Electronic Supplementary Information

Coordination of Sorbitol to $Ga(OTf)_3$ in Liquid Phase: An Experimental and Theoretical Study

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Phys. Chem. Chem. Phys.

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ii. **Figure S2**. Optimized geometric structures and the Gibbs free energies of formation (ΔGs) and the coordinated patterns of Ga^{3+} cation with OTf^- for $[Ga(OTf)_n]^{3-n}$ (n=1~3) species in the sorbitol solution. For clarity, hydrogen atoms on carbon are not shown. Bond lengths are reported in Å. 2

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v. **Table S1.** Zero-point energies (*ZPE*, hartree), thermal correction to Gibbs free energy (G_0, hartree), total energies (E_c, hartree) corrected by *ZPE*, sum of electronic and thermal free energies (G_c, hartree) with *ZPE* and thermal corrections, and relative energies (E_r, kJ mol^{-1}) and relative Gibbs free energies (G_r, kJ mol^{-1}) relative to SBT-I-1 for fifteen conformers of sorbitol at M06/6-311++G(d,p) level in sorbitol solution. 6

vi. **Table S2.** Zero-point energies (*ZPE*, hartree), thermal correction to Gibbs free energy (G_0, hartree), total energies (E_c, hartree) corrected by *ZPE*, sum of electronic and thermal free energies (G_c, hartree) with *ZPE* and thermal corrections, and relative energies (E_r, kJ mol^{-1}) and relative Gibbs free energies (G_r, kJ mol^{-1}) relative to Ga^{3+} and OTf^- for $[Ga(OTf)_n]^{3-n}$ (n= 1, 2, 3) at M06/6-311++G(d,p) level in sorbitol solution. 7

vii. **Table S3.** Zero-point energies (*ZPE*, hartree), thermal correction to Gibbs free energy (G_0, hartree), total energies (E_c, hartree) corrected by *ZPE*, sum of electronic and thermal free energies (G_c, hartree) with *ZPE* and thermal corrections, and relative energies (E_r, kJ mol^{-1}) and relative Gibbs free energies (G_r, kJ mol^{-1}) relative to Ga^{3+} and SBT for $[Ga(SBT)_m]^{3+}$ (m= 1, 2) at M06/6-311++G(d,p) level in sorbitol solution. 8

viii. **Table S4.** Zero-point energies (*ZPE*, hartree), thermal correction to Gibbs free energy (G_0, hartree), total energies (E_c, hartree) corrected by *ZPE*, sum of electronic and thermal free energies (G_c, hartree) with *ZPE* and thermal corrections, and relative energies (E_r, kJ mol^{-1}) and relative Gibbs free energies (G_r, kJ mol^{-1}) relative to Ga^{3+}, SBT and OTf^- for $[Ga(OTf)_n(SBT)_m]^{3-n}$ (n = 1,2; m = 1, 2) at M06/6-311++G(d,p) level in sorbitol solution. ...10

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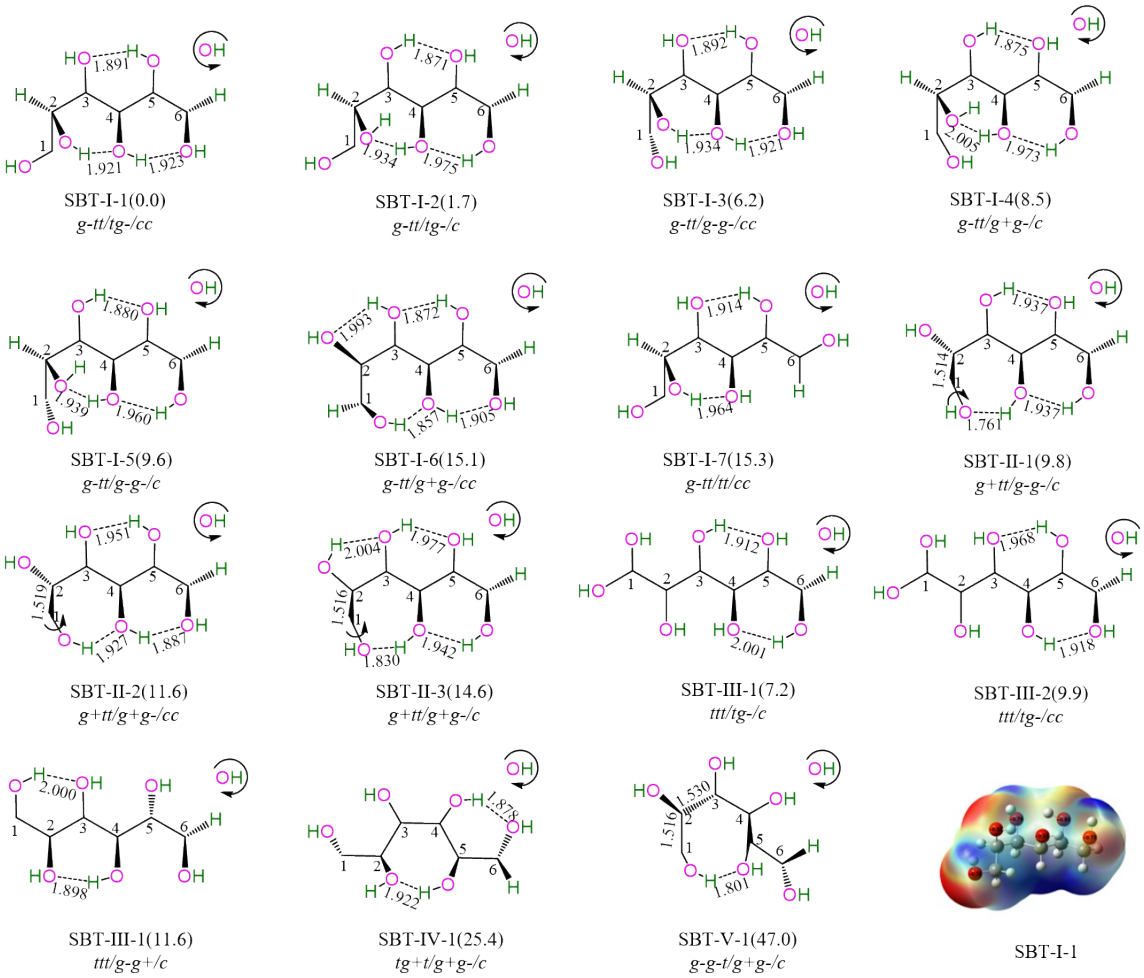


