EMSL Strategic Plan to Maximize Scientific Impact of the Radiochemistry Annex

PEMP Notable Outcomes Goal 2.3

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July 2014

Prepared for the U.S. Department of Energy's
Office of Biological and Environmental Research
under Contract DE-AC05-76RL01830

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1.0 Introduction

The Radiochemistry Annex at the Environmental Molecular Sciences Laboratory (EMSL) is a new facility built to support scientific research that will enable cost-effective solutions to the U.S. Department of Energy's (DOE) challenges associated with legacy nuclear waste in the subsurface environment and the nuclear fuel cycle. The Annex affords real-time, *in situ* study and characterization of radionuclide speciation and complexation at the molecular scale, revealing the mechanisms of contaminant fate and transport in environmental samples. Likewise, tools in the Annex can be used to probe and understand the fundamental chemistry of radionuclides during separations and other chemical processes used in the nuclear fuel cycle.

This capability is one of three capital assets recently funded by the Office of Biological and Environmental Research (BER) to foster high-impact science for the benefit of BER's science missions, those of other DOE offices, and the greater scientific community. In order to jump start interest within the scientific community (particularly in the BER community) accelerate the impact of this capability, and maximize the scientific benefit of that investment, the Performance Evaluation Management Plan (PEMP) for fiscal year 2014 for DOE-Battelle Prime Contract for the Management and Operations of Pacific Northwest National Laboratory (DE-AC05-76RL01830) includes a Notable Outcome to develop and execute a strategy for high-impact science using this capability. This strategic plan, developed in the broader context of the overall EMSL Strategic Plan, serves as the deliverable for that PEMP Notable Outcome.

1.1 Goals

The goals by which the success of the capability will be measured are:

- **Maximize Scientific Impact** by ensuring that EMSL's new assets are effectively used for high- quality science that positions EMSL to be distinctive and by leading the science agenda in its Science Themes and Leadership Areas,
- **Ensure Strategic Alignment** with BER/DOE science mission areas,
- **Maximize Utilization** of the assets at a facility/capability level to maximize the benefit of the investment, and
- **Target Outreach** to cultivate new users and ultimately demonstrate leadership by using these capabilities to perform distinctive, high-impact science.

1.2 Document Organization

Section 2 of this strategic plan describes the science vision for the Radiochemistry Annex in the context of the Science Themes and Leadership Areas in the overarching EMSL Strategic Plan. Section 3 outlines a specific, actionable 2-3-year plan for achieving the goals in Section 1.1. Section 4 sets metrics for measuring performance against the goals. Section 5 establishes the vision for sustainment of this capability.

2.0 Science Vision for the Radiochemistry Annex

Historical weapons production activities and current and future nuclear energy production activities present several national, decadal challenges of direct interest to DOE that are well suited to the Radiochemistry Annex's capabilities. The engineering challenges include:

- Prediction of the movement and reactivity of radionuclides in complex and diverse terrestrial and subsurface environments
- Design of cost-effective remediation strategies for groundwater, soils, and sediments contaminated with radionuclides
- Selective removal of problematic radionuclides from chemical processing streams to reduce the environmental impact and cost of the nuclear fuel cycle
- Design of the next-generation nuclear wasteforms that enable lower-cost, long-term geologic storage.

Housing these complementary capabilities with sample preparation and analysis facilities in a contiguous laboratory space is rare and, in concert with EMSL's own technical leadership of research areas, will make EMSL the premier facility for researchers to address these science challenges.

EMSL's Radiochemistry Annex will provide the fundamental data and understanding necessary for 1) the development and parameterization of predictive fate and transport models used by policy makers and engineers for the design of cost-effective subsurface remediation strategies and 2) significantly increasing the ability to design separations processes for fission products and radionuclides in the nuclear fuel cycle and advanced wasteforms.

