

Supplementary data for:
Cyrene™, a sustainable solution for graffiti paint removal

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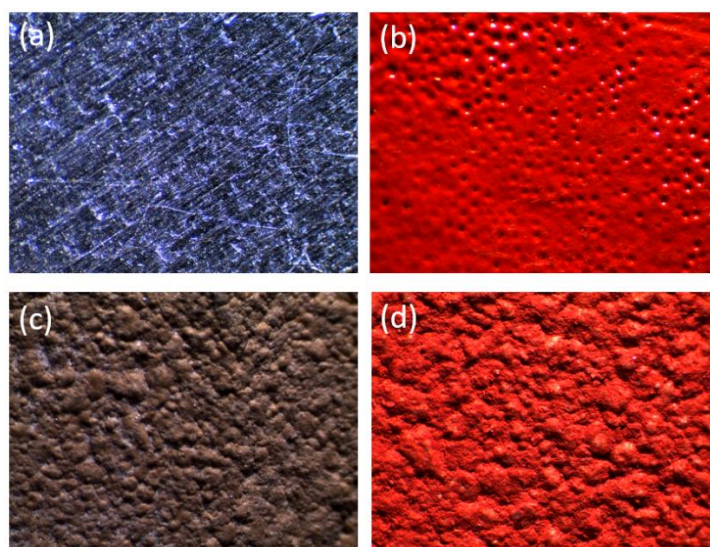


Figure S1. Pristine aluminium foil (a) and ceramic tile (c) and painted substrates (b and d).

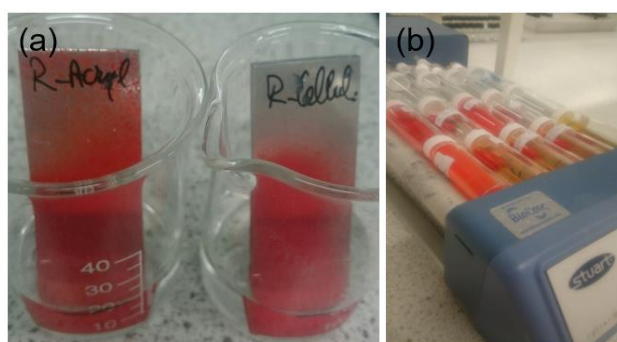


Figure S2. Immersion tests by dip-in (a) and immersion using Stuart rollers (b).

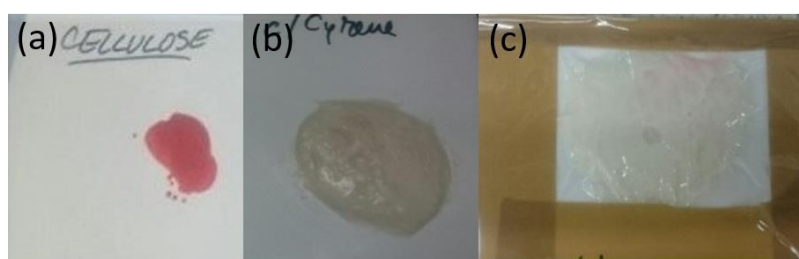


Figure S3. Poultice test: (a) the stained surface was covered by (b) poultices made of solvent and talc and covered using plastic foil. After 24 hours, (c) the plastic foil was removed, and results were assessed.