Figure S1. The geometric structures and the relative Gibbs free energies (G_r, kJ mol^{-1}) relative to SBT-I-1 for fifteen conformers of sorbitol in liquid solution calculated at the 6-311++g(d,p) level in sorbitol solution under 433.15K and ambient pressure (433.15 K and 1.0 atm). For clarity, hydrogen atoms on carbon are not shown. Bond lengths are reported in Å.

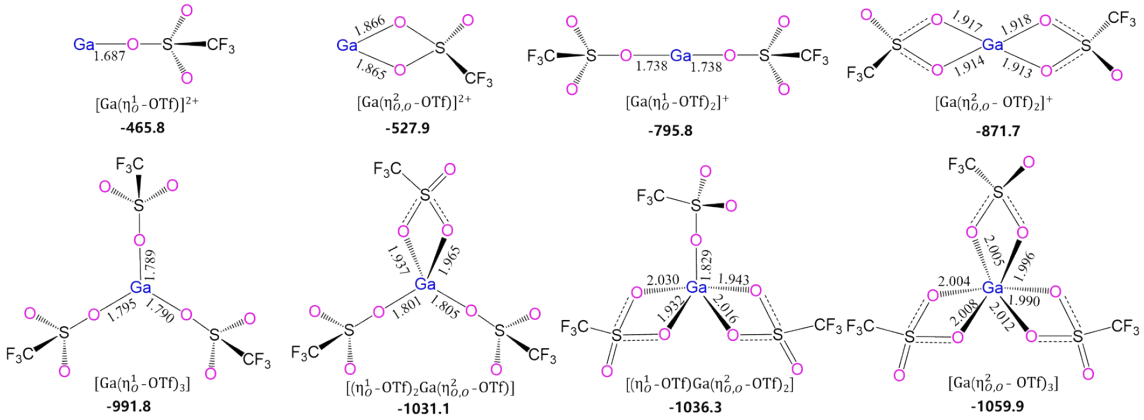


Figure S2. Optimized geometric structures and the Gibbs free energies of formation (ΔGs) and the coordinated patterns of Ga^{3+} cation with OTf^- for $[Ga(OTf)_n]^{3-n}$ (n=1~3) species in the sorbitol solution. For clarity, hydrogen atoms on carbon are not shown. Bond lengths are reported in Å.

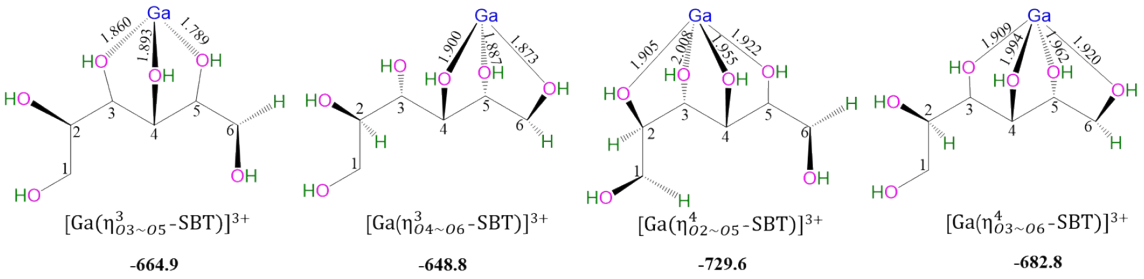
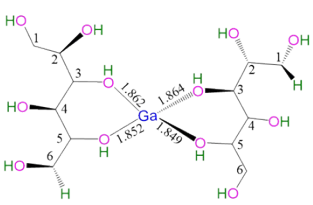
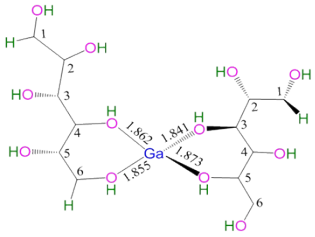


Figure S3. optimized geometric structures and the Gibbs free energies of formation (ΔGs, kJ mol^{-1}) of $[Ga(SBT)]^{3+}$ complex in the sorbitol solution. For clarity, hydrogen atoms on carbon are not shown. Bond lengths are reported in Å.



