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Laboratory for Circular Process Engineering

SOLVENT TECHNIQUES FOR CLOSED-LOOP RECYCLING OF PLASTICS

Rita Kol – 03/07/2023

Supervisor: Prof. Steven De Meester
Co-Supervisors: Prof. Dimitris Achilias, Prof. Angeliki Lemonidou

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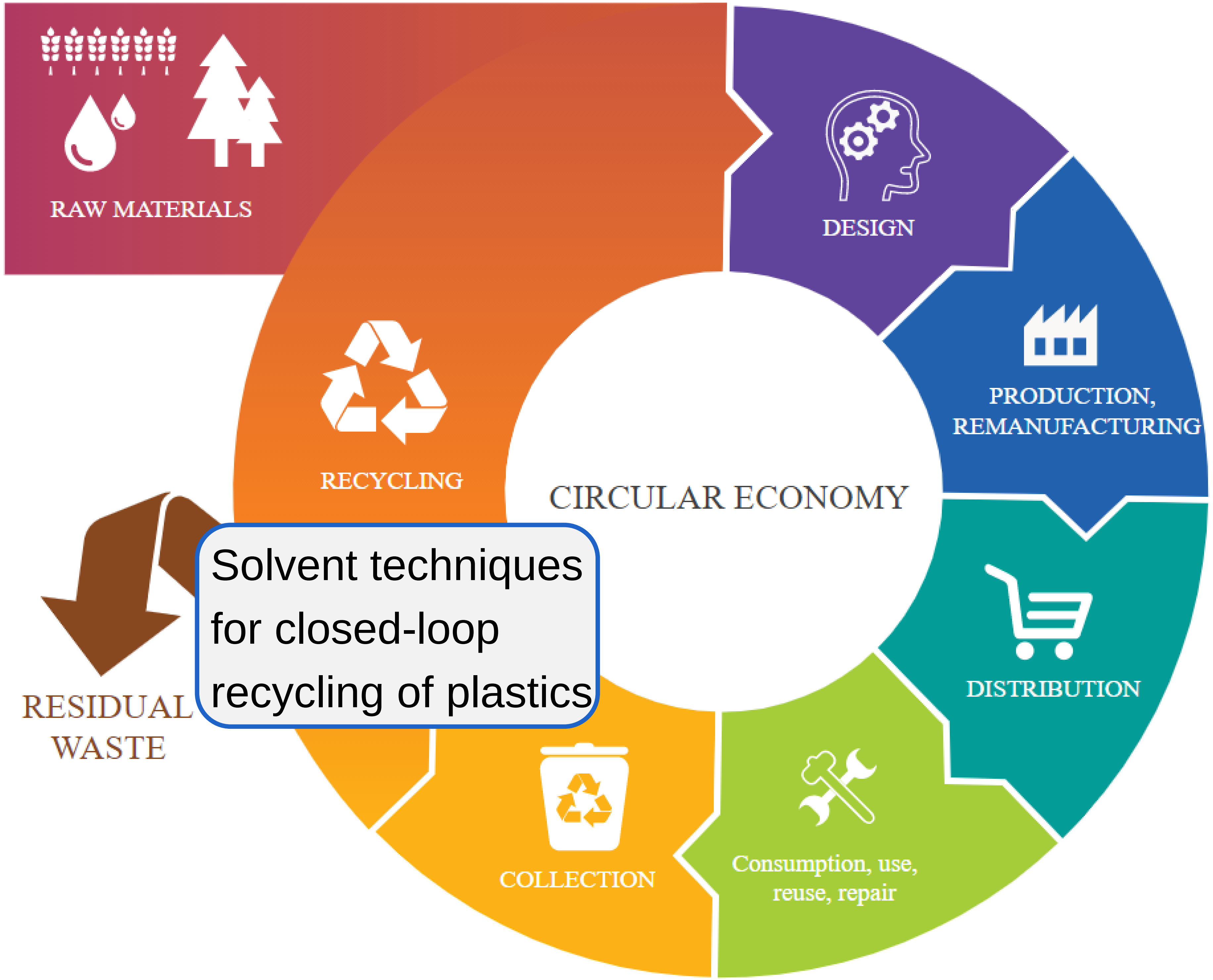
INEOS STYROLUTION

C-PlaNeT CIRCULAR PLASTICS NETWORK FOR TRAINING

This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No. 859885

ESR 9 – RITA KOL



Solvent techniques for closed-loop recycling of plastics

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CHALLENGES IN PLASTIC RECYCLING

- Unknown composition
- Different types of plastic
- Degraded polymers

- Additives
- Multilayer structures
- Contaminants

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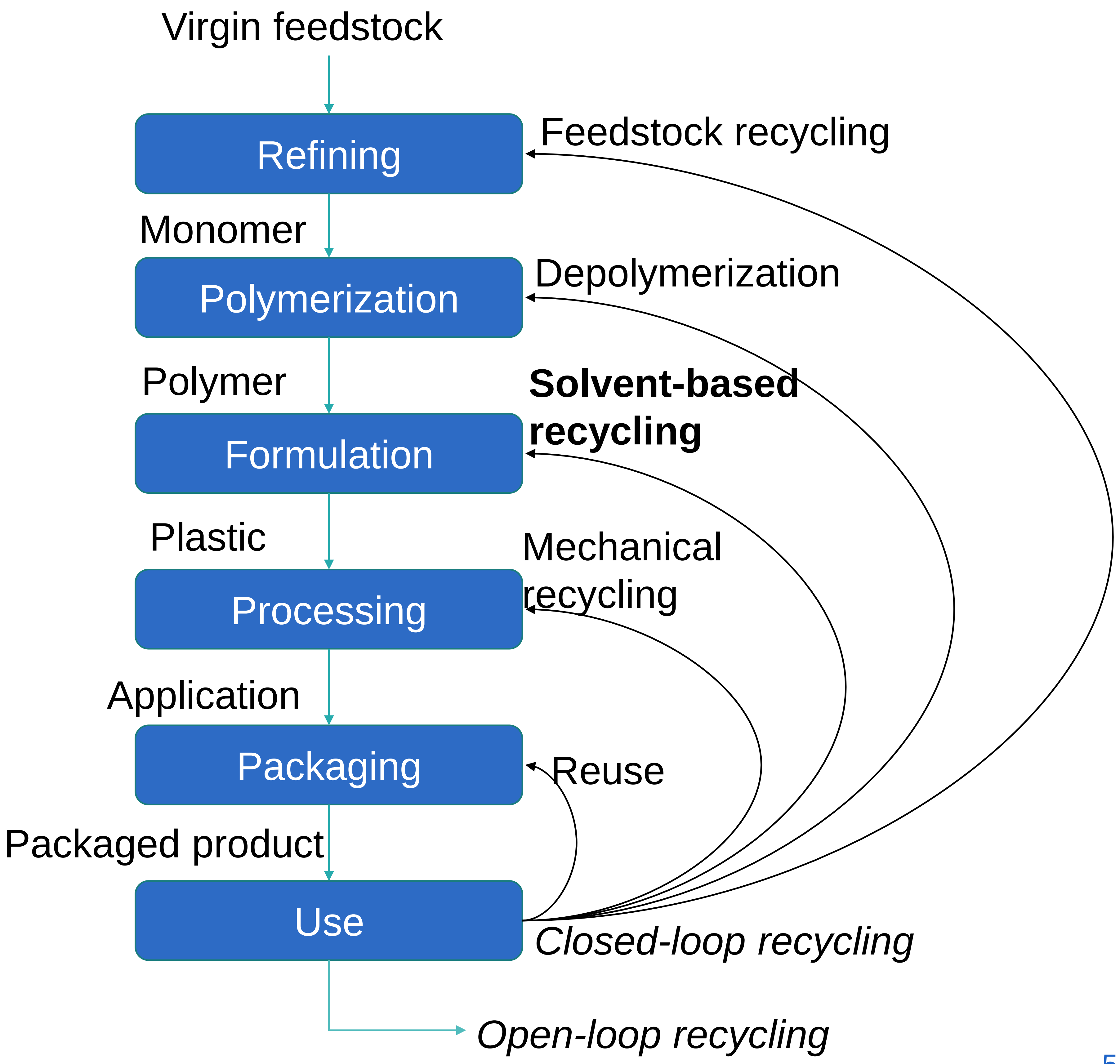
SOLVENT-BASED RECYCLING

- **Solvent-based recycling/ Dissolution recycling** (physical recycling):

  Polymer structure remains intact ≠ chemical recycling



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DISSOLUTION-PRECIPITATION TECHNIQUE

