Molten Salt Reactor
PROGRAM

Modeling Molten Salt Thermophysical Properties

(and the tools we need)

Vanda Glezakou, *PhD*

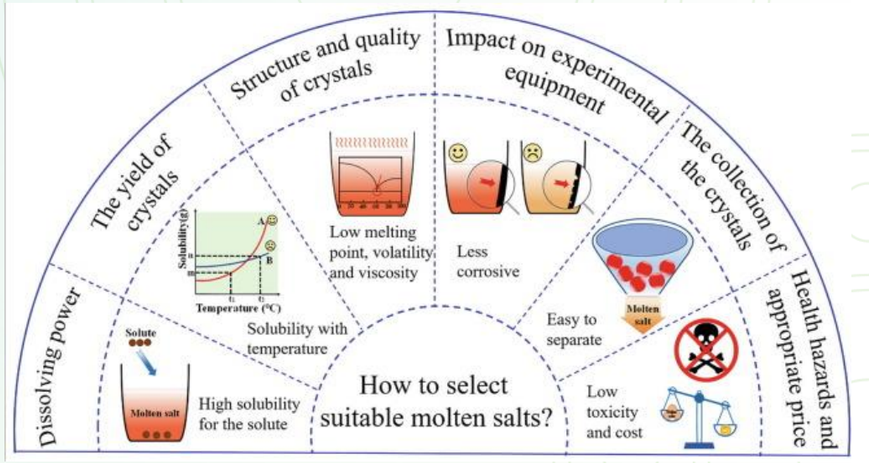
Distinguished Scientist & Section Head of Chemical Transformations

Collaborators: B. A. Smith, A. Birri

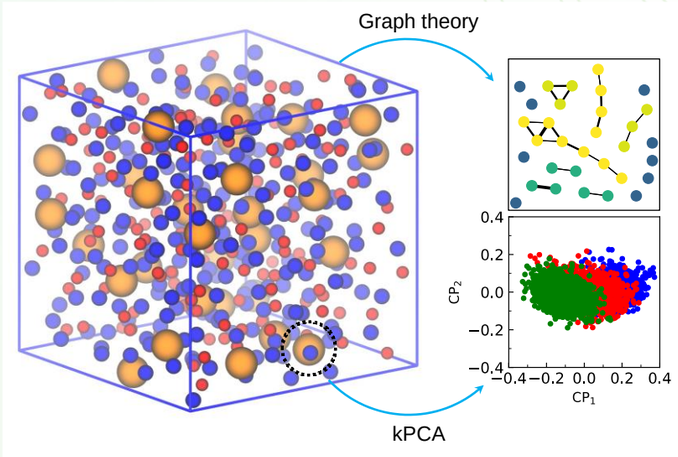
Argonne NATIONAL LABORATORY
INL Idaho National Laboratory
Los Alamos NATIONAL LABORATORY
OAK RIDGE National Laboratory
Pacific Northwest NATIONAL LABORATORY
Sandia National Laboratories
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Atomistic simulations for accurate prediction and validation of thermophysical properties of molten salts

- Why do we need to simulate any properties in molten salts or MSR-relevant systems?
  - Experimental work involving molten salts is expensive and time-consuming; only a few samples can be processed/measured
  - Exp. observations are often affected by underlying chemical reactivity; crucial for proper reactor design, performance and safety
  - Computed properties can be used to:
    - Enhance databases
    - Validate computational models and experimental measurements
    - Understand physico-/chemical-properties
    - Provide a base for physics and chemistry-informed reduced models and AI-based approaches



Liu et al. Mater. Today, 2022 128



Ab initio molecular dynamics (AIMD) allows us to observe the dynamic behavior of systems

Energy calculation (Force Field)

$$E_{pot}\{x_i\}$$

$$F_i = -\partial E_{pot}/\partial x_i$$

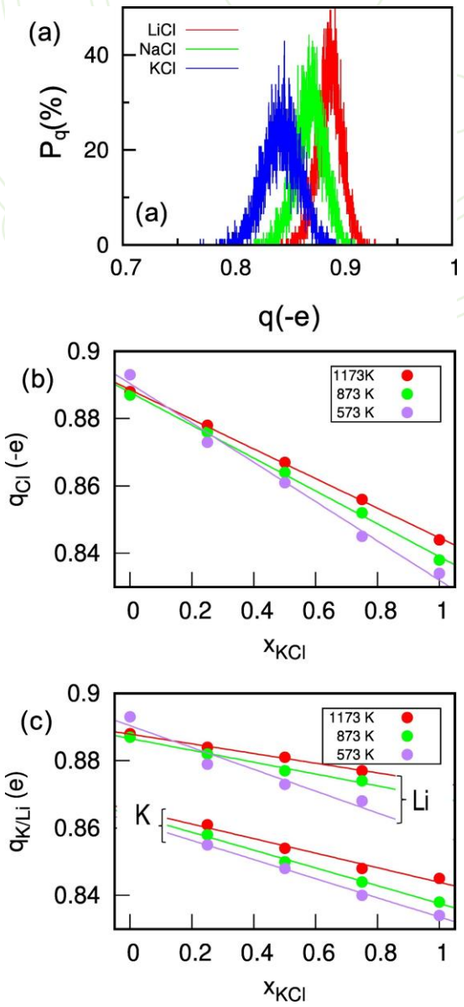
$$a_i = F/m_i$$

$$v_i(t+dt) = v(t)_i + a_i\,dt$$

$$x_i(t+dt) = x(t)_i + v_i\,dt$$

Trajectory (positions, velocities)

Reactivity Properties



Nguyen, M.T., Glezakou, V.A., Lonergan, J., McNamara, B., Paviet, P.D. and Rousseau, R., 2021. Ab initio molecular dynamics assessment of thermodynamic and transport properties in (K, Li) Cl and (K, Na) Cl molten salt mixtures. Journal of Molecular Liquids, 326, p.115262.

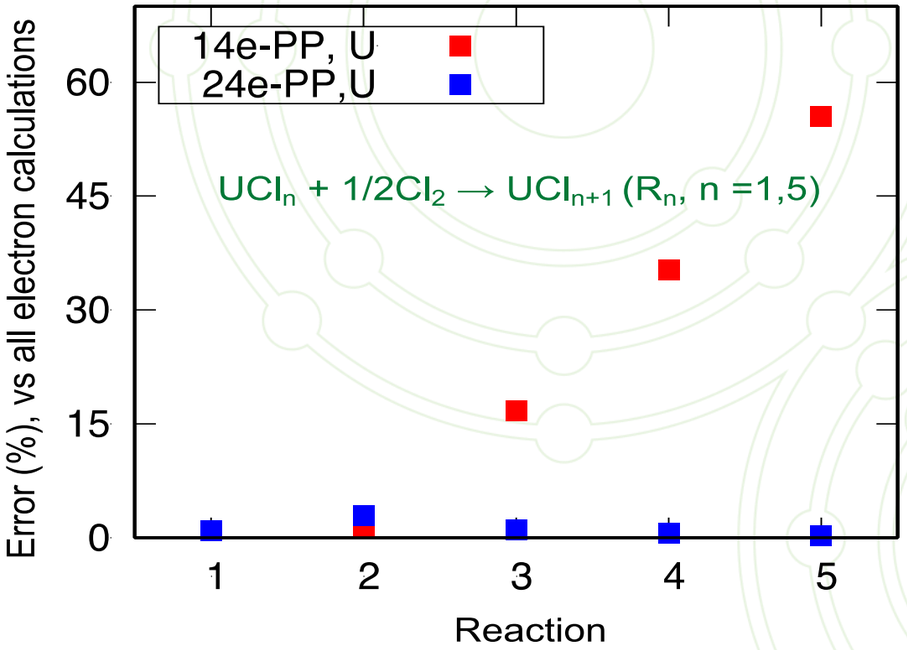
The work on this program benefits from our investment in fundamental sciences

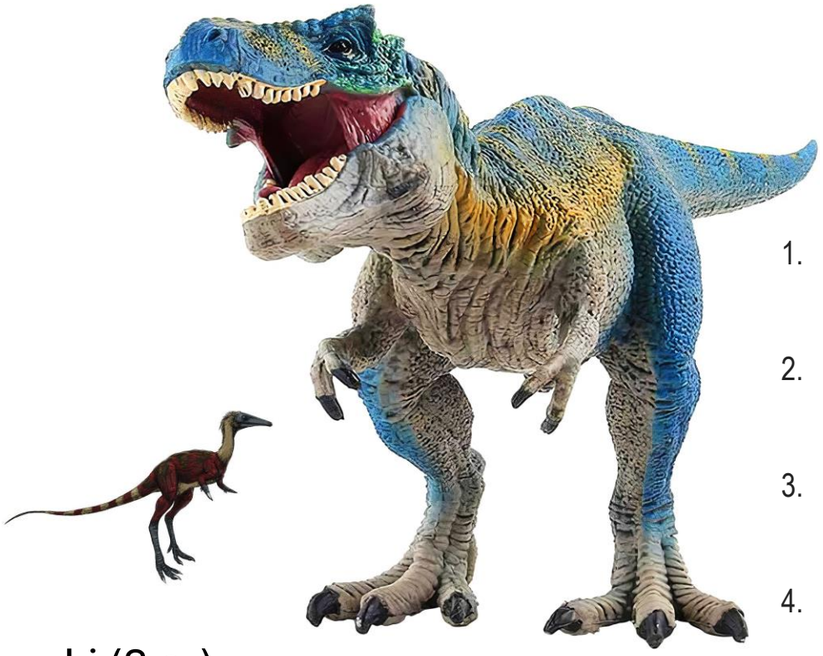
- The accuracy of simulated properties depends on:
  - The underlying complexity of the simulation system
  - Simulation time: the bigger the system, the longer the simulation has to run
  - The mathematical representation of the system, especially when heavy elements are included (relativistic effects)



DFT calculations of heavy elements in condense phase are very challenging and costly

- Large number of electrons
- Highly-contracted *f*-orbitals
- Relativistic effects
- High computational cost





Li (3 e-)

An (89-103 e-)

1. Lu, J.B., Cantu, D.C., Nguyen, M.T., Li, J., Glezakou, V.A. and Rousseau, R., **2019**. Norm-conserving pseudopotentials and basis sets to explore lanthanide chemistry in complex environments. JCTC, 15(11), pp.5987-5997.
2. Shiery, R.C., Fulton, J.L., Balasubramanian, M., Nguyen, M.T., Lu, J.B., Li, J., Rousseau, R., Glezakou, V.A. and Cantu, D.C., **2021**. Coordination sphere of lanthanide aqua ions resolved with ab initio molecular dynamics and X-ray absorption spectroscopy. Inorg. Chem., 60(5), pp.3117-3130.
3. Lu, J.B., Cantu, D.C., Xu, C.Q., Nguyen, M.T., Hu, H.S., Glezakou, V.A., Rousseau, R. and Li, J., **2021**. Norm-conserving pseudopotentials and basis sets to explore actinide chemistry in complex environments. JCTC, 17(6), pp.3360-3371.
4. Nguyen, M.-T, Zhang, J., Cantu, D., Rousseau, R., and Glezakou V.-A. Tailored Computational Approaches to Interrogate Heavy Element Chemistry and Structure in Condensed Phase Rare Earth Elements and Actinides: Progress in Computational Science Applications. , 219-245, DOI:10.1021/bk-2021-1388.ch011



We have developed PP/basis sets for An and Ln series

Goedecker-Teter-Hutter (GTH) pseudopotentials

local

$$V_{loc}^{pp}(r) = -\frac{Z_{ion}}{r}\,\mathrm{erf}\left(\alpha^{PP} r\right) + \sum_i^4 C_i^{pp}\left(\sqrt{2}\alpha^{PP} r\right)^{2i-2} \times \exp\left[-\left(\alpha^{PP} r\right)^2\right]$$

