Development of a machine learning model for polyethylene pyrolysis using a detailed reaction mechanism

NATIONAL ENERGY TECHNOLOGY LABORATORY

2024 Clearwater Clean Energy Conference

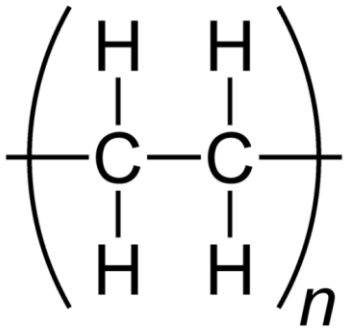
June 16-19th, Clearwater, FL

Ross Houston, Hang Zhou, and Mehrdad Shahnam
National Energy Technology Laboratory

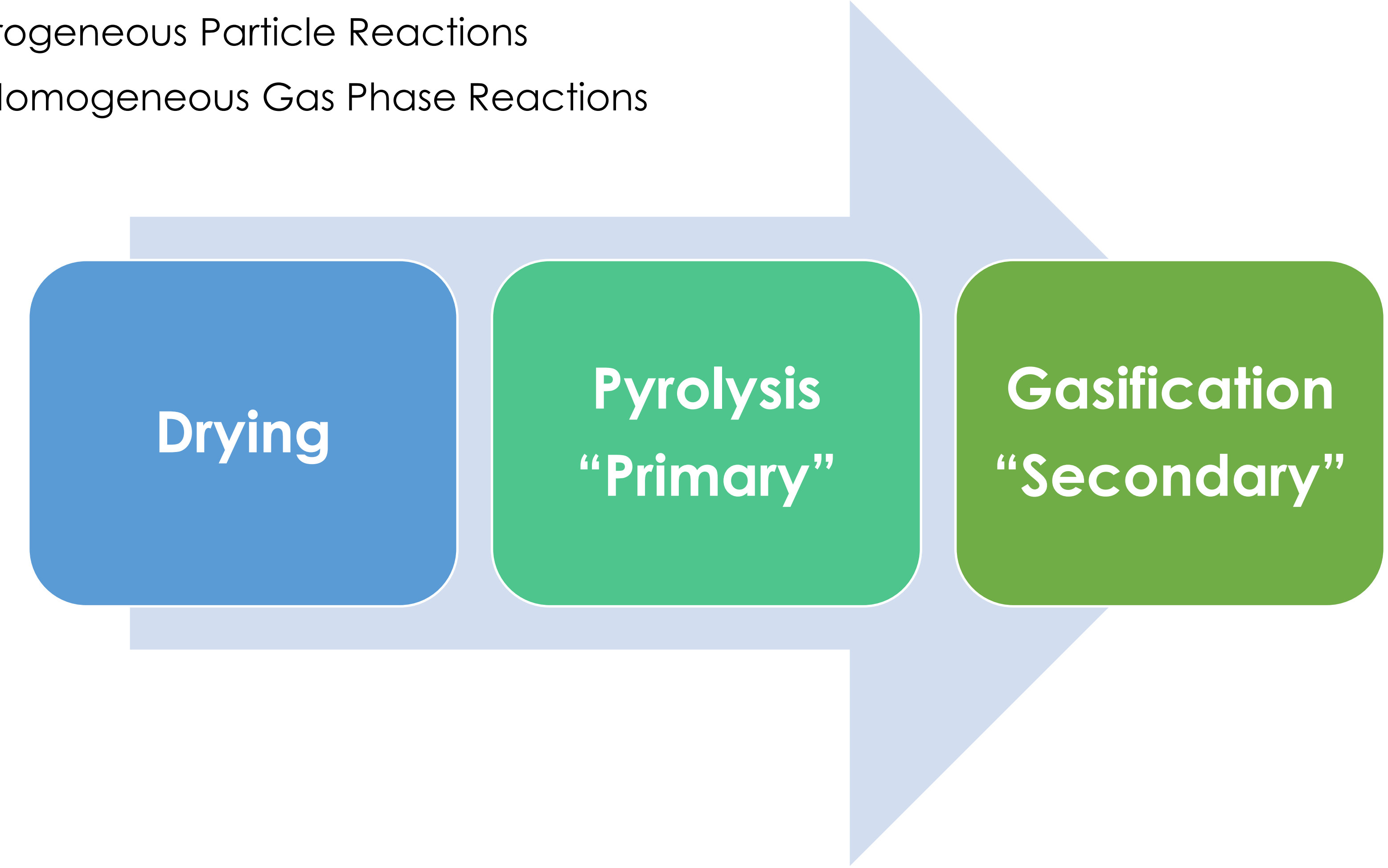
Gasification of high-density polyethylene (HDPE)

Necessary to subclassify gasification for accurate kinetic modeling

- Kinetically the gasification process can be thought of as pyrolysis/primary and gasification/secondary reactions
- Pyrolysis – Heterogeneous Particle Reactions
- Gasification – Homogeneous Gas Phase Reactions



HDPE



- Char, Ash
- Tar (C_{4+})
- Light gases (C_0-C_4)

Current state of HDPE pyrolysis kinetics

Collaboration with CRECK Modeling Group at Polytechnic University of Milan

- HDPE characterized by two lumped functional groups:
  - Mid-Chains (P-P)
  - End-Chains (P-)
- Representative Mid Chains
  - P-C20H40-P(L)
  - P-C40H80-P(L)

