Supplementary Information (SI) for Materials Advances.


# Finding low-toxicity biopolymer solvents with high melting temperatures and thermally induced phase separation of poly(ε-caprolactone) – **SUPPLEMENT**

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## 1. LIST OF PCL AND PLA DISSOLUTION CANDIDATES FOUND USING HSP

PCL and PLA dissolution candidates at T = 80 °C found using HSP parameters, including:

- Evaluated HSP parameters changed due to elevated temperature,
- melting ($T_m$) and boiling ($T_b$) temperature,
- toxicity (1 … lowest, 3 … highest, - … no data found),
- approximate purchase cost bracket $ … <100 $ per 100 g $$ … 100-200 $ per 100 g $$$ … > 200 $ per 100 g,
- and Relative Energy Difference (RED) calculated for standard as well as heated HSP parameters.

**Table 1A** PCL dissolution candidates and evaluated data, for PCL HSP taken from [1]

| Solvent | $\delta_D$ (Heated) ($Mpa^{1/2}$) | $\delta_P$ (Heated) ($Mpa^{1/2}$) | $\delta_H$ (Heated) ($Mpa^{1/2}$) | $T_m$ (°C) | $T_b$ (°C) | Toxicity | Costs | RED | RED (Heated) |
|---|---|---|---|---|---|---|---|---|---|
| BENZOIC ACID | 17.3 | 6.8 | 9.0 | 122.37 | 249.25 | 2 | $ | 0.28 | 0.25 |
| TETRAMETHYLTHIOUREA | 16.5 | 5.9 | 9.6 | 79.30 | 245.00 | 2 | $$ | 0.44 | 0.37 |
| GLYCIDYL METHACRYLATE | 15.5 | 8.3 | 5.2 | 76.00 | 189.00 | 3 | $$ | 0.91 | 0.80 |
| STEARIC ACID | 15.5 | 3.2 | 5.0 | 69.40 | 358.00 | 1 | $$ | 1.11 | 0.98 |
| METHYL VINYL SULFIDE | 15.6 | 4.8 | 5.5 | 66.60 | - | - | - | 0.62 | 0.53 |
| 1,3,5-TRIOXANE | 17.8 | 9.0 | 7.9 | 64.00 | 114.50 | 1 | $ | 0.65 | 0.63 |
| TRICHLOROBIPHENYL | 18.3 | 5.2 | 3.7 | 61.15 | 347.50 | 2 | $$$ | 1.08 | 0.96 |
| TRICHLOROACETIC ACID | 17.4 | 5.7 | 10.4 | 56.85 | 197.63 | 1 | $$ | 0.74 | 0.64 |
| INDOLE | 18.8 | 7.4 | 5.9 | 53.00 | 253.00 | 2 | $$ | 0.96 | 0.90 |
| PENTAFLUOROBENZOPHENONE | 18.4 | 7.9 | 4.9 | 37.00 | - | 2 | $$$ | 0.90 | 0.83 |
| METHYL P-TOLUATE | 18.1 | 6.4 | 3.5 | 33.00 | 217.00 | 1 | $ | 1.05 | 0.92 |
| 4-(TRIFLUOROMETHYL) ACETOPHENONE | 17.9 | 6.0 | 3.2 | 31.50 | 180.98 | 1 | $$$ | 1.08 | 0.93 |
| 4-VINYLPYRIDINE | 17.2 | 7.1 | 6.2 | 25.00 | 62.00 | 2 | $ | 0.11 | 0.10 |
| CYCLODECANONE | 16.0 | 7.8 | 3.7 | 22.85 | 236.05 | 1 | $$$ | 1.01 | 0.86 |
| TRANS-ANETHOLE | 18.1 | 4.2 | 7.9 | 22.00 | 235.50 | 1 | $ | 0.52 | 0.50 |
| 1-CHLORO-4-ETHOXYBENZENE | 18.4 | 6.2 | 4.0 | 21.00 | 213.00 | 2 | $$$ | 1.00 | 0.89 |
| DIVINYL SULFIDE | 15.7 | 4.5 | 5.1 | 20.00 | 84.00 | 2 | - | 0.69 | 0.59 |
| PENTAMETHYLENE SULFIDE | 17.6 | 6.2 | 8.1 | 19.00 | 142.00 | 2 | $$$ | 0.17 | 0.16 |
| 4-ACETYLMORPHOLINE | 17.4 | 5.2 | 7.1 | 14.50 | 207.97 | 2 | $$ | 0.09 | 0.09 |

**Table 1B** PCL dissolution candidates and evaluated data, for PCL HSP taken from [1] – continuation

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| VINYL PYRROLIDONE | 15.6 | 9.1 | 5.4 | 13.00 | 219.00 | 3 | $ | 1.03 | 0.93 |
| ETHYLENE DIBROMIDE | 18.3 | 3.4 | 7.9 | 10.00 | 131.00 | 3 | $ | 0.80 | 0.77 |
| ETHYL CINNAMATE | 17.5 | 8.0 | 3.7 | 7.75 | 271.75 | 1 | $ | 0.92 | 0.81 |
| HEXAMETHYLPHOSPHORAMIDE | 17.6 | 8.4 | 10.3 | 7.10 | 235.00 | 3 | $$ | 1.03 | 0.92 |
| 1-DECANOL | 16.7 | 2.5 | 9.1 | 6.75 | 230.20 | 2 | $ | 0.93 | 0.85 |
| 2,5-DIMETHYLPYRROLE | 17.4 | 7.5 | 6.2 | 6.50 | 171.00 | 2 | $$$ | 0.19 | 0.18 |
| METHYLENE DIIODIDE | 16.9 | 3.8 | 5.0 | 5.20 | 181.00 | 2 | $$ | 0.51 | 0.45 |
| 1-BROMO | 18.6 | 7.6 | 4.8 | 4.00 | 233.00 | 1 | $ | 0.98 | 0.90 |
| DIBENZYL ETHER | 16.5 | 3.6 | 6.7 | 3.60 | 288.30 | 2 | $ | 0.37 | 0.34 |

