Innovative Decontamination Technology for Use in Gaseous Diffusion Plant Decommissioning

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ABSTRACT
The results of bench scale tests demonstrated that TechXtract® RadPro™ technology (hereinafter referred to as RadPro®) can provide 100% coverage of complex mockup gaseous diffusion plant (GDP) equipment and can decontaminate uranium (U) deposits with 98% to 99.99% efficiency. Deployment tests demonstrated RadPro® can be applied as foam, mist/fog, or steam, and fully cover the internal surfaces of complex mockup equipment, including large piping. Decontamination tests demonstrated that two formulations of RadPro®, one with neutron attenuators and one without neutron attenuators, could remove up to 99.99% of uranyl fluoride deposits, one of the most difficult to remove deposits in GDP equipment. These results were supplemented by results from previous tests conducted in 1994 that showed RadPro® could remove >97% of U and Tc-99 contamination from actual GDP components. Operational use of RadPro® at other DOE and commercial facilities also support these data.

INTRODUCTION
The United States Department of Energy (DOE) is seeking to develop state-of-the-art alternatives for removal of uranium and technetium deposits in the uranium enrichment equipment of the gaseous diffusion system at the Portsmouth (PORTS) Gaseous Diffusion Plant (GDP) in Piketon, Ohio. Current methods used to remove uranium/technetium deposits at PORTS GDP rely on gas-phased techniques that (1) are expensive to implement because the equipment needs to be operating, (2) are time-consuming because of the long durations required for deposit removal, and (3) fail to remove deposits in areas of poor gas flow. Also, existing techniques used at the DOE Oak Ridge site that rely on mechanical removal have risks to workers due to radiation exposures and potential fire hazard.

The DOE study consists of three phases. Phase I involves literature searches to compile and evaluate technical information on existing and emerging alternative approaches to decontamination and select the apparent best for additional testing, Phase II performs laboratory-scale testing on the selected technology judged superior in Phase I, and Phase III is a floor-level demonstration that the technology works on GDP equipment at PORTS. This paper discusses the results of Phase I and Phase II.

Background
The PORTS GDP is in Piketon, Ohio. The facility is operated by the United States Enrichment Corporation (USEC) under a lease agreement with DOE. The GDP was built between 1953 and 1956 to provide enriched and highly enriched uranium for military and commercial power industry use. The GDP site consists of three process buildings with total ground coverage of 93 acres. The entire site occupies 640 acres. The buildings, collectively, contain 2,820 separative stages. A total of 1,200 stages are shut down.

The processing equipment consists of stages arranged in series for the enriching process. Each stage consists of a motor, a converter (or condenser), a compressor, control valve, attendant cooling system, and associated piping. Stages are grouped together in combinations of eight, ten, or twelve stages depending on the size of the equipment. Each grouping is referred to as a cell. Ten cells constitute a unit and several units are located within each of the three process buildings. The processing equipment is referred to as cascades.

The processing cascades contain uranium compounds, technetium-99, and other fission products from enrichment activities and are unevenly deposited in the cascade system. These contaminants/deposits need to be removed to facilitate D&D activities and proper waste disposition. Removal of deposits is made difficult by the internal configuration of the equipment, interactions between contaminants and substrates, and the nature of the deposits. In addition to solid buildup of uranyl fluoride (UO_2F_2) and other contaminants, chemical and electrostatic bonding has occurred between the contaminants and the substrate surface. Another factor affecting decontamination is that contaminants have sequestered themselves within the void spaces of the substrate itself in micropores and/or microcapillaries. Disruption of the openings to these voids could result in the entrapment of contaminants below the surface, further complicating effective decontamination.

Purpose
The purpose of the study is to identify next-generation technologies that are safer, more cost-effective, pose significantly less hazards in a D&D environment, and are compatible with the ultimate disposition paths and disposal sites for materials generated as part of the cleanup efforts. This is accomplished through a comprehensive literature review of documentation available in the public domain, down selecting the apparent-best technology based on technical criteria developed as part of the study, and testing the selected technology in a laboratory scale demonstration.

LITERATURE REVIEW AND TECHNICAL EVALUATION
Technical evaluation criteria were developed in order to conduct a comparative evaluation of the various decontamination technologies available for GDP decontamination. The criteria used to evaluate technologies are grouped as follows:
- **Management Factors:** health hazards, fire potential, waste disposal, and transportation constraints
- **Economic Factors:** life cycle costs and energy consumption
- **Feasibility Factors:** ease of use, technology maturity and commercial availability
- **Performance Factors:** overall cleaning effectiveness, subsurface decontamination effectiveness, principle of decontamination, coverage/deployability, maintenance simplicity, and criticality controls

Table I presents the evaluation matrix used for scoring the different technologies.