In the subsurface, wasteforms, and nuclear fuel cycle, the chemical behavior of radionuclides depends on the interplay of a complex array of variables, such as radionuclide oxidation state, the reducing or oxidizing conditions prevalent, pH, chemical solubility, and the presence and identity of complexants. The impact of these variables on chemical behavior and speciation of the contaminant can be determined using the suite of molecular-scale analyses housed in the Radiochemistry Annex. EMSL's spectroscopic capabilities (e.g., nuclear magnetic resonance, electron paramagnetic resonance, X-ray photoelectron spectroscopy, laser-induced fluorescence) provide molecular detail of the oxidation state, chemical bonding environment, identity of next nearest neighbors, and presence of complexing ligands. EMSL's advanced imaging capabilities (e.g., electron microprobe, scanning electron microscope, focused ion beam/scanning electron microscope, transmission electron microscope) provide spatially resolved quantitation of elemental abundance, mineral associations, and phase segregation at micron to sub-angstrom resolution. Housing these complementary capabilities with sample preparation and analysis facilities in a contiguous laboratory space is rare and, in concert with EMSL's technical leadership of research areas, will make EMSL the premier facility for researchers to address these science challenges.

2.1 Key Science Challenges

Some scientific challenges for radioactive elements are fundamentally similar to those of all elements, especially multivalent elements, in the natural environment—redox equilibrium, aqueous complexation, biological reactivity, formation of surface complexes, precipitation, and incorporation in solid phase materials. EMSL's Radiochemistry Annex is well equipped to study those phenomena. Consistent with the overall EMSL Strategic Plan, the Terrestrial and Subsurface Ecosystems Science Theme, and the proposed Leadership Areas in Hydrobiogeochemical Element Cycling, our vision is to perform high-impact science and lead the technical communities in three key scientific topics as they relate to the radiochemistry and the engineering challenges already described:

Radiochemistry. The fundamental processes that control the environmental reactivity of radionuclides include surface complexation, redox reactions, colloid formation, defect formation, and mineral associations. The Radiochemistry Annex's advanced spectroscopic and imaging capabilities were designed specifically to identify the molecular-level mechanisms that govern these processes and lead to predictive understanding of how they operate under varied terrestrial and subsurface environments. These same processes control the chemical behavior of actinides and fission products in separations, nuclear waste reprocessing, or incorporation in long-term wasteforms. In addition, both the inclusion of radionuclides in glass or crystalline host materials (natural or man-made) and the occurrence of radionuclides as nanoparticles can significantly affect chemical reactivity due to the formation of structural defects. The incorporation of radionuclides in mineral phases often creates distortion or defects in the crystalline lattice via a mismatch of ionic radius or valence, incorporation as interstitial atoms, and transmutation. Even in glass hosts, the incorporation of radionuclides at modest concentrations can lead to inhomogeneous distributions that affect glass performance. Similarly, numerous studies on non-radiological samples have shown that when materials occur as nanoparticles, their chemical reactivity can differ significantly from bulk material. The high concentration of defect structures on the surface of nanoparticles is thought to be a contributing factor in their enhanced reactivity.

EMSL's Radiochemistry Annex is ideally suited to measure both the defect structure and the resulting change in chemical reactivity. Defects are easily imaged using the advanced imaging capabilities in the Radiochemistry Annex. Additionally, the analytical chemistry suite is well suited to measure impacts on chemical reactivity through solubility experiments under tightly controlled environmental conditions. Through these means, EMSL will understand the fundamental mechanisms of environmental radiochemistry and become a leader in predicting, understanding, and controlling the complex systems involved in fate and transport of radionuclides in the environment.

Role of Organics in Radiochemistry. The formation of organic complexes with radionuclides forms the basis for separations chemistry in the nuclear fuel cycle and is significant in the fate and transport of radionuclides in the environment. Observed contaminant transport in the subsurface and rivers can depart significantly from predicted behavior due to the presence of organic molecules in these systems. Contaminants can form organic complexes that can prevent them from adhering to mineral surfaces, forming colloids or precipitating and facilitating their transport. Alternatively, they can become entrained in biofilms or soil organic matter that inhibits further dispersal. Currently, there is little ability to predict if organic complexes or colloids will have a significant impact on contaminant transport in a given scenario, leading to large uncertainties in performance assessment models. Lack of molecular characterization of the types of natural-occurring organic molecules has inhibited complete understanding of complex and colloid formation. Only operational definitions based on standard sequential extractions, which are known to change the native organic chemistry, are used to assess whether or not broad classes of organic molecules, such as humic or fulvic acids, are forming contaminant complexes. Scientists using EMSL's high-resolution mass spectrometry capabilities, which are ideal for molecular characterization of natural organic matter, can identify the specific or types of molecules that are binding to contaminants. To refine estimates of contaminant transport, high-resolution mass spectrometry examination of native soils then could be used to assess the levels of these organics in soils and the subsurface. With this insight, specific soil or sediment remediation strategies also can be designed to release contaminants bound by the identified organic molecules.