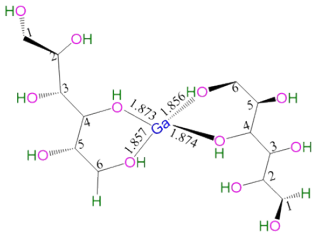
$[Ga(\eta^2_{O3,O5}\text{-SBT})_2]^{3+}$

-909.6



$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^2_{O4,O6}\text{-SBT})]^{3+}$

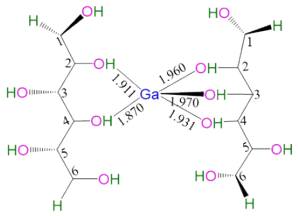
-893.7



$[Ga(\eta^2_{O4,O6}\text{-SBT})_2]^{3+}$

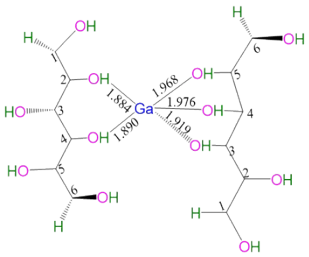
-862.7

(a)



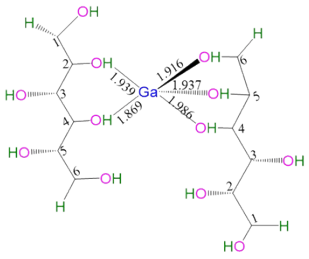
$[(\eta^2_{O2,O4}\text{-SBT})Ga(\eta^3_{O2\sim O4}\text{-SBT})]^{3+}$

-980.7



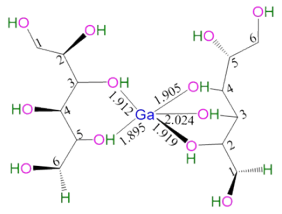
$[(\eta^2_{O2,O4}\text{-SBT})Ga(\eta^3_{O3\sim O5}\text{-SBT})]^{3+}$

-986.4



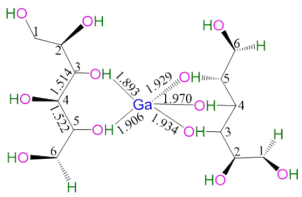
$[(\eta^2_{O2,O4}\text{-SBT})Ga(\eta^3_{O4\sim O6}\text{-SBT})]^{3+}$

-955.5



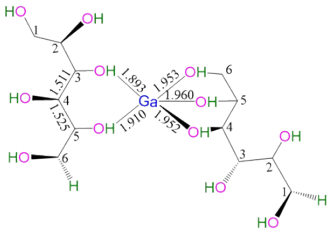
$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^3_{O2\sim O4}\text{-SBT})]^{3+}$

-980.6



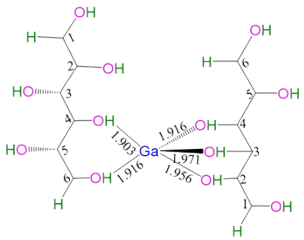
$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^3_{O3\sim O5}\text{-SBT})]^{3+}$

-968.7



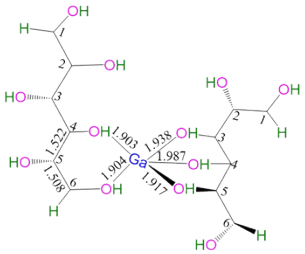
$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^3_{O4\sim O6}\text{-SBT})]^{3+}$

-946.6



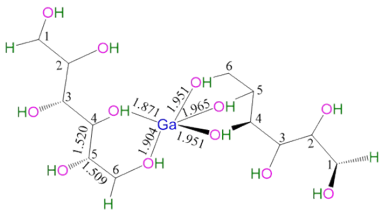
$[(\eta^2_{O4,O6}\text{-SBT})Ga(\eta^3_{O2\sim O4}\text{-SBT})]^{3+}$

-961.2



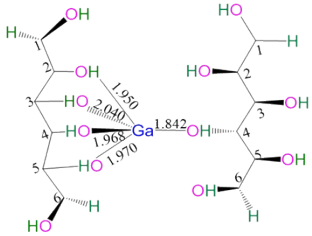
$[(\eta^2_{O4,O6}\text{-SBT})Ga(\eta^3_{O3\sim O5}\text{-SBT})]^{3+}$

-956.4



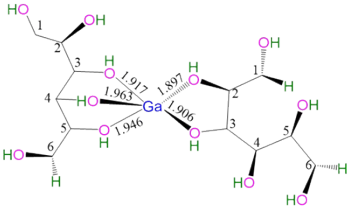
$[(\eta^2_{O4,O6}\text{-SBT})Ga(\eta^3_{O4\sim O6}\text{-SBT})]^{3+}$