HMW (P-P) ⟶ (P-) ⟶ LMW Products

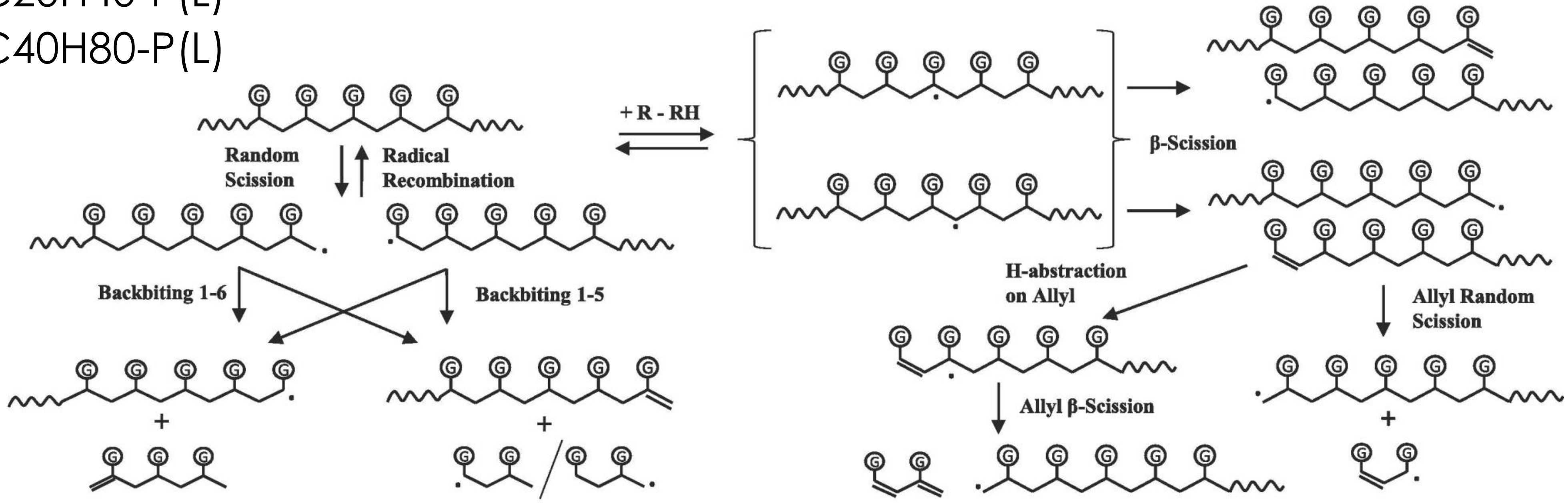


Image credit: A. Locaspi, et al., *Waste Management* **156** (2023) 107-117

Current state of HDPE pyrolysis kinetics

Collaboration with CRECK Modeling Group at Polytechnic University of Milan

- Low-molecular weight (LMW) characterized by real species up to C_5
- Larger species (C_{6+}) described by lumped paraffin and olefin species

Current Reaction Schemes:

- 71 species, 1377 reactions (71_1377)
- 71_969
- 42_737
- 10_10

Table 1. Classification of Mid- and End-Chain species for HDPE

	Paraffin	Olefin
Mid-Chain (MC)	P-$C_{20}H_{40}$-P(L)	
End-Chain (EC)	P-$C_{20}H_{41}$(L)	P-$C_{24}H_{23}$(L)
MC radical	P-$C_{20}H_{39}$-P(L)	
EC internal radical	P-$C_{20}H_{40}$(L)	P-$C_{24}H_{22}$(L)
EC position specific radical	P-$C_{20}H_{40}$-T(L)	P-$C_{24}H_{22}$-A(L)

Effect of Simplifying Kinetic Schemes

NATIONAL ENERGY TECHNOLOGY LABORATORY

Increase computational efficiency but decrease species accuracy

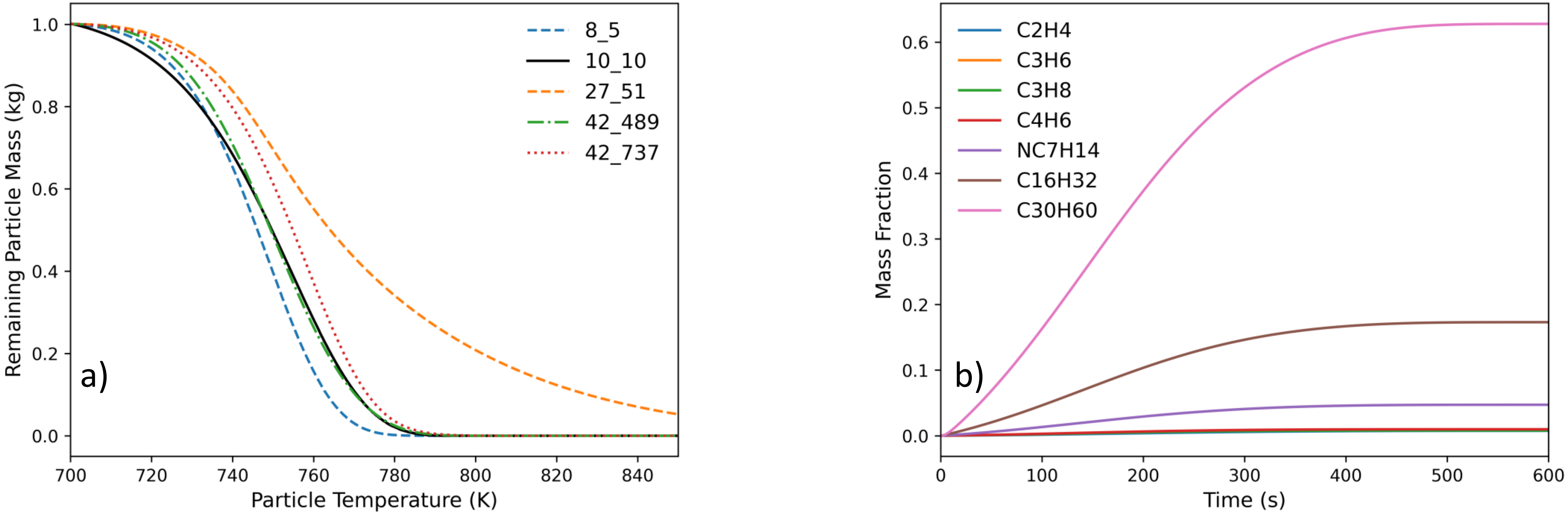


Figure 1. a) Mass loss of an HDPE particle using different reactions schemes, and b) mass fractions of the product gases using a simplified 10 species, 10 reaction scheme. *Note:* a) X_Y format represents a scheme with X species and Y reactions.