Long-range

Short-range

$$\frac{1}{\sqrt{2} r_{loc}^{pp}}$$

nonlocal

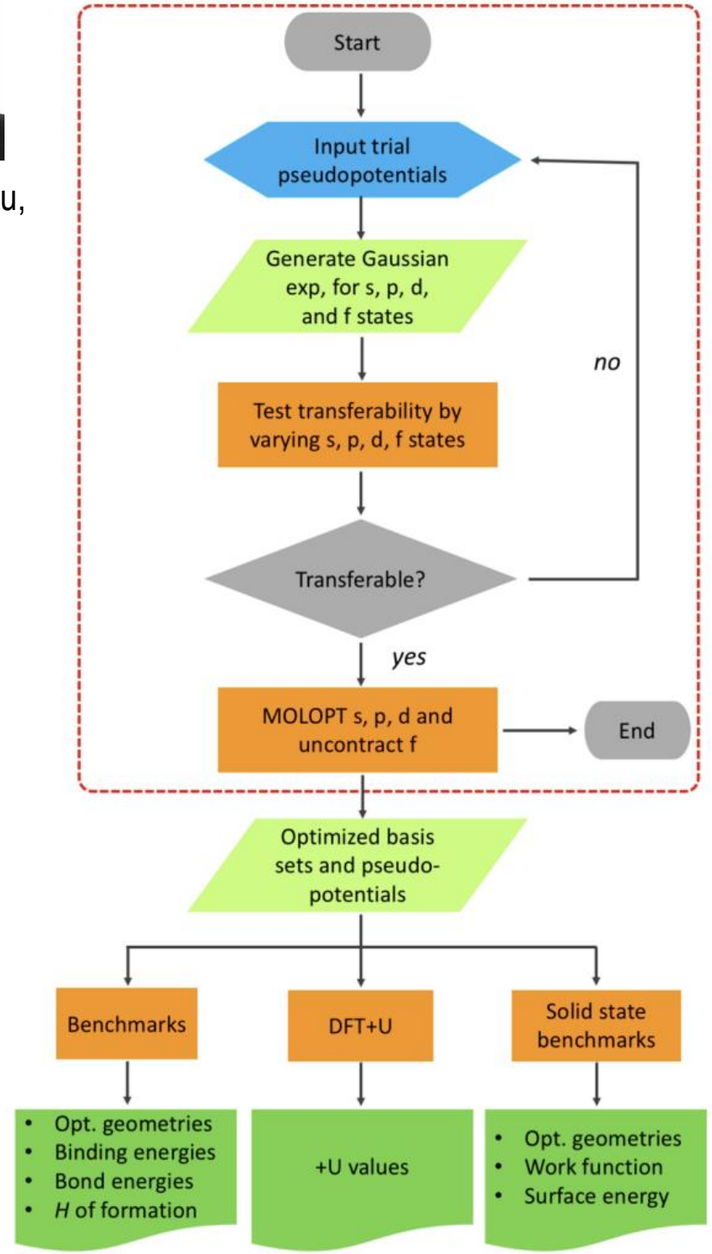
$$V_{nl}^{pp}(\mathbf{r}, \mathbf{r}') = \sum_{lm} \sum_{ij} < \mathbf{r}|p_i^{lm} > h_{ij}^{l} < p_j^{lm}|\mathbf{r}' >$$

Gaussian-type projector

Goedecker et al. PRB (1996); VandeVondele and Hutter, JCP (2007)



R. Rousseau, ORNL



Lu, J.B., Cantu, D.C., Nguyen, M.T., Li, J., Glezakou, V.A. and Rousseau, R., 2019.. JCTC, 15(11), p 5987.
Lu, J.B., Cantu, D.C., Xu, C.Q., Nguyen, M.T., Hu, H.S., Glezakou, V.A., Rousseau, R. and Li, J., 2021, JCTC 17(6), p.3360.

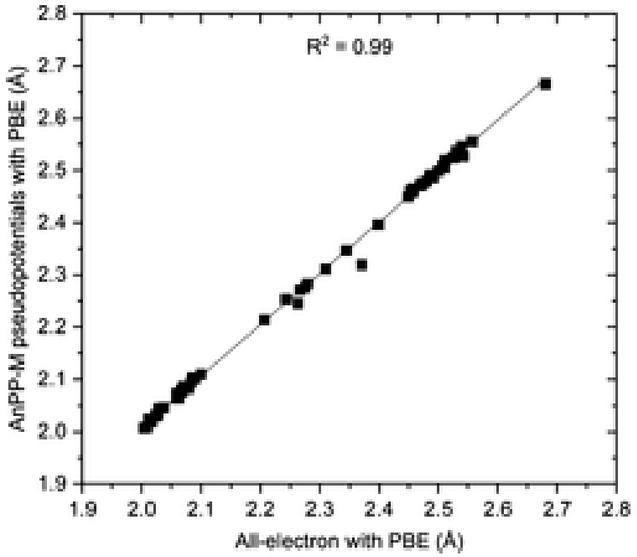
New AnPP-S/M/L PP and companion MOLOPT bases (AnBS-S/M/L)

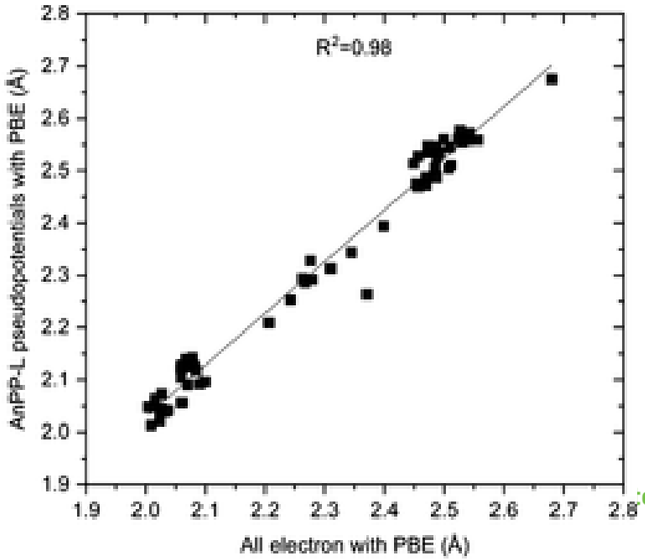
Table 2. Electronic Configurations of Core, Semi-Core, and Valence Regions for Small-Core, Medium-Core, and Large-Core Pseudopotentials for Uranium

[a][Kr] and [Xe] mean the electronic configurations of Kr and Xe atoms.

core type	core[a]	semi-core	valence
small-core (SC)	[Kr]$4d^{10}4f^{14}$	$5s^25p^65d^{10}6s^26p^6$	$5f^36d^17s^2$
medium-core (MC)	[Xe]$4f^{14}$	$5d^{10}6s^26p^6$	$5f^36d^16s^2$
large-core (LC)	[Xe]$4f^{14}5d^{10}$	$6s^26p^6$	$5f^36d^16s^2$

Available in CP2K

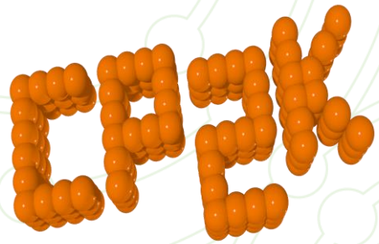




MAD for bond energies ~ 3 kcal/mol for the AnPP/AnBS-M

Computational Details

- AIMD simulations using the CP2K package
  - NVT ensemble
  - Canonical sampling through stochastic velocity scaling thermostat
    - T = 873K
  - Interatomic potentials calculated using density functional theory
    - PBE-D3 density functional
    - Gaussian plane wave hybrid basis set scheme
    - MOLOPT basis sets of double zeta valence polarized quality for all elements
    - Norm-Conserving pseudo-potentials of the GTH type
  - Will Cs/I still adsorb at the same sites (compared to the previous study)
  - These simulations will be used to train ML-based interatomic potentials for larger classical MD simulations



Combined AIMD and machine learning (ML) approaches can help overcome computational costs

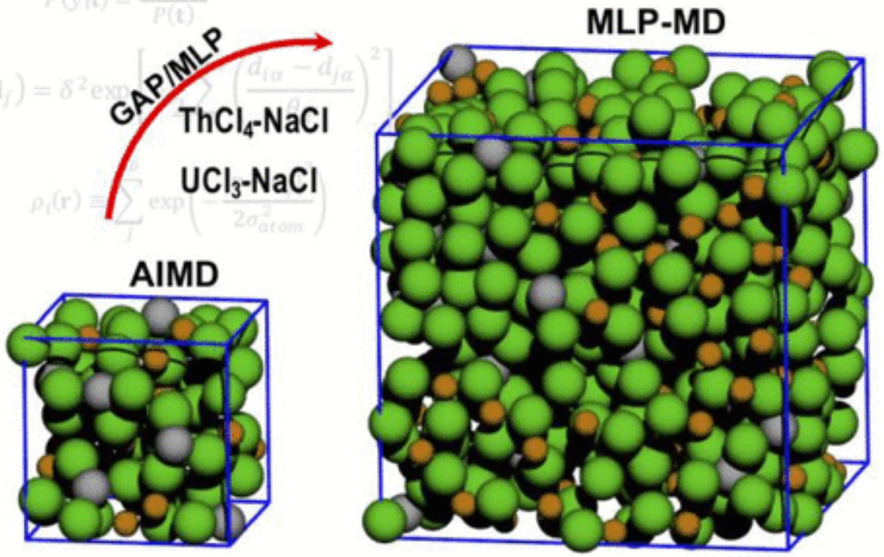


R. Rousseau, ORNL



M.-T. Nguyen (PNNL)

- ML potentials (Gaussian Approximation Potentials - GAPs) are obtained by "training" on the potential energy surfaces generated by AIMD[1, 2]
- ML (GAP) potentials, of DFT accuracy, significantly accelerate MD simulations.
- ML potentials allow to obtain long molecular dynamics trajectories (~ ns) for large systems (~10^3 atoms) [3]

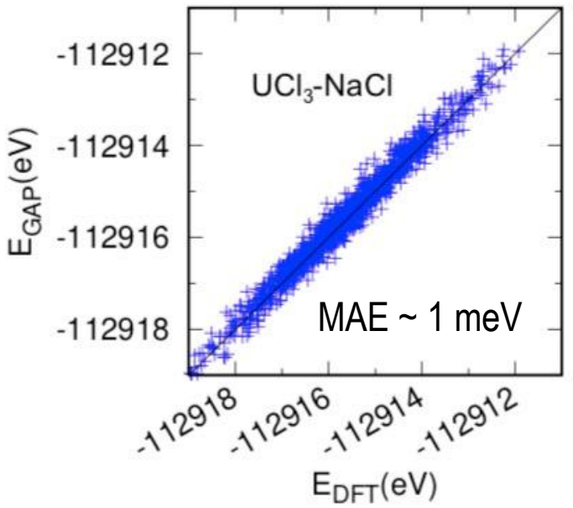


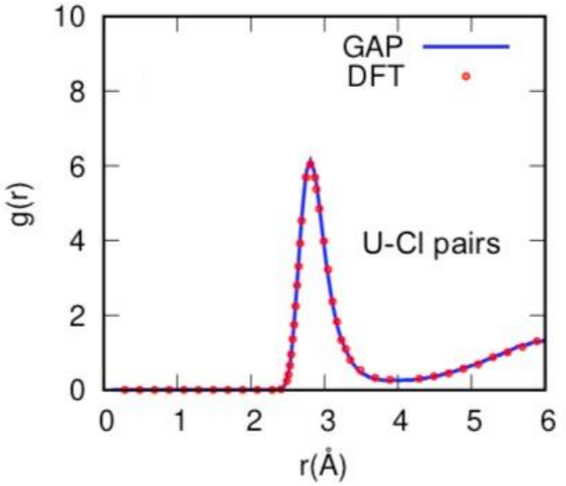
~10-10^2 atoms

~10^3-10^4 atoms

AIMD vs MLP-MD Performance

	AIMD	MLP-MD
Time per MD step / 64 CPUs	~24 hours	~ 1 second





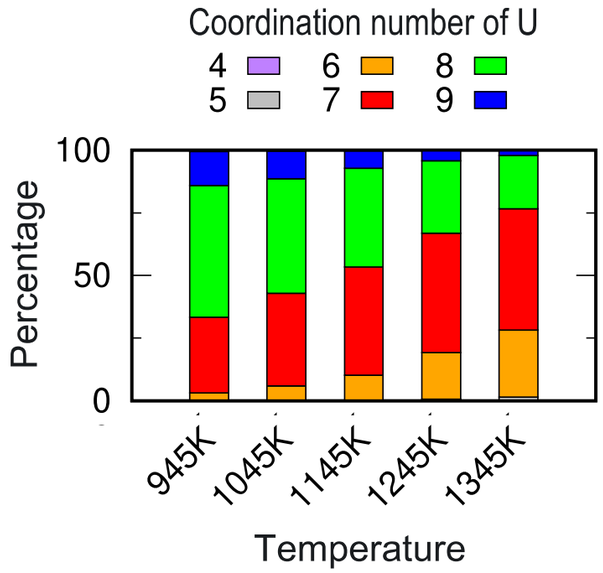
1. Bartok et al. Phys. Rev. Lett.(2010), 104 (13), 136403
2. Berstein et al. https://github.com/libAtoms/QUIP
3. Nguyen et al., ACS Appl. Mater. Interfaces, 2021, https://doi.org/10.1021/acsami.1c11358