-979.6



$[(\eta^4_{O2\sim O5}\text{-SBT})Ga(\eta^1_{O4}\text{-SBT})]^{3+}$

-895.2

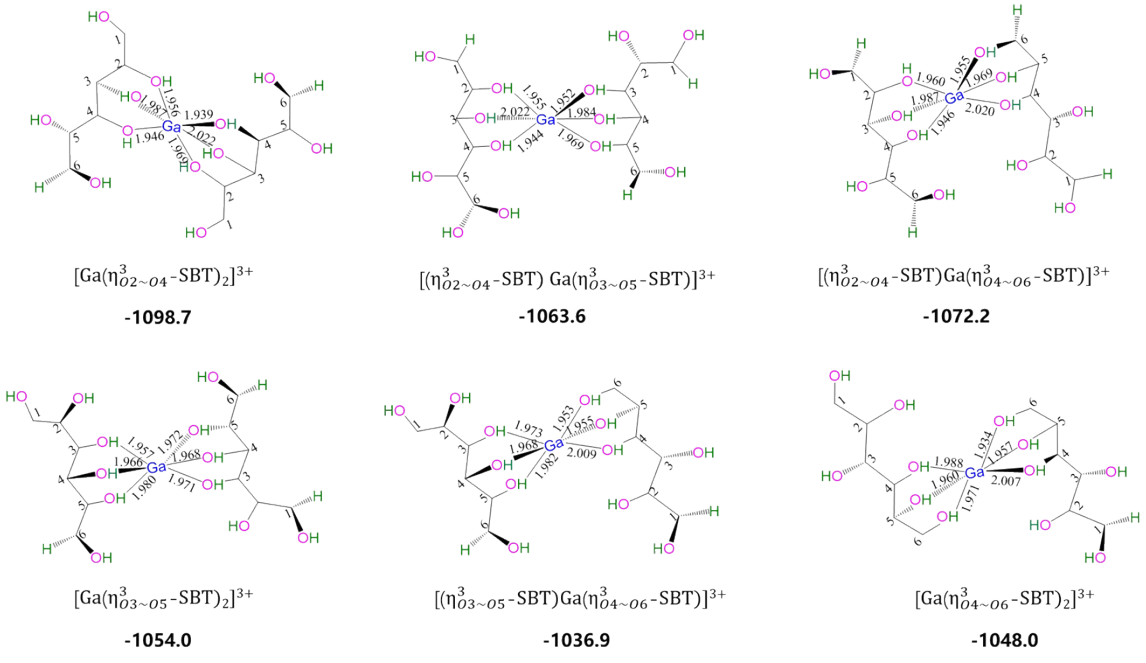


$[(\eta^3_{O3\sim O5}\text{-SBT})Ga(\eta^2_{O2,O3}\text{-SBT})]^{3+}$

-957.6

(b)

Continued from **Fig.S4**



(c)

Figure S4. Optimized geometric structures and the Gibbs free energies of formation (ΔGs, kJ mol^{-1}) of $[Ga(SBT)_2]^{3+}$ in the sorbitol solution. (a) four-coordination $[Ga(SBT)_2]^{3+}$ complexes; (b) five-coordination $[Ga(SBT)_2]^{3+}$ complexes; (c) six-coordination $[Ga(SBT)_2]^{3+}$ complexes. For clarity, hydrogen atoms on carbon are not shown. Bond lengths are reported in Å.

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Species	*ZPE*	E_c	G_0	G_c	E_r	G_r
SBT-I-1	0.21938	-688.03746	0.15320	-688.10365	0.0	0.0
SBT-I-2	0.21936	-688.03765	0.15400	-688.10301	0.5	1.7
SBT-I-3	0.21936	-688.03546	0.15355	-688.10127	5.3	6.2
SBT-I-4	0.21985	-688.03570	0.15512	-688.10043	4.6	8.5
SBT-I-5	0.21970	-688.03479	0.15448	-688.10000	7.0	9.6
SBT-I-6	0.22077	-688.03425	0.15712	-688.09791	8.4	15.1
SBT-I-7	0.21804	-688.03069	0.15090	-688.09783	17.8	15.3
SBT-II-1	0.21908	-688.00718	0.15256	-688.09993	79.5	9.8
SBT-II-2	0.22018	-688.03439	0.15535	-688.09921	8.1	11.6
SBT-II-3	0.22030	-688.03407	0.15628	-688.09809	8.9	14.6
SBT-III-1	0.21750	-688.03291	0.14949	-688.10092	11.9	7.2
SBT-III-2	0.21869	-688.03326	0.15205	-688.09990	11.0	9.9
SBT-III-3	0.21757	-688.03059	0.14892	-688.09924	18.0	11.6
SBT-IV-1	0.21800	-688.02654	0.15058	-688.09396	28.7	25.4
SBT-V-1	0.21924	-688.02106	0.15453	-688.08576	43.1	47.0

Table S2. Zero-point energies (*ZPE*, hartree), thermal correction to Gibbs free energy (G_0, hartree), total energies (E_c, hartree) corrected by *ZPE*, sum of electronic and thermal free energies (G_c, hartree) with *ZPE* and thermal corrections, and relative energies (E_r, kJ mol^{-1}) and relative Gibbs free energies (G_r, kJ mol^{-1}) relative to Ga^{3+} and OTf^- for $[Ga(OTf)_n]^{3-n}$ (n= 1, 2, 3) at M06/6-311++G(d,p) level in sorbitol solution.

$$Ga^{3+} + nOTf^- \rightarrow [Ga(OTf)_n]^{3-n}$$

$$\Delta G^\theta = G^\theta_{[Ga(OTf)_n]^{3-n}} - nG^\theta_{OTf^-} - G^\theta_{Ga^{3+}}$$

Species	*ZPE*	E_c	G_0	G_c	E_r	G_r
OTf^-	0.02747	-961.53419	-0.02407	-961.58573		
Ga^{3+}	0.00000	-1923.94153	-0.02450	-1923.96603	0.0	0.0
$[Ga(\eta^1_O\text{-}OTf)]^{2+}$	0.02852	-2885.67096	-0.02970	-2885.72918		
$[Ga(\eta^1_O\text{-}OTf)]^{2+} - OTf^-$	0.00105	-1924.13677	-0.00563	-1924.14345	-512.6	-465.8
$[Ga(\eta^2_{O,O}\text{-}OTf)]^{2+}$	0.02867	-2885.69505	-0.02911	-2885.75283		
$[Ga(\eta^2_{O,O}\text{-}OTf)]^{2+} - OTf^-$	0.00120	-1924.16086	-0.00504	-1924.16710	-575.9	-527.9
$[Ga(\eta^1_O\text{-}OTf)_2]^+$	0.05857	-3847.35874	-0.02328	-3847.44059		
$[Ga(\eta^1_O\text{-}OTf)_2]^+ - 2*OTf^-$	0.00363	-1924.29036	0.02486	-1924.26912	-915.8	-795.8
$[Ga(\eta^2_{O,O}\text{-}OTf)_2]^+$	0.05866	-3847.38914	-0.02171	-3847.46950		
$[Ga(\eta^2_{O,O}\text{-}OTf)_2]^{2+} - 2*OTf^-$	0.00372	-1924.32076	0.02644	-1924.29803	-995.7	-871.7
$Ga(\eta^1_O\text{-}OTf)_3$	0.08872	-4808.99900	-0.01329	-4809.10100		
$Ga(\eta^1_O\text{-}OTf)_3 - 3*OTf^-$	0.00630	-1924.39643	0.05893	-1924.34380	1194.3	-991.8
$(\eta^1_O\text{-}OTf)_2Ga(\eta^2_{O,O}\text{-}OTf)$	0.08861	-4809.01431	-0.01303	-4809.11595		
$(\eta^1_O\text{-}OTf)_2Ga(\eta^2_{O,O}\text{-}OTf) - 3*OTf^-$	0.00620	-1924.41174	0.05919	-1924.35875	-1234.5	-1031.1
$(\eta^1_O\text{-}OTf)Ga(\eta^2_{O,O}\text{-}OTf)_2$	0.08924	-4809.02160	-0.00709	-4809.11794		
$(\eta^1_O\text{-}OTf)Ga(\eta^2_{O,O}\text{-}OTf)_2 - 3*OTf^-$	0.00683	-1924.41903	0.06513	-1924.36074	-1253.7	-1036.3
$Ga(\eta^2_{O,O}\text{-}OTf)_3$	0.08872	-4809.02804	-0.01016	-4809.12692		
$Ga(\eta^2_{O,O}\text{-}OTf)_3 - 3*OTf^-$	0.00630	-1924.42547	0.06206	-1924.36972	-1270.6	-1059.9