Nuclear magnetic resonance spectroscopy will complement the high-resolution mass spectrometry results by identifying the ligand moieties directly involved in radionuclide binding. In this regard, coupled mass spectrometry and nuclear magnetic resonance approaches also will afford development of new radionuclide organic complexes important in actinide/fission product separations strategy.

Technetium Chemistry. The element Technetium (Tc) displays rich and varied chemistry due to the range of oxidation states it can access and its ability to be present in both anionic and cationic forms under environmental conditions.

Understanding Tc speciation is crucial to several challenges relevant to BER's interests, including subsurface fate and transport, nuclear fuel cycle separations, and waste tank chemistry.

As an environmental contaminant, Tc is particularly problematic due to its long half-life and because it is predominantly present as the highly soluble anion pertechnetate (TcO_4^-). As pertechnetate, Tc migrates freely in groundwater with other negatively charged species, such as nitrate. The negative charge reduces the tendency for Tc to sorb to oxide surfaces or to be incorporated into precipitating secondary phases or alteration minerals. However, under reducing conditions such as those provided by Fe(II)/Fe(III) coupling or by microbial respiration, pertechnetate is reduced to the poorly soluble oxide $Tc(IV)O_2.nH_2O$. In the Tc(IV) oxidation state, Tc can be incorporated into a variety of Fe-oxides commonly found in subsurface environments. However, the long-term stability of these Fe-oxides containing Tc requires additional study. Determination of the Tc speciation and heterogeneity and localization of geochemical gradients in the subsurface is essential for accurate prediction of Tc fate and transport for repository and waste remediation strategic planning.

The presence of Tc as the anion pertechnetate (TcO_4^-) creates challenges for vitrification in low-activity waste, or LAW, streams due to the high volatility within the melter environment. Two approaches to entrain Tc in melt involve maintaining Tc in a reduced oxidation state by sequestering it in a reduced solid phase or maintaining the entire melt chemistry at a reduced oxidation state. If Tc is successfully incorporated in a sequestering-reduced phase during LAW vitrification, its influence on wasteform performance is not well understood. Maintaining the entire melt under reducing conditions likely will alter the chemistry of other radionuclides and problematic metals in glass chemistry, such as copper (Cr), uranium (U), and plutonium (Pu). Under either scenario, there are long-term wasteform performance challenges that require understanding the chemical form of radiological and non-radiological constituents in the wasteform, as well as their impact on wasteform performance and upon their release in the geologic setting of the repository.

Scientists using EMSL's Radiochemistry Annex will be able to determine the chemical speciation and evaluate the impact on wasteform performance using advance spectroscopic (e.g., nuclear magnetic resonance or X-ray photoelectron spectroscopy) and imaging (e.g., transmission electron microscope or electron microprobe) techniques. The Radiochemistry Annex is particularly suited to address Tc speciation due to the high sensitivity and spectral resolution of the 750 MHz nuclear magnetic resonance, which provides information about the Tc oxidation state, composition and structure of ligands in solution, or coordinating atoms in solids. The stability of Tc-containing Fe-oxides can be approached through solubility studies, X-ray photoelectron spectroscopy, and advanced imaging techniques—all available within the Radiochemistry Annex.

The goal is that within the next 2-3 years, we will publish impactful scientific results in these three areas in top-tier journals resulting from the unique integration of complementary experimental probes that provide comprehensive understanding of the molecular processes needed for accurate predictive models of contaminant geochemical behavior. Through journal publications, workshops, and presentations at national meetings, we will demonstrate leadership, attract leaders in these fields as users, and increase overall utilization of the Radiochemistry Annex.

