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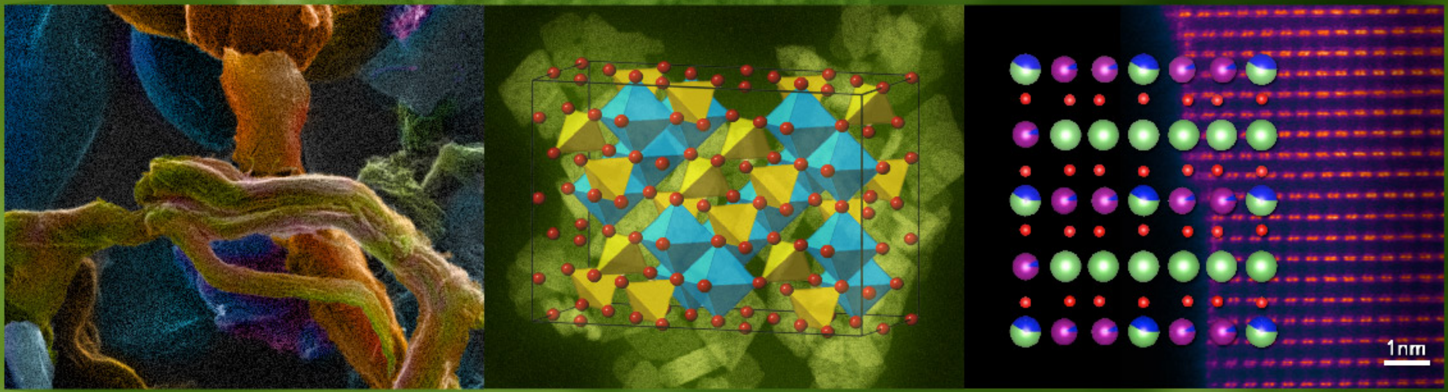
EMSL

## Energy Materials and Processes

# An EMSL Science Theme Advisory Panel Workshop

Workshop Dates
July 7-8, 2014

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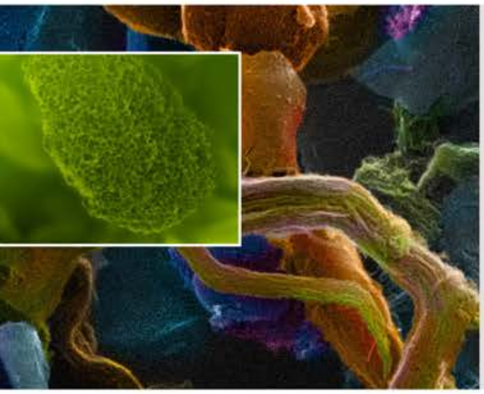
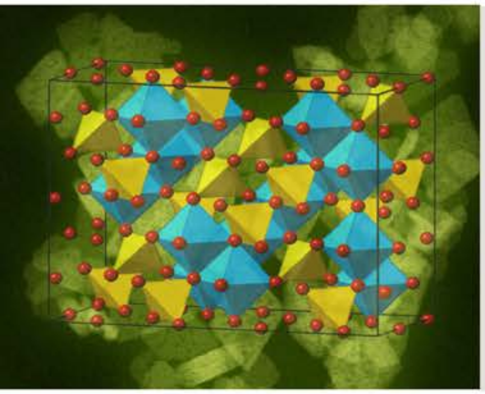
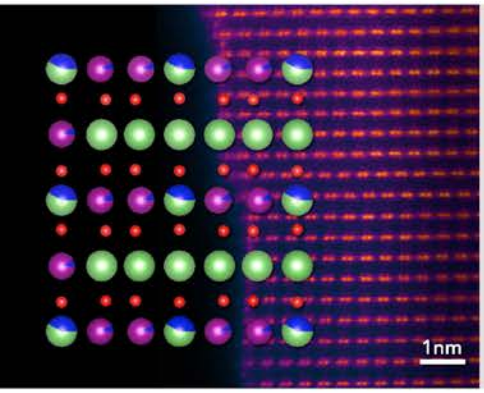
# About the cover



A colorized helium microscope image shows the structure of a novel porous carbon composite electrode that is being studied for use in a supercapacitor. A supercapacitor is an energy storage method that can deliver a sudden burst of power in a very short time. This research is critical for low-cost, higher energy, and power density storage technology for hybrid cars, electric trains, airplanes, computers, and even smart phones.



The crystallographic structure and reactive surface sites of transitional alumina catalysts and catalyst supports were an unresolved problem for 50 years, but has recently been solved combining electron microscopy, nuclear magnetic resonance, and computer modeling. The derived crystallographic model of delta-Al2O3 is shown as an overlay of an electron micrograph.