Over thirty different decontamination technologies were evaluated which included mechanical techniques, wet chemistry techniques, gas-phased techniques, and non-traditional/emerging techniques. Both quantitative and qualitative comparisons were used to complete the down selection process and identify the alternative technology that provided the most advantageous use for GDP application. Safety and performance are considered the most important technical criteria for evaluating technologies for GDP application and are weighted accordingly. The safety factors include "Health/Safety Hazards" and "Safeguards for Criticality." Performance factors include "Effectiveness/Percent Contaminant Removal" and "Ability to Remove Subsurface Contaminants." Each of these technical criteria is given a weighting of 3X. The results of the evaluation are summarized in Table II. A more detailed discussion plus supporting references are presented in the Phase I Topical Report (1).

The wet chemistry technology using RadPro® chemical extraction/decontamination solutions emerged as the apparent best alternative. The RadPro® technology uses three separate patent-protected formularies used in sequence. It consists of highly buffered acidic solutions with specialized chemical and physical properties that utilize affinity shift mechanisms with carrier solutions to shift the equilibrium of radionuclides from the substrate to the carrier. The solutions migrate along grain boundaries to penetrate the micro-pores and micro-capillaries of the substrate (Kirkendall effect) and break the chemical and electrostatic bonds between the substrate and the contaminant. The RadPro® solutions then sequester the contaminants and activate capillary rise for effective removal and recovery (Young and Laplace equation).

RadPro® has a documented decontamination efficiency of >95% for numerous contaminants on a variety of substrates; presents minimal health hazards to workers (mild caustic), no fire hazard, and is criticality safe using neutron absorbers and industry-accepted controls. RadPro® is not regulated under the Resource Conservation and Recovery Act (RCRA) and is characterized by the contaminants removed from the affected substrate, can be applied

Table I. Evaluation Matrix for Gaseous Diffusion Plant Decontamination Attributes

Factors and Technical Criteria	Attribute Analysis	Score
Management Factors		
Health/Safety Hazards	• Moderately to highly toxic; risk of amputation or severe injury • Mildly toxic; physical burn • Non-toxic; negligible health or safety risks	1 2 3
Fire Hazard/Exothermic Potential	• Self-igniting; highly flammable • Supports combustion • Non-flammable; non-combustible	1 2 3
Regulatory Compliance/Waste Handling	• Prohibited disposal in regulated landfills • RCRA-regulated • Non-regulated	1 2 3
Transportation Constraints	• Public transportation prohibited • Requires special packaging • Suitable for over-the-road conveyance	1 2 3
Economic Factors		
Life Cycle Costs	• Requires operating system with attendant maintenance and repair for operability • Suitable for idle/out-of-service systems	1 3
Energy Consumption	• Requires operating system with attendant energy costs • Suitable for out-of-service systems	1 3
Feasibility Factors		
Ease of Use	• Requires disassembly prior to decontamination • Requires multiple technological steps (e.g., acid wash followed by grinding) • Suitable for *in situ* application	1 2 3
Maturity of technology	• Conceptual; research and development effort • Demonstrated on laboratory scale • Proven technology; record of performance	1 2 3
Commercial Availability	• Not available commercially • Special order product • Off-the-shelf commodity	1 2 3
Performance Factors		
Effectiveness/Percent Contaminant Removal	• <50% removal efficiency • 51% - 90% removal efficiency • >90% removal efficiency	1 2 3
Coverage Ability of Cleaning Agent	• Requires disassembly prior to decontamination • Subject to Shadow effects • *In situ* application provides complete coverage	1 2 3
Ability to Remove Subsurface Contaminants	• Does not remove subsurface contaminants • Requires abrading surface to remove subsurface contamination • Penetrates micro-pores, capillaries, micro-grains to remove subsurface contaminants	1 2 3
Maintenance Simplicity	• Complex deployment/equipment (e.g., robotics) • Large special-purpose equipment required; multiple technological steps for application (e.g., acid wash followed by grinding) • Easy deployment without high maintenance costs	1 2 3
Safeguards for Criticality	• Requires extensive criticality protection due to presence of liquid moderators • Criticality safe due to presence of neutron absorbers • Liquid moderators not used	1 2 3

Table II. Results of Down Selection Evaluation

Factors	Management				Economic		Feasibility			Performance					
Decontamination Type	Health/Safety Effects	Fire Hazard/ Exothermic Effects	Regulatory Compliance /Waste Handling	Transportation Constraints	Life Cycle Costs	Energy Consumption	Ease of Use	Maturity of Technology	Commercial Availability	Effectiveness Percent Removal	Cleaning Agent Coverage	Removal of Subsurface Contaminants	Maintenance Simplicity	Criticality Safeguards	Total Score
Mechanical:	X 3									X 3		X 3		X 3	
High Pressure Water	3	3	1	3	1	2	1	3	3	6	1	3	2	3	35
Ultra High Pressure Water	3	3	1	3	1	2	1	3	3	6	1	3	2	3	35
High Pressure Steam	3	3	3	3	3	1	1	3	3	3	1	3	2	3	35
Sponge Blasting	6	3	2	3	2	2	1	3	3	6	1	3	2	9	46
Strippable Coatings	9	3	3	3	3	3	1	3	3	6	1	3	3	6	50
Dry-Ice Blasting	6	3	3	3	1	2	1	3	3	6	1	3	2	6	43
Scabbling, Needle Guns, Surface Grinders	3	2	2	3	3	3	1	3	3	9	1	3	3	9	48
Gas Phase:															
Chlorine Trifluoride Capenhurst	3	1	1	1	1	1	3	3	2	6	3	6	1	9	41
Chlorine Trifluoride LTLT	3	1	1	1	1	1	3	3	2	6	3	3	1	9	38
Non Traditional/ Emerging Technologies															
Laser Ablation	3	2	3	3	1	2	1	1	2	9	1	6	2	9	45
Biological (geobacter sulfurreducens)	9	3	3	3	1	3	2	1	1	3	2	3	2	3	39
Metal Binding Ligands	9	3	3	3	2	3	2	1	1	3	2	3	2	3	40

