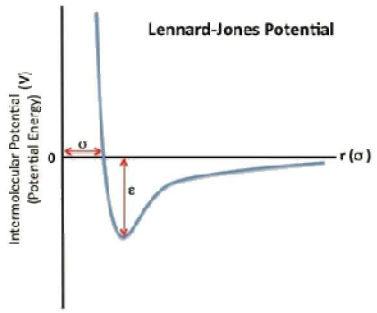
SAND2020-8122C

Using Machine Learning to Predict Self-Diffusion in Lennard Jones Fluids



ACS National Meeting
San Francisco, CA
August 17, 2020

Division/Committee: Chemical Information

**Due to the COVID-19 pandemic this presentation was pre-recorded and will be broadcast*

PRESENTED BY

Joshua P. Allers

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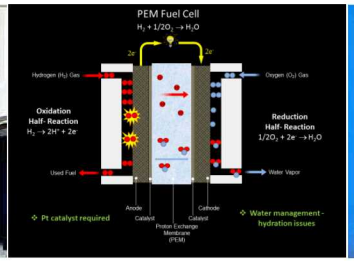
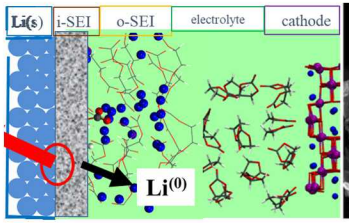
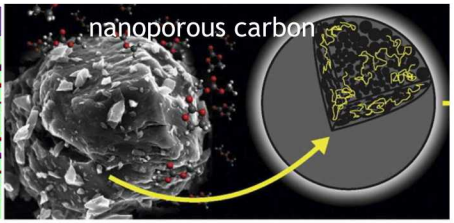
Diffusion of Mixtures Absorbed into Materials: Interest at Sandia National Laboratories

H_2O/MeOH Fuel Cells



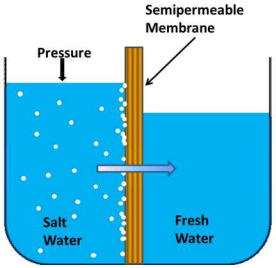
Batteries

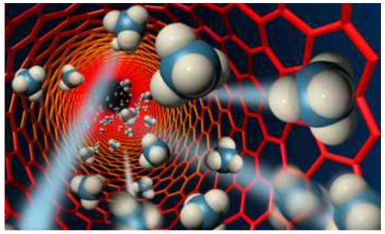




Separation Membranes

Robert Service, Science 2006





LLNL, Carbon Nanotubes

- Understanding the diffusion of chemical species in porous materials critical for the design and performance optimization for different materials.

Current Methods for Calculating Diffusion

Calculating diffusion experimentally

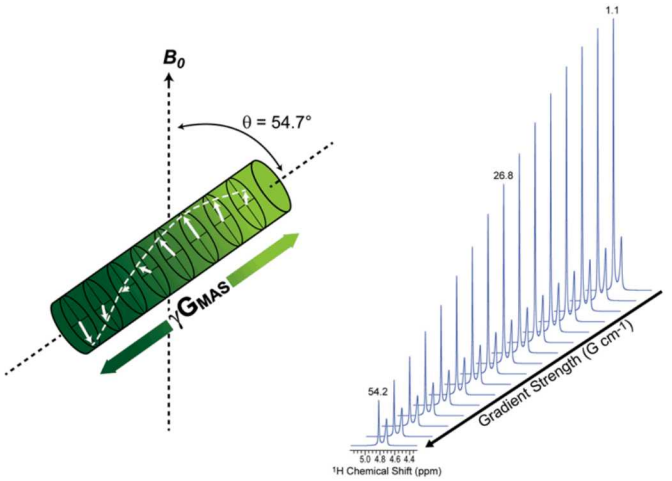
- NMR diffusometry, absorption, CT, etc.
- expensive and time-consuming

Calculating diffusion computationally

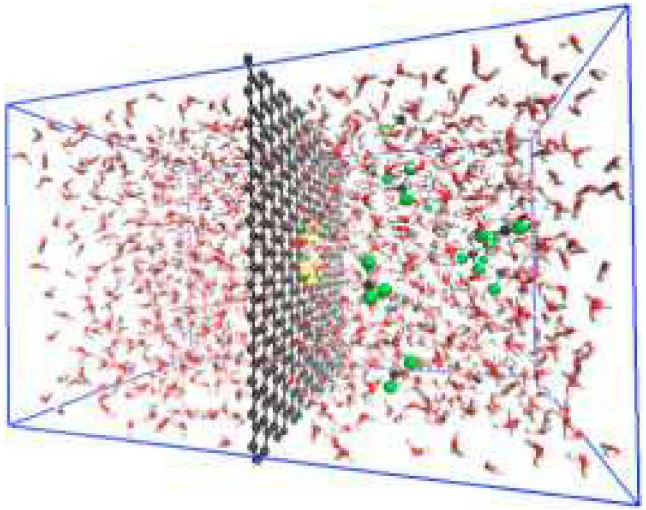
- Molecular dynamics and DFT
- Accurate, but time-consuming
- Require large amounts of computation

Consider studying 10 different compounds

- Forming all possible binary mixtures
- Testing 10 different compositions
- 450 experiments or simulations



Todd Alam and Janelle Jenkins. Advanced Aspects of Spectroscopy. **2012.**



Jafar Azamat *et. al.* Chemical Engineering Science. **2015.**

Challenge - Universal Model for Diffusion

Maxwell Stephen (MS) Model
(dominant diffusion model)

$$\frac{1}{D_{\mathrm{i,self,s}}} = \frac{1}{D_{i,s}} + \frac{x_i}{D_{ii}} + \frac{x_j}{D_{ij}}$$

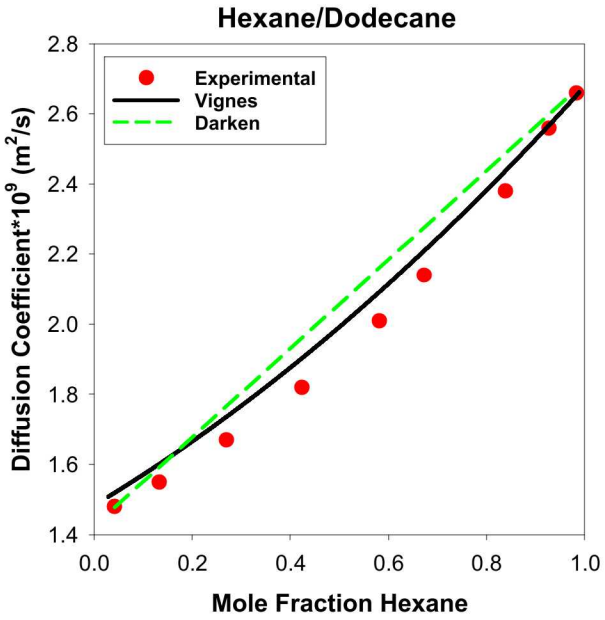
surface self exchange

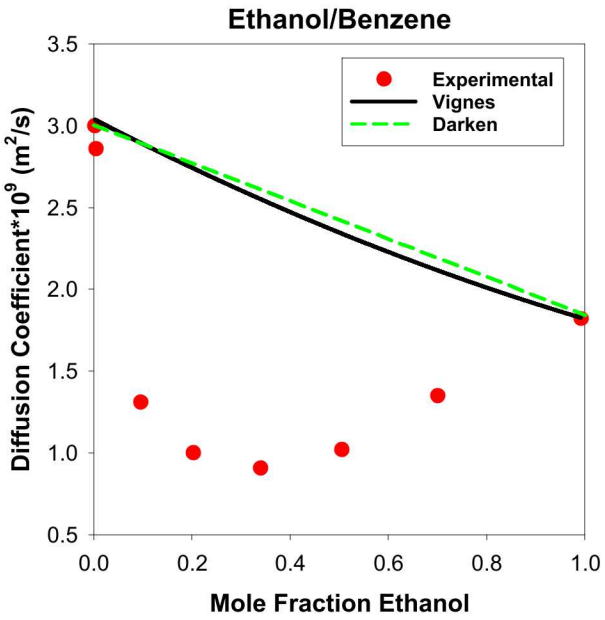
Darken Relationship
(semi-empirical - linear)

$$D_{ij} = D_i D_j \sum_{k=1}^{N} \frac{x_k}{D_k}$$

Vignes Model
(semi-empirical- power law)

$$D_{ij} = \left(D_{ij}^{x_j \to 1}\right)^{x_i} \left(D_{ij}^{x_j \to 1}\right)^{x_j} \prod_{k,k\neq i,j}^{i=N} \left(D_{ij}^{x_k \to 1}\right)^{x_k}$$



