Implementation of Detailed Polyethylene Pyrolysis Kinetics into CFD Simulations using Machine Learning

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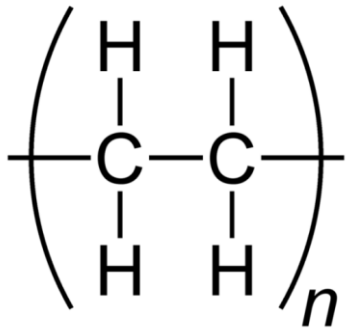
Legal Disclaimer

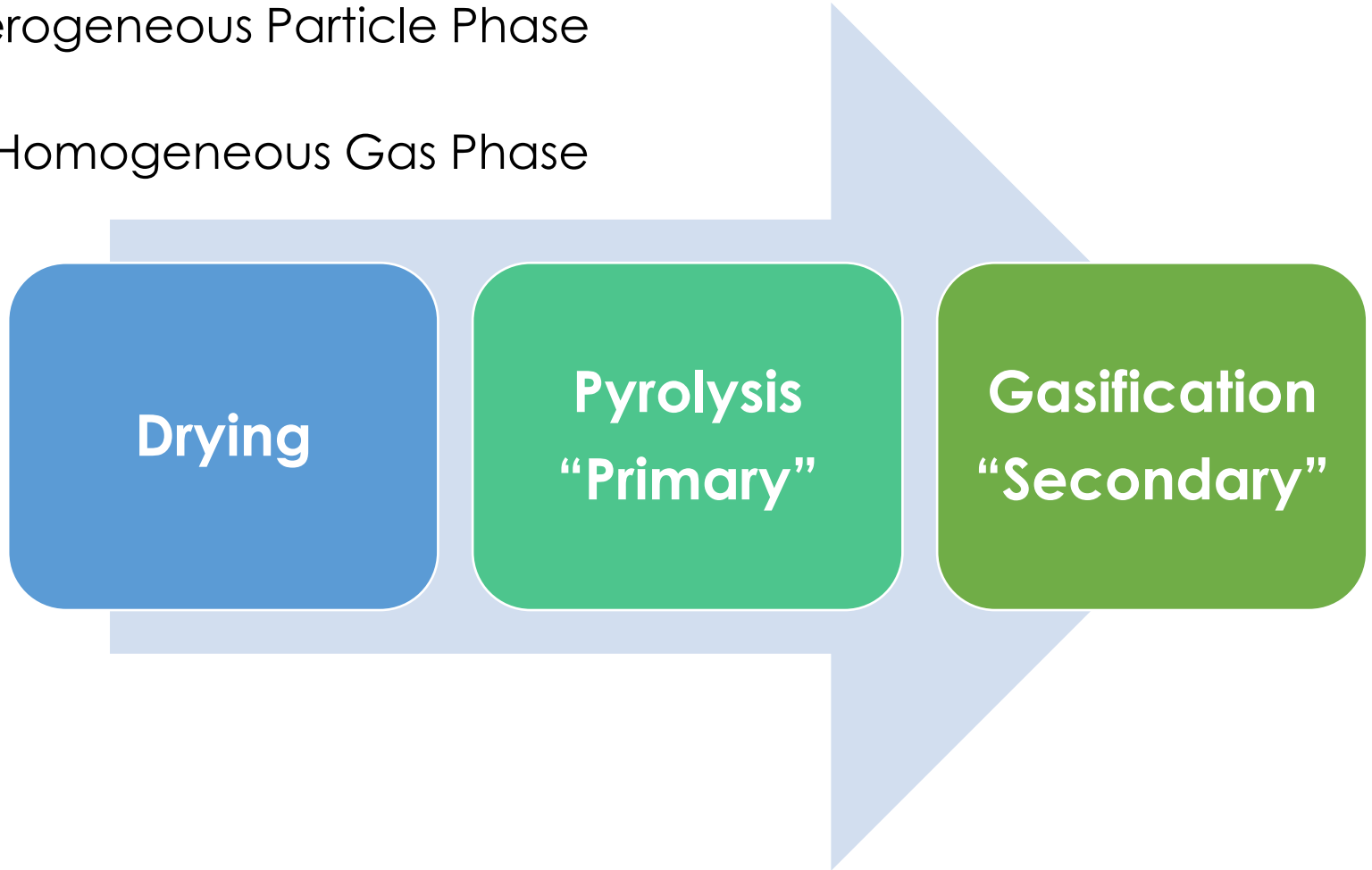
Gasification of high-density polyethylene (HDPE)

Necessary to subclassify gasification for accurate kinetic modeling

- Reaction Subclasses:
  - Pyrolysis – Heterogeneous Particle Phase
  - Gasification – Homogeneous Gas Phase