Electron microscopy has provided information about the atomic structure and chemistry of a lithium- and manganese-rich nickel oxide electrode for lithium ion batteries. Understanding the electrode structure and chemistry, especially their dynamic evolution during charging and discharging, can help researchers address the challenge associated with battery capacity and voltage fading.

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# Energy Materials and Process

# An EMSL Science Theme Advisory Panel Workshop

Workshop Dates: July 7-8, 2014

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

Pacific Northwest National Laboratory
Richland, Washington 99352

# Acronyms and Abbreviations

| | |
|---|---|
| BER | Office of Biological and Environmental Research |
| D-TEM | dynamic transmission electron microscopy |
| DEMS | differential electrochemical mass spectrometry |
| DNP | dynamic nuclear polarization |
| DOE | U.S. Department of Energy |
| EFRC | Energy Frontier Research Centers |
| EMSL | Environmental Molecular Sciences Laboratory |
| EMP | Energy Materials and Processes |
| FT-MS | Fourier transform mass spectrometry |
| IIC | Institute for Integrated Catalysis |
| MD | molecular dynamics |
| MSC | Molecular Science Computing |
| NMR | nuclear magnetic resonance |
| PNNL | Pacific Northwest National Laboratory |
| SciDAC | Scientific Discovery through Advanced Computing |
| SIMS | secondary ion mass spectrometry |
| STAP | Science Theme Advisory Panel |
| TEM | transmission electron microscopy |
| XPS | x-ray photoelectron spectroscopy |

# Contents

# 1.0 Introduction

The Environmental Molecular Sciences Laboratory (EMSL) is a national Biological and Environmental Research (BER) U.S. Department of Energy (DOE) user facility with an objective to accelerate scientific innovation for DOE by:

- focusing science on critical problems,
- providing forefront enabling capabilities,
- assembling teams of staff and users with essential expertise to address challenges, and
- marketing these results and capabilities for immediate impact.

Starting in 2012, EMSL initiated a process of realigning and refocusing Science Themes with updated priorities of BER, DOE, and with societal needs. Today EMSL has four BER-aligned Science Themes to focus its scientific resources and user program on the discovery of molecular-scale solutions to pressing challenges in biological, environmental, and energy research:

- Biosystem Dynamics and Design
- Atmospheric Aerosol Systems
- Terrestrial and Subsurface Ecosystems
- Energy Materials and Processes (EMP).

The EMP was identified as one of the EMSL Science Themes for relevance to BER and DOE, and fits with EMSL capabilities. Sustained economic progress for the nation depends on sustained availability of secure, reliable, and affordable energy supplies. Energy and environmental issues relevant to energy are central to BER and DOE missions. New energy technologies are necessary to address three great challenges: 1) the danger to political and economic security; 2) the risk to the global environment from climate change; 3) the lack of access by the world's poor to modern energy services needed for economic advancement. These critical needs have triggered research and technology development around the world. Over the past year we have identified areas where we believe EMSL can enable important progress.

Several research areas needed for development of new energy materials fall within historic EMSL research strengths, including the key role of interfaces, the relevance of oxide films and surfaces, and the importance of catalytic processes. Interfacial chemistry and new materials are priority areas in battery research where focus on charge and mass transport at interfaces, over-potential issues, and the nature of solid/electrolyte interactions is needed for both more advanced understanding and development of predictive models. It was noted that improving the efficacy of catalysts for producing or upgrading biofuels or bio-derived chemicals would be a significant step toward enhancing the economic viability of biofuel production. BER's Bioenergy Centers have the technology goal of enabling sustained and viable biofuel technologies through a better understanding of the biological mechanisms underlying biofuel production. Catalysis and bio-catalysis could play a useful role in enhancing the quality and value of biofuels, and other value-added bioproducts.

EMSL has significant and growing user and research activity in energy storage materials, such as batteries and fuel cells, as well as catalytic processes including upgrading bioproducts and energy conversion. Both batteries and catalytic upgrading of bioproducts require understanding and controlling liquid-mediated interfacial processes. While developing challenging goals for EMP, we identified understanding the complex role of solvents in reactions and interactions at solid/liquid interfaces in energy storage and catalytic processes as an important target or challenge area for high-impact EMP research. The initial focus is in two important areas and builds upon current research:

- Optimize catalytic processes to enhance the production and quality of biofuels and renewable chemicals.
- Provide a predictive understanding of the molecular processes needed to design advanced energy storage materials (such as batteries).

Science Theme Advisory Panels (STAP) of leaders in the research community provide input to EMSL regarding areas of science where the user facility can significantly enhance scientific progress, types of capabilities needed to advance that science over the next decade, and the best ways to engage these capabilities to make progress. The EMP Science Theme encompasses parts of science included in the former Science of Interfacial Phenomena Science Theme. Although longer-term goals and target areas described above were developed through interactions with the user community and seemed appropriate for the evolving research portfolio in EMSL, we asked this particular STAP to critically examine these directions in light of current science and needs, and recommend either alteration or refinement of these directions to enhance the overall impact of EMSL. It was equally important to address whether EMSL has or could develop expertise and capabilities to enable significant progress.