- Simplifying reaction schemes reduces computational cost
- Lose information about the detailed composition of products and remaining plastic
- Can lead to significantly different behavior
- Lumped species are not always easily attributed to experimental observations

Machine learning (ML) approach for HPDE reaction kinetics

NATIONAL ENERGY TECHNOLOGY LABORATORY

Increase computational speed while maintaining detailed speciation

Conventional Approach

- Take a single reaction:

A => B + C

$$k = AT^{\beta} e^{\frac{-E_a}{RT}}$$

$$\frac{dX_A}{dt} = AT^{\beta} e^{\frac{-E_a}{RT}} C_A$$

- Create the full set of ODEs

$$\frac{dX_{mn}}{dt} = \sum dX_{mn,production} - \sum dX_{mn,consumption}$$

- Quickly becomes cumbersome with 700+ reactions

Machine Learning Approach

- Three variables present:
  - Particle species concentration
  - Temperature
  - Time Step

$$f\left(X_{1,0}, \dots, X_{mn}, T_p, dt\right) = X_{0,0} \dots X_{mn}$$

- Predict final mass fractions of all species for a given time step

Data generation for model training

Generate high-fidelity composition data over a range of operating conditions

- HDPE_42_737 scheme
- 1-kg pure HDPE particle
- Initial temperature: 300 K
- Max temperature: 1000 K
- Fixed heating rate:
  - 5, 10, 15 K/min
- Timestep:
  - 1E-6 to 1E-3 s
- 49 total runs
- 4+ million data points

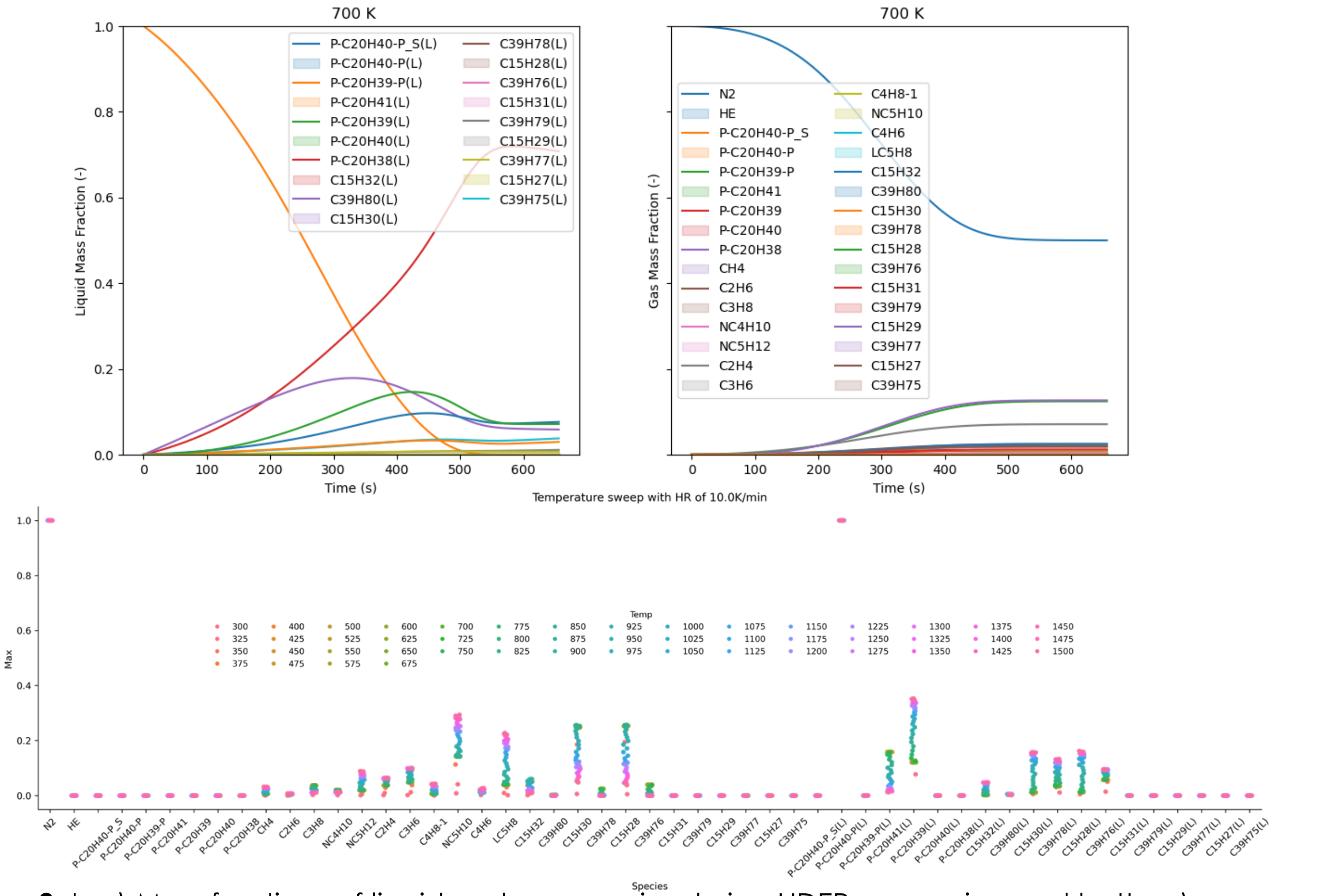
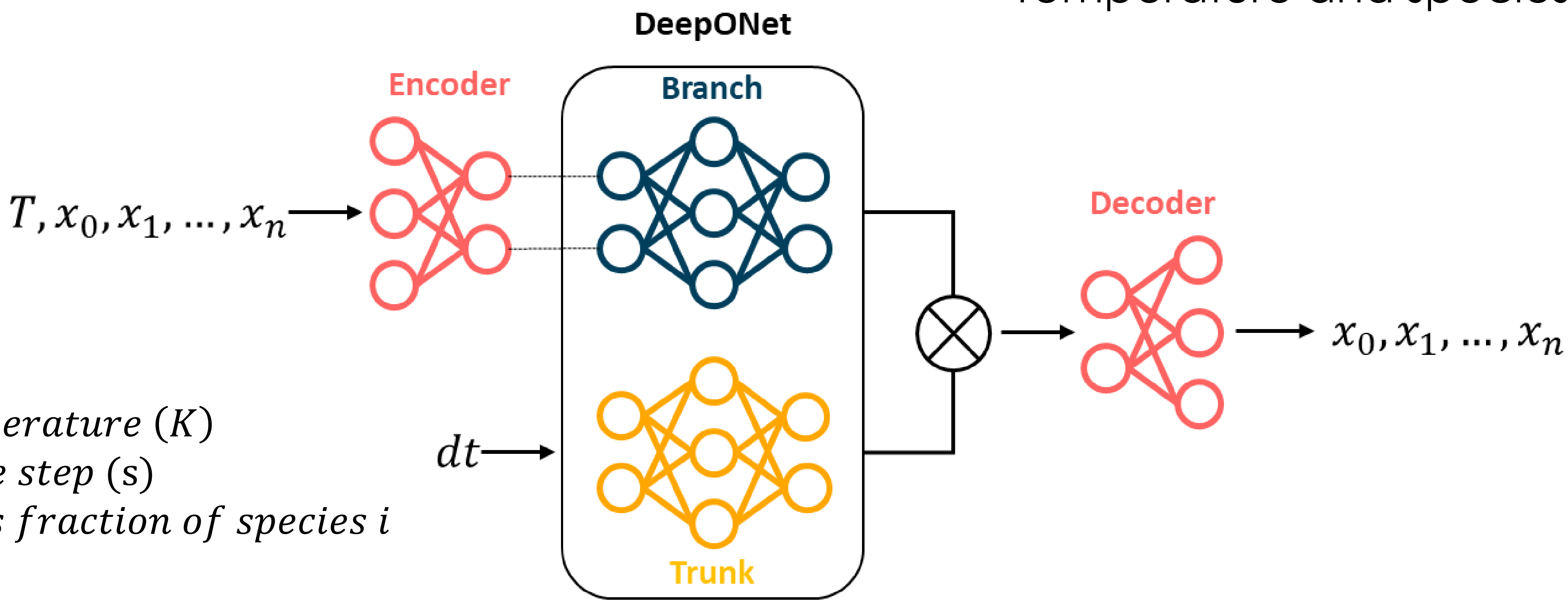


