Supplementary material

Freeze concentration of Aqueous [DBNH][OAc] Ionic Liquid Solution

Nahla Osmanbegovic [1,*], Lina Yuan [2], Heike Lorenz [2] and Marjatta Louhi-Kultanen [1]

[1] Department of Chemical and Metallurgical Engineering, School of Chemical Engineering, Aalto University, P.O. Box 16100, FI-00076 Aalto, Finland; nahla.osmanbegovic@aalto.fi ; marjatta.louhi-kultanen@aalto.fi

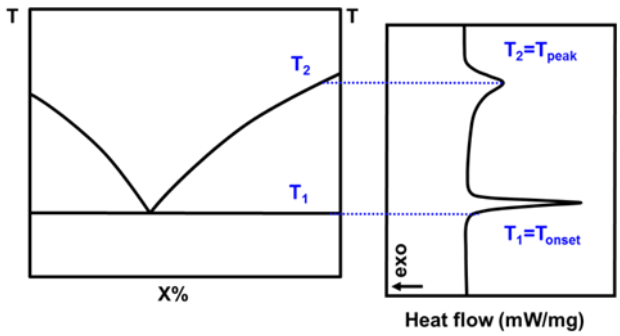
[2] Max Planck Institute for Dynamics of Complex Technical Systems, Sandtorstrasse 1, 39106 Magdeburg, Germany; lina.yuan@novartis.com; lorenz@mpi-magdeburg.mpg.de

Correspondence: nahla.osmanbegovic@aalto.fi;

This file contains data obtain by DSC, as well as calculated parameters and the experimental data obtained by freeze crystallization experiments.

DIFFERENTIAL SCANNING CALORIMETRY (DSC)

Figure S1 Schematic diagram of extracting thermal data from a DSC curve.



For a binary eutectic phase diagram, at a fixed concentration, the onset of the first thermal event corresponds to a eutectic temperature, while the peak of the second thermal event corresponds to a liquidus temperature.

Table S1 Liquidus and glass transition (or eutectic temperature) temperatures extracted from DSC curves.

Water content/wt%	Liquidus temperature/°C	Glass transition or eutectic temperature/ °C	Region of the phase diagram
100.00	0.00	//	V
96.51	-2.13	//	V
92.16	-2.43	//	V
85.17	-6.90	//	V
72.46	-13.03	-70.80	V
64.14	-25.40	-69.22	V
54.30	-38.28	//	V
21.62	//	-76.25	II
20.46	//	-75.80	II
14.20	23.32	-73.24	III
13.28	22.75	-72.95	III
11.30	//	-72.50	II
8.70	//	-70.95	II
1.88	43.62	-72.22	I

0.49	54.55	//	I

//: no available data.

LAYER FREEZE CRYSTALLIZATION

Table S2 Layer freeze crystallization data of aqueous 3 wt.% [DBNH][OAc] solutions with a freezing time of 40 minutes.

Experiment number	**#**	**1**	**2**	**3**	**4**	**5**
Sub-cooling (Freezing point)	ΔT, ℃ (T_f = -0.65 ℃)	1.23	1.54	1.82	2.05	2.35
Mass of sample	m, g	8.00	10.71	10.69	13.18	13.31
Height of sample	H, mm	52.5	54	50	53	49
Thickness of sample	d, mm	4.75	5.5	4.15	6.35	6.6
Cooling area	A_C, mm^2	1727	1774	1649	1743	1617
Overall ice growth rate	R_{Gice}, $kg_{(ice)}\ m^{-2}\ s^{-1}$	$1.91 \cdot 10^{-3}$	$2.49 \cdot 10^{-3}$	$2.67 \cdot 10^{-3}$	$3.1 \cdot 10^{-3}$	$3.37 \cdot 10^{-3}$
Impurity	C_{imp}, wt%	1.06	1.21	1.29	1.51	1.62
Distribution coefficient	K, -	0.31	0.34	0.37	0.43	0.46
Crystal yield	Y, %	3.26	4.36	4.35	5.35	5.4

Table S3 Layer freeze crystallization data of aqueous 3 wt.% [DBNH][OAc] solutions with a freezing time of 60 minutes.

