Determination of polyethylene (PE) and polypropylene (PP) content in post-consumer recycled flexible plastics using machine learning assisted differential scanning calorimetry (DSC)

Amir Bashirgonbadi, Yannick Ureel, Laurens Delva, Kevin M. Van Geem, Kim Ragaert

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C-PlaNeT
CIRCULAR PLASTICS NETWORK FOR TRAINING

COMPOSITION OF RECYCLED FLEXIBLES


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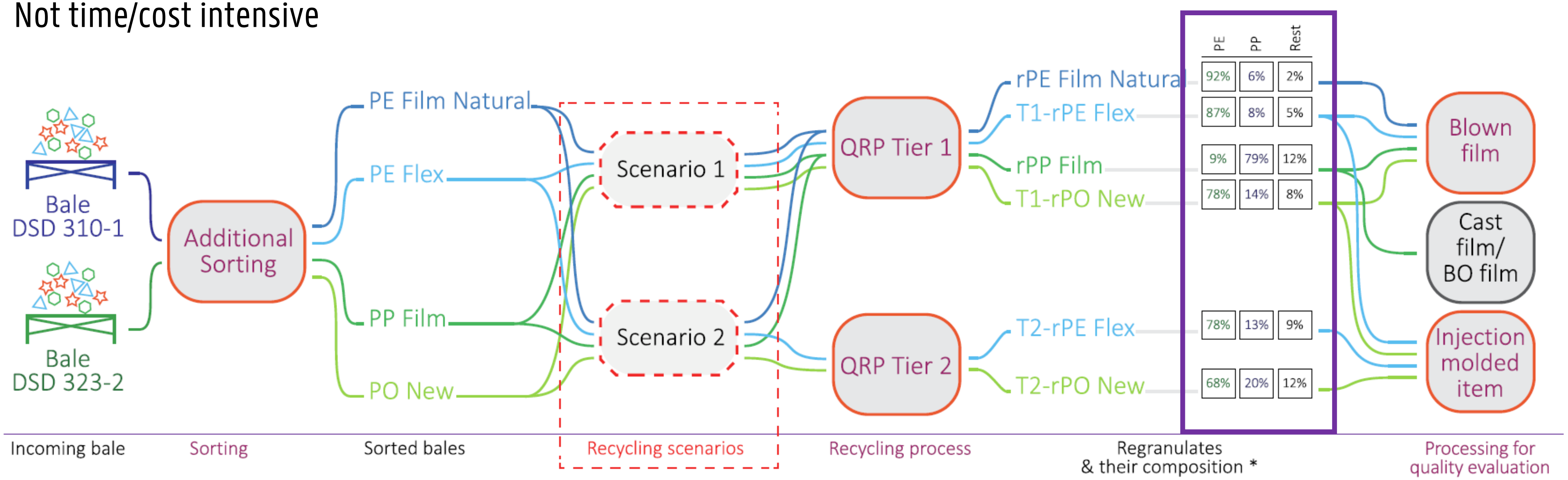
Quality evaluation and economic assessment of an improved mechanical recycling process for post-consumer flexible plastics

Amir Bashirgonbadi [a,c], Irdanto Saputra Lase [b], Laurens Delva [a], Kevin M. Van Geem [a], Steven De Meester [b,c], Kim Ragaert [c,*]

Cross contamination of PE and PP

A proper technique:

- Accurate
- Accessible
- Not time/cost intensive



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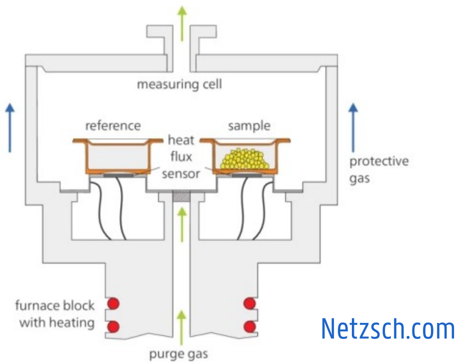
DETERMINATION OF CRYSTALLINITY/COMPOSITION

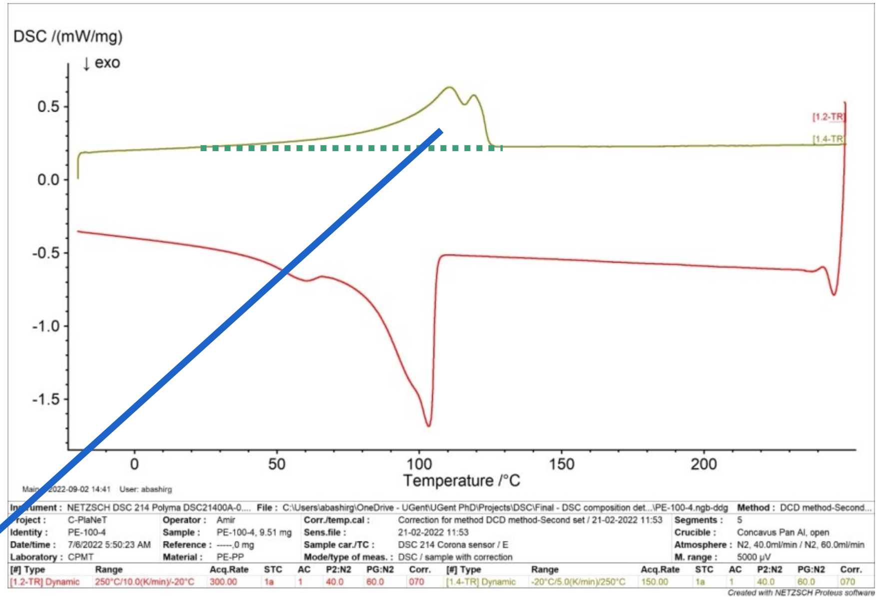
In a blend with a known composition:

$$\varphi_i = \frac{\Delta H_{m,i}}{\Delta H^0_{m,i} \times \%Xc,i} \times 100$$

- If we want to determine the composition in a blend, we should have a known (or a relatively accurate estimation of) crystallinity for each constituent.
- Remark: Crystallinity of each constituent changes with its content in the blend