Therefore, to more fully define the role of EMSL in energy research, we asked the STAP for input in the following areas:

- **Challenges, Gaps and Opportunities**: What are the most critical science gaps/challenges? What space should EMSL help fill? Is our 5-10 year focus/target appropriate and how could it be improved? Should intermediate objectives be identified?
- **Needed Capabilities**: What capabilities are needed to address the identified challenges? How might current capabilities be best deployed to address needs? What new capabilities are needed to move science forward over the next decade?
- **Critical Teams**: What teams need to be assembled to enable progress? What mechanisms should EMSL employ to assemble the needed sets of skills and expertise?

# 2.0 Workshop Overview

The workshop agenda was constructed to encourage researchers with a variety of expertise to provide input on the desired topics. The workshop was made up of a series of sessions to answer a set of high-level questions:

- Is the EMP leadership target (understanding and controlling solvent-mediated interfacial processes in sustainable catalysts and batteries) appropriate for EMSL, DOE, and the nation? If not, what would be a better target to pursue?
- Within the agreed upon target areas, what are the most critical science gaps/challenges? What space should EMSL work to help fill? Should intermediate objectives be identified?
- What capabilities are needed to address the identified challenges? How should current capabilities be best deployed to address needs? What new capabilities would move science forward over the next decade?
- What teams should be assembled to enable progress? What mechanisms should EMSL employ to assemble the needs sets of skills and expertise?

This report summarizes the workshop and outlines recommendations that will be used to plan EMSL's strategic investments in new facilities and development of scientific expertise for the EMP Science Theme. It will also influence the EMSL calls for research proposals in 2015 and in the future.

## 2.1 Plenary and Introductory Talks

The workshop began with an introduction and outline of the charge and objectives for the meeting. EMSL Interim Chief Science Officer, Karl Mueller, presented the overall context and described the four EMSL Science Themes, how EMSL fits into Pacific Northwest National Laboratory (PNNL), and where the EMSL User Program sits within BER. EMSL's strategic planning process was discussed, including input from STAPs as well as other advisory groups, BER, and the EMSL-appointed Wiley Research Fellows. EMP Science Lead and STAP Co-chair, Don Baer, provided some history of the EMP Science Theme and how it has evolved to meet changing needs and BER research priorities.

The Associate Director of PNNL's Institute for Integrated Catalysis (IIC), Chuck Peden, gave an introduction to the IIC, which has more than 100 PNNL staff participants and ranges from fundamental to applied research. In particular, he highlighted how basic research performed by the IIC has benefited greatly from EMSL collaboration, and the overall history and continued cooperation between the two organizations.

A plenary talk from STAP Co-chair and member of EMSL's Science Advisory Committee, Mark Barteau, provided further perspective drawing on both recent history and allied science initiatives. Common themes of rational design and predictive control were noted in both EMSL's goals and DOE reports, and discussion documents of research needs. While considerable progress has been made over the past decade in the design of functional materials, including catalysts, driven by computational advances, significant challenges remain in design, synthesis, and understanding performance in complex reactive environments. Parallels were highlighted between EMSL priorities and the Materials Genome Initiative strategic plan. These included requirements for *in situ* spectroscopic and microscopic techniques, an understanding of interfacial reaction mechanisms and new computational modeling tools. The panel stressed the need to be aware of the broader impact of research since the importance and support of individual research topics (e.g., biomass conversion) may wax and wane over time, while the underlying principles of design and control are more enduring and can be applied in many different contexts. Opportunities for EMSL to make an impact in the EMP Science Theme were also emphasized by focusing on critical science challenges and by incorporating a ‘model, make, maintain, and measure’ approach in the study of complex interfacial phenomena. Following the plenary talk, workshop members were invited to give short presentations to share their thoughts on scientific gaps, challenges, and needs. Many ideas that were presented are incorporated into the breakout session reports summarized in Section 3.

## 2.2 Breakout Session 1: Scientific Gaps

Several topics and issues were discussed in detail during a breakout session designed to identify critical research areas, important areas for EMSL activity, and challenges that must be addressed. The workshop participants were divided into two groups focusing on the following topics:

- Catalysis

  Mark Barteau, Chuck Peden, Susannah Scott (discussion lead), Chris Jones (discussion lead), Cathy Tway, Roger Rousseau, Rick Orth, John Holiday, Eric Bylaska, Libor Kovarik (recorder)

- Energy Conversion and Storage Materials

  Stan Whittingham (discussion lead), Shirley Meng, John Rehr, Jun Liu, Perla Balbuena, Theva Thevuthasan, Mark Bowden (recorder), Niri Govind, Karl Mueller

The groups were asked to address four questions in their assigned areas:

1. What are the most critical science gaps/challenges?
2. What space should EMSL work to help fill?
3. Is our 5-10 year focus/target appropriate and how can it be improved?
4. Are there intermediate objectives that should be identified?

