

## Supporting Information

### Design and Synthesis of Tröger's Base Ditopic Receptors: Host-Guest Interactions, a Combined Theoretical and Experimental Study

Manda Bhaskar Reddy,<sup>[a]</sup> Myadaraboina Shailaja,<sup>[a]</sup> Alla Manjula,<sup>\*,[a]</sup> Joseph Richard Premkumar,<sup>[b]</sup> Garikapati Narahari Sastry,<sup>\*,[b]</sup> Katukuri Sirisha,<sup>[c]</sup> and Akella Venkata Subramanya Sarma<sup>[c]</sup>

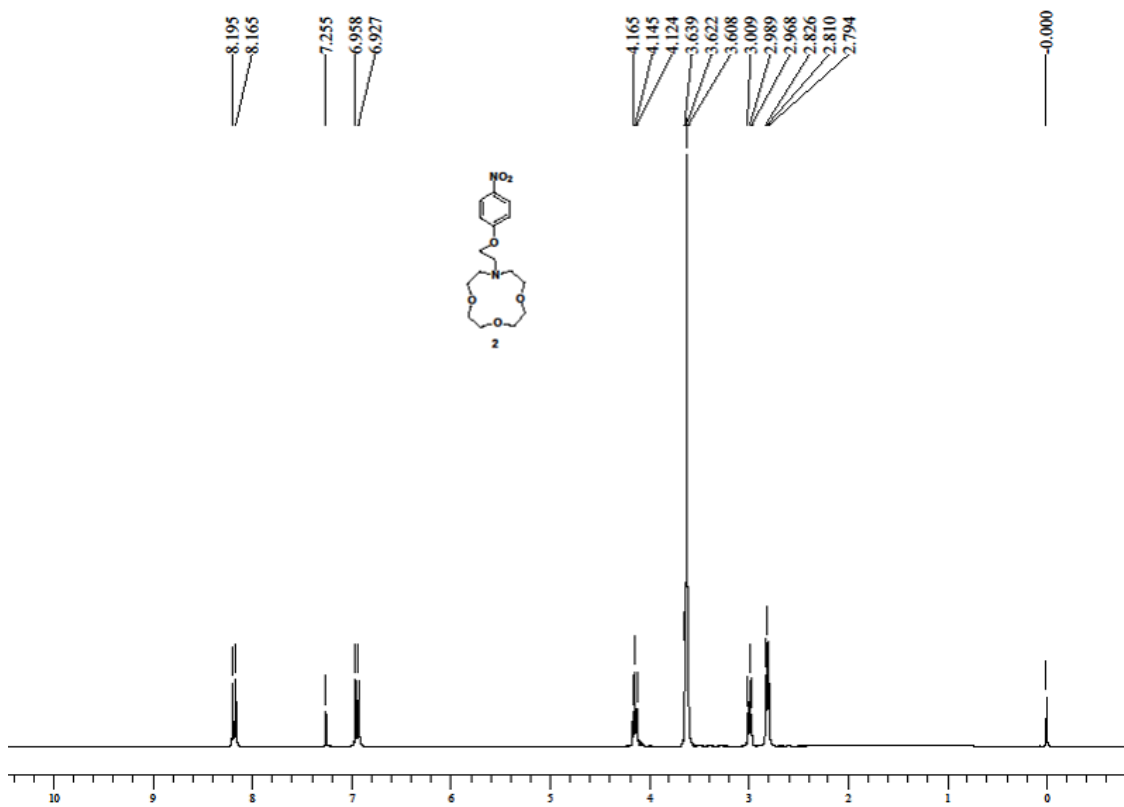
<sup>a</sup> Dr. A. Manjula, M. Bhaskar Reddy, M. Shailaja, Crop Protection Chemicals Division, CSIR-Indian Institute of Chemical Technology, Hyderabad, India, E-mail: manjula@iict.res.in.

<sup>b</sup> Dr. G. N. Sastry, J. R. Premkumar, Centre for Molecular Modelling, CSIR-Indian Institute of Chemical Technology, Hyderabad, India, E-mail: gnsastry@iict.res.in.

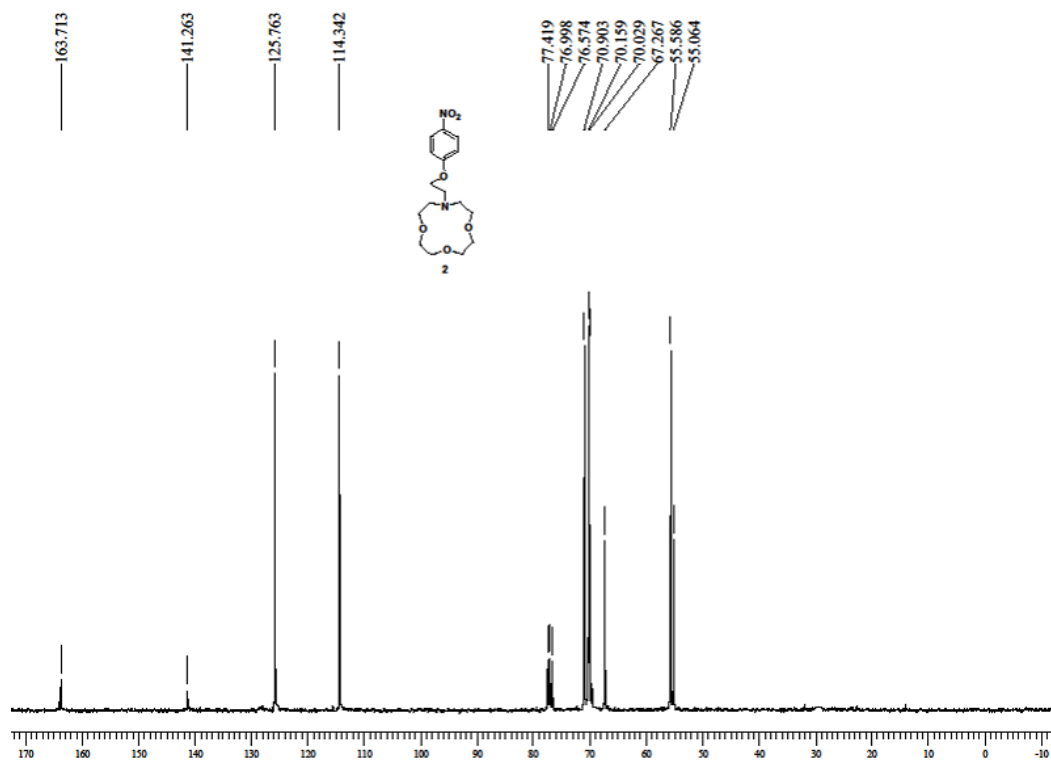
<sup>c</sup> Dr. A. V. S. Sarma, K. Sirisha, Centre for Nuclear Magnetic Resonance and Structural Chemistry, CSIR-Indian Institute of Chemical Technology, Hyderabad, India.