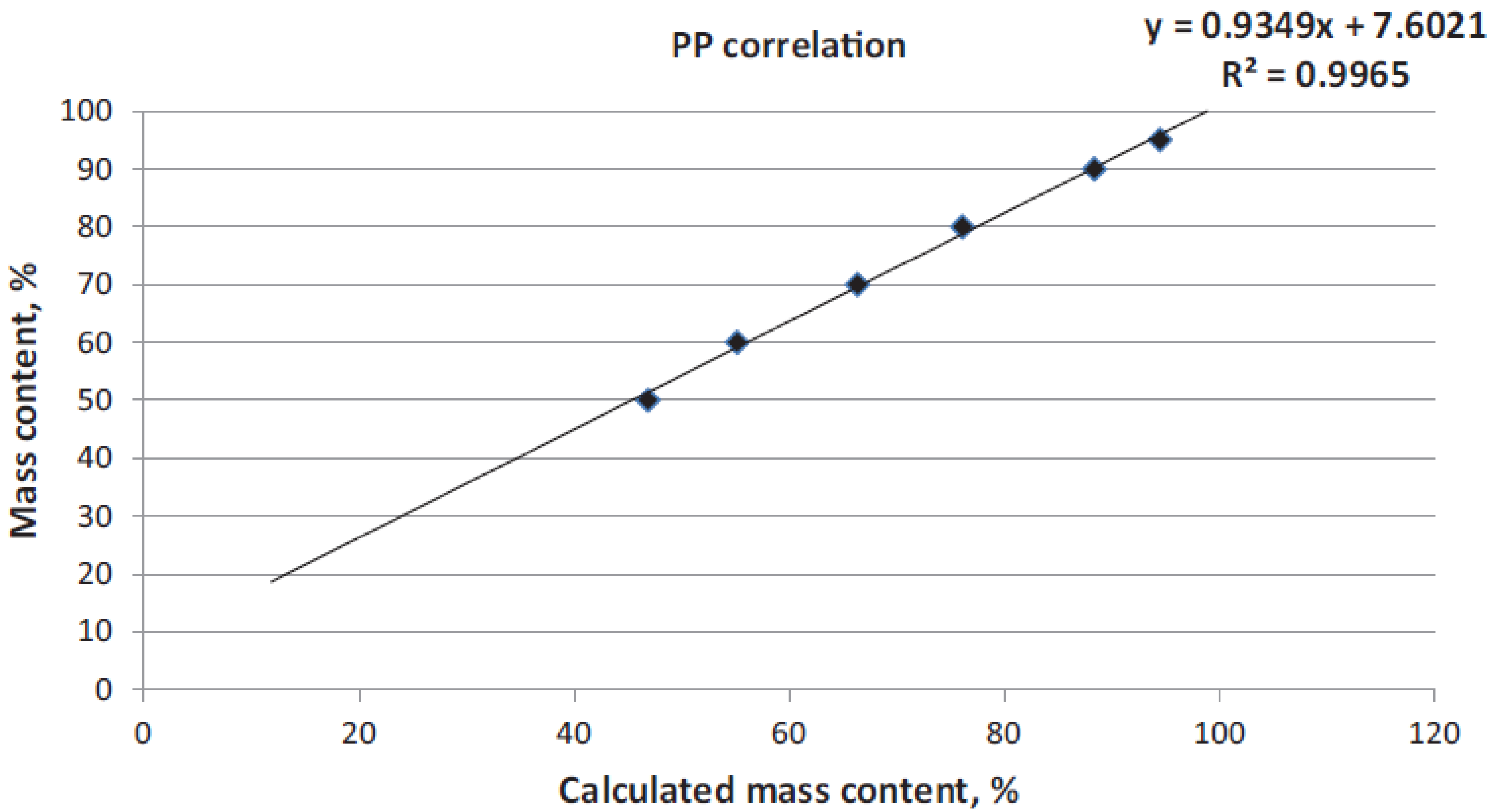


Netzsch.com



The enthalpy of fusion of a substance is a measure of the energy input, typically heat, which is necessary to convert a substance's crystals from solid to liquid state.

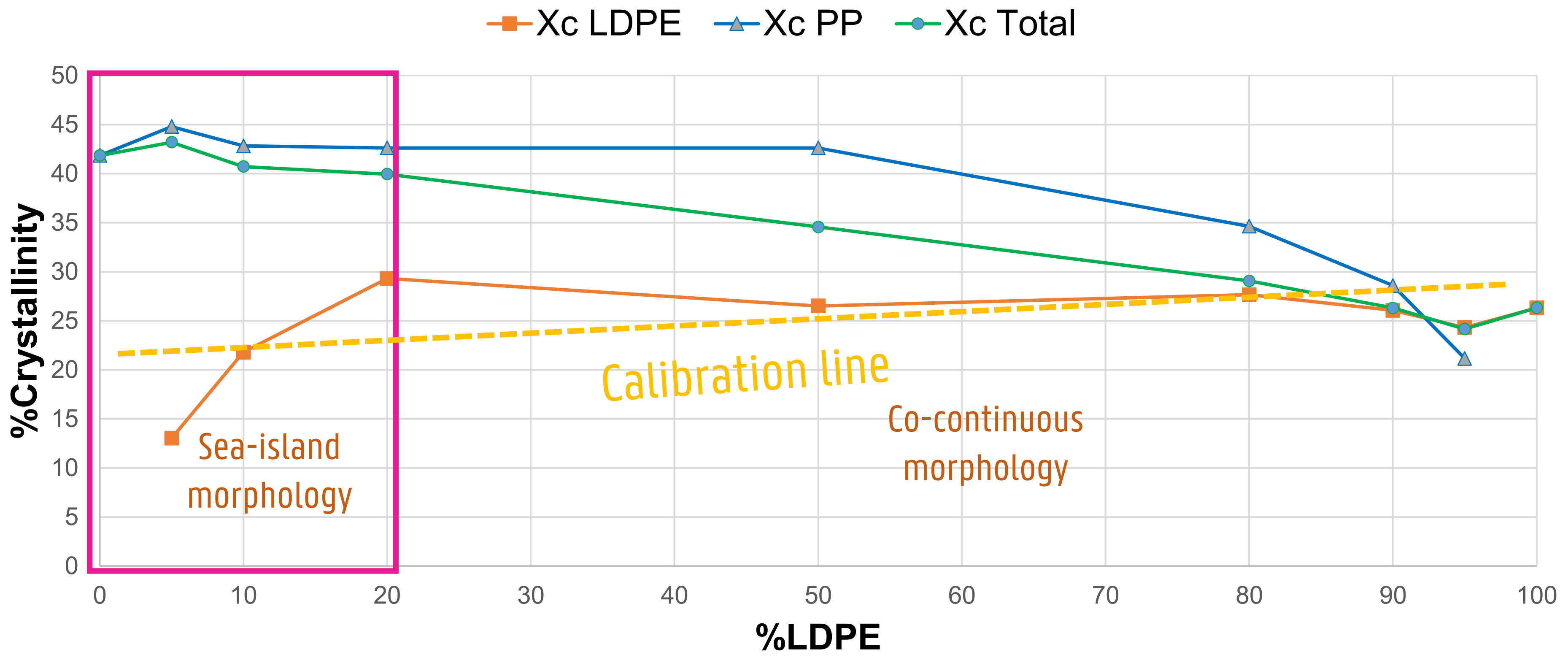
CALIBRATION LINES IN THE LITERATURE



Kisiel et al., 2018
https://journals.sagepub.com/doi/10.1177/1477760618797541

CRYSTALLINITY CHANGES AGAINST COMPOSITION-RQ DATA

- Co-continuous vs sea-island morphology
- For example, the crystallinity of LDPE+PP blends:



DEVELOPMENT OF A CALIBRATION CURVES

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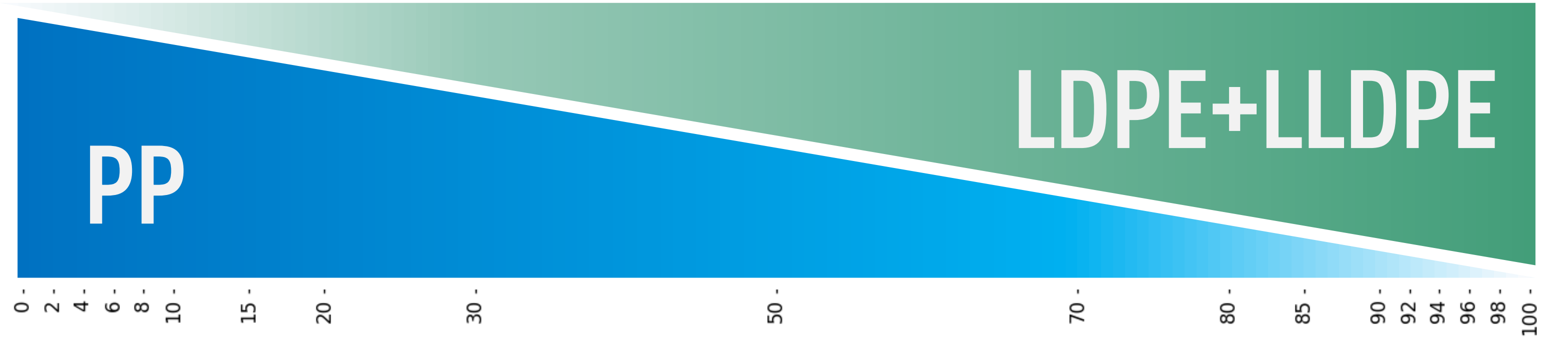
PREPARATION OF CALIBRATION BLENDS