3.0 Strategic Approach

EMSL is implementing a multi-pronged approach to achieve the goals and science vision discussed in Section 2, and thereby generate *high-impact science*, increase *alignment* with BER/DOE science mission areas, maintain high rates of facility *utilization*, and establish *leadership* within the scientific community through distinctive science and scientific capability. Several of the actions within the approach are highly tactical and short-term, with the intention of immediately increasing the external visibility of the capability and its utilization by projects aligned to the vision. Others are longer-term efforts, guided by our overall scientific vision for EMSL, to lead and drive scientific progress within specific technical communities. Both types of actions are described in this section, and many immediate actions are complete or under way.

3.1 Immediate Actions

3.1.1 External validation through advisory group input

The Radiochemistry Annex has an advisory committee that includes members from the EMSL Science Advisory Community, science leaders, and experts in radiological sample handling and experimental design.

Radiochemistry Annex Advisory Committee

Advisory Member	Institution	Role	Research Focus
Andrew Felmy	PNNL	Chair	Radionuclide thermochemistry and modeling
Sue Clark	Washington State University	EMSL SAC member	Actinide separations chemistry
Ian Farnan	Cambridge	EMSL SAC member	Radiation effects in oxide materials
Steve Conradson	LANL[1]	BES Heavy Element PI	Application of synchrotron radiation to radionuclide research
Lynda Soderholm	ANL[2]	BES Heavy Element PI	Actinide chemistry
John Bargar	SSRL[3]	BER-SFA PI	Radionuclide environmental chemistry
Annie Kersting	LLNL	BER-SFA PI	Heavy element chemistry
Edgar Buck	PNNL	Technical specialist	TEM of radionuclides
Herman Cho	PNNL	Technical specialist	MAS-NMR of radionuclides

Advisory Committee members have been asked to provide the following guidance and validation:

- Vet and enrich the science vision,
- Identify near-term scientific opportunities to produce high-impact science and key scientists who are potential new users, and
- Review proposals for quality and alignment with the vision.

[1] Los Alamos National Laboratory
[2] Argonne National Laboratory
[3] Stanford Synchrotron Radiation Lightsource

In addition, the advisory group was asked to help promote the capability within the research communities they represent; target and cultivate high-impact users; and facilitate outreach opportunities, including themed workshops or symposia.

3.1.2 Review of existing EMSL projects for acceleration/additional access

EMSL has reviewed the existing user projects and proposals submitted through the 2014 Call for Proposals. Projects and proposals with clear BER alignment and potential for high-impact science (listed in the "Candidates from Existing User Projects" table) are being approached for additional attention, staff time, funding, and priority access to key instrumentation to accelerate their progress and heighten the probability of success.

Candidates from Existing User Projects

Researcher	Institution	BER Role	Focus
Peter Burns	Notre Dame	BER PI	Fundamental factors affecting the co-precipitation of $U(VI)O_2^{2+}$ and $Np(V)O_2^{2+}$ in low-temperature carbonate, sulfate, and borate mineral phases.
Jon Chorover	University of Arizona	BER PI	Predicting reactive contaminant migration in Hanford sediments that have received acidic uranium and strontium-bearing waste streams from these cribs.
Hailiang Dong	Miami University	BER PI	Fe speciation in Fe-bearing phyllosilicates resulting from multiple cycles of Fe redox and the subsequent reactivity for U and Cr reduction.
John Zachara	PNNL	BER SBR/SFA PI	Molecular-scale understanding of mineral surfaces that mediate redox processes in the subsurface and fundamental understanding of biochemical, biotransformational, and transport processes.
Harish Veeramani	Virginia Polytechnic Institute	BER co-PI	The role of framboidal-pyrite-associated biofilms in the sequestration of uranium.
Zheming Wang	PNNL	BER co-PI	Bonding, energetics, and excited states of uranyl compounds key to spent nuclear fuel and U(VI) remediation using combined experimental and computational techniques.
Ian Farnan	Cambridge	UK EPSCR/ NDA funded	Alteration rates in spent nuclear fuel in contact with repository groundwater and the retention of radionuclides in the alteration products.
Alexandra Navrotsky	University of California, Davis		Determine oxidation states and occupancy of cerium, uranium, and iron in the garnet structure, a candidate wasteform for radionuclides.
Sue Clark	Washington State University		Solvation of trivalent actinides and lanthanides in aqueous to aqueous methanol solutions in the presence of simple organic carboxylate ligands.