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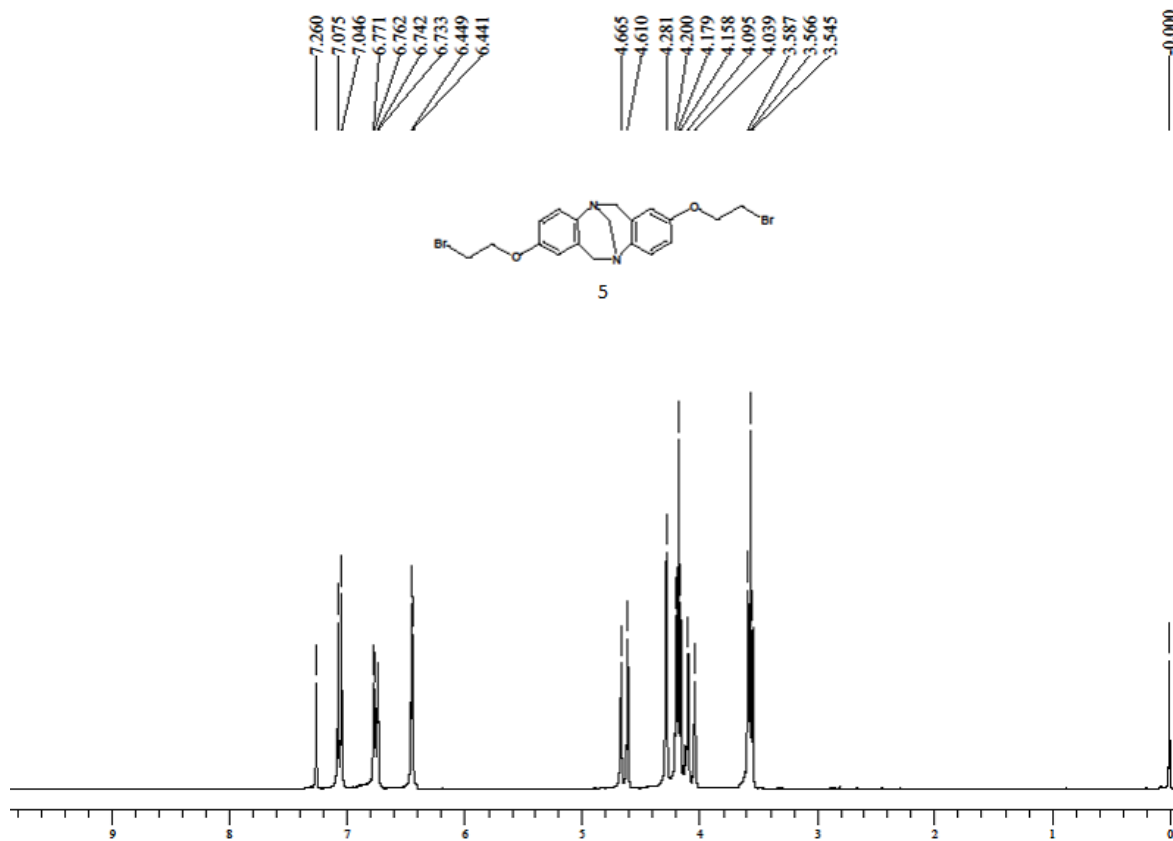
<b><sup>1</sup>H and <sup>13</sup>C NMR Spectra</b>	<b>2-5</b>
<b>Optimized structures of complexes</b>	<b>6-24</b>
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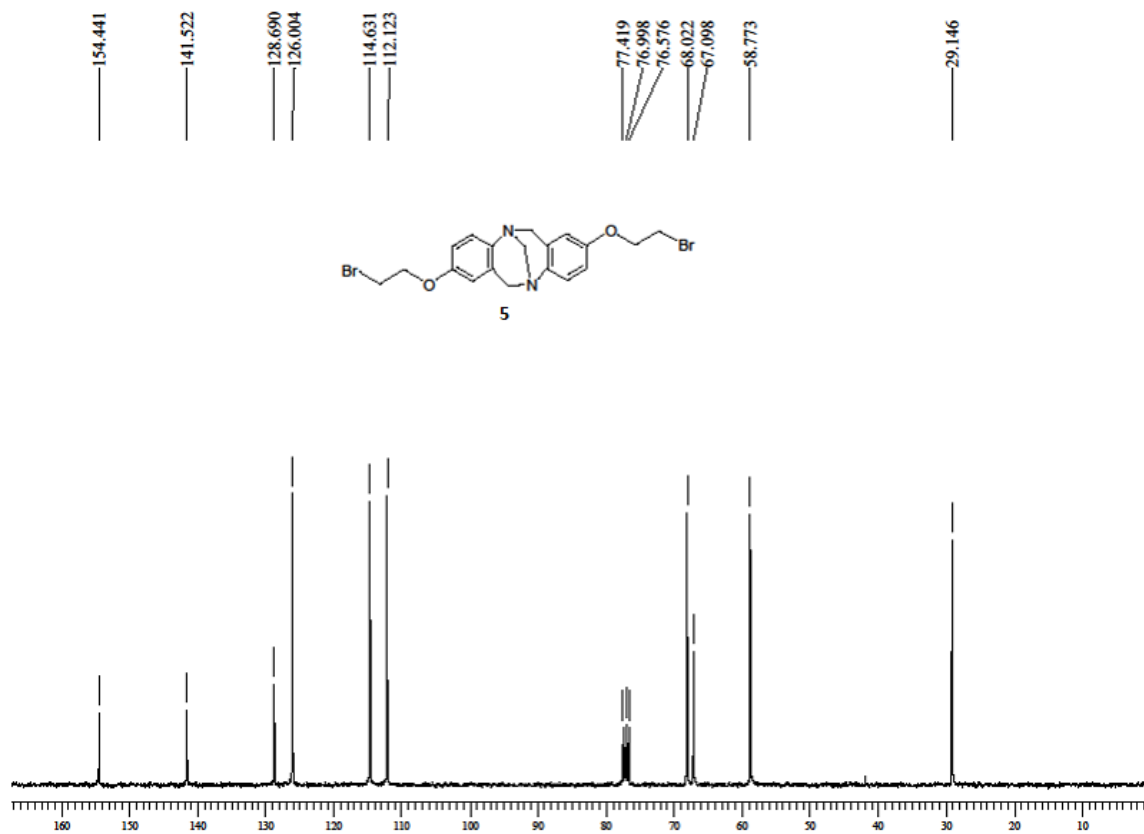
<sup>1</sup>H NMR spectra of 2 in CDCl<sub>3</sub>