HDPE



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

Current state of HDPE pyrolysis kinetics

Collaboration with CRECK Modeling Group at Politecnico di Milano

- 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

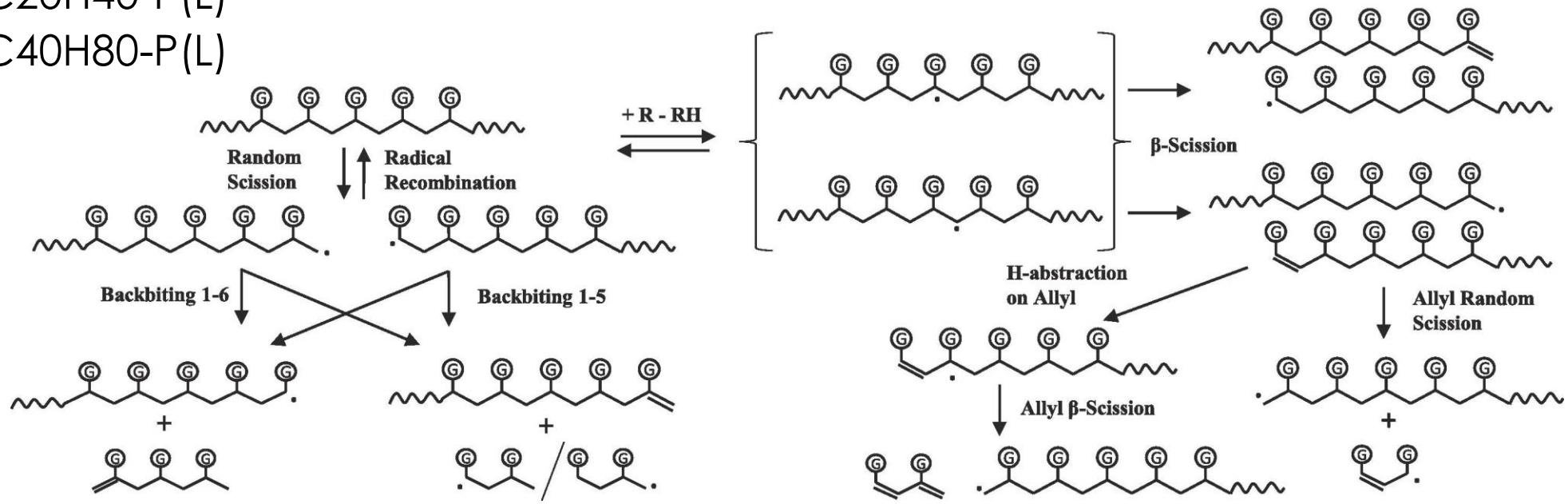


Image credit: A. Locaspi, et al., *Waste Management* **156** (2023) 107-117

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 Primary Reaction Schemes:

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

*Focus of ML modeling work

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

	Paraffin	**Olefin**
Mid-Chain (MC)	P-$C_{20}H_{40}$-P(L) P-$C_{40}H_{80}$-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)

Machine learning (ML) approach for HPDE reaction kinetics

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

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

- 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

- 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 operating conditions
- 4+ million data points