MLP-MD is a promising method for the computation of many properties in MS

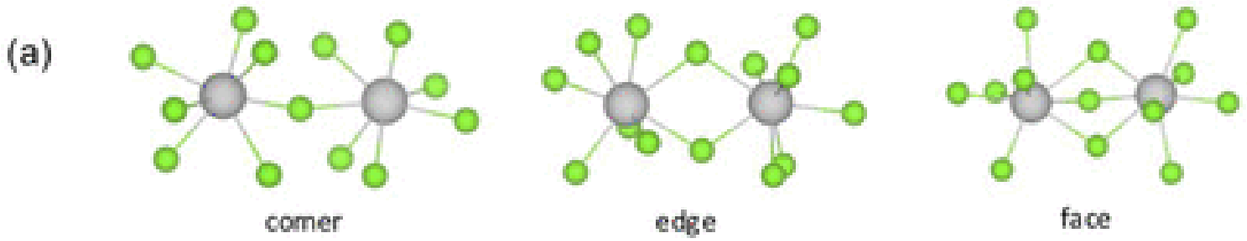
Coordination number CN=7



$(UCl_7)^{4-}$

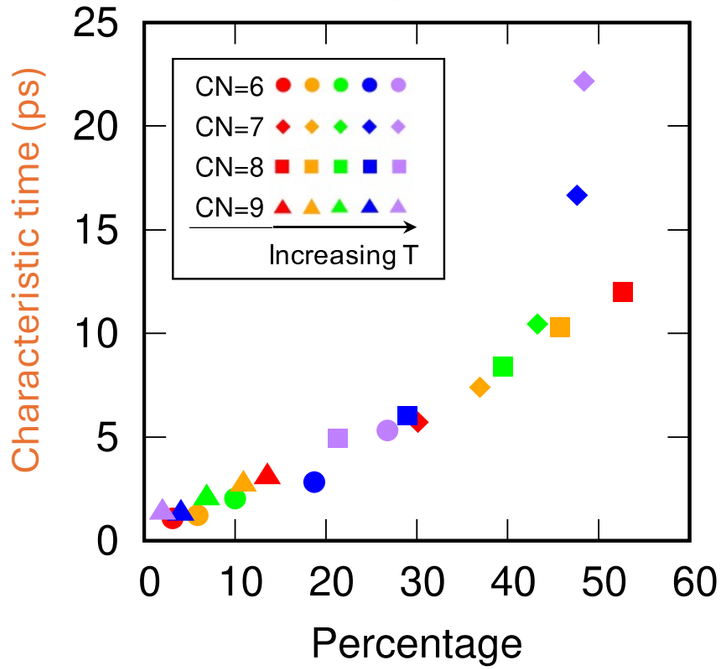


- MLP-MD helps identify speciation in actinide molten salts
  - At about 1000 K, the dominant $[ThCl_7]^{3-}$ is replaced by $[ThCl_6]^{2-}$ and the major $[UCl_8]^{5-}$ population is equal to the $[UCl_7]^{4-}$ population
  - Cation-cation correlations differ between Na-Na and An-An pairs with T
- Characteristic time of ~ 1-20 ps allows for detection using scattering methods.



$ThCl_4$-NaCl and UCl_3-NaCl

UCl_3-NaCl



Nguyen et al., ACS Appl. Mater. Interfaces, 2021

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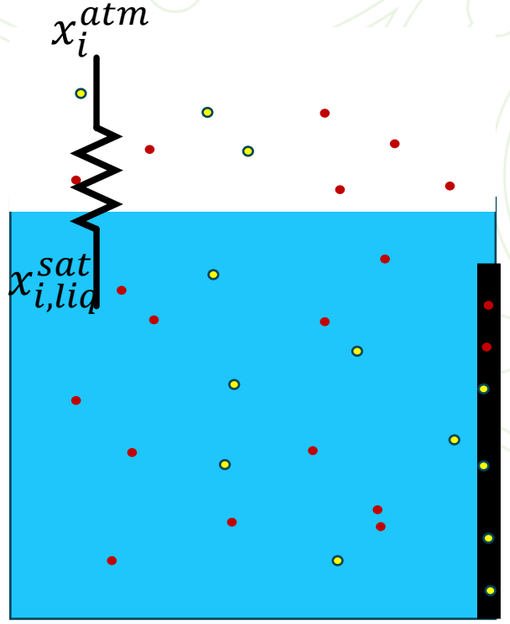
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GRTR – Generalized Radionuclide Transport and Retention



D. Luxat
(Sandia)

- MELCOR: flagship code to assess accidents leading to radiological release at nuclear facilities, used by the U.S. Nuclear Regulatory Commission as well as a range of organizations in the U.S. and over 30 nations. MELCOR is reactor safety technology of global significance.
- Track fission products; how much is released from liquid to atmosphere
- Characterize evolution of fission products between different physico-chemical forms
  - Fission product evolution from a liquid pool to an atmosphere
    - Influenced by solubility and vapor pressure
  - Insoluble fission product deposition on structures
- GRTR mass transport modeling characterizes
  - Concentration of radionuclide forms
  - Concentration gradients between radionuclide forms
  - Resistance to mass transfer between radionuclide forms using standard correlation-based interfacial mass transport theory



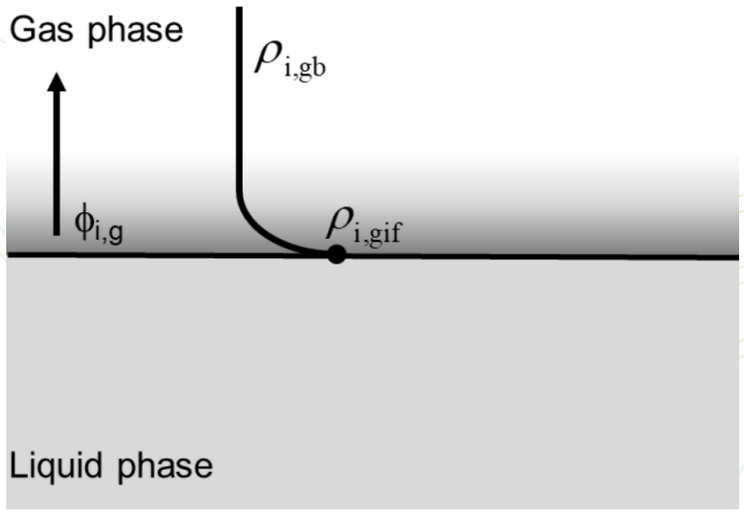
Herranz, L. E.; García, M.; Kissane, M. P. Prog. Nucl. Energy 2012, 54 (1), 138–149.
Serp, J.; et al. Prog. Nucl. Energy 2014, 77, 308–319.

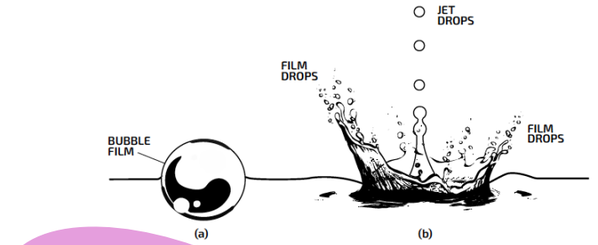
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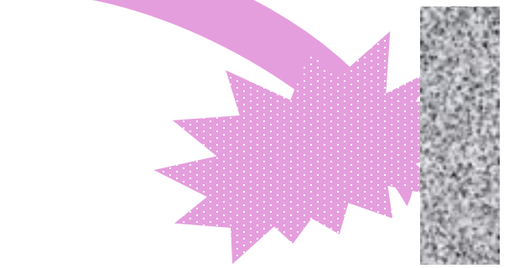
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GRTR – Range of Mass Transport Processes

- Evolution of fission products from molten salts primarily focused on vaporization
  - Provides ability to perform best estimate evaluations of release from molten salts
  - Demonstration calculations have focused on direct comparison to MSRE for the maximum hypothetical accident
  - Exercise of model will be performed next year
- Mass transfer interfaces
  - Liquid-gas atmosphere interfaces
  - Liquid-solid structure interfaces
  - Gas atmosphere-solid structure interfaces
  - Model how new interfaces are defined
- Sparging gas flows (i.e., helium gas injection) will result in fission products entrained in the gas bubble formed by injection
- Jet breakup when contaminated fluids are released into a gas atmosphere (e.g., due to a pipe break)







Lewis, B. J.; Dickson, R.; Iglesias, F. C.; Ducros, G.; Kudo. J. Nucl. Mater. 2008, 380 (1-3), 126–143.
Serp, J.; et al. Prog. Nucl. Energy 2014, 77, 308–319.