Wet Chemistery															
Electrolitic	6	1	1	3	2	2	1	2	1	9	1	6	2	3	40
Formic Acid	3	1	2	2	3	3	1	3	3	3	1	3	3	3	34
Oxalic/Nitric Acid	3	1	2	2	2	3	1	3	3	3	1	3	3	3	33
Tartaric Acid	9	3	3	3	3	3	1	3	3	3	1	3	3	3	44
Nitric Permanganate	3	1	1	2	2	3	1	3	3	3	1	3	3	3	32
Turco Alkaline Rust Remover	6	3	2	2	3	3	1	3	3	3	1	3	3	3	39
Nitric Acid	3	1	2	2	2	3	1	3	3	3	1	3	3	3	33
Citric Acid	9	3	3	3	3	3	1	3	3	3	1	3	3	3	44
Alkaline Permanganate	3	3	2	2	2	3	1	3	3	3	1	3	3	3	35
Aluminum Nitrate	6	2	2	2	3	3	1	3	3	6	1	3	3	3	41
Nitric/Hydrofluoric Acid	3	1	2	2	2	3	1	3	3	6	3	3	3	3	42
Tech Extract	9	3	3	2	3	3	3	3	3	6	3	9	3	3	56
Radpro	9	2	3	2	3	3	3	3	3	9	1	9	3	6	61
Fluoroboric Acid	3	1	2	2	2	3	1	3	3	6	1	3	3	3	36
Cerium Nitrate	3	1	2	2	2	3	1	3	3	6	1	3	3	3	36
Corpex 921	0	0	3	2	3	3	1	3	0	9	1	3	3	3	34
Corpex 918	0	0	3	2	3	3	1	3	0	6	1	3	3	3	31
Hydrogen Peroxide	6	1	3	2	3	3	1	3	3	3	1	3	3	3	38
LOMI-Low Oxidation State	6	3	2	2	2	2	1	3	3	6	2	3	2	3	40

Note: A zero used in Table II denotes the attribute is not applicable/not available (NA)

in situ to idle GDP components and, thereby, does not require pre-decontamination disassembly nor operating equipment, thus eliminating the need for pre-decontamination radiation exposure protection, maintenance and/or repair costs.

Laboratory scale testing was performed to confirm the effectiveness of the RadPro® technology and its utility for use in GDP applications. The laboratory scale testing examined two aspects of decontamination:
1. Deployment effectiveness, defined as whether the RadPro® formulation and surrogate solutions could be delivered to the surfaces of the mockup GDP components with 100% coverage. Deployment methods were foam, steam, and mist/fog. Liquid delivery was not included as part of the deployment demonstration; however previous applications at other DOE and commercial sites have shown that liquid can be effectively used when applied with hand-held spray bottles and/or garden sprayers.
2. Decontamination effectiveness, defined as whether RadPro® formulations could remove greater than 95% of U and Tc-99 deposits and sub-surface contamination.

Results of these two testing efforts are discussed in the following sections.

DEPLOYMENT TEST RESULTS
The deployment tests successfully demonstrated the ability to (1) deliver RadPro® and surrogate solutions to the internal surfaces of mockup GDP equipment and provide 100% coverage of internal surfaces, (2) penetrate into the nooks and crannies of internally complex configurations, (3) retain contact with surfaces for sufficient dwell time, and (4) recover the spent cleaning agents.

The deployment testing was performed in February 2005 at an NRC licensed facility in Oak Ridge, Tennessee. The following test equipment was used for the demonstration:
- One glove box containing internal test fixtures.
- Two aluminum radiator style fixtures.
- One large piping fixture.
- Associated hoses, nozzles, piping, and valves.
- One commercial wet/dry spray steamer unit.
- One foam generator.
- Three vacuum units.
- One waste collection drum.
- One hand-held TURBOFOGGER fogging unit.