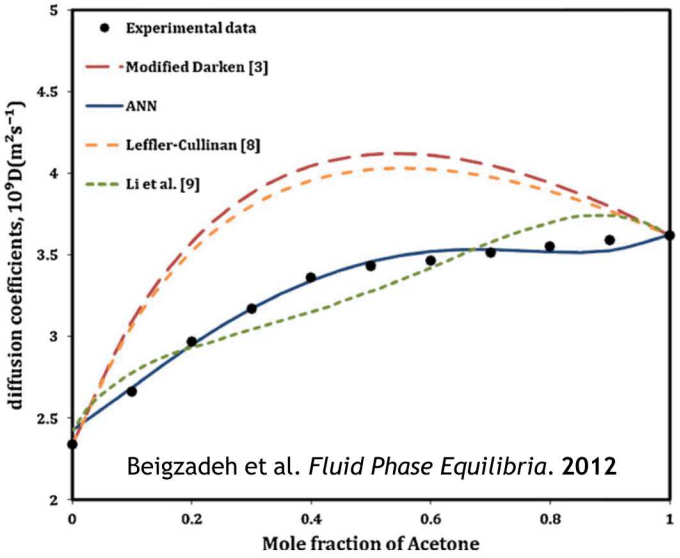


Goal: Develop a generalized model that can accurately predict diffusion in multi-component mixtures

Potential of Machine Learning

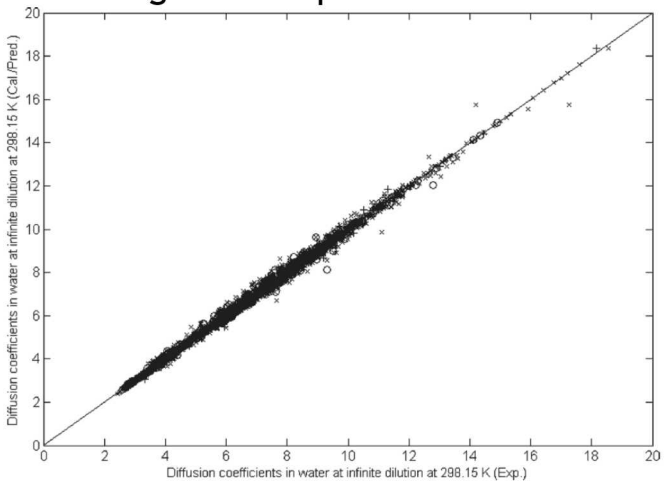
- Rapidly growing field in materials science
- Powerful tools for prediction
  - Random Forests
  - Artificial Neural Networks (ANN)
  - Symbolic Regression
  - Genetic Engineering
- Already showing promise in literature

acetone-chloroform mixture



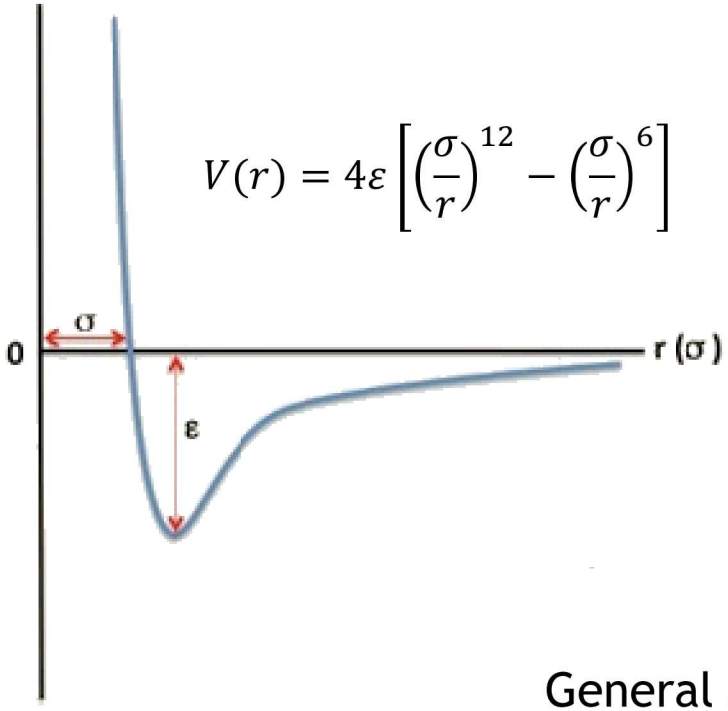
Beigzadeh et al. *Fluid Phase Equilibria*. **2012**

organic compounds in water



Gharagheizi et al. *Journal of Chemical Engineering Data*. **2011**

Start Simple – Lennard Jones (LJ)

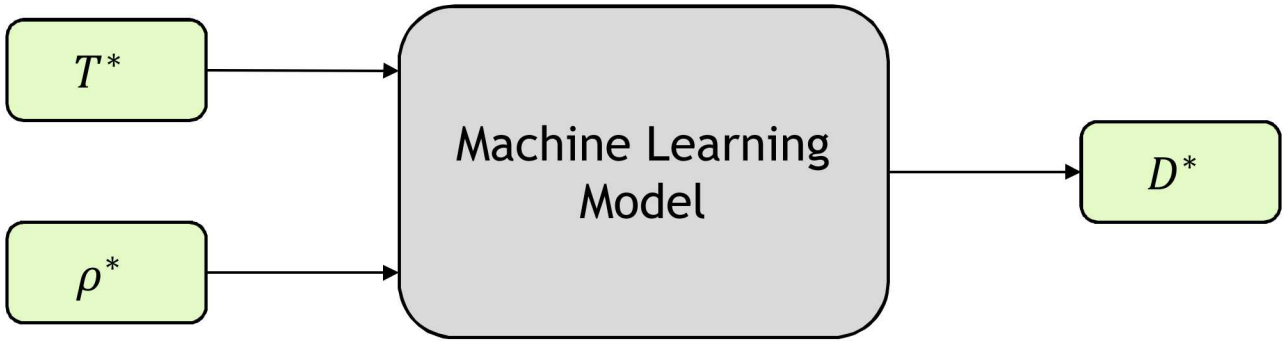


$$V(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right]$$

$$T^* = \frac{Tk}{\varepsilon} \qquad \rho^* = \frac{N\sigma^3}{V} \qquad D^* = D\frac{\sqrt{\frac{m}{\varepsilon}}}{\sigma}$$