**Table 2A** PLA dissolution candidates and evaluated data, for PLA HSP taken from [2]

| Solvent | $\delta_D$ (Heated) ($Mpa^{1/2}$) | $\delta_P$ (Heated) ($Mpa^{1/2}$) | $\delta_H$ (Heated) ($Mpa^{1/2}$) | $T_m$ (°C) | $T_b$ (°C) | Toxicity | Costs | RED | RED (Heated) |
|---|---|---|---|---|---|---|---|---|---|
| BENZOIC ACID | 17.3 | 6.8 | 9.0 | 122.37 | 249.25 | 2 | $ | 0.45 | 0.37 |
| p-BROMOBENZONITRILE | 19.4 | 9.1 | 5.3 | 113.00 | 237.00 | 2 | $$ | 0.90 | 0.86 |
| 3-METHYLINDOLE | 19.0 | 7.0 | 5.7 | 95.00 | 265.00 | 1 | $$$ | 0.36 | 0.35 |
| 4-CHLOROBENZONITRILE | 18.6 | 7.8 | 3.7 | 95.00 | 223.00 | 2 | $$ | 0.59 | 0.52 |
| TETRAMETHYLTHIOUREA | 16.5 | 5.9 | 9.6 | 79.30 | 245.00 | 2 | $$ | 1.07 | 0.90 |
| 2,4-DICHLOROBENZALDEHYDE | 18.8 | 8.6 | 4.9 | 71.00 | 233.00 | 1 | $ | 0.48 | 0.46 |
| 1,3,5-TRIOXANE | 17.8 | 9.0 | 7.9 | 64.00 | 114.50 | 2 | $ | 0.37 | 0.34 |
| TRICHLOROBIPHENYL | 18.3 | 5.2 | 3.7 | 61.15 | 347.50 | 3 | $$$ | 0.61 | 0.54 |
| TRICHLOROACETIC ACID | 17.4 | 5.7 | 10.4 | 56.85 | 197.63 | 2 | $$$ | 1.08 | 0.90 |
| 2,4-DICHLORO-5-NITROBENZOTRIFLUORIDE | 18.9 | 7.5 | 3.4 | 55.00 | - | 1 | $$ | 0.84 | 0.75 |
| INDOLE | 18.8 | 7.4 | 5.9 | 53.00 | 253.00 | 2 | $ | 0.26 | 0.26 |
| o-CHLOROTHIOPHENOL | 19.2 | 6.9 | 9.1 | 49.00 | 205.00 | 2 | $$ | 0.89 | 0.80 |
| 4-CHLOROBENZALDEHYDE | 18.9 | 7.1 | 5.1 | 47.50 | 213.50 | 2 | $ | 0.38 | 0.36 |
| p-BROMOBENZOYL CHLORIDE | 19.2 | 6.4 | 5.0 | 42.00 | 247.00 | 2 | $ | 0.56 | 0.53 |
| 3,4-DICHLORONITROBENZENE | 19.1 | 7.1 | 3.7 | 41.00 | 255.00 | - | $$$ | 0.80 | 0.72 |
| 4-ETHOXYACETOPHENONE | 17.9 | 10.1 | 5.8 | 37.00 | 268.50 | 2 | $$$ | 0.56 | 0.54 |
| PENTAFLUOROBENZOPHENONE | 18.4 | 7.9 | 4.9 | 37.00 | - | 2 | $ | 0.26 | 0.24 |
| 4'-METHOXY ACETOPHENONE | 18.0 | 11.0 | 6.4 | 36.00 | 250.00 | 1 | $$ | 0.89 | 0.86 |
| 5-BROMO-2-NITROBENZOTRIFLUORIDE | 19.0 | 5.9 | 4.5 | 34.00 | 95.00 | 1 | $$$ | 0.60 | 0.55 |
| METHYL P-TOLUATE | 18.1 | 6.4 | 3.5 | 33.00 | 217.00 | 1 | $ | 0.54 | 0.46 |
| 4-(TRIFLUOROMETHYL) ACETOPHENONE | 17.9 | 6.0 | 3.2 | 31.50 | 180.98 | 1 | $$$ | 0.65 | 0.55 |

**Table 2B** PLA dissolution candidates and evaluated data, for PLA HSP taken from [2] – continuation

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| 2,6-DICHLOROANISOLE | 18.9 | 8.2 | 5.9 | 31.00 | 205.93 | 2 | $$ | 0.39 | 0.38 |
| DIPHENYL ETHER | 18.7 | 3.1 | 5.3 | 26.88 | 258.31 | 2 | $ | 0.95 | 0.91 |
| p-BROMOTOLUENE | 18.4 | 6.7 | 3.7 | 26.80 | 184.35 | 1 | $ | 0.49 | 0.42 |
| 2-BROMOTHIOPHENE | 19.1 | 5.1 | 4.2 | 25.00 | 150.00 | 2 | $ | 0.83 | 0.77 |
| 4-VINYLPYRIDINE | 17.2 | 7.1 | 6.2 | 25.00 | 62.00 | 3 | $ | 0.08 | 0.06 |
| TRANS-ANETHOLE | 18.1 | 4.2 | 7.9 | 22.00 | 235.50 | 1 | $ | 0.52 | 0.49 |
| 1-CHLORO-4-ETHOXYBENZENE | 18.4 | 6.2 | 4.0 | 21.00 | 213.00 | 2 | $$$ | 0.43 | 0.38 |
| p-CHLOROACETOPHENONE | 18.7 | 7.5 | 3.7 | 20.00 | 232.00 | 2 | $ | 0.62 | 0.55 |
| ACETOPHENONE | 18.7 | 8.4 | 3.4 | 19.45 | 202.11 | 1 | $ | 0.83 | 0.73 |
| PENTAMETHYLENE SULFIDE | 17.6 | 6.2 | 8.1 | 19.00 | 142.00 | 2 | $$$ | 0.21 | 0.18 |
| p-CHLOROBENZOYL CHLORIDE | 18.9 | 6.6 | 4.7 | 16.00 | 246.70 | 1 | $ | 0.46 | 0.43 |
| 4-ACETYLMORPHOLINE | 17.4 | 5.2 | 7.1 | 14.50 | 207.97 | 2 | $$ | 0.21 | 0.19 |
| 2,6-DICHLOROANISOLE | 18.8 | 8.2 | 5.9 | 10.10 | 205.93 | 1 | $$$ | 0.34 | 0.34 |
| ETHYLENE DIBROMIDE | 18.3 | 3.4 | 7.9 | 10.00 | 131.00 | 3 | $ | 0.79 | 0.75 |
| ETHYL CINNAMATE | 17.5 | 8.0 | 3.7 | 7.75 | 271.75 | 1 | $ | 0.51 | 0.43 |
| p-CHLOROTOLUENE | 18.2 | 6.1 | 2.4 | 7.50 | 162.50 | 1 | $ | 1.03 | 0.87 |
| HEXAMETHYLPHOSPHORAMIDE | 17.6 | 8.4 | 10.3 | 7.10 | 235.00 | 3 | $$ | 1.06 | 0.90 |
| 2,5-DIMETHYLPYRROLE | 17.4 | 7.5 | 6.2 | 6.50 | 171.00 | 2 | $$$ | 0.06 | 0.04 |
| NITROBENZENE | 19.0 | 8.4 | 3.7 | 5.76 | 210.80 | 3 | $ | 0.88 | 0.80 |
| METHYLENE DIIODIDE | 16.9 | 3.8 | 5.0 | 5.20 | 181.00 | 2 | $$ | 0.76 | 0.69 |
| 4-BROMOPHENETOLE | 18.6 | 7.6 | 4.8 | 4.00 | 230.85 | 1 | $ | 0.29 | 0.27 |
| DIBENZYL ETHER | 16.5 | 3.6 | 6.7 | 3.60 | 288.30 | 2 | $ | 0.95 | 0.86 |
| DIMETHYL PHTHALATE | 17.7 | 10.6 | 4.5 | 2.00 | 284.00 | 1 | $ | 0.95 | 0.88 |
| o-DIBROMOBENZENE | 19.7 | 6.4 | 4.8 | 1.80 | 221.00 | - | $$ | 0.94 | 0.88 |
| o-BROMOANISOLE | 18.8 | 8.2 | 6.1 | 1.30 | 216.00 | 2 | $ | 0.34 | 0.33 |