Glove Box Details
The glove box was constructed of Plexiglass™ panels secured with stainless steel framing, which comprised the 0.9 m x 0.9 m x 1.8 m (3 ft x 3 ft x 6 ft) housing unit. The clear Plexiglass™ sidewalls and ceiling allowed for viewing. The top of the glove box was hinged for placement of the simulated GDP component (i.e., the radiator) within the glove box. One of the sidewalls contained four glove ports to manipulate fixtures inside the glove box. The glove box housing unit rested on stainless steel legs. The floor of the glove box was graded for drainage. A stainless steel drip pan collected liquid wastes beneath the glove box. A 13-mm (0.5-in.) drain was positioned in the center of the drip pan to facilitate liquid collection. A 0.24-m^3 (55-gal) drum was used for waste collection. Access ports were located at each end of the glove box for introduction and recovery of cleaning solutions.

Pipe Fixture Details
The piping fixture was 4.8-mm (0.18-in.) carbon steel. The fixture was curved mid-way along its length forming a 0.9-m (3-ft) diameter radius elbow (90°) typical of any industrial piping system. The 5.2-m (17-ft) piping fixture consisted of three sections joined by flanges for disassembly and transport. Viewing windows were located along the pipe's length on both sides. External lights at two of the viewing windows illuminated the pipe interior for observation. Three other ports on the top of the pipe fixture served as injection ports, vacuum ports, and ports for make-up air. Two injection/recovery tubes extended from the ports to verify the area to be covered with RadPro® and surrogate solutions could be segregated and controlled. A butterfly valve was located near the entry port. The Plexiglass™ ends of the piping fixture contained ports for introducing and recovering solutions.

The glove box and pipe fixture deployment tests relied on vacuum systems for cleaning agent recovery. Pipe fixture topside piping and valving was configured so the ports could be used for foam injection and recovery. Two injection/recovery tubes were inserted into the pipe through the topside ports. The injection/recovery tubes descended into the interior of the pipe fixture to the bottom. A 0.24-m^3 (55-gal) metal drum was installed between the test fixture and vacuum system to capture the foam or liquid from steam. Supporting equipment consisted of a commercial wet/dry spray steamer unit, a foam generator, three vacuum units, and associated nozzles, hoses, and other conveyance devices.

All deployment tests were conducted using non-radioactive materials, (i.e., test fixtures, surfactants for foam, and tap water for fog and steam.) RadPro® solutions were used for the foam expansion tests, since it had not been deployed as foam in previous testing. However, RadPro® had been deployed as a mist and as steam at other DOE and commercial sites. RadPro® is an aqueous-based formula displaying the deployment properties of water. Therefore, in order to keep cost down, RadPro® solutions were not employed in the mist and steam testing since the deployment test purpose was to show surface contact ability and did not include demonstrating decontamination capability.

Fog/Mist Tests
A TURBOFOGGER fogging unit was used to demonstrate fog delivery. The TURBOFOGGER is manufactured by OWR, a German company that fabricates delivery systems for military application in decontaminating biological and chemical toxins.[1] The fogging unit was designed to dispense optimally sized aerosols for maximum dispersion and surface coverage in seconds.

Water was used in the demonstration to show dispersion capability. A fine fog was directed to the outside wall of the glove box to demonstrate wetting ability and coverage. The fog covered the panel and condensate was visible on the wall, indicating effective coverage. Large droplets were not observed; the wall was covered with a fine mist.

The fogging test proved successful since the wall of the glove box was covered with a thin layer of liquid. Fogging is most appropriate for large components with big void spaces and internally complex interiors. The aerosol particles distribute rapidly, especially within confined spaces, and result in a thin layer of solution in contact with interior surfaces.

Vendor design of the TURBOFOGGER allows the device to generate 3-micrometer (0.0001 in.) droplets, and deliver liquids within a confined system in a very effective manner. Research has shown that 3 micrometers (0.0001 in.) is the most effective droplet size for dispersion to transpire. This allowed RadPro® and the surrogate to enter all the nooks and crannies of complex equipment geometries.

Foam Tests with Radiator Fixture
Two tests were performed in the glove box using the radiator style test fixture. RadPro® solutions were used in the demonstration tests. In the first test, a clean radiator was inserted into the housing unit and secured in place. The dimensions of the radiator were 0.6 m x 0.6 m x 0.1 m (24 in. x 24 in. x 4 in.). The housing unit was sealed by closing the lid and latching in place. Foam was introduced at one end of the fixture while, simultaneously, vacuum was applied at the other end. The tubing entering and exiting the housing unit was clear so that the action of the foam could be observed. The introduced foam was carried into the housing unit by the combination of pressure from the spray nozzle and vacuum from the exit port. The foam expanded within the housing unit coating all of the radiator fin surfaces. The foam was drawn from the housing unit into the 0.24 m^3 (55-gallon) collection drum. The radiator was removed and visually inspected to determine the degree of coverage of all internal surfaces.

The test showed that the foam passed through the entire surface of the radiator without creating a preferential flow path as evidenced by the amount of foam present on all observable surfaces. Foam distribution was uniform throughout the radiator and exhibited 100% surface coverage.