## 2.3 EMSL Capabilities Overview

Scott Lea showcased EMSL’s experimental capabilities most relevant to energy materials. Karl Mueller discussed processes by which EMSL identifies and plans new instruments. Karl also described major capabilities under development: high-resolution mass spectrometry and dynamic transmission electron microscopy (D-TEM). Additional capabilities in the planning stage include dynamic nuclear polarization (DNP), a capability combining features of nuclear magnetic resonance (NMR) and electron paramagnetic resonance, and ultrafast transmission electron microscopy, which would increase temporal resolution of microscopy to the picosecond regime. Tim Scheibe and Dave Cowley gave an overview of EMSL’s High Performance Computation capability including developments in both software and hardware. These presentations provided background for further discussion in breakout sessions to address new capabilities that will be required to meet the science challenges identified earlier.

## 2.4 Breakout Session 2: Capability Needs

To focus discussion on capability needs and recommendations for possible development, the STAP workshop participants broke into groups to focus on the following areas:

- Experimental Capabilities Catalysis

  Mark Barteau, Chuck Peden (discussion lead), Susannah Scott, Chris Jones, Cathy Tway, Roger Rousseau, Rick Orth, John Holiday, Libor Kovarik, Nancy Washton (recorder)

- Experimental Capabilities Energy Storage

  Stan Whittingham, Shirley Meng (discussion lead), Jun Liu (discussion lead), Theva Thevuthasan (recorder), Mark Bowden, Karl Mueller, Scott Lea

- Computational Capabilities

  John Rehr (discussion lead), Perla Balbuena (discussion lead), Dave Cowley, Niri Govind (recorder), Tim Scheibe, Eric Bylaska, Roger Rousseau

The groups were asked to consider what new experimental and computational tools are necessary to address the science gaps (previously noted) structured around the following questions:

1. What capabilities are needed to address the identified challenges?
2. How might current capabilities be best deployed to address needs?
3. What new capabilities are needed to move science forward over the next decade?

## 2.5 Maximizing Impact

The final half-day discussion focused on what can be done to enable EMSL-facilitated research for the greatest impact. The ideas and recommendations identified in earlier discussions were summarized to ensure an accurate representation of the panel. New input was sought upon reflection.

Opportunities were considered to position EMSL for maximum impact in the EMP Science Theme. These included: 1) mechanisms to focus activities such as user calls, campaigns, intramural projects, visiting fellows, and partnerships, 2) short-term objectives to move toward longer-term targets, 3) best ways to utilize or deploy EMSL resources and capabilities, 4) improvements in outreach, and 5) partnerships and relationships needed to enable progress.

# 3.0 STAP Recommendations and Feedback

The workshop benefited from highly engaged participants. Many suggestions and recommendations of multiple types were presented and discussed. Summaries of each portion of the workshop are included in specific sections. Several themes emerged throughout the workshop. These common themes and high-level recommendations are summarized here before more detailed reports of the breakout sessions and general discussion.

## 3.1 Science Directions and Opportunities

The focus on interfaces, and in particular solvent-solid interfaces, was endorsed as "the strongest way for EMSL to maximize impact in the science area." The STAP recommended focus on integration across system parts and across platforms, utilizing EMSL staff assets. The participants indicated that solvent-mediated interfacial chemistry is broadly important and impactful, and a topic whereby EMSL can assert national prominence. It impacts energy storage, catalysis, and separations as well as biomass conversion, electrochemistry, and other areas (coal, petroleum, and other liquid phase processing, as well as water remediation).

The panel noted that solvent-mediated interfacial processes impact the other Science Themes in EMSL, providing a strong synergy in fundamental science. The STAP advocated embracing and expanding solvent-mediated interfacial chemistry beyond catalysis and energy conversion storage materials to include an integrated reactions-separations-systems approach. The panel encouraged EMSL to broaden the scope to include separations.

## 3.2 Specific Targets, Challenges, and Gaps

Having affirmed the importance of solvent-mediated processes, the following significant challenges were identified as important targets:

- Understand (via experiment and modeling) reactive sites at interfaces, via their structure, dynamics, and emergence including "rare" sites.
- Understand and predict phase formation at interfaces, including transformations, plating-stripping, dendrite formation, and inhibition.
- Develop higher resolution tools and novel combinations of computational and experimental *in situ* (especially spectroscopic) methods that will impact many areas of science. There is a strong need for quantifiable measurements, in addition to qualitative observations.

