2024 NETL Resource Sustainability Project Review Meeting

Multiphysics and Multiscale Simulation Methods for Electromagnetic Energy Assisted Fossil Fuel to Hydrogen Conversion

Su Yan [1], Jiefu Chen [2], and Xiaonan Shan [2]

[1] Howard University, Washington, DC 20059; su.yan@howard.edu

[2] University of Houston, Houston, TX 77204; jchen82@central.uh.edu, xshan@central.uh.edu

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TECHNICAL BACKGROUND & MOTIVATION

- Fossil fuels comprise 80% of current global primary energy demand and the energy system is the source of approximately 2/3 of global CO_2 emissions.
- Develop new methods that use electromagnetic (EM) energy to assist fossil fuel to H_2 conversion.
- **Research Questions:**
  1. What is the catalyst morphology and how does it affect EM energy deposition during H_2 production?
  2. How do physical factors, such as reaction chamber geometry, EM wave intensity and frequency, source material flow velocity, and reaction temperature, affect catalytic active sites?
  3. How to estimate the throughput, yield, and cost of dehydrogenization?

SIGNIFICANCE OF THE PROJECT RESULTS

- **Research Component:**

Direct impact on the petrochemical industry by providing theoretically rigorous and technically sound computational approaches to guide low-cost hydrogen generations.

1. Develop a computational platform to provide fundamental understanding of using alternative energy processes (microwave, radio frequency, plasma and other EM inputs) for low-cost hydrogen production from fossil fuels;
2. Provide thorough and comprehensive studies on how to improve the H2 conversion efficiency of EM energy assisted approaches using multiphysics and multiscale computation, imaging capabilities, and optimization;
3. Investigate and optimize the H2 production using EM energy and estimate the conversion costs;
4. Publish on technical journals and conferences to support technology transfer.

SIGNIFICANCE OF THE PROJECT RESULTS

- **Education Component:**

Education and research opportunities for our African American and other underrepresented minority students.

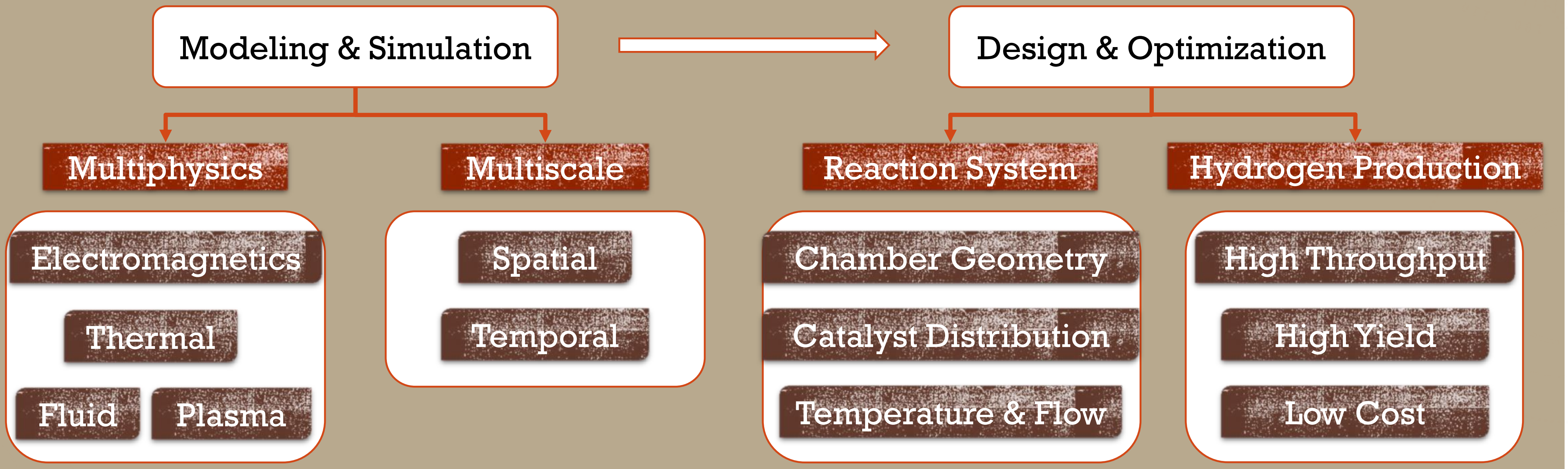
1. Support one graduate student and multiple undergraduate students in Howard University and one graduate student in the University of Houston;
2. Provide education and training in an interdisciplinary research team that integrates EM, chemical engineering, computer graphics, material sciences, and computational science and engineering;
3. Offer an excellent opportunity for our students to be exposed and involved in fossil energy related education and research, and hence, develop their interests and capabilities in creating and executing new research ideas that support the mission of DOE.

PROJECT OBJECTIVES

- **Objective:**

  Develop, implement, validate, and apply multiphysics and multiscale simulation methods for efficient electromagnetic (EM) energy assisted conversion from fossil fuel to low-cost hydrogen.

- **Two Major Thrust Areas:**



RESEARCH TASKS

1. **Understanding 3D structures of catalysts and their supports:**

   Utilize 3D optical imaging and FIB-SEM sectioning and imaging to determine catalyst distribution and support morphology in order to characterize 3D structure;

2. **Characterization of EM hotspots within heterogeneous catalysis:**

   Use computational methods to develop 3D nodal discontinuous Galerkin (NDG) methods for the modeling and simulation of coupled EM-thermal-fluid-plasma problems involving multiscale media;

3. **Multiphysics investigation of EM energy assisted catalytic active sites enhancement:**

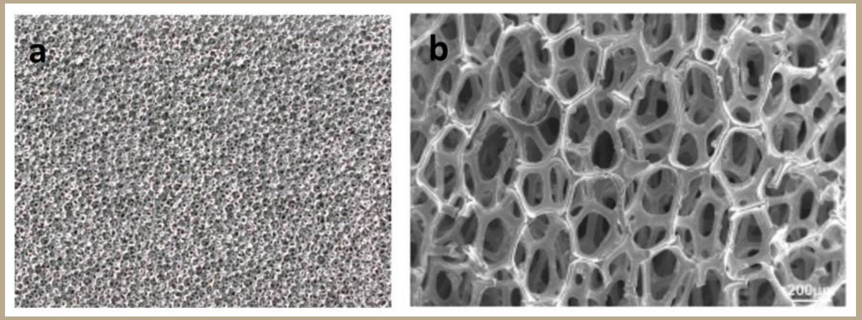
   Develop multiscale simulation methods to couple physical with chemical phenomena based on the coupled modeling of the NDG method, reactive forcefield molecular dynamics (ReaxFF MD) simulations, and density functional theory (DFT) simulations;

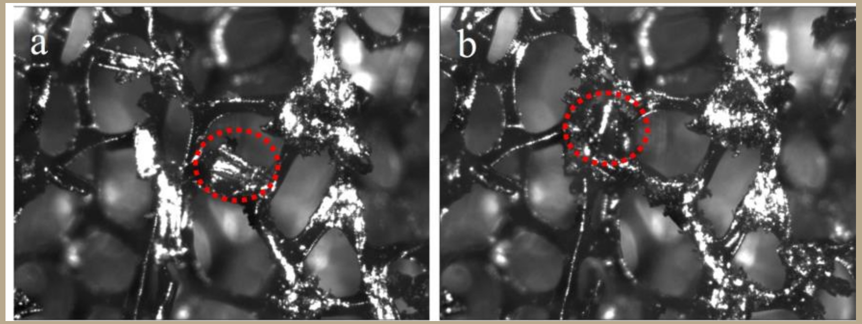
4. **System design and optimization for high-yield and low-cost hydrogen generation:**

   Use the methods developed above to design an optimized EM-assisted catalytic system for an improved hydrogen conversion efficiency with a lowered cost.

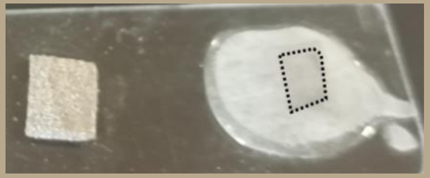
PROJECT STATUS: 3D IMAGING & RECONSTRUCTION

- Optical 3D Imaging to Obtain Morphology of Catalyst and Study the Localized Chemical Reactions:

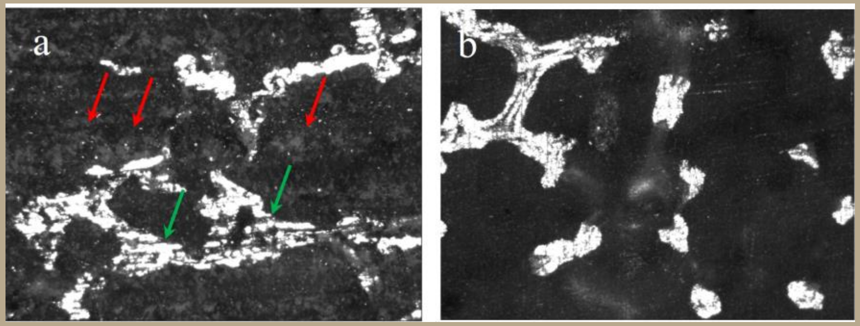




Movement of Ni foam. The red dashed circles indicate the same region before (a) and after (b) the polishing. The structure shape and pattern are totally different.



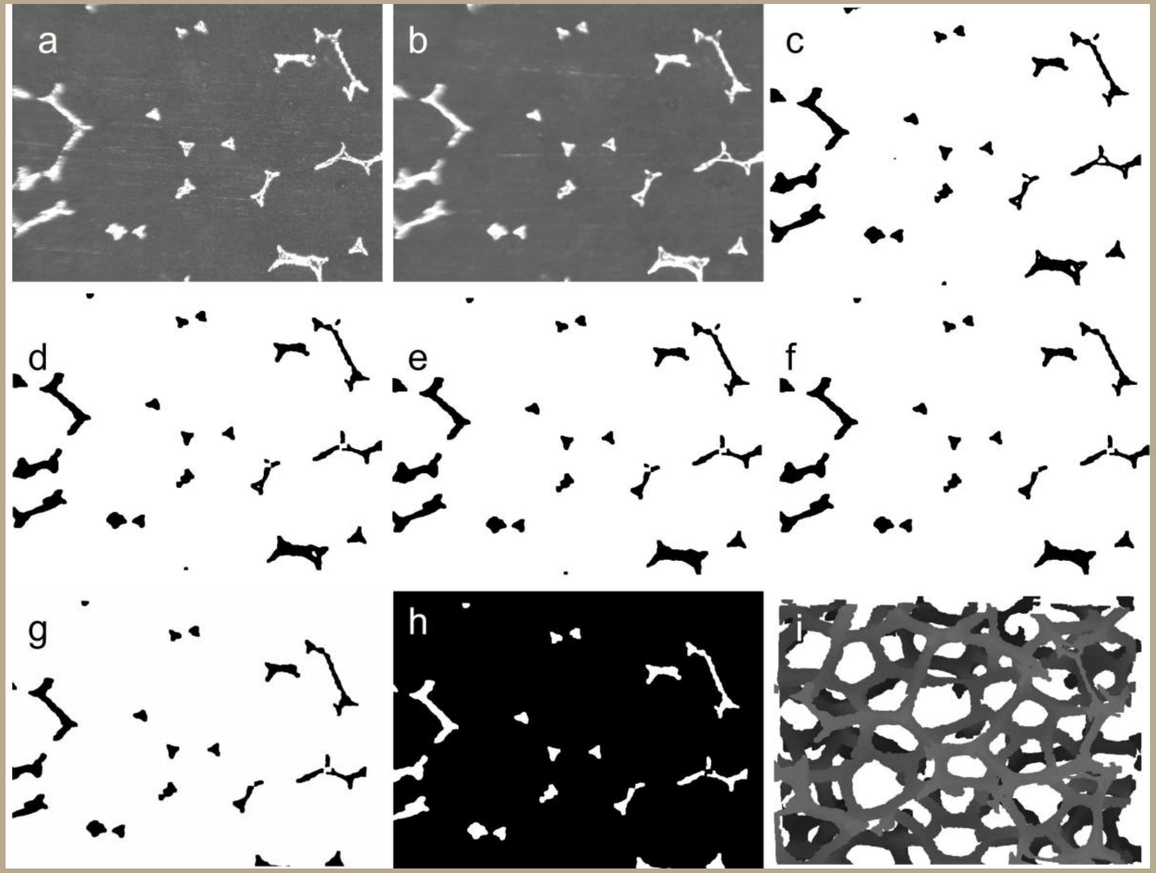
Ni foam with (right) and without (left) epoxy support.



Polished cross-section of Ni foam. (a) Polished in air. (b) Polished in water. Green arrows showed the scratch line, and the red arrows show epoxy residue.

PROJECT STATUS: 3D IMAGING & RECONSTRUCTION

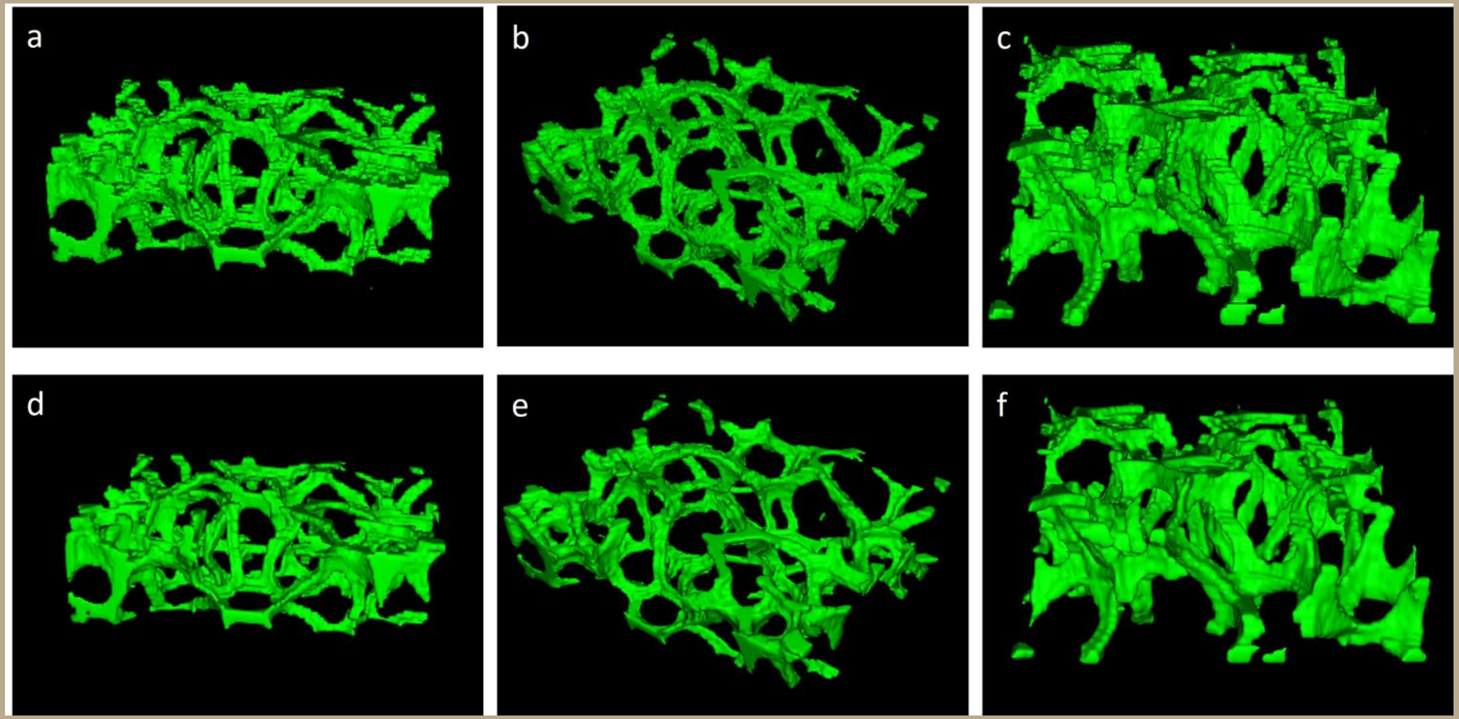
- Optical 3D Imaging to Obtain Morphology of Catalyst and Study the Localized Chemical Reactions:



Modified image segmentation. (a) The raw image of the cross-section of Ni foam. (b) Using median filter to remove the bright particles on the surface. (c) Segmentation by setting a threshold. (d) Using dilate and erode to smooth the boundaries. (e) Fill the holes to remove white area in black part. (f) Remove small particles. (g) Image alignment of adjacent pictures. (h) Switch the color. (i) Build the 3D stack in z direction.