**Table 3A** PLA dissolution candidates and evaluated data, for PLA HSP taken from [3]

| **Solvent** | $\delta_D$ **(Heated)** ($Mpa^{1/2}$) | $\delta_P$ **(Heated)** ($Mpa^{1/2}$) | $\delta_H$ **(Heated)** ($Mpa^{1/2}$) | $T_m$ (°C) | $T_b$ (°C) | **Toxicity** | **Costs** | **RED** | **RED (Heated)** |
|---|---|---|---|---|---|---|---|---|---|
| 2,4,5-TRICHLOROTHIOPHENOL | 20.0 | 4.4 | 8.3 | 117.00 | - | 1 | $$$ | 0.59 | 0.54 |
| 3-METHYLINDOLE | 19.0 | 7.0 | 5.7 | 95.00 | 265.00 | 1 | $$$ | 0.50 | 0.49 |
| NAPHTHALENE | 18.3 | 2.0 | 5.4 | 80.28 | 217.99 | 3 | $ | 0.30 | 0.27 |
| 2,4,6-TRICHLOROPHENOL | 19.3 | 5.0 | 9.9 | 69.50 | 244.50 | 3 | $ | 0.95 | 0.82 |
| TRICHLOROBIPHENYL | 18.3 | 5.2 | 3.7 | 61.15 | 347.50 | 3 | $$$ | 0.49 | 0.42 |
| 2,4-DICHLORO-5-NITROBENZOTRIFLUORIDE | 18.9 | 7.5 | 3.4 | 55.00 | - | 1 | $$ | 1.10 | 1.00 |
| INDOLE | 18.8 | 7.4 | 5.9 | 53.00 | 253.00 | 2 | $ | 0.61 | 0.59 |
| p-DICHLOROBENZENE | 18.8 | 5.5 | 2.5 | 52.99 | 174.06 | 3 | $ | 0.93 | 0.79 |
| o-CHLOROTHIOPHENOL | 19.2 | 6.9 | 9.1 | 49.00 | 205.00 | 2 | $$ | 1.03 | 0.93 |
| 4-CHLOROBENZALDEHYDE | 18.9 | 7.1 | 5.1 | 47.50 | 213.50 | 2 | $ | 0.57 | 0.55 |
| p-BROMOBENZOYL CHLORIDE | 19.2 | 6.4 | 5.0 | 42.00 | 247.00 | 2 | $ | 0.42 | 0.40 |
| 3,4-DICHLORONITROBENZENE | 19.1 | 7.1 | 3.7 | 41.00 | 255.00 | - | $$$ | 0.87 | 0.80 |
| PENTAFLUOROBENZOPHENONE | 18.4 | 7.9 | 4.9 | 37.00 | - | 2 | $ | 0.98 | 0.92 |
| 5-BROMO-2-NITROBENZOTRIFLUORIDE | 19.0 | 5.9 | 4.5 | 34.00 | 95.00 | 1 | $$$ | 0.37 | 0.34 |
| METHYL P-TOLUATE | 18.1 | 6.4 | 3.5 | 34.00 | 218.15 | 1 | $ | 0.86 | 0.75 |
| 4-(TRIFLUOROMETHYL) ACETOPHENONE | 17.9 | 6.0 | 3.2 | 31.50 | 180.98 | 1 | $$$ | 0.93 | 0.80 |
| p-DIVINYLBENZENE | 17.7 | 1.0 | 6.4 | 31.00 | 200.55 | 2 | $$$ | 0.74 | 0.67 |
| 2,6-DICHLOROANISOLE | 18.9 | 8.2 | 5.9 | 31.00 | 205.93 | 2 | $$ | 0.97 | 0.94 |
| DIPHENYL ETHER | 18.7 | 3.1 | 5.3 | 26.88 | 258.31 | 2 | $ | 0.08 | 0.07 |
| p-BROMOTOLUENE | 18.4 | 6.7 | 3.7 | 26.80 | 184.35 | 1 | $ | 0.78 | 0.69 |
| 2-BROMOTHIOPHENE | 19.1 | 5.1 | 4.2 | 25.00 | 150.00 | 2 | $ | 0.31 | 0.28 |
| 4-VINYLPYRIDINE | 17.2 | 7.1 | 6.2 | 25.00 | 62.00 | 3 | $ | 1.09 | 0.98 |

**Table 3B** PLA dissolution candidates and evaluated data, for PLA HSP taken from [3] – continuation

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| TRANS-ANETHOLE | 18.1 | 4.2 | 7.9 | 22.20 | 235.50 | 1 | $ | 0.33 | 0.27 |
| 1-CHLORO-4-ETHOXYBENZENE | 18.4 | 6.2 | 4.0 | 21.00 | 213.00 | 2 | $$$ | 0.58 | 0.51 |
| p-CHLOROACETOPHENONE | 18.7 | 7.5 | 3.7 | 20.00 | 232.00 | 2 | $ | 1.02 | 0.93 |
| PENTAMETHYLENE SULFIDE | 17.6 | 6.2 | 8.1 | 19.00 | 142.00 | 2 | $$$ | 0.84 | 0.74 |
| 1,2,4-TRICHLOROBENZENE | 19.2 | 5.9 | 2.9 | 16.89 | 213.50 | 2 | $ | 0.84 | 0.74 |
| p-CHLOROBENZOYL CHLORIDE | 18.9 | 6.6 | 4.7 | 16.00 | 246.70 | 1 | $ | 0.49 | 0.46 |
| 4-ACETYLMORPHOLINE | 17.4 | 5.2 | 7.1 | 14.50 | 207.97 | 2 | $$ | 0.60 | 0.51 |
| 1,4-DIOXANE | 18.1 | 1.8 | 6.8 | 11.77 | 101.28 | 3 | $ | 0.39 | 0.35 |
| 2,6-DICHLOROANISOLE | 18.8 | 8.2 | 5.9 | 10.10 | 205.93 | 1 | $$$ | 0.97 | 0.94 |
| ETHYLENE DIBROMIDE | 18.3 | 3.4 | 7.9 | 10.00 | 131.00 | 3 | $ | 0.27 | 0.22 |
| BROMOFORM | 20.4 | 4.0 | 5.6 | 9.00 | 149.50 | 2 | $$ | 0.53 | 0.51 |
| 2,5-DIMETHYLPYRROLE | 17.4 | 7.5 | 6.2 | 6.50 | 171.00 | 2 | $$$ | 1.10 | 1.00 |
| 4-CHLOROBENZOTRICHLORIDE | 19.3 | 5.4 | 3.2 | 5.80 | 245.00 | 3 | $ | 0.67 | 0.59 |
| METHYLENE DIIODIDE | 16.9 | 3.8 | 5.0 | 5.20 | 181.00 | 2 | $$ | 0.87 | 0.74 |
| 4-BROMOPHENETOLE | 18.6 | 7.6 | 4.8 | 4.00 | 230.85 | 1 | $ | 0.80 | 0.75 |
| o-DIBROMOBENZENE | 19.7 | 6.4 | 4.8 | 1.80 | 221.00 | - | $$ | 0.57 | 0.55 |
| o-BROMOANISOLE | 18.8 | 8.2 | 6.1 | 1.30 | 216.00 | 2 | $ | 0.97 | 0.93 |