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[1] The fogging unit was on loan to EAI Government Services for use in testing its suitability for dispensing RadPro® for radiological decontamination in the event of a Radiological Dispersion Device (RDD) incident.

The second test was performed to determine if the foam and simulated entrapped contaminants (indicated by red chalk) could be transported to a waste collection drum. The test consisted of applying colored chalk to one surface of a clean, dry radiator and inserting the radiator into the housing unit such that the chalk surface was nearest the foam entry port. Foam was again introduced and allowed to expand. Following expansion, the foam was drawn through the radiator via vacuum and captured in the collection drum. The chalk material was observed passing through the radiator as evidenced by the colored foam exiting the housing unit. In much the same manner, contaminants would be removed from the internal surfaces of the GDP equipment, sequestered in the RadPro® solutions and conveyed to a collection device. It is recognized that removing long-standing contamination differs from recently applied chalk; however, the surface removal, suspension, and conveyance of the chalk particles simulates contaminant behavior in the RadPro® solutions as demonstrated in previous DOE supported research in which uranium and technetium were removed from contaminated nickel surfaces of GDP components.

Steam Testing in Pipe Fixture
In this test, hot steam that contained a surrogate solution was introduced into the pipe test fixture through an entry port located at one end. The fixture was placed under negative pressure by means of a vacuum system attached at the opposite end of the fixture. Hot steam was introduced for approximately 20 seconds and allowed to coat the interior surface. The steam mist was conveyed throughout the entire 5.2-m (17-ft) length of the pipe fixture and coated the interior surfaces as was observed through the viewing windows. After the initial 20-second injection, additional steam was introduced to further coat the interior wall surfaces and to heat up the pipe. (Heat enhances the chemical and physical reactions of the RadPro® solutions resulting in faster and more effective cleaning.) After a short (20 to 60 minute) dwell time was simulated, the water condensate was removed through one of the topside vacuum ports and the main vacuum port at the end of the fixture.

The test successfully demonstrated that steam can traverse the length of the pipe and effectively coat the interior surfaces within 8 minutes. The direction of the steam mist was controlled by the flow of the air directed by vacuum and the amount of make-up air introduced into the system. The dispersal of the steam aerosol showed effective distribution that covered 100% of the interior surfaces without relying on the pressure stream from the nozzle, thereby eliminating shadow effect concerns. Test results showed that steam deployment of the RadPro® solutions could remove widely distributed contaminants present throughout the cascade system.

Foam Testing in Pipe Fixture
The foam test was performed to demonstrate that RadPro® could be injected as a foam, since this had not been demonstrated before. Another test objective was to ensure RadPro® solutions could cover an area and be controlled between two points, allowing the solutions to reach a simulated deposit. Two injection/recovery tubes were located 1.7 m (5.5 ft) apart with separate entry ports for each. Foam that contained RadPro® solutions was injected into the pipe at the first topside port through an injection tube that extended from the port to the bottom of the pipe. The foam was allowed to flow to the vicinity of the second tube.

Foam flow was facilitated by vacuum at the exit port at the opposite end of the pipe fixture. The flow of foam was controlled by installing a vacuum at the second topside port such that the plug of foam was positioned between the first and second injection/recovery tubes near the bottom of the fixture. In this way, the foam plug remained stationary with foam injection and removal near equilibrium. This configuration would allow localized placement of the RadPro® solutions (suspended in the foam plug) to react with a thick deposit for a sufficient dwell time and remove the deposit.

During all testing conducted within the piping fixture (foam, steam, and mist/fog) a vacuum system maintained the system under continuous negative pressure conditions. This is important during the deployment and recovery of decontamination materials since the negative pressure keeps the system from dispersing any displaced contaminants or from allowing decontamination agents to be released outside of the system. Pressure was continuously monitored using a magnahelic mounted on the side of the piping fixture. During the testing the vacuum was maintained at ~12.7 cm (5 in.) of water below standard atmospheric conditions.

The foam test successfully demonstrated that foam could be used to place RadPro® solutions on a simulated localized deposit and control dwell time for effective deposit removal. The application shown at the DOE-observed test demonstration was limited to deposits located on the bottom of the GDP component. Should the deposit be

located on the side wall or top interior of the component, then the entire volume of the GDP component would need to be filled with foam. This would require a larger foam unit if the component is as large as or larger than the pipe fixture.

Deployment Test Results

The deployment tests showed that RadPro® and/or surrogate cleaning solutions can be distributed and coat the interior surfaces throughout the text fixtures simulating GDP equipment. The three types of carriers tested (i.e., steam, foam, and mist/fog) provided effective coverage, as assessed by visual inspection. The test fixtures simulated large cavernous equipment (i.e., a pipe mockup) as well as internally complex structures (i.e., a radiator mockup with multiple fins and interstitial spaces and surfaces). Based on past experience and the observed behavior of the solutions in the fixtures, the solutions remained in contact with the surfaces sufficiently long for the physical and chemical reactions to occur that are required for decontamination. Spent solutions were recovered using vacuum collection systems that worked well for all carrier types.