Table S3. Zero-point energies (*ZPE*, hartree), thermal correction to Gibbs free energy (G_0, hartree), total energies (E_c, hartree) corrected by *ZPE*, sum of electronic and thermal free energies (G_c, hartree) with *ZPE* and thermal corrections, and relative energies (E_r, kJ mol^{-1}) and relative Gibbs free energies (G_r, kJ mol^{-1}) relative to Ga^{3+} and SBT for $[Ga(SBT)_m]^{3+}$ (m= 1, 2) at M06/6-311++G(d,p) level in sorbitol solution.

$$Ga^{3+} + mSBT \rightarrow [Ga(SBT)_m]^{3+}$$

$$\Delta G^{\theta} = G^{\theta}_{[Ga(SBT)_m]^{3+}} - mG^{\theta}_{SBT} - G^{\theta}_{Ga^{3+}}$$

Species	*ZPE*	E_c	G_0	G_c	E_r	G_r
SBT	0.21938	-688.03746	0.15320	-688.10365		
Ga^{3+}	0.00000	-1923.94153	-0.02450	-1923.96603	0.0	0.0
$[(\eta^3_{O3\sim O5}\text{-SBT})Ga]^{3+}$	0.21872	-2612.25432	0.15012	-2612.32293		
$[(\eta^3_{O3\sim O5}\text{-SBT})Ga]^{3+} - SBT$	-0.00066	-1924.21686	-0.00308	-1924.21928	-722.9	-664.9
$[(\eta^3_{O4\sim O6}\text{-SBT})Ga]^{3+}$	0.21985	-2612.24951	0.15257	-2612.31679		
$[(\eta^3_{O4\sim O6}\text{-SBT})Ga]^{3+} - SBT$	0.00047	-1924.21204	-0.00063	-1924.21314	-710.2	-648.8
$[(\eta^4_{O2\sim O5}\text{-SBT})Ga]^{3+}$	0.22010	-2612.28170	0.15421	-2612.34758		
$[(\eta^4_{O2\sim O5}\text{-SBT})Ga]^{3+} - SBT$	0.00071	-1924.24423	0.00101	-1924.24393	-794.7	-729.6
$[(\eta^4_{O3\sim O6}\text{-SBT})Ga]^{3+}$	0.22099	-2612.26406	0.15532	-2612.32973		
$[(\eta^4_{O3\sim O6}\text{-SBT})Ga]^{3+} - SBT$	0.00160	-1924.22659	0.00212	-1924.22608	-748.4	-682.8
$[Ga(\eta^2_{O3,O5}\text{-SBT})_2]^{3+}$	0.44050	-3300.41595	0.33666	-3300.51979		
$[Ga(\eta^2_{O3,O5}\text{-SBT})_2]^{3+} - 2*SBT$	0.00173	-1924.34102	0.03027	-1924.31249	-1048.9	-909.6
$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^2_{O4,O6}\text{-SBT})]^{3+}$	0.43955	-3300.40844	0.33425	-3300.51374		
$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^2_{O4,O6}\text{-SBT})]^{3+} - 2*SBT$	0.00078	-1924.33351	0.02786	-1924.30644	-1029.2	-893.7
$[Ga(\eta^2_{O4,O6}\text{-SBT})_2]^{3+}$	0.44023	-3300.39986	0.33818	-3300.50192		
$[Ga(\eta^2_{O4,O6}\text{-SBT})_2]^{3+} - 2*SBT$	0.00146	-1924.32494	0.03178	-1924.29462	-1006.6	-862.7
$[(\eta^2_{O2,O4}\text{-SBT})Ga(\eta^3_{O2\sim O4}\text{-SBT})]^{3+}$	0.44332	-3300.44920	0.34567	-3300.54685		
$[(\eta^2_{O2,O4}\text{-SBT})Ga(\eta^3_{O2\sim O4}\text{-SBT})]^{3+} - 2*SBT$	0.00455	-1924.37427	0.03927	-1924.33955	-1136.2	-980.7
$[(\eta^2_{O2,O4}\text{-SBT})Ga(\eta^3_{O3\sim O5}\text{-SBT})]^{3+}$	0.44207	-3300.44801	0.34105	-3300.54903		
$[(\eta^2_{O2,O4}\text{-SBT})Ga(\eta^3_{O3\sim O5}\text{-SBT})]^{3+} - 2*SBT$	0.00331	-1924.37308	0.03465	-1924.34173	-1133.0	-986.4
$([\eta^2_{O2,O4}\text{-SBT})Ga(\eta^3_{O4\sim O6}\text{-SBT})]^{3+}$	0.44235	-3300.43659	0.34167	-3300.53726		
$[(\eta^2_{O2,O4}\text{-SBT})Ga(\eta^3_{O4\sim O6}\text{-SBT})]^{3+} - 2*SBT$	0.00358	-1924.36166	0.03527	-1924.32996	-1103.0	-955.5
$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^3_{O2\sim O4}\text{-SBT})]^{3+}$	0.44158	-3300.44409	0.33885	-3300.54682		
$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^3_{O2\sim O4}\text{-SBT})]^{3+} - 2*SBT$	0.00282	-1924.36916	0.03245	-1924.33952	-1122.7	-980.6
$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^3_{O3\sim O5}\text{-SBT})]^{3+}$	0.44165	-3300.44038	0.33972	-3300.54230		
$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^3_{O3\sim O5}\text{-SBT})]^{3+} - 2*SBT$	0.00288	-1924.36545	0.03332	-1924.33500	-1113.0	-968.7
$[(\eta^2_{O3,O5}\text{-SBT})Ga(\eta^3_{O4\sim O6}\text{-}$	0.44315	-3300.43467	0.34394	-3300.53388		

