}_2\text{F}_2$ .

The results of the RCUT tracer study showed the feasibility of using  $\text{SF}_6$  as a tracer gas to detect the presence of radioactive contamination through the measurement of predictable decomposition byproducts of  $\text{SF}_6$  radiolysis [2]. Significant progress has been made in the understanding of  $\text{SF}_6$  decomposition in a variety of exposure scenarios, including combinations of various products to influence and enhance the reactions such as  $\text{H}_2\text{O}$  and  $\text{O}_2$ . For operational implementation, a gaseous tracer comprised of a blend of  $\text{SF}_6$  and  $\text{O}_2$  is recommended.



**Figure 2.** GC/MS Chromatogram of both a irradiated sample and a non-irradiated sample (control) of  $\text{SF}_6$  with the irradiated sample showing a peak that corresponds to  $\text{SO}_2\text{F}_2$  whereas the control does not show any  $\text{SO}_2\text{F}_2$ .

## CONCLUSION

RCUT and PCUT are both effective technologies that can be used to detect contamination within pipelines without the need for mechanical or human inspection. These methods can be used to detect, locate, and/or estimate the volume of a variety of radioactive materials and hazardous chemicals such as chlorinated solvents, petroleum products, and heavy metals. While further optimization is needed for RCUT, the key first step of identification of a tracer compound appropriate for the application of detecting radioactive pipeline contamination through the detection of decomposition products of  $\text{SF}_6$  has been demonstrated. Other tracer gases that will also undergo radiolysis will be considered in the future. The next step for the RCUT development

process is conducting laboratory scale tests using short pipelines to define analytical requirements, establish performance boundaries, and develop strategies for lower exposure levels. Studies to identify additional analytical techniques using equipment that is more field rugged than a GC/MS would also be beneficial.

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