Dissolution Mechanism of Cellulose in Benzyltriethylammonium/Urea Deep Eutectic Solvent (DES): DFT-Quantum Modeling, Molecular Dynamics and Experimental Investigation

(Supplementary Data)

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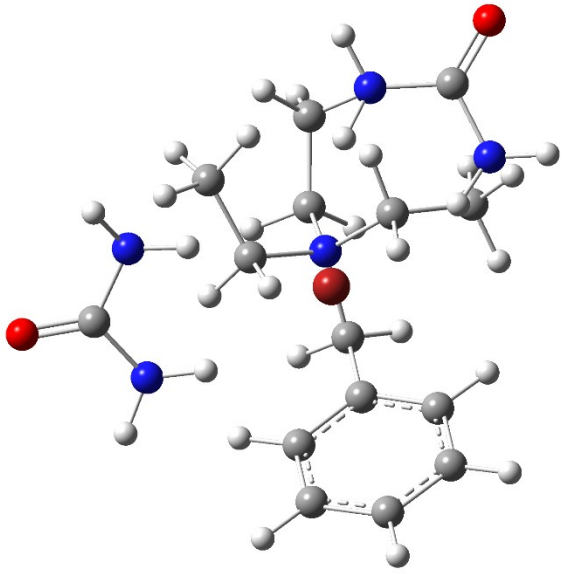
BCP	*d(°A)*	*ρ(r) (a.u.)*	*□²ρ(r)(a.u.)*	*V(r) (a.u.)*	*G(r)(a.u.)*	*H(r) (a.u.)*
CELLULOSE-DES						
BCP100 $H_{cell}(98)\cdots O_{urea}(49)$	1.89	0.039148	0.127159	-0.34805E-01	0.33298E-01	-0.15079E-02
BCP105 $O_{cell}(67)\cdots H_{urea}(52)$	3.2	0.0105340	0.036392	-0.664657E-02	0.78723E-02	0.12258E-02
BCP122 $H_{cell}(62)\cdots N_{urea}(48)$	1.96	0.004295	0.012793	-0.22686E-02	0.27335E-02	0.46491E-03
BCP144 $H_{cell}(67)\cdots Br_{urea}(37)$	4.12	0.007477	0.019154	-0.28826E-02	0.38355E-02	0.95295E-03
BCP181 $H_{cell}(95)\cdots Br(37)$	2.56	0.006143	0.018004	-0.29733E-02	0.37372E-02	0.76391E-03
BCP233 $O_{cell}(85)\cdots H_{urea}(43)$	2.42	0.018264	0.069110	-0.12577E-01	0.14927E-01	0.23504E-02
BCP247 $O_{cell}(89)\cdots H_{urea}(45)$	2.24	0.016178	0.055002	-0.10116E-01	0.11933E-01	0.18173E-02
DES						
BCP123 $H_{urea}(53)\cdots Br(37)$	2.67	0.011946	0.0331574	-0.57622E-02	0.70258E-02	0.12636E-02
BCP128 $H_{urea}(51)\cdots Br(37)$	2.52	0.013133	0.034870	-0.61743E-02	0.74459E-02	0.12716E-02
BCP197 $H_{urea}(42)\cdots Br(37)$	2.69	0.012391	0.033674	-0.55057E-02	0.69621E-02	0.14564E-02

Table SD1: AIM Topological parameters values of DES-CELL system at hydrogen BCPs

	η_{inh} (ml/g)	M_w	DP
Regenerated cellulose	114	31644	195
Commercial cellulose	122	34390	212