**Table 4A** PLA dissolution candidates and data, for PLA HSP taken from [4]

| **Solvent** | $\delta_D$ **(Heated)** ($Mpa^{1/2}$) | $\delta_P$ **(Heated)** ($Mpa^{1/2}$) | $\delta_H$ **(Heated)** ($Mpa^{1/2}$) | $T_m$ (°C) | $T_b$ (°C) | **Toxicity** | **Costs** | **RED** | **RED (Heated)** |
|---|---|---|---|---|---|---|---|---|---|
| BENZOIC ACID | 17.3 | 6.8 | 9.0 | 122.37 | 249.05 | 2 | $ | 0.59 | 0.53 |
| 2,4,5-TRICHLOROTHIOPHENOL | 20.0 | 4.4 | 8.3 | 117.00 | - | 1 | $$$ | 1.01 | 0.95 |
| 3-METHYLINDOLE | 19.0 | 7.0 | 5.7 | 95.00 | 265.00 | 1 | $$$ | 0.66 | 0.63 |
| NAPHTHALENE | 18.3 | 2.0 | 5.4 | 80.28 | 217.99 | 3 | $ | 0.51 | 0.47 |
| TETRAMETHYLTHIOUREA | 16.5 | 5.9 | 9.6 | 79.30 | 245.00 | 2 | $$ | 1.01 | 0.87 |
| 2,4,6-TRICHLOROPHENOL | 19.3 | 5.0 | 9.9 | 69.50 | 244.50 | 3 | $ | 0.92 | 0.83 |
| TRICHLOROBIPHENYL | 18.3 | 5.2 | 3.7 | 61.15 | 347.50 | 3 | $$$ | 0.66 | 0.56 |
| METHOXY ANILINE | 18.9 | 6.4 | 10.3 | 57.70 | 243.00 | 3 | $ | 1.07 | 0.95 |
| TRICHLOROACETIC ACID | 17.4 | 5.7 | 10.4 | 56.85 | 197.63 | 2 | $$$ | 0.86 | 0.72 |
| INDOLE | 18.8 | 7.4 | 5.9 | 53.00 | 253.00 | 2 | $ | 0.65 | 0.63 |
| o-CHLOROTHIOPHENOL | 19.2 | 6.9 | 9.1 | 49.00 | 205.00 | 2 | $$ | 0.92 | 0.86 |
| 4-CHLOROBENZALDEHYDE | 18.9 | 7.1 | 5.1 | 47.50 | 213.50 | 2 | $ | 0.75 | 0.70 |
| p-BROMOBENZOYL CHLORIDE | 19.2 | 6.4 | 5.0 | 42.00 | 247.00 | 2 | $ | 0.75 | 0.70 |
| PENTAFLUOROBENZOPHENONE | 18.4 | 7.9 | 4.9 | 37.00 | - | 2 | $ | 0.89 | 0.83 |
| 5-BROMO-2-NITROBENZOTRIFLUORIDE | 19.0 | 5.9 | 4.5 | 34.00 | 95.00 | 1 | $$$ | 0.71 | 0.65 |
| METHYL P-TOLUATE | 18.1 | 6.4 | 3.5 | 34.00 | 218.15 | 1 | $ | 0.91 | 0.78 |
| 4-(TRIFLUOROMETHYL) ACETOPHENONE | 17.9 | 6.0 | 3.2 | 31.50 | 180.98 | 1 | $$$ | 0.96 | 0.81 |
| p-DIVINYLBENZENE | 17.7 | 1.0 | 6.4 | 31.00 | 200.55 | 2 | $$$ | 0.69 | 0.65 |
| 2,6-DICHLOROANISOLE | 18.9 | 8.2 | 5.9 | 31.00 | 205.93 | 2 | $$ | 0.99 | 0.95 |
| DIPHENYL ETHER | 18.7 | 3.1 | 5.3 | 26.88 | 258.31 | 2 | $ | 0.37 | 0.34 |
| p-BROMOTOLUENE | 18.4 | 6.7 | 3.7 | 26.80 | 184.35 | 1 | $ | 0.89 | 0.78 |

**Table 4B** PLA dissolution candidates and data, for PLA HSP taken from [4] – continuation

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| 4-VINYLPYRIDINE | 17.2 | 7.1 | 6.2 | 25.00 | 62.00 | 3 | $ | 0.49 | 0.45 |
| 2-BROMOTHIOPHENE | 19.1 | 5.1 | 4.2 | 25.00 | 150.00 | 2 | $ | 0.77 | 0.69 |
| TRANS-ANETHOLE | 18.1 | 4.2 | 7.9 | 22.20 | 235.50 | 1 | $ | 0.07 | 0.06 |
| 1-CHLORO-4-ETHOXYBENZENE | 18.4 | 6.2 | 4.0 | 21.00 | 213.00 | 2 | $$$ | 0.69 | 0.61 |
| PENTAMETHYLENE SULFIDE | 17.6 | 6.2 | 8.1 | 19.00 | 142.00 | 2 | $$$ | 0.25 | 0.23 |
| p-CHLOROBENZOYL CHLORIDE | 18.9 | 6.6 | 4.7 | 16.00 | 246.70 | 1 | $ | 0.74 | 0.68 |
| 4-ACETYLMORPHOLINE | 17.4 | 5.2 | 7.1 | 14.50 | 207.97 | 2 | $$ | 0.09 | 0.07 |
| 1,4-DIOXANE | 18.1 | 1.8 | 6.8 | 11.77 | 101.28 | 3 | $ | 0.40 | 0.39 |
| 2,6-DICHLOROANISOLE | 18.8 | 8.2 | 5.9 | 10.10 | 205.93 | 1 | $$$ | 0.95 | 0.92 |
| ETHYLENE DIBROMIDE | 18.3 | 3.4 | 7.9 | 10.00 | 131.00 | 3 | $ | 0.13 | 0.13 |
| 1-DECANOL | 16.7 | 2.5 | 9.1 | 6.75 | 229.85 | 2 | $ | 0.87 | 0.75 |
| 2,5-DIMETHYLPYRROLE | 17.4 | 7.5 | 6.2 | 6.50 | 171.00 | 2 | $$$ | 0.55 | 0.51 |
| METHYLENE DIIODIDE | 16.9 | 3.8 | 5.0 | 5.20 | 181.00 | 2 | $$ | 0.48 | 0.40 |
| 4-BROMOPHENETOLE | 18.6 | 7.6 | 4.8 | 4.00 | 230.85 | 1 | $ | 0.82 | 0.76 |
| DIBENZYL ETHER | 16.5 | 3.6 | 6.7 | 3.60 | 288.30 | 2 | $$ | 0.55 | 0.46 |
| o-BROMOANISOLE | 18.8 | 8.2 | 6.1 | 1.30 | 216.00 | 2 | $ | 0.93 | 0.90 |