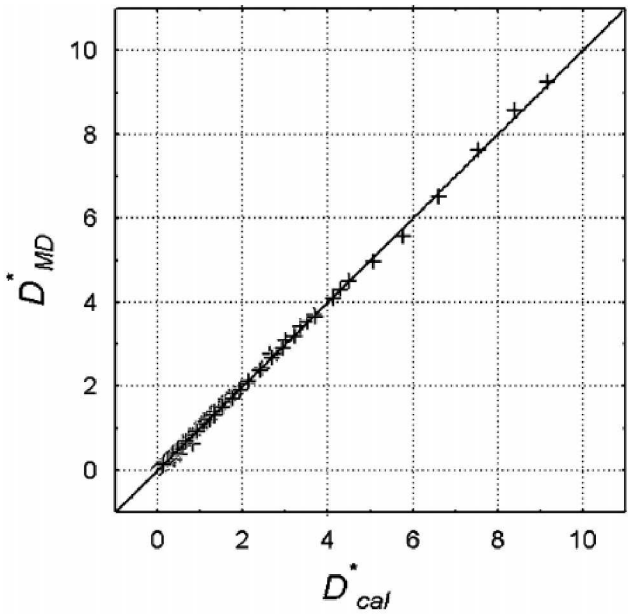
- Explained by two parameters
  - σ - Distance where potential becomes zero
  - ε - Well depth and measure of attractive force
- Allows model development over multiple phases

General Model Architecture



Lennard Jones Molecular Dynamics (MD)

- Many MD simulations have been performed for LJ systems
- Zhu's empirical equation is one of the better models
  - Has 8 adjustable parameters
- Will act as the "gold standard" for comparison



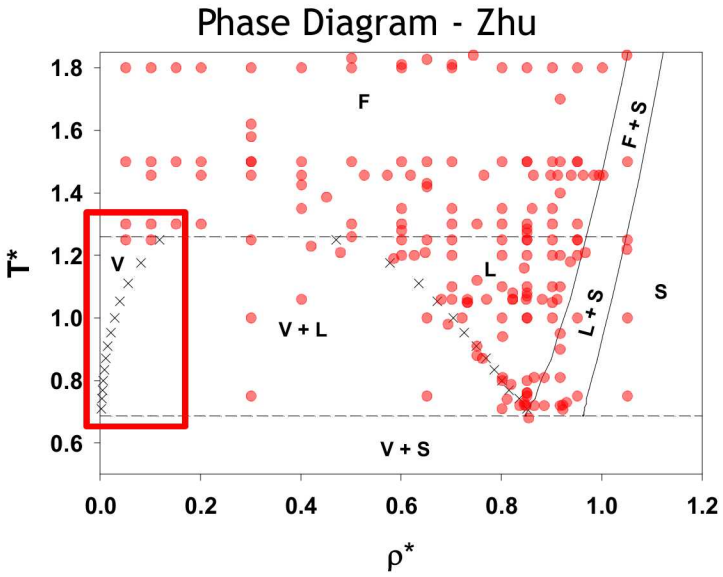
$$D^*_{cal} = \frac{3\sqrt{T^*}}{8\rho^*\sqrt{\pi}} A \times B$$

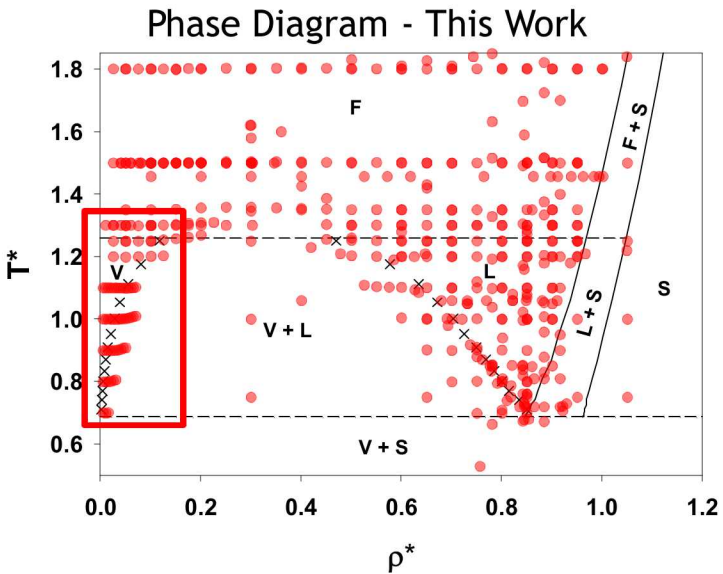
$$A = \left(1 - \frac{\rho^*}{a(T^*)^b}\right)\left[1 + (\rho^*)^c\left(\frac{P1(\rho^* - 1)}{P2(\rho^* - 1) + (T^*)^{(P3+P4\rho^*)}} + P5\right)\right]$$

$$B = exp\left(\frac{-\rho^*}{2T^*}\right)$$

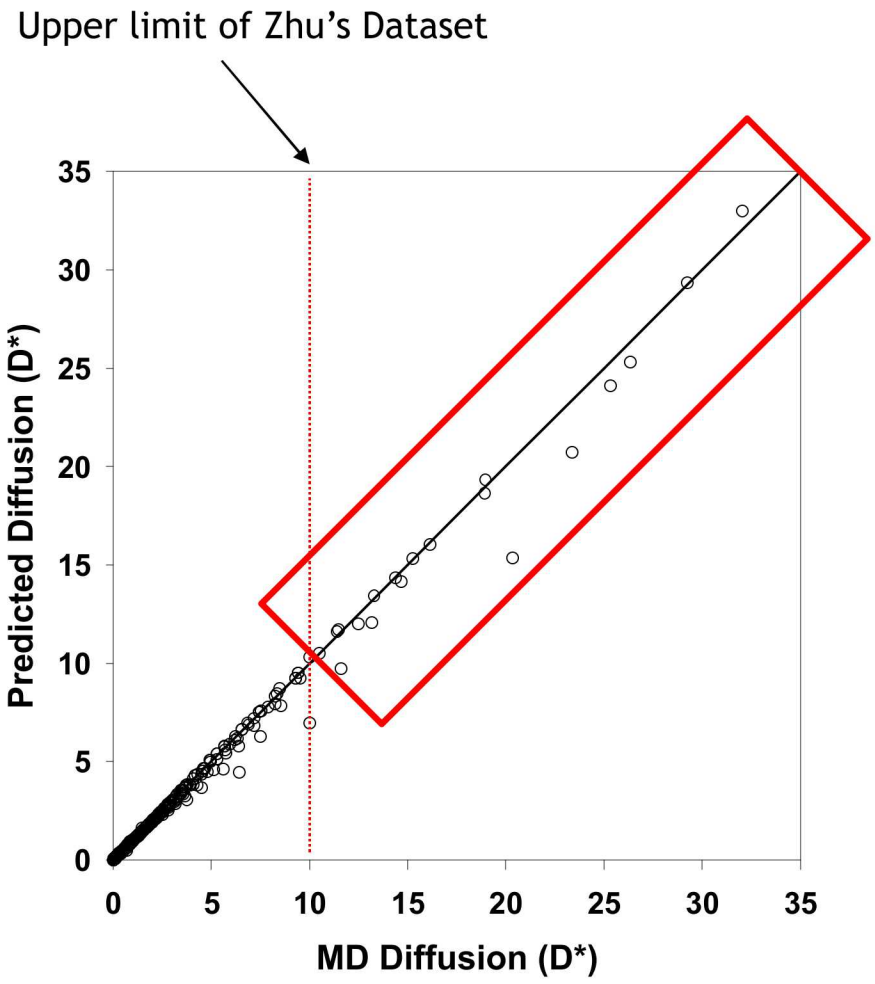
Yu Zhu, Xiaohua Lu, Jian Zhou, Yanru Wang, Jun Shi, "Prediction of diffusion coefficients for gas, liquid and supercritical fluid: application to pure real fluids and infinite binary solutions based on the simulation of Lennard-Jones fluid". *Fluid Phase Equilibria*. **2002**. https://doi.org/10.1016/S0378-3812(01)00669-0

