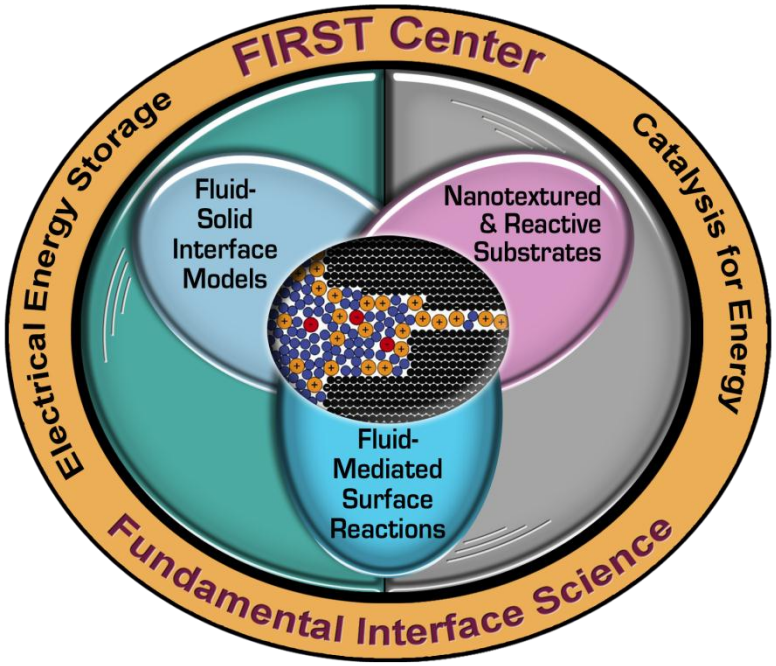
Fluid Interface Reactions, Structures and Transport (FIRST) Center

David J. Wesolowski (Oak Ridge National Lab)

U.S. DEPARTMENT OF ENERGY | Office of Science

FIRST Center Vision and Goal

To develop quantitative and predictive models of the unique nanoscale environment at fluid-solid interfaces to enable transformative advances in electrical energy storage and heterogeneous catalysis.



RESEARCH PLAN AND DIRECTIONS

Our multidisciplinary team integrates advanced materials synthesis, neutron and X-ray scattering, various spectroscopies, macroscopic experiments, and multiscale molecular modeling to provide a predictive capability for controlling and designing new interfacial systems for 21st century energy needs.