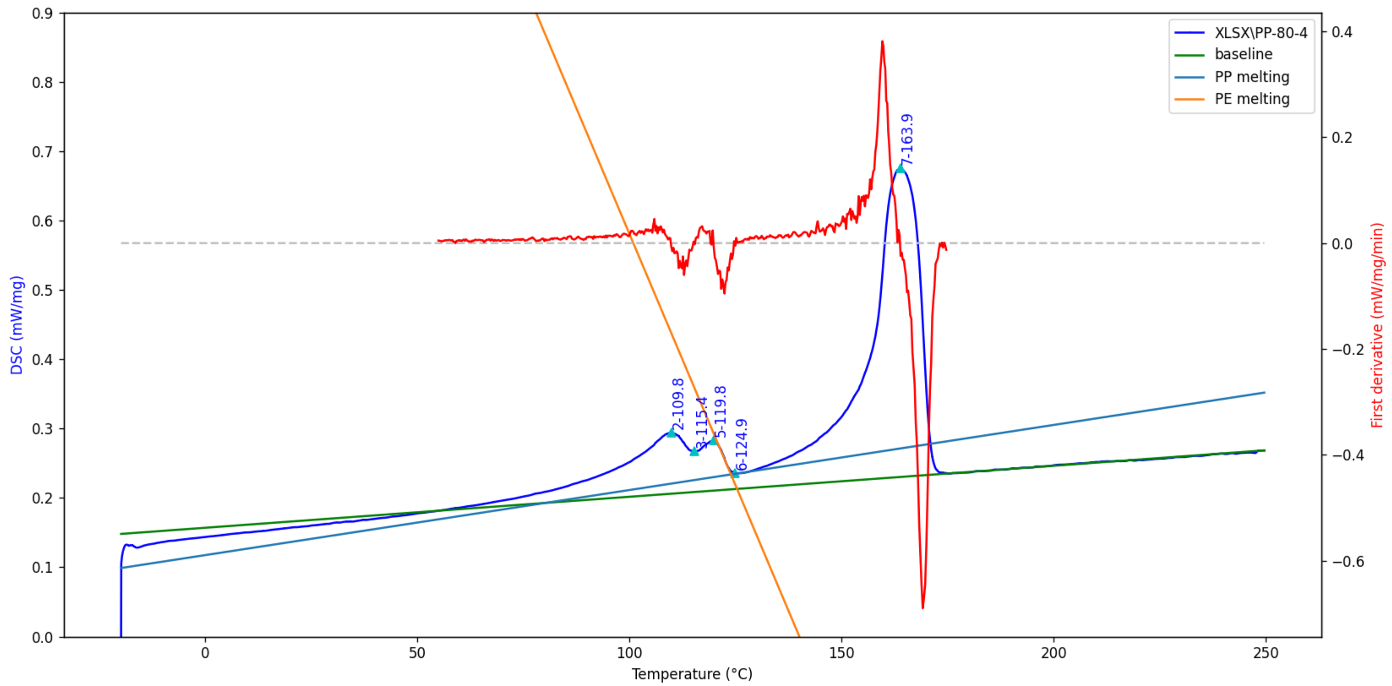
- Extrusion temperature: 210 °C (PE>70%), 230 °C (PE<70%)
- Screw speed: 100 rpm
- Residence time: 80 s
- Feeding amount: 2.8 g

- PE fraction: 50:50 blend of LDPE (i2= 1.0 dg/min) and LLDPE (i2= 0.9 dg/min) (both conventional film blowing grades)
- PP fraction: Homo PP (i2=3.0 dg/min) (conventional (biaxially) oriented PP film extrusion grade)
- 19 compositions, 3 extrusions at each composition, 2 sets of blending