## 3.3 Methods

The panel noted that EMSL should build upon areas where it has expertise and leadership, including NMR, microscopy, and computation. Several themes drew widespread support from workshop participants:

- Incorporate an increased number of researchers (staff, postdocs, students).
- Continued *in situ* probe development will be essential to retain EMSL's current leadership position. Additional development will allow measurements under more realistic operating conditions and be sensitive enough to detect

rare interface sites that contribute strongly to catalytic activity. EMSL users need to be more aware of microfabrication capabilities that might be used for development of a wide range of *in situ* probes.

- Multimodality, including computation, is important and needs to incorporate length scales beyond the molecular, and the integration of information from different length scales. For example, the combination of Differential Electrochemical Mass Spectrometry (DEMS), Time of Flight (TOF) secondary ion mass spectrometry (SIMS), x-ray photoelectron spectroscopy (XPS) and sum frequency generation spectroscopy would generate comprehensive complementary information from both *in situ* and *ex situ* environments.

- A hierarchy is needed having computation codes covering multiple length and time scales, well defined workflows, and more physics codes. More rapid linking of modeling and experiment is desirable, ultimately leading to sophisticated data analysis on the fly during experimental measurement to guide the next experiment.

- New capabilities such as dynamic nuclear polarization (DNP), DEMS, and ultrafast TEM received support from participants.

## 3.4 Maximizing EMSL's Impact

Discussion on enhancing the impact of EMSL took several forms and covered the following areas:

- Partner with multiple entities for greatest success. Identify and engage these partners in a systematic and deliberate process involving a persistent presence at critical meetings, involvement with major DOE programs, and EMSL user campaigns.

- Attract and train high-level users to increase visibility and impact. The participants stressed the value of engaging emerging scientists who are likely to form the next generation of scientific leaders by including the use of social media and targeted training sessions on science and instrumentation.

- Recognize the significant synergy that solvent-mediated interfacial processes have with EMP and other EMSL Science Themes.

- Increase community impact by engaging PNNL expertise in techno-economic analysis (and life-cycle assessment) and consider engaging EMSL users early in their work to consider techno-economics of their work, when relevant. This is one way of helping advance projects with greatest opportunity for successful impact.

- Raise awareness about EMSL. STAP members had the impression that great work is conducted in EMSL, yet too few people know.

# 4.0 Summaries of Breakout and Discussion Sessions

## 4.1 Breakout Session 1: Scientific Gaps

Susannah Scott and Chris Jones, discussion leads; Libor Kovarik, recorder

### 4.1.1 Catalysis

The breakout group supported the importance placed on solvent-mediated interfacial chemistry, its potential for broad impact, and an area in which EMSL can assert national prominence. They proposed emphasis on projects that impact both catalysis and separations, and impact biomass conversion and electrochemistry, two previously defined themes, plus other areas (coal, petroleum, and other liquid phase processing, as well as water remediation).

The discussion started on global energy issues and evolved further. The inter-relationship was noted between energy, environment, $CO_2$, water, food, and national security issues, suggesting EMSL consider opportunities at the intersection of some or all of these areas. For example, the basic science of catalytic aqueous pollutant degradation was identified as an important cross-cutting opportunity.

They suggested both top-down and bottom-up approaches to best use strengths and capabilities. An example of a top-down approach is embracing an integrated catalysis-separations-system within solvent-mediated interfacial chemistry, recognizing energy challenges at interfaces are broader than chemical transformations alone. An example of a bottom-up approach is development of novel computational and experimental (especially *in situ* spectroscopic) techniques, a strength EMSL should continue to leverage; however, these developments should not be too narrow and have the capacity to impact a wide array of fields.

The group advocated engaging PNNL expertise in techno-economic analysis and life-cycle assessment early in projects to ensure resources are committed to projects with a reasonable chance of commercial or economic impact. This would not apply to all projects; however, it would be particularly valuable for more applied projects where industrial links are likely and may also steer fundamental projects in a relevant direction.

In addition to the general, longer-term issues discussed above, the group identified the following specific areas in computation and experiment for near-term focus:

- Computation: advancing non-equilibrium simulation methods and developing the ability to model amorphous solids are important. Extending simulation to longer timescales is a major bottleneck for many models with liquid systems, as is the need to incorporate larger length scales. These are important scientific challenges for catalysis, which will impact many other areas of research.

- Experimentation: designing materials capturing not only static, but also dynamic structures is rarely considered, yet is an essential component of most catalytic systems. The development of *operando* flow reactors is needed to bridge this knowledge gap.

### 4.1.2 Energy Storage

Stan Whittingham, discussion lead; Mark Bowden, recorder

The group identified two major challenges in electrochemical storage systems that tie strongly to the EMP focus in solvent-mediated interfacial chemistry:

- Understand and control metal plating and stripping. This is a key problem in batteries, particularly those with large storage capacities that cannot rely on intercalation at electrodes. It is a complex problem dependent on properties of both metal and electrolyte. Advances are needed to avoid loss of metal and formation of dendrites, which can short-circuit batteries.