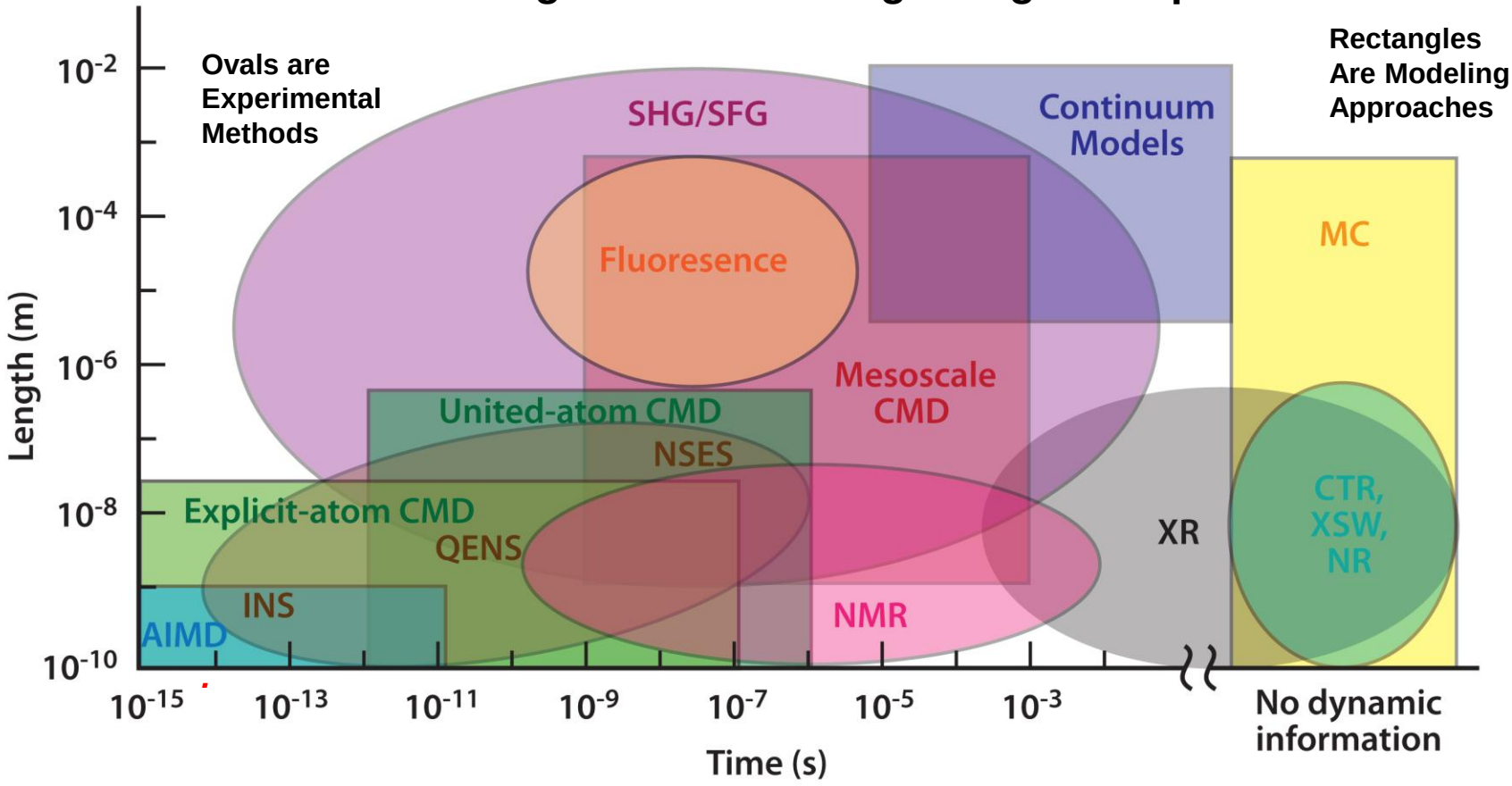
OAK RIDGE National Laboratory | VANDERBILT UNIVERSITY | Argonne NATIONAL LABORATORY | Drexel UNIVERSITY | UNIVERSITY of VIRGINIA | NORTHWESTERN UNIVERSITY

an Office of Basic Energy Sciences
Energy Frontier Research Center

Thrust 1: Fluid-Solid Interface (FSI) Model Development
Peter T. Cummings, Leader

Goal is to develop quantitative and predictive Fluid-Solid Interface (FSI) models for relevant fluid/substrate combinations (e.g, water, ionic liquids, polar organics, carbon, oxides) guided by experimental input.

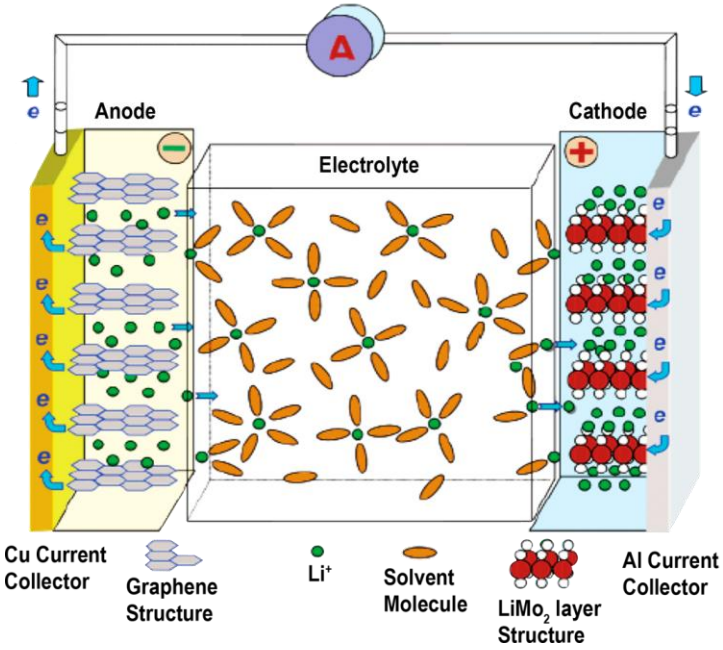
- **Integrated Theory, Modeling, Simulation, and Experiments, *ITMSE*, across the relevant time and length scales is the guiding concept.**