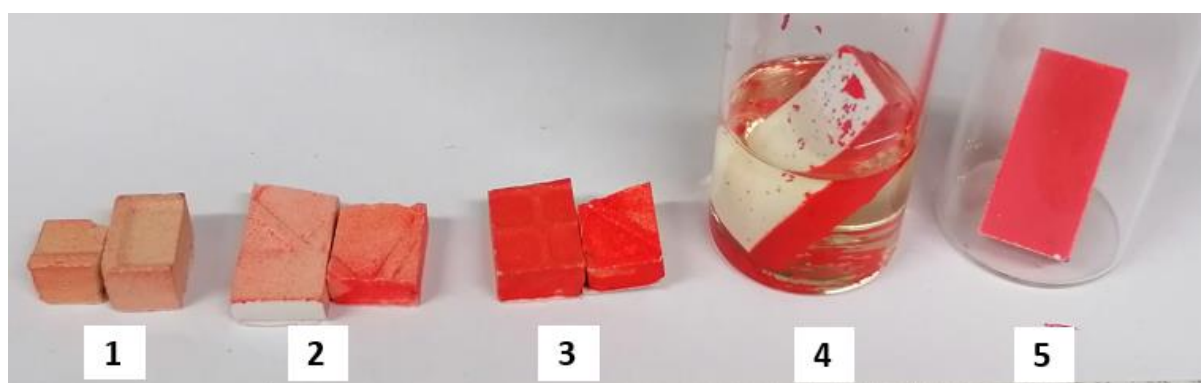


Figure S4. Scores from 1 (good cleaning) to 5 (no change) given to the cleaned tiles.

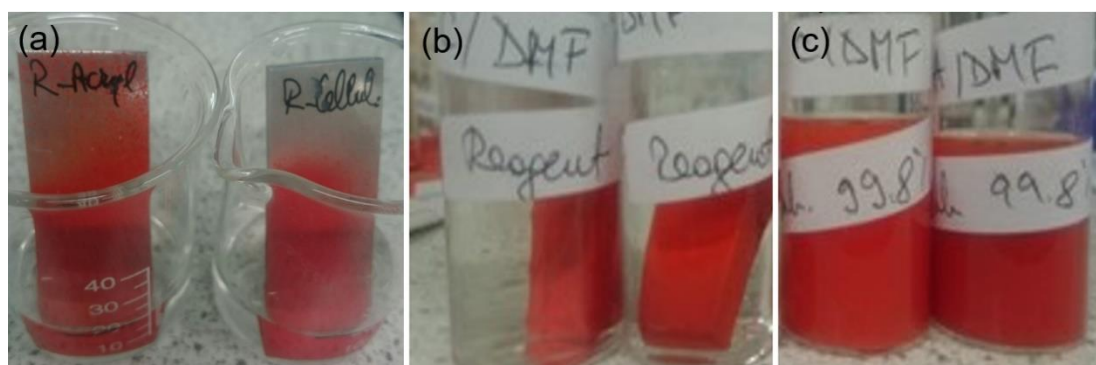


Figure S5. DMF technical grade (a, b) and anhydrous (c) used as paint stripper of acrylic and cellulose-based paints from aluminium foil (a) and ceramic tiles (b, c).

Table S1. Water content determination *via* volumetric Karl Fisher titration.

Sample name	Sample no.	Measured water content (%)	Average water content (%)	Standard deviation (%)
Cyrene 98.5%	1	0.12	0.13	0.01
	2	0.14		
	3	0.13		
NMP 99.5%	1	0.18	0.19	0.01
	2	0.19		
	3	0.2		
DMF min. 98%	1	1.95	1.97	0.03
	2	1.96		
	3	2		
DMF $\geq 99.8\%$	1	0.17	0.12	0.05
	2	0.09		
	3	0.09		
DCM $\geq 99\%$	1	0	<0.01	0
	2	0		
	3	0		
DMAc $\geq 99.5\%$	1	0.22	0.18	0.04
	2	0.16		
	3	0.15		
DMSO $\geq 99\%$	1	0.03	0.03	0.02
	2	0.04		
	3	0.01		