**Table 5A** PLA dissolution candidates and data, for PLA HSP taken from [5]

| **Solvent** | $\delta_D$ **(Heated)** ($Mpa^{1/2}$) | $\delta_P$ **(Heated)** ($Mpa^{1/2}$) | $\delta_H$ **(Heated)** ($Mpa^{1/2}$) | $T_m$ (°C) | $T_b$ (°C) | **Toxicity** | **Costs** | **RED** | **RED (Heated)** |
|---|---|---|---|---|---|---|---|---|---|
| p-BROMONITROBENZENE | 19.9 | 9.7 | 5.4 | 127.00 | 256.00 | 1 | $$$ | 1.06 | 1.00 |
| p-BROMOBENZONITRILE | 19.4 | 9.1 | 5.3 | 113.00 | 237.00 | 2 | $$ | 0.67 | 0.64 |
| 4-CHLOROBENZONITRILE | 18.6 | 7.8 | 3.7 | 95.00 | 223.00 | 2 | $$ | 0.52 | 0.49 |
| 3-METHYLINDOLE | 19.0 | 7.0 | 5.7 | 95.00 | 265.00 | 1 | $$$ | 0.79 | 0.76 |
| 3,5-DIFLUOROBENZONITRILE | 17.9 | 11.0 | 2.9 | 86.50 | 188.63 | 2 | $$$ | 0.48 | 0.42 |
| p-CHLORONITROBENZENE | 19.4 | 9.4 | 3.8 | 83.50 | 242.00 | 3 | $ | 0.81 | 0.76 |
| 2,4-DICHLOROBENZALDEHYDE | 18.8 | 8.6 | 4.9 | 71.00 | 233.00 | 1 | $ | 0.32 | 0.31 |
| 1,3,5-TRIOXANE | 17.8 | 9.0 | 7.9 | 64.00 | 114.50 | 2 | $ | 0.36 | 0.31 |
| 2,3-DICHLORONITROBENZENE | 18.8 | 12.4 | 4.0 | 62.00 | 258.00 | 2 | $$$ | 0.73 | 0.70 |
| 2,4-DICHLORO-5-NITROBENZOTRIFLUORIDE | 18.9 | 7.5 | 3.4 | 55.00 | - | 1 | $$ | 0.87 | 0.80 |
| INDOLE | 18.8 | 7.4 | 5.9 | 53.00 | 253.00 | 2 | $ | 0.59 | 0.57 |
| p-NITROTOLUENE | 19.1 | 9.4 | 3.6 | 51.60 | 238.50 | 3 | $ | 0.68 | 0.62 |
| 4-CHLOROBENZALDEHYDE | 18.9 | 7.1 | 5.1 | 47.50 | 213.50 | 2 | $ | 0.71 | 0.69 |
| 3,4-DICHLORONITROBENZENE | 19.1 | 7.1 | 3.7 | 41.00 | 255.00 | - | $$$ | 1.00 | 0.93 |
| 4-CHLORO-2-NITROTOLUENE | 18.9 | 11.6 | 3.5 | 36.54 | 266.35 | 1 | $ | 0.76 | 0.71 |
| 3,4-DICHLOROPHENYLACETONITRILE | 19.5 | 10.6 | 4.0 | 38.00 | - | 2 | $$ | 0.89 | 0.84 |
| 4-METHOXYACETOPHENONE | 18.0 | 11.0 | 6.4 | 37.50 | 255.00 | 1 | $$ | 0.15 | 0.15 |
| 4-ETHOXYACETOPHENONE | 17.9 | 10.1 | 5.8 | 37.00 | 268.50 | 2 | $$$ | 0.02 | 0.03 |
| PENTAFLUOROBENZOPHENONE | 18.4 | 7.9 | 4.9 | 37.00 | - | 2 | $ | 0.28 | 0.27 |
| 1,3-DICHLORO-2-FLUOROBENZENE | 18.5 | 8.9 | 2.5 | 37.00 | - | 1 | $$$ | 0.70 | 0.61 |
| 6-CHLORO-2-NITROTOLUENE | 19.3 | 9.4 | 3.5 | 34.00 | 238.00 | 2 | $ | 0.82 | 0.76 |

**Table 5B** PLA dissolution candidates and data, for PLA HSP taken from [5] – continuation

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| METHYL P-TOLUATE | 18.1 | 6.4 | 3.5 | 34.00 | 218.15 | 1 | $ | 0.85 | 0.79 |
| 4-(TRIFLUOROMETHYL) ACETOPHENONE | 17.9 | 6.0 | 3.2 | 31.50 | 180.98 | 1 | $$$ | 1.04 | 0.96 |
| 2,6-DICHLOROANISOLE | 18.9 | 8.2 | 5.9 | 31.00 | 205.93 | 2 | $$ | 0.46 | 0.45 |
| 2,6-DIFLUOROBENZONITRILE | 17.9 | 11.0 | 2.9 | 30.00 | 197.00 | 2 | $$$ | 0.48 | 0.42 |
| CYCLOOCTANONE | 16.2 | 9.4 | 4.1 | 29.00 | 196.00 | 2 | $ | 0.63 | 0.53 |
| 2,4-DICHLORONITROBENZENE | 19.4 | 8.5 | 3.8 | 29.00 | 258.00 | 3 | $ | 0.88 | 0.82 |
| p-BROMOTOLUENE | 18.4 | 6.7 | 3.7 | 26.80 | 184.35 | 1 | $ | 0.76 | 0.71 |
| 4-VINYLPYRIDINE | 17.2 | 7.1 | 6.2 | 25.00 | 62.00 | 3 | $ | 0.45 | 0.41 |
| CYCLOHEPTANONE | 16.4 | 10.4 | 4.4 | 25.00 | 178.50 | 1 | $$ | 0.49 | 0.41 |
| DIISOPROPYL METHYLPHOSPHONATE | 15.6 | 9.8 | 5.2 | 25.00 | 121.10 | 2 | $$$ | 0.97 | 0.84 |
| DIPROPYL SULFOXIDE | 16.2 | 12.7 | 6.8 | 24.75 | 201.85 | 1 | $$$ | 1.09 | 0.98 |
| 2-CHLOROCYCLOHEXANONE | 17.6 | 12.7 | 4.7 | 23.00 | 161.50 | 1 | $$$ | 0.52 | 0.50 |
| CYCLODECANONE | 16.0 | 7.8 | 3.7 | 22.85 | 236.05 | 1 | $$$ | 1.01 | 0.87 |
| 1-CHLORO-4-ETHOXYBENZENE | 18.4 | 6.2 | 4.0 | 21.00 | 213.00 | 2 | $$$ | 0.87 | 0.83 |
| p-CHLOROACETOPHENONE | 18.7 | 7.5 | 3.7 | 20.00 | 232.00 | 2 | $ | 0.66 | 0.62 |
| ACETOPHENONE | 18.7 | 8.4 | 3.4 | 19.45 | 202.11 | 1 | $ | 0.55 | 0.50 |
| PENTAMETHYLENE SULFIDE | 17.6 | 6.2 | 8.1 | 19.00 | 142.00 | 2 | $$$ | 1.07 | 0.97 |
| p-CHLOROBENZOYL CHLORIDE | 18.9 | 6.6 | 4.7 | 16.00 | 246.70 | 1 | $ | 0.89 | 0.85 |
| VINYL PYRROLIDONE | 15.6 | 9.1 | 5.4 | 13.00 | 219.00 | 3 | $ | 0.99 | 0.85 |
| TRICRESYL PHOSPHATE | 18.1 | 12.1 | 4.1 | 11.00 | 410.00 | 3 | $ | 0.43 | 0.41 |
| 2,6-DICHLOROANISOLE | 18.8 | 8.2 | 5.9 | 10.10 | 205.93 | 1 | $$$ | 0.41 | 0.40 |
| 2,4-DIFLUORONITROBENZENE | 18.5 | 10.8 | 3.4 | 9.00 | 203.00 | 2 | $$ | 0.45 | 0.41 |
| 2,5-DIMETHYLPYRROLE | 17.4 | 7.5 | 6.2 | 6.50 | 171.00 | 2 | $$$ | 0.31 | 0.29 |