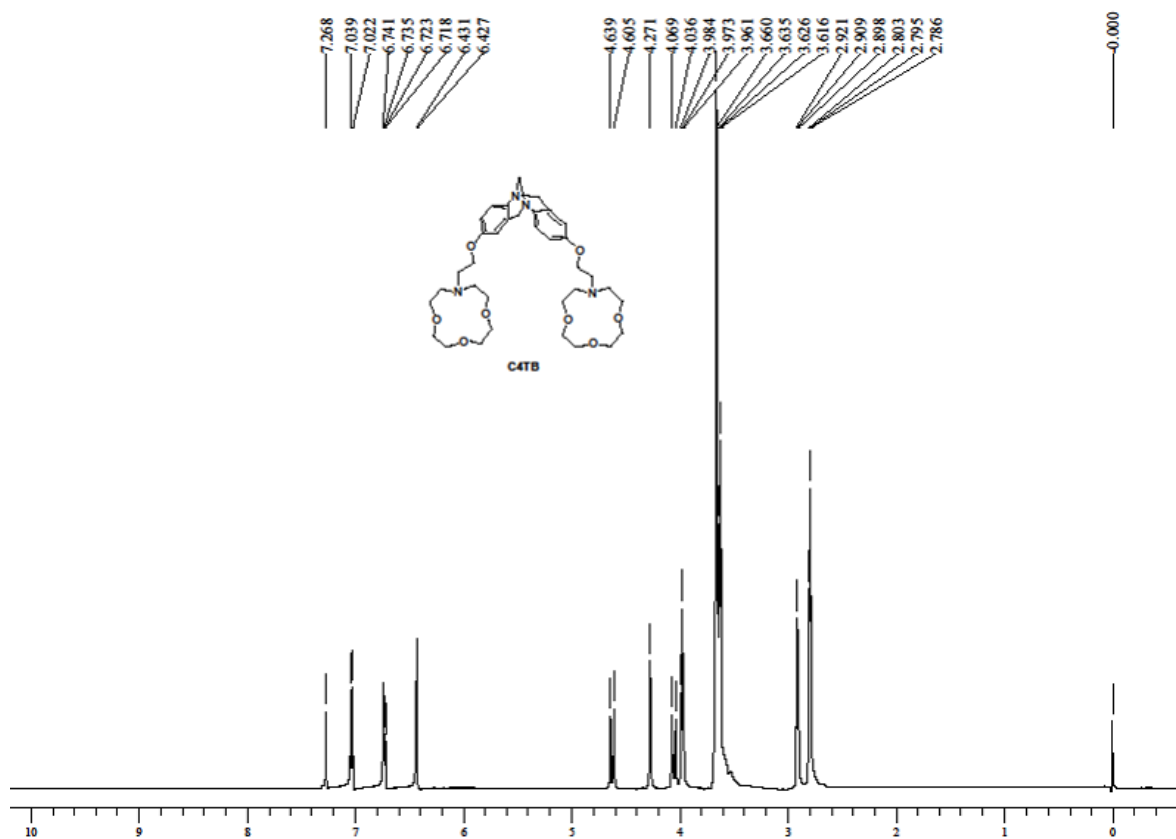
<sup>13</sup>C NMR spectra of 2 in CDCl<sub>3</sub>



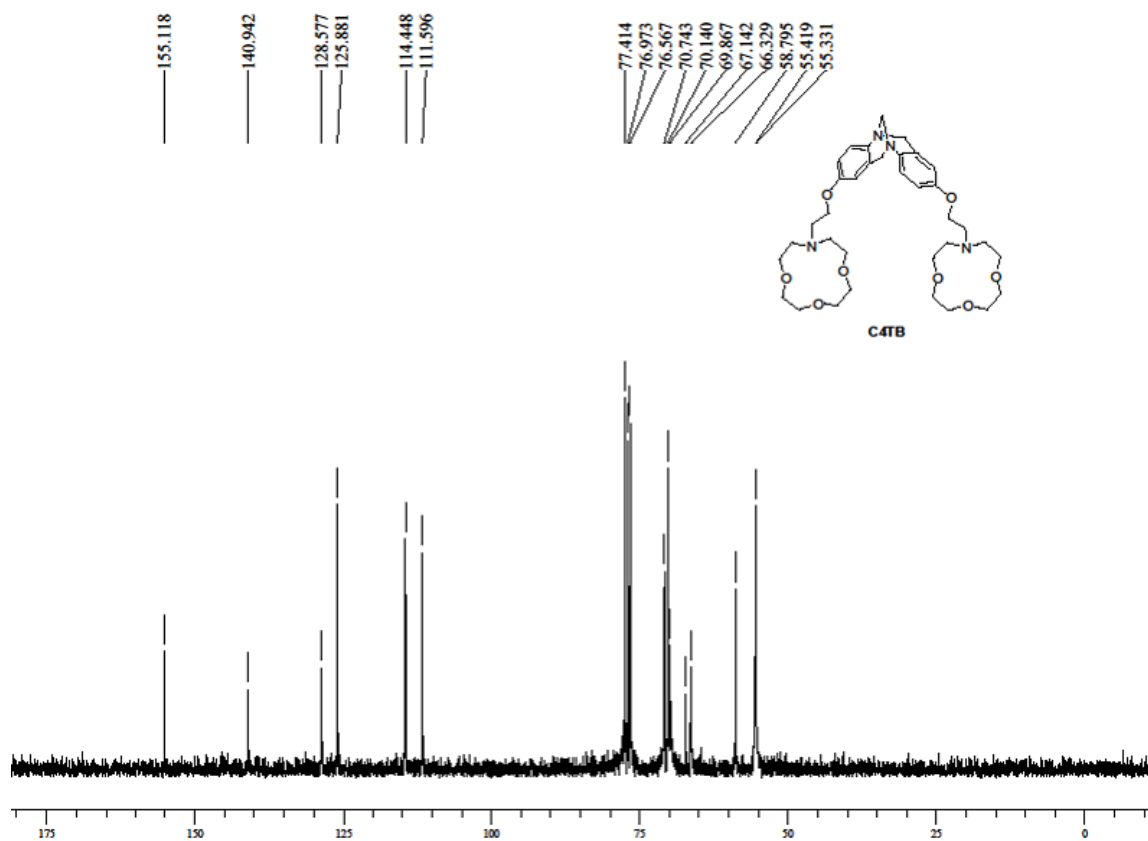
<sup>1</sup>H NMR spectra of 5 in CDCl<sub>3</sub>



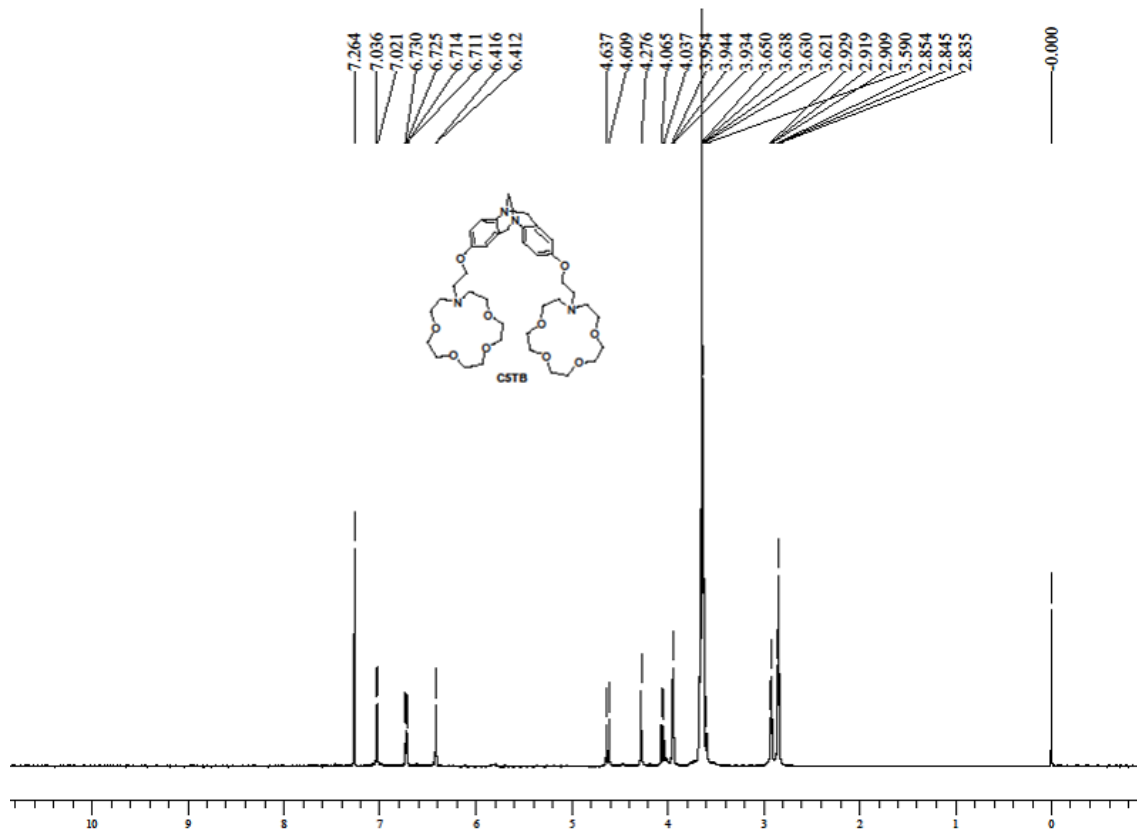
<sup>13</sup>C NMR spectra of 5 in CDCl<sub>3</sub>