Expanded Diffusion Dataset



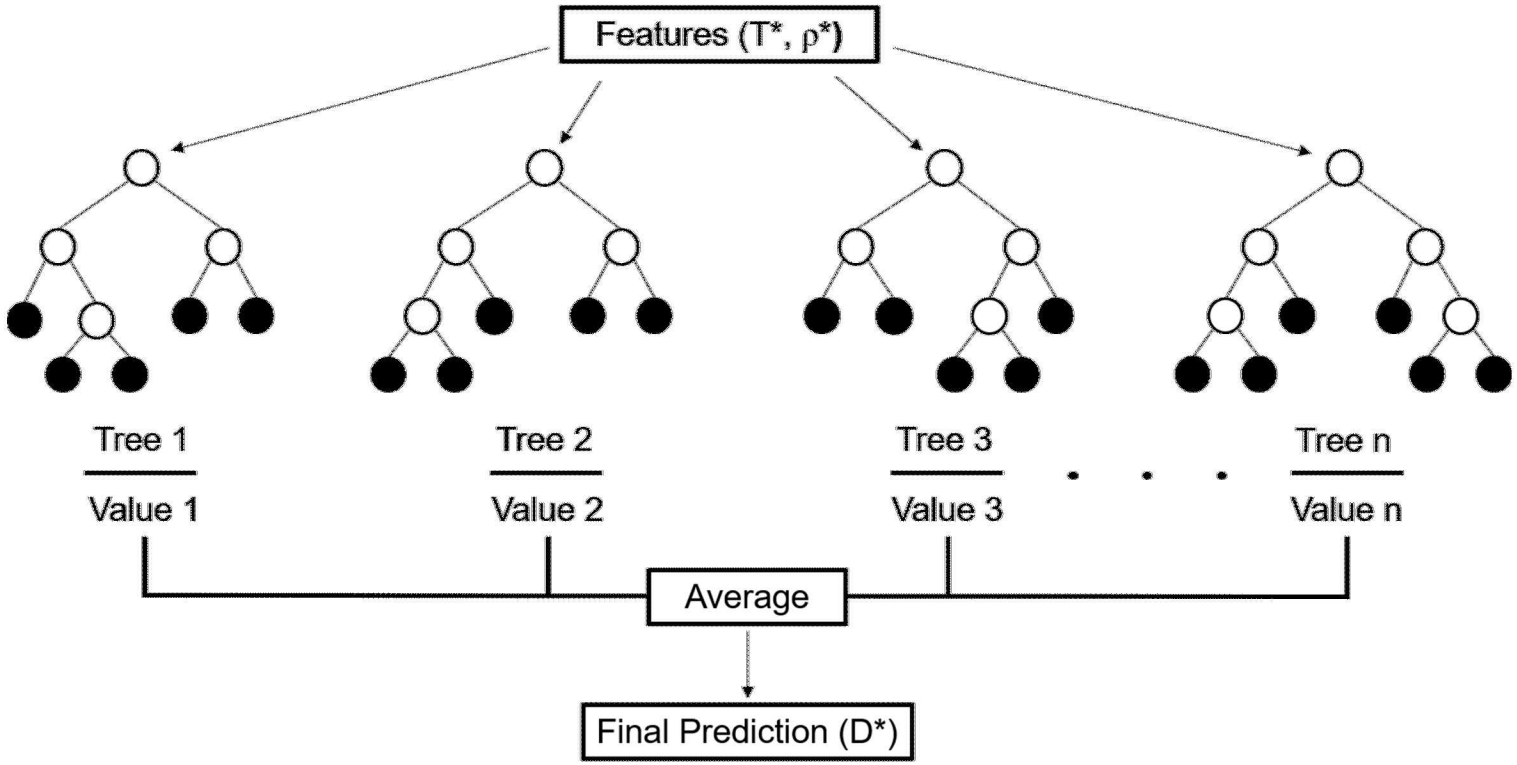


V - vapor
L - liquid
F - supercritical
S - solid



Joshua P. Allers, Jacob A. Harvey, Fernando H. Garzon, Todd M.
"Machine learning prediction of self-diffusion in Lennard Jones fl
Chem. Phys. 153, 034102 (2020).

Random Forest Architecture

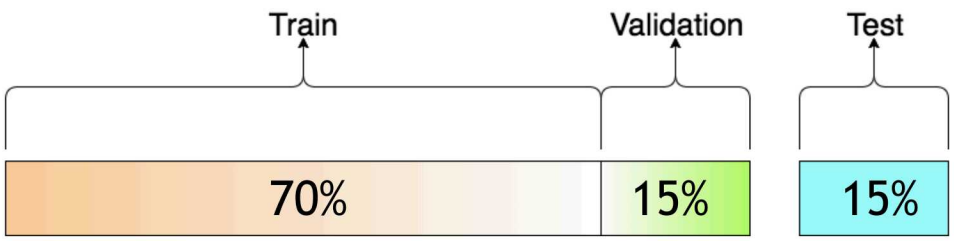


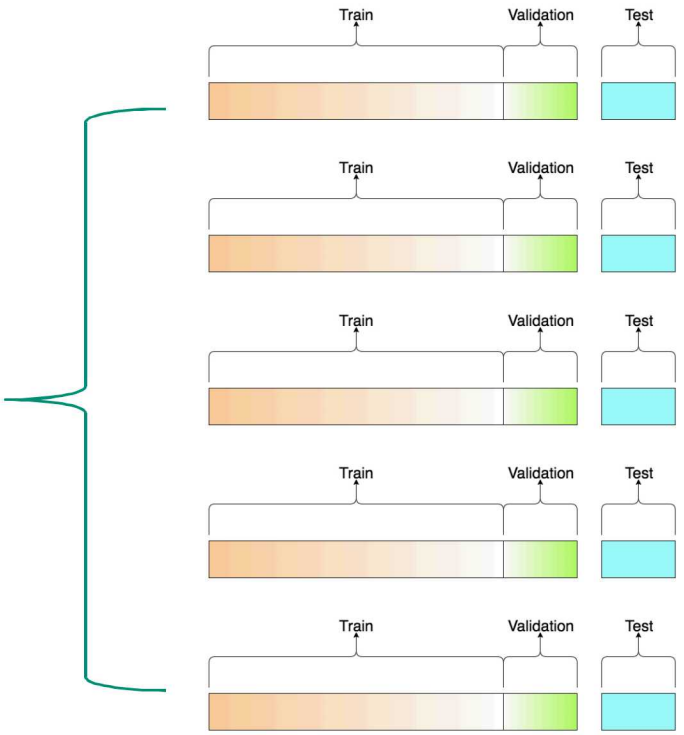
- Software: Python Scikit-Learn
- Optimal number of trees: 235

Joshua P. Allers, Jacob A. Harvey, Fernando H. Garzon, Todd M. Alam. "Machine learning prediction of self-diffusion in Lennard Jones fluids." *J Chem. Phys.* 153, 034102 (2020).