- Understand reactive sites at interfaces. This is a broader challenge and impacts many other fields including catalysis, and requires study of structure, dynamics, and emergent phenomena. The challenge arises because the reactive sites, which control properties, are in many cases minority species and difficult to probe.

The group discussed the experimental and computational approaches needed to meet these challenges. They noted the need for *operando* studies of real systems that provided quantitative information ("measure" rather than "observe") over smaller (nm) and faster (ns) timescales. These studies should be integrated over multiple experiments (e.g., spectroscopy and microscopy) with accurate modeling at multiple length and time scales.

The focus on interfaces, in particular solvent-solid interfaces, was endorsed as the strongest way for EMSL to maximize impact in the EMP Science Theme. EMSL's efforts should focus on integration across system parts and across platforms, utilizing staff assets. Computational capabilities should extend in both spatial and temporal regimes for better linkages between experiment and modeling. Multi-scale computations and more physics codes in NWChem are needed. In pursuing these challenges, EMSL should partner better with other national groups (JCESR, Energy Frontier Research Centers (EFRC), and other principal investigators) to avoid duplicating effort and to deliver impact more quickly. Metrics of progress are needed to demonstrate impact, such as:

- Increase EFRC users, EFRC interactions (e.g., joint publications), and synergies.

- Make EMSL a critical and indispensable partner in large DOE programs such as Hubs and EFRCs. EMSL should be sought for both its researchers and tools.

- Improve efficiency of workflow; shorten the time between project initiation and publication.

Two intermediate objectives were discussed as priorities to help EMSL meet long-term goals:

- Improve EMSL's electrochemical expertise through hiring, developing existing staff, and partnering with external groups.

- Integrate codes for spectroscopy that apply to multiple length scales and include physics capabilities in addition to chemistry.

The group viewed the Wiley Research Fellows as a strong mechanism to meet these objectives.

## 4.2 Breakout Session 2: Capability Needs

### 4.2.1 Catalysis

Cathy Tway, discussion lead; Nancy Washton, recorder

The group identified several capability-related challenges that if overcome, would deliver impact and maintain EMSL's excellence in measurement and modeling of catalytic systems. These challenges were framed in the form of questions:

1. How do we interrogate with site specificity rare sites of catalytic activity?
2. How do we develop robust methods and techniques that clearly and directly show correlations and causation?
3. How do we link computational results and data processing to experimental data so experiments can be optimized on the fly in real time to produce the most meaningful and impactful results?

The discussions focused on three techniques deemed particularly relevant to catalysis: TEM, non-linear optical spectroscopy, and DNP. The first question above has particular relevance to TEM since this technique looks at isolated regions that may not be representative of the whole. The key here is locate and analyze sites responsible for catalytic activity even when the number of such sites might be relatively small.

EMSL has strength in non-linear optical spectroscopy through superior spectral resolution, which leads to accurate line shapes and meaningful data interpretation. Value needs to be demonstrated to the catalyst community by studying a high-profile catalysis problem and publishing the results in a widely-read high-impact journal.

The group recognized the value of DNP to catalytic studies and noted that such an instrument would benefit many scientific communities including geoscientists, biologists, and materials chemists. There is no DNP instrument available at a user facility in the United States and the group strongly supported EMSL's efforts to develop this capability.

Discussions also included opportunities to use analytical techniques and methods in new ways or in new areas from their traditional user base. This may result in unanticipated and highly impactful breakthroughs in catalysis as well as other science areas. The group suggested EMSL host a "capability boot camp" and invite graduate students and postdocs from catalysis groups to brainstorm with EMSL's staff instrument scientists. They saw two potential benefits:

- instructing students on experiments and information obtainable from EMSL
- identifying new experiments or samples that could be analyzed to advance knowledge in unexpected ways.

### 4.2.2 Energy Storage

Shirley Meng and Jun Liu, discussion leads; Theva Thevuthasan, recorder

The group considered TEM capabilities central to understanding science challenges in energy storage. While advances by EMSL staff and users were very valuable, capabilities are far from optimized. The liquid cells, for example, have severe limitations on the amount of fluid that can be supplied to the region of interest. New developments (channel design, biasing ports, etc.) are necessary to gain information critical to unravelling the chemistry at the interface in a solvent environment under operating conditions approaching that of real batteries. A direct detector for TEM would also be a valuable addition to EMSL's microscopy capabilities for more rapid observation and reduced beam exposure. The group looked forward to successful use of the D-TEM and encouraged further increases in temporal resolution. Investment in sample environments and in the ability to transfer samples between the TEM and other instruments (e.g., atom probe tomography, focused ion beam/scanning electron microscope, NMR) was also viewed as a productive avenue for development. The group recognized the critical importance for skilled scientists to operate microscopes and interpret results in this area, and recommended EMSL allocate more staff time to these studies.