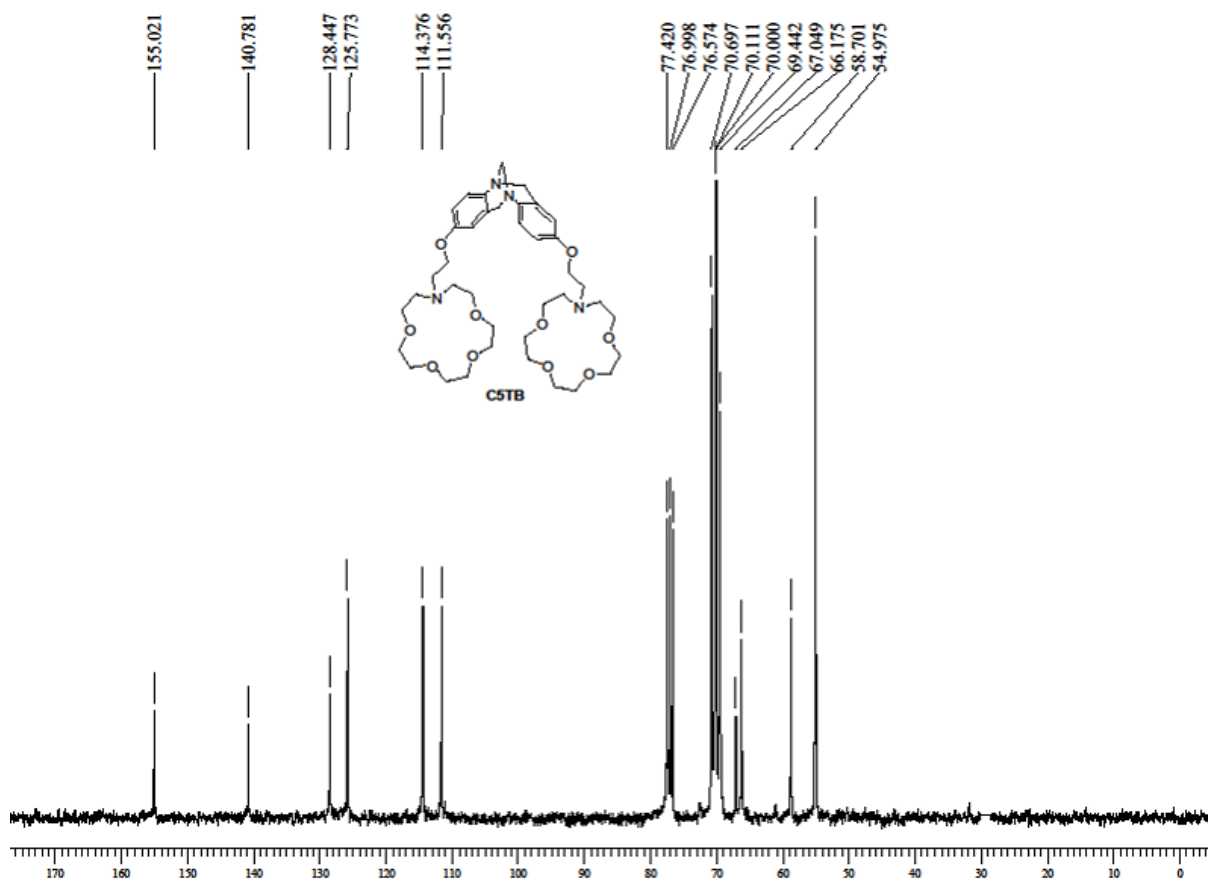
<sup>1</sup>H NMR spectra of C4TB in CDCl<sub>3</sub>



<sup>13</sup>C NMR spectra of C4TB in CDCl<sub>3</sub>



$^1\text{H}$  NMR spectra of C5TB in  $\text{CDCl}_3$

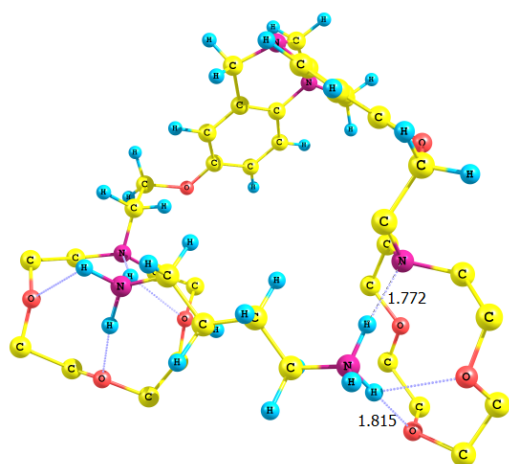


$^{13}\text{C}$  NMR spectra of C5TB in  $\text{CDCl}_3$

**Table S1.** The M06-2X/6-31G(d) structures of the host-guest complexes studied given in Bohr units. In the figure the hydrogen atoms of crown ethers removed for the clarity. The numbers in the figure shortest N...H and O...H hydrogen bond distances (Å).

C4TB...A1				
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8	7.488402057	-7.057079659	1.278135070	
1	8.804254168	-5.805385636	4.740130088	
1	6.832164472	-8.505271170	4.869684033	
8	9.091248743	-6.250402878	-3.593340423	
1	9.801498477	-3.254100593	-5.792503373	
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6	10.076109678	-8.308841713	-2.201099919	
1	10.264806377	-9.820029130	1.554532062	
1	11.361161145	-6.665713627	1.365693634	
1	12.042995068	-8.695721535	-2.738409489	
1	8.922350704	-9.934960375	-2.746170594	
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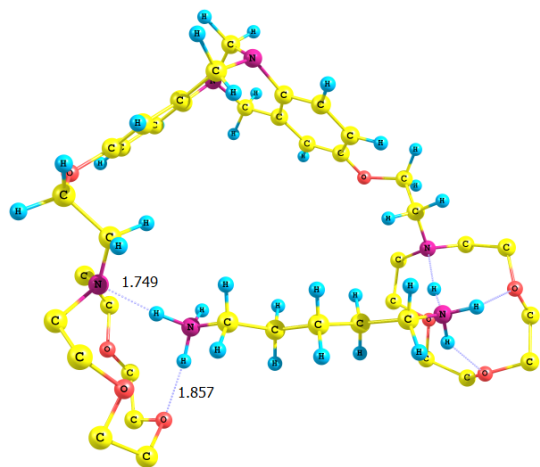
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C4TB...A3