Figure 2. top) Mass fractions of liquid and gas species during HDEP conversion and bottom) maximum mass of each species during conversion over a range of temperatures using a 1-kg basis.

Architecture of the ML model

Implementation of DeepONet structure with physics informed loss functions

- 20 input features
- 35 output features
  - Liquid and gas species
- Trunk Network
  - Handles dt for time dependence
- Branch Network
  - Temperature and species fractions



$T = Temperature\ (K)$
$dt = Time\ step\ (s)$
$x_i = Mass\ fraction\ of\ species\ i$

Physics-informed loss functions

NATIONAL ENERGY TECHNOLOGY LABORATORY

Couple mass-conservation and time-informed loss functions for training

- Training and validation loss calculating using the mean absolute error (MAE)
- Chemical reactions must conserve mass
- ML model is useless if it doesn't abide by physical laws
- Introduce new loss functions
  - Sum of gas species (1)
  - Rate of gas production (2)
  - Sum of liquid species (3)

Standard MAE in PyTorch

$$\frac{1}{BN_{spcs}}\sum_{i=1}^{B}\sum_{j=1}^{N_{spcs}}\left|X_j^{(1)} - \hat{X}_j^{(1)}\right|$$

Additional physics-informed MAE functions

(1) $$\frac{1}{BN_g}\sum_{i=1}^{B}\sum_{j=1}^{N_g}\left|\sum X_{g,j}^{(1)} - \sum \hat{X}_{g,j}^{(1)}\right|$$

(2) $$\frac{1}{BN_g}\sum_{i=1}^{B}\sum_{j=1}^{N_g}\left|\frac{X_{g,j}^{(1)}}{dt} - \frac{\hat{X}_{g,j}^{(1)}}{dt}\right|$$

(3) $$\frac{1}{BN_l}\sum_{i=1}^{B}\sum_{j=1}^{N_l}\left|\sum X_{l,j}^{(1)} - \sum \hat{X}_{l,j}^{(1)}\right|$$

Model inference in comparison to CFD results

Performance of isolated ML model compared to previous MFiX results

- 1-kg single HDPE Particle
- Initial Particle Temperature: 650 K
- Heating Rates of 5, 10, 20 K/min
- Time step: $1x10^{-3}$ s