Table SD2: M_w and DP of Commercial and regenerated Cellulose

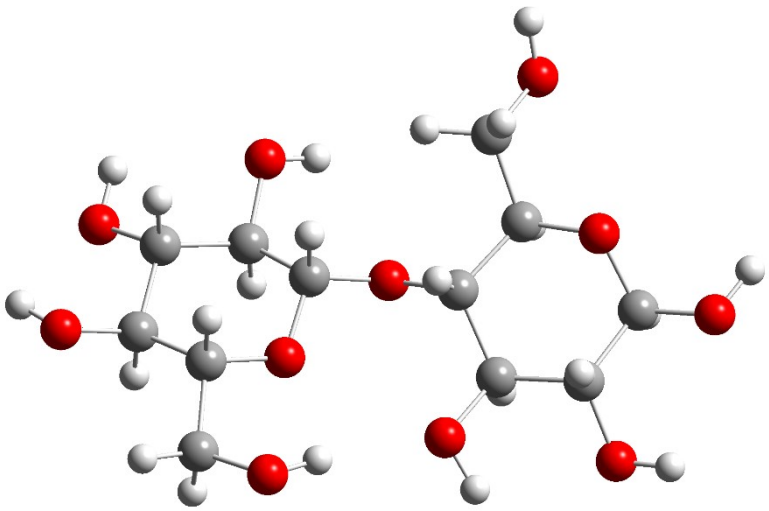


Atomic type	X	Y	Z
C	-0.5521	2.09611	0.00723
C	-1.40068	2.067	-1.09896
C	-0.87248	2.13891	-2.3894
C	0.50705	2.25353	-2.56797
C	1.35426	2.28414	-1.46085
C	0.83819	2.1857	-0.15958
C	1.74749	2.25013	1.04277
N	2.33256	0.9083	1.56102
C	3.1053	1.26469	2.83102
C	4.12734	0.25705	3.35426
C	1.1386	-0.01737	1.85662

C	3.21816	0.25925	0.49769
C	4.43964	1.06322	0.06919
C	1.41739	-1.33292	2.56831
H	-0.97777	2.03682	1.00485
H	-2.47329	1.98627	-0.95168
H	-1.53194	2.10314	-3.25101
H	0.92517	2.31249	-3.56794
H	2.42393	2.37988	-1.61801
H	2.61123	2.88444	0.84193
H	1.21503	2.66717	1.90098
H	3.61369	2.20853	2.61978
H	2.34242	1.46667	3.58828
H	4.57177	0.69451	4.25445
H	3.69242	-0.70052	3.63268
H	4.93828	0.074	2.64802
H	0.69745	-0.22959	0.88132
H	0.44196	0.58878	2.44114
H	3.51894	-0.70962	0.89454
H	2.55738	0.05082	-0.34536
H	4.18888	2.01043	-0.41415
H	5.1364	1.27152	0.88519
H	4.96699	0.45161	-0.66844
H	0.49133	-1.9136	2.50426
H	1.66245	-1.21042	3.62606
H	2.19023	-1.9254	2.07333
Br	0.5899	-1.67635	-1.33579
N	-1.29466	-3.27382	1.11626
C	-2.42888	-2.48706	1.20252
N	-2.3267	-1.2749	0.54121
O	-3.41068	-2.79857	1.87512
H	-1.44083	-4.23367	1.39368
H	-0.66108	-3.11106	0.33446
H	-3.21546	-0.81686	0.40189

H	-1.64966	-1.19999	-0.21664
N	3.96506	-1.90962	-2.09237
C	4.40895	-2.86655	-1.19706
N	3.47536	-3.21707	-0.23461
O	5.5599	-3.30097	-1.20009
H	4.53634	-1.83161	-2.92156
H	2.95834	-1.803	-2.21709
H	3.69688	-4.07802	0.24543
H	2.49167	-3.05536	-0.45563

Table SD3: Optimized geometry and Cartesian coordinates of DES at B3LYP/6-311+G(d,p) level.



Atomic type	X	Y	Z
C	-2.77252	-1.37138	0.49157
C	-3.12233	-2.78323	-0.01041
C	-4.644	-2.99066	-0.0768
C	-5.30167	-1.86358	-0.86334
C	-3.62248	-0.30092	-0.23009
H	-2.71655	-2.88253	-1.02112
H	-2.99517	-1.31006	1.56422
H	-4.91687	-1.84292	-1.89389
H	-3.26786	-0.17721	-1.25831