PROJECT STATUS: 3D IMAGING & RECONSTRUCTION

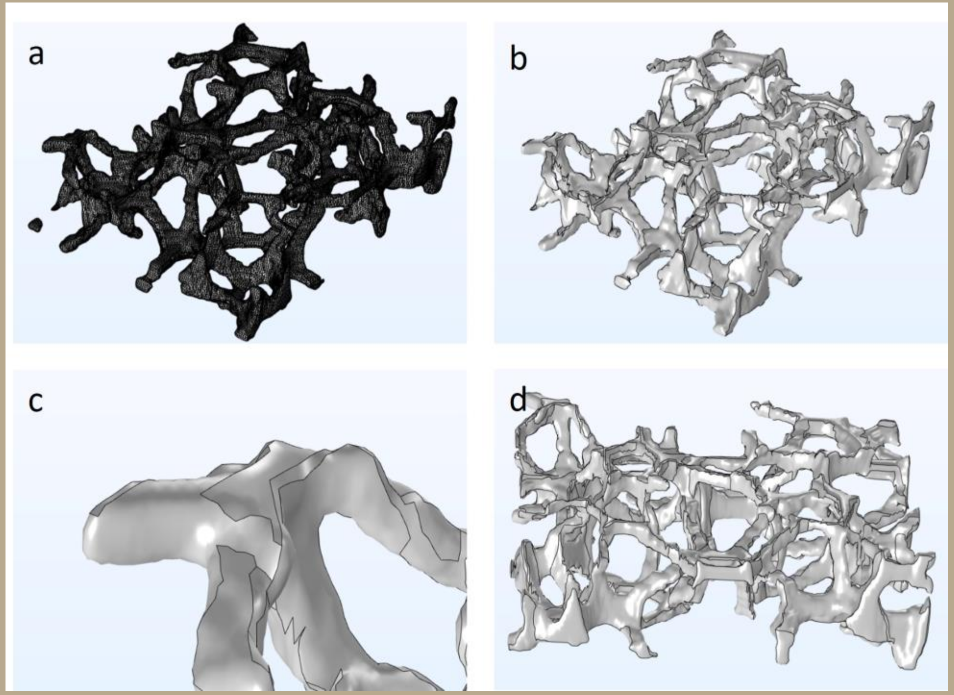
- Optical 3D Imaging to Obtain Morphology of Catalyst and Study the Localized Chemical Reactions:



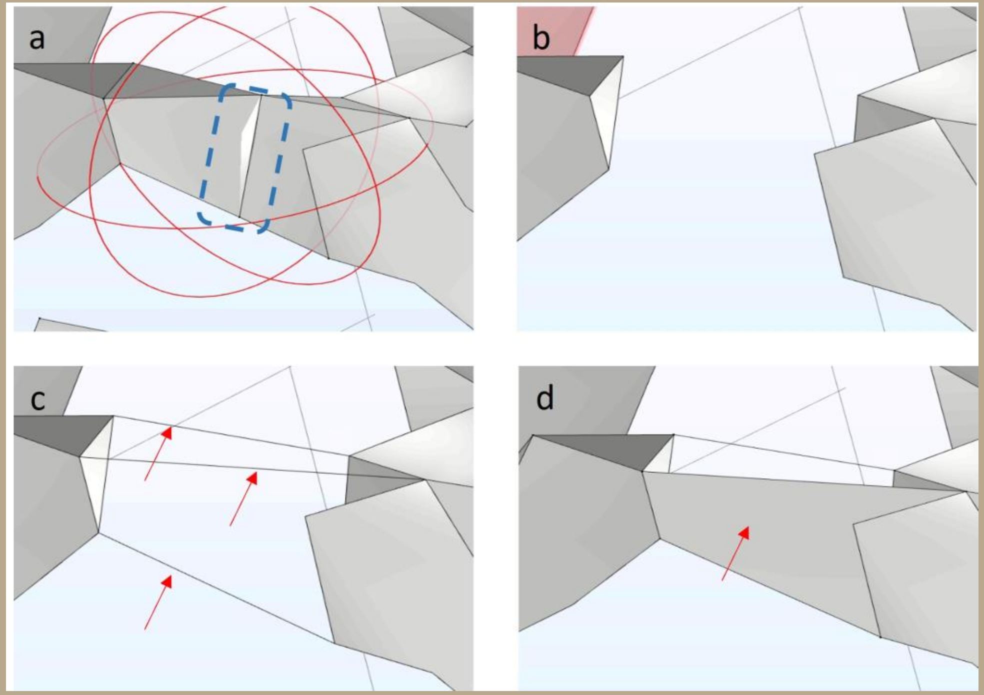
3D reconstructed Ni foam viewing from different angles. a-c before the smooth, and d-f after the smooth. The entire structure is ~ 1.8 mm wide and ~0.5 mm thick.

PROJECT STATUS: 3D IMAGING & RECONSTRUCTION

- Optical 3D Imaging to Obtain Morphology of Catalyst and Study the Localized Chemical Reactions:



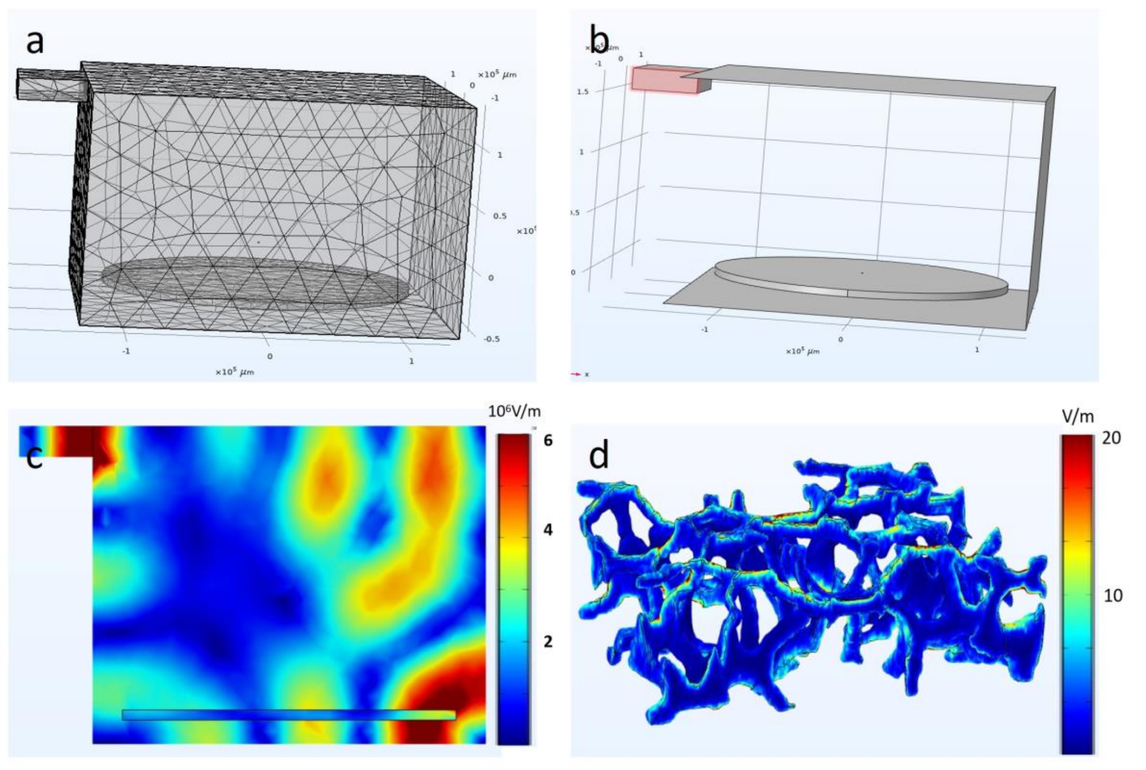
Imported the 3D geometry into the MESHLAB to remove the extra small domains that is caused by the image reconstruction errors Nickel foam. 3D model in COMSOL (a-b) 3D geometry with (a) and without the mesh (b). (c) A zoomed in region. (d) 3D geometry with different angle.



Process to remove self-intersecting surfaces and edges. (a) Using balling function to split surface. (b) Delete the intersecting surfaces. (c) Generate new edges. (f) Create new surfaces based on the new edges.

PROJECT STATUS: COMSOL SIMULATION

- Optical 3D Imaging to Obtain Morphology of Catalyst and Study the Localized Chemical Reactions:

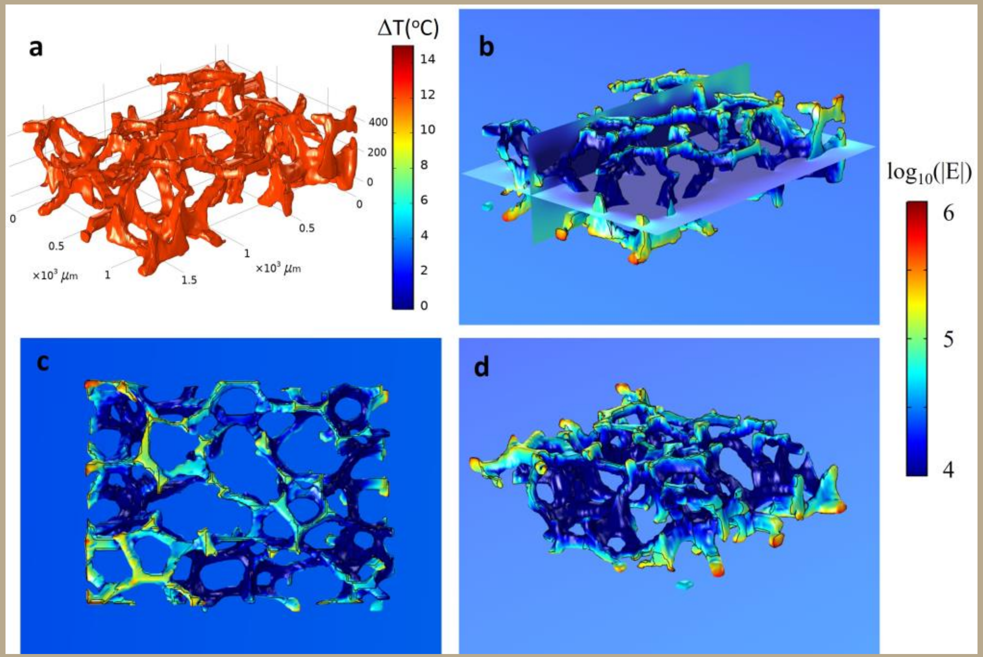


We used the RF module in the COMOSL. The input power of the port is set to 1,000 W, and the frequency is set to 2.45 GHz (**Fig. 1-12 b**). All the boundaries of the microwave is set to impedance boundary. The model includes the waveguide, steel container, glass plate and nickel foam. The rectangular power port (red arrow in Fig. 1-13a) is used to excite the microwave using a transverse electric (TE) boundary condition at 2.45 GHz.