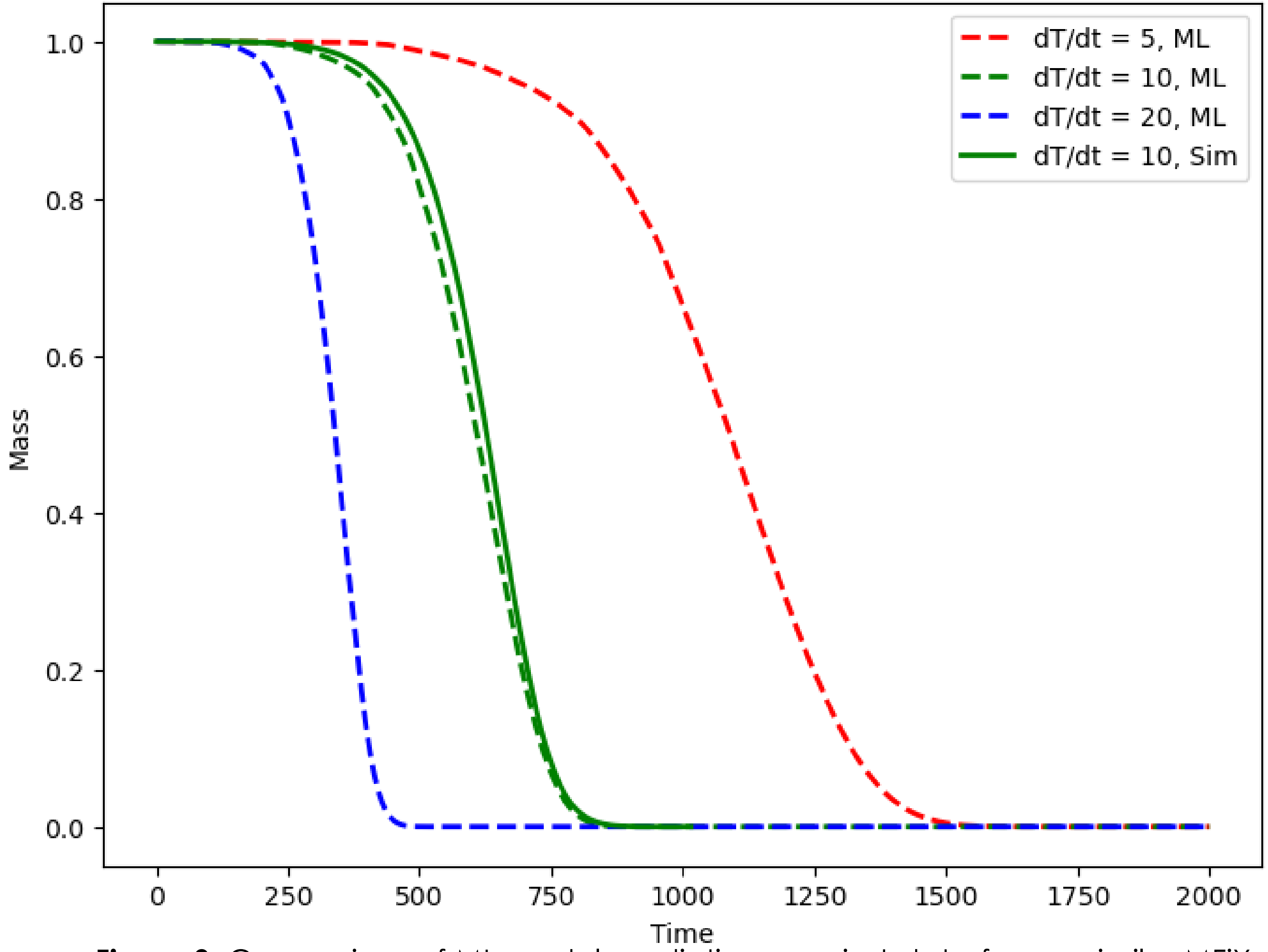


Figure 3. Comparison of ML model predictions against data from a similar MFiX single particle simulation.

Model inference in comparison to CFD results

NATIONAL ENERGY TECHNOLOGY LABORATORY

Performance of isolated ML model compared to previous MFiX results

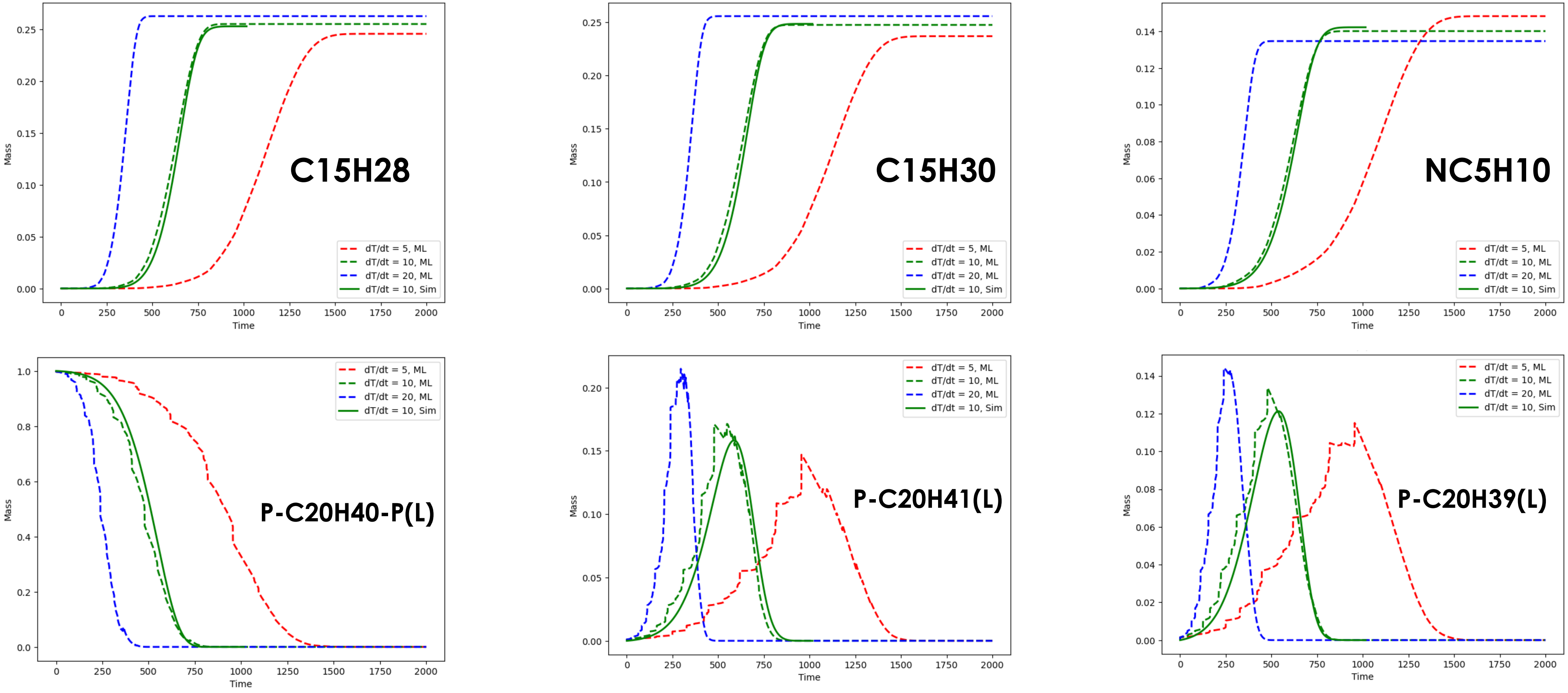
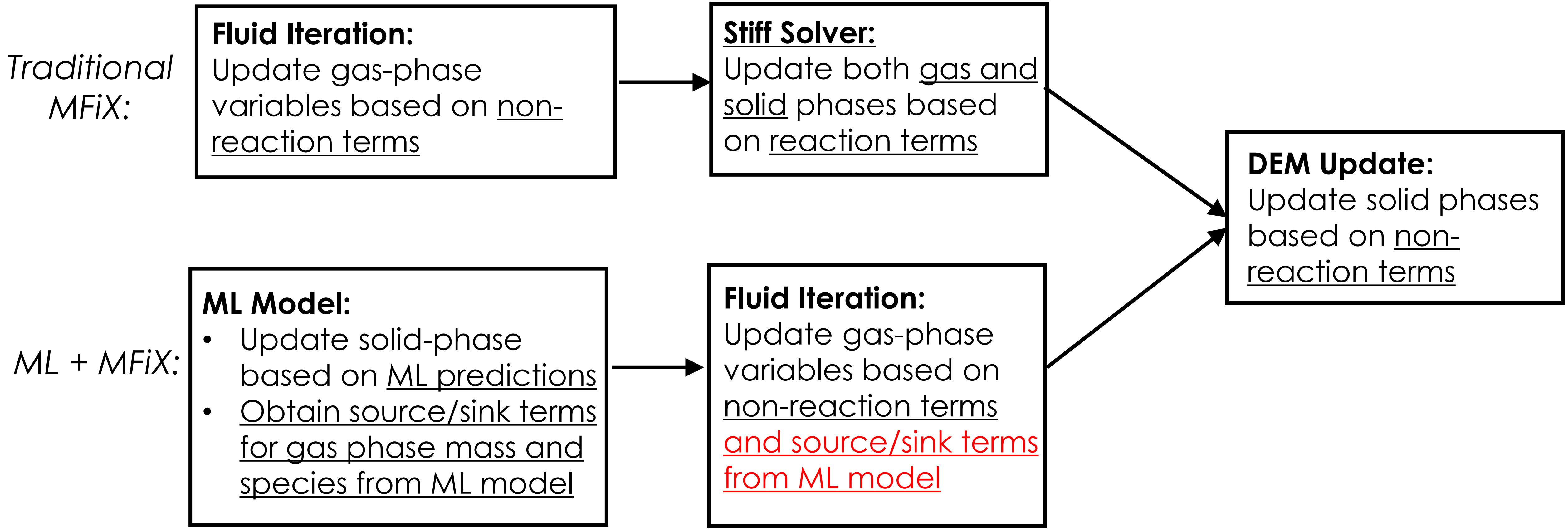