SBT)]$^{3+}$						
[($^{2}\eta_{O3,O5}$-SBT)Ga($^{3}\eta_{O4\sim O6}$-SBT)]$^{3+}$ − 2*SBT	0.00438	-1924.35974	0.03754	-1924.32658	-1098.0	-946.6
[($^{2}\eta_{O4,O6}$-SBT)Ga($^{3}\eta_{O2\sim O4}$-SBT)]$^{3+}$	0.44139	-3300.43824	0.33946	-3300.54017		
[($^{2}\eta_{O4,O6}$-SBT)Ga($^{3}\eta_{O2\sim O4}$-SBT)]$^{3+}$ − 2*SBT	0.00262	-1924.36331	0.03306	-1924.33287	-1107.4	-963.1

Continued from **Table S3**

Species	*ZPE*	E_c	G_0	G_c	E_r	G_r
[($^{2}\eta_{O4,O6}$-SBT)Ga($^{3}\eta_{O3\sim O5}$-SBT)]$^{3+}$	0.44130	-3300.43239	0.33608	-3300.53761		
[($^{2}\eta_{O4,O6}$-SBT)Ga($^{3}\eta_{O3\sim O5}$-SBT)]$^{3+}$ − 2*SBT	0.00253	-1924.35747	0.02968	-1924.33031	-1092.0	-956.4
[($^{2}\eta_{O4,O6}$-SBT)Ga($^{3}\eta_{O4\sim O6}$-SBT)]$^{3+}$	0.44247	-3300.44435	0.34039	-3300.54643		
[($^{2}\eta_{O4,O6}$-SBT)Ga($^{3}\eta_{O4\sim O6}$-SBT)]$^{3+}$ − 2*SBT	0.00371	-1924.36942	0.03399	-1924.33914	-1123.4	-979.6
[($^{4}\eta_{O2\sim O5}$-SBT)Ga($^{1}\eta_{O4}$-SBT)]$^{3+}$	0.44160	-3300.41365	0.34096	-3300.51429		
[($^{4}\eta_{O2\sim O5}$-SBT)Ga($^{1}\eta_{O4}$-SBT)]$^{3+}$ − 2*SBT	0.00283	-1924.33872	0.03456	-1924.30699	-1042.8	-895.2
[($^{3}\eta_{O3\sim O5}$-SBT)Ga($^{2}\eta_{O2,O3}$-SBT)]$^{3+}$	0.44098	-3300.43482	0.33773	-3300.53807		
[($^{3}\eta_{O3\sim O5}$-SBT)Ga($^{2}\eta_{O2,O3}$-SBT)]$^{3+}$ − 2*SBT	0.00221	-1924.35989	0.03133	-1924.33077	-1098.4	-957.6
[Ga($^{3}\eta_{O2\sim O4}$-SBT)$_2$]$^{3+}$	0.44190	-3300.49148	0.34158	-3300.59179		
[Ga($^{3}\eta_{O2\sim O4}$-SBT)$_2$]$^{3+}$ − 2*SBT	0.00313	-1924.41655	0.03518	-1924.38449	-1247.2	-1098.7
[($^{3}\eta_{O2\sim O4}$-SBT) Ga($^{3}\eta_{O3\sim O5}$-SBT)]$^{3+}$	0.44213	-3300.47831	0.34202	-3300.57841		
[($^{3}\eta_{O2\sim O4}$-SBT) Ga($^{3}\eta_{O3\sim O5}$-SBT)]$^{3+}$ − 2*SBT	0.00336	-1924.40338	0.03562	-1924.37112	-1212.6	-1063.6
[($^{3}\eta_{O2\sim O4}$-SBT)Ga($^{3}\eta_{O4\sim O6}$-SBT)]$^{3+}$	0.44285	-3300.48157	0.34272	-3300.58170		
[($^{3}\eta_{O2\sim O4}$-SBT)Ga($^{3}\eta_{O4\sim O6}$-SBT)]$^{3+}$ − 2*SBT	0.00408	-1924.40664	0.03632	-1924.37440	-1221.2	-1072.2
[Ga($^{3}\eta_{O3\sim O5}$-SBT)$_2$]$^{3+}$	0.44160	-3300.47326	0.34008	-3300.57478		
[Ga($^{3}\eta_{O3\sim O5}$-SBT)$_2$]$^{3+}$ − 2*SBT	0.00284	-1924.39833	0.03368	-1924.36748	-1199.3	-1054.0
[($^{3}\eta_{O3\sim O5}$-SBT)Ga($^{3}\eta_{O4\sim O6}$-SBT)]$^{3+}$	0.44310	-3300.46997	0.34479	-3300.56828		
[($^{3}\eta_{O3\sim O5}$-SBT)Ga($^{3}\eta_{O4\sim O6}$-SBT)]$^{3+}$ − 2*SBT	0.00433	-1924.39504	0.03840	-1924.36098	-1190.7	-1036.9
[Ga($^{3}\eta_{O4\sim O6}$-SBT)$_2$]$^{3+}$	0.44221	-3300.47227	0.34198	-3300.57250		
[Ga($^{3}\eta_{O4\sim O6}$-SBT)$_2$]$^{3+}$ − 2*SBT	0.00345	-1924.39734	0.03558	-1924.36521	-1196.7	-1048.0