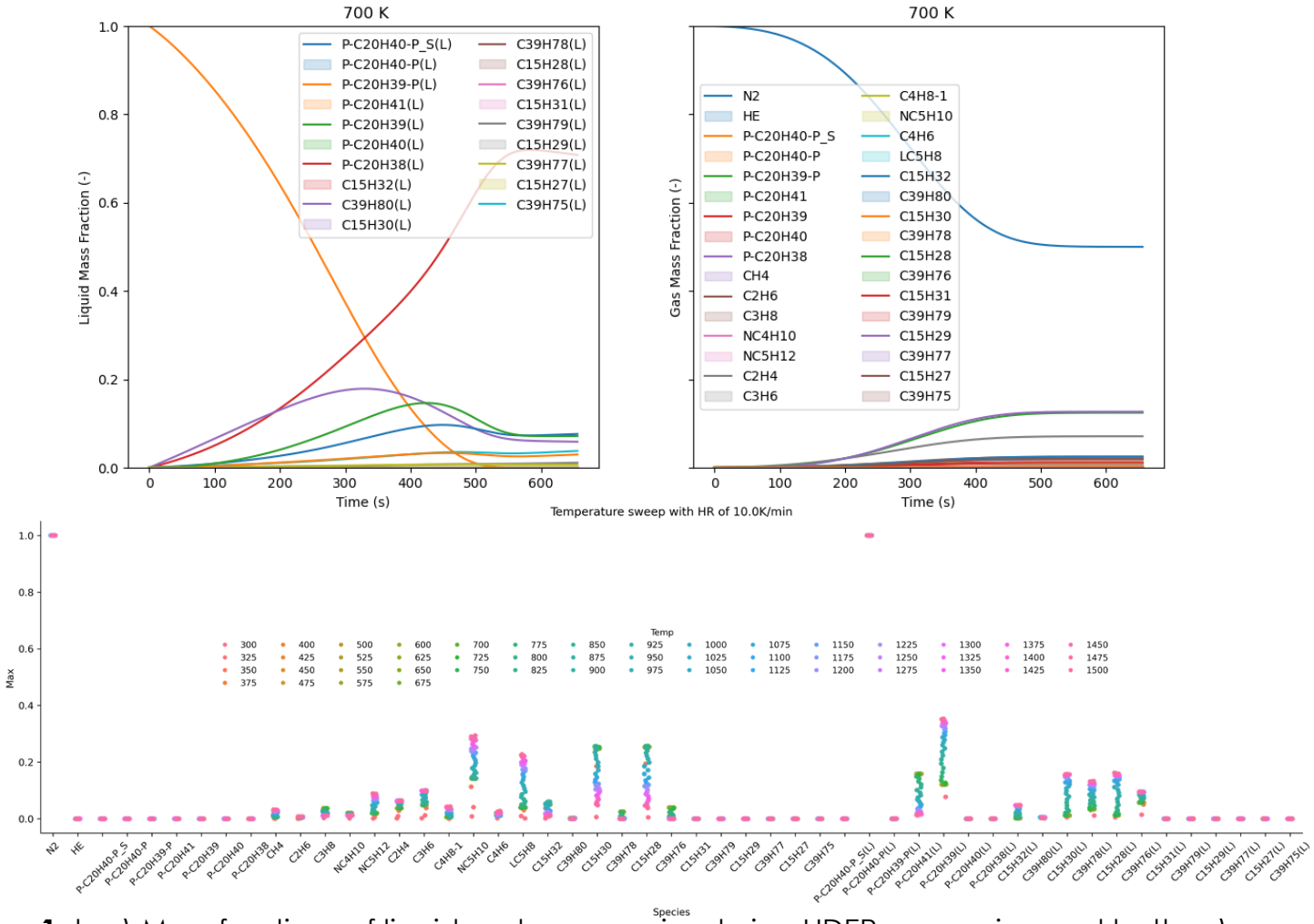


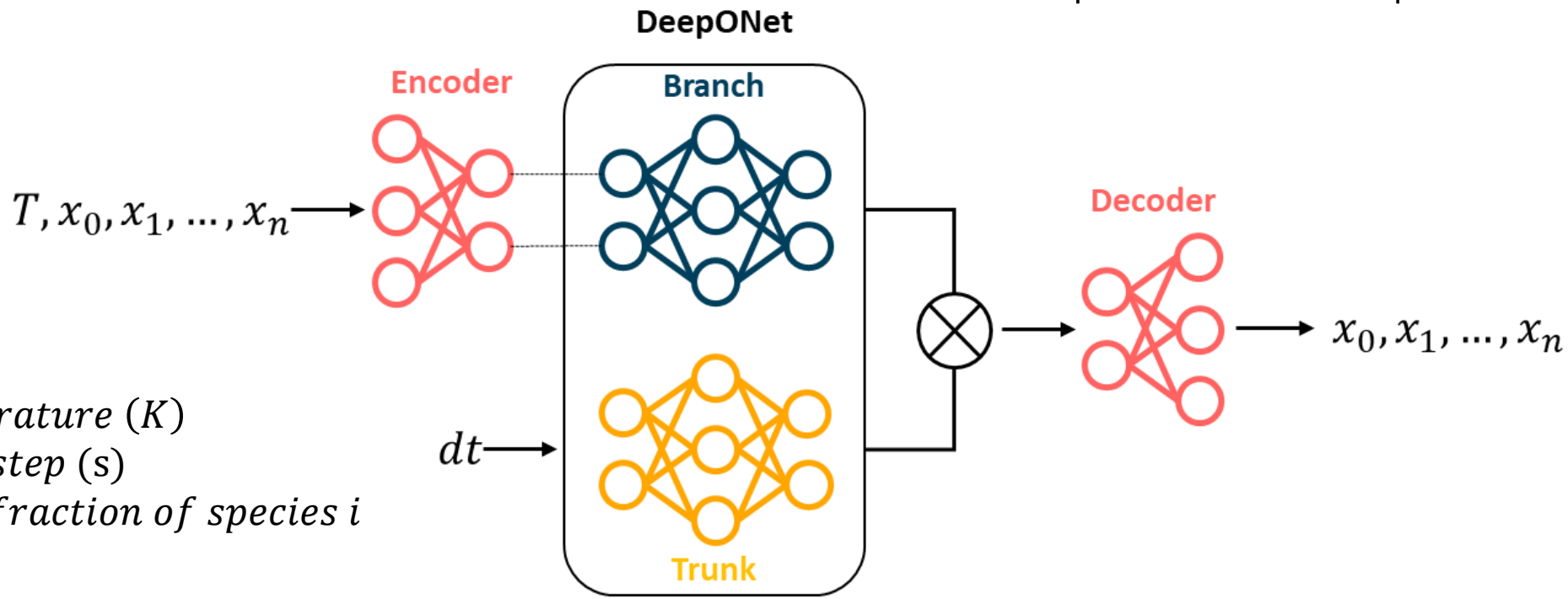
Figure 1. top) Mass fractions of liquid and gas species during HDEP conversion and bottom) maximum mass of each species during conversion over a range of temperatures using a 1-kg basis.