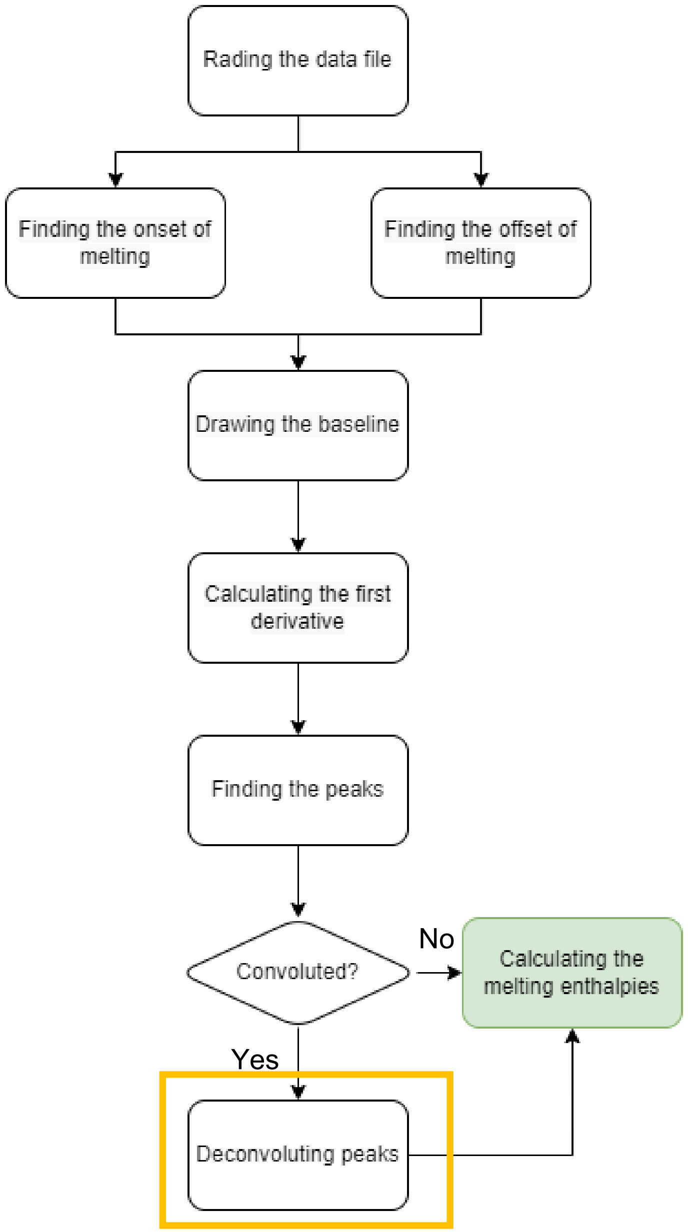
xplore-together.com



INTEGRATED DATA ANALYSIS TECHNIQUE



$$\varphi_i = \frac{\Delta H_{m,i}}{\Delta H^0_{m,i} \times \%Xc,i} \times 100$$



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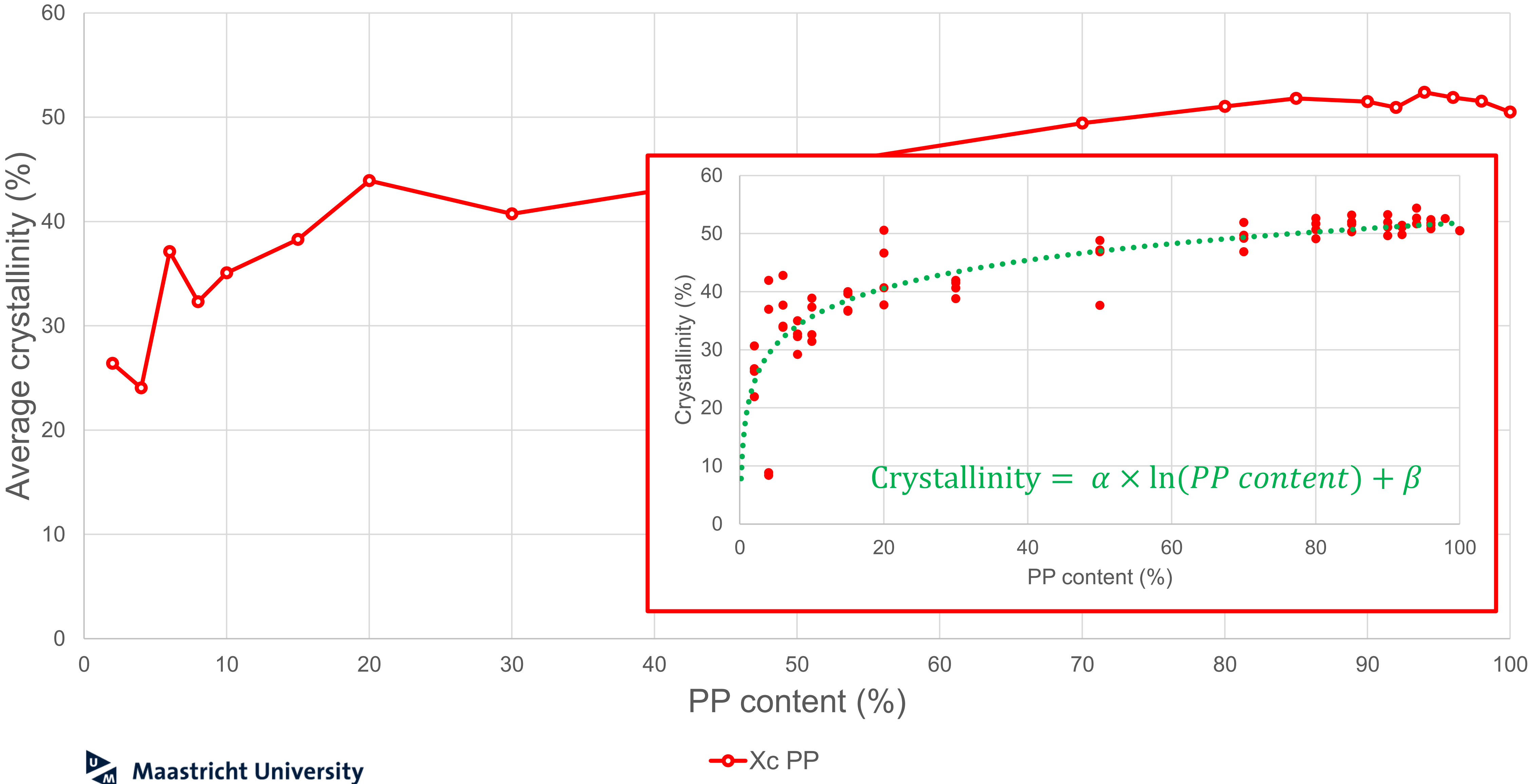
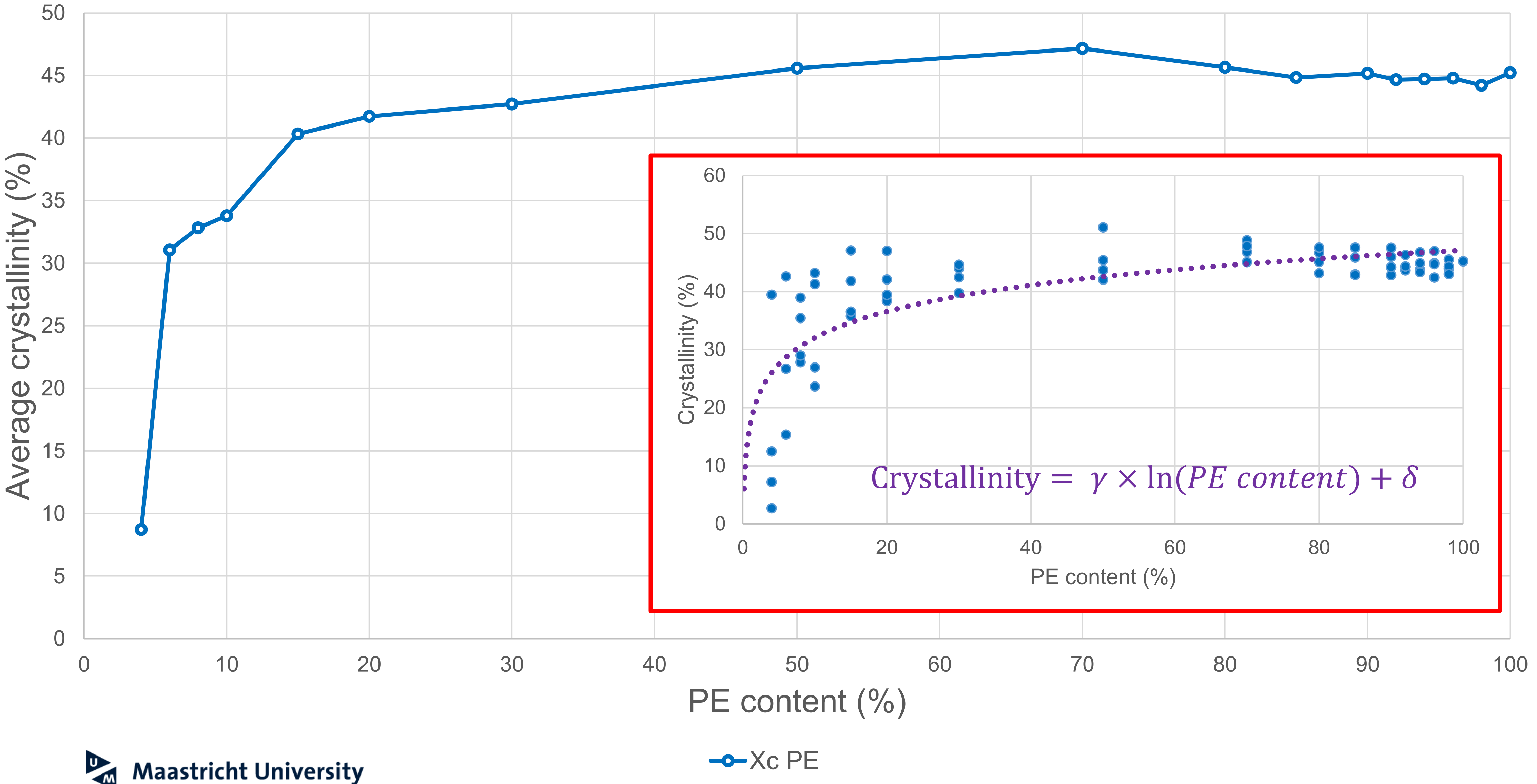
PP CRYSTALLINITY EVOLUTION AGAINST COMPOSITION



PE CRYSTALLINITY EVOLUTION AGAINST COMPOSITION



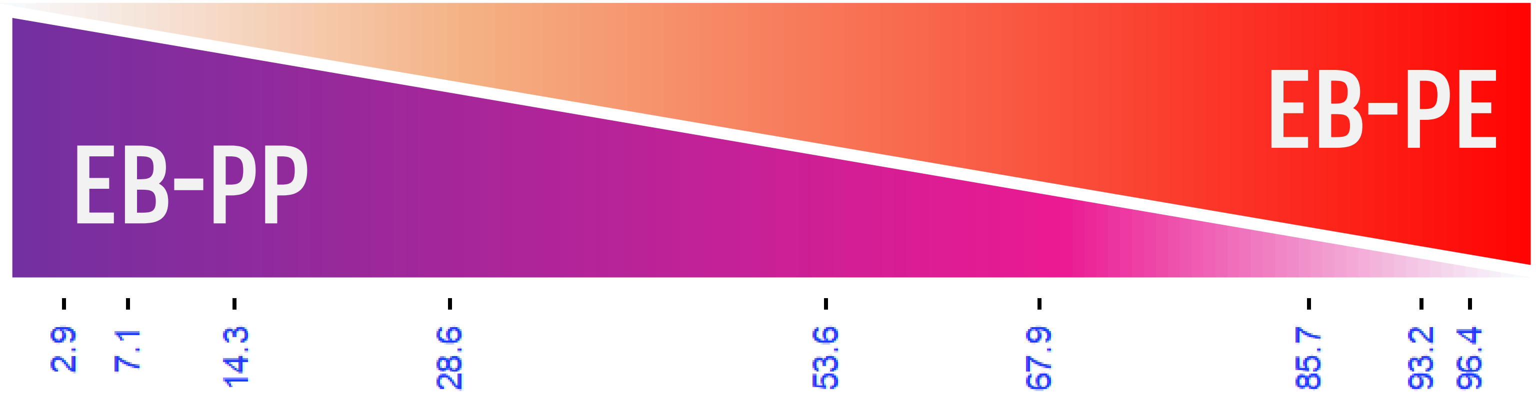
VALIDATION BLENDS

PREPARATION OF VALIDATION BLENDS

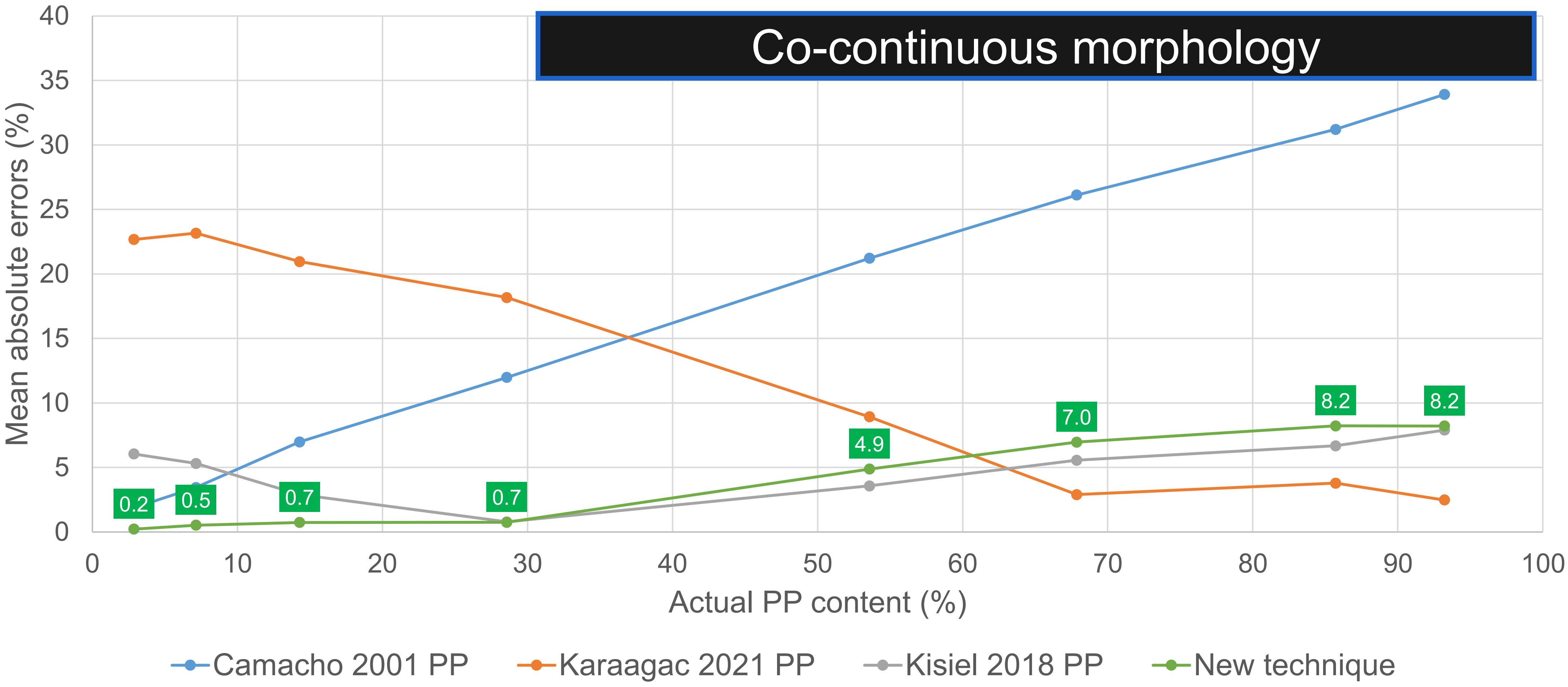
- Extrusion temperature: 210 °C (PE>70%), 230 °C (PE<70%)
- Screw speed: 100 rpm
- Residence time: 80 s
- Feeding amount: 2.8 g