**Table 5C** PLA dissolution candidates and data, for PLA HSP taken from [5] – continuation

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| ETHYL CINNAMATE | 17.5 | 8.0 | 3.7 | 6.00 | 271.00 | 1 | $ | 0.33 | 0.29 |
| NITROBENZENE | 19.0 | 8.4 | 3.7 | 5.76 | 210.80 | 3 | $ | 0.66 | 0.61 |
| 4-BROMOPHENETOLE | 18.6 | 7.6 | 4.8 | 4.00 | 230.85 | 1 | $ | 0.43 | 0.42 |
| o-BROMOANISOLE | 18.8 | 8.2 | 6.1 | 1.30 | 216.00 | 2 | $ | 0.43 | 0.41 |
| DIMETHYL PHTHALATE | 17.7 | 10.6 | 4.5 | 1.03 | 283.70 | 1 | $ | 0.10 | 0.09 |

## 2. SUPPLEMENTARY MEASUREMENT DATA

**Table 6** Cloud point measurement data used in the PCL/MPTOL phase diagram construction, where $\varphi_{PCL}$ stands for volume fraction of PCL in the solution, $T_{cloud}$ stands for measured cloud point temperature and $T_{dev}$ stand for corresponding standard deviation of measured $T_{cloud}$ .

| $\phi_{PCL}$ (-) | 0.045 | 0.065 | 0.084 | 0.090 | 0.093 | 0.093 | 0.111 | 0.112 | 0.116 | 0.133 | 0.139 | 0.151 | 0.159 | 0.169 | 0.178 | 0.188 | 0.206 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $T_{cloud}$ (°C) | -4.380 | -3.090 | -2.000 | 2.210 | -0.430 | 3.370 | 1.870 | 2.230 | 2.540 | 4.520 | 4.800 | 7.320 | 6.000 | 9.340 | 8.890 | 10.040 | 10.190 |
| $T_{std}$ (°C) | 0.470 | 7.110 | 0.190 | 0.450 | 1.110 | 0.720 | 0.160 | 0.850 | 1.060 | 0.090 | 0.070 | 0.630 | 0.300 | 0.260 | 1.220 | 0.110 | 0.250 |

**Table 7** Freezing point measurement data used in the PCL/MPTOL phase diagram construction, where $\varphi_{PCL}$ stands for volume fraction of PCL in the solution, $T_{freeze}$ stands for measured freezing point temperature and $T_{dev}$ stand for corresponding standard deviation of measured $T_{freeze}$.

| $\varphi_{PCL}$ (-) | 0.038 | 0.055 | 0.093 | 0.113 | 0.135 | 0.168 |
|---|---|---|---|---|---|---|
| $T_{freeze}$ (°C) | -3.140 | -0.870 | -3.840 | -8.140 | -9.490 | -11.050 |
| $T_{std}$ (°C) | 4.990 | 0.700 | 0.670 | 0.290 | 1.270 | 1.240 |

**Table 8** Porous sample characterization data, where $d_p$ denotes frequency pore size average, $A_{pore}$ stands for specific pore surface area and suffix ($_{std}$) denotes standard deviation of the respective measurement.

| $w_{PCL}$ | $d_p$ | $d_{p\text{-std}}$ | Porosity | Porosity$_{std}$ | $A_{pore}$ | $A_{pore\text{-std}}$ |
|---|---|---|---|---|---|---|
| (-) | (nm) | (nm) | (%) | (%) | ($m^2$/g) | ($m^2$/g) |
| 0.12 | 58.1 | 28.4 | 71.8 | 2.7 | 19.8 | 3.7 |
| 0.14 | 99 | 54.6 | 72.7 | 9.2 | 18.0 | 3.1 |
| 0.16 | 64.1 | 48.2 | 74.0 | 7.8 | 22.4 | 9.1 |

**Table 9** Data, used in the manuscript, for comparison of PCL solution wt.% influence on pore volume percentage (Vol. perc.) at different pore sizes $d_p$. Where values of $d_p$ denote pore range in ± 0.5 µm, e.g. $d_p$= 0.5 µm describes range from 0 – 1 µm.

| 12 PCL wt.% | | | 14 PCL wt.% | | | 16 PCL wt.% | | |
|---|---|---|---|---|---|---|---|---|
| $d_p$ (µm) | Vol. perc. (%) | Vol perc.$_{std}$ (%) | $d_p$ (µm) | Vol. perc. (%) | Vol perc.$_{std}$ (%) | $d_p$ (µm) | Vol. perc. (%) | Vol perc.$_{std}$ (%) |
| 0.5 | 31.14 | 2.86 | 0.5 | 48.58 | 8.79 | 0.5 | 38.48 | 5.12 |
| 1.5 | 16.30 | 1.58 | 1.5 | 20.26 | 0.55 | 1.5 | 20.36 | 1.11 |
| 2.5 | 10.35 | 0.42 | 2.5 | 5.85 | 5.09 | 2.5 | 10.83 | 2.18 |
| 3.5 | 5.76 | 0.20 | 3.5 | 3.13 | 1.01 | 3.5 | 4.47 | 0.86 |
| 4.5 | 3.21 | 0.28 | 4.5 | 1.49 | 0.75 | 4.5 | 4.48 | 4.25 |
| 5.5 | 3.26 | 0.36 | 5.5 | 1.51 | 0.58 | 5.5 | 1.62 | 0.72 |
| 6.5 | 2.57 | 0.30 | 6.5 | 1.09 | 0.49 | 6.5 | 1.14 | 0.52 |
| 7.5 | 1.88 | 0.16 | 7.5 | 0.91 | 0.36 | 7.5 | 0.94 | 0.28 |
| 8.5 | 1.53 | 0.08 | 8.5 | 0.67 | 0.29 | 8.5 | 0.76 | 0.27 |
| 9.5 | 1.21 | 0.14 | 9.5 | 0.62 | 0.17 | 9.5 | 0.66 | 0.26 |
| 10.5 | 1.05 | 0.07 | 10.5 | 0.58 | 0.17 | 10.5 | 0.59 | 0.14 |
| 11.5 | 0.99 | 0.17 | 11.5 | 0.52 | 0.20 | 11.5 | 0.53 | 0.20 |
| 12.5 | 0.85 | 0.14 | 12.5 | 0.44 | 0.18 | 12.5 | 0.55 | 0.22 |
| 13.5 | 0.83 | 0.09 | 13.5 | 0.46 | 0.12 | 13.5 | 0.49 | 0.11 |
| 14.5 | 0.73 | 0.18 | 14.5 | 0.45 | 0.21 | 14.5 | 0.45 | 0.13 |
| 15.5 | 0.62 | 0.08 | 15.5 | 0.41 | 0.08 | 15.5 | 0.41 | 0.13 |
| 16.5 | 0.61 | 0.05 | 16.5 | 0.40 | 0.03 | 16.5 | 0.36 | 0.09 |
| 17.5 | 0.60 | 0.18 | 17.5 | 0.37 | 0.13 | 17.5 | 0.36 | 0.10 |
| 18.5 | 0.54 | 0.06 | 18.5 | 0.36 | 0.12 | 18.5 | 0.30 | 0.10 |
| 19.5 | 0.52 | 0.08 | 19.5 | 0.33 | 0.05 | 19.5 | 0.31 | 0.09 |