DECONTAMINATION TEST RESULTS

Two sets of laboratory-scale tests were done to confirm the decontamination effectiveness of the RadPro® technology for GDP application. The 2005 tests, done as part of the Phase II study, demonstrated RadPro®'s ability to remove large uranyl fluoride deposits from metal surrogates. These tests were designed to simulate decontamination of large uranium deposits associated with in-leakage of moist ambient air into GDP components. Removal efficiencies of >98% were realized. The addition of neutron absorbers in the Phase II tests did not affect the RadPro® decontamination effectiveness. Tests performed in 1994 on actual GDP contaminated components demonstrated RadPro®'s ability to remove uranium and technetium surface scale and subsurface contamination. Removal efficiencies of > 97% were realized.

Phase II Decontamination Tests

The Phase II decontamination tests were conducted in May 2005 at the Materials and Chemistry Laboratory, Inc. (MCL) test facility in the Eastern Tennessee Technology Park (ETTP) in Oak Ridge, Tennessee. Coupons plated with uranium were decontaminated using RadPro® solutions to demonstrate contaminant removal ability. An additional aspect of the Phase II testing was to empirically show the incorporation of neutron attenuators (i.e., boron) did not affect the decontaminating ability of the cleaning agents.

The decontamination testing was conducted at the MCL facility located in building K1006 within the ETTP complex at Oak Ridge, TN. The facility was licensed by the U.S. Nuclear Regulatory Commission (NRC) to handle radioactive materials and was formerly a DOE laboratory supporting GDP activities. This laboratory was used to provide independent review and quality assured data.

Since actual deposits from a gaseous diffusion facility were not available, MCL developed deposits that were credible representatives of the authentic materials. Deposits found in DOE GDPs are uranyl fluoride deposits on various metal substrates. Bulk contamination is due to uranyl fluoride (at varying degrees of hydration), sometimes admixed at the immediate metal surface with some reduced uranium (typically UF_4 hydrate) and substrate fluoride corrosion products.

Uranium hexafluoride (UF_6) reacts rapidly with atmospheric moisture to produce uranyl fluoride (UO_2F_2) and HF:
$UF_6 + 2H_2O = UO_2F_2 + 4HF$

Uranyl fluoride hydrate ($UO_2F_2 * nH_2O$), a yellow-colored compound, is the form of uranium deposit most commonly found in GDP equipment. Uranyl fluoride hydrate is due to in-leakage of moist air during operation or after shutdown. For example, massive deposits (estimated up to 1,300 kg) at the K-29 Building of the former Oak Ridge GDP were primarily comprised of (partially) hydrated uranyl fluoride, developed as a result of moist air leakage into UF_6 gas process pipes. Hannon et al. (1998) examined these massive deposits by NDA, by minimally intrusive *in-situ* fiber optic camera, and also by intrusive examination of cut cross-sections. In general, deposits observed in piping were annular but somewhat irregular in cross-sectional distribution. The color of the deposit material (which is stated to be related to the hydration level and the residual HF content) was predominantly orange or yellow-green.

Based upon evidence from laboratory studies and from the examination of similar massive deposits, the orange-colored component is believed to be an intermediate oxyfluoride, such as $U_2O_3F_6$, which is a product of the incomplete hydrolysis of UF_6. The intermediate oxyfluoride hydrates are rather easily converted in moist air to form the usual UO_2F_2.

Detailed characterization was previously conducted on removed deposit material from the K-33 cascade at the Oak Ridge GDP. This analysis revealed the material was predominantly in the form of gravel-sized, hard yellow-green "chunks" and "flakes". The x-ray diffractogram for the sample was an excellent match for fully hydrated uranyl fluoride, $UO_2F_2.2H_2O$, as determined by comparison to the International Center for Diffraction Data (ICDD) standard reference pattern 47-0577.

Sample development for testing included deposition of the uranium materials on metal coupons that represent the three types of metal substitutes encountered in gaseous diffusion plants: mild steel, mild steel plated with nickel, and copper. Coupons were exposed to fluorine prior to deposition of the uranyl fluoride to duplicate fluoride corrosion products found on process parts removed from the gaseous diffusion plant environment. Coupons were then coated with a saturated uranyl fluoride solution and allowed to dry in a low humidity environment. Subsequently, the coupons plus deposit were cycled through alternating conditions of high and low relative humidity to duplicate conditions that result from in-leakage of moist air into the gaseous diffusion plants.

After exposure, three coupons were removed for examination by x-ray diffraction (XRD) to verify the chemical composition of the deposits Coupons were analyzed for total radioactivity to establish a baseline contamination level. Surfaces were then examined under a stereomicroscope and photographed to document starting conditions. Test coupons were maintained in a low humidity environment until testing started. Prior to decontamination, coupons were removed from the low humidity storage environment and weighed. Activity on the coupon surface was measured directly with use of a Tennelec S5E gas proportional counter (GPC).