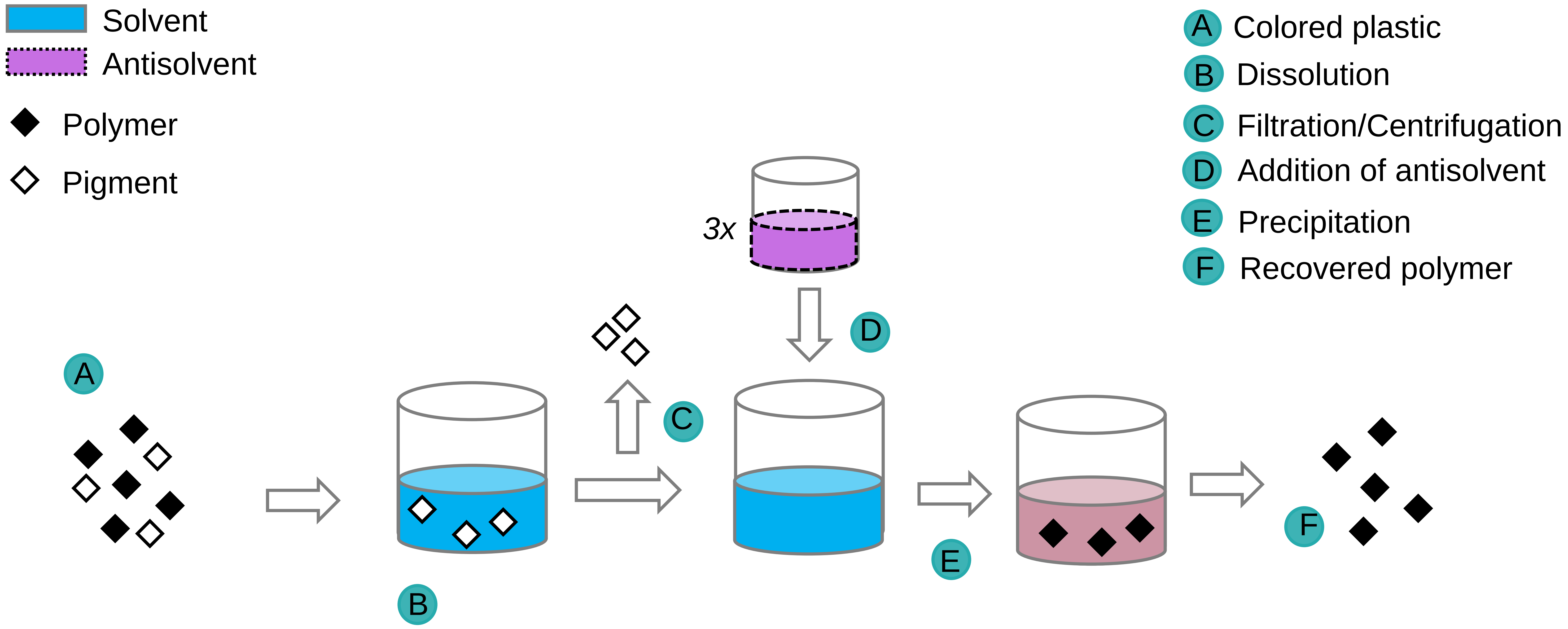

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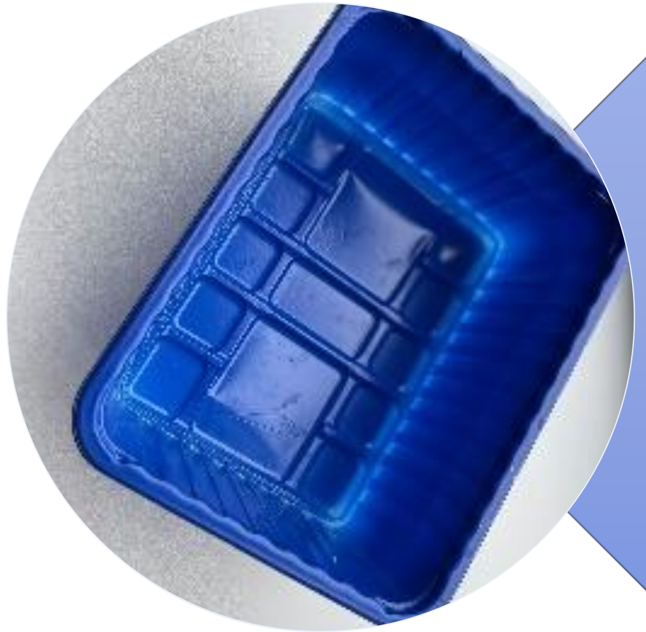
Figure redrawn and adapted from J. Sherwood (2020) Closed-loop recycling of polymers using solvents. Johnson Matthey Technology Review. pp. 4-15

OVERVIEW

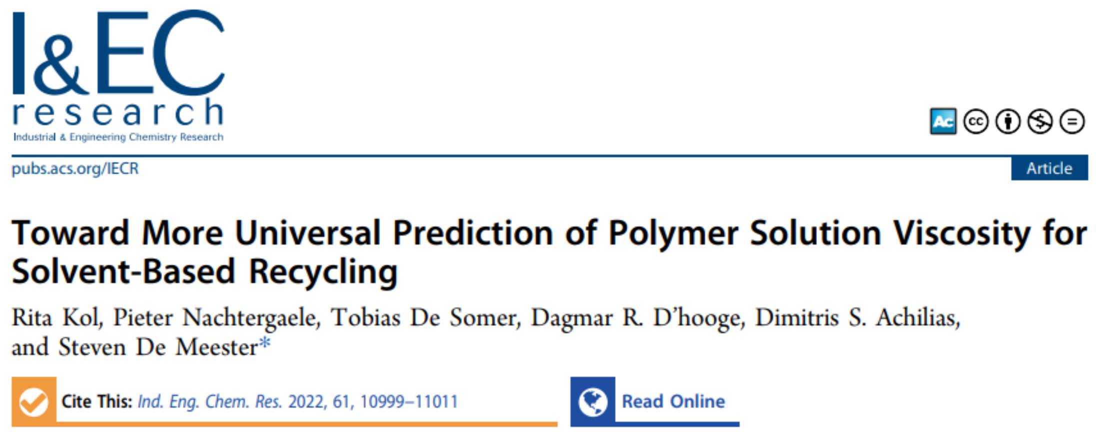
Rheology/Viscosity of polymer solutions

- Review & Implications for dissolution recycling (e.g. filtration & centrifugation)
- Polymer entanglements & concentration range for dissolution recycling
- Prediction of viscosity



Removal of insoluble particles from PS-based waste

- Pigments & rubber particles from HIPS
- Filtration & Centrifugation
- Modeling & Scale-up consideration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Toward More Universal Prediction of Polymer Solution Viscosity for Solvent-Based Recycling

Rita Kol, Pieter Nachtergaele, Tobias De Somer, Dagmar R. D'hooge, Dimitris S. Achilias, and Steven De Meester*

Cite This: Ind. Eng. Chem. Res. 2022, 61, 10999−11011
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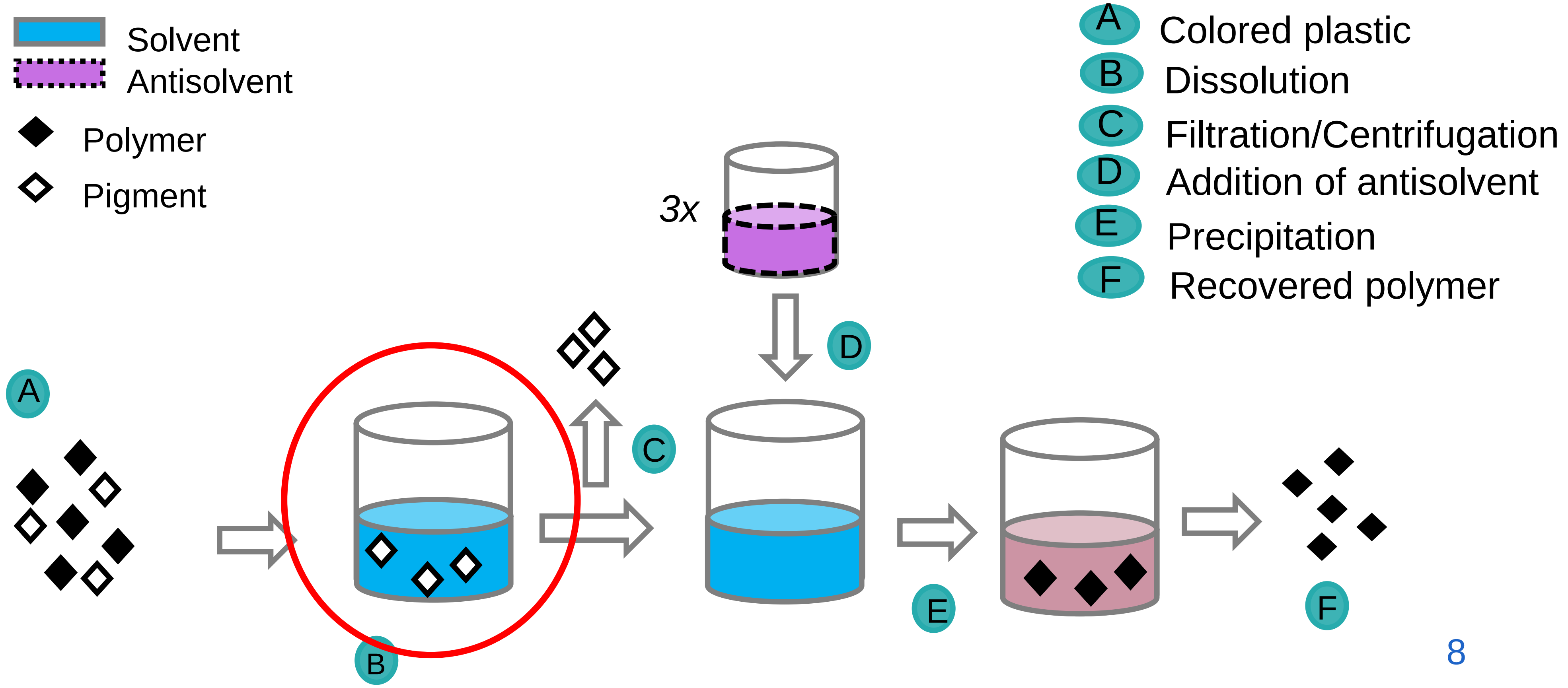
VISCOSITY OF POLYMER SOLUTIONS



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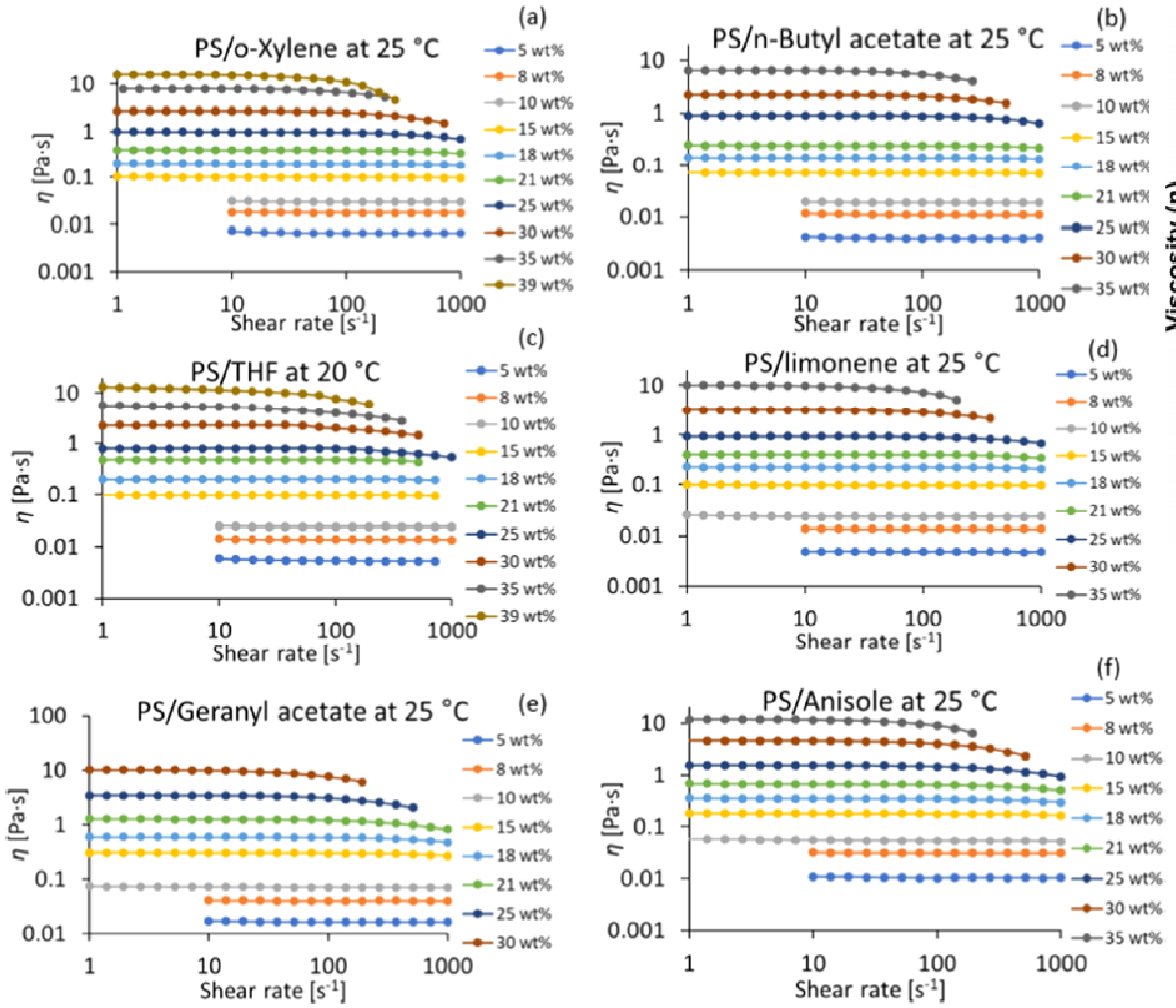
VISCOSITY MEASUREMENT OF POLYSTYRENE SOLUTIONS