**Table 10** Data, used in the manuscript, for comparison of PCL solution pore frequency at different pore sizes $d_p$. Where values of $d_p$ denote pore range in ± 5 nm, e.g. $d_p$ = 5 nm describes range from 0 – 10 nm.

| 12 PCL wt.% | | | 14 PCL wt.% | | | 16 PCL wt.% | | |
|---|---|---|---|---|---|---|---|---|
| $d_p$ (nm) | Frequency (%) | Frequency$_{std}$ (%) | $d_p$ (nm) | Frequency (%) | Frequency$_{std}$ (%) | $d_p$ (nm) | Frequency (%) | Frequency$_{std}$ (%) |
| 5 | 35.26 | 23.83 | 5 | 8.65 | 14.94 | 5 | 22.04 | 37.95 |
| 15 | 20.92 | 28.15 | 15 | 32.88 | 25.77 | 15 | 23.99 | 24.60 |
| 25 | 3.32 | 3.29 | 25 | 8.97 | 14.30 | 25 | 13.05 | 15.40 |
| 35 | 4.56 | 4.99 | 35 | 2.62 | 4.54 | 35 | 7.37 | 3.34 |
| 45 | 5.93 | 4.27 | 45 | 2.86 | 4.96 | 45 | 4.26 | 3.71 |
| 55 | 6.38 | 8.15 | 55 | 5.97 | 7.30 | 55 | 3.38 | 5.10 |
| 65 | 5.31 | 5.44 | 65 | 2.37 | 1.45 | 65 | 3.40 | 5.10 |
| 75 | 1.45 | 0.84 | 75 | 2.74 | 3.88 | 75 | 2.86 | 4.14 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 85 | 2.79 | 2.95 | 85 | 4.95 | 5.26 | 85 | 3.15 | 4.69 |
| 95 | 2.03 | 0.58 | 95 | 1.75 | 1.27 | 95 | 1.21 | 1.46 |
| 105 | 2.25 | 2.13 | 105 | 2.33 | 3.00 | 105 | 1.85 | 2.62 |
| 115 | 0.50 | 0.45 | 115 | 0.92 | 1.13 | 115 | 1.39 | 1.90 |
| 125 | 0.66 | 0.61 | 125 | 1.42 | 2.08 | 125 | 1.43 | 2.06 |
| 135 | 1.04 | 1.34 | 135 | 0.95 | 0.66 | 135 | 0.86 | 1.12 |
| 145 | 0.51 | 0.39 | 145 | 1.09 | 0.90 | 145 | 0.65 | 0.76 |
| 155 | 0.51 | 0.28 | 155 | 0.72 | 0.66 | 155 | 0.78 | 0.93 |
| 165 | 0.58 | 0.43 | 165 | 0.75 | 0.42 | 165 | 0.73 | 0.75 |
| 175 | 0.43 | 0.33 | 175 | 0.68 | 0.66 | 175 | 0.62 | 0.83 |
| 185 | 0.41 | 0.36 | 185 | 0.73 | 0.59 | 185 | 0.47 | 0.72 |
| 195 | 0.33 | 0.30 | 195 | 0.56 | 0.32 | 195 | 0.48 | 0.65 |
| 205 | 0.34 | 0.21 | 205 | 0.45 | 0.24 | 205 | 0.38 | 0.49 |
| 215 | 0.32 | 0.25 | 215 | 0.42 | 0.22 | 215 | 0.32 | 0.44 |
| 225 | 0.26 | 0.19 | 225 | 0.40 | 0.21 | 225 | 0.29 | 0.37 |
| 235 | 0.25 | 0.21 | 235 | 0.52 | 0.45 | 235 | 0.29 | 0.37 |
| 245 | 0.19 | 0.16 | 245 | 0.53 | 0.45 | 245 | 0.26 | 0.32 |
| 255 | 0.17 | 0.14 | 255 | 0.39 | 0.22 | 255 | 0.29 | 0.38 |
| 265 | 0.19 | 0.15 | 265 | 0.32 | 0.19 | 265 | 0.19 | 0.24 |
| 275 | 0.15 | 0.12 | 275 | 0.68 | 0.92 | 275 | 0.23 | 0.30 |
| 285 | 0.17 | 0.14 | 285 | 1.46 | 2.30 | 285 | 0.19 | 0.24 |
| 295 | 0.13 | 0.09 | 295 | 2.62 | 4.28 | 295 | 0.18 | 0.23 |
| 305 | 0.12 | 0.08 | 305 | 2.04 | 3.28 | 305 | 0.18 | 0.22 |
| 315 | 0.12 | 0.10 | 315 | 1.36 | 2.12 | 315 | 0.16 | 0.21 |
| 325 | 0.10 | 0.08 | 325 | 0.68 | 0.94 | 325 | 0.15 | 0.19 |
| 335 | 0.11 | 0.09 | 335 | 0.35 | 0.41 | 335 | 0.11 | 0.13 |
| 345 | 0.09 | 0.08 | 345 | 0.21 | 0.18 | 345 | 0.11 | 0.13 |
| 355 | 0.09 | 0.08 | 355 | 0.18 | 0.11 | 355 | 0.14 | 0.18 |
| 365 | 0.08 | 0.08 | 365 | 0.16 | 0.12 | 365 | 0.10 | 0.14 |
| 375 | 0.07 | 0.04 | 375 | 0.15 | 0.12 | 375 | 0.10 | 0.14 |
| 385 | 0.08 | 0.07 | 385 | 0.16 | 0.11 | 385 | 0.08 | 0.08 |
| 395 | 0.05 | 0.04 | 395 | 0.14 | 0.10 | 395 | 0.10 | 0.13 |