The basic protocol for uranium decontamination using RadPro® was conducted in petri dishes. Dwell time was from 60 minutes to overnight. The following sequence represents one decontamination cycle:

- An initial 0300/0200 RadPro® blend was applied to the samples and allowed to dwell. (0300/0200 mix = 1 parts 0300 to 5 part 0200.)
- A second application of 0300/0200 RadPro® blend was applied to the samples and allowed to dwell.
- The samples were rinsed (rinse = 20% 0300 and 80% DI water).
- 0100 was applied to the samples and allowed to dwell.
- A final rinse was applied.

In addition to the protocol as described above, decontamination was conducted using a series of deployment methods and neutron attenuators. The deployment methods included applying a RadPro® formulation without neutron attenuators as a liquid (mist), steam, and foam. The liquid was applied as a mist from a spray bottle and a small hand held steamer was used for the steam applications. The foam application used standard class A-1 fire fighting foam and was generated by adding 3% foam agent to the decontamination chemicals in a Nalgene bottle and shaking the mixture. The foam was then applied to the samples. The entire sequence was then repeated using a RadPro® formulation that contained neutron attenuators (i.e., boron). This sequence included adding 3.3% sodium tetra-borate deca-hydrate ($Na_2B_4O_7*10H_2O$) to the 0100 and 5% boric acid (H_3BO_3) to the 0300/0200 mixture and rinsate.

The purpose of the decontamination tests was to show the level of effectiveness of two types of RadPro® decontamination formulations (with and without neutron attenuators) in removing uranium deposits that would be encountered within the GDP equipment. Additionally, the effectiveness of applying RadPro® as a spray/liquid, foam, and steam was evaluated to determine whether the deployment method would affect deposit removal.

During the decontamination tests, two decontamination cycles were performed on all test coupons. Table III shows the results of the testing using the RadPro® formulation without neutron attenuators.

Table III. Decontamination Test Results Using RadPro®Formulation without Neutron Attenuators

Application Type	Substrate	Initial DPM (Combined alpha and beta)	Final DPM (Combined alpha and beta)	% Removal
Spray/Liquid	Steel	2.42E+05	4,046	98.33
Spray/Liquid	Nickel	2.70E+05	1,446	99.46
Spray/Liquid	Copper	2.38E+05	1,428	99.40
Foam	Steel	2.56E+05	579	99.77
Foam	Nickel	2.60E+05	2,043	99.21
Foam	Copper	2.26E+05	2,196	99.03
Steam	Steel	2.47E+05	1,063	99.57
Steam	Nickel	3.08E+05	133	99.96
Steam	**Copper**	2.29E+05	**565**	99.75

As shown in Table III, the method of application did not measurably affect the level of deposit removal/decontamination. All of the deployment methods were able to remove from 98.33% to 99.96% of the uranyl fluoride deposit and significantly reduce the remaining contamination from the coupon surfaces.[2]

The testing was repeated using neutron attenuators and keeping all other parameters the same. Table IV shows the testing results.

Table IV. Phase II Decontamination Test Results Using RadPro® Formulation with Neutron Attenuators

Application Type	Substrate	Initial DPM (Combined alpha and beta)	Final DPM (Combined alpha and beta)	% Removal
Spray/Liquid	Steel	2.68E+05	704	99.74
Spray/Liquid	Nickel	2.68E+05	175	99.93
Spray/Liquid	Copper	2.35E+05	65	99.97
Foam	Steel	2.43E+05	902	99.63
Foam	Nickel	2.43E+05	1,341	99.45
Foam	Copper	2.37E+05	280	99.88
Steam	Steel	2.66E+05	4,074	98.47
Steam	Nickel	2.82E+05	38	99.99
Steam	Copper	2.54E+05	930	99.63

Table IV shows the addition of neutron absorbers did not measurably affect the level of deposit removal/decontamination. Regardless of the addition of neutron attenuators or method of deployment used, the decontamination formula was able to decontaminate 98.47% to 99.99% of the uranyl fluoride deposit and significantly reduce the remaining contamination from the coupon surfaces. In both cases the substrates could be decontaminated to free release levels, if desired, by conducting another decontamination cycle.

The 1994 Decontamination Test Result on Actual GDP Components
The July–August 1994 test results were obtained from studies conducted at Oak Ridge, Tennessee in which actual gaseous diffusion plant components were used. The 1994 tests successfully demonstrated RadPro®'s ability to effectively remove uranium and technetium deposits from stainless steel and nickel plated surfaces typical of cascade equipment. The tests were conducted in the July–August 1994 timeframe at the Oak Ridge K-25 facility. Three materials were decontaminated: a stainless steel valve disk and two nickel-plated valves. Results of the tests show that RadPro® achieved greater than 97% contaminant removal efficiencies for both alpha and beta activity.

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[2] It should be noted that due to the approximate 2.54 mm (0.1 in.) thickness of the deposit material, the initial combined alpha and beta DPM measurements were affected by self-absorption of the deposit material, making the initial measurement an underestimate of the actual combined alpha and beta reading. This makes the reduction in overall contamination even larger.