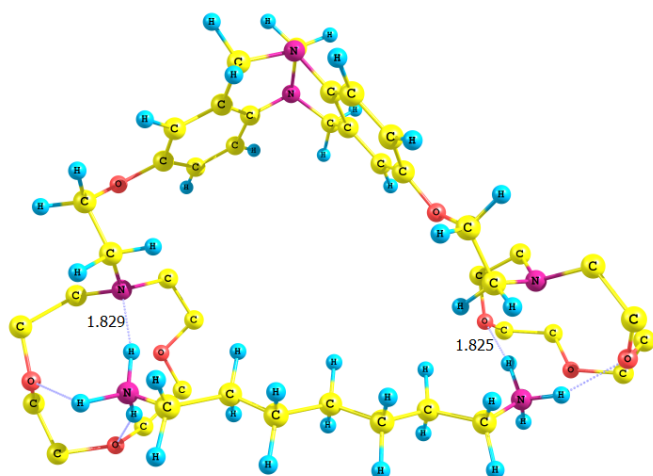
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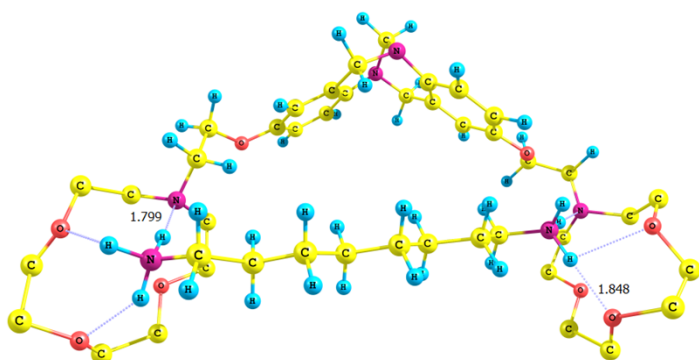
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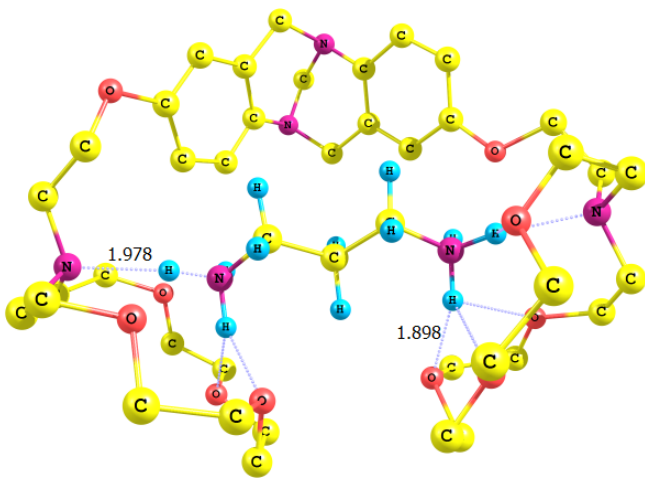
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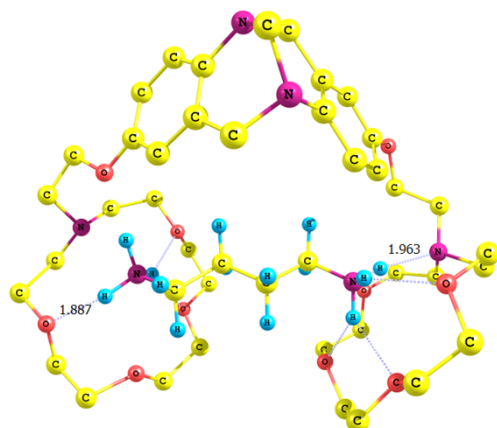
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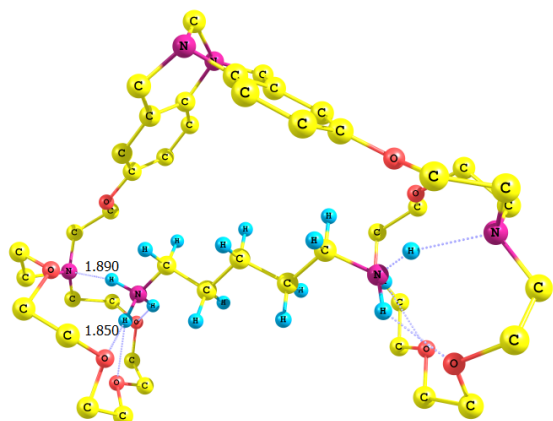
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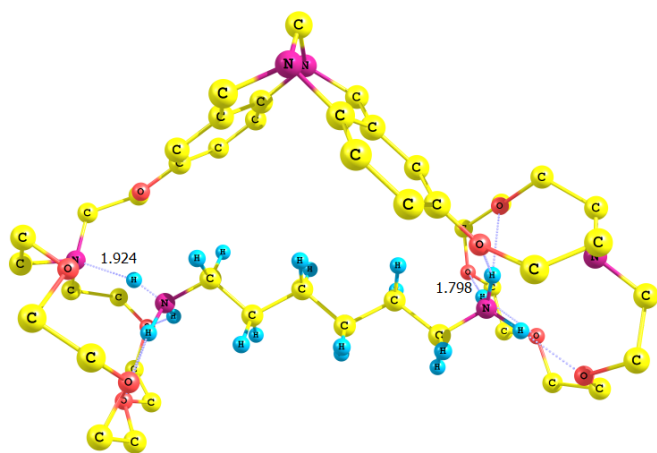
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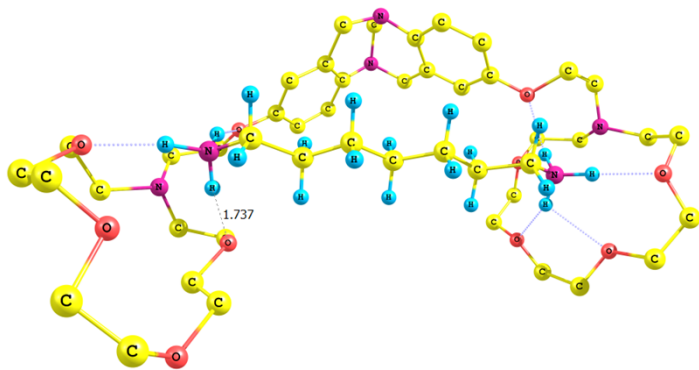
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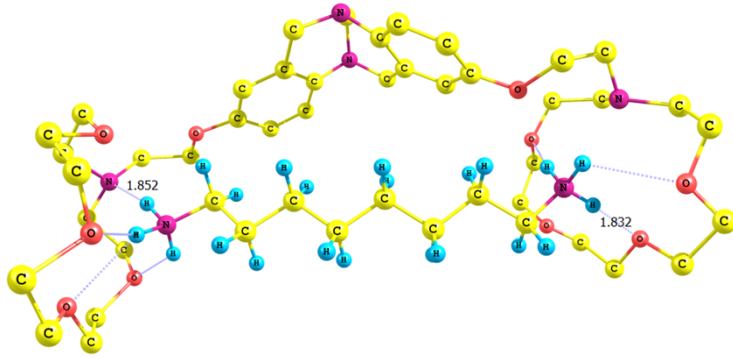