3.1.3 Special Call for Proposals

A Special Science Call (http://www.emsl.pnl.gov/access/calls/science_call/) opened in April 2014 for research proposals in selected topics that would be enabled by utilization of all three new capabilities (the Radiochemistry Annex, Quiet Wing, and High Resolution and Mass Accuracy Spectrometry Capability). Proposals are being reviewed as received to accelerate scientific advancement and impact. To increase project's alignment to high-impact, BER-relevant science, principal investigators are being contacted, cultivated, and encouraged to respond to the Special Call. Candidate projects are organized in "High-impact, BER-aligned Target Users" table.

High-impact, BER-aligned Target Users

Researcher	Institution	BER Role	Prior EMSL User?
John Bargar	SLAC	SBR SFA lead	Yes
Dan Kaplan	SRNL	SBR co-PI	Yes
Ken Kemner	ANL	SBR SFA lead	No
Annie Kersting	LLNL	SBR SFA lead	No
Brian Powell	Clemson University	SBR PI	No
Mavrik Zavarin	LLNL	SBR SFA lead	No
Udo Becker	University of Michigan	None	Yes
Dave Clark	LANL	None	No
Abe Clearfield	Texas A&M University	None	No
Rod Ewing	Stanford University	None	No
Wayne Lukens	LBNL	None	No
Bruce Moyer	Oak Ridge National Laboratory	None	Yes
Heino Nitsche	University of California, Berkeley	None	Yes
David Shuh	Lawrence Berkeley National Laboratory	None	Yes

3.1.4 Outreach and Longer-Term Action Plan

The broader scientific community also will be engaged through technical meetings and conferences, symposia, and workshops to raise visibility of the Radiochemistry Annex's capabilities. In the past, both oral presentations of science results and poster presentations highlighting EMSL capabilities have been successful mechanisms for reaching new users and strengthening scientific collaborations. In the near term, EMSL will promote the Radiochemistry Annex at several upcoming scientific meetings that are well aligned to the science vision and user base. The "Targeted Meetings to Promote the Radiochemistry Annex" table denotes these opportunities.

Targeted Meetings to Promote the Radiochemistry Annex

Fiscal Year 2014 BER PI Meetings and Workshops	**Dates**
DOE-Terrestrial Ecosystem Science/Subsurface Biogeochemical Research Joint PI Meeting	May 6-7, 2014
Fiscal Year 2014 Workshops and Regional Meetings	
EMSL Technetium Chemistry	July 24-25, 2014
Fiscal Year 2014 Society Meetings	
Actinides Separation Conference	May 23-26, 2014
Goldschmidt Conference	June 8-11, 2014
American Chemistry Society – *plan to organize special session on Tc chemistry in 2015*	August 10-14, 2014
Plutonium Futures	September 7-12, 2014
Fiscal Years 2015-2016 Meetings	**Dates**
American Association for the Advancement of Science (AAAS); *proposing special session on "*Imaging Molecules in the Environment*" in 2015*	February 12-16, 2015
Actinides Separation Conference	May 2015
American Chemistry Society – *plan to organize special session on Tc chemistry in 2015*	August 16-20, 2015 August 21-25, 2016
American Society for Microbiology (ASM)	May 30-June 2 2015 May 2016
Plutonium Futures	June 7-11, 2015 June 12-16, 2016
Goldschmidt Conference	June 8-13, 2014 August 16-21, 2015 (with permission) June 26-July 1, 2016 (with permission)
International Society for Microbial Ecology (ISME)	August 2016
DOE-Terrestrial Ecosystem Science/Subsurface Biogeochemical Research Joint PI Meeting	2015
EMSL Technetium Chemistry	2015
Migration Conference – *plan to organize special session on integration of experiment and computational chemistry in 2015*	2015

EMSL also will host a series of workshops to identify science challenges and showcase the Radiochemistry Annex. The first workshop will feature Tc chemistry and will be held in July 2014. Preliminary plans include hosting a workshop to explore science challenges involving the interplay between organic complexants, microbial communities, and radionuclide environmental chemistry in fiscal year 2015.