Figure 4. Comparison of ML model predictions against data from a similar MFiX single particle simulation for the three largest species for each phase.

Implementation of Neural Network into MFiX

Replace conventional stiff chemistry solver with the ML model



ML model is integrated into MFiX to replace the stiff solver for reacting terms

- Solid phases from reactions are predicted directly by ML model
- Source/sink terms for gas phases are predicted from ML model and used in the following fluid iteration

Scheme provided by Hang Zhou.

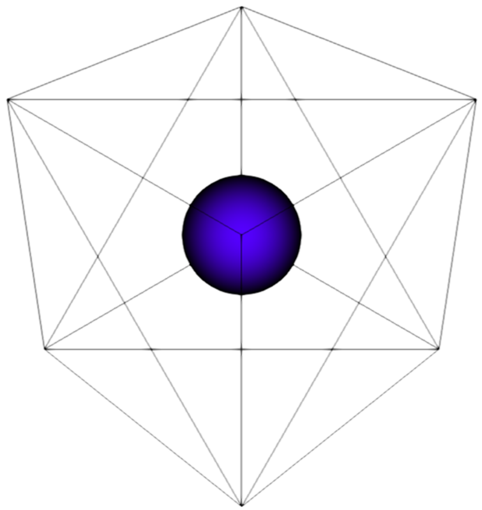
Performance in a Single-Particle Simulation

Testing kinetic performance in an ideal environment

- Single, 1-kg HDPE particle
- Fixed temperature ramp
- 10 K/min
- Full particle conversion at 1600 s
- Test of kinetic performance

ML Performance:

- Error of particle mass (w.r.t initial mass):
  - 0.8% average
  - 9.1 % max
- Error during conversion (1100 – 1500 s):
  - 3.2% average
  - 9.0 % max



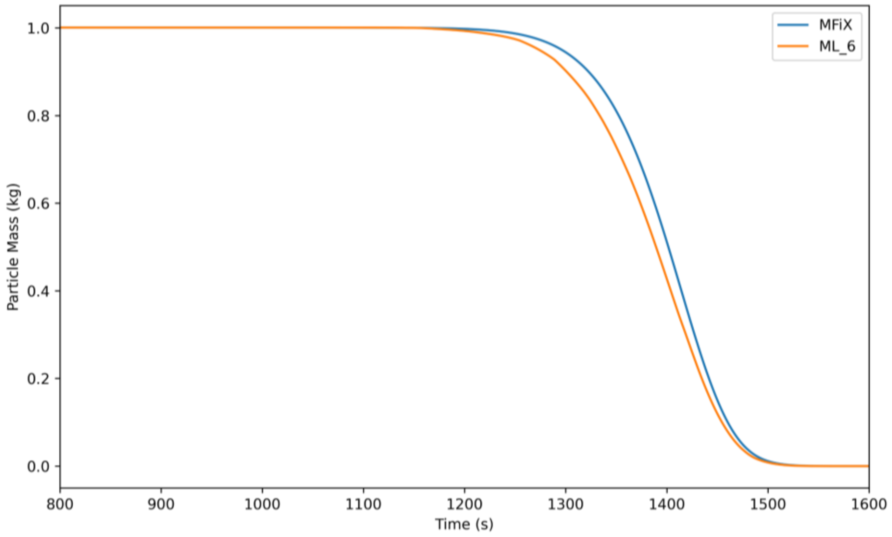


Figure 5. Top) Geometry setup of the MFiX simulation and bottom) prediction of particle mass loss compared to the stiff-solver.