H	-5.03719	-2.972	0.95174
O	-4.99933	-0.62606	-0.21907
O	-4.95213	-4.23138	-0.69693
H	-4.38091	-4.88377	-0.26672
O	-2.63612	-3.8032	0.81903
O	-1.39536	-1.06364	0.23719
H	-0.90202	-1.06628	1.08249
O	-3.56116	0.92462	0.49202
C	-6.82041	-1.95934	-0.90338
H	-7.20237	-1.99647	0.12634
H	-7.11464	-2.87433	-1.41574
O	-7.39234	-0.86926	-1.61784
C	-2.31039	4.08593	-1.22377
C	-3.25376	2.90414	-0.96964
C	-2.63289	1.92187	0.04775
C	-2.1843	2.68409	1.30947
C	-1.86425	4.72103	0.08823
H	-4.19451	3.28514	-0.5488
H	-1.41822	3.71034	-1.74211
H	-3.05044	3.13952	1.80531
H	-2.72015	5.17823	0.60809
H	-1.7564	1.44409	-0.39395
O	-1.27419	3.73869	0.91361
O	-3.51245	2.22901	-2.19139
O	-3.01351	5.01908	-2.03663
H	-2.41227	5.73083	-2.28234
O	-0.8865	5.69703	-0.22806
H	-0.76874	6.29377	0.51958
C	-1.4662	1.7468	2.27192
H	-0.63677	1.26196	1.74867
H	-2.18299	0.97544	2.56694
O	-0.97597	2.43845	3.40441
H	-0.29145	1.86477	3.79928

H	-3.90807	2.86293	-2.80149
H	-7.01401	-0.0641	-1.2449
H	-1.75387	-4.13272	0.56831

Table SD4: Optimized geometry and Cartesian coordinates of cellobiose at at B3LYP/6-311+G(d,p) level.

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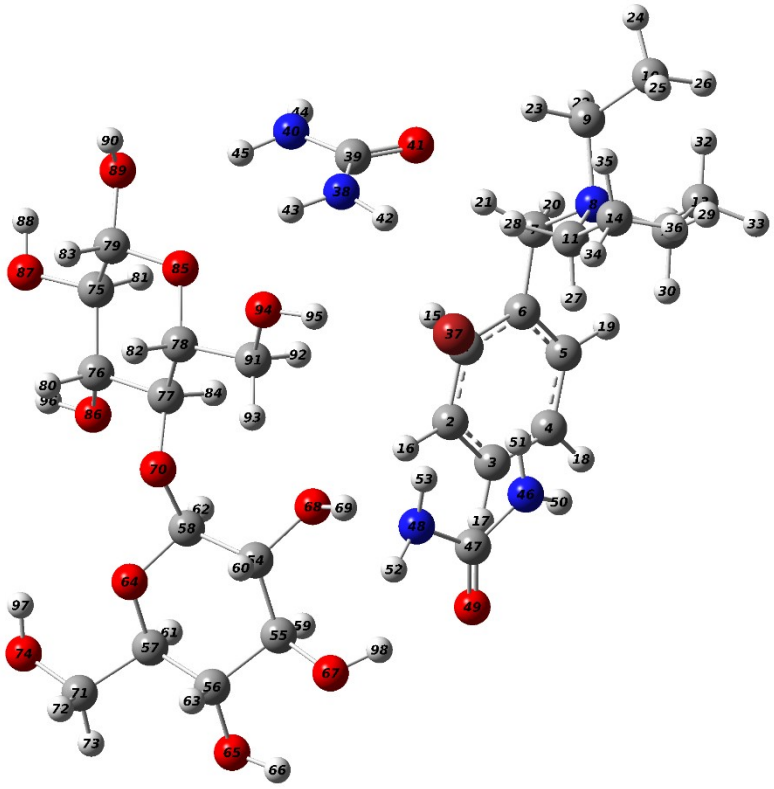

Atomic type	X	Y	Z
C	1.73746	-0.74863	-1.47652
C	0.63815	-1.57985	-1.6479
C	0.75369	-2.69288	-2.50108
C	1.96219	-2.95284	-3.17427
C	3.05797	-2.10315	-2.99004
C	2.96181	-1.00423	-2.13104
C	4.09624	-0.01845	-1.953