- PE fraction: engineered blend of 8 different PEs
- PP fraction: engineered blend of 5 different PPs
- 9 compositions, 4 extrusions at each composition, single set of blending

xplore-together.com



COMPOSITION DETERMINATION FOR VALIDATION BLENDS



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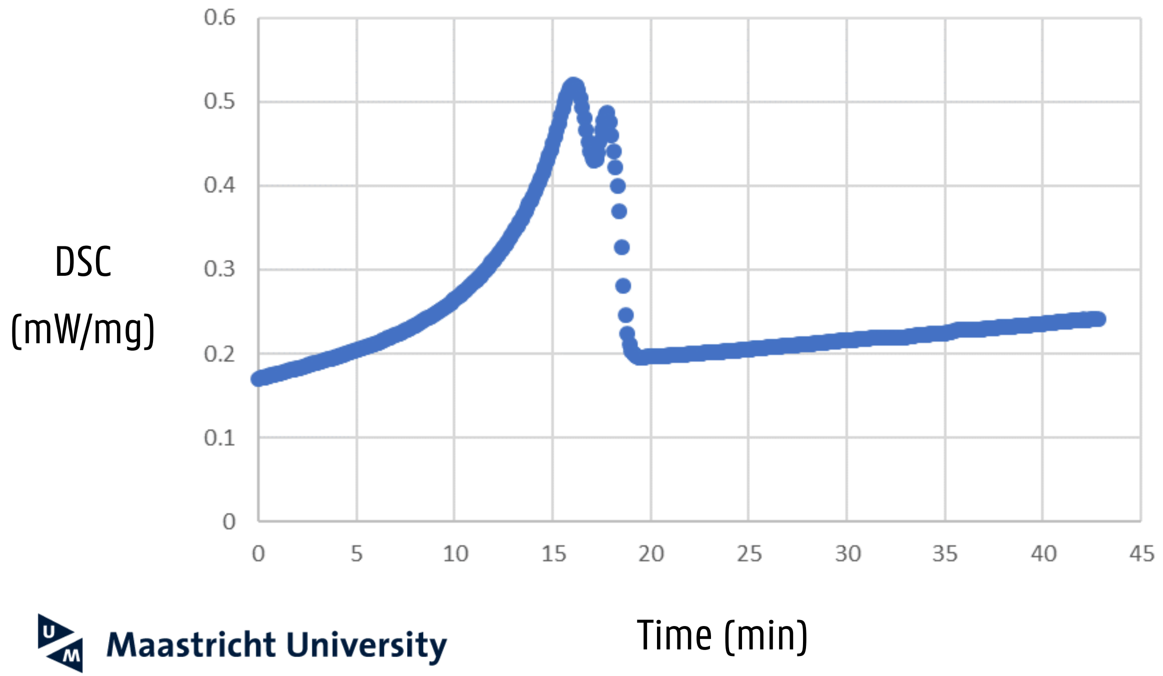
MACHINE LEARNING ASSISTED COMPOSITION DETERMINATION

MACHIN LEARNING METHODOLOGY: DATA

Calibration blends | Validation blends | Supporting data

– Inputs:
  - 429 Datapoints per curve between 30.5-245°C (0-42.8 min)
  - Composition



HDPE
LLDPE
LDPE
PP

Known composition

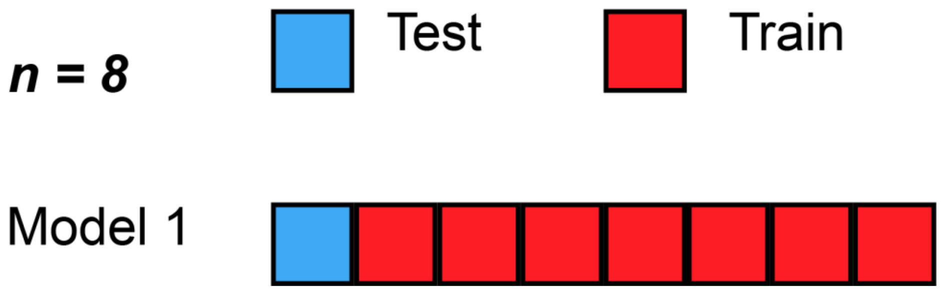
MACHIN LEARNING METHODOLOGY: MODEL

- PLS: Partial Least Squared Regression (16 components)

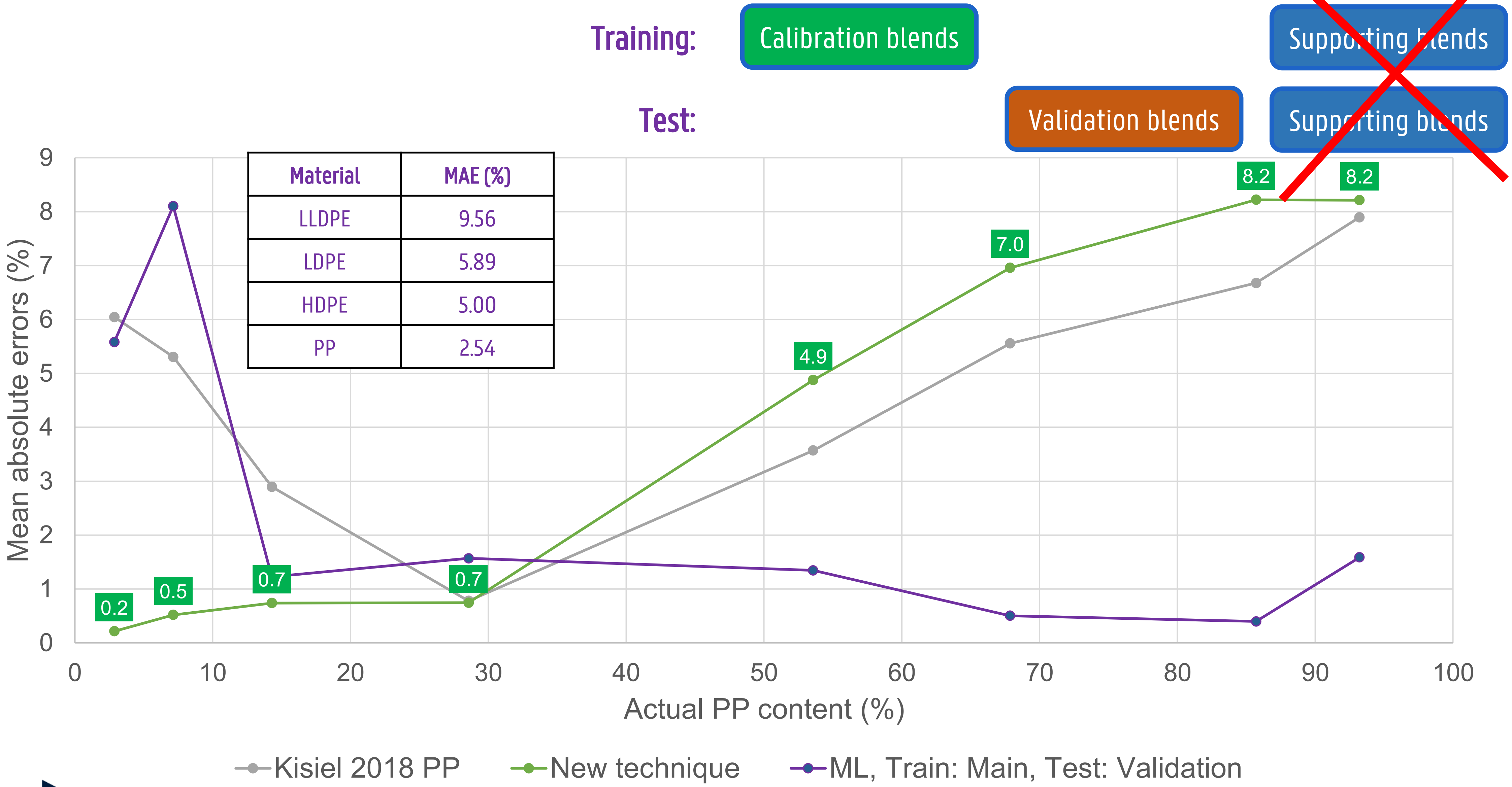
Finds a linear transformation P&Q between X (variables) and Y (output) to ensure a linear relation between Q.X and P.Y