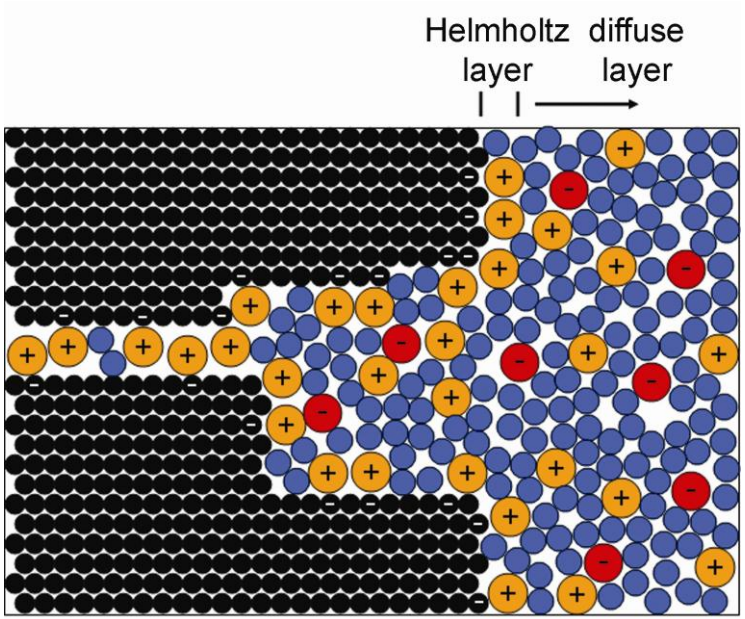
Thrust 2: Nanotextured and Reactive Substrates
Sheng Dai, Leader

Goal is to understand and control ion transport, charge/discharge processes and substrate evolution at nanotextured and reactive battery and supercapacitor electrode/electrolyte interfaces.

Ion transport in electrolyte, intercalation into electrodes and Solid-Electrolyte Interphase (SEI) formation are all poorly-understood, but limiting phenomena in, for instance, Li-ion batteries.



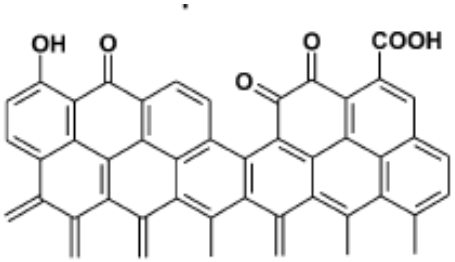
In electrochemical capacitors "super-capacitance" is postulated to result when electrode nanopores are filled with 'bare' ions, free of their electrolyte solvation shells.



Thrust 3: Fluid-Mediated Surface Reactions
Steven H. Overbury, Leader

Goal is to understand and control fluid-mediated proton-coupled electron transfer (PCET) in photocatalytic and electrocatalytic reactions at fluid-solid interfaces, such as CO_2 and O_2 reduction in $H_2O \pm CO_2$ and other dense fluids.

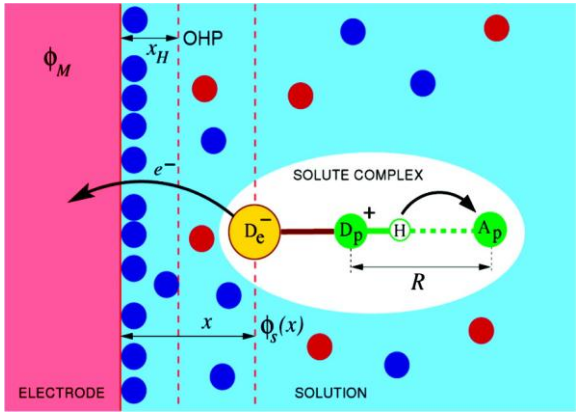
Functional groups at graphene edges.