N	5.10768	-0.32385	-0.82068
C	6.04966	0.90618	-0.8081
C	7.36068	0.75642	-0.06311
C	4.26294	-0.44915	0.47225
C	5.85541	-1.61243	-1.04563
C	6.72552	-1.67974	-2.3005
C	4.97269	-0.61652	1.81109
H	1.66157	0.1033	-0.8203
H	-0.29654	-1.38427	-1.1225
H	-0.09596	-3.35849	-2.63884
H	2.04028	-3.81453	-3.84957
H	3.97507	-2.29659	-3.53193
H	4.69328	0.06432	-2.86242
H	3.70508	0.9799	-1.70358
H	6.26158	1.1305	-1.85686
H	5.42802	1.72306	-0.42717
H	7.89945	1.7014	-0.17501
H	7.25462	0.57533	1.00123
H	7.98781	-0.02983	-0.49135
H	3.6052	-1.29745	0.29145
H	3.65998	0.44583	0.48733
H	6.47156	-1.77019	-0.16025
H	5.09834	-2.39483	-1.06408
H	6.15027	-1.60009	-3.22346
H	7.50448	-0.91782	-2.31517
H	7.22044	-2.65337	-2.3148
H	4.17023	-0.81003	2.52682
H	5.47993	0.29916	2.12658
H	5.67102	-1.45359	1.86708
Br	1.48701	-0.31299	2.57418
N	1.65574	2.95231	1.03803
C	2.4027	3.31828	-0.0114
N	2.00752	4.46484	-0.66152

O	3.40676	2.66135	-0.3491
H	1.83752	2.06743	1.47829
H	0.75104	3.33278	1.19615
H	2.48859	4.69475	-1.51958
H	1.07251	4.84766	-0.52682
N	1.82285	-3.65589	1.42142
C	0.50471	-3.95804	1.51446
N	-0.09907	-3.46389	2.6538
O	-0.06991	-4.60816	0.68895
H	2.17666	-3.80557	0.52028
H	2.08357	-2.78667	1.83548
H	-1.1057	-3.52851	2.66493
H	0.30106	-2.67865	3.13287
C	-2.77252	-1.37138	0.49157
C	-3.12233	-2.78323	-0.01041
C	-4.644	-2.99066	-0.0768
C	-5.30167	-1.86358	-0.86334
C	-3.62248	-0.30092	-0.23009
H	-2.71655	-2.88253	-1.02112
H	-2.99517	-1.31006	1.56422
H	-4.91687	-1.84292	-1.89389
H	-3.26786	-0.17721	-1.25831
H	-5.03719	-2.972	0.95174
O	-4.99933	-0.62606	-0.21907
O	-4.95213	-4.23138	-0.69693
H	-4.38091	-4.88377	-0.26672
O	-2.63612	-3.8032	0.81903
O	-1.39536	-1.06364	0.23719
H	-0.90202	-1.06628	1.08249
O	-3.56116	0.92462	0.49202
C	-6.82041	-1.95934	-0.90338
H	-7.20237	-1.99647	0.12634
H	-7.11464	-2.87433	-1.41574

O	-7.39234	-0.86926	-1.61784
C	-2.31039	4.08593	-1.22377
C	-3.25376	2.90414	-0.96964
C	-2.63289	1.92187	0.04775
C	-2.1843	2.68409	1.30947
C	-1.86425	4.72103	0.08823
H	-4.19451	3.28514	-0.5488
H	-1.41822	3.71034	-1.74211
H	-3.05044	3.13952	1.80531
H	-2.72015	5.17823	0.60809
H	-1.7564	1.44409	-0.39395
O	-1.27419	3.73869	0.91361
O	-3.51245	2.22901	-2.19139
O	-3.01351	5.01908	-2.03663
H	-2.41227	5.73083	-2.28234
O	-0.8865	5.69703	-0.22806
H	-0.76874	6.29377	0.51958
C	-1.4662	1.7468	2.27192
H	-0.63677	1.26196	1.74867
H	-2.18299	0.97544	2.56694
O	-0.97597	2.43845	3.40441
H	-0.29145	1.86477	3.79928
H	-3.90807	2.86293	-2.80149
H	-7.01401	-0.0641	-1.2449
H	-1.75387	-4.13272	0.56831

Table SD5: Optimized geometry and Cartesian coordinates of DES-CELL system at B3LYP/6-311+G(d,p) level.

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