Performance Assessed with Cross Validation (CV)

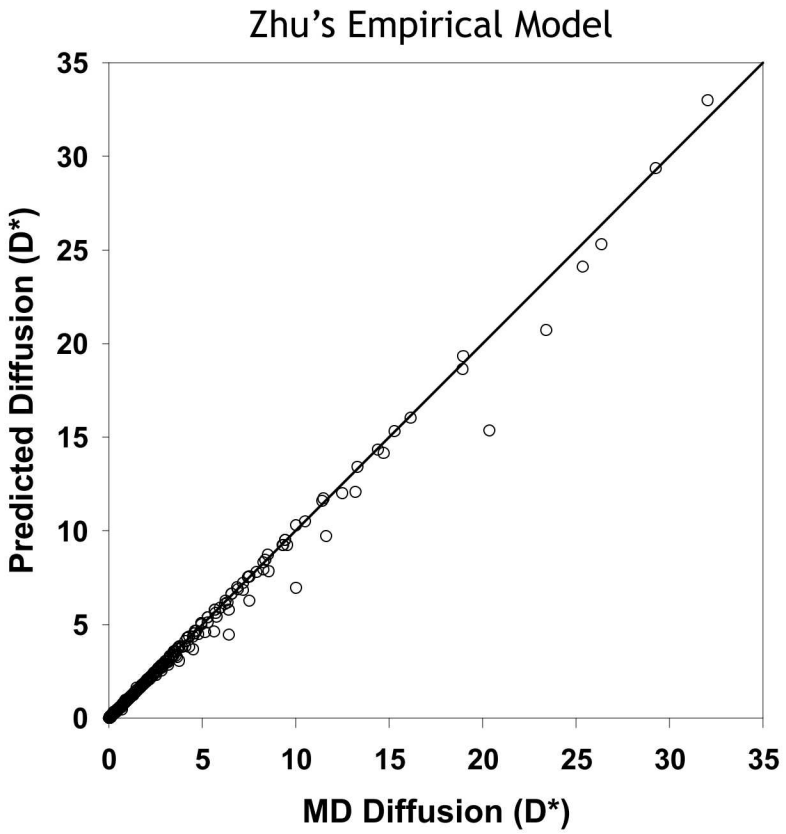
- Splits dataset in even train/validate/test sets
  - ML models train on the 70%
  - Validation set used to adjust hyperparameters (model parameters)
  - Test set gave final predictive power
- 5 different splits of the data
  - Randomly divided
- Performance metrics:
  - Mean Square Error (MSE)
  - Correlation Coefficient (R)