Experimental conditions:

- ❑ Solvents:
  - *o-Xylene*
  - *Butyl acetate*
  - *THF*
  - *Limonene*
  - *Geranyl acetate*
  - *Anisole*
- ❑ *T*: 25, 40 and 50 °C
- ❑ Shear rate: 1 -1000 s^{-1}
- ❑ $c_{polymer}$: 5 and 39 wt%



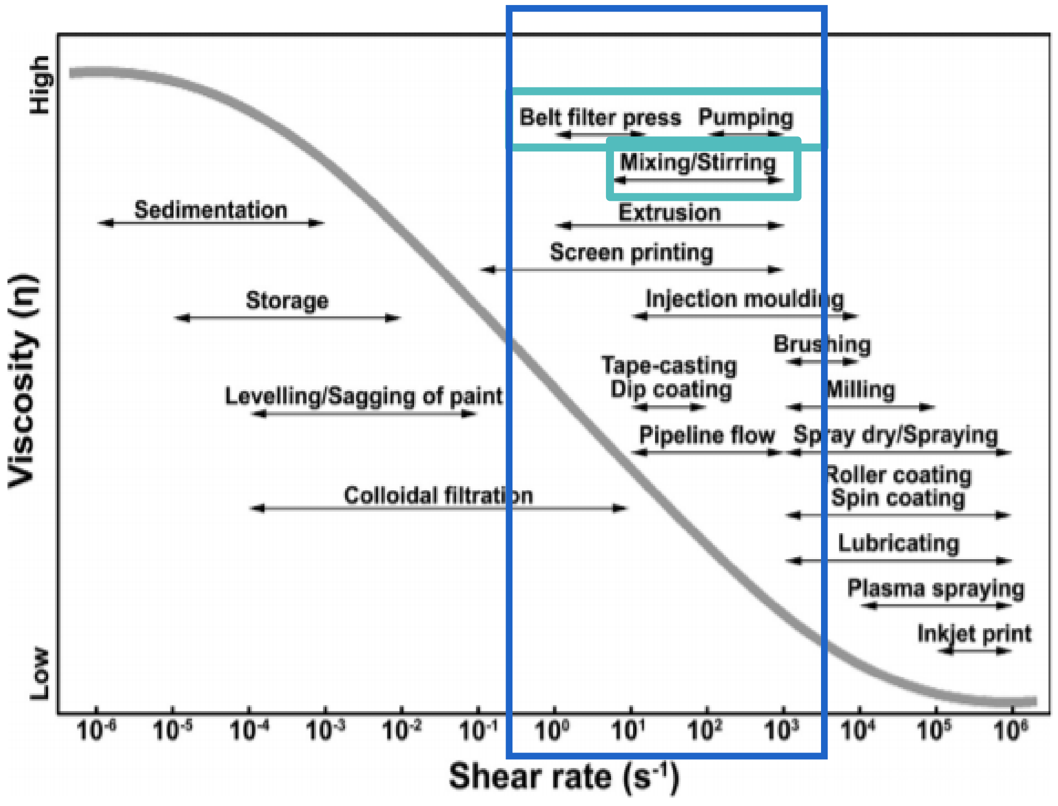


Figure from V. Carnicer et al. (2020), Open Ceramics 5,100052.

- ❑ Newtonian behaviour up to 15 wt%
- ❑ Visible gels at higher polymer concentration (>25 wt%)

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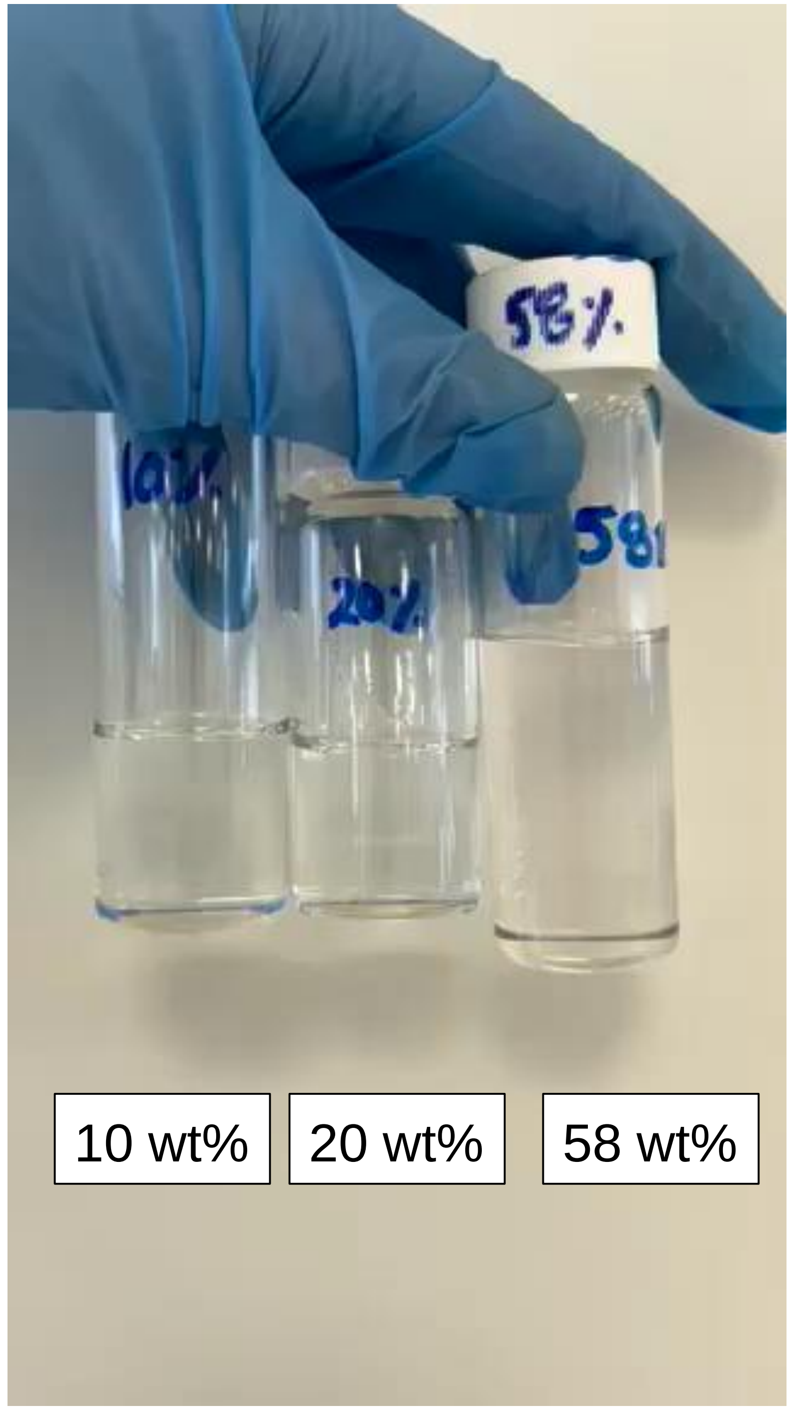
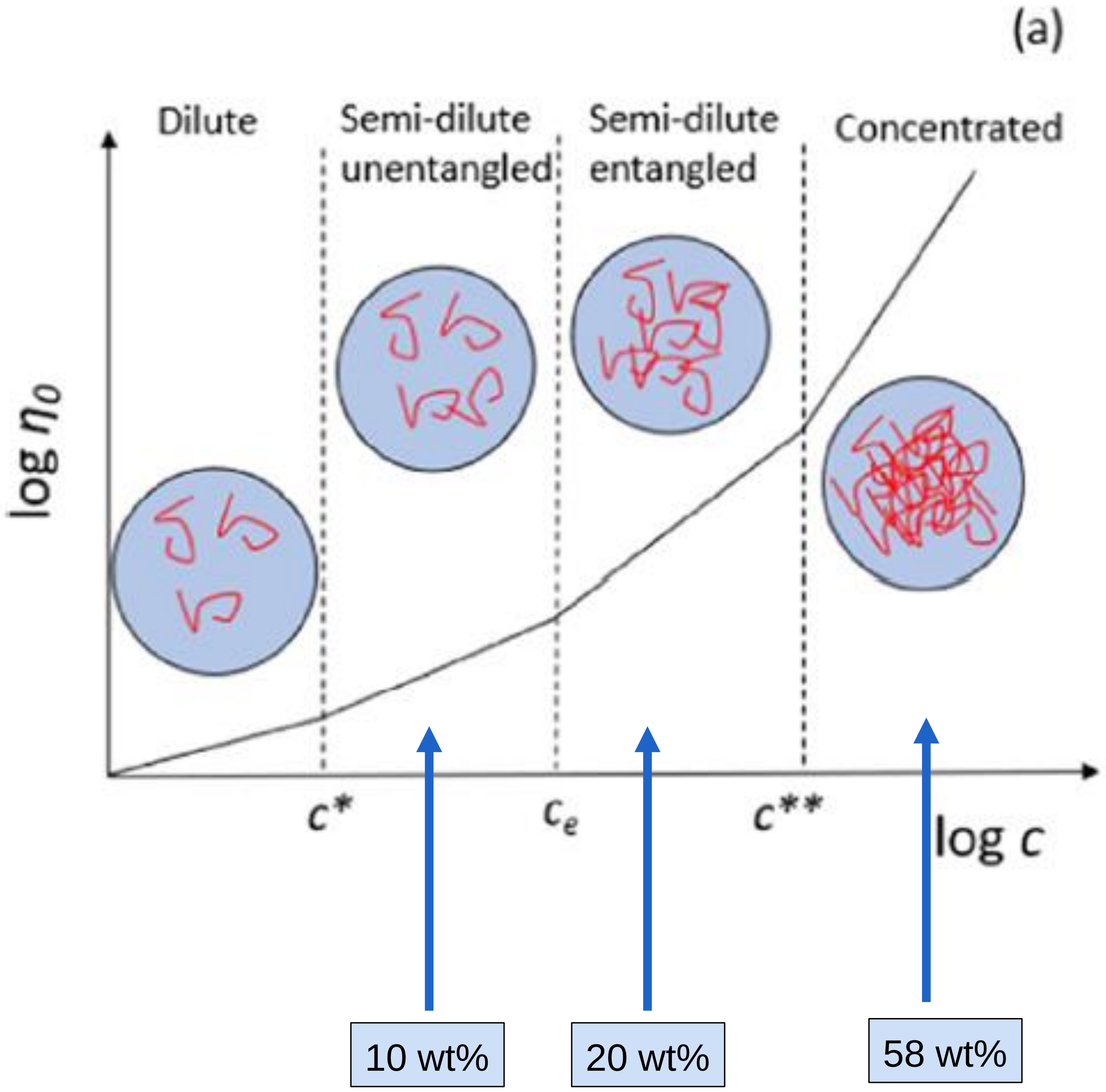
VISCOSITY OF PS SOLUTIONS