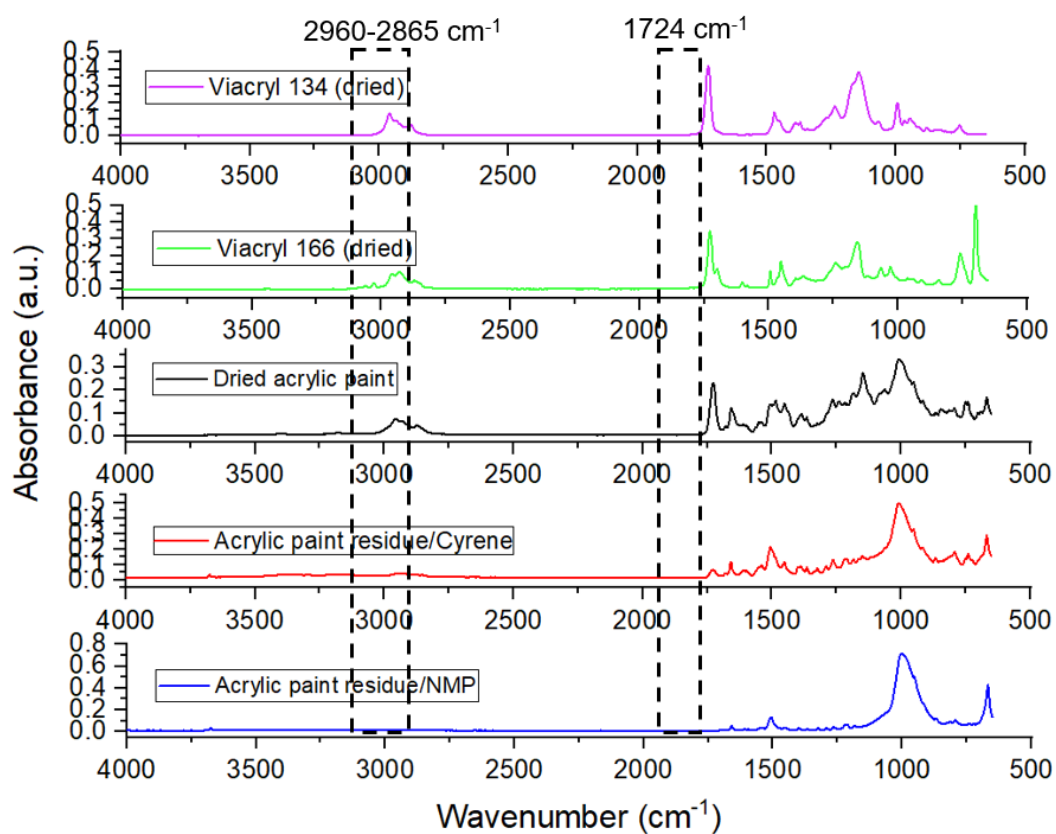


Figure S6. The infrared spectra for the dry acrylic red paint (black line) and residues after removal with Cyrene (red line) and NMP (blue line). Two acrylic resins (green and purple lines) are shown here for comparison.

Table S2. The inorganic content of the acrylic paint residues remaining after dissolution in Cyrene and NMP.

Element	Dried acrylic paint (%)	Solid residue resulted from cleaning using Cyrene	Solid residue resulted from cleaning using NMP
Mg	1	2.3	3.5
Ca	0.3	0.14	0.59
Al	0.2	0.5	0.9
P	0.14	0.07	0.11
Fe	0.08	0.2	0.3
Ba	0.05	0.1	0.2
Na	0.04	0.0026	0.0024
K	0.023	0.027	0.05
Zn	0.003	0.006	0.02
Mn	0.003	0.004	0.01
Cr	0.0003	0.0007	0.001
Pb	0.0003	0.0001	0.0006
Ni	0.0003	0.0007	0.0008
Sn	0.00017	0.0023	0.0004
Cu	0.0001	0.001	0.004
Se	0.00005	<0.0000005	0.00008
Cd	0.000035	0.000026	0.00005
Co	0.00003	0.00007	0.0001
As	0.000027	0.000034	0.0001

< represents a lower than the limit of quantification (LOQ), which is three times the limit of detection (LOD). Numbers in red represent a lower concentration than of the element from the original paint, suggesting the solubility of those elements in solvent. Number in blue and black represent higher concentrations than the elements from the original paint, indicating a chemical change.