$$PY = A(QX) + B$$

- 10-Fold Cross-validation (no bias on reported error)



MACHINE LEARNING TECHNIQUE, TRAIN: MAIN DATA



Material	MAE (%)
LLDPE	9.56
LDPE	5.89
HDPE	5.00
PP	2.54

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MACHINE LEARNING TECHNIQUE, TRAIN: ALL DATA, CROSS VALIDATION

***n* = 8**

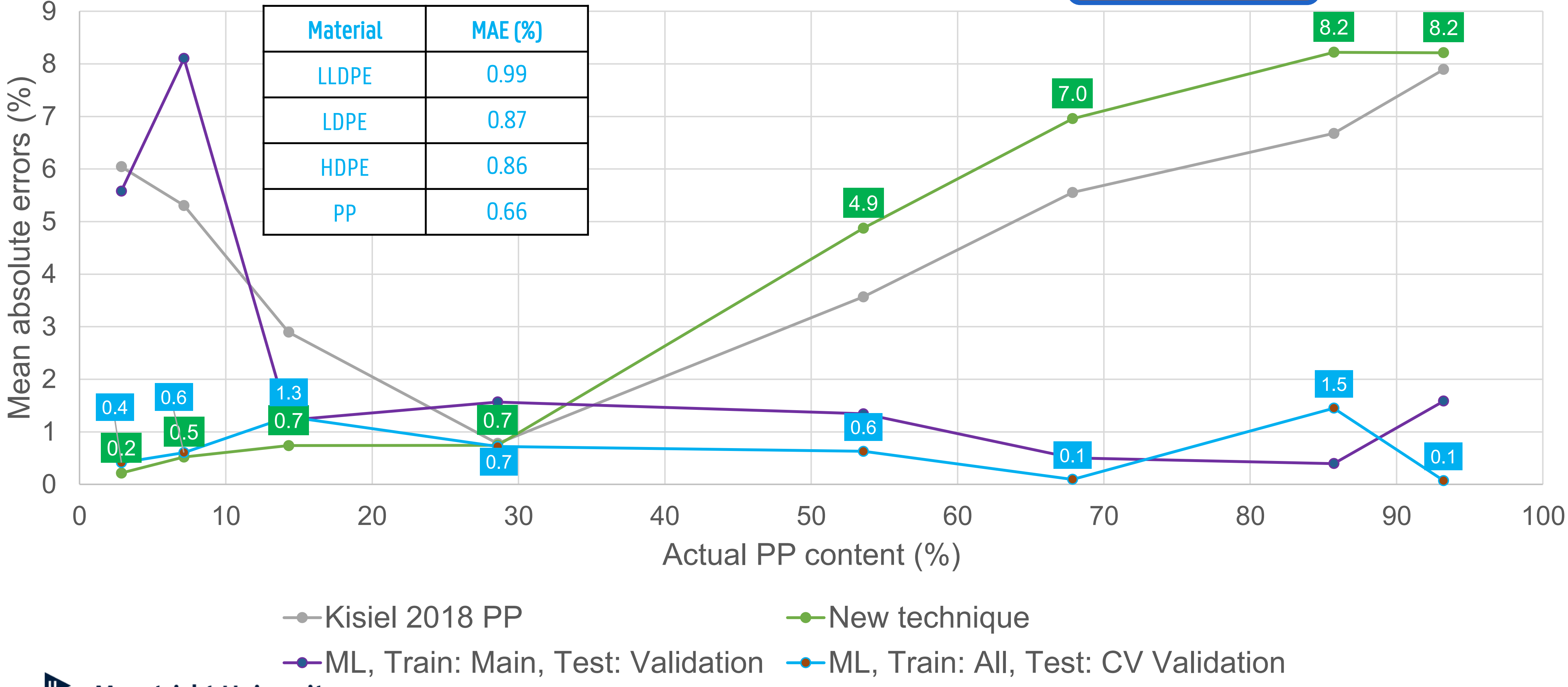
Test Train

Model 1

Training: Calibration blends | Validation blends | Supporting blends

Test: Validation blends

Material	MAE (%)
LLDPE	0.99
LDPE	0.87
HDPE	0.86
PP	0.66



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CONCLUSIONS

- A non-linear calibration curve based on the crystallinity of the constituents gives a higher accuracy for the determination of the composition. However, it can be used only if the material under investigation is of the same nature as the calibration curve; e.g., both being from the film applications.

- AI-assisted technique gives even a higher accuracy as it takes more features into account when determining the composition. Additionally, by (reasonably) improving the training dataset the model can become independent from the choice of the materials in the training dataset.

- AI-assisted can differentiate between not only PE and PP, but also to distinguish the subcategories namely LDPE, LLDPE, and HDPE, which is not possible by the conventional DSC-based technique, neither via FTIR-based techniques.

THANKS!

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Laboratory for Chemical Technology (LCT)

Maastricht University

Circular Plastics research group

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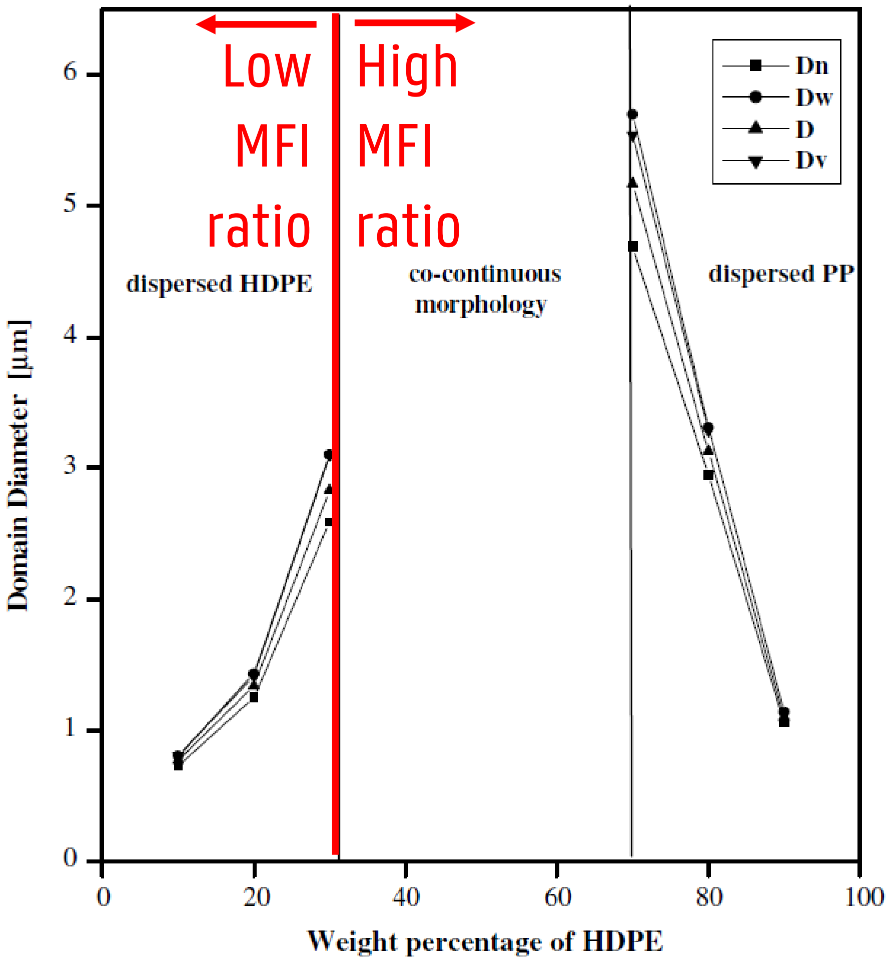
CRYSTALLINITY IN SEA-ISLAND STRUCTURES

- the particle size will be smaller when the volume fraction of the dispersed phase is smaller. Again, as the concentration of the dispersed phase decreases, the probability that a collision will result in coalescence becomes minimum.