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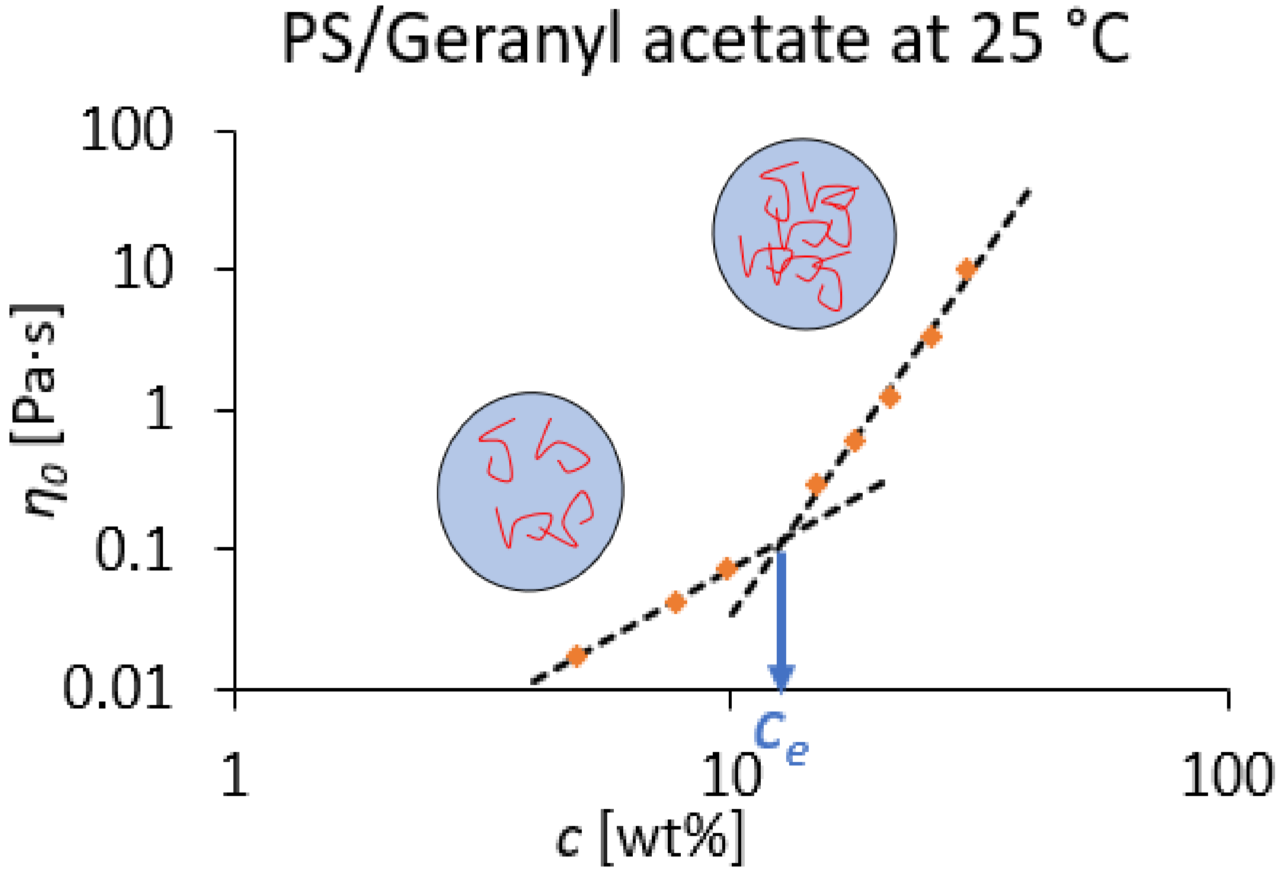
DETERMINATION OF ENTANGLEMENT CONCENTRATION



c_e – entanglement concentration

Table 2. Solubility Limit of Polystyrene in the Different Solvents at Room Temperature and Entanglement Concentrations, c_e

solution	PS solubility limit [wt %]	entanglement concentration [wt %]			
		temperature			
		20 °C	25 °C	40 °C	50 °C
PS/*o*-xylene	53.9 ± 1.0		13.9	14.1	14.6
PS/*n*-butyl acetate	62.7 ± 1.2		13.6	13.5	13.4
PS/THF	57.0 ± 0.2	13.0			
PS/limonene	47.1 ± 0.4		13.6	13.5	13.3
PS/geranyl acetate	40.9 ± 0.1		12.8	12.9	13.0
PS/anisole	58.5 ± 0.7		13.9	13.8	14.0

Experimental results: c_e = 13 – 15 wt%

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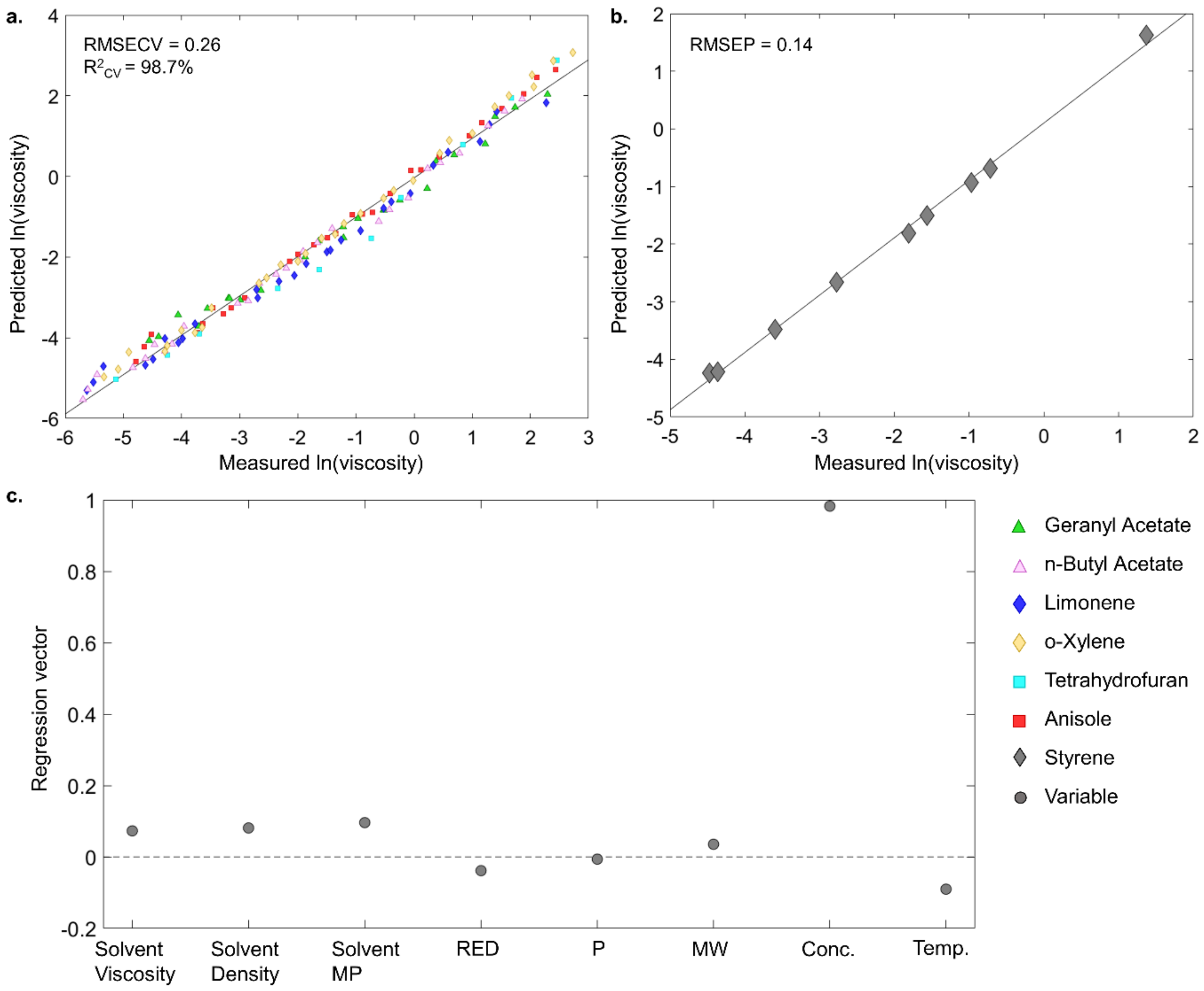
PREDICTION OF VISCOSITY

Regression analysis - Partial least squares regression model



$$\ln(\eta_0) = -8 + 0.38\ln\eta_s + 0.0038\,\rho_s + 0.0055\,MP$$
$$-0.69\,RED - 0.01P + 0.0024\,Mw + 0.23\,c$$
$$-0.019\,T + \varepsilon$$

- ❑ c → positive contribution, T → negative contribution
- ❑ Validation with external dataset