Table S4. Zero-point energies (*ZPE*, hartree), thermal correction to Gibbs free energy (G_0, hartree), total energies (E_c, hartree) corrected by *ZPE*, sum of electronic and thermal free energies (G_c, hartree) with *ZPE* and thermal corrections, and relative energies (E_r, kJ mol^{-1}) and relative Gibbs free energies (G_r, kJ mol^{-1}) relative to Ga^{3+}, SBT and OTf^- for $[Ga(OTf)_n(SBT)_m]^{3-n}$ (n = 1,2; m = 1, 2) at M06/6-311++G(d,p) level in sorbitol solution.

$$Ga^{3+} + nOTf^- + mSBT \rightarrow [Ga(OTf)_n(SBT)_m]^{3-n}$$

$$\Delta G^\theta = G^\theta_{[Ga(OTf)_n(SBT)_m]^{3-n}} - nG^\theta_{OTf^-} - mG^\theta_{SBT} - G^\theta_{Ga^{3+}}$$

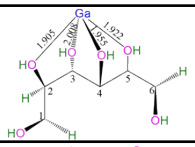
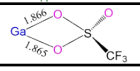
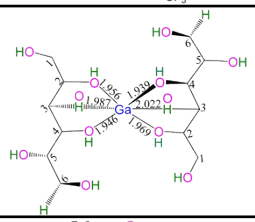
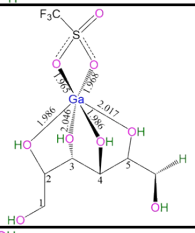
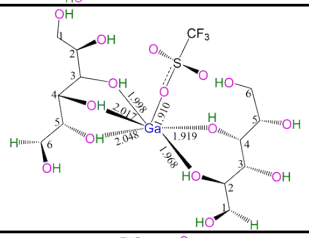
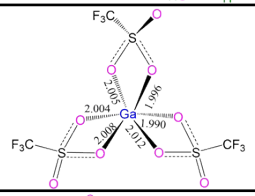
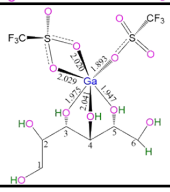
Species	*ZPE*	E_c	G_0	G_c	E_r	G_r
SBT	0.21938	-688.03746	0.15320	-688.10365		
OTf^-	0.02747	-961.53419	-0.02407	-961.58573		
Ga^{3+}	0.00000	-1923.94153	-0.02450	-1923.96603	0.0	0.0
$[(\eta^1_O$-OTf)Ga($\eta^4_{O2\sim O5}$-SBT)]$^{2+}$	0.25068	-3573.93642	0.16100	-3574.02610		
$[(\eta^1_O$-OTf)Ga($\eta^4_{O2\sim O5}$-SBT)]$^{2+}$ – SBT – OTf^-	0.00382	-1924.36477	0.03187	-1924.33672	-1111.2	-973.2
$[(\eta^1_O$- OTf)Ga($\eta^4_{O3\sim O6}$-SBT)]$^{2+}$	0.25216	-3573.92595	0.16530	-3574.01282		
$[(\eta^1_O$- OTf)Ga($\eta^4_{O3\sim O6}$-SBT)]$^{2+}$ – SBT – OTf^-	0.00531	-1924.35429	0.03617	-1924.32343	-1083.7	-938.4
$[(\eta^2_{O,O}$- OTf)Ga($\eta^4_{O2\sim O5}$-SBT)]$^{2+}$	0.25053	-3573.95198	0.16252	-3574.03999		
$[(\eta^2_{O,O}$- OTf)Ga($\eta^4_{O2\sim O5}$-SBT)]$^{2+}$ – SBT – OTf^-	0.00367	-1924.38033	0.03340	-1924.35061	-1152.1	-1009.7
$[(\eta^2_{O,O}$- OTf)Ga($\eta^4_{O3\sim O6}$-SBT)]$^{2+}$	0.25215	-3573.93721	0.16476	-3574.02460		
$[(\eta^2_{O,O}$- OTf)Ga($\eta^4_{O3\sim O6}$-SBT)]$^{2+}$ – SBT – OTf^-	0.00530	-1924.36556	0.03563	-1924.33522	-1113.3	-969.3
$[(\eta^1_O$-OTf) ($\eta^2_{O2,O4,}$-SBT)Ga($\eta^3_{O3\sim O5}$-SBT)]$^{2+}$	0.47426	-4262.06017	0.35705	-4262.17737		
$[(\eta^1_O$-OTf) ($\eta^2_{O2,O4,}$-SBT)Ga($\eta^3_{O3\sim O5}$-SBT)]$^{2+}$ - 2*SBT – OTf^-	0.00802	-1924.45105	0.07473	-1924.38434	-1337.7	-1098.3
$[(\eta^2_{O,O}$- OTf) (η^1_{O4}-SBT)Ga($\eta^2_{O4,O6}$-SBT)]$^{2+}$	0.47400	-4262.00372	0.35546	-4262.12226		
$[(\eta^2_{O,O}$- OTf) (η^1_{O4}-SBT)Ga($\eta^2_{O4,O6}$-SBT)]$^{2+}$ – 2*SBT – OTf^-	0.00776	-1924.39460	0.07314	-1924.32923	-1189.5	-953.6
$[(\eta^1_O$-OTf)$_2$Ga($\eta^2_{O2,O4}$-SBT)]$^+$	0.28145	-4535.54195	0.17268	-4535.65072		
$[(\eta^1_O$-OTf)$_2$Ga($\eta^2_{O2,O4}$-SBT)]$^+$ – SBT – 2*OTf^-	0.00712	-1924.43610	0.06762	-1924.37560	-1298.5	-1075.3
$[(\eta^2_{O,O}$-OTf)$_2$Ga($\eta^2_{O2,O4}$-SBT)]$^+$	0.28213	-4535.54119	0.17453	-4535.64879		
$[(\eta^2_{O,O}$-OTf)$_2$Ga($\eta^2_{O2,O4}$-SBT)]$^+$ – SBT – 2*OTf^-	0.00780	-1924.43535	0.06948	-1924.37367	-1296.5	-1070.3
$[(\eta^1_O$-OTf) ($\eta^2_{O,O}$-OTf)Ga($\eta^3_{O3\sim O5}$-SBT)]$^+$	0.28093	-4535.55419	0.16835	-4535.66677		
$[(\eta^1_O$-OTf) ($\eta^2_{O,O}$-OTf)Ga($\eta^3_{O3\sim O5}$-SBT)]$^+$ – SBT – 2*OTf^-	0.00660	-1924.44835	0.06329	-1924.39165	-1330.6	-1117.5
$[(\eta^1_O$-OTf)$_2$Ga($\eta^4_{O2\sim O5}$-SBT)]$^+$	0.28186	-4535.55152	0.17454	-4535.65884		
$[(\eta^1_O$-OTf)$_2$Ga($\eta^4_{O2\sim O5}$-SBT)]$^+$ – SBT – 2*OTf^-	0.00753	-1924.44567	0.06949	-1924.38372	-1323.6	-1096.6
$[(\eta^1_O$-OTf)$_2$Ga($\eta^4_{O3\sim O6}$-SBT)]$^+$	0.28239	-4535.53989	0.17483	-4535.64744		
$[(\eta^1_O$-OTf)$_2$Ga($\eta^4_{O3\sim O6}$-SBT)]$^+$ – SBT – 2*OTf^-	0.00806	-1924.43405	0.06978	-1924.37233	-1293.1	-1066.7