Number of experiment	**#**	**1**	**2**	**3**	**4**	**5**
Sub-cooling (Freezing point)	ΔT, ℃ (T_f = -0.65℃)	1.25	1.50	1.73	1.98	2.28
Mass of sample	m, g	10.54	12.55	14.86	16.24	17.59
Height of sample	H, mm	52.5	52.50	53.00	53.00	53.70
Thickness of sample	d, mm	5	6.65	6.7	7.5	8
Cooling area	A_C, mm^2	1727	1727	1743	1743	1765
Overall ice growth rate	R_{Gice}, $kg_{(ice)}\ m^{-2}\ s^{-1}$	$1.68 \cdot 10^{-3}$	$1.99 \cdot 10^{-3}$	$2.34 \cdot 10^{-3}$	$2.55 \cdot 10^{-3}$	$2.73 \cdot 10^{-3}$
Impurity	C_{imp}, wt%	1.04	1.24	1.3	1.47	1.46
Distribution coefficient	K, -	0.3	0.35	0.37	0.42	0.41
Crystal yield	Y, %	4.3	5.11	6.05	6.6	7.15

Table S4 Layer freeze crystallization data of aqueous 6 wt.% [DBNH][OAc] solution with a freezing time of 40 minutes.

Number of experiment	**#**	**1**	**2**	**3**	**4**	**5**
Sub-cooling (Freezing point)	ΔT, ℃ (T_f = -1.28℃)	1.11	1.43	1.63	1.98	2.18
Mass of sample	m, g	6.27	8.04	8.55	10.04	12.46
Height of sample	H, mm	50	51	51.3	47.4	53
Thickness of sample	d, mm	4.85	4	4.5	6	6
Cooling area	A_C, mm^2	1649	1680	1689	1567	1743
Overall ice growth rate	R_{Gice}, $kg_{(ice)}\ m^{-2}\ s^{-1}$	$1.55 \cdot 10^{-3}$	$1.94 \cdot 10^{-3}$	$2.06 \cdot 10^{-3}$	$2.59 \cdot 10^{-3}$	$2.90 \cdot 10^{-3}$
Impurity	C_{imp}, wt%	2.07	2.67	2.39	2.98	2.79
Distribution coefficient	K, -	0.3	0.4	0.35	0.46	0.42

Crystal yield	Y, %	2.61	3.33	3.55	4.15	5.15

Table S5 Layer freeze crystallization data of aqueous 6 wt.% [DBNH][OAc] solutions with a freezing time of 60 minutes.

Number of experiment	**#**	**1**	**2**	**3**	**4**	**5**
Sub-cooling (Freezing point)	ΔT, °C (T_f = -1.28°C)	1.12	1.39	1.69	1.97	2.22
Mass of sample	m, g	8.64	10.5	12.96	15.18	17.33
Height of sample	H, mm	50.7	51	54	54.2	53.5
Thickness of sample	d, mm	4.3	5.5	6.35	6.5	7.5
Cooling area	A_C, mm^2	1670	1680	1774	1780	1758
Overall ice growth rate	R_{Gice}, $kg_{(ice)}\ m^{-2}\ s^{-1}$	$1.41 \cdot 10^{-3}$	$1.70 \cdot 10^{-3}$	$1.98 \cdot 10^{-3}$	$2.30 \cdot 10^{-3}$	$2.67 \cdot 10^{-3}$
Impurity	C_{imp}, wt%	2.01	2.29	2.43	2.9	2.6
Distribution coefficient	K,-	0.28	0.33	0.36	0.44	0.39
Crystal yield	Y, %	3.6	4.37	5.38	6.27	7.18

SUSPENSION FREEZE CRYSTALLIZATION

Table S6 Suspension freeze crystallization data of aqueous 3 wt.% [DBNH][OAc] solutions with a freezing time of 40 minutes

Number of experiment	**#**	**1**	**2**	**3**	**4**	**5**
Sub-cooling (Freezing point)	ΔT, °C (T_f = -0.65 °C)	1.09	1.37	1.51	1.66	1.85
Mass of ice crystals	m_{ice}, g	61.18	65.97	66.9	70.96	74.43
Impurity of ice sample	C_{imp}, wt%	0.91	1.01	0.67	0.88	0.5
Mother liquor concentration	$C_{moth.liquor}$, wt%	3.91	4.01	4.03	4.12	4.2
Cooling area	A_C, mm^2	14946	14946	14946	14946	14946
Overall ice growth rate	R_{Gice}, $kg_{(ice)}\ m^{-2}\ s^{-1}$	$1.71 \cdot 10^{-3}$	$1.84 \cdot 10^{-3}$	$1.86 \cdot 10^{-3}$	$1.98 \cdot 10^{-3}$	$2.07 \cdot 10^{-3}$
Distribution coefficient	K,-	0.3	0.34	0.22	0.29	0.17
Crystal yield	Y, %	24.92	26.87	27.25	28.9	30.32