Table V shows the test results for the highest readings of alpha and beta contamination from the Oak Ridge tests performed in 1994. Complete test data are presented in the Phase II Topical Report (2).

Table V. The 1994 Test Results Summary for the Highest Contamination Levels

Substrate	Contaminant	Sample Location	Pre-Decon Levels (dpm)	Post-Decon Levels (dpm)	Removal Efficiency (%)
Stainless steel valve disk					
	Alpha	6	3,217	<29	99.1
	Beta	2	14,127,781	<284	>99.9
ValveVB-5					
	Alpha	13	2,631	<31	98.8
	Beta	4	189,796	<1,108	99.4
Valve VB-6					
	Alpha	4	599	<16	97.3
	Beta	4	52,600	<1,284	97.6

Criticality Evaluation Results
Results of the criticality analysis showed that the conventional controls of mass, volume, and geometry are applicable to criticality prevention when using RadPro® solutions for decontaminating GDP equipment. Volume and geometry appear to be best suited for GDP application in a D&D environment. Isolating GDP components and limiting the volume introduced in the component to sub-critical amounts of liquid is operationally feasible and can be safely implemented using standard field techniques. The cleaning solution coats the interior surfaces and, with time, condenses into the bottom taking the shape of the bottom surface of the affected component. Volume controls can be used successfully to prevent criticality concerns and still adequately coat the interior surfaces for effective decontamination. Geometry controls are best used for solution collection and recovery. The use of small diameter collection tubing and thin diameter collection tanks (e.g., pencil tanks) can be used to recover the spent cleaning solutions using vacuum systems after the contaminants are removed from the substrate surface.

To achieve an added level of protection, neutron attenuators can be included in the RadPro® solution without affecting its ability to remove contamination. Boronated RadPro® solution can be used to decontaminate GDP equipment. The addition of boron provides defense-in-depth for decontamination operations during D&D actions and the double contingency necessary for safe and effective GDP clean up.

Waste Management Considerations
Disposal of liquid wastes generated as part of the decontamination process is a significant concern in any decontamination project. The materials used in the RadPro formulation possess no hazardous, flammable, or reactive components or characteristics that would classify the spent RadPro® as hazardous waste other than the contaminants. As a result, the waste stream from a project can be characterized based solely on the contaminants extracted.

Previous experience with RadPro® solutions has shown that little liquid waste is generated (i.e., 0.01-0.03 gallons per ft^2 of surface area cleaned) when the solutions are applied as liquids (e.g., using hand-held spray bottles or garden sprayers) and recovered using vacuum systems. These wastes could be solidified and disposed in approved repositories. However, use of steam and foams for application in large systems such as GDP cascades could generate greater amounts of liquid. Deployment tests showed that, on average, about 1.5 liters (0.4 gal) per minute of free liquid was generated using the steam system employed during the laboratory scale demonstration. .

The foam demonstration tests indicate that, on average, 3.8 liters (1 gallon) of RadPro® liquid generated between 75 and 125 liters (20 and 33 gallons) of foam. Conservation of mass would indicate that, based on the foam expansion rate, the same ratio would apply to liquid generation. Hence, modest amounts of free liquids would result from the deployment of foam. For both the steam and foam applications, it is expected that the generation of free liquid could be managed such that the resultant amount of solidified waste per surface area cleaned is manageable.

CONCLUSIONS

Results of the Phase I literature review and down selection effort determined that the RadPro® technology has advantages over other conventional and emerging decontamination technologies for use in GDP applications. The results of Phase II laboratory-scale tests demonstrated that RadPro® technology can provide 100% coverage of complex mockup GDP equipment and can decontaminate uranium deposits with 98% to 99.99% efficiency. Laboratory tests of the RadPro® formulation with neutron attenuators also showed >98% removal efficiency.

RadPro® can be used to decontaminate the entire cascade as a stand-alone technology. However, its use could augment DOE's existing capability for GDP D&D. Its major advantages are:
- A short dwell time (from hours to several days) for contaminant removal, reducing cost and schedule.
- A non-hazardous chemical formulation, enhancing worker safety and waste management.
- Ease of application, allowing it to cover areas other technologies cannot reach.
- Inclusion of neutron attenuators (i.e., boron) in the RadPro® formulation, minimizing criticality risks.
- Generation of small volumes of contaminants, reducing waste disposal costs.
- Use on inoperable equipment eliminating the necessity to repair, test and start up the GDP allowing significant cost and schedule savings to be recognized.

Focused niche application of RadPro® would eliminate the current gas-phase methodologies used at PORTS and provide DOE with greater capability and flexibility for D&D. The addition of RadPro® technology to the DOE's existing decontamination methods would provide expanded capability at less cost.

REFERENCES

1. Alpha Group and Associates, LLC / EAIGS Team *Alternatives for Characterization and Removal of Deposits at Portsmouth Gaseous Diffusion Plant, Phase I Final Topical Report,* June 2004.

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