The group encouraged greater university collaboration and partnership to augment EMSL staff time in NMR spectroscopy, perhaps using graduate students or postdocs. The panel encouraged development of *in situ* magic-angle spinning-NMR probes to obtain critical data. Collaboration with the Joint Center for Energy Storage Research and EFRCs might assist both probe development and NMR usage to advance battery science. The group was supportive of EMSL's plans to develop a DNP capability and felt this was an important addition to instrumentation and expertise. The DNP capability should include *in situ* environments and interface with other instruments through multi-modal sample transfer.

The DEMS was seen as a valuable addition to EMSL's capabilities. This technique provides a detailed chemical composition of electrolytes and gases in addition to helping unravel interfacial reactions. It is not widely available, especially in user facilities, and could become a powerful drawing card for users in energy storage, providing complementary information to TOF-SIMS and XPS in particular.

*In situ* and *operando* capabilities in general were encouraged, particularly for non-linear spectroscopy (second harmonic generation and sum-frequency generation), TOF-SIMS, and XPS. X-ray methods, such as high-resolution tomography, were seen as valuable, but the principal advantage for EMSL is data integration with other capabilities. Otherwise the group thought the instruments already available at synchrotrons were sufficient and offered advantages from the increased x-ray flux.

### 4.2.3 Computational Capabilities

John Rehr and Perla Balbuena, discussion leads; Niri Govind, recorder

The group discussed computational capabilities in the context of the breakout questions and identified challenges:

- Understanding and controlling materials synthesis
- Dynamic evolution of materials interfaces under operating conditions
- Integrated computation encompassing the whole system over multiple scales.

To meet these and future challenges, the following needs were identified:

- Software people and applications scientists to promote theory-experiment integration
- New codes that address problems related to identified challenges, in addition to what NWChem can currently compute
- New code integration via an open-source model

- More staff members and users who can utilize high-performance computing resources effectively and better integration with computer science and applied mathematics groups at PNNL, other national laboratories, and academia
- Partnerships with other national laboratories, academia, and industry.

EMSL's current capabilities would be best deployed by providing researchers with quick access to Cascade and by ensuring effective usage of large-scale resources. The panel identified the following specific new capabilities to advance scientific objectives:

- Develop and implement a hierarchy of methods with increasing accuracy and complexity to tackle a broad range of problems.
- Provide workflows to guide usage of NWChem and additional physics codes.
- Deliver easy-to-use interfaces.

## 4.3 Maximizing Impact, Teaming, and Other Discussion Topics

Day 2 of the workshop was devoted to ways to focus and optimize the impact of EMSL, looking at short- and long-term objectives, types of partnerships that should be formed and any other topics the STAP members thought would be valuable to EMSL. To the extent possible, the wide range of topics is captured here.

**Critical meetings** in target areas should be attended for a persistent presence via talks and if possible, display booths. The North American Catalysis Society meeting would be an appropriate meeting for catalysis. To increase meeting impact, perhaps hold evening workshops or team with other user facilities on a user poster session.

**Partnerships** are important to success in these areas. A mechanism should be found to foster long-term partnerships. It is useful to establish specific contacts with EFRCs and other significant DOE programs. For example, Shirley Meng is an appropriate contact between EMSL and the NorthEast Center for Chemical Energy Storage EFRC. Success will also come from partnerships with leading universities and research groups, and by increasing the number of industrial users.

**Campaigns** were viewed as a good way to highlight the presence and value of new capabilities as well as a way to form partnerships with leading research teams.

**Attracting and training users** is viewed as important. Several approaches might be useful including:

- Conduct scientific “boot camp” on science and tools.
- Focus on new faculty members and cultivate those likely to be the next generation scientific leaders.
- Push social media for the new generation of scientists.
- Make videos on topics such as sample preparation to attract users to capabilities new to them and valuable to the scientific community.

**Broader implications** of solvent-mediated processes including the current description:

- Liquid-mediated interfacial processes would be a more general description in some ways and highly relevant to all EMSL Science Themes and many DOE challenges.
- Separation materials and processes go beyond EMSL’s historic focus on catalysis and energy conversion, and couple directly to catalytic and energy conversion systems. EMSL was encouraged to broaden its scope to include separation.
- Significant synergy with other EMSL Science Themes exists. Ammonia production and use contribute significantly to energy use and greenhouse gas production. In effect, there is a tight coupling among food production, environmental catalysis, water purification, and chemical separations.

**Clean room and micro-fabrication capabilities** at EMSL are important for *in situ* cell/probe designs. This is a very important capability for the user community and should be publicized. Moreover, current *in situ* cells are optimized for the measurement technique while future cells will be further optimized for the operation of the system being studied. Thus there will be continual development and additional need for designing new generations of *in situ* cells. EMSL has a strong position in this area, but will need to work to maintain its leading position.