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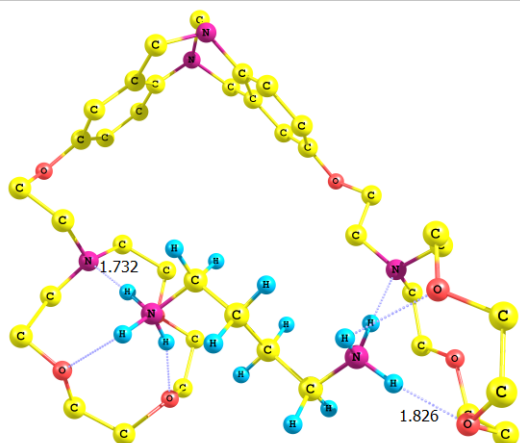


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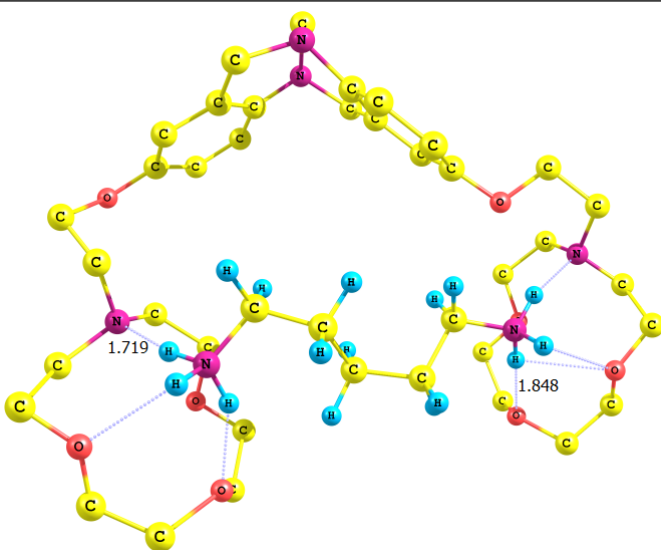
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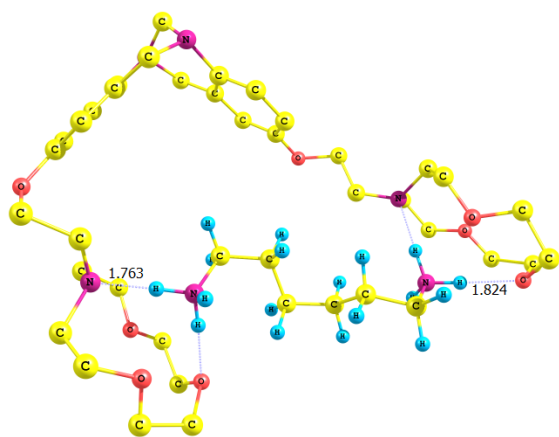
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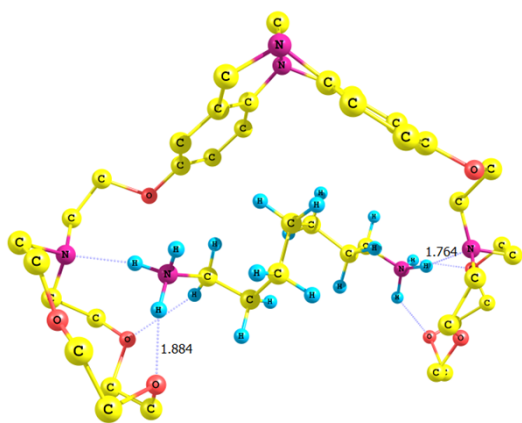
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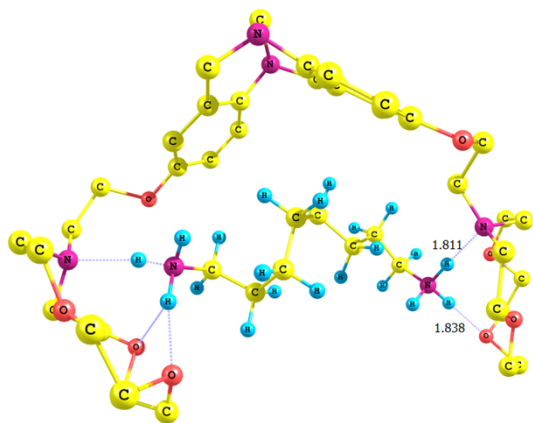
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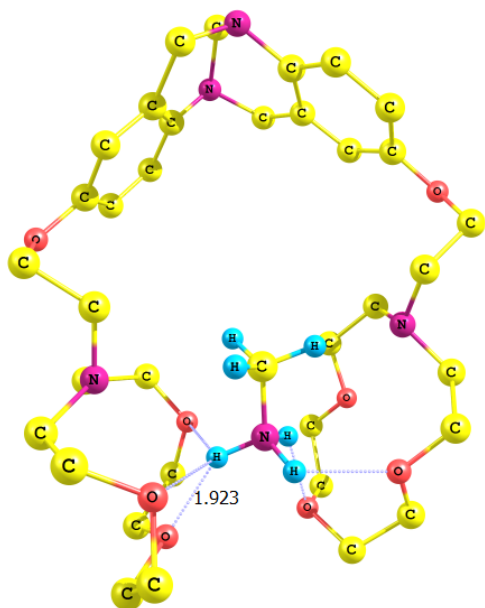
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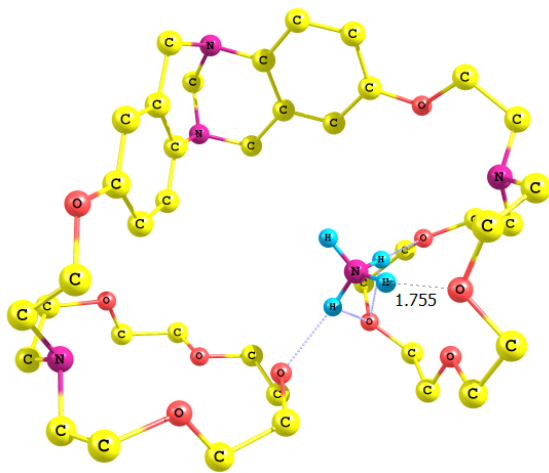
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6	-8.998614375	-3.302171443	-4.424113101
1	-12.027089245	-3.902061179	-1.758590343
1	-9.770580670	-6.376022413	-1.816848706
1	-9.887749350	-4.323513538	-5.997820212
1	-9.492579300	-1.300590684	-4.598493236
1	-5.681020879	9.051747801	-3.456312613
6	-4.567607554	7.375687461	-2.940770796
6	-2.487368840	8.104679497	-1.080872243
8	-6.198558245	5.471654357	-1.984786765
1	-3.742732711	6.609384675	-4.677069932
7	-0.543982192	6.146352675	-0.898863284
1	-1.684456393	9.935878782	-1.670876822
1	-3.314598281	8.385507896	0.788624229
6	-8.142715901	6.463603654	-0.442798704
6	0.495864099	5.806750018	1.637014822
6	1.377069784	6.384868220	-2.866699207
6	-9.665007348	4.301876175	0.661929440
1	-9.402739837	7.669562529	-1.574778586
1	-7.340270216	7.629602383	1.076212179
6	-1.064138719	4.049798067	3.284270069
1	0.772535102	7.625911748	2.612447913
1	2.360948061	4.930880919	1.474568307
1	0.409849442	6.699486810	-4.671013360
1	2.370659363	4.573097208	-3.045499411
6	3.337104918	8.493112673	-2.534230265
8	-8.248865589	2.782615849	2.350480869
1	-11.355314332	5.058327819	1.598386933
1	-10.284408504	3.040026764	-0.857257187
8	-3.594821969	4.883322185	3.447096419
1	-1.027238040	2.117334035	2.509238639
1	-0.203686005	3.979783719	5.177352211
1	4.268411018	8.897665213	-4.339659357
1	2.443636801	10.243310744	-1.896086806
6	-7.745421799	3.904803171	4.736439453
6	-4.971997577	3.553410513	5.310953948
1	-8.906420314	2.994561847	6.194251560
1	-8.204097981	5.924829546	4.718494615
1	-4.522316491	4.267498280	7.210092662
1	-4.492974716	1.533300990	5.232651262
8	2.775655988	-8.292128976	0.430060061
8	5.168795531	7.939766889	-0.631410365
1	-5.003941505	1.609857570	0.314588354
1	-5.616052099	-1.266114523	-0.862703377
1	-5.986009425	1.386172594	-2.580751539
7	-4.870687477	0.494677351	-1.277046587
6	-2.254477119	0.279428113	-2.234268390
1	-1.143692404	-0.704933934	-0.809014372
1	-2.303284962	-0.831109049	-3.966574534
1	-1.522571126	2.181475115	-2.531847321