Furthermore, EMSL will conduct outreach to potential users within programs funded by DOE's Offices of Environmental Management, Basic Energy Sciences, and Nuclear Energy by asking their program managers to promote the Radiochemistry Annex in upcoming proposal calls or periodic program mailings, and/or by helping to publicize the Special Science Call. Philip Wilk of DOE's Office of Basic Energy Sciences Heavy Element program has toured the Radiochemistry Annex and sent a brochure on the Radiochemistry Annex along with information regarding this Special Call to Mark Gilbertson and Karen Skubal, both of DOE's Office of Environmental Management.

Over the longer term, as lead time for planning permits and both the user base and the EMSL scientific base expand, EMSL will host on-site workshops and conduct specialized sessions at national society meetings to bring together researchers interested in science enabled by the capability. Ultimately, these workshops will be opportunities to achieve not only visibility but recognition of EMSL's unique abilities and contributions to the areas described in the science vision. Also, these forums will create excitement within the user community to collaborate with EMSL. For example, plans are underway to organize a symposium at the American Chemical Society 2015 national meeting, highlighting research conducted in the Radiochemistry Annex.

4.0 Target Metrics

The long-term goals and outcomes of this effort are for these capabilities to truly distinguish EMSL and its science to BER and the broader scientific community—for EMSL to be seen as the go-to place, or BER hub, for sophisticated radiological characterization and study for difficult scientific questions. A leading indicator of success will be that this capability soon joins other EMSL capabilities, such as mass spectrometry, nuclear magnetic resonance, and computation, which are routinely over-subscribed and sought-after. Additional indicators will be recognition of these capabilities in major invited talks (both by EMSL staff and users), review papers that highlight EMSL's pre-eminent capability, leadership in forward-looking capability/science workshops, and inclusion in National Academy or other reports and projections on science and technology direction. EMSL will strive to reach this level of accomplishment within the next three years and to sustain such preeminence for the next decade and beyond.

We propose a set of specific metrics aligned to the four goals listed in Section 1.1. There are four primary metrics well aligned to the goals; however, some of these (in particular, publications and special sessions at symposia) tend to be lagging indicators. In these cases, secondary metrics that are more leading or real-time indicators are also proposed to help manage performance against the primary metrics.

- Science impact:
  - Primary: High-impact publications featuring the capability in the Radiochemistry Annex
  - Secondary: Number of BER-focused pre-publication science accomplishments submitted
  - Secondary: Number of distinguished participants on user projects
- Utilization: General utilization of major equipment in the Radiochemistry Annex at the capability level
- Alignment: Utilization by projects aligned to BER missions and interests
- Outreach and leadership:
  - Primary: Number of workshops, special sessions, tutorials held
  - Secondary: Number of proposals submitted by potential users.

These metrics are benchmarked against year-to-date performance in the "Metrics and Targets" table.

Metrics and Targets

Primary Metrics are in **bold**. These metrics are well aligned, but some tend to be lagging indicators.

Secondary metrics are in *italics*. These are not the direct focus, but are more leading indicators.

Goal	Metric	FY14 YTD (Q2)	FY14 Target	FY15 Target	FY16 Target
Science Impact	**High-impact publications**[1]	3	6	8	12
	Number of BER-focused pre-publication science accomplishments submitted	0	3	6	6
	Number of distinguished participants in user projects	6	8	10	12
Utilization	**General utilization**[2]	39%	50%	75%	75%
Alignment	**BER-aligned utilization**[3]	20%	40%	50%	65%
Outreach and Leadership	**Number of workshops, special sessions, tutorials held**	0	1, July 2014	1	1
	Number of proposals submitted by potential users	15	20	20	25

1 Defined as number in top 10% journals.
2 Defined at the capability level for radiological work across all major instruments based on available operating hours (hours used/hours available as a percentage).
3 Defined as percent utilization by projects aligned to BER science interests (a value of 4+ on alignment during proposal review). May be funded by other agencies.

5.0 Sustainment

This strategic plan focuses on accelerated utilization and impact of Radiochemistry Annex over the next 2-3 years. This capability will require continued stewardship as it matures (i.e., 4-8 years and beyond). The EMSL Strategic Plan will include stewardship needs, further outreach, and other leadership actions for this capability. Annual measures will be added to EMSL's manual dashboard. Furthermore, EMSL commits to implementing similar approaches for accelerating new capabilities in the future.