Table S7 Suspension freeze crystallization data of aqueous 3 wt.% [DBNH][OAc] solutions with a freezing time of 60 minutes

Number of experiment	**#**	**1**	**2**	**3**	**4**	**5**
Sub-cooling (Freezing point)	ΔT, °C (T_f = -0.65 °C)	1.04	1.34	1.51	1.67	1.87
Mass of ice crystals	m_{ice}, g	70.07	74	80.19	85.21	87.44
Impurity of ice sample	C_{imp}, wt%	0.76	0.87	0.45	0.7	1.08
Mother liquor concentration	$C_{moth.liquor}$, wt%	4.1	4.19	4.34	4.47	4.53
Cooling area	A_C, mm^2	14946	14946	14946	14946	14946
Overall ice growth rate	R_{Gice}, $kg_{(ice)}\ m^{-2}\ s^{-1}$	$1.30 \cdot 10^{-3}$	$1.38 \cdot 10^{-3}$	$1.49 \cdot 10^{-3}$	$1.58 \cdot 10^{-3}$	$1.63 \cdot 10^{-3}$
Distribution coefficient	K,-	0.25	0.29	0.15	0.23	0.36
Crystal yield	Y, %	28.54	30.14	32.66	34.71	35.62

Table S8 Suspension freeze crystallization data of aqueous 6 wt.% [DBNH][OAc] solutions with a freezing time of 40 minutes

Number of experiment	**#**	**1**	**2**	**3**	**4**	**5**
Sub-cooling (Freezing point)	ΔT, °C (T_f = -1.28 °C)	1.02	1.26	1.41	1.57	1.89
Mass of ice crystals	m_{ice}, g	39.62	42.53	48.93	52.63	54.43
Impurity of ice sample	C_{imp}, wt%	1.42	0.92	1.07	0.77	1.07
Mother liquor concentration	$C_{moth.liquor}$, wt%	7.13	7.23	7.46	7.6	7.67
Cooling area	A_C, mm^2	14946	14946	14946	14946	14946
Overall ice growth rate	R_{Gice}, $kg_{(ice)}\ m^{-2}\ s^{-1}$	$1.10 \cdot 10^{-3}$	$1.19 \cdot 10^{-3}$	$1.36 \cdot 10^{-3}$	$1.47 \cdot 10^{-3}$	$1.52 \cdot 10^{-3}$
Distribution coefficient	K,-	0.24	0.15	0.18	0.13	0.18
Crystal yield	Y, %	16.86	18.1	20.82	22.4	23.16

Table S9 Suspension freeze crystallization data of aqueous 6 wt.% [DBNH][OAc] solutions with a freezing time of 60 minutes

Number of experiment	**#**	**1**	**2**	**3**	**4**	**5**
Sub-cooling (Freezing point)	ΔT, °C (T_f = -1.28 °C)	1	1.26	1.38	1.57	1.76
Mass of ice crystals	m_{ice}, g	46.47	51.59	57.45	58.67	61.79
Impurity of ice sample	C_{imp}, wt%	1.75	0.68	1.02	1.06	1.2
Mother liquor concentration	$C_{moth.liquor}$, wt%	7.37	7.56	7.79	7.84	7.97
Cooling area	A_C, mm^2	14946	14946	14946	14946	14946
Overall ice growth rate	R_{Gice}, $kg_{(ice)}\ m^{-2}\ s^{-1}$	$8.64 \cdot 10^{-4}$	$9.59 \cdot 10^{-4}$	$1.07 \cdot 10^{-3}$	$1.09 \cdot 10^{-3}$	$1.15 \cdot 10^{-3}$
Distribution coefficient	K,-	0.29	0.11	0.17	0.18	0.2
Crystal yield	Y, %	19.77	21.95	24.45	24.97	26.29