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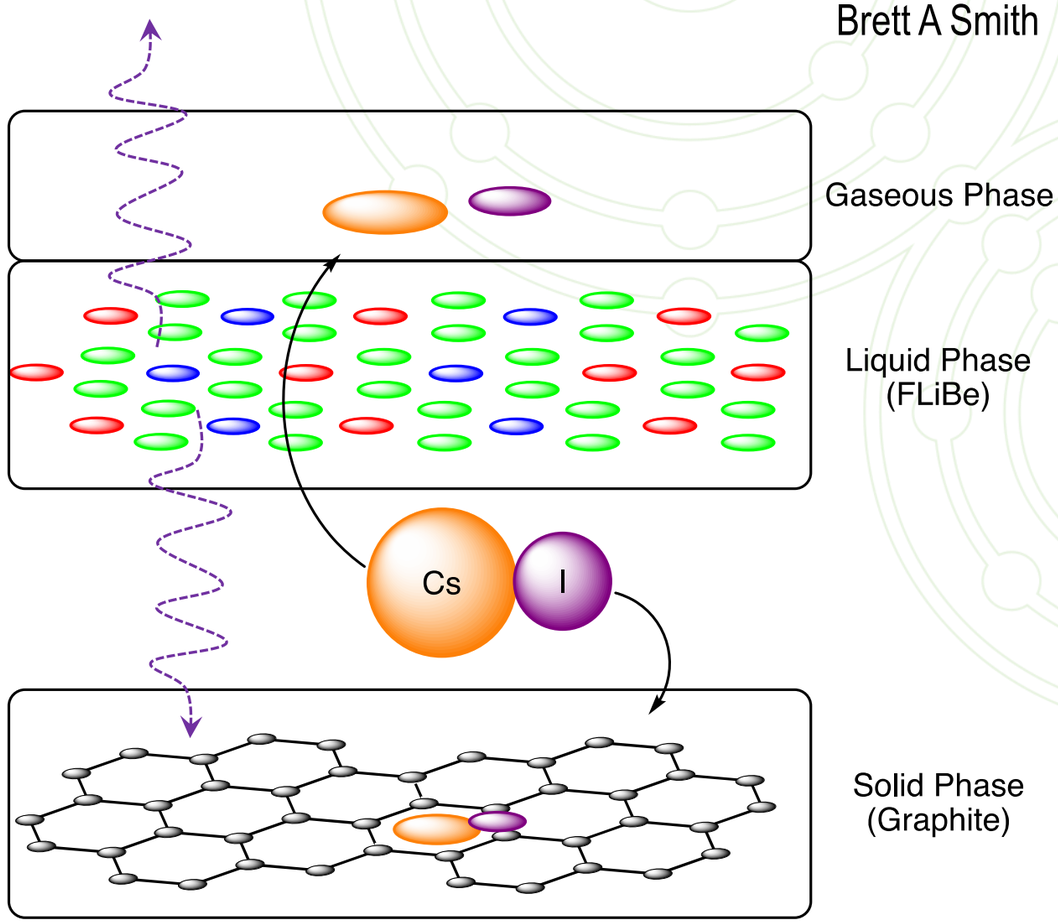
Radionuclide Transport Across MSR Interfaces

ORNL



Brett A Smith

- Mass transport of radionuclides across interfaces in MSR's is critical in evaluation of distribution of radionuclides in MSR's
- Radionuclide deposition in graphite interfaces can deteriorate the interface
- Radionuclide release from MSR operation can pose serious health threats
- Mass transfer across the liquid-gas interface ultimately determined by species diffusivity and gas phase convection



R. J. Kedl, 1972, Report ORNL-TM-3884.
B.F. Myers & W.E. Bell, 1979, Report, GA-A-13990

Cesium and Iodine Adsorption to Graphite

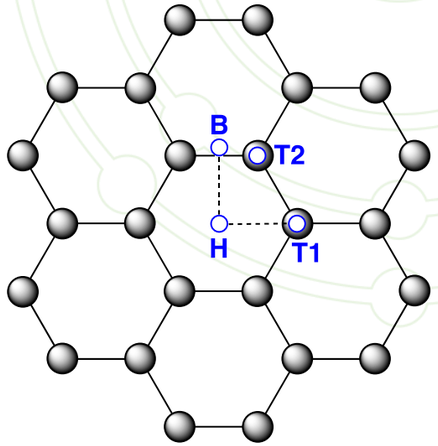
- 4 proposed binding sites: B, H, T1 and T2
- Cs preferential to site H
  - E_{ads} = 1.44 eV
- I preferential to site B
  - E_{ads} = 0.54 eV
- Calculated using density functional theory
  - Projector augmented wave (PAW) pseudopotential
  - Generalized gradient approximation (GGA)
  - 6x6x1 k-point grid

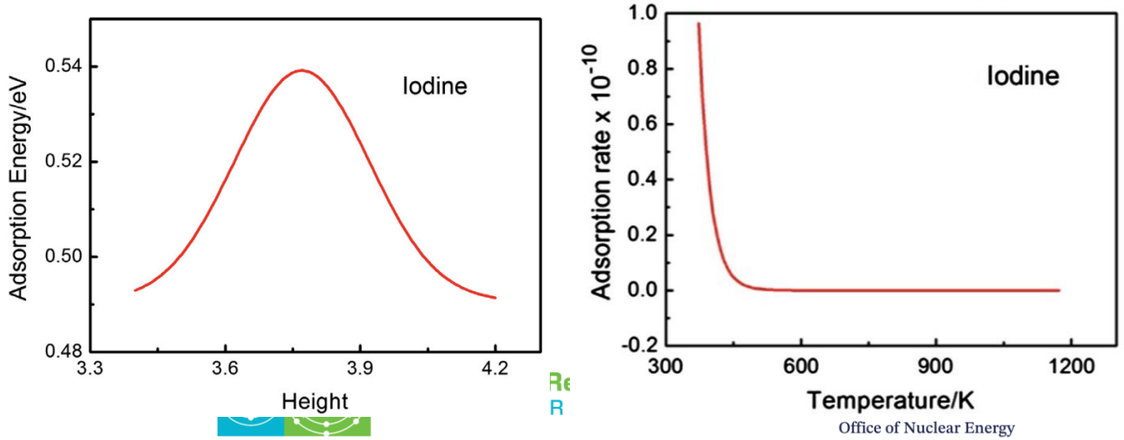


B. Smith (ORNL)



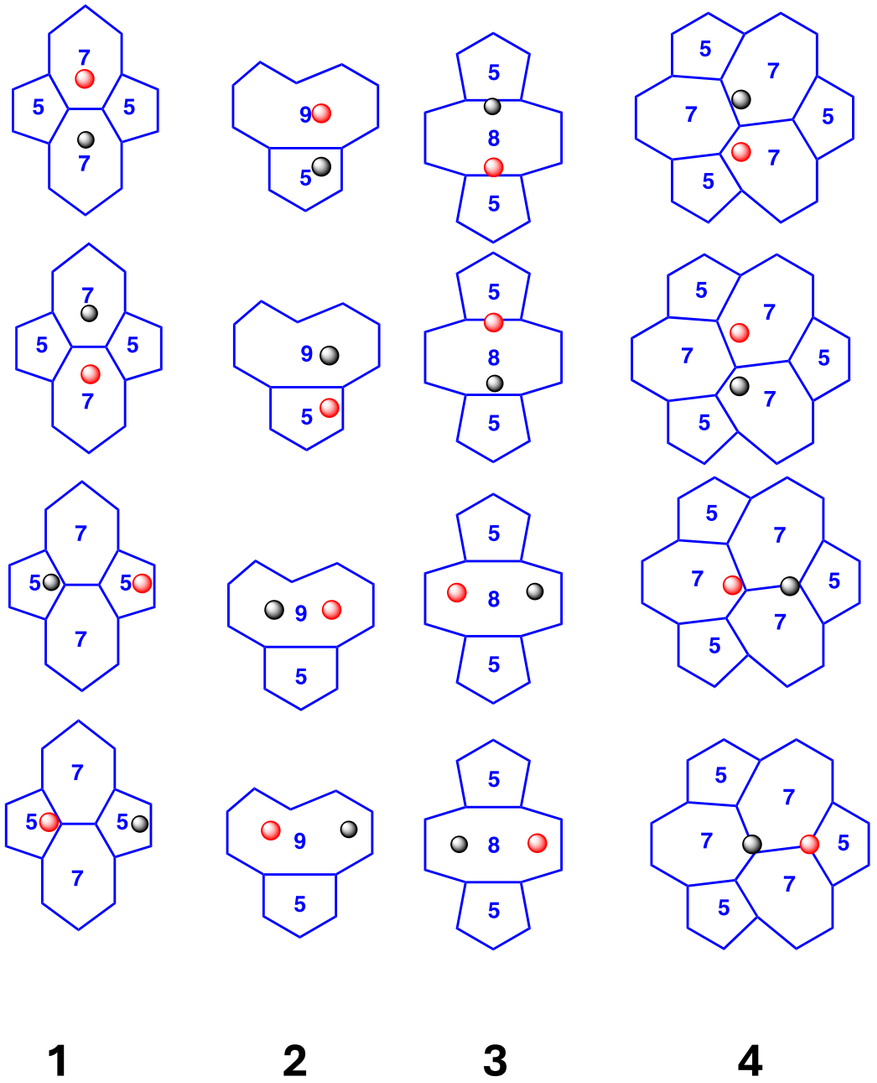
D. Luxat (Sandia)

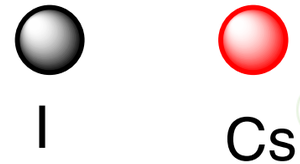


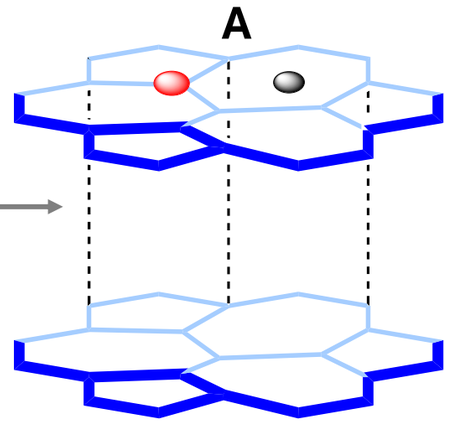


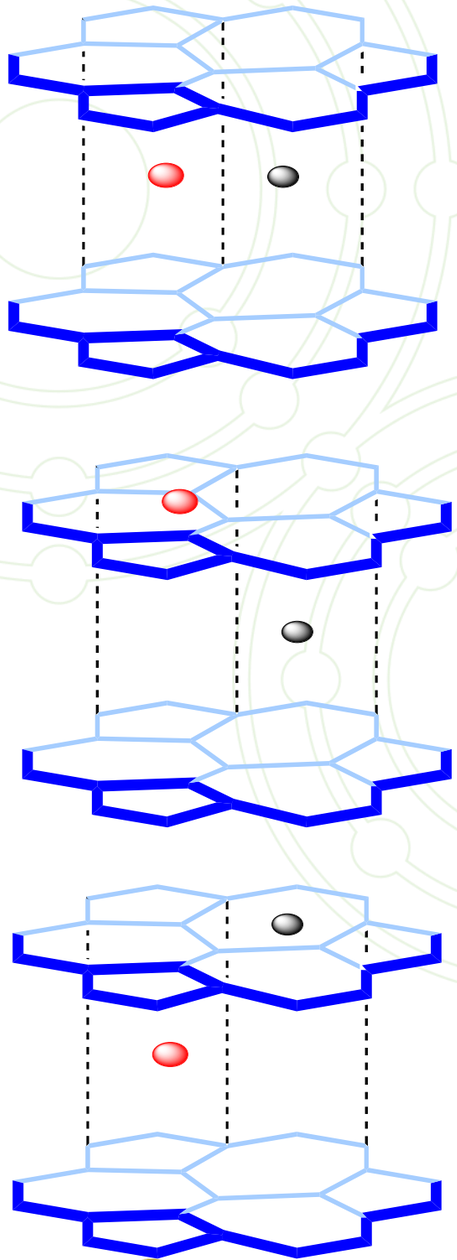
X. Luo, C. Fang, X. Li, W. Lai, T. Liang, *J. Nucl. Mater.* **2013** 441 (1-3), 113-118

Defect Exploration







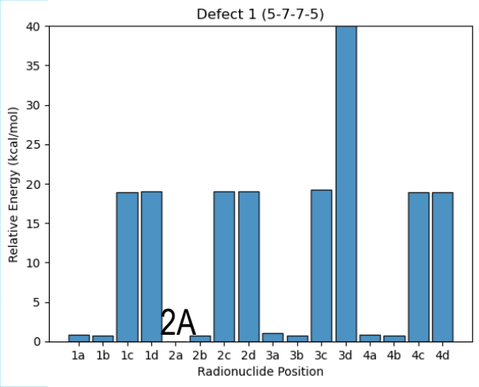


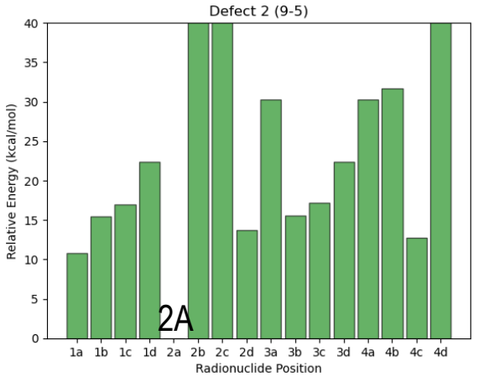
- 4 simple arrangements per defect
- 4 configurations per arrangement
- 16 Structures per defect
- 64 optimizations