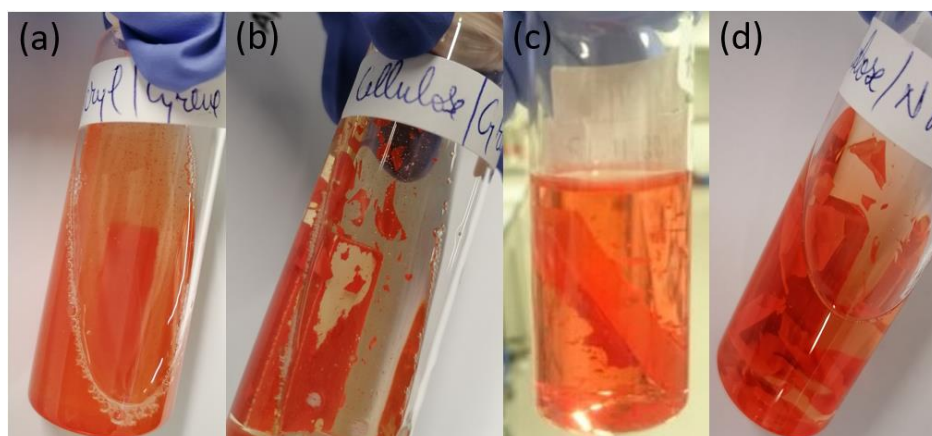


Figure S7. The removal process of acrylic (a, c) and cellulose-based (b, d) paint using Cyrene (a, b) and NMP (c, d) from a tile.

Table S3. Dynamic and apparent viscosities of Cyrene and Cyrene-water mixtures using different methods and the final temperatures after mixing Cyrene and water at different ratios.

Sample	Final temperature (°C) after mixing Cyrene with water (200 mL total volume)	Dynamic viscosity at 20 °C (cP)	Apparent (shear) viscosity at 20 °C (cP)
Cyrene	20	46	11.67
75% Cyrene in water	34	79	28.64
50% Cyrene in water	38	45.8	11.89
25% Cyrene in water	32	16.8	3.67

Only the dynamic viscosity was measured for NMP (12 cP).

Table S4. Hansen Solubility Parameters, the scores given and relative energy distance (RED) of the neat solvents for acrylic paint removal.

Solvent	δ_D	δ_P	δ_H	Score	RED
<i>N</i> -Methyl-2-Pyrrolidone (NMP)	18	12.3	7.2	2**	0.726
<i>N,N'</i> -Dimethylformamide (DMF)	17.4	13.7	11.3	2***	0.784
<i>N,N'</i> -Dimethylacetamide	16.8	11.5	9.4	2	0.993
Cyrene	18.9	12.4	7.1	1	0.96
Dimethyl Sulfoxide (DMSO)	18.4	16.4	10.2	2	0.96
Acetone	15.5	10.4	7	3	1.935
Methylene Chloride (DCM)	17	7.3	7.1	3	2.177
Dimethyl Carbonate	15.5	8.6	9.7	3	2.213
Acetonitrile	15.3	18	6.1	3	2.286
Propylene Carbonate	20	18	4.1	3	2.413
Tetrahydrofuran (THF)	16.8	5.7	8	3	2.628
OME _{e(3-5)}	15.6	7.1	6.1	3	2.669
Benzyl Alcohol	18.4	6.3	13.7	4	2.747
1,1-Dichloroethane	16.5	7.8	3	4	2.764
Ethyl Acetate	15.8	5.3	7.2	3	2.982
Acetic Acid	14.5	8	13.5	4	3.114
2-Methylfuran	17.3	2.8	7.4	4	3.451
Propionic Acid	14.7	5.3	12.4	3	3.467
1-Butanol	16	5.7	15.8	3	3.488
2-Propanol	15.8	6.1	16.4	3	3.566
1-Propanol	16	6.8	17.4	4	3.6
Chlorobenzene	19	4.3	2	3	3.71
Ethanol	15.8	8.8	19.4	3	3.829
2-Pinene (dl)	16.9	1.8	3.1	5	4.186
Diethyl Ether	14.5	2.9	4.6	4	4.214
2,2,5,5,-Tetramethyloxolane (TMO)	15.6	2.3	2.4	4	4.352
Toluene	18	1.4	2	3	4.409
Cyclohexene	17.2	1	2	4	4.54
Methanol	14.7	12.3	22.3	5	4.643
β -Pinene	16.2	1	1.8	4	4.674

Heptane	15.3	0	0	5	5.365
Hexane	14.9	0	0	5	5.447
Lactic Acid (dl)	17	8.3	28.4	5	6.323
Water	15.5	16	42.3	5	10.537

* Wrongly positioned solvents inside or outside the Hansen space. The solvent with scores "1" and "2" are/should be inside the Hansen sphere. The solvents with scores "3-5" are considered bad solvents are are/should be positioned outside the Hansen sphere. Hansen solubility parameters of the solvents are sourced from HSPiP database and expressed in MPa^{1/2}. **NMP received score "2" because despite being covered in a yellow layer, it can be easily removed. ***DMF anhydrous 99.8%.