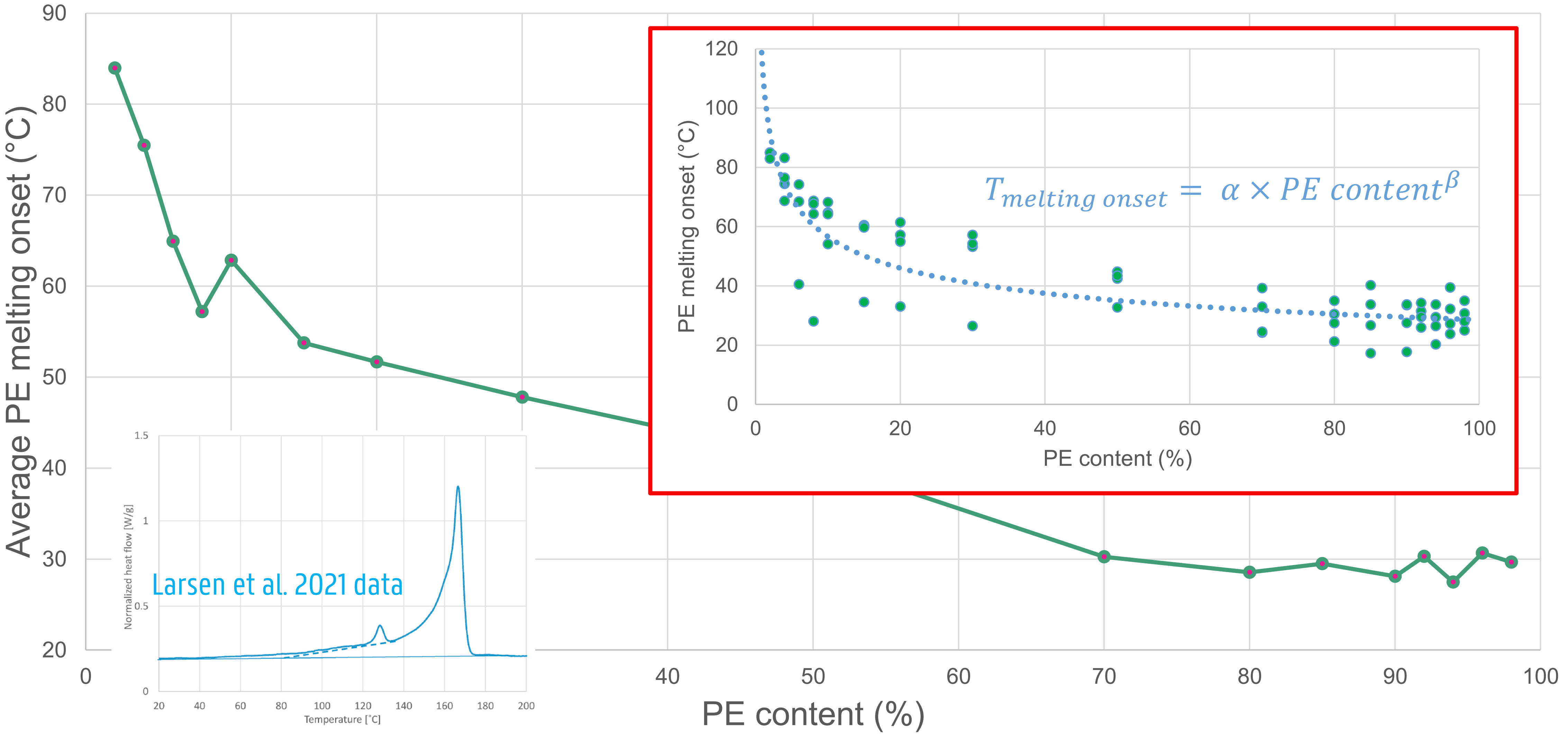
$$R^* = \frac{12 p \nu \phi_d}{\pi \sigma} \left(1 - \frac{4 p \phi_d E_{dk}}{\pi \sigma} \right) \quad (7)$$

where R^* = radius of particles, σ = shear stress, ν = interfacial tension, γ = shear rate, ϕ_d = volume fraction of the dispersed phase, E_{dk} = bulk breaking energy and p is the probability that a collision will result in a coalescence.



Jose et al., 2004
https://doi.org/10.1016/j.eurpolymj.2004.02.026

PE MELTING ONSET TEMPERATURE



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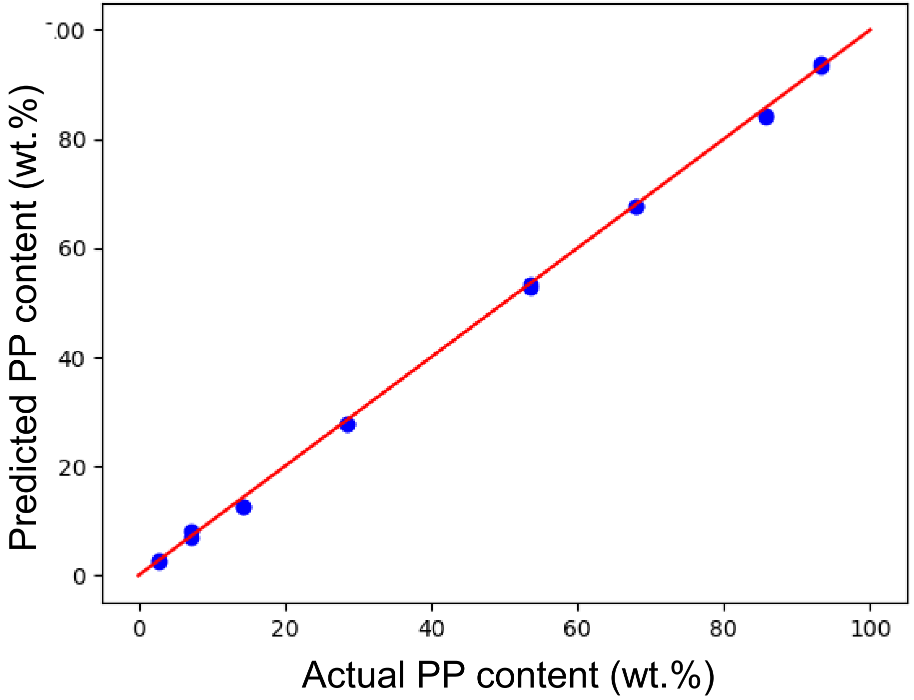
RESULTS AND DISCUSSION (PUBLISH II)

- Cross-validation on **validation** data trained on **all data**

Material	RMSE (%)	MAE (%)
LLDPE	1.19	0.99
LDPE	1.15	0.87
HDPE	1.07	0.86
PP	0.94	0.66



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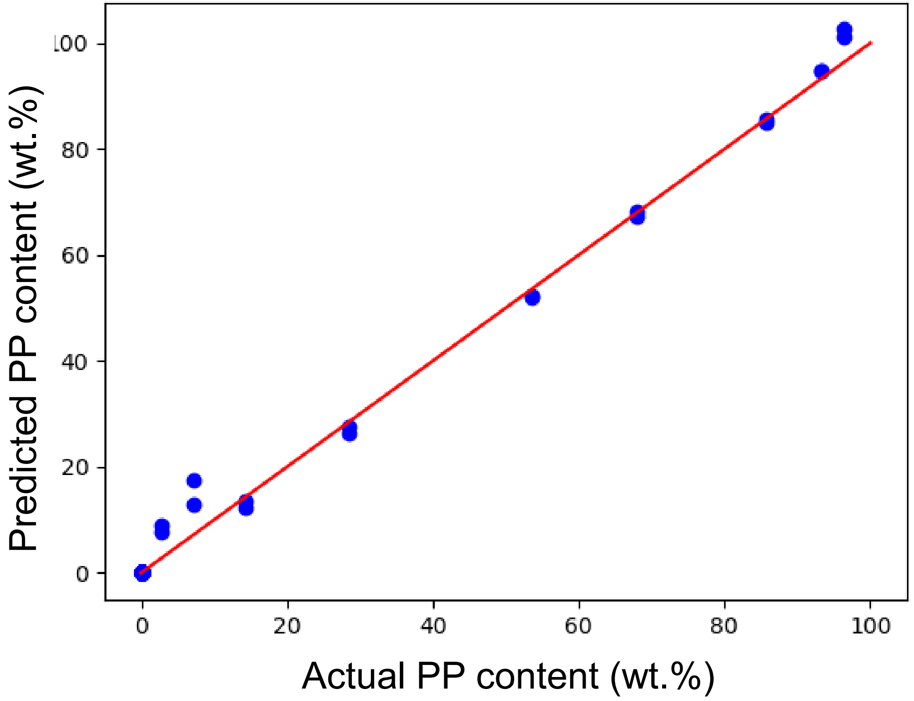
RESULTS AND DISCUSSION