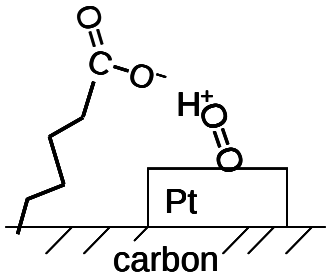


$O_2 + 4H^+ + 4e^- \Leftrightarrow 2H_2O$

$CO_2 + 2H^+ + 2e^- \Leftrightarrow HCOOH$

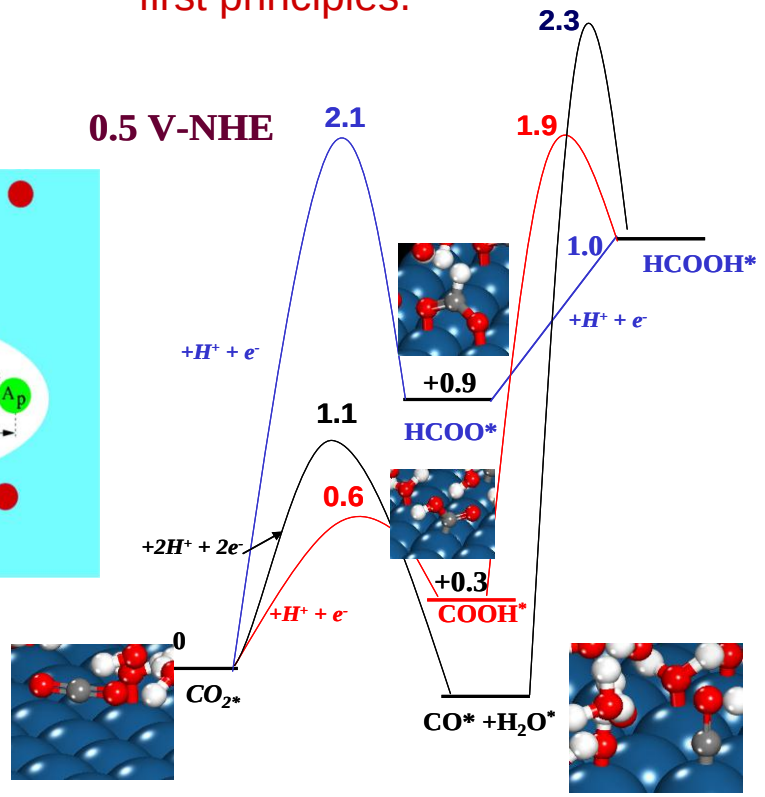
PCET at FSI





Role of surface anchored proton donor ligands in oxygen reduction reaction rates?

Energetics of CO_2 reduction from first principles.