Table S5. Hansen Solubility Parameters, the scores given and relative energy distance (RED) of the neat solvents for cellulose-based paint removal.

Solvent	δ_D	δ_P	δ_H	Score	RED
Dimethyl Sulfoxide (DMSO)	18.4	16.4	10.2	1	0.312
<i>N,N'</i> -Dimethylformamide (DMF)	17.4	13.7	11.3	1**	0.394
<i>N,N'</i> -Dimethylacetamide (DMAc)	16.8	11.5	9.4	1	0.712
Cyrene	18.9	12.4	7.1	1	0.734
<i>N</i> -Methyl-2-Pyrrolidone (NMP)	18	12.3	7.2	1	0.737
Propylene Carbonate	20	18	4.1	2	1.002*
Benzyl Alcohol	18.4	6.3	13.7	4	1.006
Acetonitrile	15.3	18	6.1	3	1.016
Dimethyl carbonate	15.5	8.6	9.7	3	1.053
Acetone	15.5	10.4	7	3	1.072
Ethanol	15.8	8.8	19.4	3	1.139
Methylene Chloride (DCM)	17	7.3	7.1	5	1.145
1-Propanol	16	6.8	17.4	5	1.175
Acetic Acid	14.5	8	13.5	2	1.182*
2-Propanol	15.8	6.1	16.4	5	1.220
1-Butanol	16	5.7	15.8	5	1.222
Tetrahydrofuran (THF)	16.8	5.7	8	3	1.248
Methanol	14.7	12.3	22.3	5	1.294
OME ₍₃₋₅₎	15.6	7.1	6.1	4	1.327
Propionic Acid	14.7	5.3	12.4	5	1.369
1,1-Dichloroethane	16.5	7.8	3	5	1.417
2-Methylfuran	17.3	2.8	7.4	5	1.513
Chlorobenzene	19	4.3	2	5	1.674
Lactic Acid (dl)	17	8.3	28.4	5	1.790
Diethyl Ether	14.5	2.9	4.6	5	1.824
β -Pinene	16.9	1.8	3.1	5	1.832
2,2,5,5-Tetramethyloxolane (TMO)	15.6	2.3	2.4	5	1.903
Toluene	18	1.4	2	5	1.903
Cyclohexene	17.2	1	2	5	1.953
Heptane	15.3	0	0	5	2.245
Hexane	14.9	0	0	5	2.271
Water	15.5	16	42.3	5	3.068

* Wrongly positioned solvents inside or outside the Hansen space. Hansen solubility parameters of the solvents are sourced from HSPiP database and expressed in MPa^{1/2}. **DMF anhydrous 99.8%.