RMSECV - root-mean-square error of cross-validation

R^2_{CV} - cross-validated coefficient of determination

RMSEP - rootmean- square error of prediction

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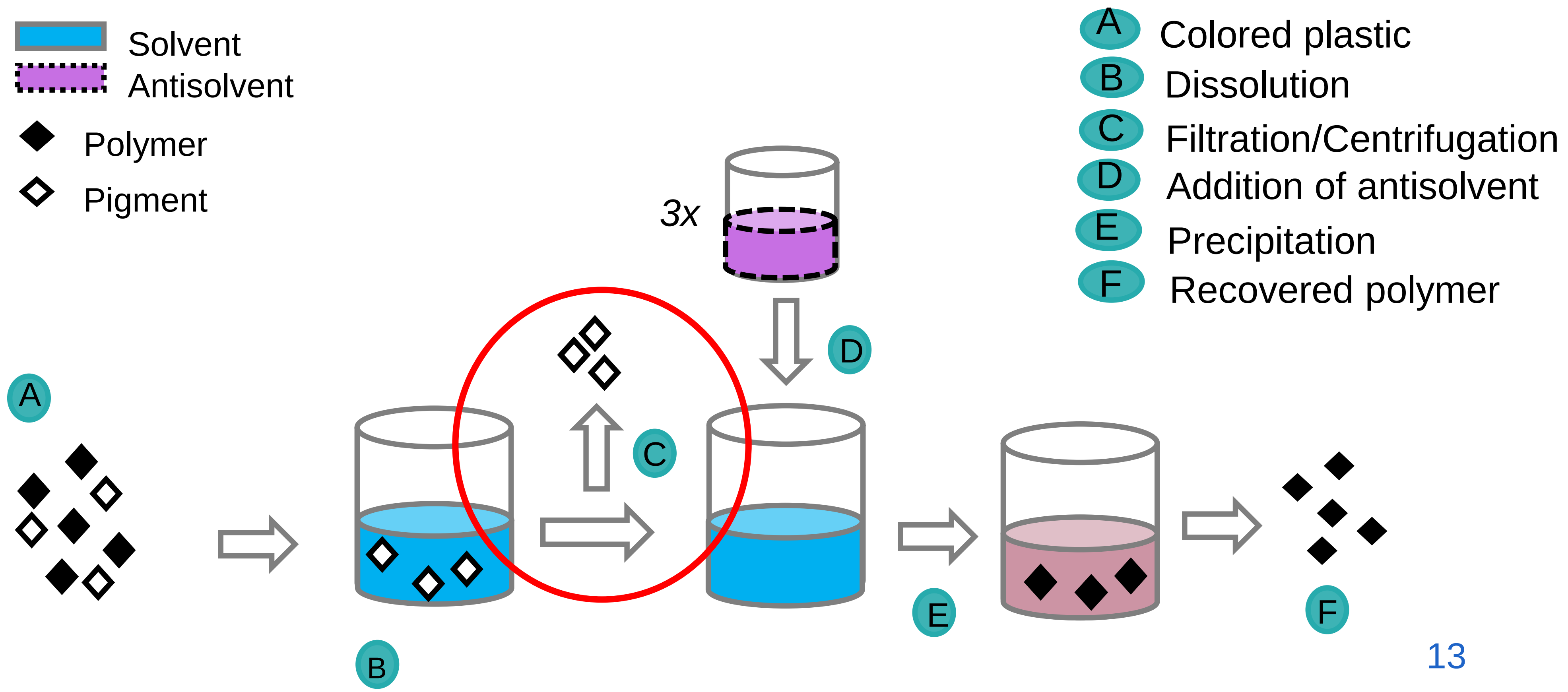
ELSEVIER
Separation and Purification Technology
Volume 325, 15 November 2023, 124682

Removal of undissolved substances in the dissolution-based recycling of polystyrene waste by applying filtration and centrifugation

Rita Kol [a] [b], Elisabetta Carrieri [a], Sergei Gusev [a], Michiel Verswyvel [c], Norbert Niessner [d], Angeliki Lemonidou [e], Dimitris S. Achilias [b], Steven De Meester [a]

REMOVAL OF INSOLUBLE PARTICLES



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EXPERIMENTAL PROCEDURE

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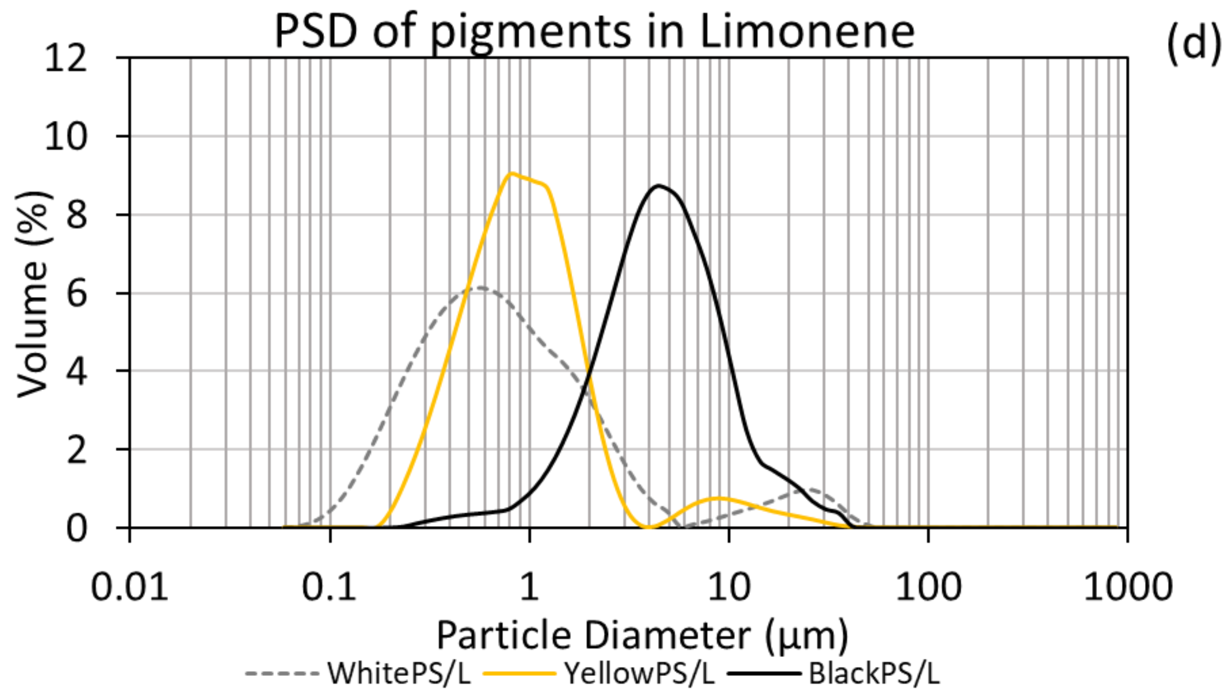
INSOLUBLE PARTICLES