## 3. UNPROCESSED SEM IMAGES USED IN MANUSCRIPT

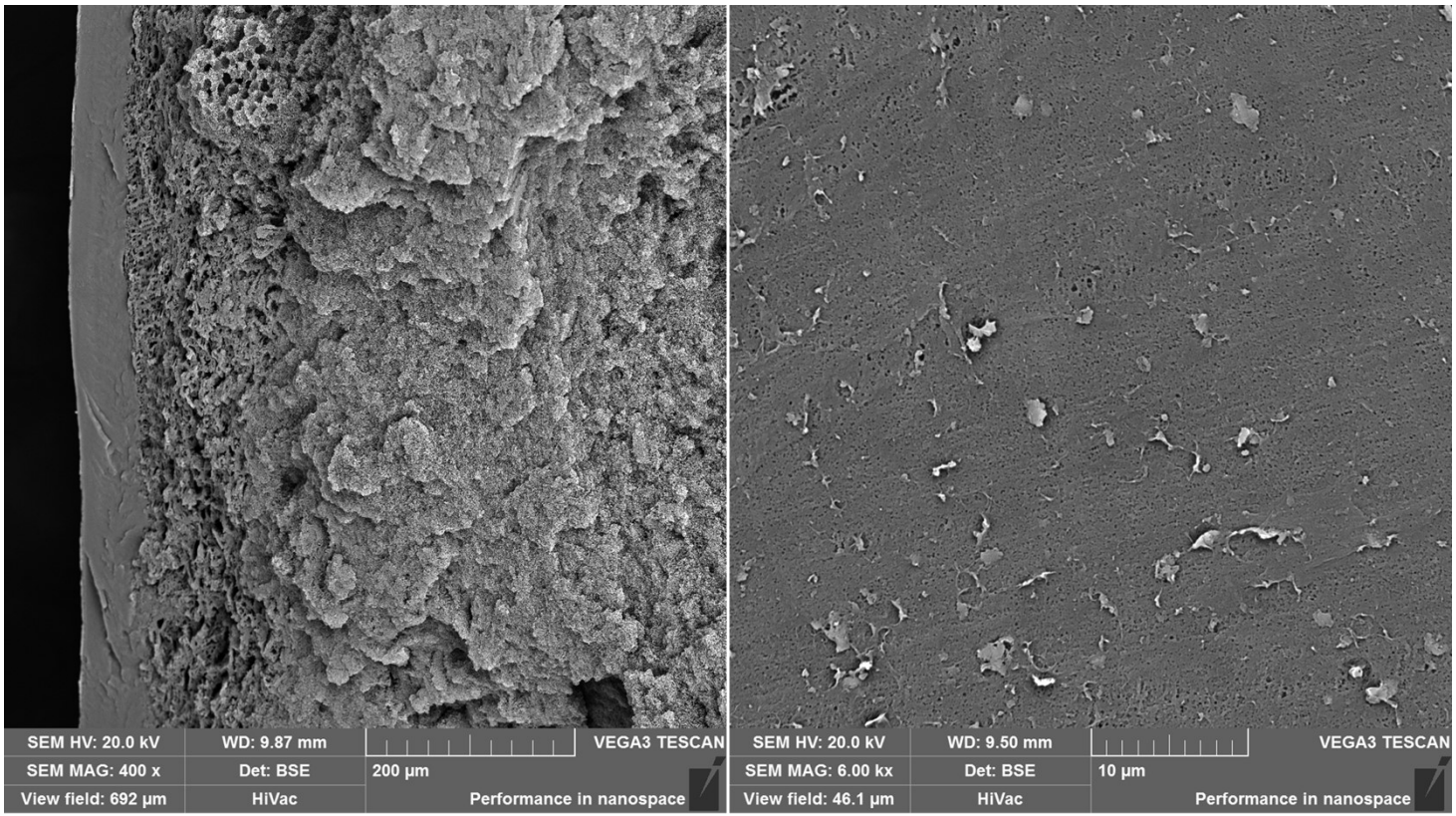


**Figure 1** *unprocessed SEM images used in the manuscript to describe spatial morphology differences.*

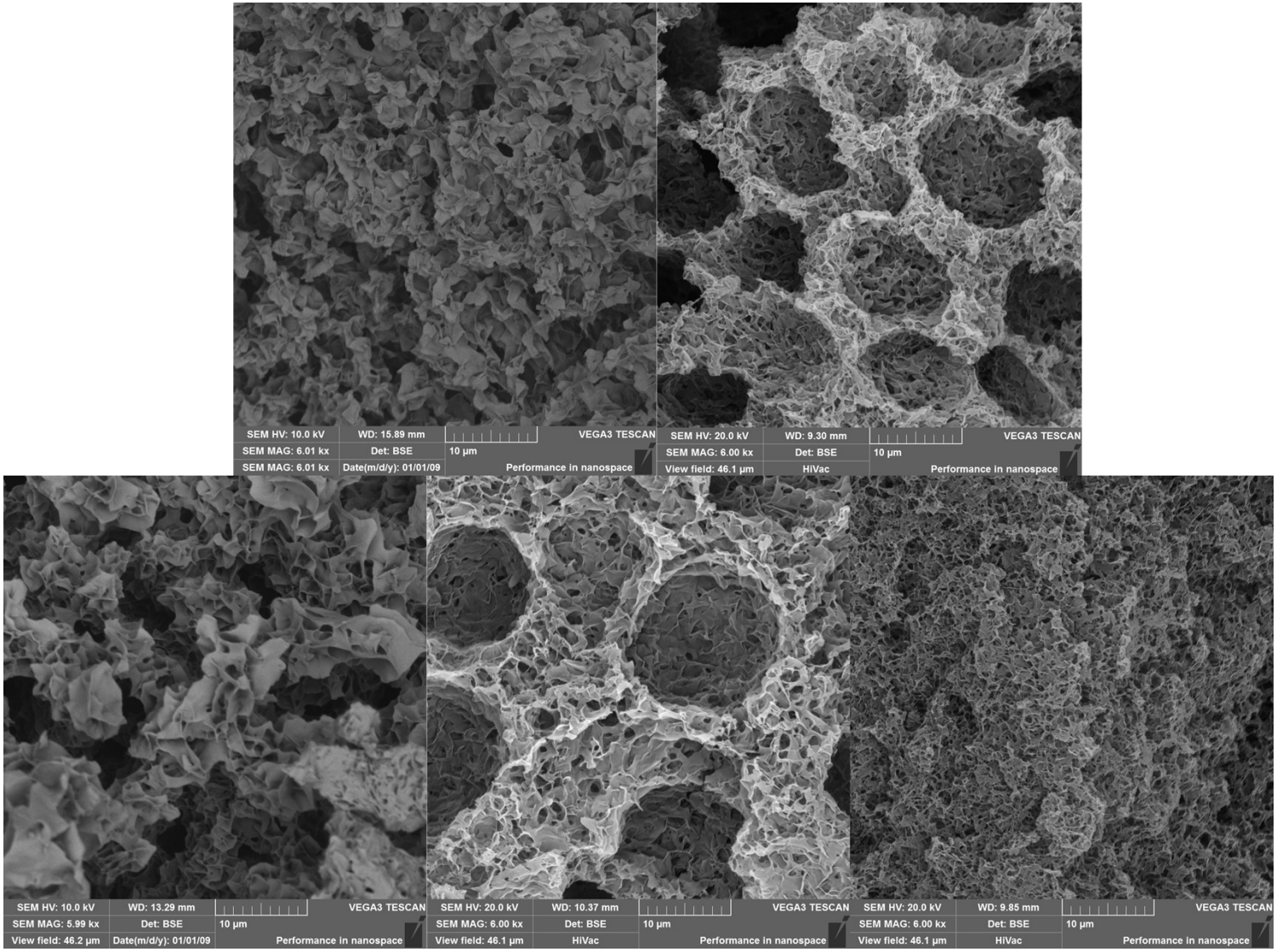


***Figure 2*** *unprocessed SEM images used in the manuscript for description of PCL concentration influence on morphology*.

## 4. REFERENCES


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