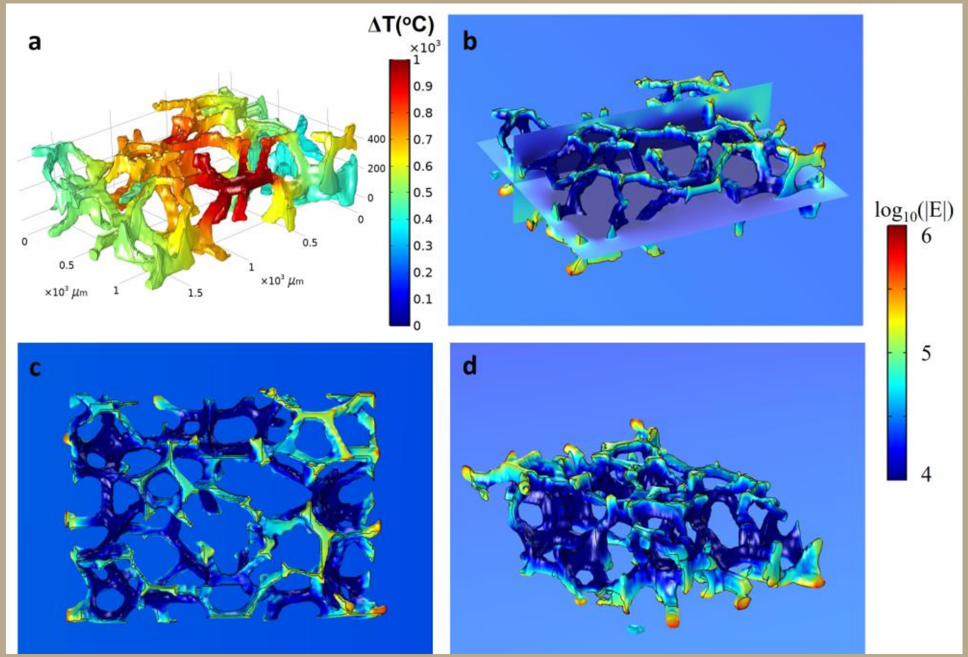
Property	Carbon black	Nickel	Unit
Relative permittivity	62-19j	1	1
Relative permeability	1	3	1
Electrical conductivity	1	2e5	S/cm
Thermal conductivity	0.35	200	W/(m·K)
Density	2000	8900	kg/m³
Heat capacity	1262	350	J/(kg·K)

PROJECT STATUS: COMSOL SIMULATION

- Optical 3D Imaging to Obtain Morphology of Catalyst and Study the Localized Chemical Reactions:



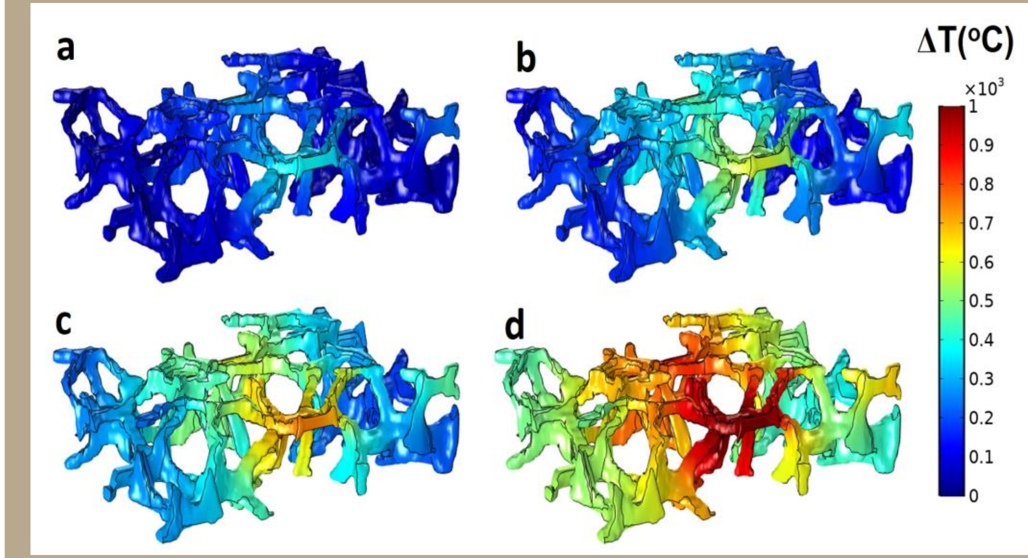
Electric field and temperature distribution on **Ni foam**. (a) Temperature distribution after 5 seconds of microwave heating. (b) The electric field distribution around the 3D Ni foam. (c-d) Electric field distribution at different angles.

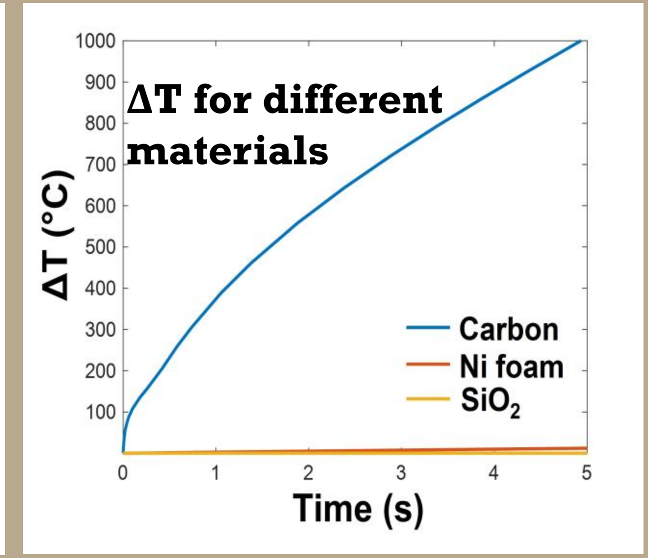


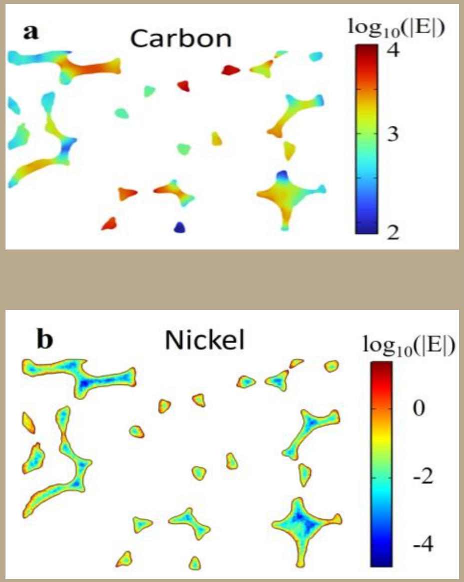
Electric field and temperature distribution on **carbon foam**. (a) Temperature distribution after 5 seconds of microwave heating. (b) The electric field distribution around the 3D Ni foam. (c-d) Electric field distribution at different angles.

PROJECT STATUS: COMSOL SIMULATION

- Optical 3D Imaging to Obtain Morphology of Catalyst and Study the Localized Chemical Reactions:





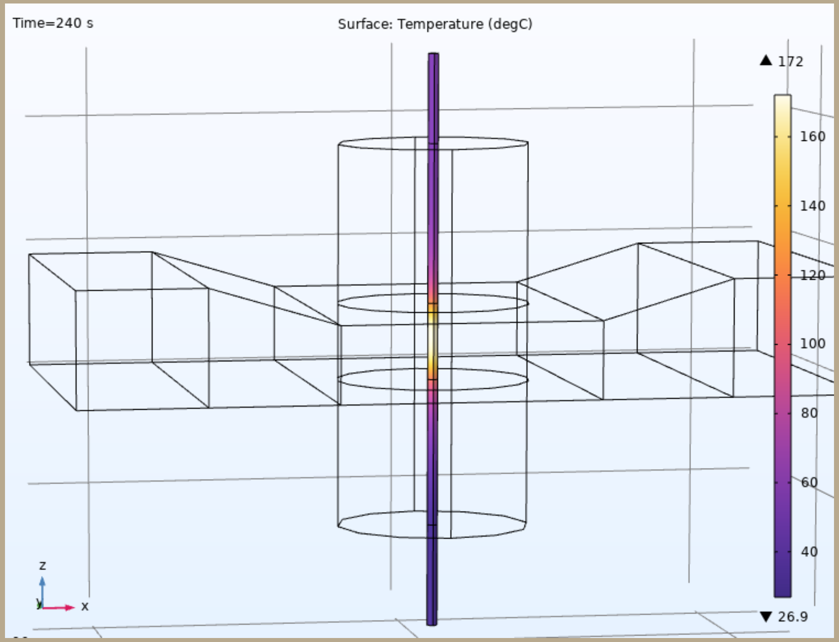


Temperature distribution changes over time on carbon foam (a-d), and the heating time for a-d are 1 s, 2 s, 3 s, and 5 s, respectively. (e) Temperature vs time curve.

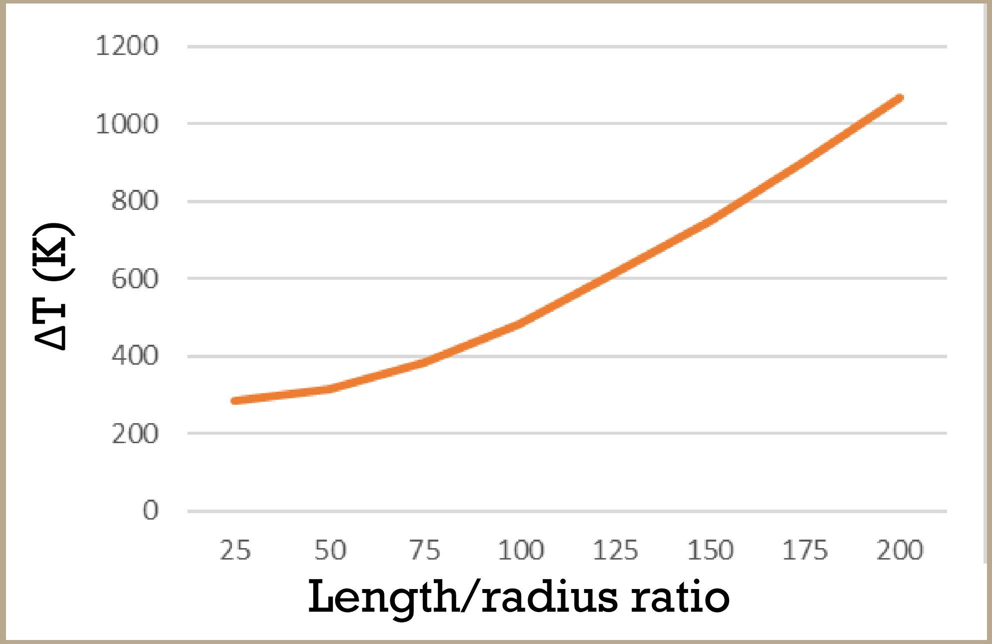
Electric field distribution across a cross section of the carbon (a) and Ni foam (b). E-field of carbon foam is much bigger than that of Ni foam. The penetration depth of E-field in carbon is much bigger than the Ni foam.

PROJECT STATUS: COUPLED SIMULATION MODEL

- Development of Coupled Physical-Chemical Simulation Model:



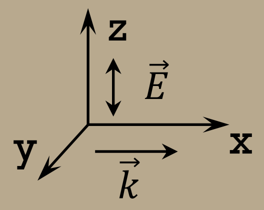
COMSOL Multiphysics is used, and RF module is used to simulate the microwave's generation, and plasma module is coupled to simulate the plasma's generation. The center rod is carbon material.



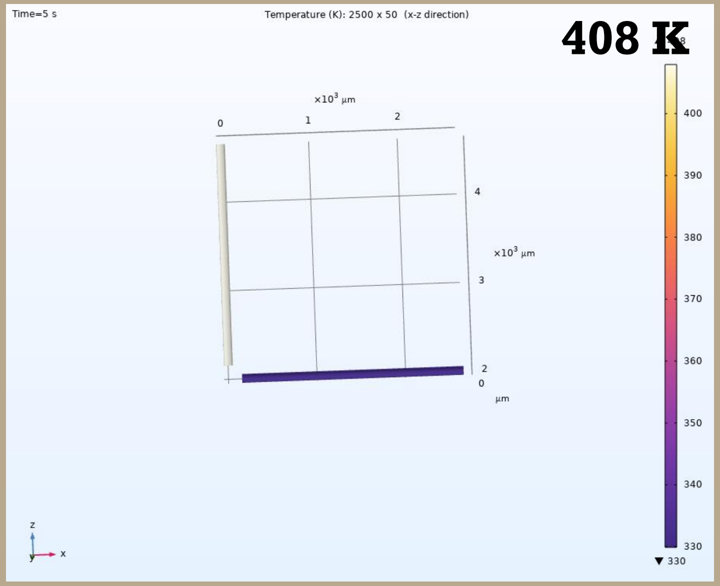
Heating of carbon material: the conductivity of carbon materials (such as carbon black) is in between metal (reflect EM) and insulator (transmit EM), so it will observe EM and convert the EM energy to heat very efficiently. Geometry of the carbon material is also very important.

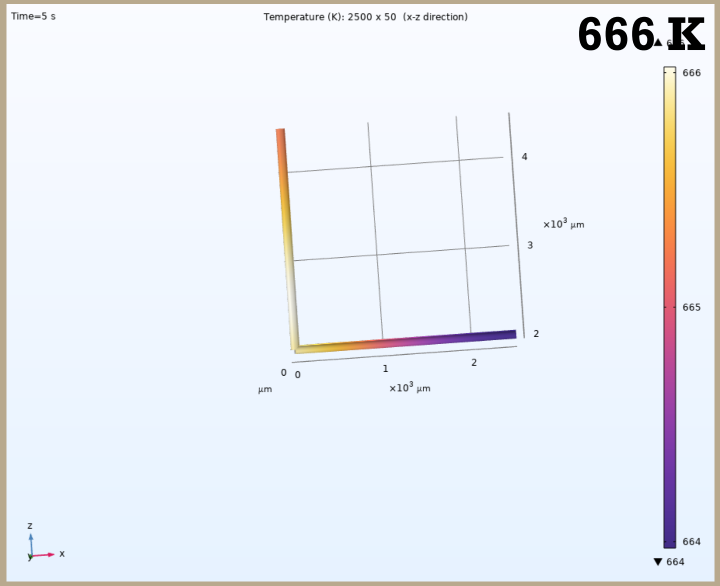
PROJECT STATUS: COUPLED SIMULATION MODEL

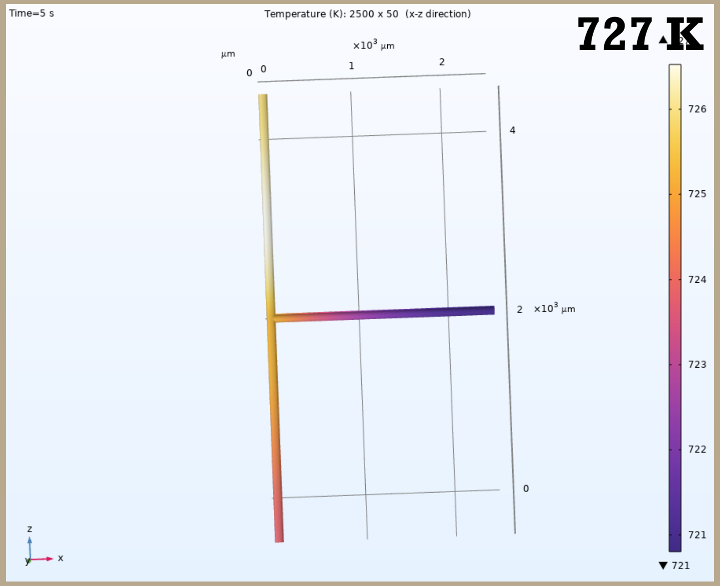
- Development of Coupled Physical-Chemical Simulation Model:



Geometry of the carbon materials affect the heating effect

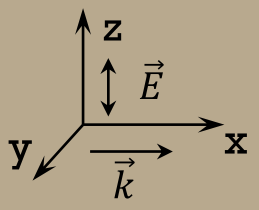




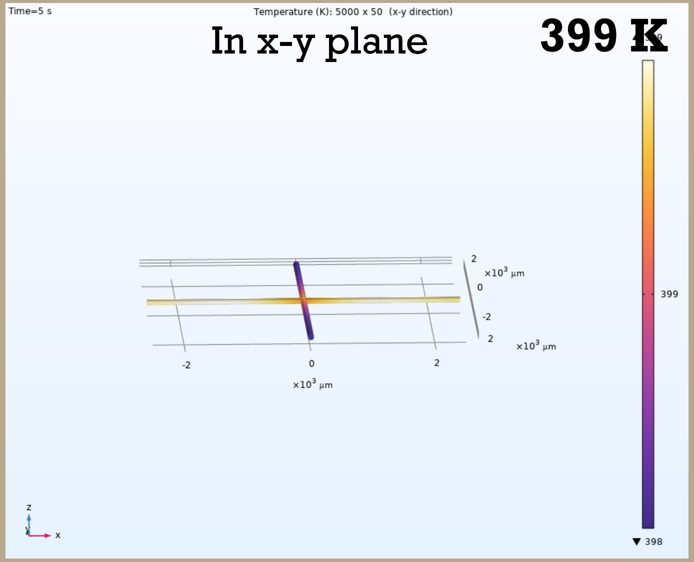


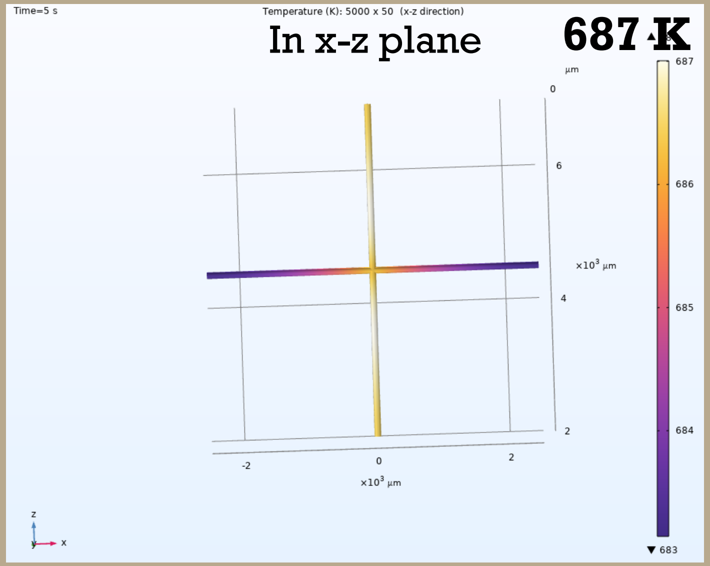
PROJECT STATUS: COUPLED SIMULATION MODEL

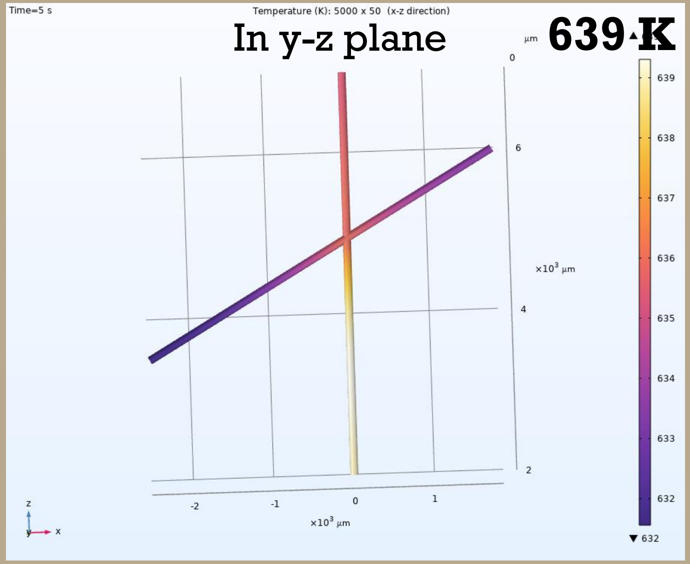
- Development of Coupled Physical-Chemical Simulation Model:



Orientation of the carbon materials affect the heating effect







PROJECT STATUS: MULTISCALE EM MODELING

- Numerical Simulation of Multiscale Electromagnetic Problems:
  - Overall large size of the simulation domain: large system to solve;
  - Sub-wavelength geometrical structures cause numerical instability: numerical breakdown;
- An all-frequency stable formulation is proposed, implemented, and validated:
  - All four Maxwell's equations and the current continuity equation are formulated
  - Inhomogeneous Coulomb gauge is enforced.

$$\nabla \times \frac{1}{\mu} \nabla \times \boldsymbol{A} - \omega^2 \tilde{\epsilon} \boldsymbol{A} + \mathrm{j}\omega\tilde{\epsilon}\nabla\phi + \frac{\epsilon_{\mathrm{r}}}{\mu_0}\nabla\chi = \boldsymbol{J}_{\mathrm{imp}}$$

$$-\mathrm{j}\omega\nabla \cdot \tilde{\epsilon}\boldsymbol{A} - \nabla \cdot \tilde{\epsilon}\nabla\phi = \rho_{\mathrm{imp}}$$

$$-\frac{1}{\mu_0}\nabla \cdot \epsilon_{\mathrm{r}}\boldsymbol{A} + \chi = 0$$

where $\tilde{\epsilon} = \epsilon - \mathrm{j}\sigma/\omega$ denotes the complex permittivity and χ denotes an auxiliary variable introduced to enforce the inhomogeneous Coulomb gauge $\nabla \cdot \epsilon\boldsymbol{A} = 0$.

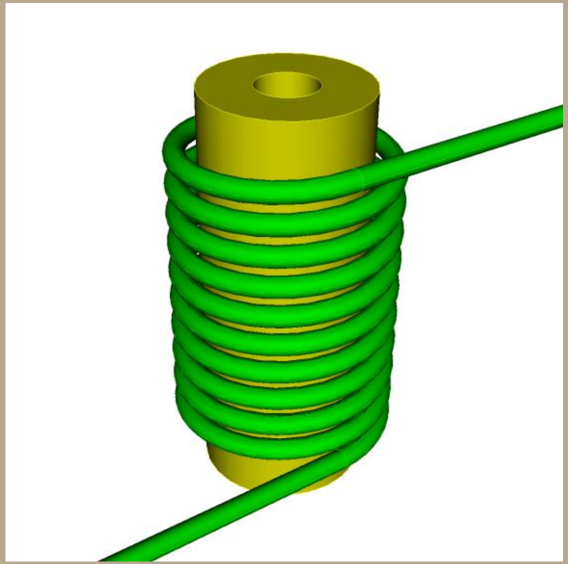
The electric field intensity $\boldsymbol{E}$ and the magnetic flux density $\boldsymbol{B}$ can be recovered as

$$\boldsymbol{E} = -\mathrm{j}\omega\boldsymbol{A} - \nabla\phi$$

$$\boldsymbol{B} = \nabla \times \boldsymbol{A}$$

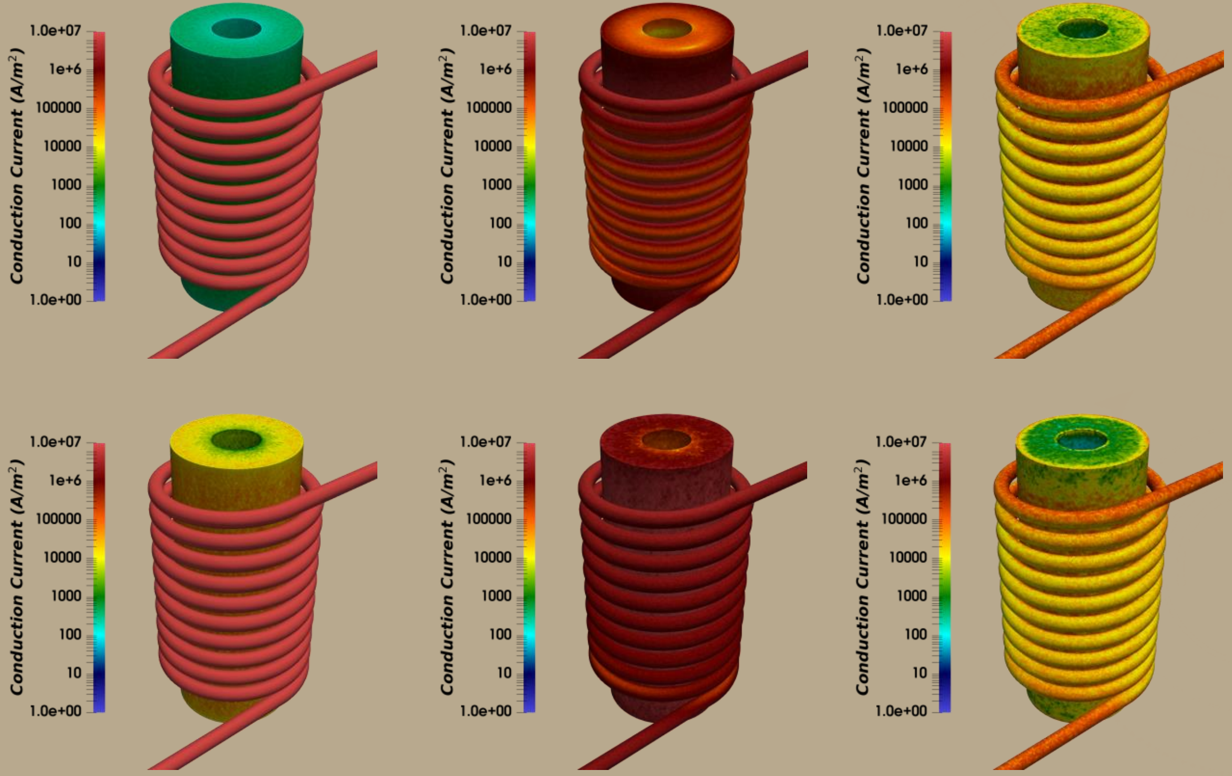
PROJECT STATUS: MULTISCALE EM MODELING

- Validation Example: Induction Heating
  - Copper conduction wire connected to a 1-V source
  - Aluminum/magnetic heating core.



$\mu_r = 1$

$\mu_r = 1000$



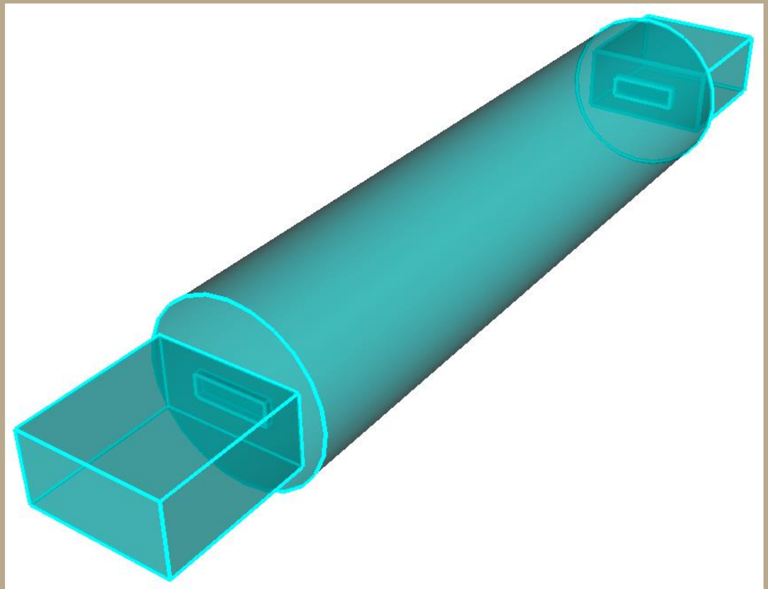
$f = 1$ Hz

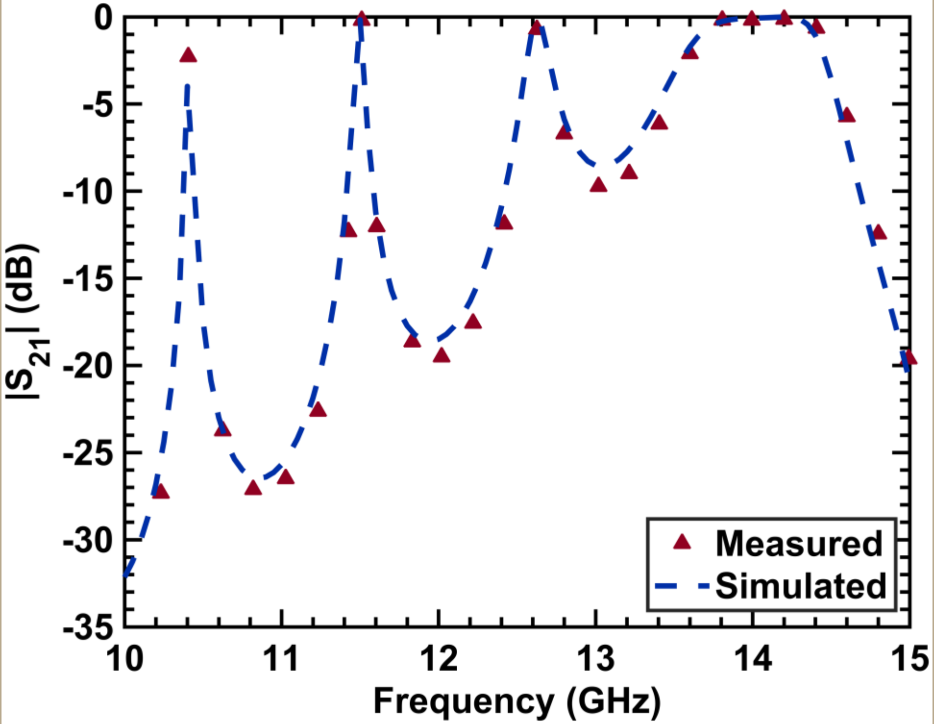
$f = 1$ kHz

$f = 100$ kHz

PROJECT STATUS: MULTISCALE EM MODELING

- Validation Example: Cylindrical Cavity Resonator
  - WR75 waveguide
  - Internal resonator connected through two rectangular slots
  - S-parameter is calculated in a wide frequency band.