Performance in a Single-Particle Simulation

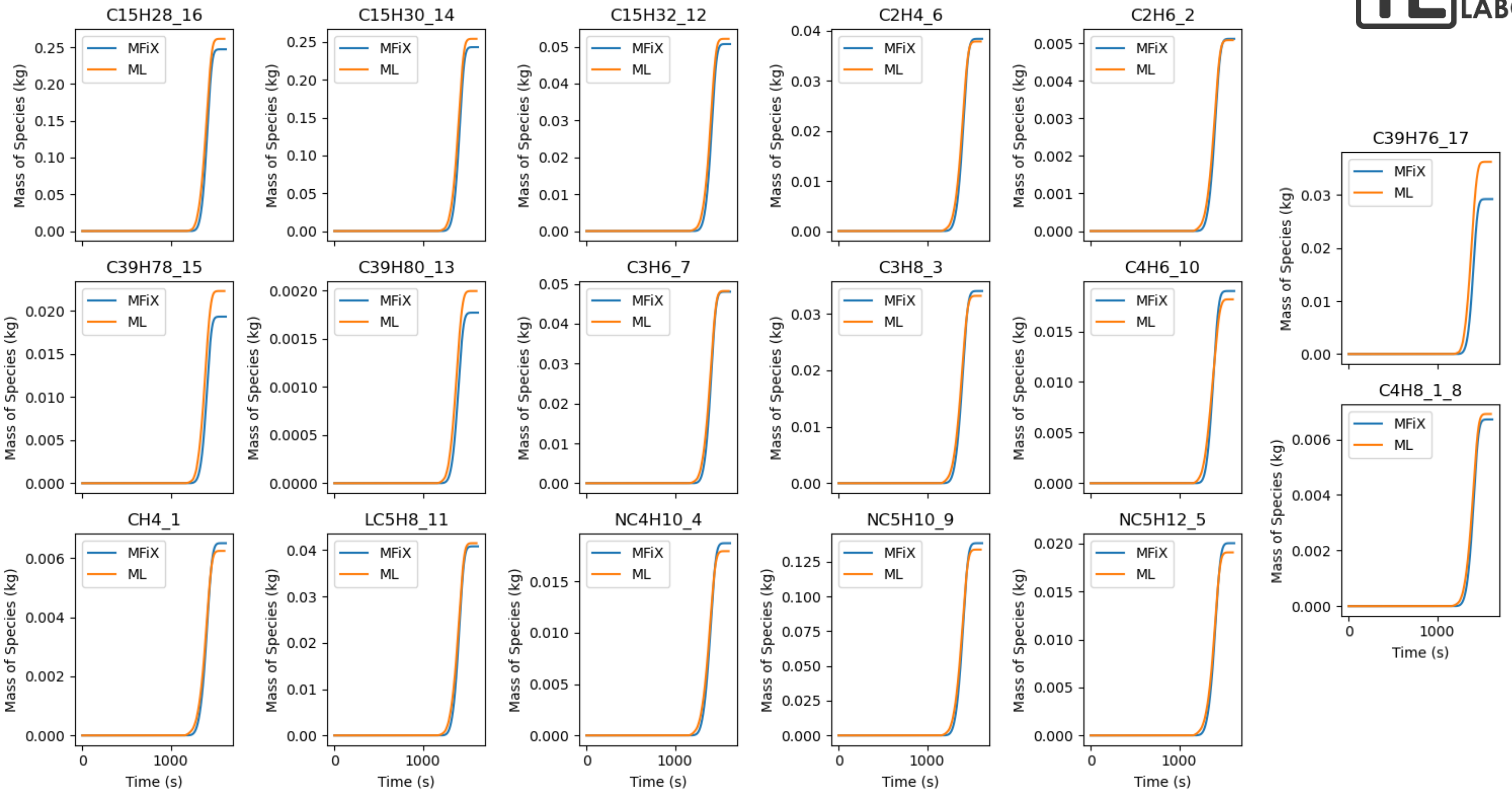


Figure 6. Comparison of ML model predictions against data from a similar MFiX single particle simulation for gas products.