- Validation on **validation** data trained on **main data**

(training on main + other is worse)

Material	RMSE (%)	MAE (%)
LLDPE	11.48	9.56
LDPE	7.29	5.89
HDPE	6.14	5.00
PP	3.98	2.54



RESULTS AND DISCUSSION

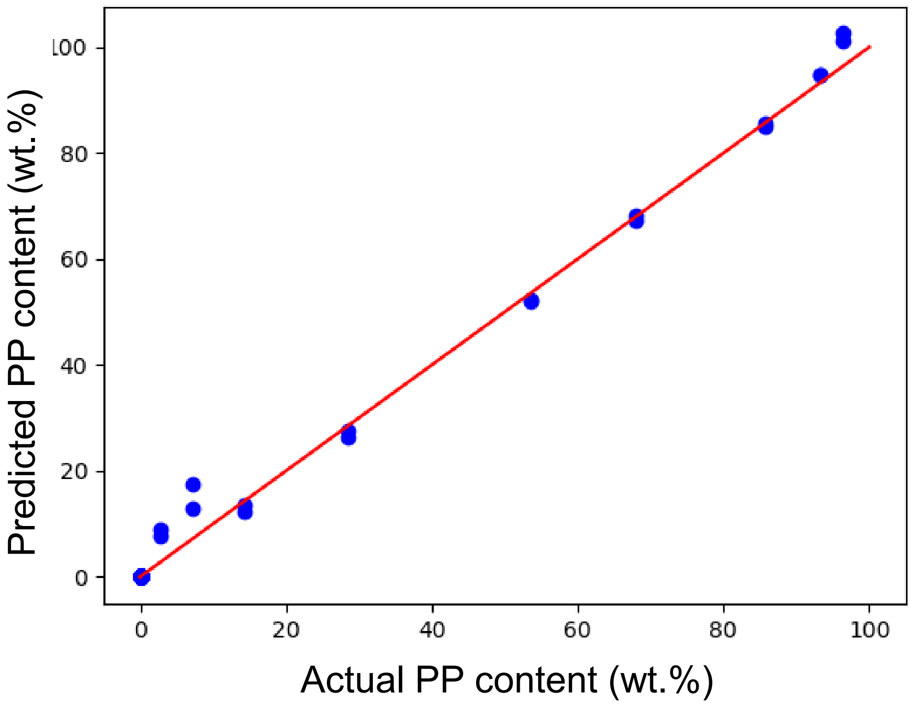
- Validation on **validation** data trained on **main data**

All Data

Material	RMSE (%)	MAE (%)
LLDPE	1.19	0.99
LDPE	1.15	0.87
HDPE	1.07	0.86
PP	0.94	0.66

Main Data

Material	RMSE (%)	MAE (%)
LLDPE	11.48	9.56
LDPE	7.29	5.89
HDPE	6.14	5.00
PP	3.98	2.54



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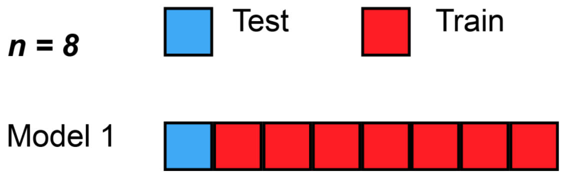
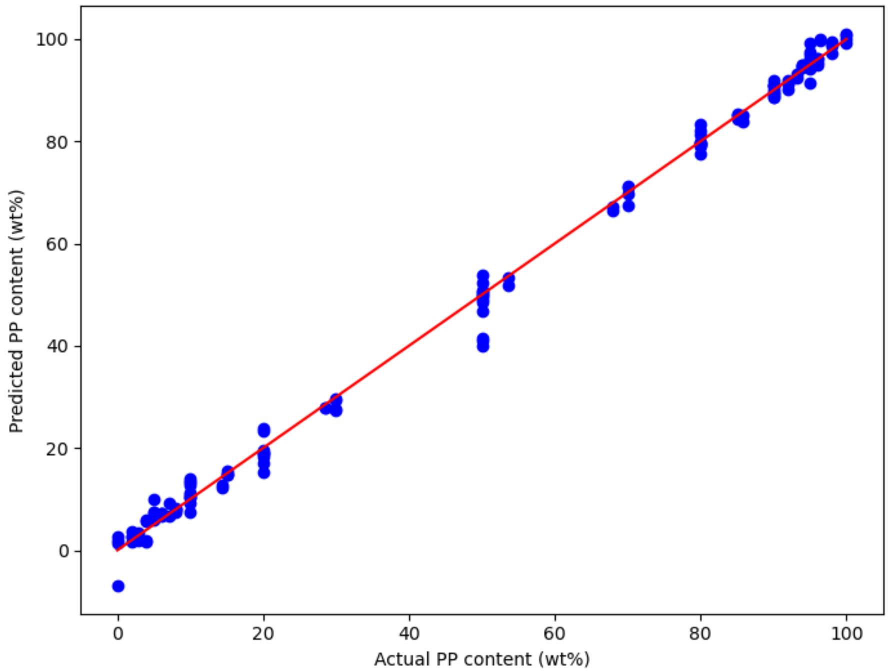
MACHINE LEARNING TECHNIQUE, TRAIN: ALL DATA, CROSS VALIDATION



Training: Calibration blends | Validation blends | Supporting blends

Test: Calibration blends | Validation blends | Supporting blends

Material	RMSE (%)	MAE (%)
LLDPE	1.47	1.05
LDPE	1.62	1.06
HDPE	1.58	0.91
PP	2.07	1.41



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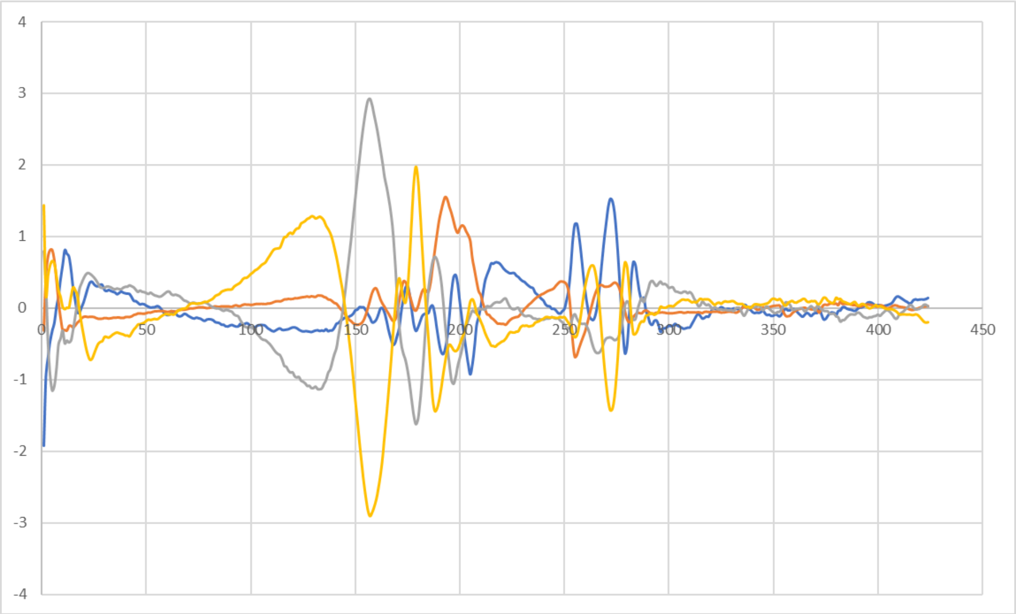
MODEL INSIGHTS

“Latent variable”

DSC (mW/mg)



Time×10 (min)

PP-curve