Architecture of the ML model

Implementation of DeepONet structure with physics-based 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



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

Physics-based loss functions

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

- Training and validation loss calculated using the mean absolute error (MAE)
- Chemical reactions must conserve mass
- Mass conservation can inform ML training
- 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) $$\frac{1}{BN_l}\sum_{i=1}^{B}\sum_{j=1}^{N_l}\left|\sum X_{l,j}^{(1)} - \sum \hat{X}_{l,j}^{(1)}\right|$$

Predictions against training data

Parity plots for the major gas and liquid species – HDPE 42_737 scheme

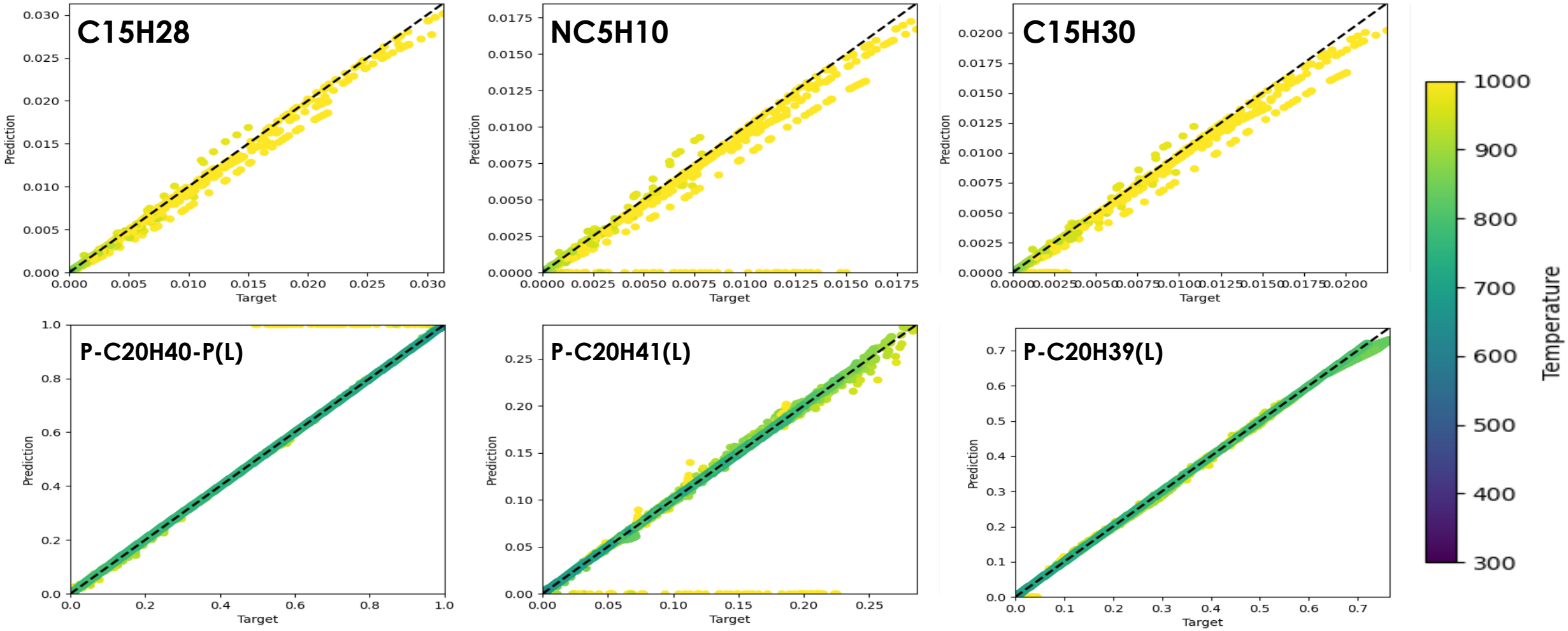


Figure 2. Comparison of ML model predictions against our training data. Note: Temperature is in Kelvin (K).

