**Fluid Interface Reactions, Structures and Transport**

**FIRST Energy Frontier Research Center**

David J. Wesolowski, Director  
Oak Ridge National Laboratory

http://web.ornl.gov/sci/first/

Mission: to develop fundamental understanding and validated, predictive models of the unique nanoscale environment at fluid-solid interfaces that will enable transformative advances in electrical energy storage and electrocatalysis.

PI's:  
- **ORNL** - Nina Balke, Sheng Dai, Nancy Dudney, Paul Kent, Alexander Kolesnikov, Daniel Lutterman, Shannon Mahurin, Eugene Mamontov, David Mullins, Michael Naguib, Gernot Rother, Robert Sacci, Raymond Unocic, Huiyuan Zhu;
- **Argonne National Lab** - Paul Fenter;  
- **Drexel Univ.** - Yury Gogotsi;  
- **Penn State Univ.** - Adri Van Duin;  
- **UC Davis** - Alexandra Navrotsky;  
- **UC Riverside** - De-en Jiang, Jianzhong Wu;  
- **Univ. Delaware** - Joel Rosenthal;  
- **Univ. Minnesota** – Matthew Neurock;  
- **Vanderbilt Univ.** – Peter Cummings, Haoxiang Liu

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**FIRST Organizational Structure**

### Steering Committee:
- Works with Director to establish research goals, coordinate activities, allocate resources and start/end projects.

### Research Thrusts:
- Thrust 1 focuses on developing accurate conceptual and predictive computational models of key interfacial phenomena.
- Thrust 2 focuses on model-driven synthesis and characterization of novel functional interfaces.

### Crosscutting Themes:
- Interfacial fluid *structure* and *transport* dominate energy and power density.
- Interfacial fluid *dynamics* and *reactivity* dominate electrocatalysis and pseudo-capacitance.
Behavior of Electrolytes in Nanoporous Carbon Independently Predicted Using Classical Density Functional Theory (CDFT) and Classical Molecular Dynamics (CMD) Simulations

Difference in Capacitance Strongly Dependent on Solvent Polarity

Aqueous or organic electrolyte (ions in a polar solvent)

Ionic liquid electrolyte (only cations and anions)


Capacitance (F/m²)
Pore size (nm)

Difference in Capacitance Strongly Dependent on Solvent Polarity
CMD and CDFT Predictions of ‘Exohedral’ and ‘Endohedral’ Curvature Effects on Capacitance

Ion packing effects observed on the flat graphene surface, which result in ‘lattice saturation’ (bell-shaped curve) are strongly reduced at curved surfaces allowing high capacitance over wide range of surface potentials.


MXenes ($M_{n+1}X_n$, n = 1-3), a New Class of 2D Transition Metal (M) Carbonitrides (X)


Capacitive Energy Storage

Etching “A” layer from $M_{n+1}AX_n$ + Sonication $\Rightarrow$ MXene

Work supported by the U.S. Department of Energy, Office of Basic Energy Sciences as part of an Energy Frontier Research Center
Structure/Stability/Capacitive Performance of MXenes Tied to Surface Chemistry (F, O, OH)

Neutron and X-ray Scattering/PDF:


Solution-Drop Calorimetry:

X-ray Diffraction/Thermogravimetry Probes of Water and Ion Non-Redox Intercalation:

Quasielastic Neutron Scattering/ CMD Simulations of Water and Ion Dynamics:
Using Scanning Probe Microscopy (SPM) to detect ion intercalation dynamics through electrode volume changes, a new method we call **Electrochemical Strain Microscopy (ESM)**

**Patented and Licensed to Asylum Research**

- 20 FIRST publications on ion intercalation dynamics in batteries, imaging theory, pore-pressure/volume effects.
- Ångström-scale vertical and nanoscale lateral resolution.
- Environmental electrochemical cells developed and used to study electrochemical capacitor charge/discharge kinetics.


This capability is now freely available to users in ORNL’s Center for Nanophase Materials Science and is in Big Demand!
A-Team Promotes Synergy, Collaboration and Communication Across the Center.

FIRST Center students, postdocs and early-career staff/faculty conduct their own monthly teleconferences and offsite meetings to conceive and coordinate research.
“Heterogeneity of Charge Storage Processes in Electrochemical Capacitors”

Q. Gao, W. Tsai, M. Naguib, Nina Balke, P. Fenter, M. Thompson, P. Cummings (ESII-5, Tue. 9:50 AM)

“High-Rate Electrical Energy Storage Enabled by Metallic Conductivity of MXenes”

M. Lukatskaya, M. Alhabeb, M. Boota, M. Nagub, Y. Xie, P. Kent, X. Sang, R Unocic, N. Balke, D. Wesolowski, Yury Gogotsi (ESIV-2, Tue. 1:50 PM)

“Electrical Double Layer in Porous Electrodes – Capacitance, Capacitive Mixing, and Electro-Osmosis”

C. Lian, J. Neal, K. Liu, D. Wesolowski, D. Henderson, Jianzhong Wu (ESI-6, Mon. 4:40 PM)

Team Competition: “Understanding Room Temperature Ionic Liquids and Their Performance in Supercapacitors”

J. Neal, K. Liu, Naresh Osti, Matthew Thompson, Katherine Van Aken, Y. Zhang (ESI-3, Mon. 3:40 PM)
“Insights into the Interaction Between 2D Transition Metal Carbide and Nitride (MXenes) with Ions and Water”

Michael Naguib, 22 co-authors (PI-9, Mon. 5:00 PM)

“Insights into the Mechanisms and Kinetics that Control Electrocatalysis at Fluid-Solid Interfaces”

Matthew Neurock, 16 co-authors (PI-6, Mon. 5:00 PM)

“Organic-Inorganic Hybrid Electrode Materials for Static and Flowable Pseudocapacitive Energy Storage”

Muhammad Boota, 8 co-authors (PII-14, Tue. 3:30 PM)

“Probing Atomic Structure Defects in 2D Materials for Energy Applications”

Xiahan Sang, 9 co-authors (PII-18, Tue. 3:30 PM)

“Quantum Capacitance, Dielectric Screening, and Edge Effects in Carbon Supercapacitors and Beyond”

Cheng Zhan, 6 co-authors (PII-7, Tue. 3:30 PM)