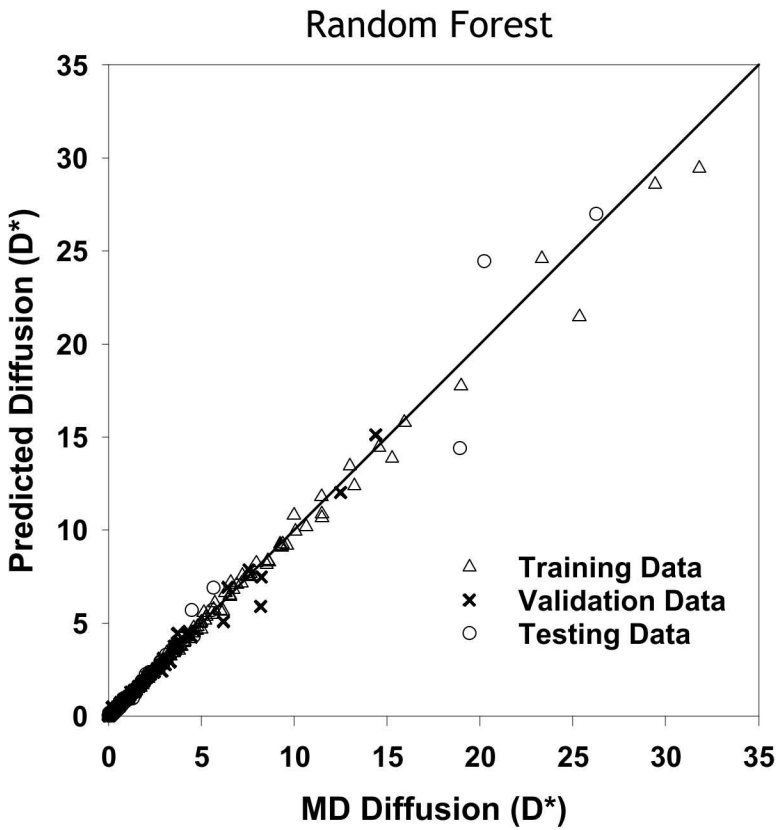




towardsdatascience.com/train-validation-and-test-sets-72cb40cba9e7

Performance of Random Forest

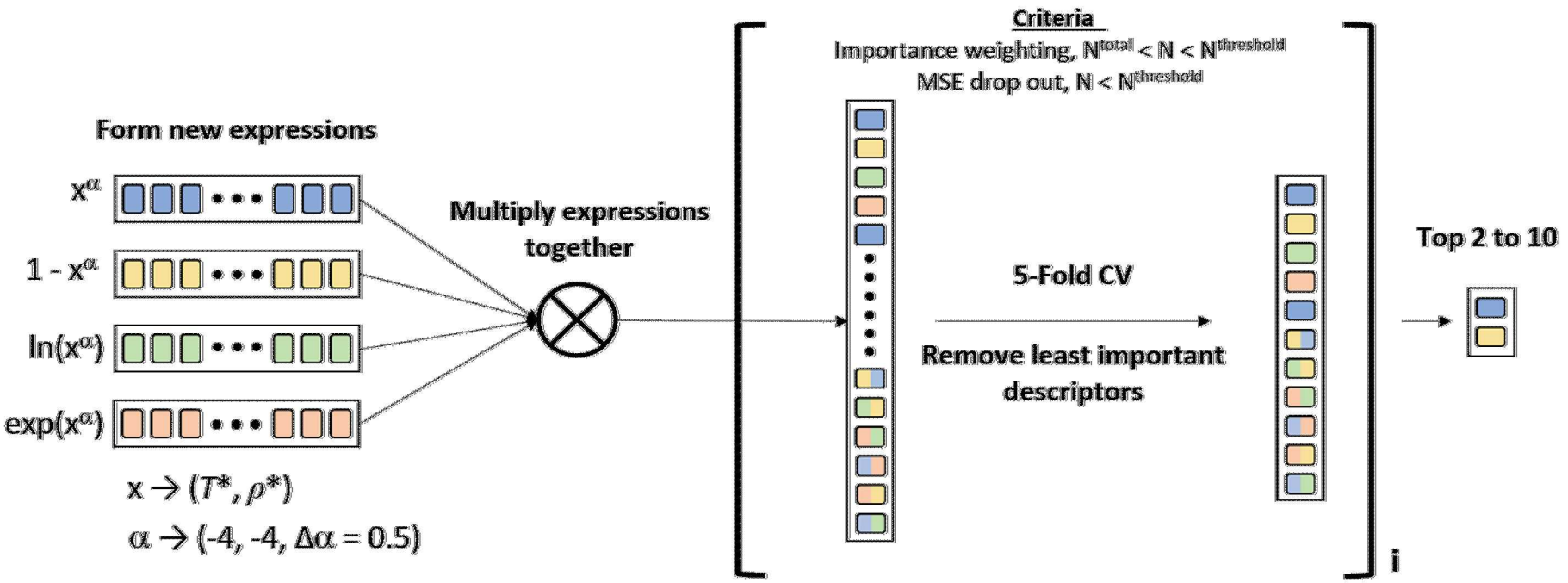




Model	Zhu	RF
Mean MSE	0.067	0.18
Std. Dev.	± 0.077	± 0.21
Mean R	0.99685	0.99624

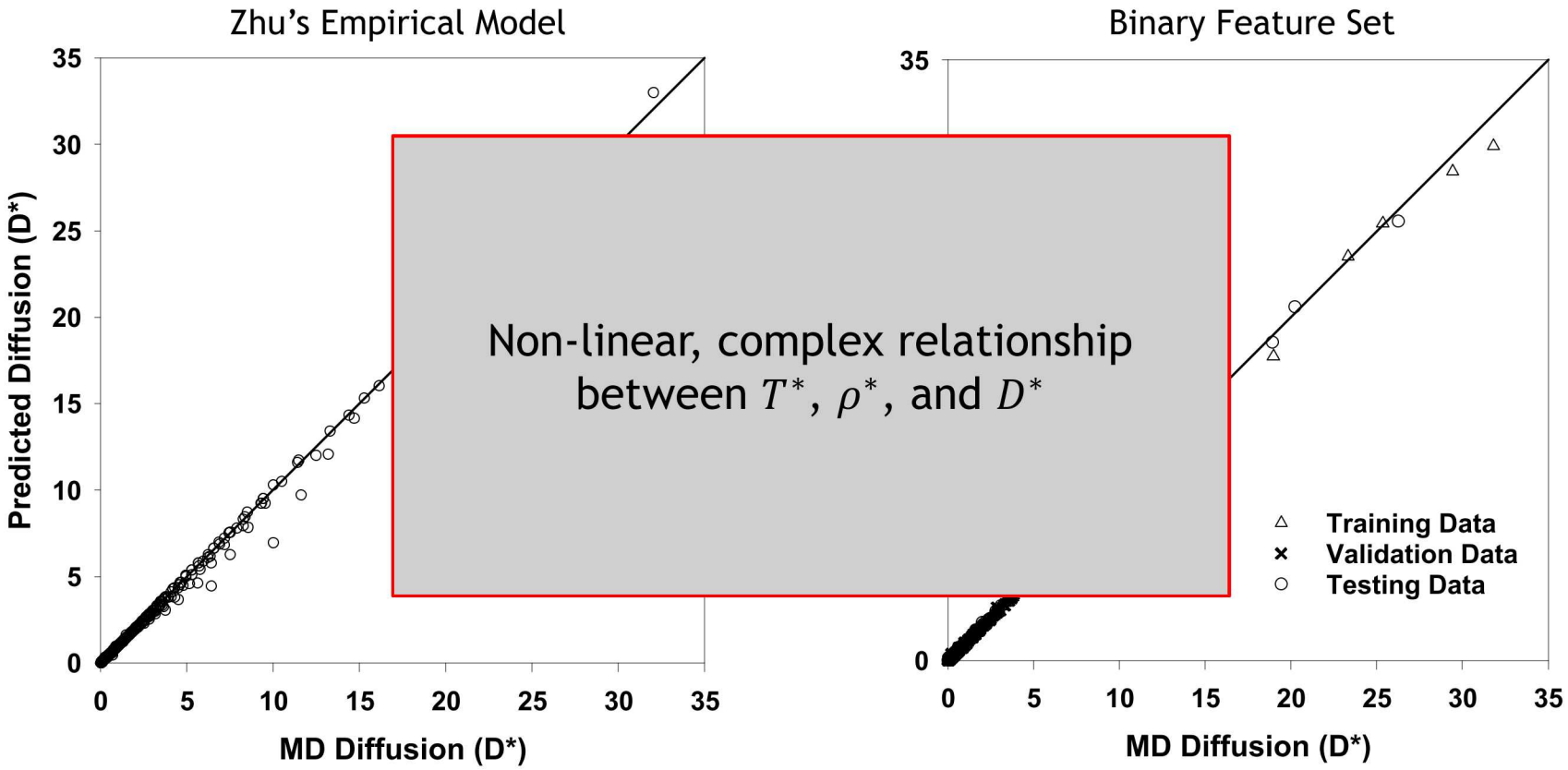
Joshua P. Allers, Jacob A. Harvey, Fernando H. Garzon, Todd M. Alam. "Machine learning prediction of self-diffusion in Lennard Jones fluids." *J Chem. Phys.* 153, 034102 (2020).

Feature Engineering Process



Solo	Binary	Self-Binary
$f(T^*)$ **or** $f(\rho^*)$	$f(T^*) * g(\rho^*)$	$f(T^*) * g(T^*)$
128 Features	4224 Features	7296 Features
e.g. $\ln(T^*)$	e.g. $\ln(T^*) * \exp(\rho^*)$	e.g. $\ln(T^*) * \exp(T^*)$

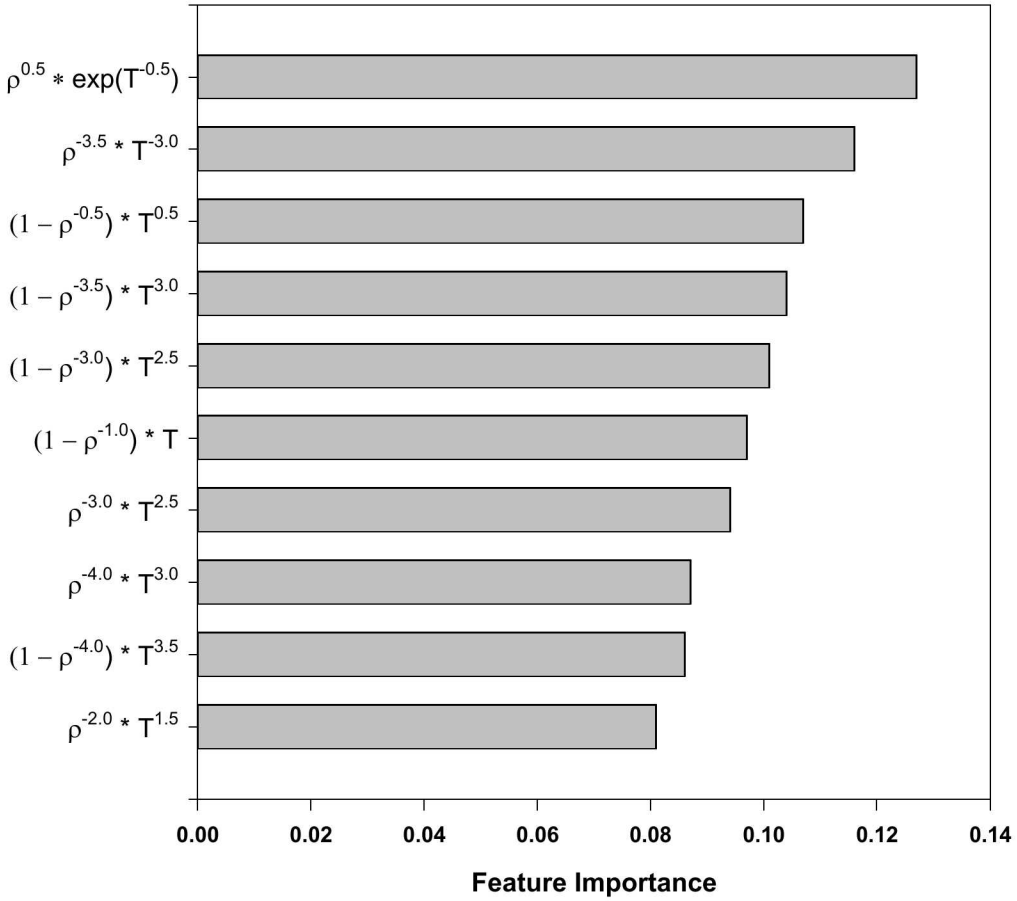
Random Forest with Feature Engineering



Model	Zhu	RF	Solo	Binary	Self-Binary
Mean MSE	0.067	0.18	0.15	0.052	0.047
Std. Dev.	± 0.077	± 0.21	± 0.18	± 0.047	± 0.072
Mean R	0.99685	0.99624	0.99642	0.99913	0.99912

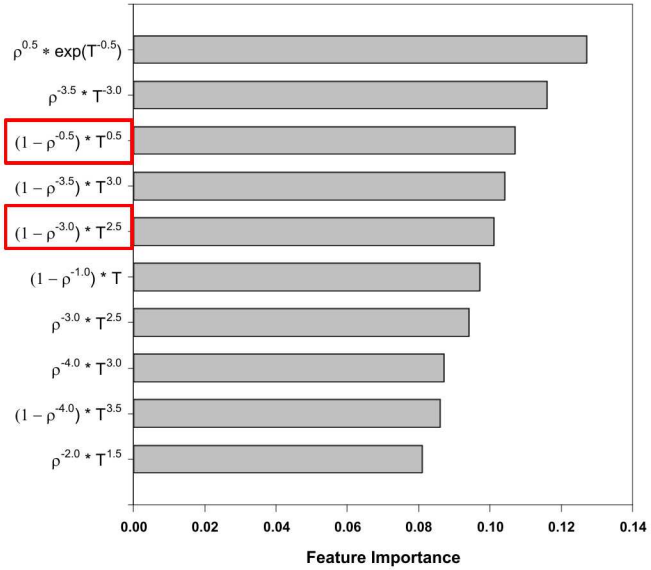
Joshua P. Allers, Jacob A. Harvey, Fernando H. Garzon, Todd M. Alam. "Machine learning prediction of self-diffusion in Lennard Jones fluids." *J Chem. Phys.* 153, 034102 (2020).