C5TB<sup>+</sup>·(NH<sub>4</sub>)<sup>+</sup>



6	-7.037560679	-2.335552034	-5.732757797
1	-10.235674361	-0.041449250	-6.102813388
1	-10.993749280	-2.999913551	-4.801593426
1	-12.611749679	1.887471545	-3.571517868
1	-12.272464495	1.831938168	-0.276410220
6	-9.726455568	4.338293085	-2.128158386
8	-3.093317037	-4.460747329	4.248655822
1	-2.258422867	-0.966645866	5.436225774
1	-3.316905637	-2.989569191	7.897068530
8	-5.777345881	-4.143692882	-4.202882879
1	-7.271888592	-3.096850825	-7.651079119
1	-5.759761981	-0.712695039	-5.818560805
1	-10.998363991	5.976893387	-2.171302720
1	-8.519544271	4.388427515	-3.816010616
6	-4.674615178	-6.606034191	4.490002177
6	-6.839884003	-6.598363793	-4.167614923
6	-4.437872196	-8.178524319	2.116141618
1	-6.662140156	-6.053045565	4.687748774
1	-4.130003707	-7.725723295	6.151490840
6	-5.386996522	-8.118115448	-2.231231600
1	-6.684505063	-7.478953427	-6.039836380
1	-8.842424743	-6.522980734	-3.636361925
8	-5.534175941	-6.819110245	0.097415375
1	-2.442601231	-8.583078748	1.699997499
1	-5.416582072	-9.990847131	2.389838192
1	-6.213504088	-10.017256052	-2.077164130
1	-3.402018895	-8.321693739	-2.816016756
1	8.146262917	-7.404394288	-5.291332923
6	7.755435457	-5.446598480	-4.714465720
6	9.843144258	-4.605307588	-2.921291501
8	5.298816238	-5.266957348	-3.674602420
1	7.756227252	-4.305995778	-6.438177390
7	9.650756924	-1.985856350	-2.065581999
1	11.663417147	-4.949611883	-3.896728372
1	9.818058146	-5.816748558	-1.242107444
6	4.633842450	-7.381906549	-2.190001558
6	11.695612409	-1.457543765	-0.276495258
6	9.703193041	-0.197952577	-4.179446498
6	1.930965479	-7.072664229	-1.284117942
1	5.914717622	-7.588483835	-0.566934804
1	4.766799791	-9.116731698	-3.329111377
6	11.155390772	0.775798659	1.465208494
1	13.508891423	-1.116315944	-1.257162891
1	11.978077311	-3.149162220	0.880772937
1	10.326810176	1.640771597	-3.469236449
1	11.115383383	-0.787819206	-5.598417176
6	7.112167061	0.226549800	-5.448010105
8	1.688222617	-5.306588681	0.723668678
1	0.788142349	-6.356898386	-2.853401205
1	1.177775502	-8.920421293	-0.709941708
8	8.981742902	0.422572967	2.970426266
1	12.825258480	1.117990241	2.656848915
1	10.802091380	2.499759773	0.376595933
1	5.659143521	-0.898596833	-4.492868891
1	7.165833390	-0.312859255	-7.440036410
6	2.300711155	-6.350170961	3.125799537
6	9.318939938	-1.190230686	5.062638284
6	3.147586188	-4.277598866	4.887538164
1	3.841332944	-7.722928391	2.941808256
1	0.650328412	-7.313226348	3.921642519
6	6.732885827	-1.933276614	6.011059633
1	10.382583549	-0.226487439	6.566635294
1	10.352251557	-2.913347093	4.542604590
8	5.599418731	-3.510651683	4.184966387
1	1.818598592	-2.678444704	4.804724702
1	3.141707251	-5.012845894	6.833957822
1	6.906780303	-2.932531591	7.827093867
1	5.584720442	-0.226659404	6.324945009
8	-8.209536612	4.383873276	0.083648721
8	6.390223704	2.816328561	-5.502530590
1	-1.353646630	-3.250474209	0.322345680
7	-2.755753394	-2.115230770	-0.436868744
1	-2.012554398	-0.417533067	-0.978271460
1	-3.610935662	-3.003953785	-1.964047022
1	-4.168293335	-1.855469036	0.896865844
6	-1.836418708	8.968894655	0.418666903
6	-3.742549408	9.520639841	-1.315663138
6	-5.903263993	8.035734734	-1.493951226
6	-6.178451561	5.959602394	0.113927800
6	-4.296469669	5.408684907	1.872924435
6	-2.123428402	6.879687301	2.033611615
6	3.429591887	6.241485261	0.928885361
6	3.349531756	7.440316090	-1.438595483
6	4.401164268	6.241409672	-3.529428001
6	5.464773864	3.837137753	-3.323504560
6	5.515146400	2.617474585	-0.981017232
6	4.520933212	3.851713209	1.116049492
7	0.394742972	10.513755099	0.458120602
7	2.372348778	7.385243805	3.153497251
6	2.053446179	9.984834023	-1.709353533
6	1.871057386	10.070688054	2.750571765
6	-0.030866784	6.192809699	3.868803891
1	-3.518208698	11.150071193	-2.545731137
1	-7.355293538	8.509625099	-2.859580609
1	-4.530678529	3.742235714	3.055055755
1	4.341832542	7.137096327	-5.378067567
1	6.357164837	0.760329362	-0.740162206
1	4.633861347	2.939523577	2.951982541
1	3.449966913	11.511014526	-1.838448274
1	0.934720835	10.032088511	-3.445945576
1	3.658683369	11.092723678	2.577463416
1	0.841528998	10.791735682	4.391059904
1	-0.514330502	6.808367153	5.788253380
1	0.220374176	4.142056379	3.929847710
1	-10.459565317	-2.572865713	3.730026194
6	-9.485016617	-1.263106079	2.446305094
6	-9.986721859	-2.158564077	-0.239195846
8	-6.856619416	-1.335303060	2.998745700
1	-10.212912611	0.642996275	2.805056345
7	-9.498900434	-0.295226222	-2.223803087
1	-11.967377683	-2.822222616	-0.320849017
1	-8.767393173	-3.798562776	-0.544328012
6	-6.312578642	-1.435847821	5.613678603
6	-9.591810702	-1.456666932	-4.736042611
6	-11.204506640	1.888231215	-2.046333250
6	-3.627744775	-2.440874022	5.918266528
1	-7.680262157	-2.676254511	6.560059047
1	-6.461232047	0.438316274	6.493444317