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

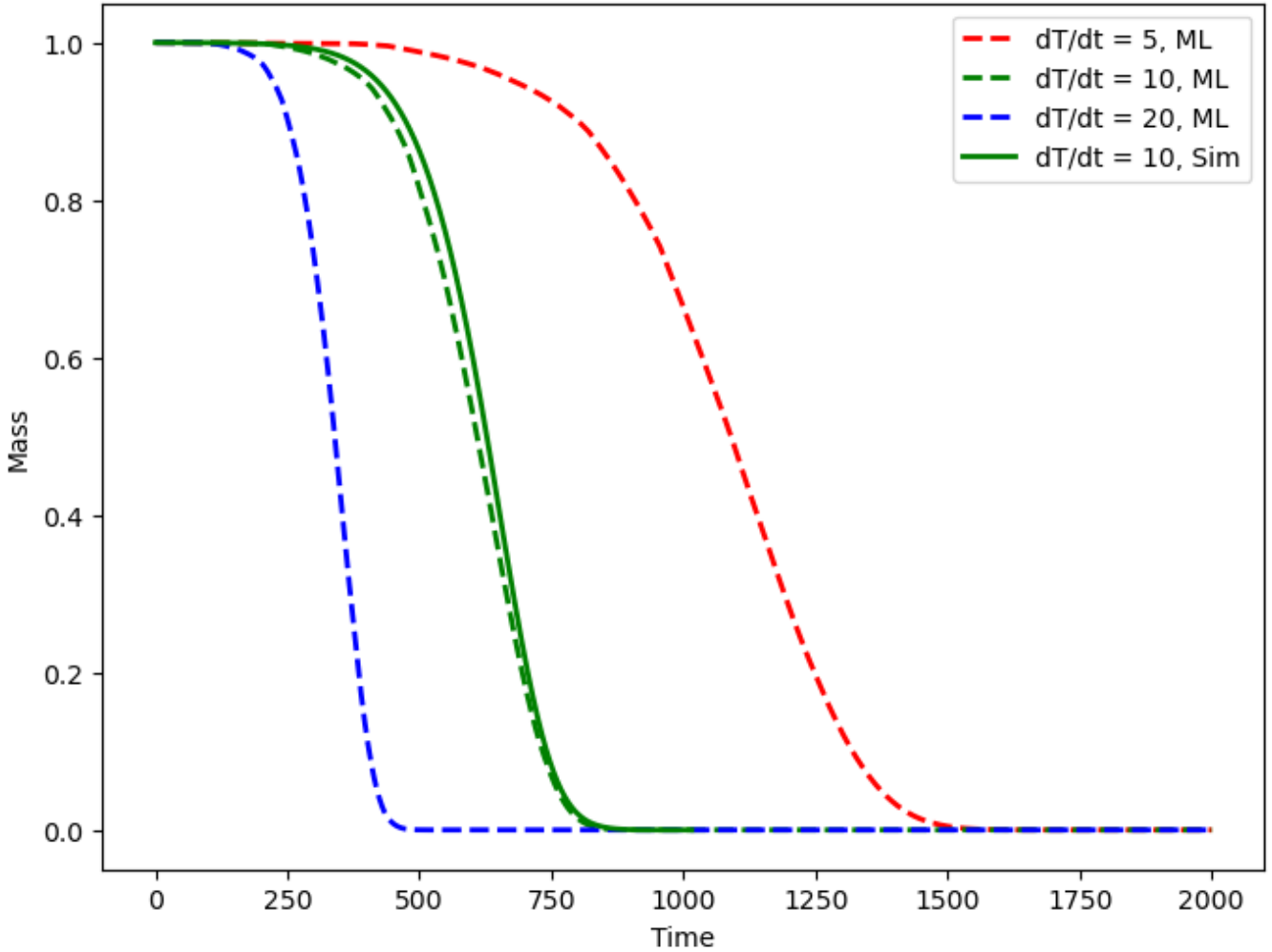


Figure 3. Comparison of isolated ML model predictions against data from a similar MFiX single particle simulation.