Most Important Features – Binary Feature Set



Observed trend: non-linear functions of T^* and ρ^* consistently show up as the most important features

Most Important Features – Binary Feature Set



Continued iteration →

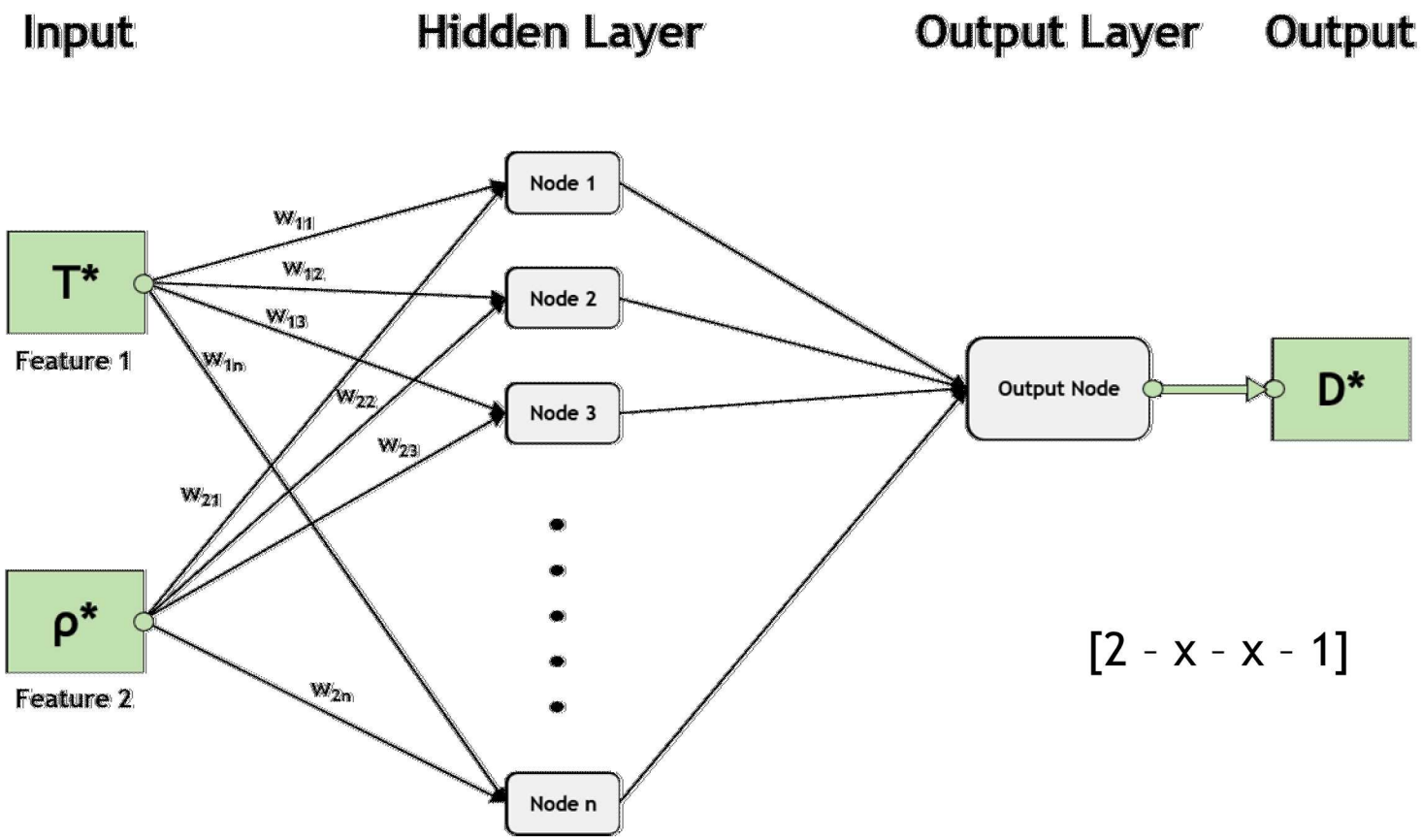
Top 2 Features

$$(1 - \rho^{-0.5}) * T^{0.5}$$

$$(1 - \rho^{-3.0}) * T^{2.5}$$

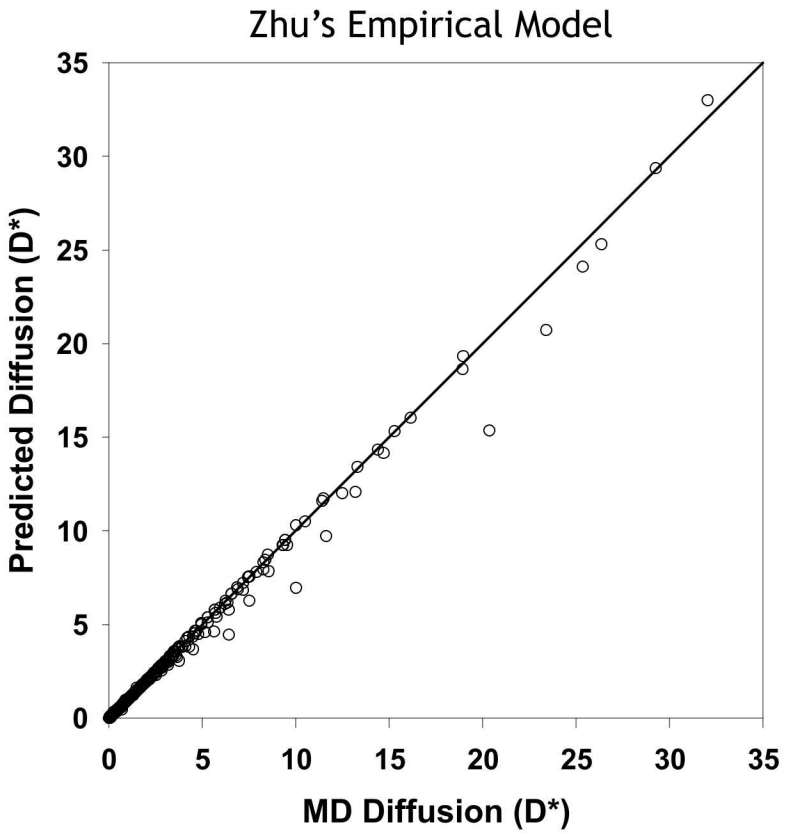
3rd and 5th ranked features become the top 2