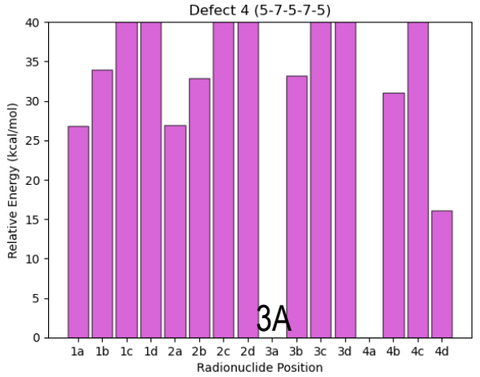
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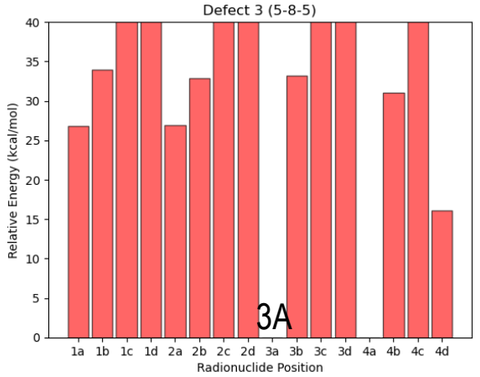
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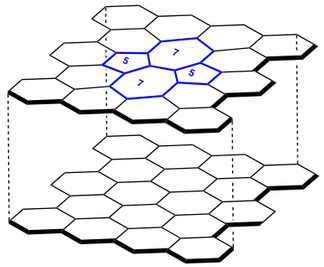
Thermophysical Properties of Caesium Iodide on Bi-Layer Graphene Defects

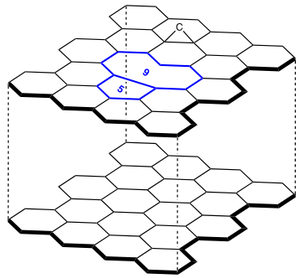


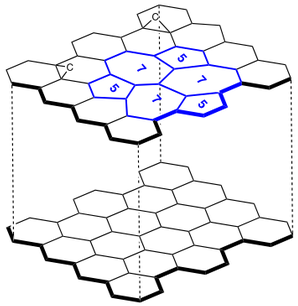


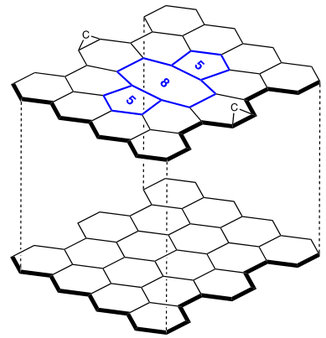


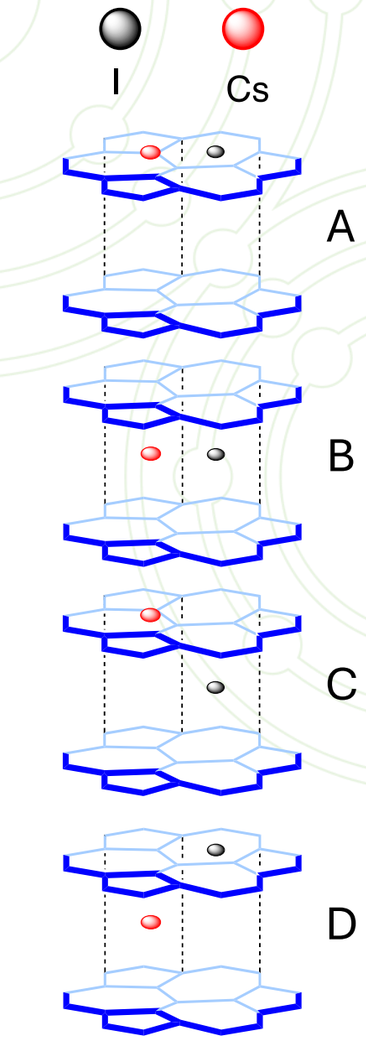












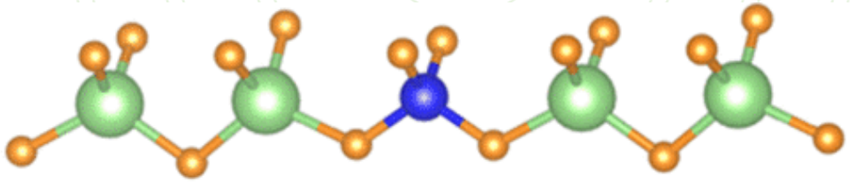
- Screening of bilayer-graphene defects with different arrangements Cs-I has shown preferences for Cs-I atoms in the same plane (configurations A and B)
- Currently testing how the addition of FLiBe Salt above graphene interface effects the energetics and transportation properties of this system.

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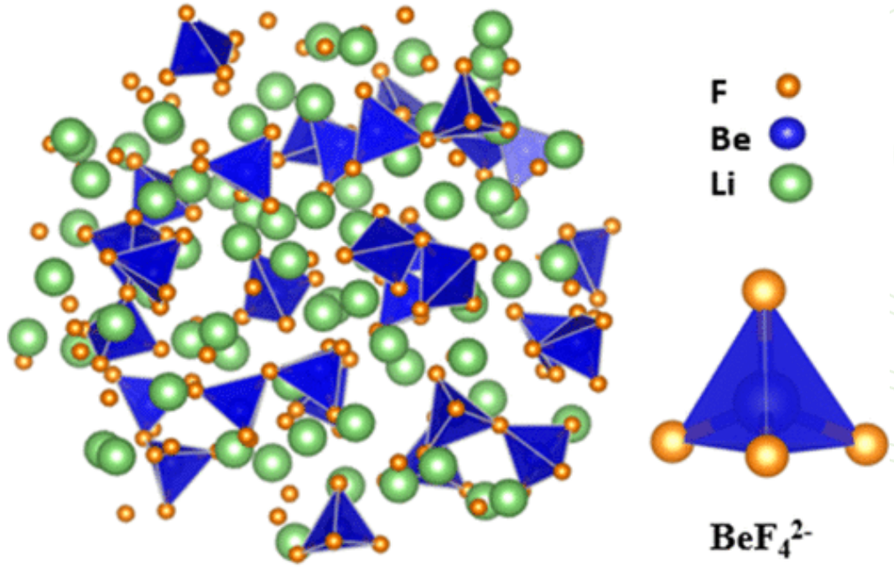
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Lithium Tetrafluoroberyllate (FLiBe)

- FLiBe has been extensively used MSR experiments as both a coolant and solvent for fissile material
  - Neutron transparency
  - High thermal efficiency
  - Low spent fuel per unit energy
  - Favorable thermophysical properties
- FLiBe has exhibited limited mass transport across graphite interfaces (avoid fluorination of graphite)
  - FLiNaK is often surrogated for FLiBe to avoid the toxicity of Be



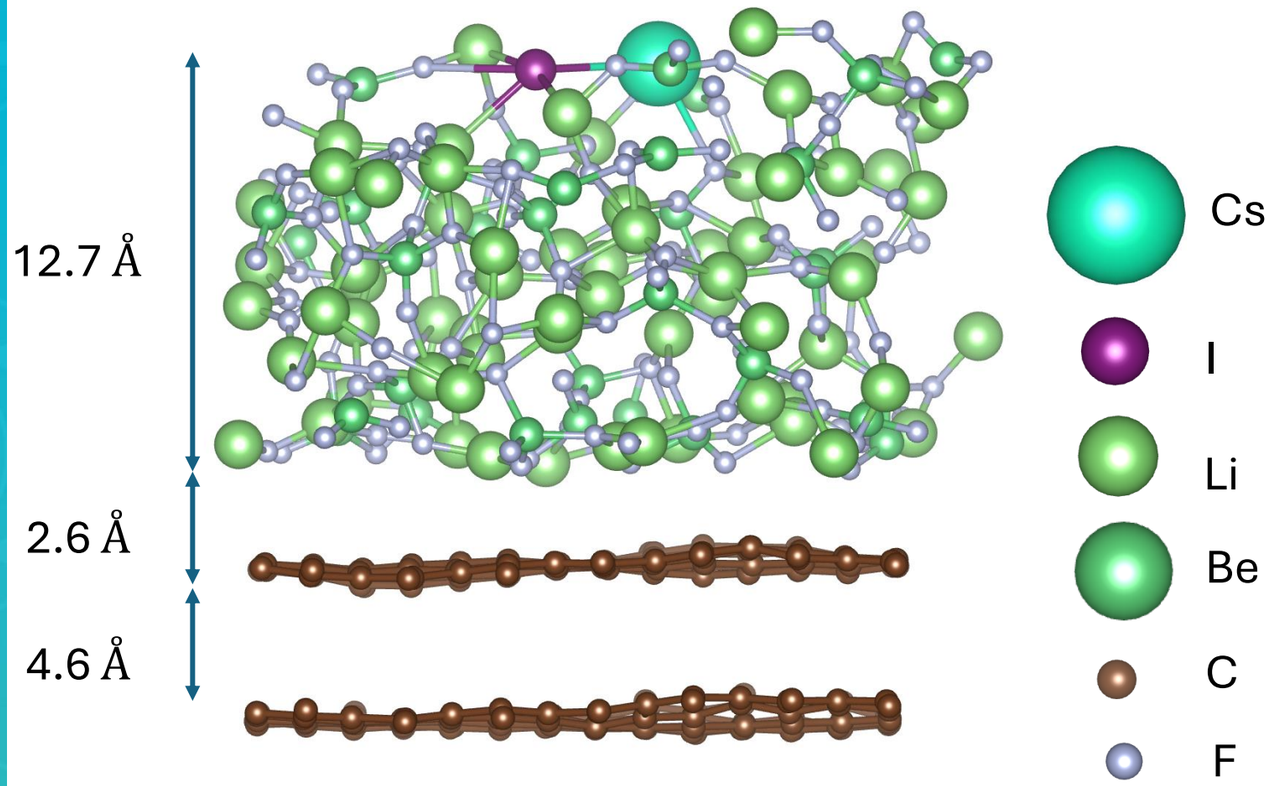
Chain of tetrahedra in the crystalline FLiBe



Amorphous FLiBe

Baral,K.; San, S.; Sakidja, R.; Couet, A.; Sridharan, K.; Ching, W.Y. ACS Omega 2021 6 (30), 19822-19835
Sprouster, D.; Zheng, G.; Lee, S.-C.; Olds, D.; Agca, C.; McFarlane, J.; Z, Y.; Khaykovich, B. ACS Appl. Energy Mater. 2022, 5 (7), 8067–8074.
Vergari, L.; Nelson, M.; Droster, A.; Contescu, C.; Gallego, N.; Scarlat, R. O. J. Nucl. Mater. 2022, 572, 154058.