Model inference in comparison to CFD results

Performance of isolated ML model compared to previous MFiX results

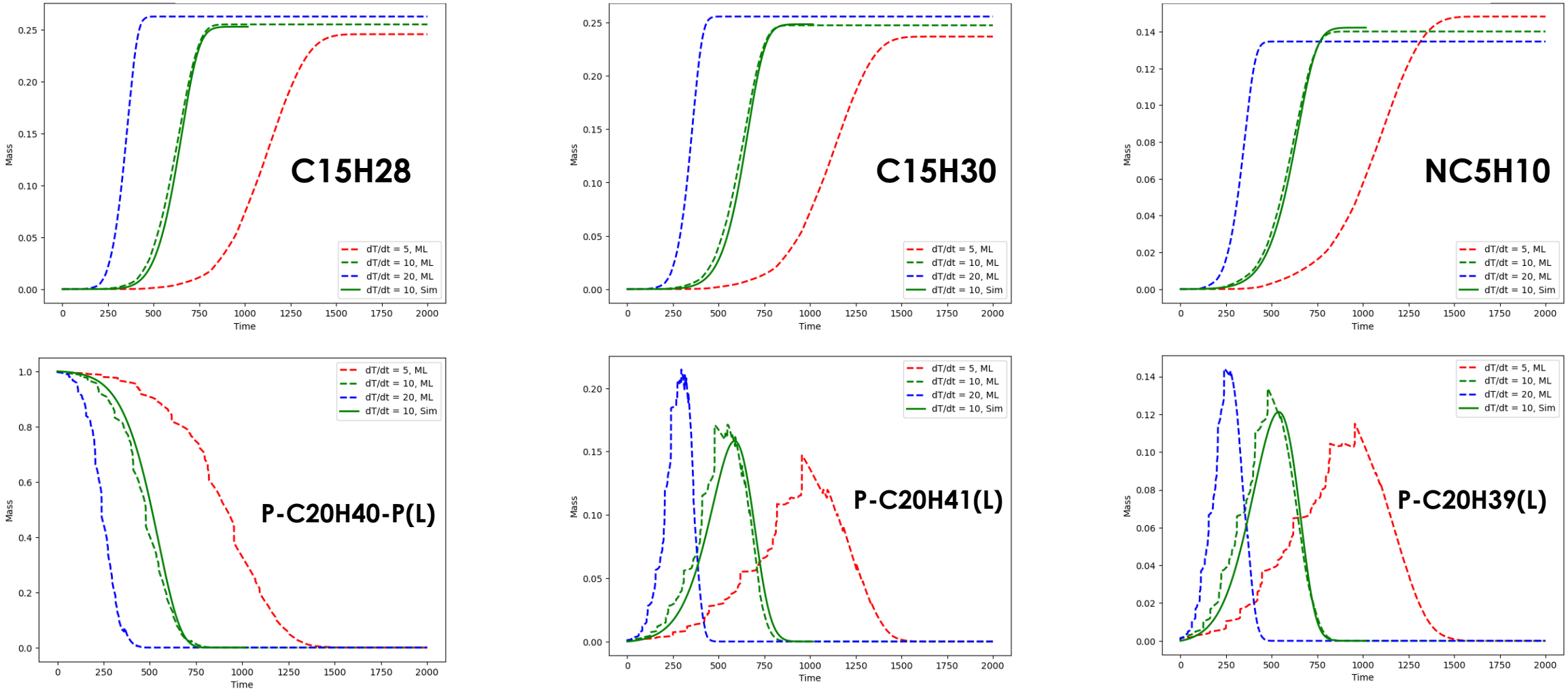


Figure 4. Comparison of isolated ML model predictions against data from a similar MFiX single particle simulation for the three largest species for each phase.

Implementation of Neural Network into MFiX

Replace conventional stiff chemistry solver with the ML model

Traditional MFiX:

Fluid Iteration:
Update gas-phase variables based on non-reaction terms

→

Stiff Solver:
Update both gas and solid phases based on reaction terms

ML + MFiX:

ML Model:
- Update solid-phase based on ML predictions
- Obtain source/sink terms for gas phase mass and species from ML model

→

Fluid Iteration:
Update gas-phase variables based on non-reaction terms and source/sink terms from ML model