Inorganic pigments

PS/Carbon black

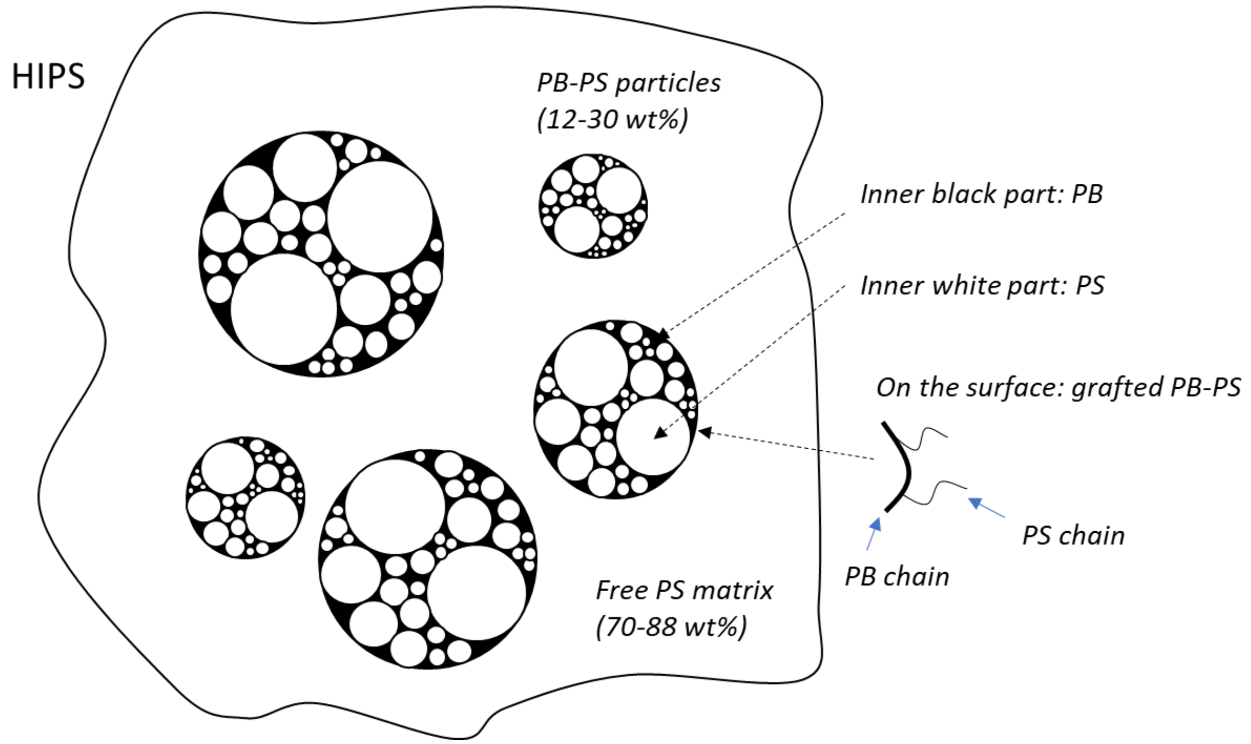
PS/Cr/Sb/Ti oxide

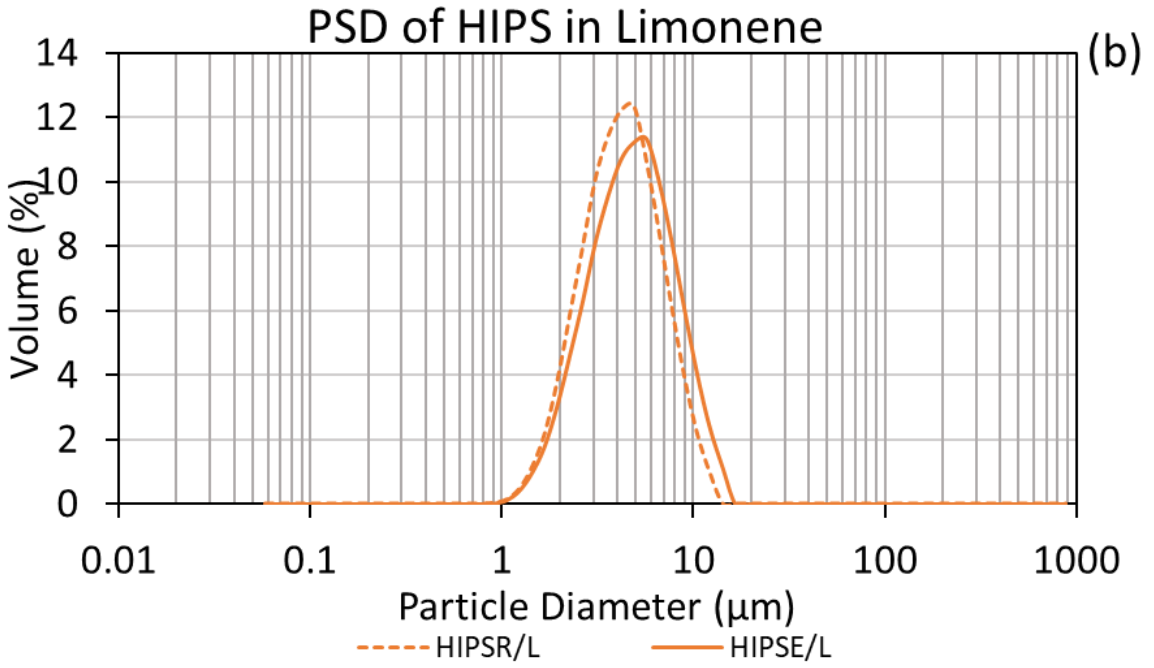
PS/TiO_2



HIPS rubber particles

HIPS





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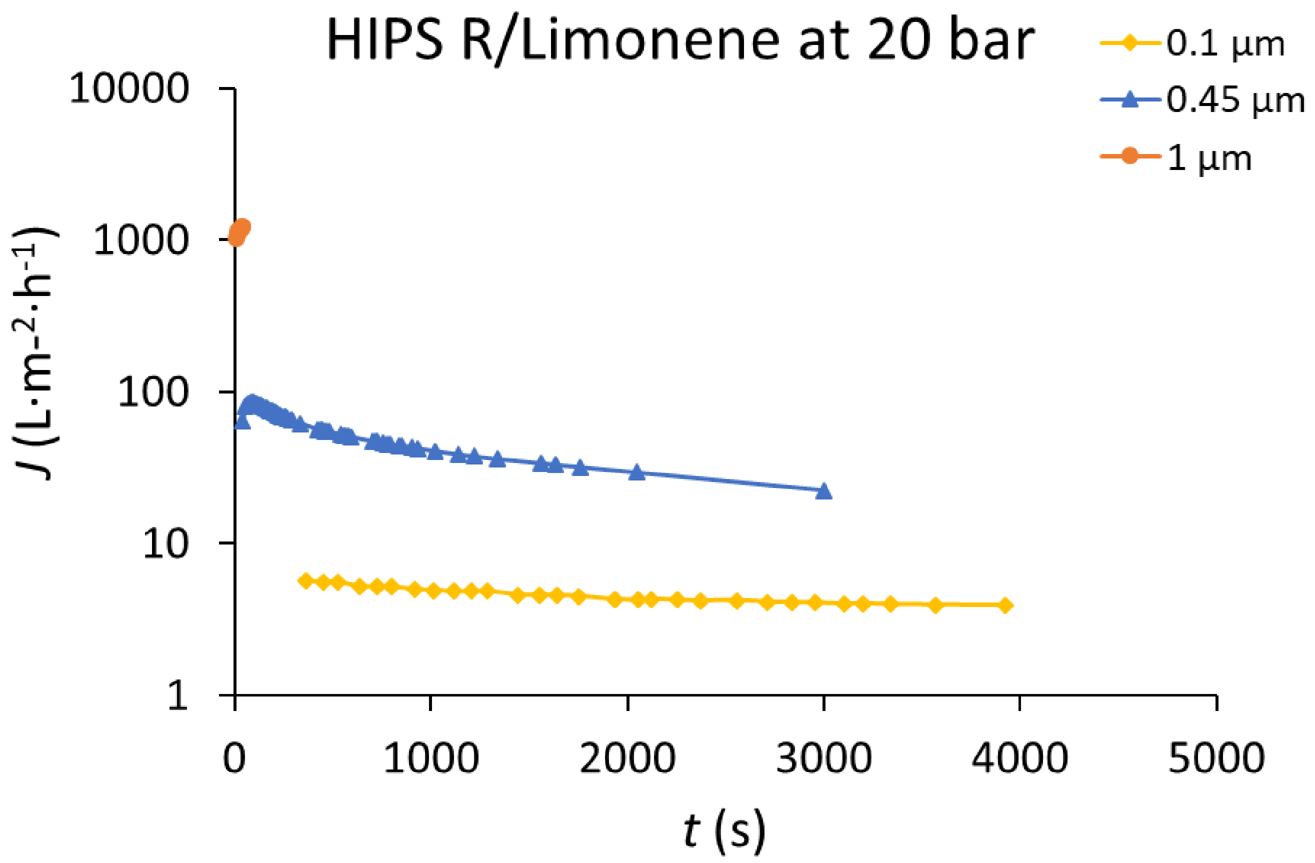
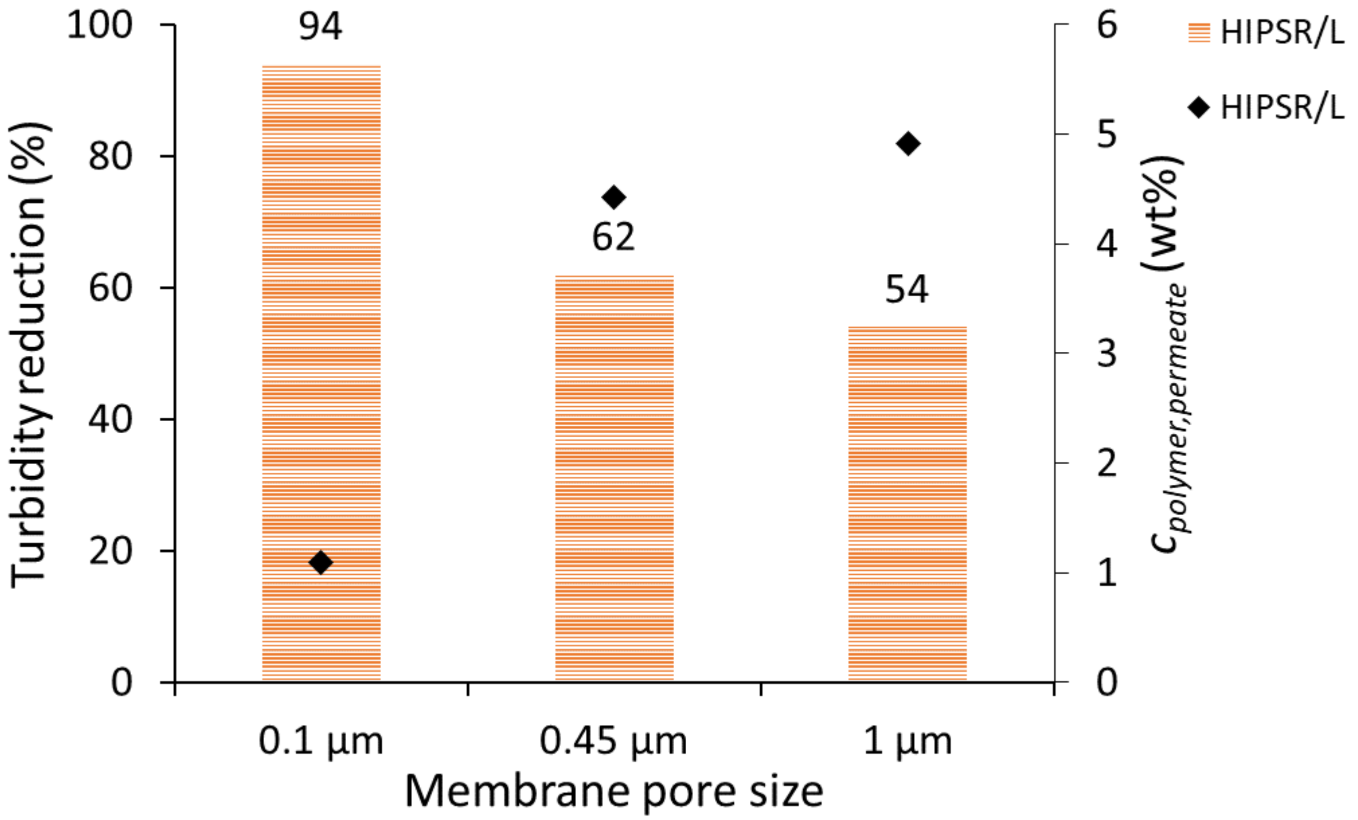
- ❑ Pigments: 0.1 < *d* < 100 µm
- ❑ Rubber particles of HIPS: 1 < *d* < 20 µm

MICROFILTRATION

Experimental conditions:

- ❑ Different membranes: 0.1 µm, 0.45 µm, 1 µm
- ❑ 5 wt% polymer solutions





- ❑ 0.1 µm: low flux but high turbidity reduction
- ❑ 1 µm: high flux but low turbidity reduction