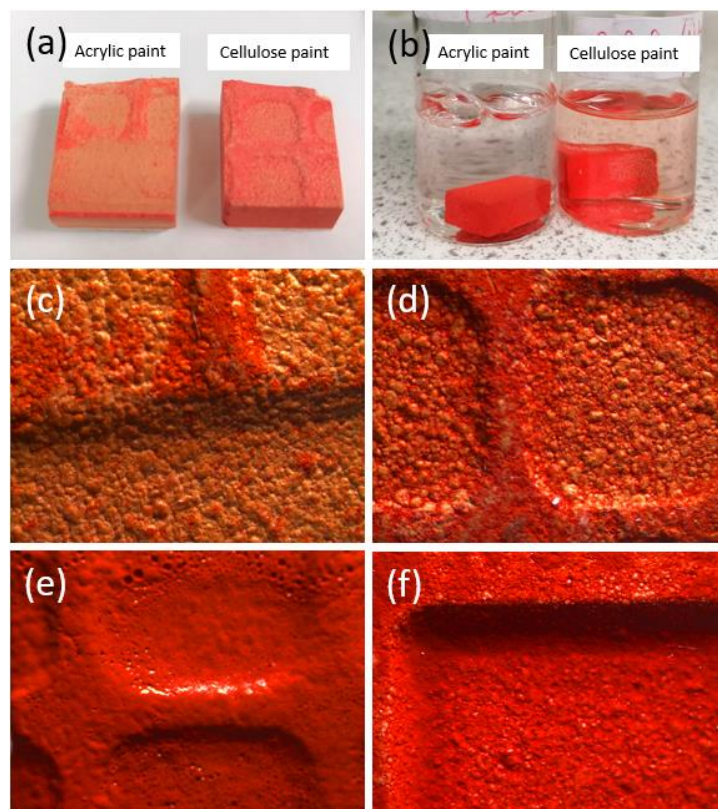


Figure S8. The use of binary solvent systems 75% Cyrene-25% water (a) and 75% NMP-25% water (b) as paint strippers for the acrylic (c, e) and cellulose paints (d, f).

Table S6. Hansen Solubility Parameters, the scores given and relative energy distance (RED) of the neat solvents and mixtures of Cyrene with greener solvents for acrylic paint removal.

Solvent	δ_D	δ_P	δ_H	Score	RED
<i>N</i> -Methyl-2-Pyrrolidone (NMP)	18	12.3	7.2	2**	0.742
<i>N,N'</i> -Dimethylformamide (DMF)	17.4	13.7	11.3	2***	0.767
50% Cyrene-50% GVL	17.9	11.9	6.9	-	0.889
75% Cyrene-25% GVL	18.4	12.1	7	-	0.892
Dimethyl Sulfoxide (DMSO)	18.4	16.4	10.2	2	0.957
<i>N,N'</i> -Dimethylacetamide	16.8	11.5	9.4	2	0.98
Cyrene	18.9	12.4	7.1	1	0.989
25% Cyrene-75% GVL	17.3	11.6	6.8	-	1.035
γ -Valerolactone (GVL)	16.8	11.4	6.7	-	1.233
75% Cyrene-25% MeTHF	18.4	10.6	6.4	-	1.336

50% Cyrene- 50% MeTHF	17.9	8.7	5.7	-	1.901
Acetone	15.5	10.4	7	3	1.92
75% Cyrene-25% Water	18.1	13.3	15.9	2	2.16
Methylene Chloride	17	7.3	7.1	3	2.183
Acetonitrile	15.3	18	6.1	3	2.26
Propylene Carbonate	20	18	4.1	3	2.428
25% Cyrene- 75% MeTHF	17.4	6.9	5	-	2.51
Tetrahydrofuran (THF)	16.8	5.7	8	3	2.633
OME ₍₃₋₅₎	15.6	7.1	6.1	3	2.682
Benzyl Alcohol	18.4	6.3	13.7	4	2.782
1,1-Dichloroethane	16.5	7.8	3	4	2.787
Ethyl Acetate	15.8	5.3	7.2	3	3.003
Acetic Acid	14.5	8	13.5	4	3.103
2-Methyl Tetrahydrofuran (MeTHF)	16.9	5	4.3	-	3.176
Propionic Acid	14.7	5.3	12.4	3	3.47
2-Methylfuran	17.3	2.8	7.4	4	3.49
1-Butanol	16	5.7	15.8	3	3.496
2-Propanol	15.8	6.1	16.4	3	3.57
1-Propanol	16	6.8	17.4	4	3.602
Chlorobenzene	19	4.3	2	3	3.759
Ethanol	15.8	8.8	19.4	3	3.821
2-Pinene (dl)	16.9	1.8	3.1	5	4.222
Diethyl Ether	14.5	2.9	4.6	4	4.23
2,2,5,5-Tetramethyloxolane (TMO)	15.6	2.3	2.4	4	4.378
Toluene	18	1.4	2	3	4.453
Cyclohexene	17.2	1	2	4	4.579
Methanol	14.7	12.3	22.3	5	4.617
β -Pinene	16.2	1	1.8	4	4.706
50% Cyrene-50% Water	17.2	14.2	24.7	-	4.918
Heptane	15.3	0	0	5	5.392
Hexane	14.9	0	0	5	5.472
Lactic Acid (dl)	17	8.3	28.4	5	6.317
25% Cyrene-75% Water	16.35	15.1	33.5	-	7.718
Water	15.5	16	42.3	5	10.525

* Wrongly positioned solvents inside or outside the Hansen space. – refers to the solvents only predicted and not tested in paint solubility. Hansen solubility parameters of the solvents are sourced from HSPiP database and expressed in MPa^{1/2}. **NMP received score “2” because despite being covered in a yellow layer, it can be easily removed. ***DMF anhydrous 99.8%.