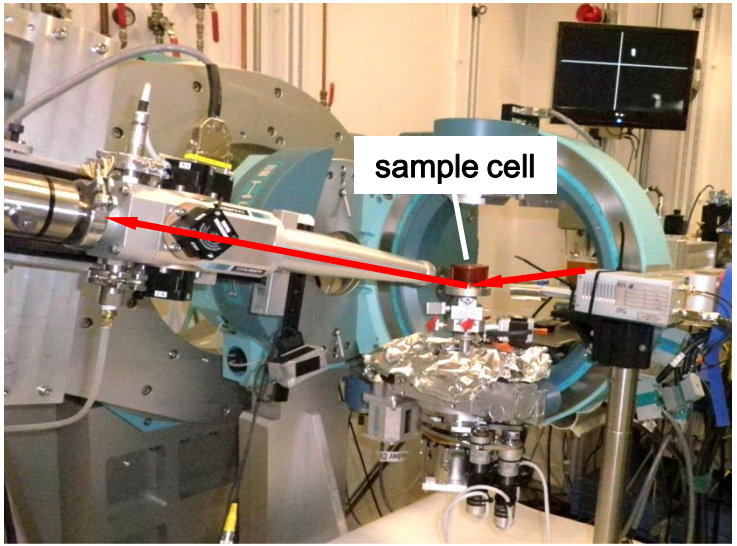


$H_2O + CO_2$ at Pt on carbon

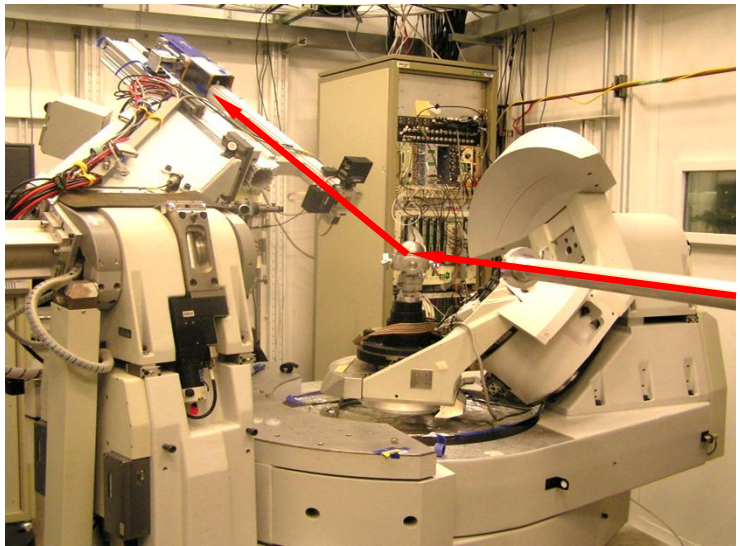
Argonne National Laboratory's Advanced Photon Source is the Western Hemisphere's Brightest X-ray source, making it an ideal facility for probing fluid-solid Interfaces *in situ.*

Fenter's Beam Lines

Sector 6

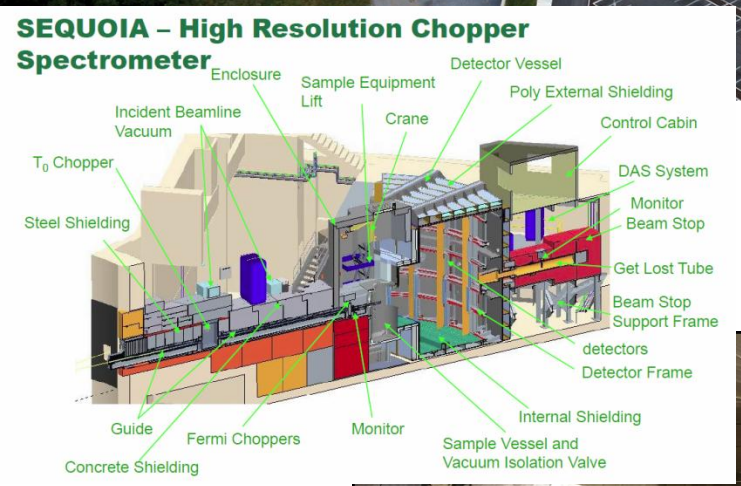


Sector 33



ORNL's SNS/HFIR and CNMS provide a wealth of unique capabilities free to users.

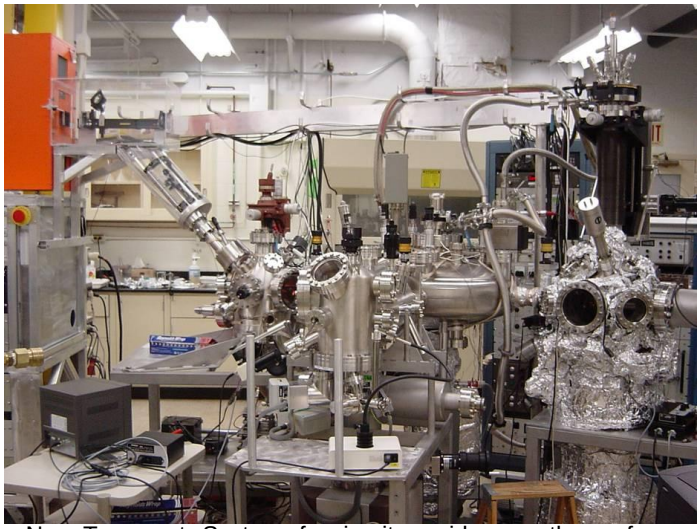
Current (Nanoman, MFP-3D, and Cypher) ambient/SPM platforms at CNMS. Figures are courtesy of Veeco and Asylum Research



Kolesnikov's INS instrument 3-stories high!

Mamontov *Inside* BASIS spectrometer

Kalinin/Balke labs in CNMS



NanoTransport System for in-situ oxide growth, surface characterization, and UHV STM and PFM studies.

ORNL's National Center for Computational Sciences, a DOE user facility, is home to the world's most powerful open-system high performance computing platform. This combined with CNMS Nanomaterials Theory Institute (Cummings, Director) and our newly-purchased Institutional Computational Cluster and 700,000 hour NERSC allocation gives FIRST Center researchers unprecedented access to a wide range of computing facilities.