**Table S2.** The hydrogen bonding distances and the corresponding hydrogen bond donor length of **C4TB...Am** (**m = 1-6**), **C4TB...Am\*** (**m = 2-6**) and **C5TB...Am** (**m = 1-6**) complexes obtained at PCM-M06-2X/6-31G(d) level of theory

<b>C4TB...Am</b>						
<b>m=</b>	<b>H...N</b>	<b>N-H</b>	<b>N-H...N</b>	<b>H...O</b>	<b>N-H</b>	<b>N-H...O</b>
1	1.712	1.079	176	1.896	1.038	160
2	1.772	1.071	174	1.815	1.039	175
3	1.749	1.071	174	1.857	1.038	173
4	1.771	1.069	174	1.845	1.038	169
5	1.829	1.064	168	1.825	1.042	160
6	1.799	1.067	173	1.848	1.038	179
<b>C4TB...Am*</b>						
2	1.732	1.077	177	1.826	1.040	167
3	1.719	1.077	171	1.848	1.038	169
4	1.760	1.070	175	1.820	1.040	171
5	1.823	1.067	171	1.842	1.039	172
6	1.811	1.064	175	1.838	1.038	173
<b>C5TB...Am</b>						
1	1.978	1.049	164	1.898	1.035	152
2	1.963	1.048	163	1.887	1.035	158
3	1.890	1.050	159	1.850	1.035	156
4	1.924	1.046	155	1.798	1.035	166
5	-	-	-	1.737	1.041	171
6	1.852	1.055	167	1.832	1.037	176

The hyphen indicates that the H...N type hydrogen bond was not seen in the **C5TB...Am** complexes.

**Table S3.** The binding energies of **C4TB...Am**, **C4TB...Am\*** and **C5TB...Am** complexes. The values are expressed in kcal/mol

<b>Method</b>	<b>C4TB...A1</b>	<b>C4TB...A2</b>	<b>C4TB...A3</b>	<b>C4TB...A4</b>	<b>C4TB...A5</b>	<b>C4TB...A6</b>
PCM-B3LYP/cc-pVTZ//B3LYP/6-31G(d)	40.49	36.25	36.82	36.08	33.84	34.92
M06-2X/cc-pVTZ//B3LYP/6-31G(d)	150.14	133.23	127.57	122.07	117.65	123.10
M06-2X/cc-pVTZ//PCM-M06-2X/6-31G(d)	142.83	128.87	122.66	117.22	113.57	120.57
PCM-M06-2X/cc-pVTZ//PCM-M06-2X/6-31G(d)	52.32	48.69	49.07	47.99	46.21	49.37
HF/6-31G(d)//B3LYP/6-31G(d)	132.42	119.68	110.28	105.35	103.65	105.74
	<b>C4TB...A1*</b>	<b>C4TB...A2*</b>	<b>C4TB...A3*</b>	<b>C4TB...A4*</b>	<b>C4TB...A5*</b>	<b>C4TB...A6*</b>
PCM-M06-2X/cc-pVTZ//PCM-M06-2X/6-31G(d)	-	51.70	54.86	52.05	57.89	59.88
	<b>C5TB...A1</b>	<b>C5TB...A2</b>	<b>C5TB...A3</b>	<b>C5TB...A4</b>	<b>C5TB...A5</b>	<b>C6TB...A6</b>
PCM-B3LYP/cc-pVTZ//B3LYP/6-31G(d)	46.23	41.65	42.07	43.32	41.28	40.43
M06-2X/cc-pVTZ//B3LYP/6-31G(d)	182.04	164.57	164.58	160.57	148.15	149.00
M06-2X/cc-pVTZ//PCM-M06-2X/6-31G(d)	173.60	161.54	161.83	157.15	144.65	148.25
PCM-M06-2X/cc-pVTZ//PCM-M06-2X/6-31G(d)	63.72	60.63	65.65	64.66	59.45	63.01
HF/6-31G(d)//B3LYP/6-31G(d)	166.50	148.75	148.56	144.06	134.51	132.45

**Table S4.** The relative energies (B3LYP/cc-pVTZ//B3LYP/6-31G(d)) of extended and folded conformations of bisammonium ions **A<sub>m</sub>** (**m** = **1-6**) showed in Figure S1

	Extended	Folded
<b>A1</b>	0.00	*
<b>A2</b>	0.00	3.12
<b>A3</b>	0.00	5.92
<b>A4</b>	0.00	5.02
<b>A5</b>	0.00	6.53
<b>A6</b>	0.00	7.60

\* The initial folded bisammonium ion collapsed to the extended structure upon geometry optimization.

**Table S5.** The binding energies of  $\text{C4-NH}_4^+$  calculated using various methods (A/cc-pVTZ//A/6-31G(d)) in three different solvents

A	Water (80)	DMSO (47)	Methanol (33)
B3LYP	19.17	19.57	20.00
B3LYP <sup>1</sup>	18.68	19.07	19.50
B3LYPD	28.91	29.32	29.76
BP86	21.45	21.87	22.32
M052X	22.91	23.41	23.89
M062X	24.54	25.02	25.48
MP2	24.03	24.46	24.92
MPW1PW91	19.79	20.20	20.65
PBE0	21.17	21.61	22.05
PM6	8.23	8.94	9.67

<sup>1</sup>-B3LYP/cc-pVTZ geometry has been taken for the binding energy calculations.

**Table S6.** The percentage contribution of various energy terms to the total host-guest binding energy.

	<b>C4-NH<sub>4</sub><sup>+</sup></b>	<b>C4-Me-NH<sub>4</sub><sup>+</sup></b>	<b>C4-Et-NH<sub>4</sub><sup>+</sup></b>	<b>C4-<i>n</i>-Pr-NH<sub>4</sub><sup>+</sup></b>	<b>C5-NH<sub>4</sub><sup>+</sup></b>	<b>TB1- NH<sub>4</sub><sup>+</sup></b>	<b>TB2- NH<sub>4</sub><sup>+</sup></b>
E <sub>es</sub>	68.44	69.72	69.45	69.36	71.58	52.15	50.65
E <sub>x</sub>	44.04	45.96	47.05	49.30	38.35	33.24	31.77
POL <sub>h</sub>	16.22	15.11	14.82	14.37	15.97	28.41	30.18
POL <sub>g</sub>	2.65	3.19	3.83	3.68	2.05	0.28	0.45
CT <sub>h-g</sub>	12.12	11.24	11.20	11.88	9.94	18.71	18.35
CT <sub>g-h</sub>	0.57	0.73	0.70	0.71	0.46	0.45	0.37