Table S7. Hansen Solubility Parameters, the scores given and relative energy distance (RED) of the neat solvents and mixtures of Cyrene with greener solvents for cellulose-based paint removal.

Solvent	δ_D	δ_P	δ_H	Score	RED
Dimethyl Sulfoxide (DMSO)	18.4	16.4	10.2	1	0.311
<i>N,N'</i> -Dimethylformamide (DMF)	17.4	13.7	11.3	1**	0.393
75% Cyrene- 25% Water	18.1	13.3	15.9	3	0.411
<i>N,N'</i> -Dimethylacetamide (DMAc)	16.8	11.5	9.4	1	0.71
Cyrene	18.9	12.4	7.1	1	0.733
<i>N</i> -Methyl-2-Pyrrolidone (NMP)	18	12.3	7.2	1	0.736
75% Cyrene- 25% GVL	18.4	12.1	7	-	0.756
50% Cyrene- 50% GVL	17.9	11.9	6.9	-	0.787
25% Cyrene- 75% GVL	17.3	11.6	6.8	-	0.843
γ -Valerolactone	16.8	11.35	6.7	-	0.902
Propylene Carbonate	20	18	4.1	2	1.001*
Benzyl Alcohol	18.4	6.3	13.7	4	1.006
Acetonitrile	15.3	18	6.1	3	1.014
Acetone	15.5	10.4	7	3	1.07
50% Cyrene- 50% MeTHF	17.9	8.7	5.7	-	1.089
Ethanol	15.8	8.8	19.4	3	1.139
Methylene Chloride	17	7.3	7.1	5	1.144
1-Propanol	16	6.8	17.4	5	1.174
Acetic Acid	14.5	8	13.5	2	1.181*
2-Propanol	15.8	6.1	16.4	5	1.22
1-Butanol	16	5.7	15.8	5	1.221
50% Cyrene- 50% Water	17.2	14.2	24.7	-	1.236
Tetrahydrofuran (THF)	16.8	5.7	8	3	1.247
25% Cyrene- 75% MeTHF	17.4	6.9	5	-	1.287
Methanol	14.7	12.3	22.3	5	1.293
OME ₍₃₋₅₎	15.6	7.1	6.1	4	1.326
Propionic Acid	14.7	5.3	12.4	5	1.368
Ethyl Acetate	15.8	5.3	7.2	-	1.39
1,1-Dichloroethane	16.5	7.8	3	5	1.416
75% Cyrene- 25% MeTHF	18.4	10.6	-	-	1.47
2-Methyl Tetrahydrofuran (2-MeTHF)	16.9	5	4.3	-	1.5
2-Methylfuran	17.3	2.8	7.4	5	1.512
Chlorobenzene	19	4.3	2	5	1.673
Lactic Acid (dl)	17	8.3	28.4	5	1.791
Diethyl Ether	14.5	2.9	4.6	5	1.823
2-Pinene (dl)	16.9	1.8	3.1	5	1.831
2,2,5,5-Tetramethyloxolane (TMO)	15.6	2.3	2.4	5	1.902
Toluene	18	1.4	2	5	1.902
Cycloheptane	17.1	1.4	1.9	-	1.929
Cyclohexene	17.2	1	2	5	1.952
β -Pinene	16.2	1	1.8	5	2.005
25% Cyrene- 75% Water	16.4	15.1	33.5	-	2.145

Heptane	15.3	0	0	5	2.244
Hexane	14.9	0	0	5	2.27
Water	15.5	16	42.3	5	3.069

* *Wrongly positioned solvents inside or outside the Hansen space. – refers to the solvents only predicted and not tested in paint solubility. Hansen solubility parameters of the solvents are sourced from HSPiP database and expressed in MPa^{1/2}. **DMF anhydrous 99.8%.*