PROJECT STATUS: THERMAL MODELING

- Numerical Simulation of Heat Transfer Problems:
  - Equilibrium and transient simulations.
  - Linear and nonlinear materials.
- Heat transfer equation with convective flow:

$$\rho C_P \frac{\partial T}{\partial t} + \rho C_P \vec{u} \cdot \nabla T = \nabla \cdot (\kappa \nabla T) + q$$

where ρ is the density of the materials, C_P is the specific heat, κ is the thermal conductivity, $\vec{u}$ is the fluid velocity, and q is the volumetric heat generation. The corresponding boundary conditions associated with the transient thermal analysis includes the Dirichlet boundary condition

$$T = T_d$$

and the air convection boundary

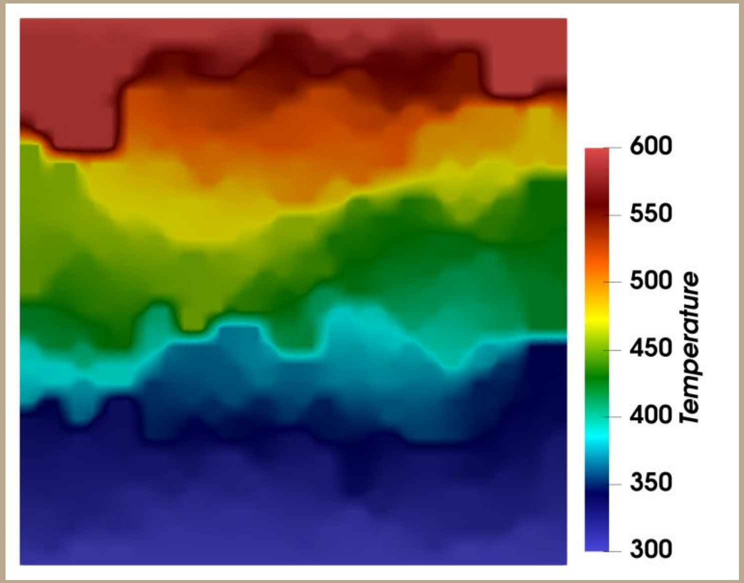
$$\hat{n} \cdot \kappa \nabla T = -h(T - T_a)$$

where h and T_a are the convective heat transfer coefficient and the ambient temperature, respectively.

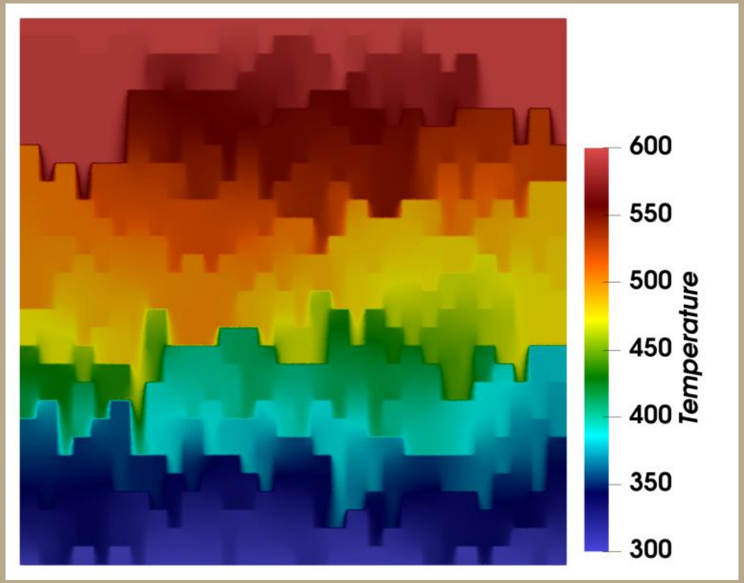
PROJECT STATUS: THERMAL MODELING

- Validation Example:
  - Equilibrium and transient simulations.
  - Linear and nonlinear materials.

Geometry of a 30-by-30 random checkerboard



Temperature profile with conductive heat transfer



Temperature profile with conductive and convective heat transfer

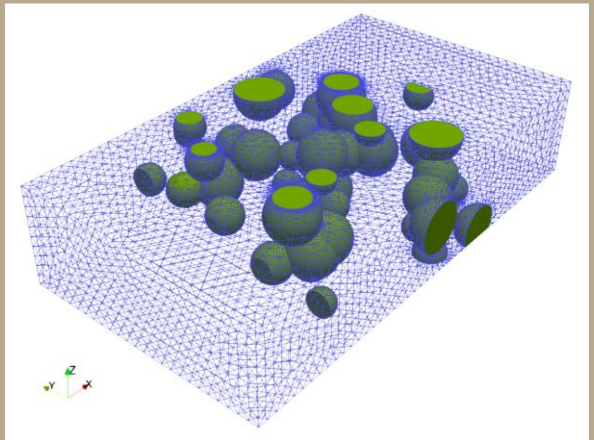
PROJECT STATUS: MULTIPHYSICS MODELING

- Numerical Simulation of Multiphysics-Multiscale Electromagnetic-Thermal Problems:
  - All-frequency stable formulation coupled with heat transfer equation.
- Multiphysics Coupling:
  - Electromagnetic power dissipation is converted to heat.
  - The volumetric heat source includes dielectric loss and conduction (Ohmic) loss:

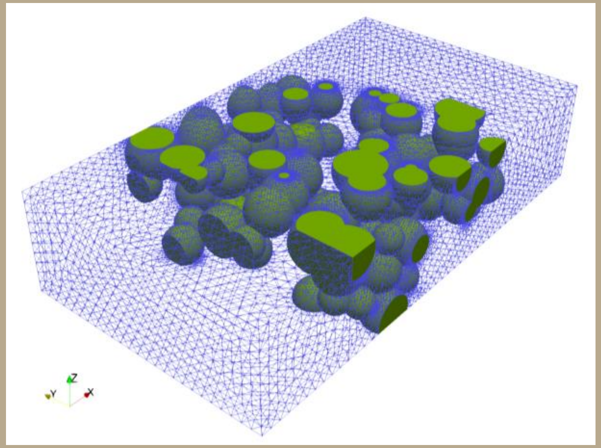
$$q = (\omega\epsilon'' + \sigma)\|\boldsymbol{E}\|^2$$

PROJECT STATUS: MULTIPHYSICS MODELING

- Microwave induced thermal effect
  - Total size: 1800 um by 1000 um by 400 um
  - Particle sizes: 50 to 100 um in radii
  - Operating frequency: 2.45 GHz
  - Microwave intensity: 10 kV/m
  - Ambient temperature: 300 K
  - Convection cooling at near and far ends
  - Thermal insulation at other four walls



50 particles



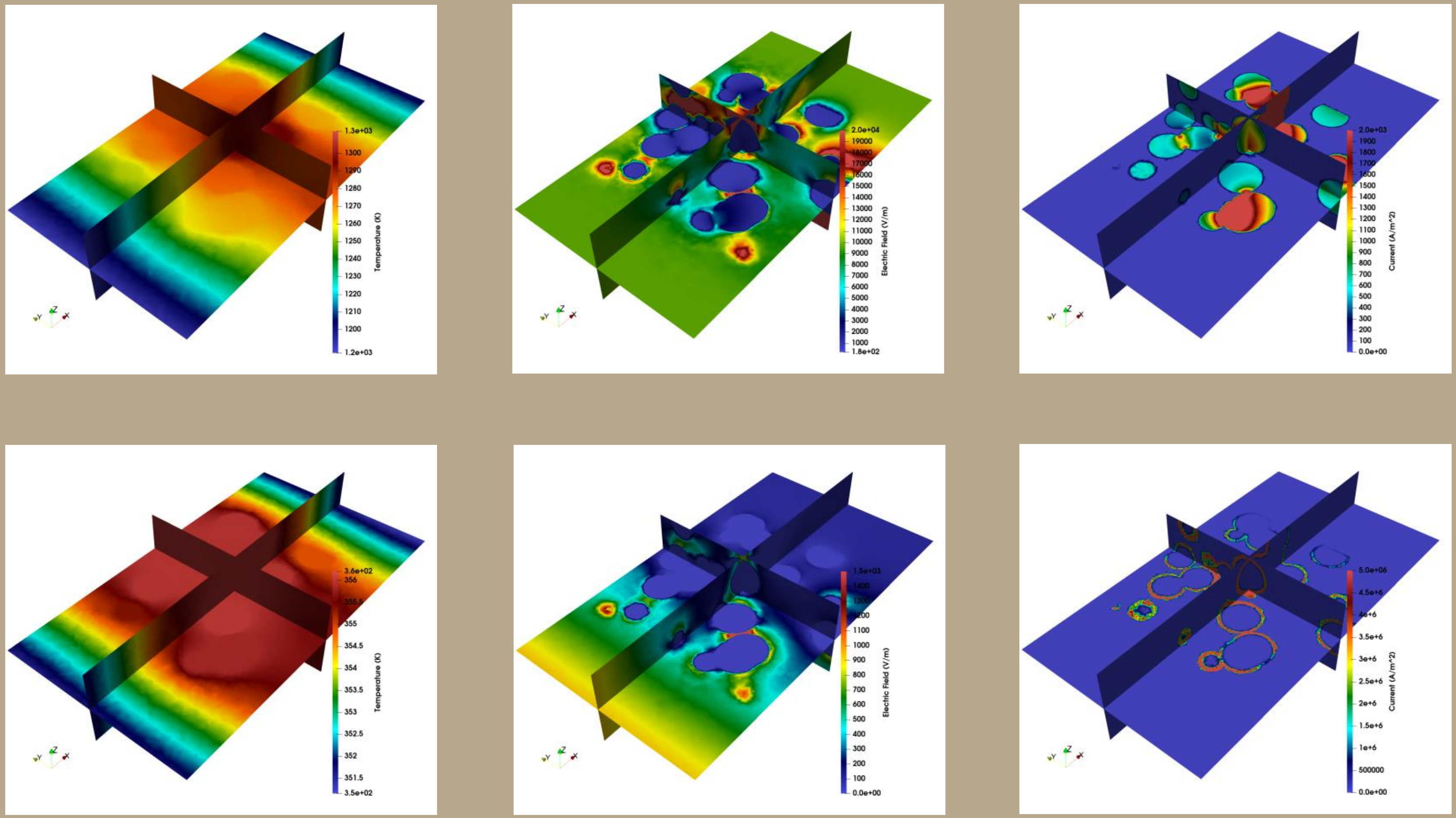
100 particles

PROJECT STATUS: MULTIPHYSICS MODELING

- 50 Particles

Carbon
(max: 1,300K)

Nickel
(max: 360 K)



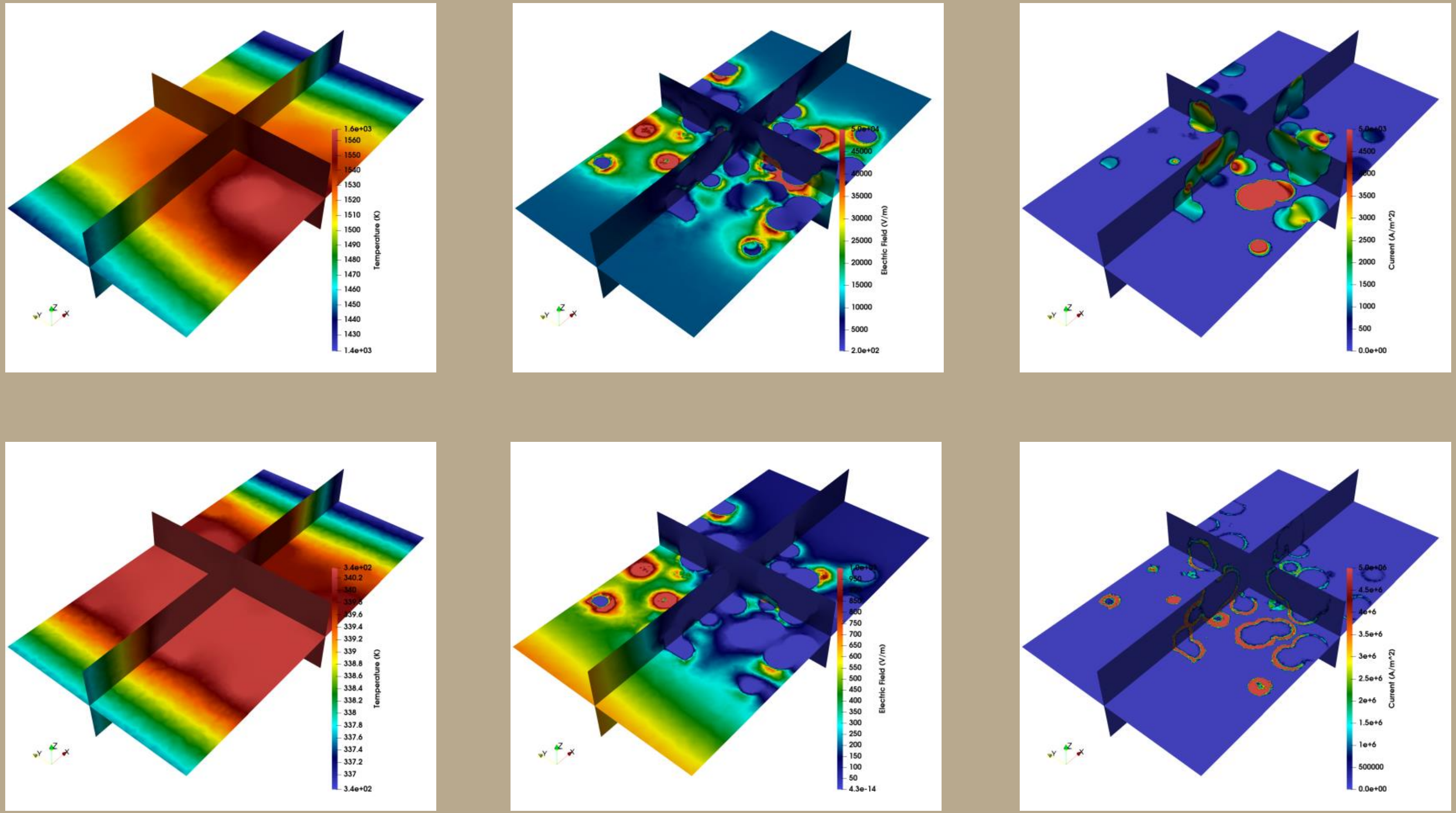
Temperature | Electric Field | Conduction Current

PROJECT STATUS: MULTIPHYSICS MODELING

- 100 Particles

Carbon
(max: 1,600K)

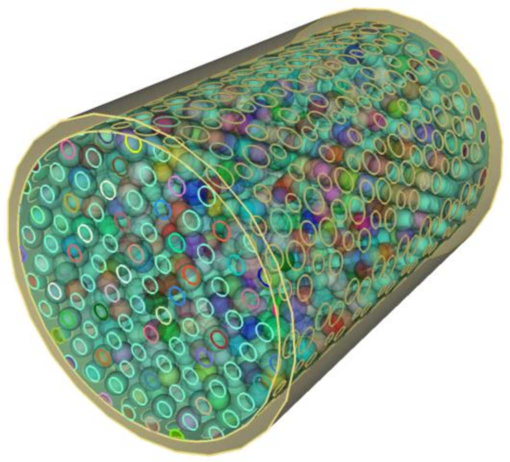
Nickel
(max: 340 K)