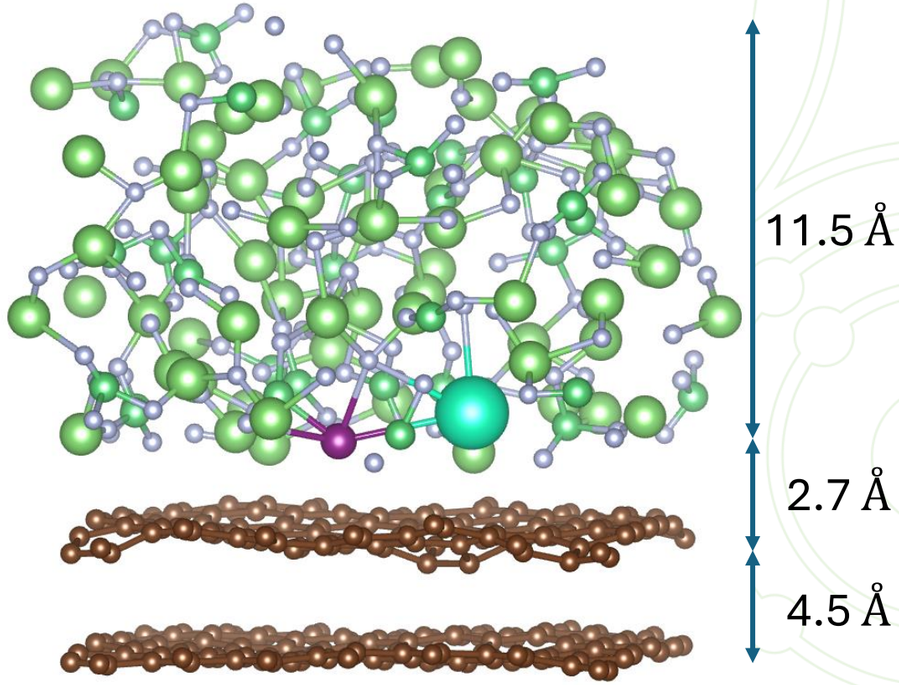
FLiBe (LiF-BeF_2)- CsI



“Top” configuration

Element	Be-F	Li-F	Cs-F
Coordination Number	4.0	4.57	3.74
Cutoff (Å)	2.85	2.45	3.25

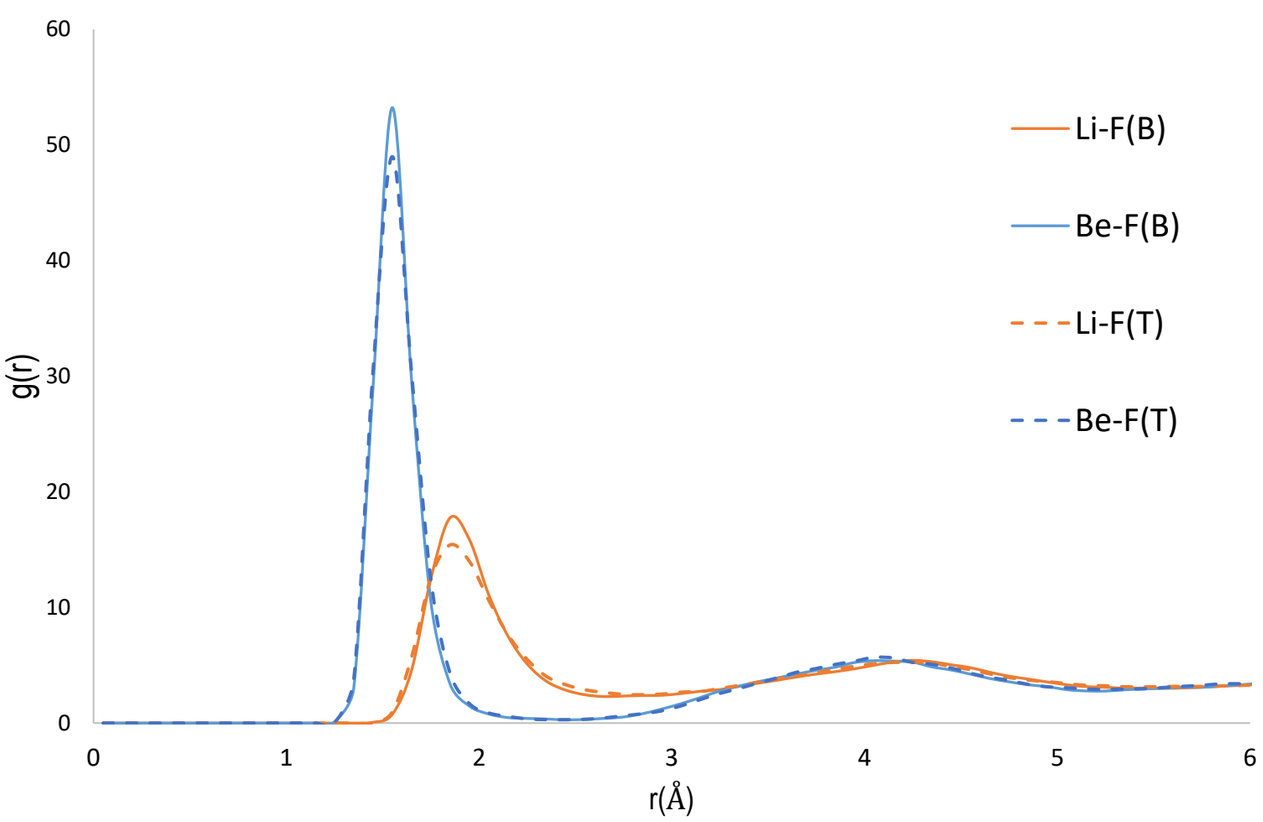
Typical CsI bonding ~3.9 Å

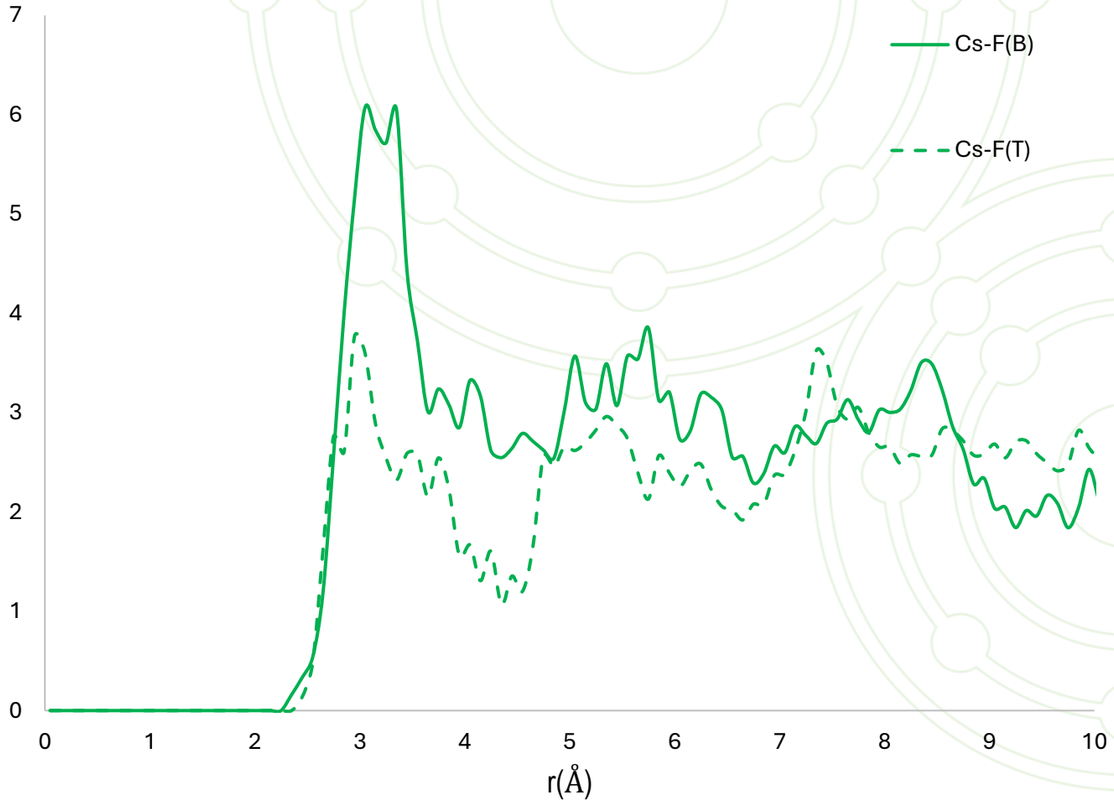


“Bottom” configuration

Element	Be-F	Li-F	Cs-F
Coordination Number	3.97	3.84	4.73
Cutoff (Å)	2.85	2.45	3.35

FLiBe-CsI Pair Distribution Function





- Be-F interactions are stronger (not much exchange of ions); well-defined solvation structure
- Li-F pairs are more transient; F ion exchange between 1st-/2nd solvation shell

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Modeling of viscosity

ORNL



T. Birri and team



B. A. Smith

PNNL



M.-T. Nguyen

- Expand on previous simulations work for viscosity calculations [1]
- Contribute to database for thermodynamic properties, benchmark AIMD models
- Expand on ML methods to extend simulation times and robust interatomic potentials with AIMD accuracy [2]
- Directly compare with upcoming measurements by Birri and team

Mix #	System	Temperature Range for Measurements (deg C)
1	NaCl-KCl (0.75-0.25)	750-825
2 ✓	NaCl-KCl (0.50-0.50)	675-825
3 ✓	NaCl-KCl (0.25-0.75)	750-825
4 ✓	NaCl-LiCl (0.75-0.25)	750-825
5	NaCl-LiCl (0.50-0.50)	650-800
6	NaCl-LiCl (0.25-0.75)	600-800
7 ✓	KCl-LiCl (0.75-0.25)	550-800
8 ✓	KCl-LiCl (0.50-0.50)	500-800
9 ✓	KCl-LiCl (0.25-0.75)	550-800
10 ✓	NaCl-KCl-LiCl (eutectic)	400-700
11 ✓	NaCl-KCl-LiCl (0.333-0.333-0.334)	600-800
12 ✓	NaCl-KCl-LiCl (0.20-0.40-0.40)	500-800
13 ✓	NaCl-KCl-LiCl (0.20-0.20-0.60)	525-800

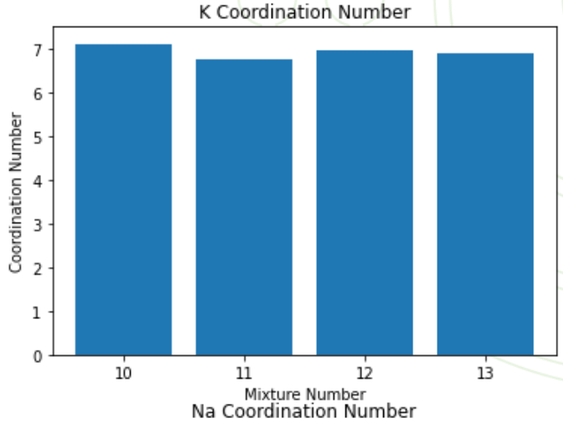
1. Nguyen, M.T., Glezakou, V.A., Lonergan, J., McNamara, B., Paviet, P.D. and Rousseau, R., 2021. Ab initio molecular dynamics assessment of thermodynamic and transport properties in (K, Li) Cl and (K, Na) Cl molten salt mixtures. Journal of Molecular Liquids, 326, p.115262.
2. Nguyen, M.T., Rousseau, R., Paviet, P.D. and Glezakou, V.A., 2021. Actinide molten salts: A machine-learning potential molecular dynamics study. ACS Applied Materials & Interfaces, 13(45), pp.53398-53408.

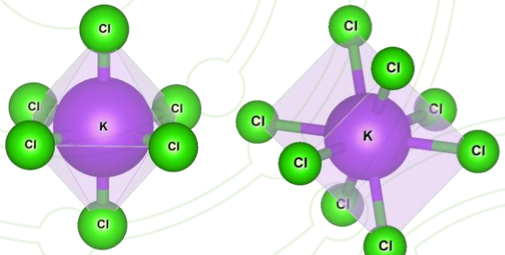
Elucidating Transport Properties of Pseudo-Ternary Molten Chloride Salts Using AIMD Simulations

Mix #	χ_{NaCl}	χ_{KCl}	χ_{LiCl}	D_{Na}	D_K	D_{Li}	D_{Cl}	D_{Mix}	η_{mix}	R_H
10	0.075	0.375	0.550	3.32	6.57	7.96	5.07	6.09	1.08	3.24
11	0.333	0.333	0.334	6.09	6.73	7.95	5.59	6.16	1.15	3.22
12	0.200	0.400	0.400	5.39	4.74	6.41	4.18	5.05	1.35	3.24
13	0.200	0.200	0.600	9.01	10.60	12.61	8.46	9.88	0.82	2.80

Diffusion coefficients (D_i where i is Na, Li, K, Cl, or mix) given in units of 10^{-5} cm^2/s, viscosity (η) is given in units of cP and the hydrodynamic radius is given in angstroms.