→

DEM Update:
Update solid phases based on non-reaction terms

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

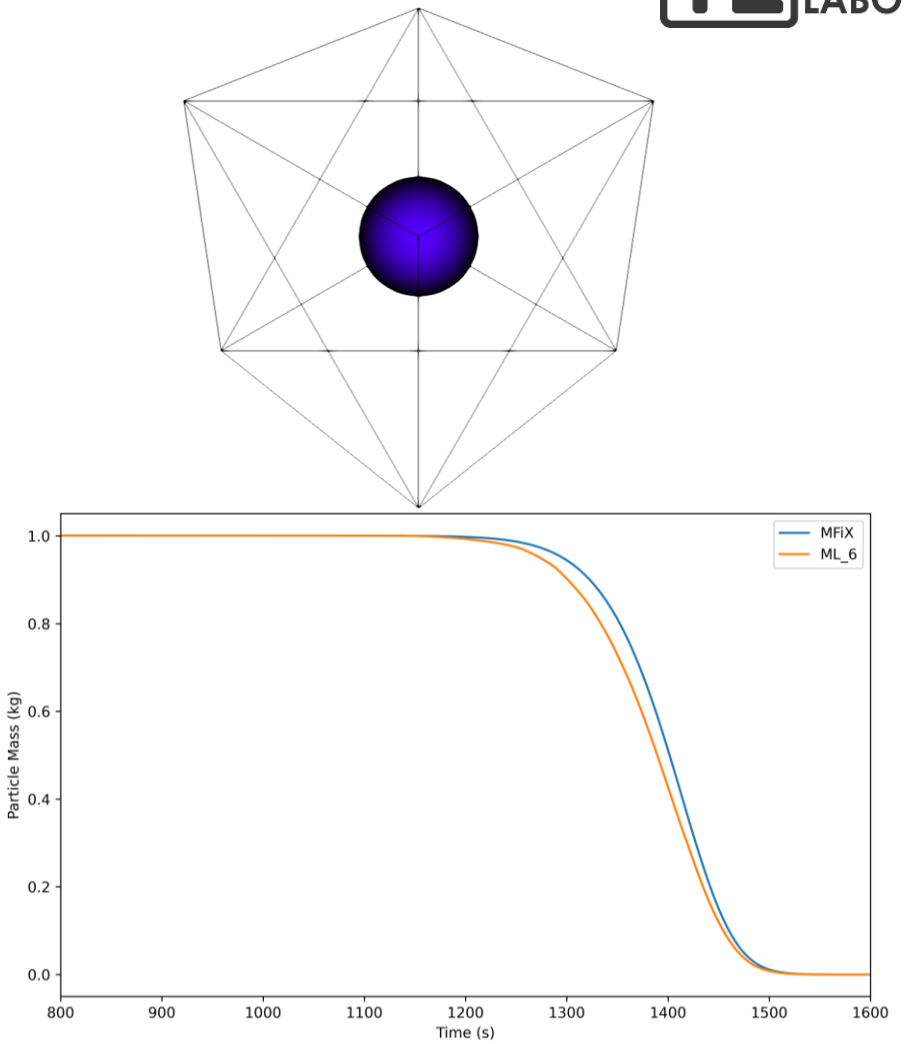


Figure 6. Top) Geometry setup of the MFiX simulation and bottom) prediction of particle mass loss compared to the stiff-solver.