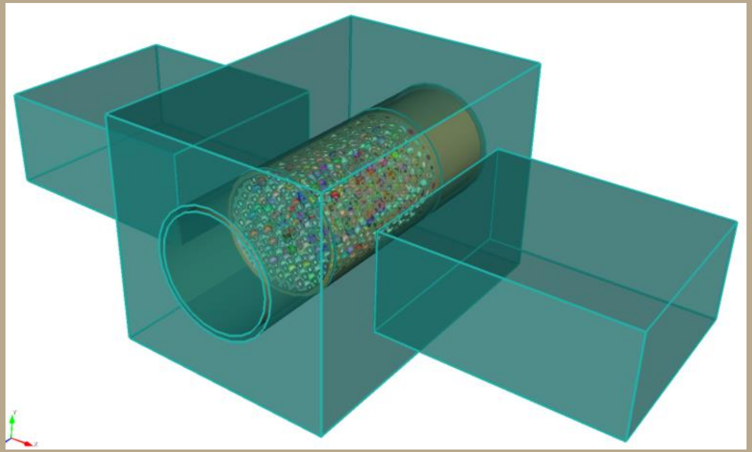


Temperature Electric Field Conduction Current

PROJECT STATUS: SIMULATION OF REACTION CHAMBER

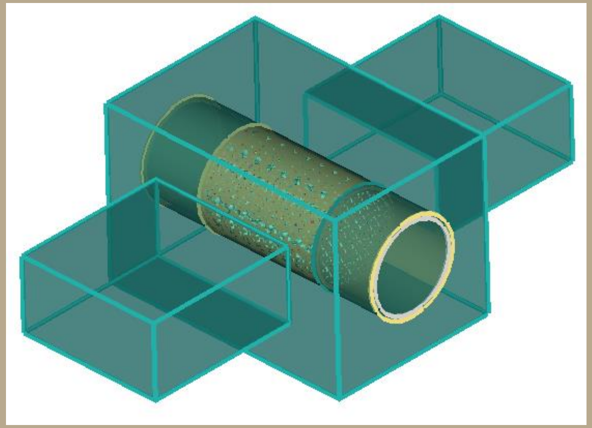
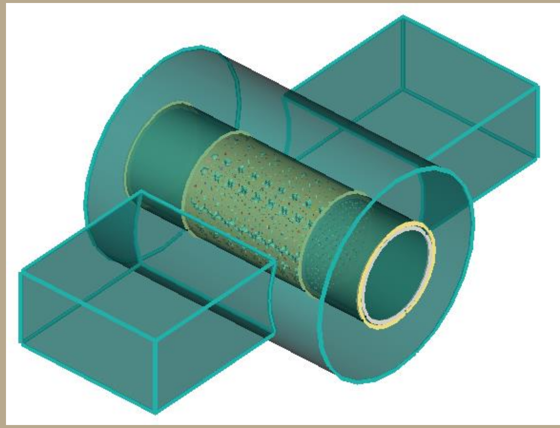
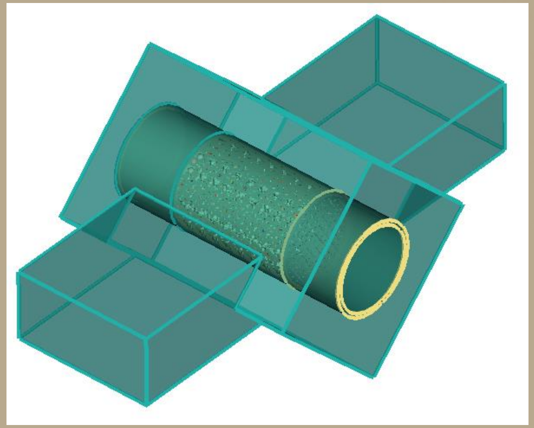
- **Microwave input from WR340 waveguide**
  - Operating frequency: 2.45 GHz
  - Input power: 2 kW
- Microwave Chamber Size:
  - 100 cm^2 by 15 cm
- Reaction Quartz Tube Size:
  - 5 cm diameter
- Catalyst Region:
  - Total size: 5 cm diameter by 7.5 cm length
  - Pore size: 2 mm radius, 2029 air pores
  - Carbon black backbone considered



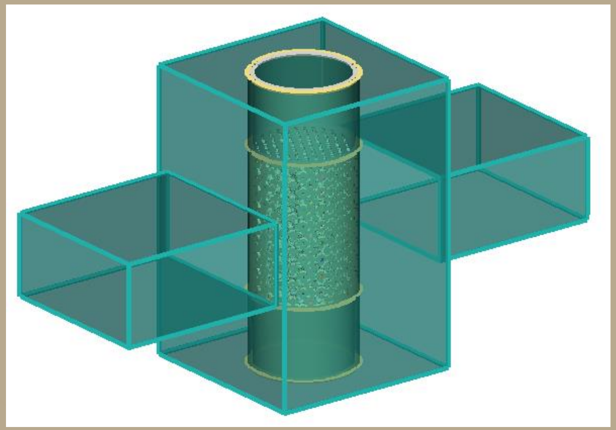
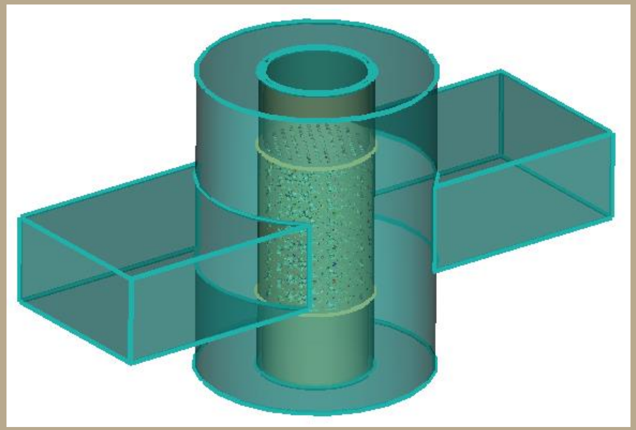
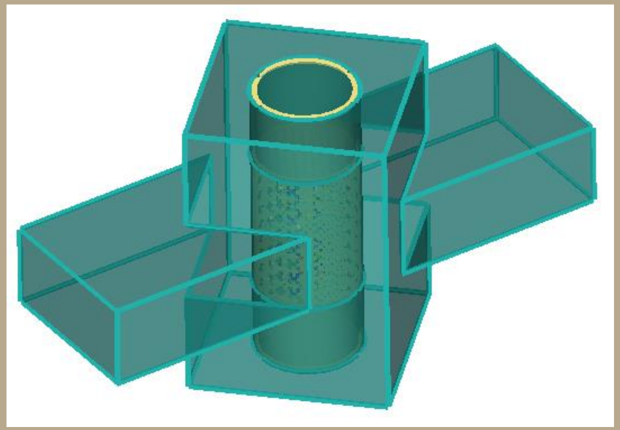


REACTION CHAMBER: CONFIGURATIONS

- **Horizontally Placed Chamber and Tube:**

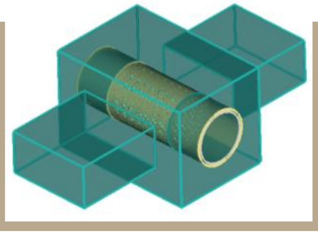




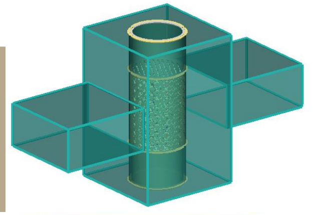
- **Vertically Placed Chamber and Tube:**

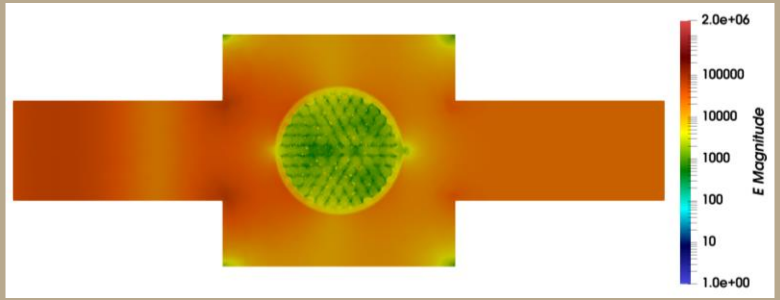
Cuboid **Cylinder** **Diamond**

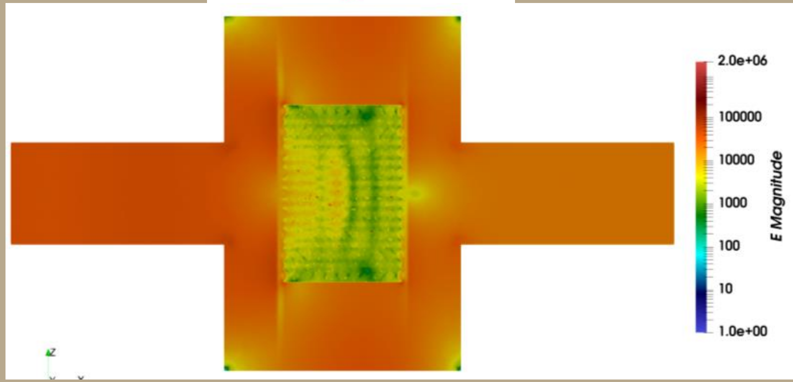
REACTION CHAMBER: CUBOID



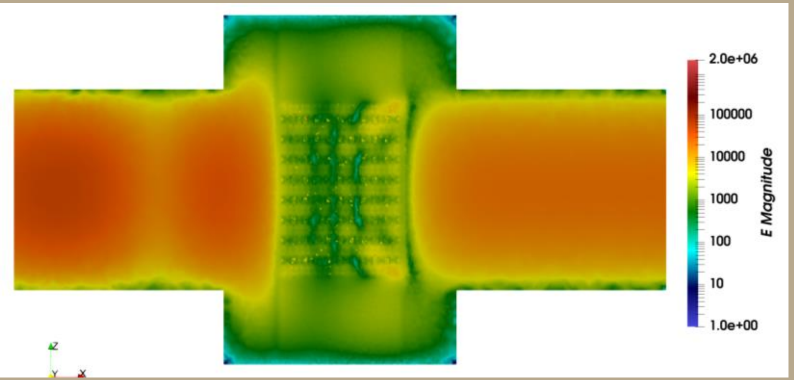


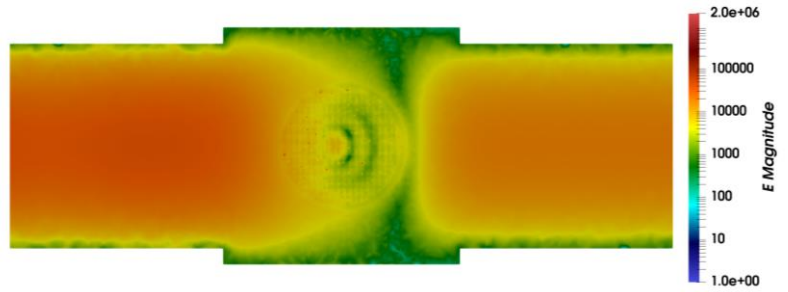
E Plane





H Plane

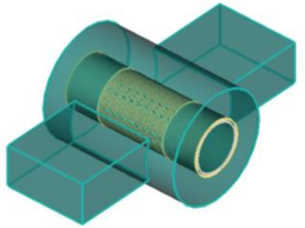


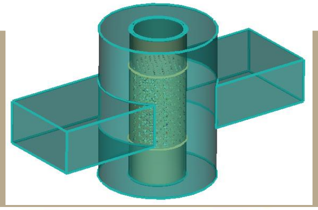


EM Energy Absorption Rate:

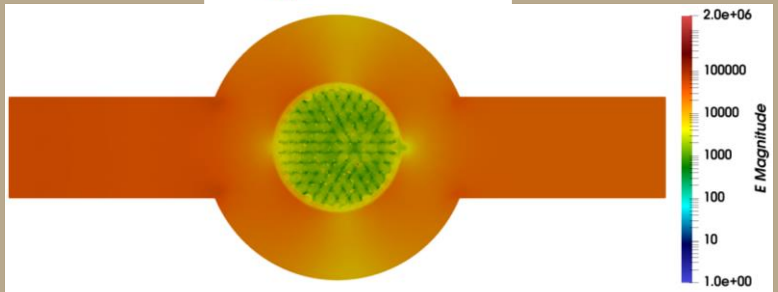
33.8% **71.0%**

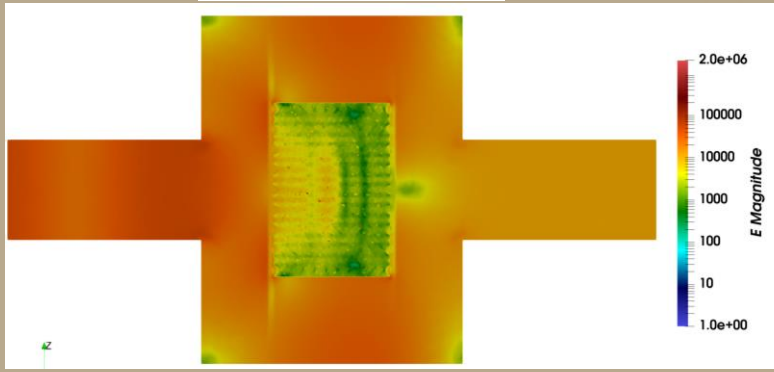
REACTION CHAMBER: CYLINDER



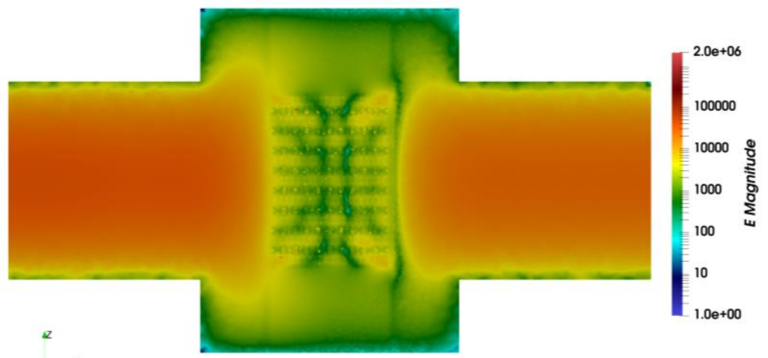


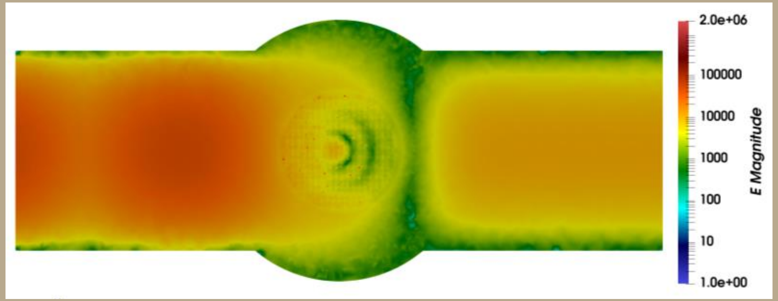
E Plane





H Plane

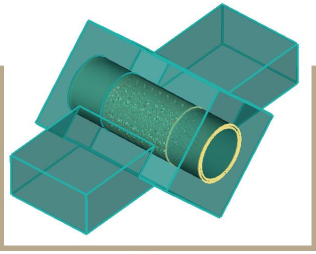


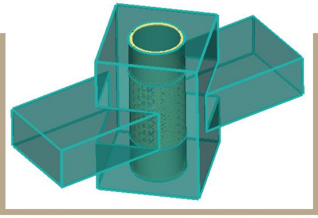


EM Energy Absorption Rate:

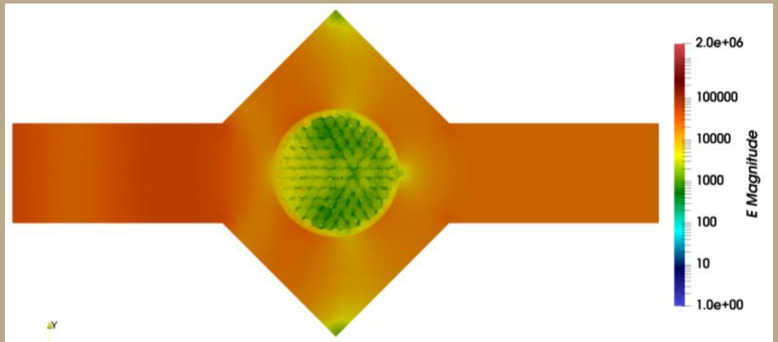
43.6% **78.0%**

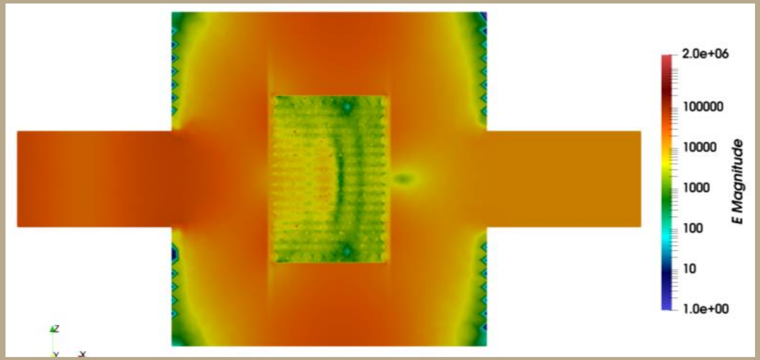
REACTION CHAMBER: DIAMOND



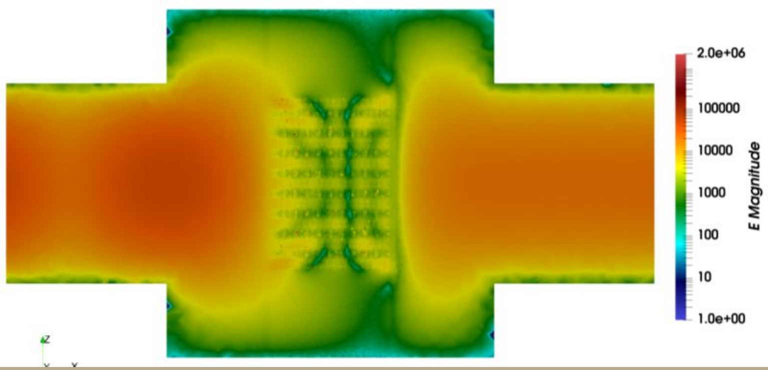


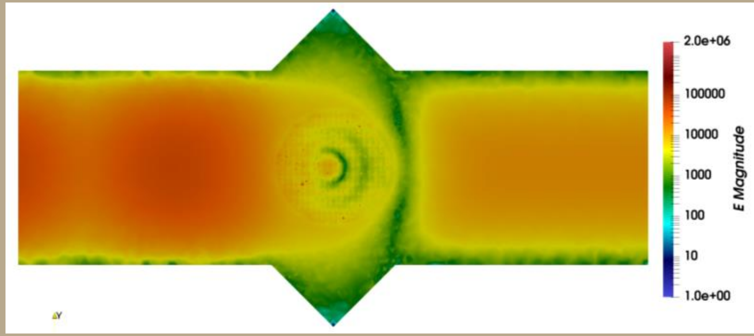
E Plane





H Plane





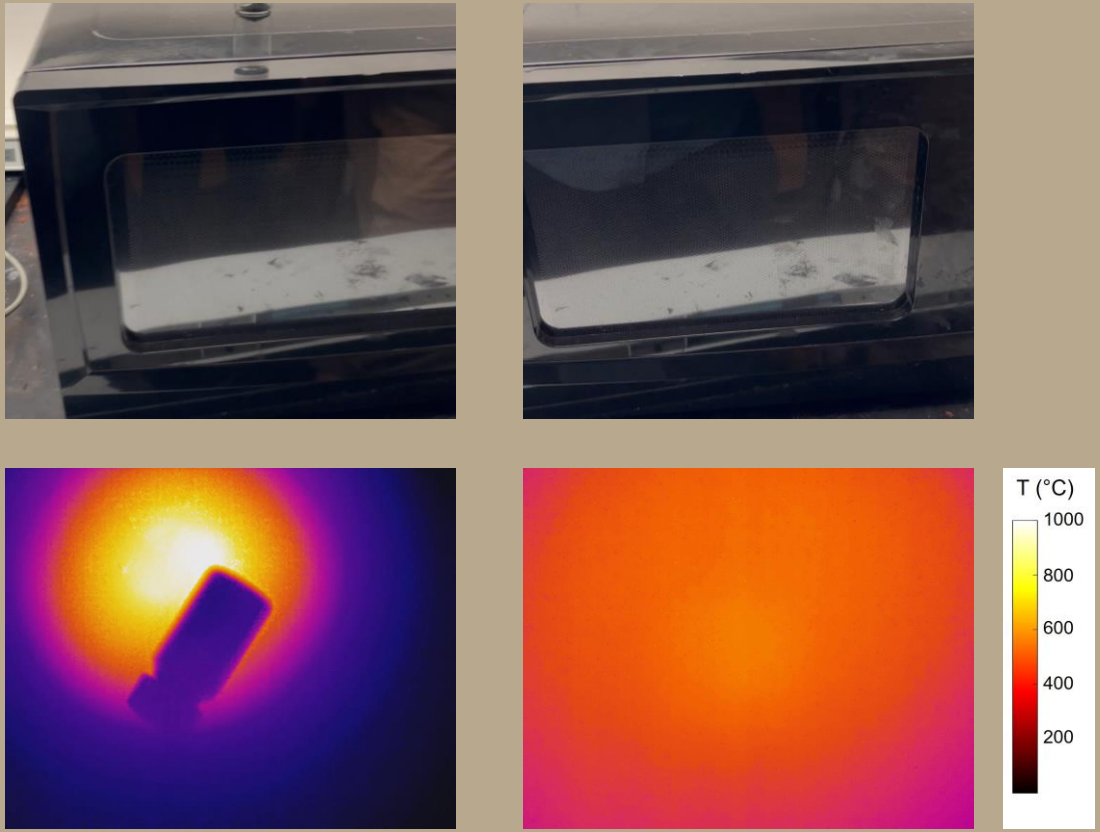
EM Energy Absorption Rate:

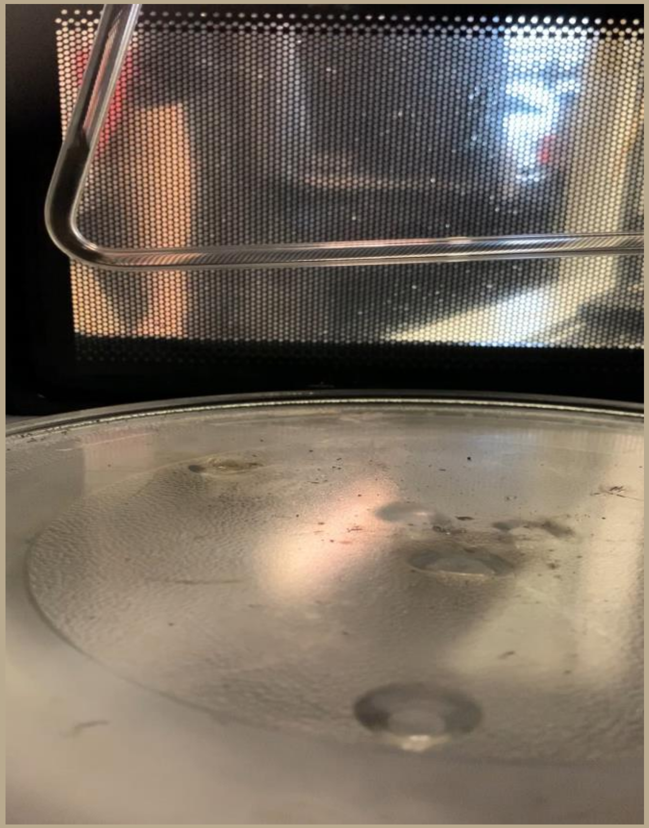
48.5% **73.5%**

PROJECT STATUS: PLASMA GENERATION

- Development of Coupled Physical-Chemical Simulation Model:

Plasma Generation

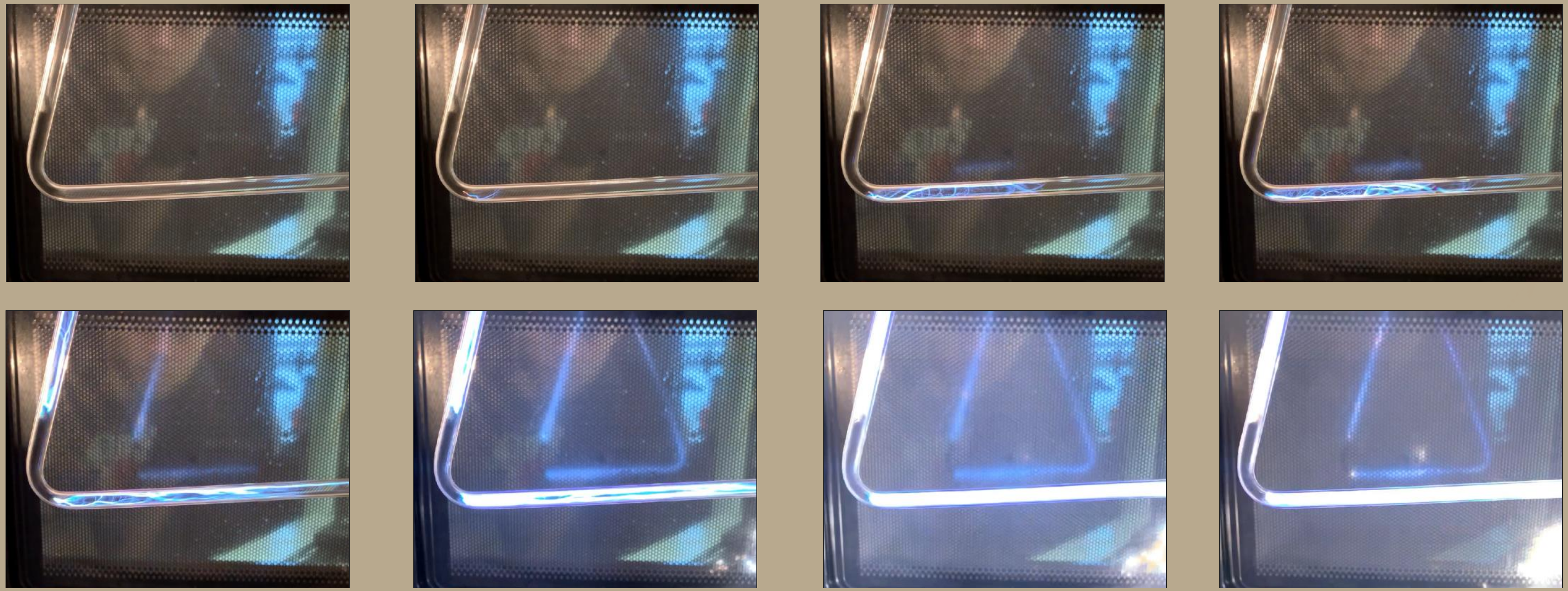




PROJECT STATUS: PLASMA GENERATION

- Development of Coupled Physical-Chemical Simulation Model:

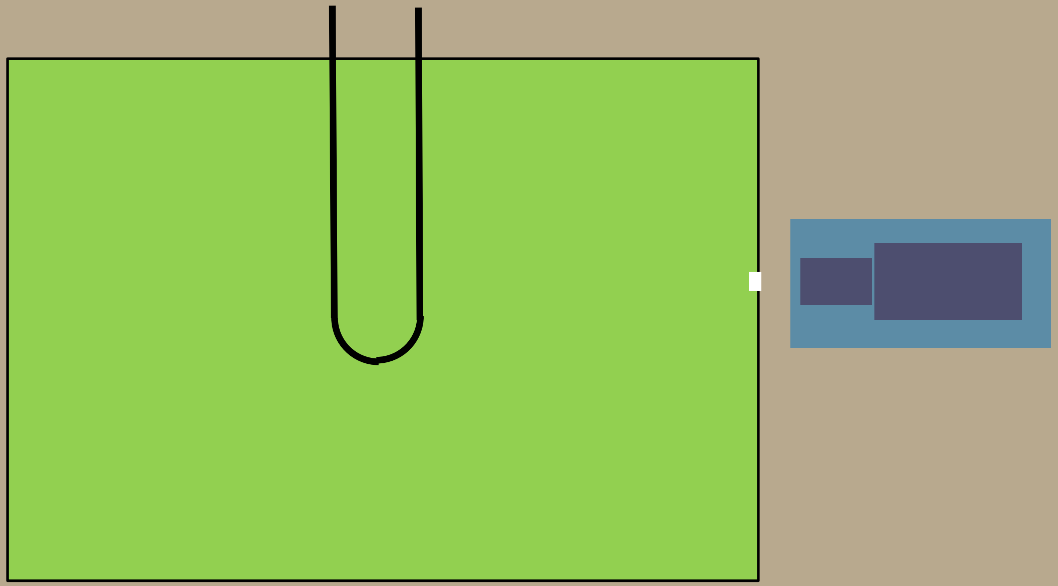
Plasma Generation

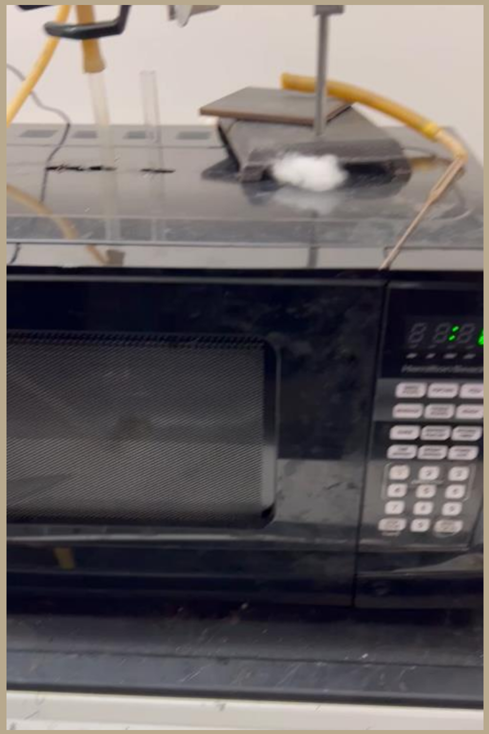


PROJECT STATUS: PLASMA GENERATION

- Development of Coupled Physical-Chemical Simulation Model:

Operando Imaging to Study the Local Plasma Generation





- We want to image the local hot spot of plasma generation from the carbon fiber bundle.
- Build experimental setup with the camera;
- Use a metal mesh to shield the camera to protect it;
- Flow Ar gas through the tube with carbon fiber, to observe the sparking process;

PROJECT STATUS: PLASMA GENERATION

- Development of Coupled Physical-Chemical Simulation Model:

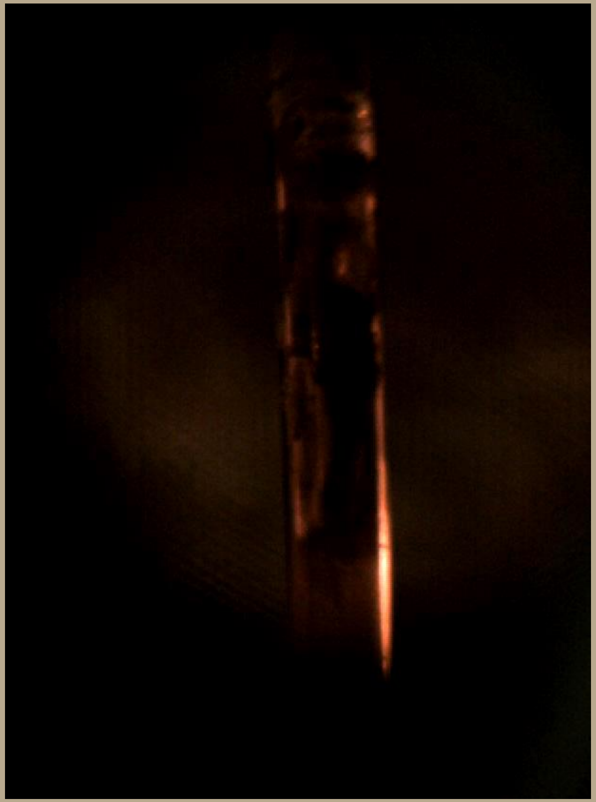
Experimental Results

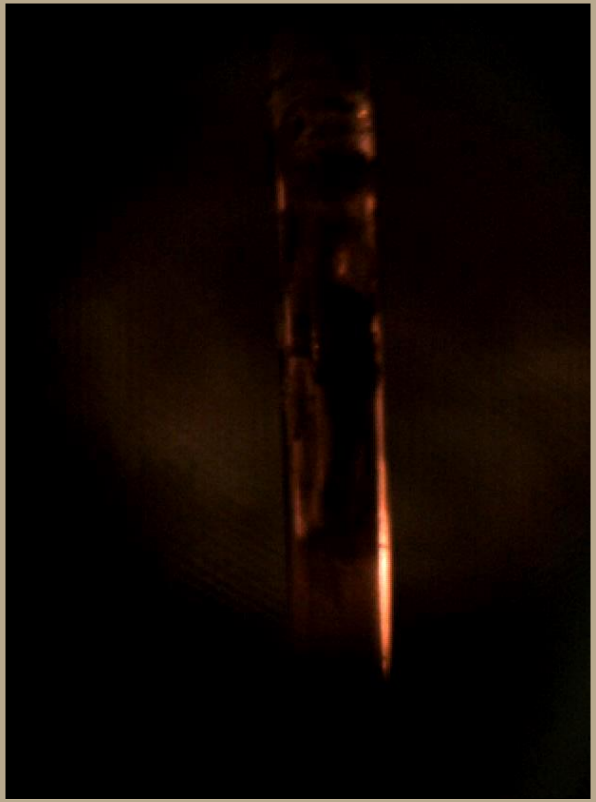
Differential Video

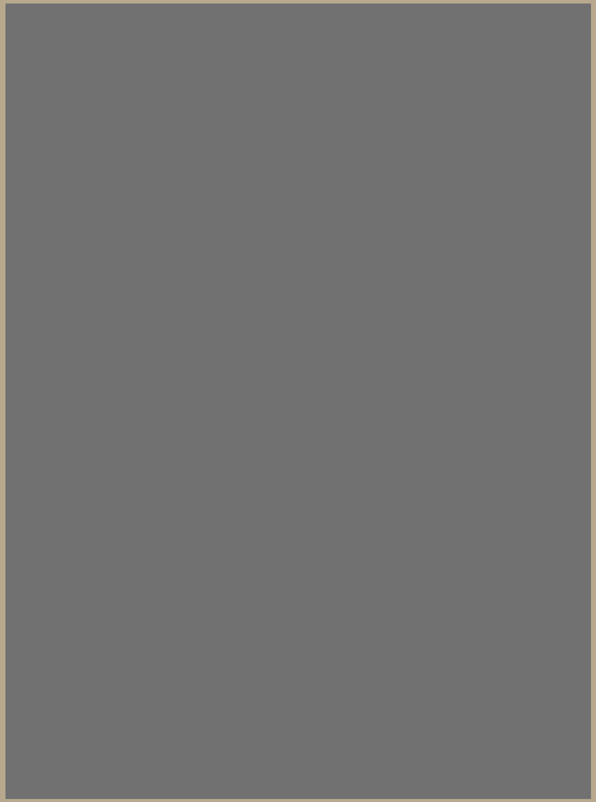
PROJECT STATUS: PLASMA GENERATION

- Development of Coupled Physical-Chemical Simulation Model:

Experimental Results



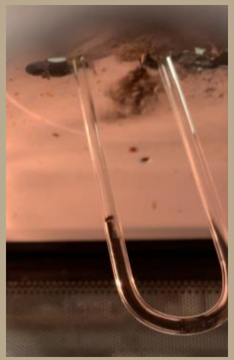




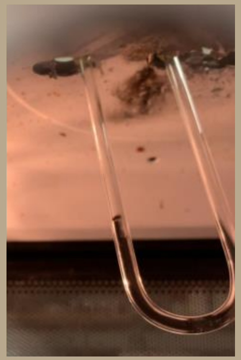
PROJECT STATUS: METHANE PYROLYSIS

- Development of Coupled Physical-Chemical Simulation Model:

Carbon in Argon

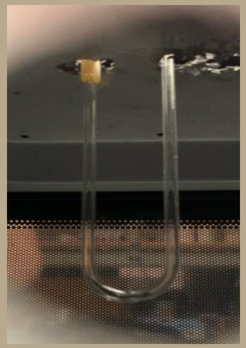


Carbon in Methane

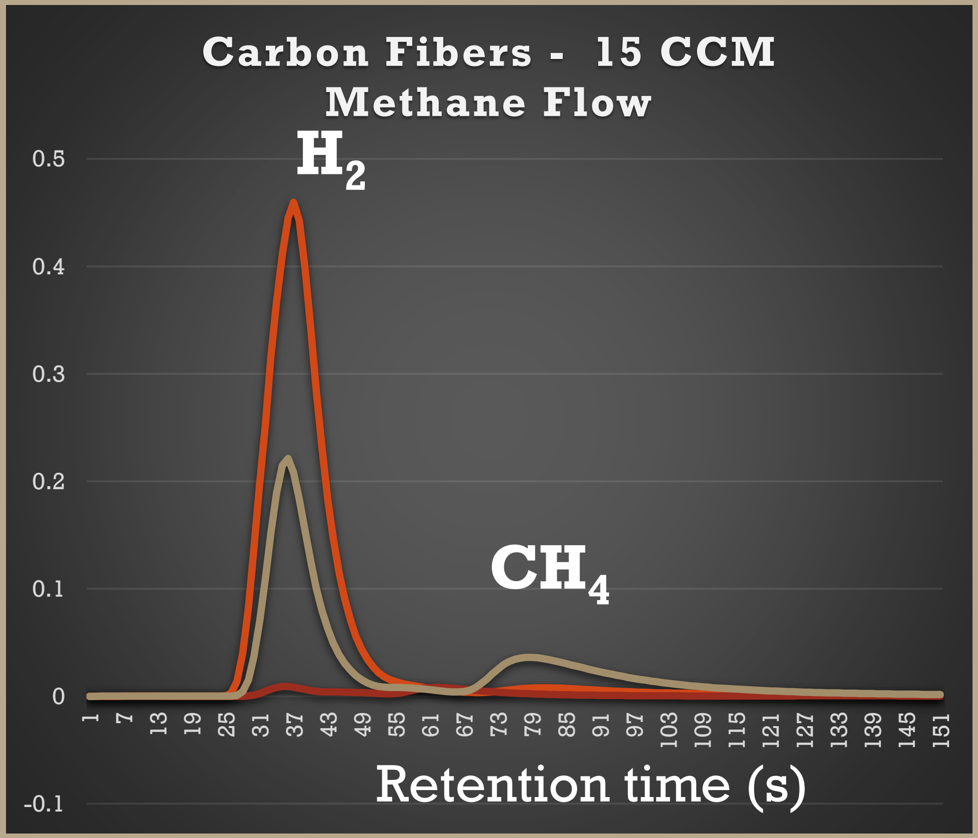


Nickel in Argon

Nickel in Methane



Methane Conversion



STUDENT TRAINING

- **Education and Training:**

  Howard:

  - One Ph.D. student
  - Three undergraduate research assistants

  Houston:

  - One Ph.D. student
  - One high-school student

PROJECT OUTCOMES

- **Publication:**

1. Shi, Y. *et al.* Electrochemical Impedance Imaging on Conductive Surfaces. *Anal. Chem.* **93**, 12320-12328 (2021), published, acknowledgement of federal support: yes.
2. Feng, G. *et al.* Probe the Localized Electrochemical Environment Effects and Electrode Reaction Dynamics for Metal Batteries using In Situ 3D Microscopy. *Adv. Energy Mater.*, 2103484 (2021), published, acknowledgement of federal support: yes.
3. Yan, S. A Continuous-Discontinuous Galerkin Method for Electromagnetic Simulations Based on an All-Frequency Stable Formulation, *Progress In Electromagnetics Research M* **106**, 153-165 (2021), published, acknowledgement of federal support: yes.
4. Xu Yang, Jonathan Koonce, Ying-Chau Liu, Guangxia Feng, Yaping Shi, Xiaoliang Li, Mubeen Syed, Xiaonan Shan. Reflection Optical Imaging to Study Oxygen Evolution Reactions, Journal of the Electrochemical Society, 2022, 169, 057507, published, acknowledgement of federal support: yes.
5. S. Yan and A. O. Idubor, “Parameter identification for symmetrical Prandtl-Ishlinskii hysteresis model using Gauss-Newton method,” in Proc. IEEE Antennas Propag. Symp., Portland, OR, USA, July 2023, published, acknowledgement of federal support: yes.
6. M. Mekonnen and S. Yan, “Comparative investigation of iterative solutions of the all- frequency stable formulation and its vector-potential-only variation,” in Proc. IEEE Antennas Propag. Symp., Portland, OR, USA, July 2023. published, acknowledgement of federal support: yes.
7. M. Mekonnen and S. Yan, “An efficient solution of low-frequency magnetic problems with volt- age sources using all-frequency stable formulation,” in 2023 IEEE MTT-S International Conference on Numerical Electromagnetic and Multiphysics Modeling and Optimization, Winnipeg, Canada, June 2023. published, acknowledgement of federal support: yes.

PROJECT OUTCOMES

- **Publication:**

8. Feng, Guangxia, Hao Jia, Yaping Shi, Xu Yang, Yanliang Liang, Mark H. Engelhard, Ye Zhang et al. "Imaging solid–electrolyte interphase dynamics using operando reflection interference microscopy." Nature nanotechnology 18, no. 7 (2023): 780-789. acknowledgement of federal support: yes.
9. Feng, Guangxia, Yaping Shi, Hao Jia, Samprash Risal, Xu Yang, Paul Ruchhoeft, Wei-Chuan Shih, Zheng Fan, Wu Xu, and Xiaonan Shan. "Progressive and instantaneous nature of lithium nucleation discovered by dynamic and operando imaging." Science Advances 9, no. 21 (2023): eadg6813. acknowledgement of federal support: yes.
10. Tian, Huajun, Guangxia Feng, Qi Wang, Zhao Li, Wei Zhang, Marcos Lucero, Zhenxing Feng et al. "Three-dimensional Zn-based alloys for dendrite-free aqueous Zn battery in dual-cation electrolytes." Nature Communications 13, no. 1 (2022): 7922. acknowledgement of federal support: yes.
11. Khochare, Suraj; Li, Xiaoliang; Yang, Xu; Shi, Yaping; Feng, Guangxia; Ruchhoeft, Paul; Shih, Wei-Chuan ; Shan, Xiaonan, “Functional Plasmonic Microscope: Characterize Metabolic Activity of Single Cells via sub-nm Membrane Fluctuations”, Analytical Chemistry, Accepted. acknowledgement of federal support: yes.
12. M. Mekonnen and S. Yan, “A Novel A-φ Formulation for Efficient Electromagnetic Computations,” in 2024 International Applied Computational Electromagnetics Society (ACES) Symposium, Orlando, FL, USA, May 2024. acknowledgement of federal support: yes.
13. M. Mekonnen and S. Yan, “A simplified all-frequency stable formulation with an implicit Coulomb gauge,” in Proc. IEEE Antennas Propag. Symp., Florence, Italy, July 2024. acknowledgement of federal support: yes.

Thank You!
All Questions Are Welcome!

Multiphysics and Multiscale Simulation Methods for Electromagnetic Energy Assisted Fossil Fuel to Hydrogen Conversion

Su Yan [1], Jiefu Chen [2], and Xiaonan Shan [2]

[1] Howard University, Washington, DC 20059; su.yan@howard.edu

[2] University of Houston, Houston, TX 77204; jchen82@central.uh.edu, xshan@central.uh.edu