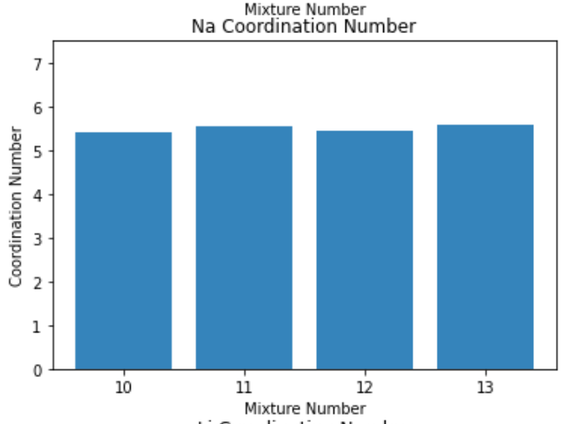
- Self Diffusion coefficients appear to decrease with the conc. of KCl, and increase with concentration of LiCl (coordination number holds the inverse of that trend)
  - Ionic radius of Li+ (73pm) is considerably smaller than K+ (152pm). Hydrodynamic radius holds the same trend.
  - K+ likely diffuses as an atomic species whereas Li+ is more likely to retain bonding to Cl- ions.
- A large majority of Na+ and K+ exhibit octahedron shapes where as Li+ exhibits more distorted tetrahedron.

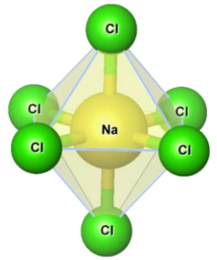




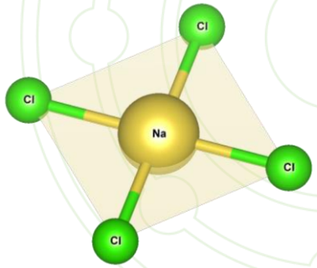
CN=6

CN=8

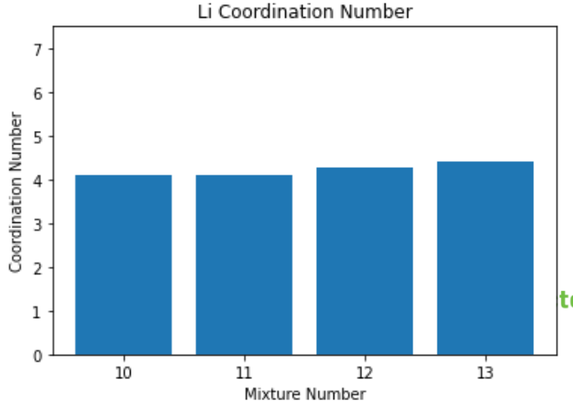


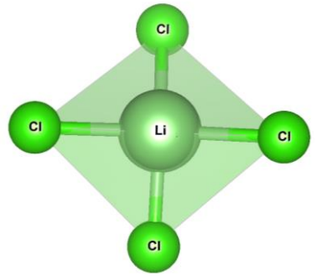


CN=6



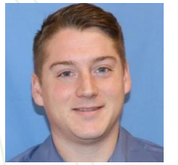
CN=4





CN=4

Thermophysical and transport properties on U-Chlorides AIMD/ML-trained interatomic potentials (in progress)



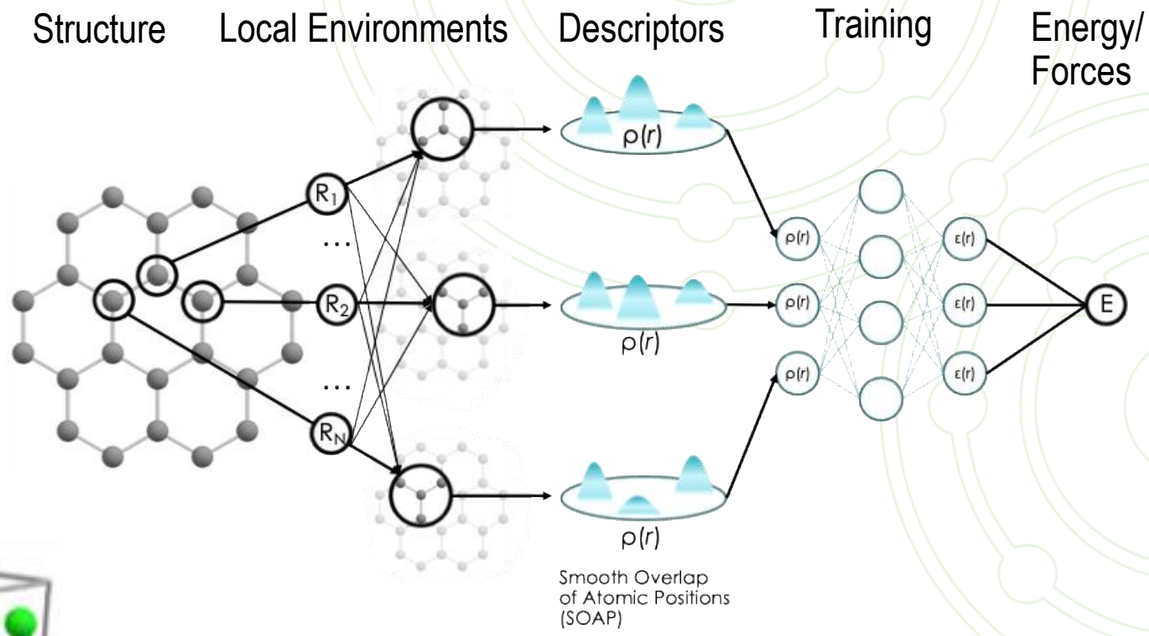
B. A. Smith

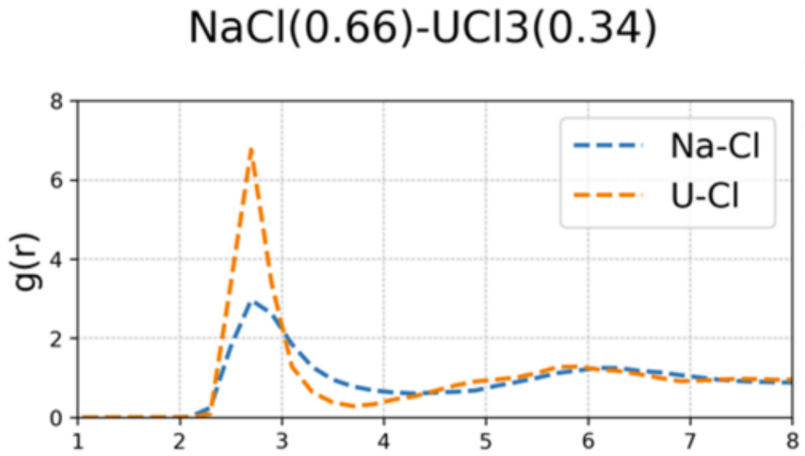
UCl_3-NaCl

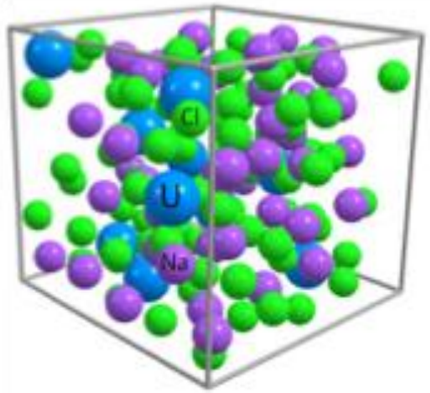
- Well-separated U atoms which diffuse slower than Na/Cl; fairly stable U-Cl associations

D_{Na} (10^{-5} cm^2s^{-1})	D_U (10^{-5} cm^2s^{-1})	D_{Cl} (10^{-5} cm^2s^{-1})
5.08	3.2	5.3

η_{mix}= 2.30 (cP, 987 K); Exp 3.0 (cP, 1025K) (Birri and co-workers);
2.29 (cP, 987K) (Desyatnik et al, 2020)







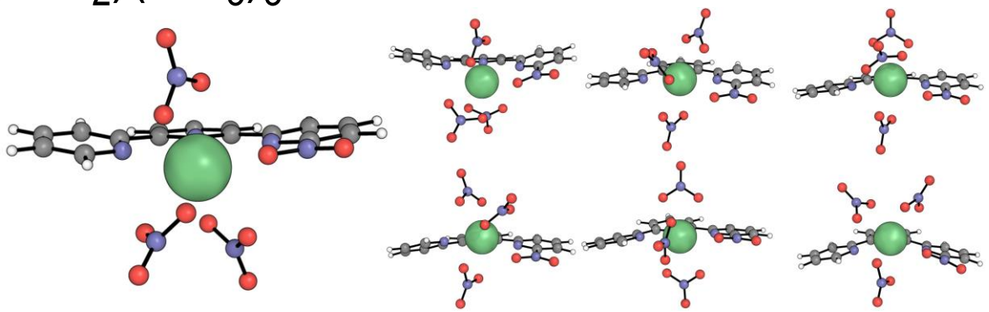
- MLIPs from AIMD to accelerate MD for the CsI/FliBe/graphene and U-based Chloride systems