**Mass spectrometry** capabilities at EMSL can be deployed in creative ways in the energy and catalysis area. These capabilities could be investigated to determine whether the composition of complex product mixtures derived from biomass processing that should be physically located at or near the reactor for immediate analysis. Similarly, measurements produced during interfacial reactions at battery/electrochemical interfaces can provide information about species evolving at these interfaces as a complement to current measurements, which provide information about products formed or retained at the interfaces under study.

# Appendix A

# Workshop Agenda

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## Energy Materials and Processes
## Science Theme Advisory Panel

**July 8-9, 2014**
**EMSL Board Room**

# AGENDA

| **Tuesday, July 8** | |
|---|---|
| 7:30 a.m. | Breakfast |
| 7:45 a.m. | Introductions |
| 8:00 a.m. | Welcome and Overview – Don Baer, Mark Barteau, Karl Mueller, Chuck Peden<br>• Welcome, EMSL Context, STAP Objectives<br>• EMSL Science Overview<br>• History of EMP, time evolution, proposed focus, current activities<br>• IIC activities and important challenges<br>• Review of STAP charge and schedule |
| 9:30 a.m. | Break |
| **Science needs, directions, opportunities, critical challenges** | |
| 9:40 a.m. | Introduction and perspective – Mark Barteau |
| 10:00 a.m. | Presentations by STAP members |
| 11:00 a.m. | Discussion - All |
| 11:10 a.m. | Small Breakout Discussions (Board room and EMSL 1075)<br>• Catalysis<br>• energy conversion & storage materials |
| 12:15 p.m. | Working Lunch will be set up in EMSL Room 1030 in the Glass Room |
| 12:45 p.m. | Small Group Reports & Discussion |
| **Capabilities, technologies, tool challenges, needs and opportunities** | |
| 1:15 p.m. | Overview of current EMSL capabilities –Scott Lea |

| | |
|---|---|
| 1:30 p.m. | EMSL Tour |
| 2:30 p.m. | Current thinking about future tools – Karl Mueller |
| 2:45 p.m. | Next Generation Computer and Codes – Dave Cowley and Tim Scheibe |
| 3:00 p.m. | Opportunity for STAP member ideas |
| 3:30 p.m. | Break |
| 3:40 p.m. | Small Group Breakout Discussions on technology needs for advancing<br>(Board room, EMSL Rooms 1075 and 1077) |
| 4:40 pm | Small group report and discussion |
| 5:15 p.m. | Adjourn for the day |
| 6:00 p.m. | Dinner STAP Members and Invited Guests (planned for Fat Olives) |
| **Wednesday, July 9** | |
| 7:30 a.m. | Breakfast |
| 8:00 a.m. | Follow up discussion and questions – Don Baer and Mark Barteau |
| 8:20 a.m. | Ways to focus/enable science, optimizing use/Impact of EMSL – Don Baer<br>• Mechanisms to focus activities, user calls, campaigns, intramural projects visiting fellows, partnerships<br>• Round Table discussion on partnerships, team formations and ways to enhance impact |
| 9:45 a.m. | Break |
| 10:00 a.m. | Putting it all together – Round Table discussion on important opportunities, need tools and methods to enable progress<br>• Refinement of EMSL EMP target area – specific objectives<br>• Short term objectives to move toward longer term target<br>• How to focus current activities and capabilities for impact<br>• Longer term capability development needs<br>• Identification of types of partnerships and relationships needed to enable progress<br>• Best ways to use or deploy EMSL resources and capabilities |
| 12:00 p.m. | Adjourn |

# Appendix B

# Workshop Attendees

# Appendix B

## Workshop Attendees

**STAP Members**

Mark Barteau, University of Michigan (Workshop Co-chair)

Perla Balbuena, Texas A & M University

Chris Jones, Georgia Institute of Technology

Jun Liu, PNNL

Shirley Meng, University of California, San Diego

Chuck Peden, PNNL

John Rehr, University of Washington

Cathy Tway, Dow Chemical Company

Susannah Scott, University of California, Santa Barbara

Stan Whittingham, Binghamton University

**Other Workshop Participants and Observers**

John Holladay, PNNL

Rick Orth, PNNL

Roger Rousseau, PNNL

Don Baer (Workshop Co-chair), EMSL

Mark Bowden, EMSL

Eric Bylaska, EMSL

Dave Cowley, EMSL

Niri Govind, EMSL

Libor Kovarik, EMSL

Scott Lea, EMSL

Karl Mueller, EMSL

Tim Scheibe, EMSL

Theva Thevuthasan, EMSL

Nancy Washton, EMSL

Hongfei Wang, EMSL