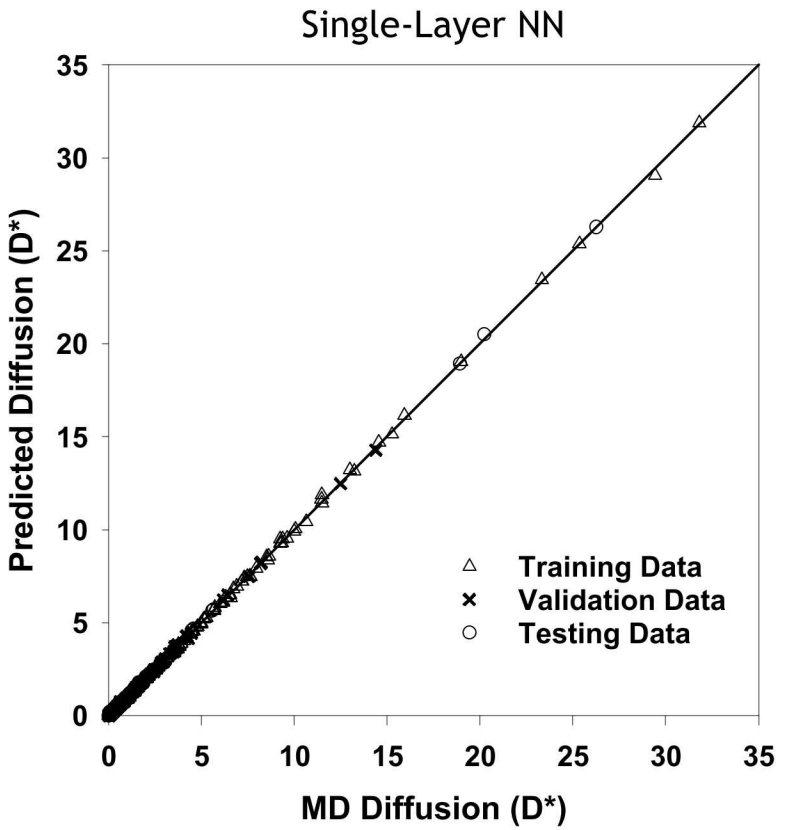
Artificial Neural Network (ANN) Architecture



- Software: MATLAB Deep Learning Toolbox

Neural Network Performance





Model	ANN(2-14-12-1)	ANN(2-14-1)	RF-FE	Zhu	RF
Mean MSE	0.0012	0.0016	0.052	0.067	0.18
Std. Dev.	± 0.0009	± 0.001	0.047	± 0.077	± 0.21
Mean R	0.99994	0.99991	0.99913	0.99685	0.99624

Joshua P. Allers, Jacob A. Harvey, Fernando H. Garzon, Todd M. Alam. "Machine learning prediction of self-diffusion in Lennard Jones fluids." *J Chem. Phys.* 153, 034102 (2020).

Conclusions

- Machine learning improved upon existing empirical relationships in LJ system
- Random Forest with T^* and ρ^* performed worse than Zhu's equation
- Employed feature engineering, which improved predictions
  - On par with Zhu's equation
  - Pointed to non-linear, complex relationships
- Artificial Neural Networks provide the best predictions
  - Will be used going forward with real systems

Ongoing Work

- Machine Learning extended to sets of binary LJ fluids
- Artificial Neural Networks assessed on a dataset of pure solutions
  - Shows excellent performance over multiple compounds and phases

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Thank you for you attention - Questions?

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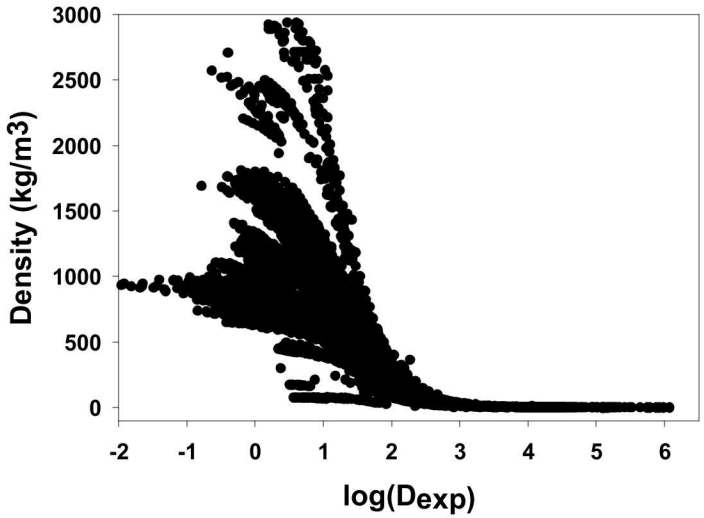
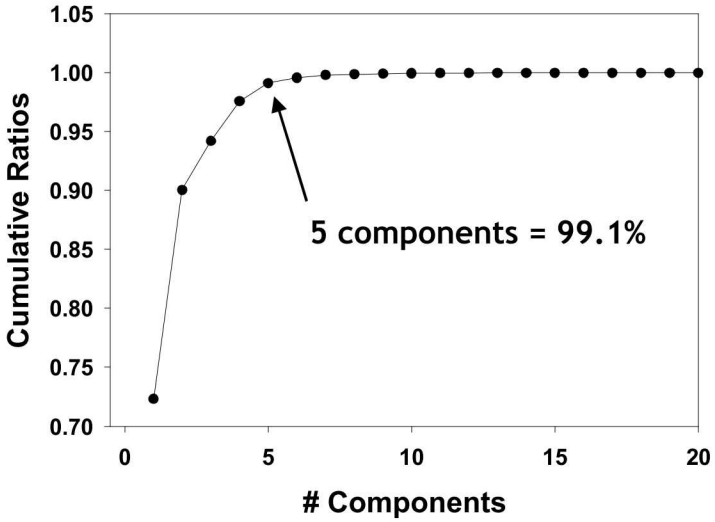
Project funding through the Sandia LDRD program

Additional Slides

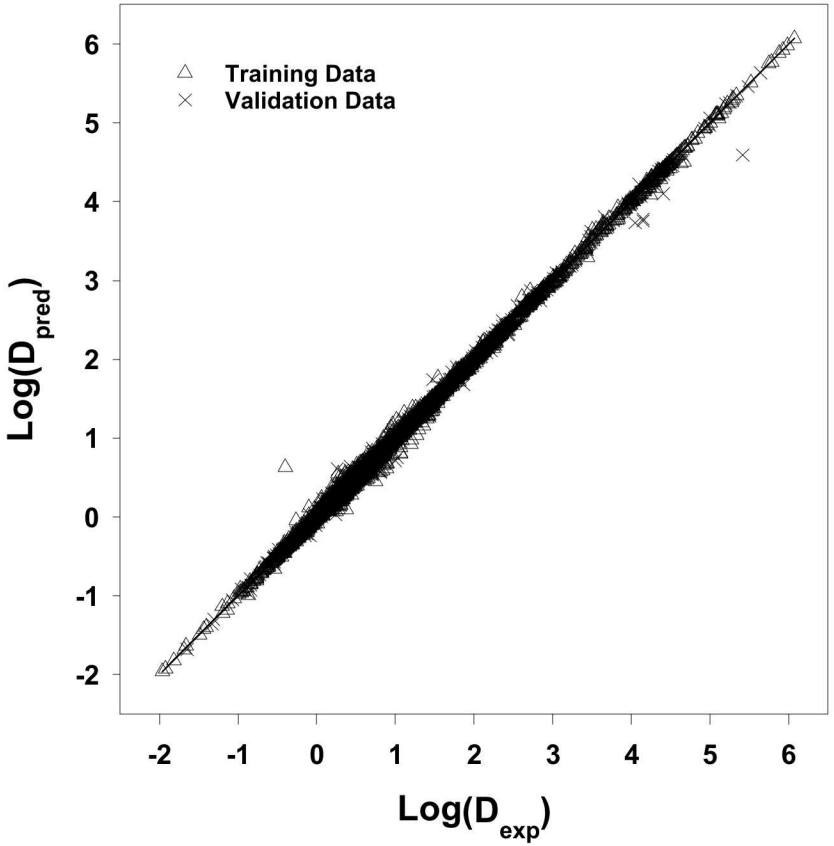
Current Work – Pure Solutions Diffusion





- 6252 points from a wide range of compounds
  - Multiple phases present
- 24 features were collected including:
  - Critical properties
  - Experimental properties
  - Phase information
  - Structural information
- Principal Component Analysis (PCA) used to reduce the dimensionality

Current Work – Pure Solution Diffusion



- Models are robust and generalized over a 5-fold cross-validation
- Shows good predictions over multiple phases

Joshua P. Allers, Fernando H. Garzon, Todd M. Alam. *In Progress*.