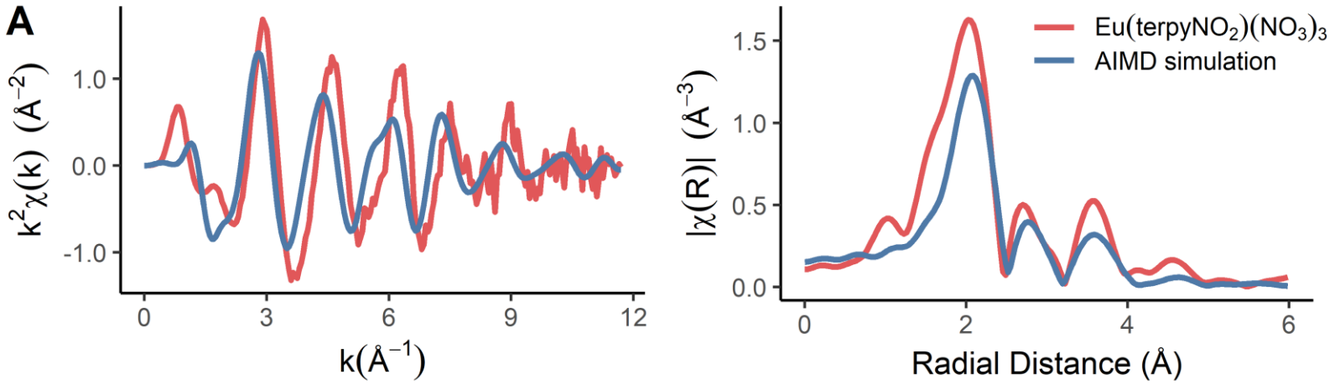
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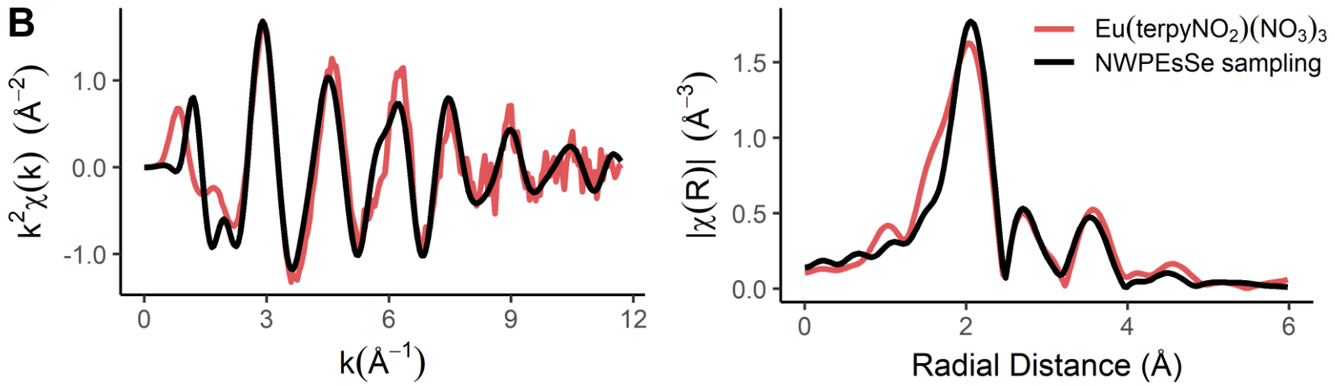
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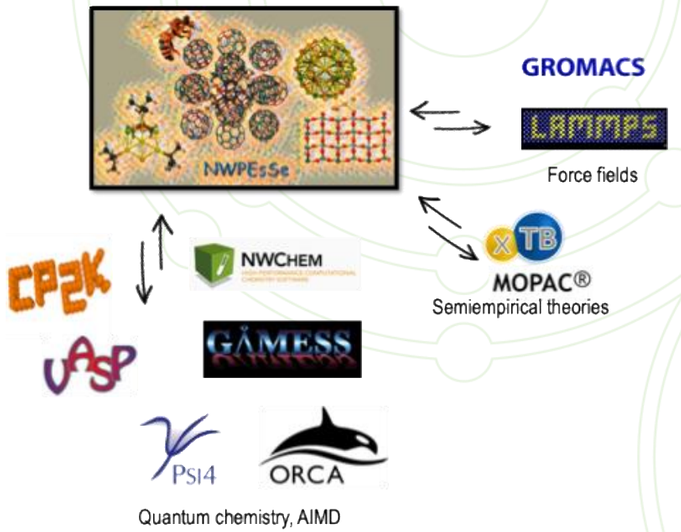
Global Optimization allows efficient configurational sampling for ensemble spectroscopic properties

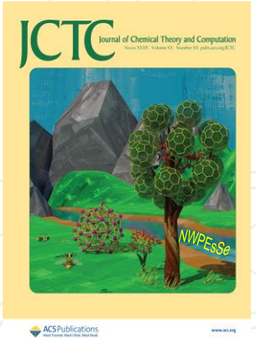
$Eu(terpyNO_2)(NO_3)_3$











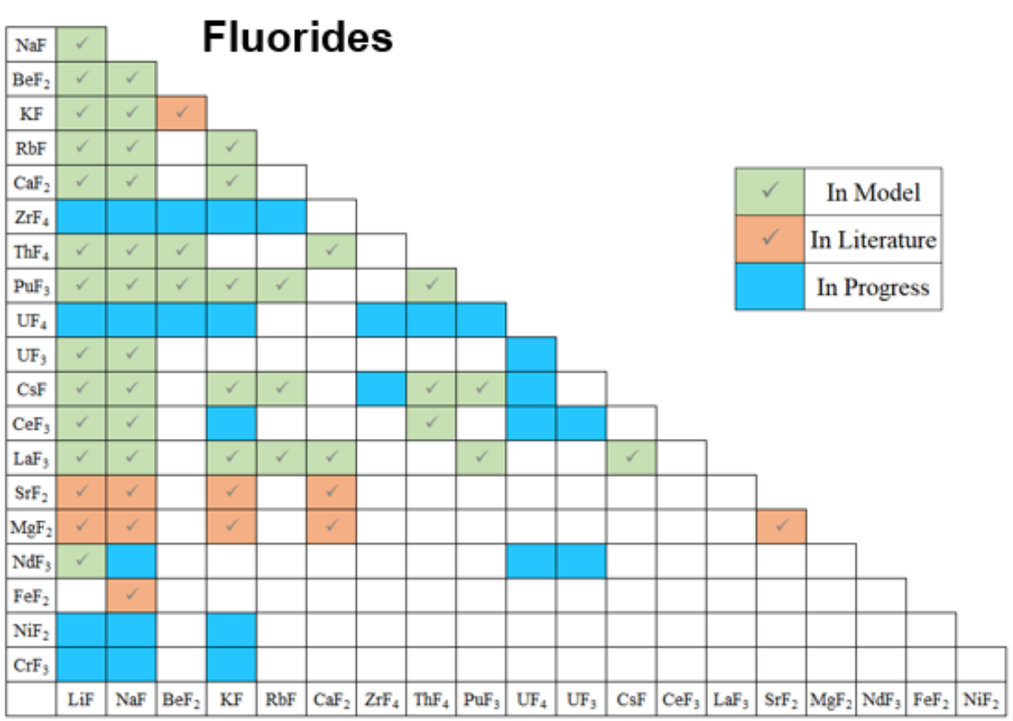
Python version allows for

Summers, T.J., Zhang, D., Sobrinho, J.A., de Bettencourt-Dias, A., Rousseau, R., Glezakou, V.A. and Cantu, D.C., 2024. Pairing a global optimization algorithm with EXAFS to characterize lanthanide structure in solution. Journal of Chemical Information and Modeling, 64(23), pp.8926-8936.

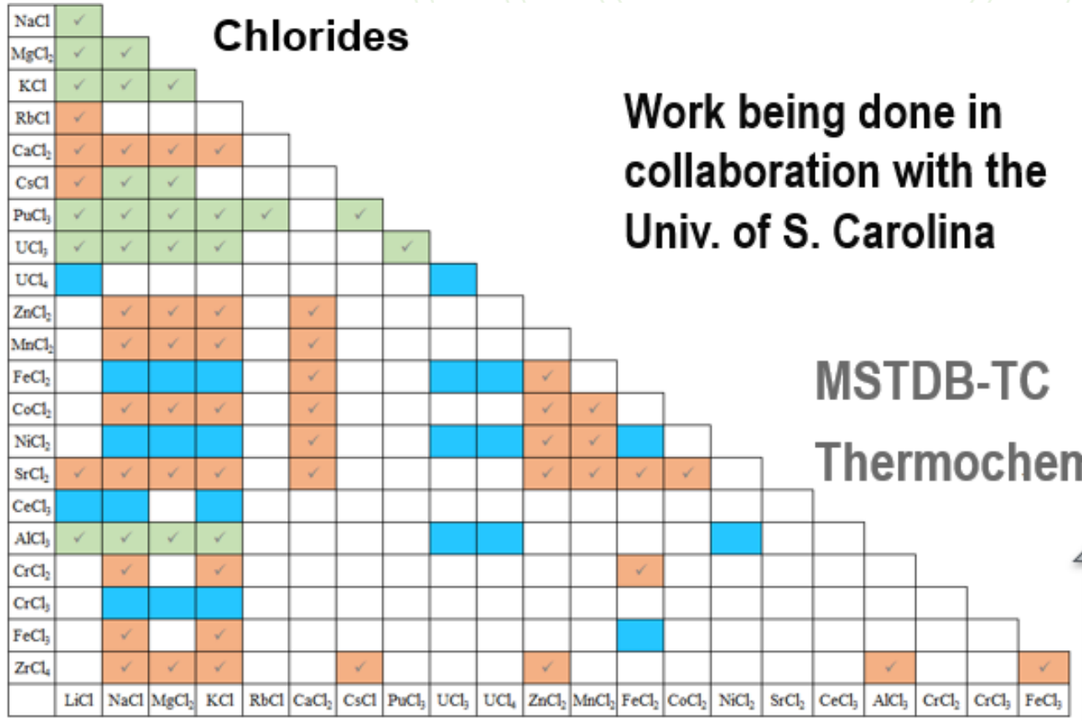
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Supplement missing entries in MSTDB through MD and ML-based methods (literature and computation)



In Model
In Literature
In Progress



Work being done in collaboration with the Univ. of S. Carolina

MSTDB-TC
Thermochemical

KF	B						
LiF	A, B, D	A, B, C, D					
NaF	A, B	A, B, C	A, B, C, D				
ThF4		A, B	A, B, D	A, B			
UF3							
UF4		A, B	A, B	A, B			
ZrF4			A	A, B, D			
	BeF2	KF	LiF	NaF	ThF4	UF3	UF4

Properties modeled
A: Density
B: Viscosity
C: Thermal conductivity
D: Heat capacity

KCl	A, B							
LiCl	A	A, B, D						
MgCl2		A, B, D	A, B					
NaCl	A, B	A, B, D	A, D	A, B				
PuCl3								
ThCl4		A			A			
UCl3		A, B	A		A, B			
UCl4		A, B	A	A	A, B			
ZrCl4		A						
	AlCl3	KCl	LiCl	MgCl2	NaCl	PuCl3	ThCl4	UCl3	UCl4

MSTDB-TP
Thermophysical

Accomplishments

- **Presentations and symposia (full or partial support)**
  - VAG was invited to present at the “Theory in Actinide Science: Chemistry and Materials” Santa Fe, NM Feb 26-Mar 1 2023.
  - Invited seminar at Istituto Italiano di Tecnologia, Genoa, Italy, January 2023.
  - MSR work was highlighted at the 2024 GRC in Chemical Separations, Galveston TX, Jan 21-26
  - Invited presentation to Spring National ACS, 2024
  - Co-organized 2025 Spring National ACS, “f-Element Chemistry and Non-Equilibrium Phenomena: Dissolution, Adsorption and Radiolysis Kinetics in Nuclear Wastes”
- **Reports**
  - Termini, N., Birri, T., Smith, B., Chesser, R., Numbers, J., Garland, K., ... & Glezakou, V. (2024). *FY24 Progress Report on Viscosity and Thermal Conductivity Measurements of Nuclear Industry Relevant Chloride Salts: An Experimental and Computational Study* (No. ORNL/TM-2024/3650). Oak Ridge National Laboratory (ORNL), Oak Ridge, TN (United States).
  - Smith, B. A., Glezakou, V.-A., Termini, N. & Birri. A. FY24 Progress Report on Atomistic Simulation for Thermophysical properties of U-containing Halide Molten Salts” (ORNL/SPR-2024/3729) Oak Ridge National Laboratory, Oak Ridge TN (United States).
- **Publications**
  - Nguyen, M. T., Glezakou, V. A., Rousseau, R., & Paviet, P. D. (2023). Exploring NaCl-PuCl3 molten salts with machine learning interatomic potentials and graph theory. *Applied Materials Today*, *35*, 101951.
  - Karlsson, T. Y., Middlemas, S. C., Nguyen, M. T., Woods, M. E., Tolman, K. R., Glezakou, V. A., ... & Paviet, P. D. (2023). Synthesis and thermophysical property determination of NaCl-PuCl3 salts. *Journal of Molecular Liquids*, *387*, 122636.
  - Birri et al “Thermochamical properties in ternary salts LiCl-NaCL-KCl” in preparation
  - Smith, B. A. et al. “CsI transport at graphene interfaces” to be submitted
  - Smith, B. A. et al. “Thermochemical and transport properties of UCl_4/NaCl(KCl)” in progress

Acknowledgements

MSR Campaign Office of Nuclear Energy for funding

Computer resources: NERSC, CADES

AT-23OR070507 Thermochemical and Thermophysical Properties Database Development- Modeling ORNL

ORNL, US DEPARTMENT OF ENERGY under contract DE-AC05-00OR22725



Patricia Paviet (PNNL)



Janelle Eddins (DOE-NE)

THANK YOU!

Questions?