Performance in a Single-Particle Simulation

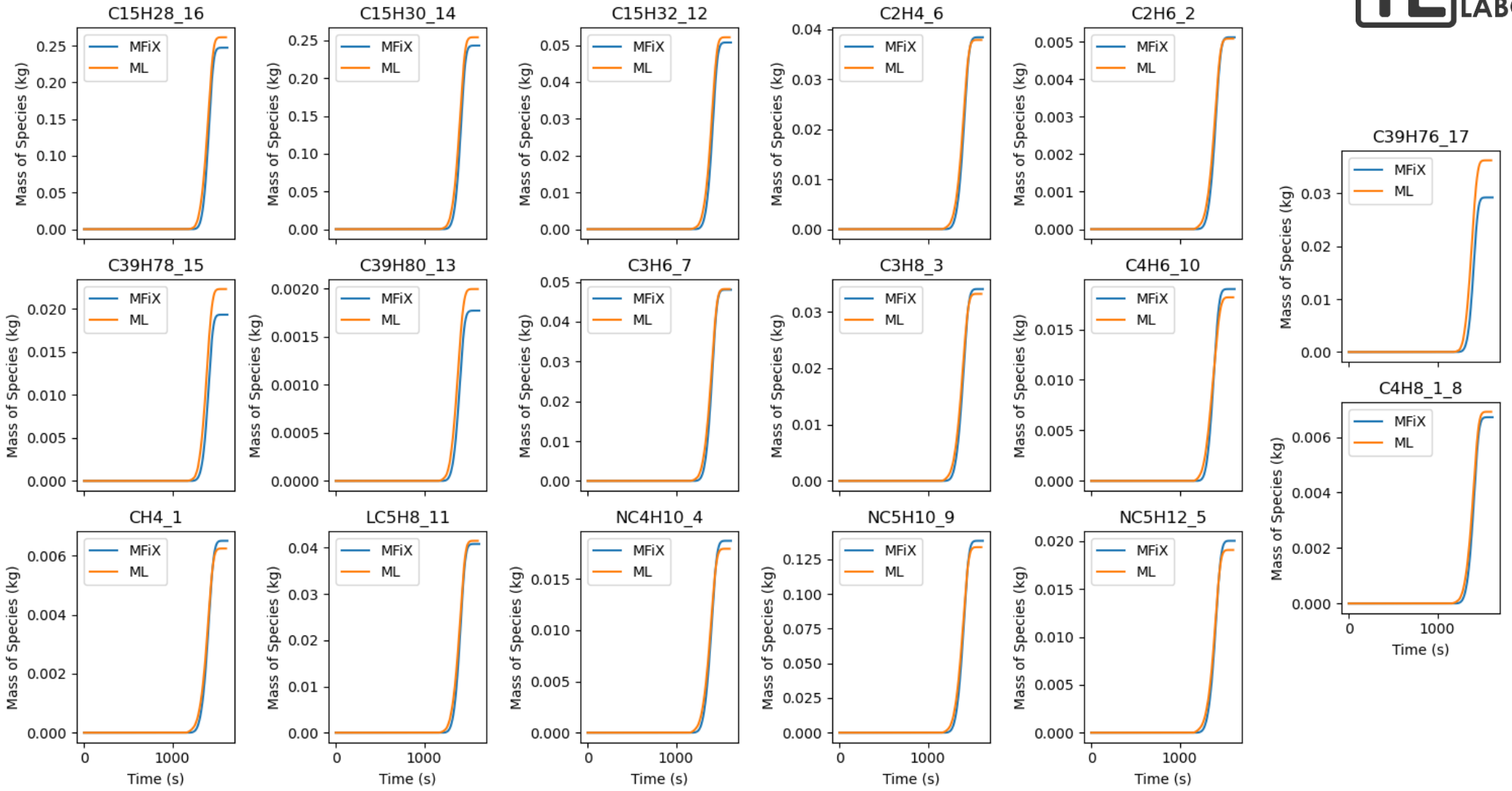


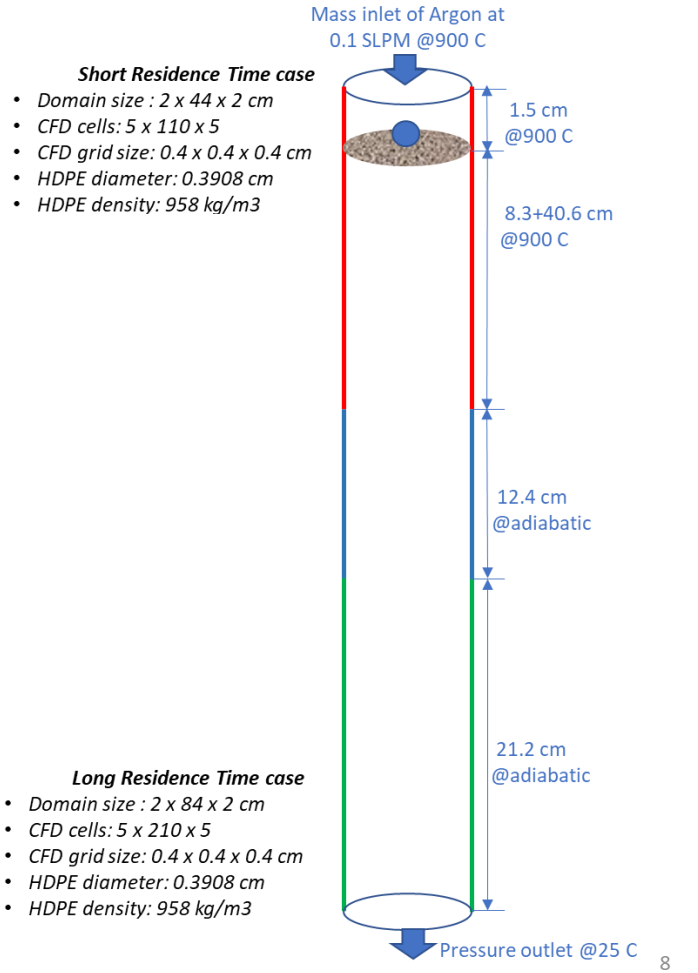
Figure 7. Comparison of gaseous product species between traditional the traditional stiff-solver and the ML implementation in MFiX.

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



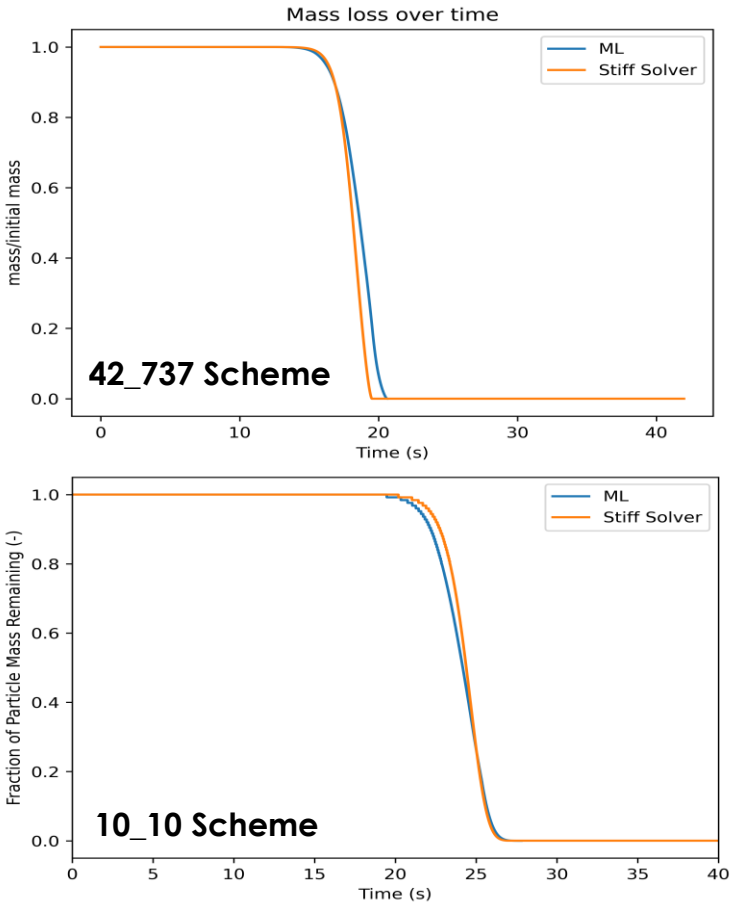


Figure 8. Left) Drop tube geometry and right) mass loss prediction for particle in a droptube reactor for ML and stiff-solver

Simulation settings in MFiX

Future Work

- Optimize the current NN to improve predictions
- Implement ML model into full scale simulations
  - Fluidized bed reactors
- Adapt similar strategies for developing a NN for large secondary gasification schemes
- Perform detailed simulations with all kinetics (primary and secondary reactions) solved via ML models while achieving the same level of accuracy

Thank you for your attention

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