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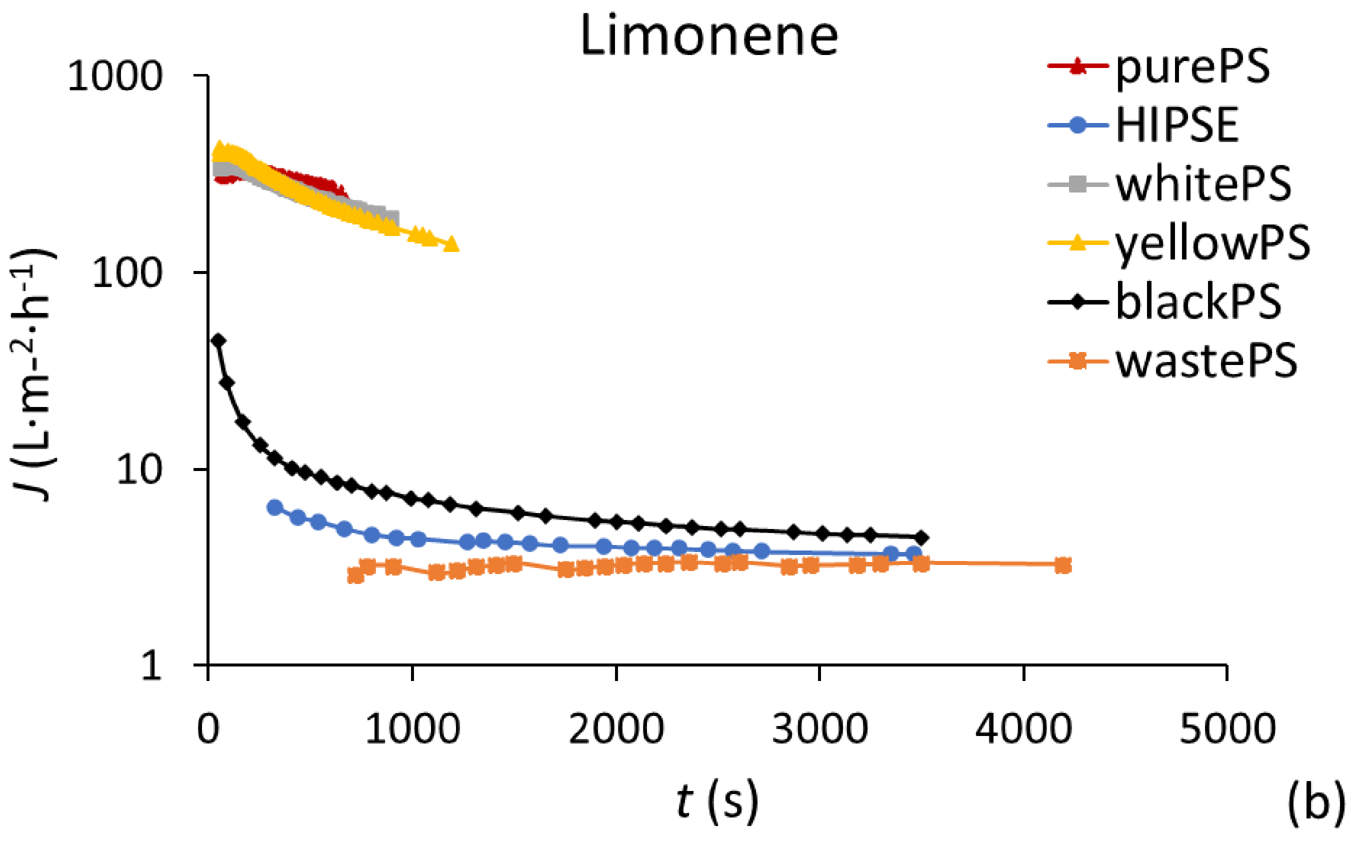
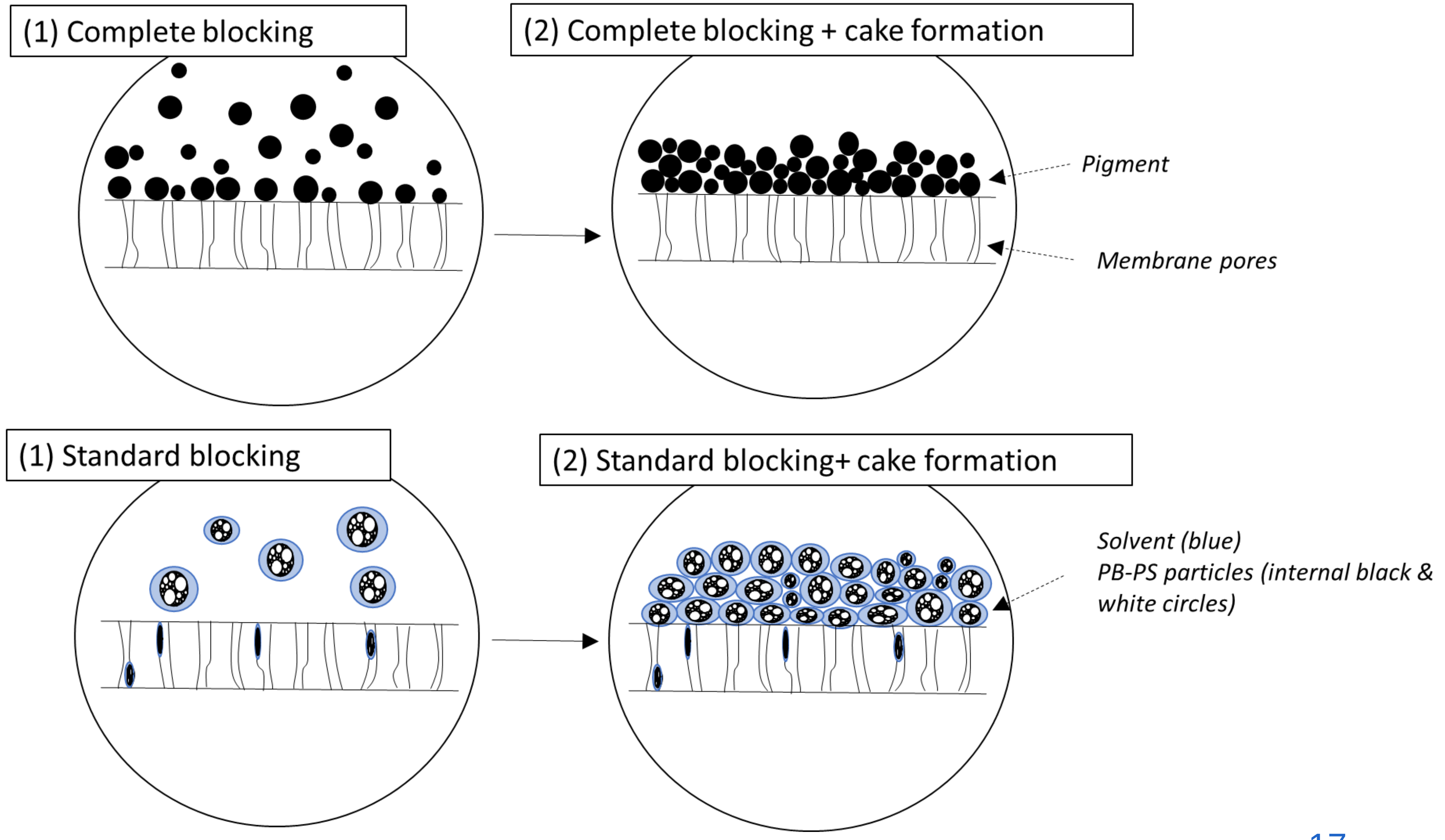
MICROFILTRATION

Experimental conditions:

- ❑ 0.1 µm membrane
- ❑ 5 wt% polymer solutions



Model for understanding underlying fouling mechanism



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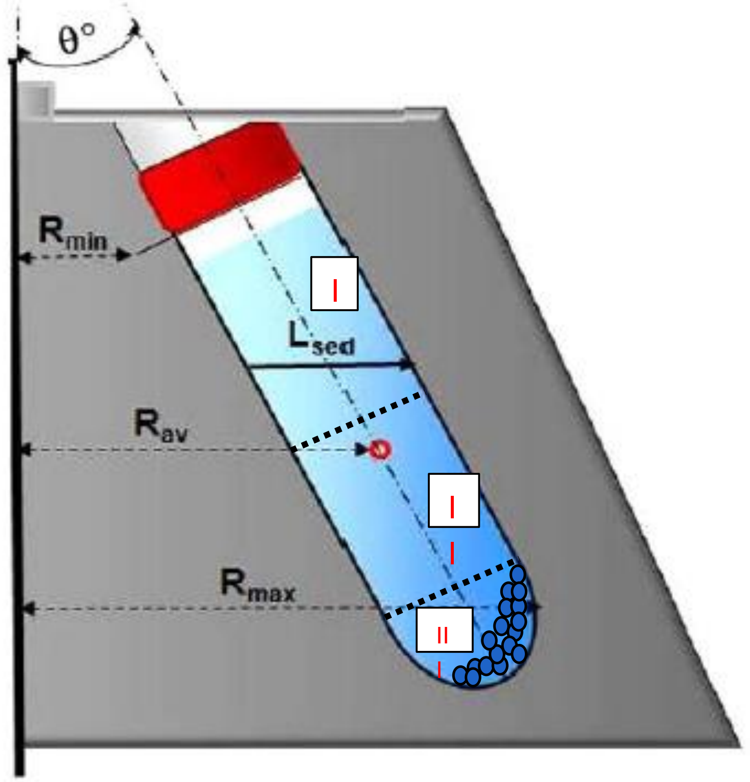
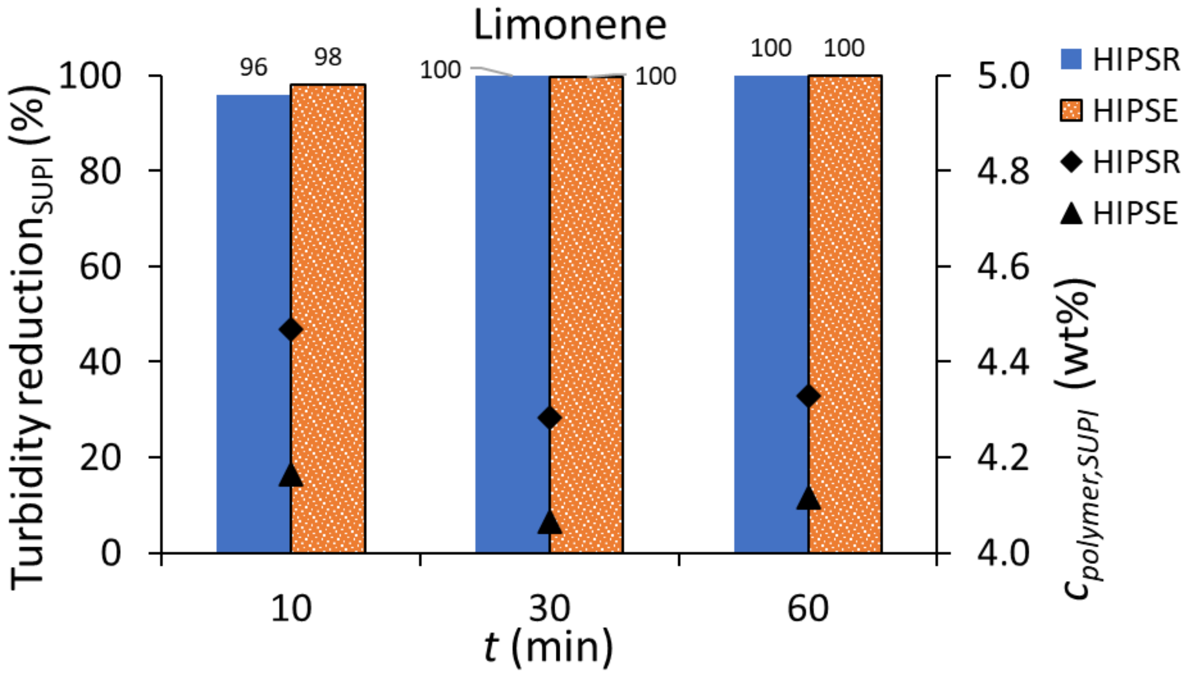
CENTRIFUGATION

Sigma Theory

Experimental conditions:

- ❑ G-force of 14052
- ❑ 5 wt% polymer solutions





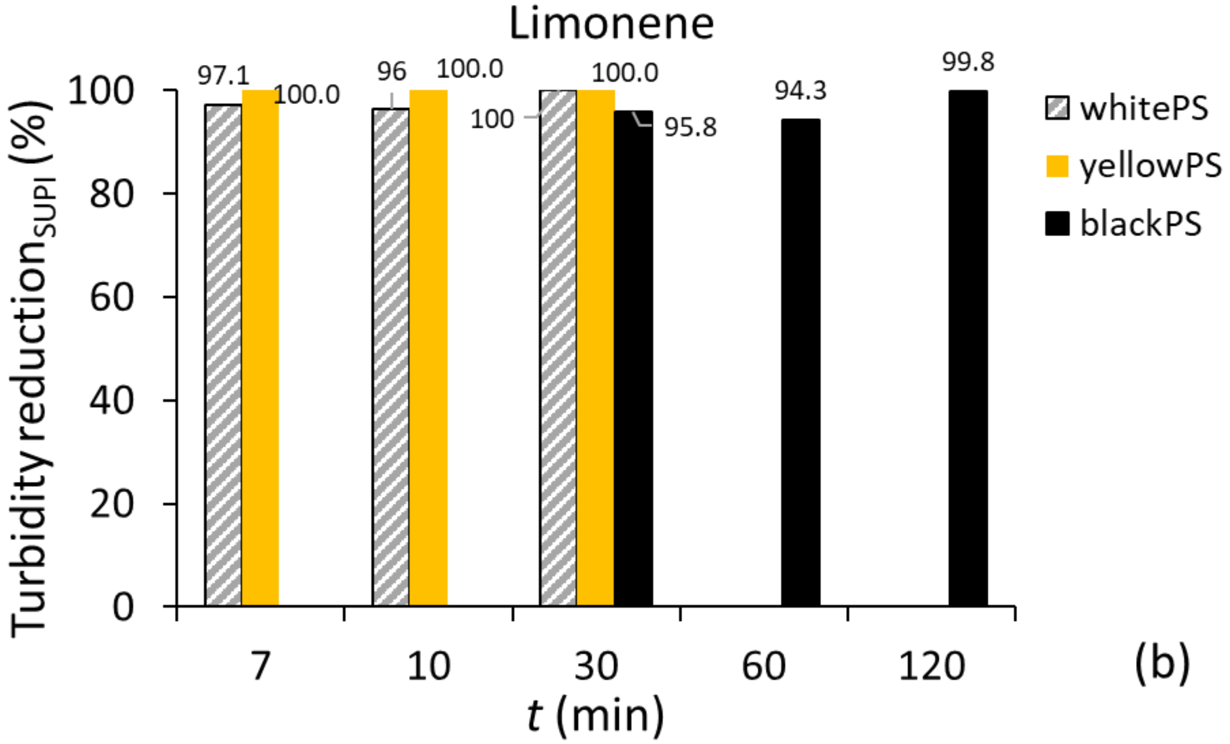
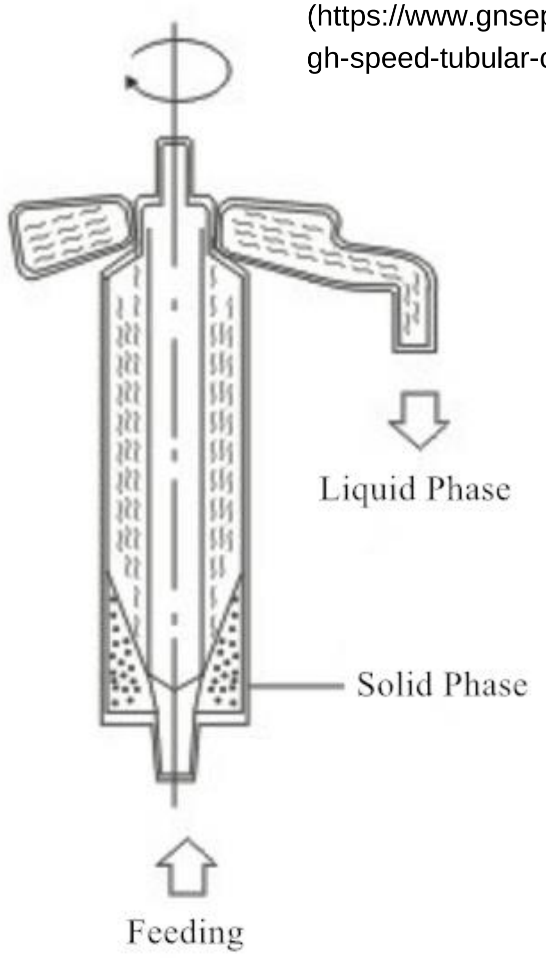


Figure from GN separation (https://www.gnseparation.com/high-speed-tubular-centrifuge).



Tubular bowl centrifuge

- ❑ 100% turbidity reduction for rubber particles and pigments (TiO2, Cr/Sb/Ti oxide) in limonene

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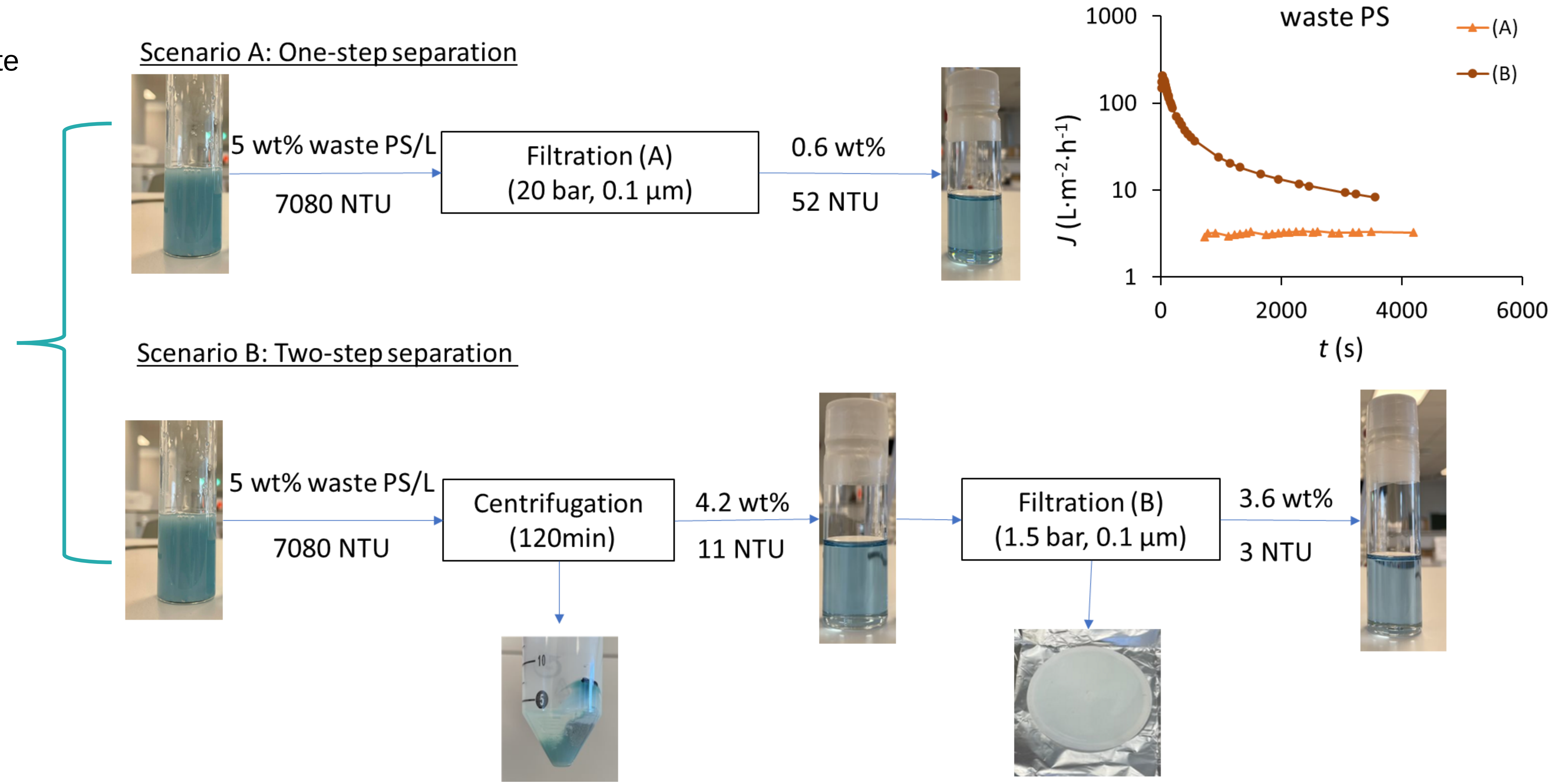
POLYSTYRENE-BASED WASTE

Rigid PS packaging waste



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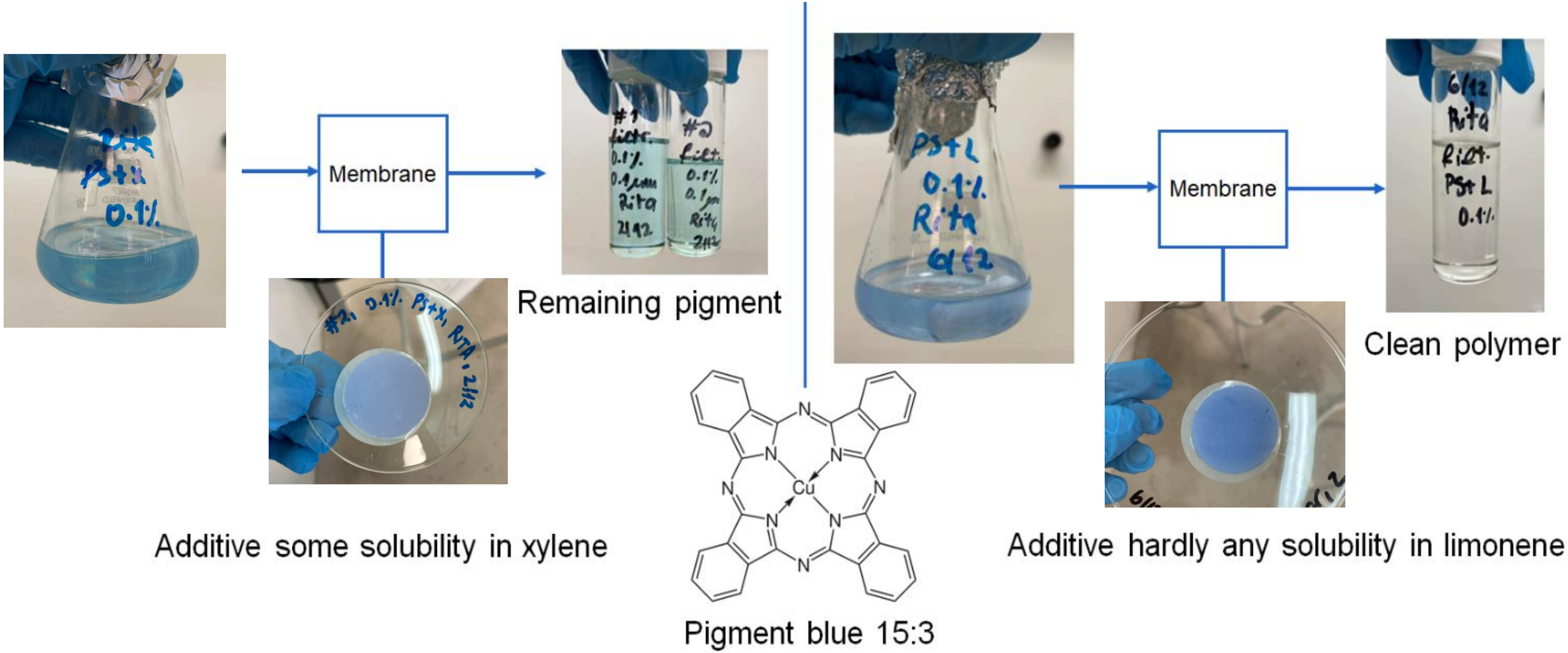
SOLVENT INFLUENCE



CONCLUSIONS

- ❑ Viscosity of polymer solutions plays an important role in dissolution recycling -> above entanglement concentration viscosity increases drastically;
- ❑ Regression model for PS viscosity prediction;
- ❑ Centrifugation is a promising technique to remove insoluble particles;
- ❑ 2 – step separation process can be beneficial to reduce resistance to filtration;
- ❑ Solvent screening for removal organic substances;

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State-Of-The-Art Quantification of Polymer Solution Viscosity for Plastic Waste Recycling

Rita Kol, Tobias De Somer, Prof. Dagmar R. D'hooge, Fabian Knappich, Prof. Kim Ragaert, Prof. Dimitris S. Achilias, Prof. Steven De Meester

First published: 29 July 2021 | https://doi.org/10.1002/cssc.202100876

Chapter Intechopen, 2021

Recent Advances in Pre-Treatment of Plastic Packaging Waste

Rita Kol, Martijn Roosen, Sibel Ügdüler, Kevin M. Van Geem, Kim Ragaert, Dimitris S. Achilias and Steven De Meester

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