Performance in a Single-Particle Simulation

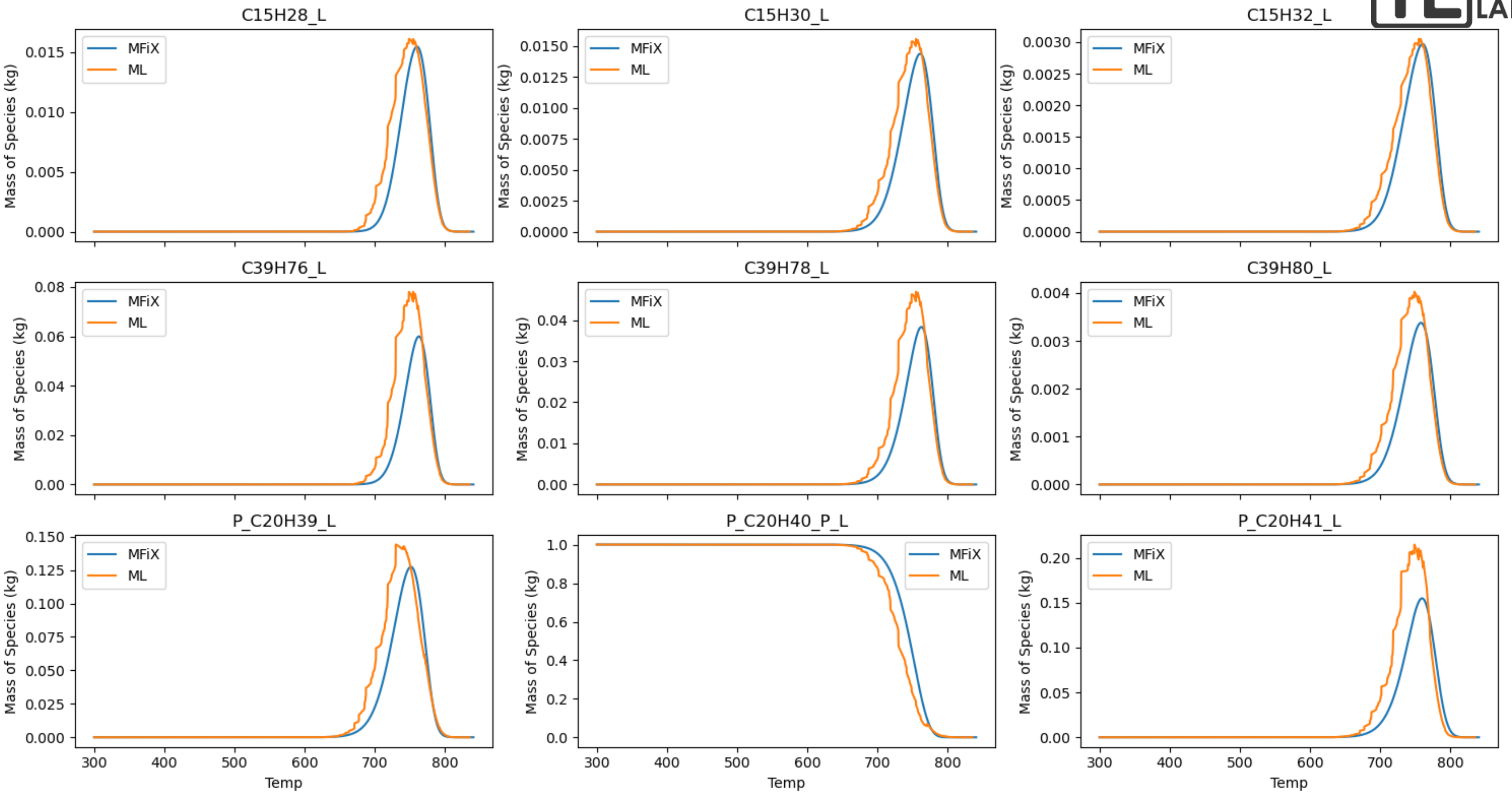


Figure 7. Comparison of ML model predictions against data from a similar MFiX single particle simulation for liquid plastic species.

Future Work

- Optimize the current NN to improve predictions
- Increase the breadth of our training data
  - More heating rates for data generations
- Implement our ML model into full scale simulations
  - Fluidized bed reactors
- Adapt similar strategies for developing a NN for large secondary gasification schemes
- Performed detailed simulations with all kinetics are solved via ML models while achieving the same level of accuracy

Thank you for your attention

VISIT US AT: www.NETL.DOE.gov
www.mfix.netl.doe.gov

@NETL_DOE

@NETL_DOE

@NationalEnergyTechnologyLaboratory

CONTACT:

Ross Houston, Research General Engineer

Ross.Houston@netl.doe.gov

O: (304) 285-0284

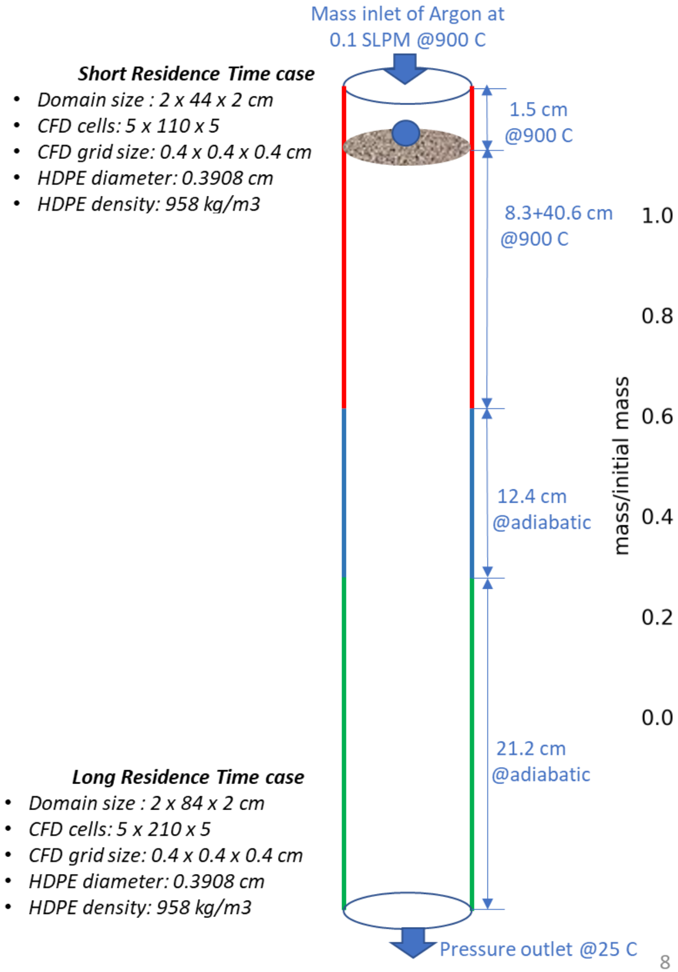
U.S. DEPARTMENT OF ENERGY

Performance in a Drop Tube Reactor

- Experimental drop tube reactor at NETL
- Currently used as a source of validation for HDPE kinetics
- Higher heating rate
  - ~23 K/s
- Full particle conversion at 20 s

ML Performance:

- ML was **25%** faster than stiff-solver
- Error of particle mass:
  - 2.3% average
  - 26 % max



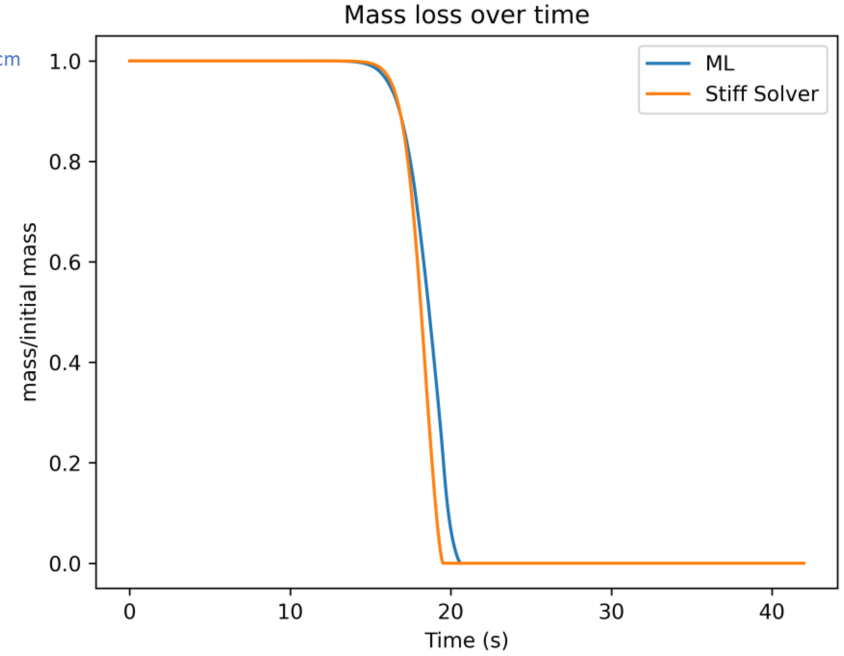


Figure 8. Left) Drop tube geometry and right) mass loss prediction for particle in a reactor for ML and stiff-solver

Simulation settings in MFiX