Table S5. In THF solution with both SBT and $Ga(OTf)_3$ compounds, the species was characterized by ESI-MS spectra.[a]

Species	m/z
$[Ga(SBT)]^{3+}$	83.67
$[Ga(OTf)]^{2+}$	109.94
$[Ga(SBT)_2]^{3+}$	144.36
$[Ga(OTf)(SBT)]^{2+}$	199.98
$[Ga(OTf)(SBT)_2]^{2+}$	291.02
$[[Ga(OTf)_2(SBT)]^+ - F^-]^{2+}$	264.95
$[[Ga(OTf)_2(SBT)]^+ - 2F^-]^{3+}$	170.30
$[[Ga(OTf)_2(SBT)]^+ - 3F^-]^{4+}$	122.98
$[[Ga(SBT)]^{3+} - OH^-]^{4+}$	58.50
$[[Ga(OTf)_3] - F^-]^+$	496.78
$[[Ga(OTf)(SBT)_2]^{2+} - OH^-]^{3+}$	188.34
$[[Ga(OTf)(SBT)]^{2+} - OH^-]^{3+}$	127.65
$[[Ga(OTf)(SBT)]^{2+} - CF_3^+ - OH^-]^{2+}$	248.02
$[[Ga(SBT)_2]^{3+} - H^+ - OH^-]^{3+}$	138.36
$[[Ga(OTf)_2(SBT)]^+ - 6F^- - H^+]^{6+}$	72.32
$[[Ga(OTf)(SBT)]^{2+} - F^-]^{3+}$	126.99
$[[Ga(OTf)(SBT)]^{2+} - OH^-]^{3+}$	127.65
$[(OTf)]^- - F^-]$	129.95
$[[Ga(OTf)(SBT)_2]^{2+} - 4OH^- - F^-]^{5+}$	70.72
$[[Ga(OTf)(SBT)_2]^{2+} - 5OH^-]^{7+}$	71.00
$[[Ga(OTf)(SBT)_2]^{2+} - 2H^+ - 6OH^- - 2F^-]^{8+}$	55.00

[a] ESI-MS (X500R QTOF, AB Sciex LP) spectra. The operating parameters were as follows: ionization voltage, 4.5 kV; interface temperature, 500 °C; nebulizer gas flow, 55 psi (N_2); detector voltage, 2.225 kV; continuum mode.

Table S6. Some typical species from experimental observation of ESI-MS spectra and the density functional theory (DFT) calculations at the M06/6-311++g(d,p), aug-cc-pvtz level.

ESI-MS		M06/6-311++g(d,p), aug-cc-pvtz	
Species	m/z	Geometric Structure	Relative Gibbs Free Energies (G_r, kJ mol^{-1})
$[Ga(SBT)]^{3+}$	83.67		–729.6
$[Ga(OTf)]^{2+}$	109.94		–527.9
$[Ga(SBT)_2]^{3+}$	144.36		–1098.7
$[Ga(OTf)(SBT)]^{2+}$	199.98		–1009.7
$[Ga(OTf)(SBT)_2]^{2+}$	291.02		–1098.3
$Ga(OTf)_3$			–1059.9
$[Ga(OTf)_2(SBT)]^{+}$			–1117.5