Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Structural and Thermochemical Investigation of 1,3-bis(λ^4-boraneyl)-1λ^4,3λ^4-imidazolidine Adduct for Chemical Hydrogen Storage

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Supporting Information

***Ab initio* Electronic Structure Calculations for All Conformers and Stereoisomers of 1,3-bis(λ⁴-boraneyl)-1λ⁴,3λ⁴-imidazolidine (IMBB)**

Table S1: Calculated coordinates at M06-2X/6-31G* level for the *cis* planar **IMBB** (A)

Atom	x	y	z
C	1.01868	0.73312	0.77287
C	1.01868	0.73312	-0.77287
C	-0.76428	-0.57060	0.00000
H	1.76172	0.04519	1.17837
H	1.16684	1.71460	1.22329
H	1.76172	0.04519	-1.17837
H	1.16684	1.71460	-1.22329
H	-1.84134	-0.72879	0.00000
H	-0.23797	-1.52736	0.00000
N	-0.31706	0.20360	1.16977
H	-0.95927	0.98491	1.31349
N	-0.31706	0.20360	-1.16977
B	-0.31706	-0.65728	2.57652
H	0.07893	0.12548	3.40879
H	-1.46893	-0.99503	2.72754
H	0.44591	-1.57282	2.35823
H	-0.95927	0.98491	-1.31349
B	-0.31706	-0.65728	-2.57652
H	-1.46893	-0.99503	-2.72754
H	0.07893	0.12548	-3.40879
H	0.44591	-1.57282	-2.35823

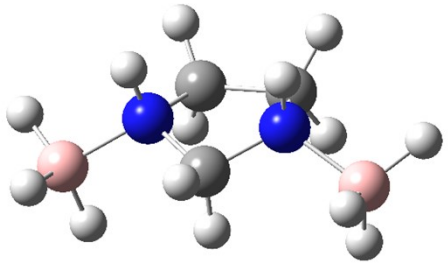

Figure S1: The optimized structure for the *cis* planar **IMBB** (A) at M06-2X/6-31G* level

Table S2: Calculated coordinates at M06-2X/6-31G* level for the *trans* planar **IMBB** (B)

Atom	x	y	z
C	-0.47715	1.20537	-0.39326
C	0.83904	1.23099	0.40542
C	-0.11451	-0.87868	0.66129
H	-1.24843	1.82342	0.06555
H	-0.34137	1.50087	-1.43256
H	0.68470	1.65481	1.40156
H	1.64778	1.75535	-0.10221
H	-0.03135	-1.94601	0.46533
H	-0.56038	-0.68458	1.64090
N	-0.94374	-0.21466	-0.35113
H	-0.71767	-0.64448	-1.25002
N	1.19809	-0.20933	0.52099
B	-2.55349	-0.39013	-0.06093
H	-3.10060	0.16303	-0.98547
H	-2.73635	-1.58565	-0.02041
H	-2.71348	0.16703	1.00649
H	1.77415	-0.37580	1.34664
B	2.04635	-0.74601	-0.78780
H	1.37165	-0.44671	-1.75358
H	3.09025	-0.13997	-0.74578
H	2.15213	-1.93878	-0.62255

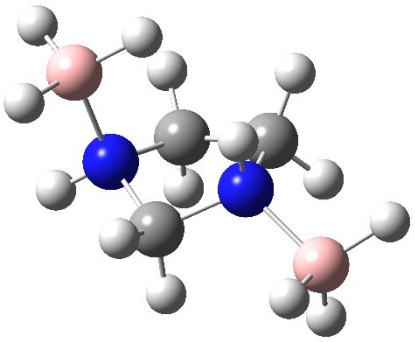

Figure S2: The optimized structure for the *trans* planar **IMBB** (B) at M06-2X/6-31G* level

Table S3: Calculated coordinates at M06-2X/6-31G* level for the *cis* enveloped **IMBB** (C)

Atom	x	y	z
C	1.01856	0.73318	0.77292
C	1.01856	0.73318	-0.77292
H	1.16680	1.71488	1.22283
H	1.76182	0.04555	1.17844
H	1.16680	1.71488	-1.22283
H	1.76182	0.04555	-1.17844
C	-0.76374	-0.57121	0.00000
H	-1.84065	-0.73013	0.00000
H	-0.23651	-1.52743	0.00000
N	-0.31715	0.20362	1.16970
N	-0.31715	0.20362	-1.16970
H	-0.95973	0.98460	-1.31324
B	-0.31715	-0.65707	-2.57680
H	0.07879	0.12572	-3.40904
H	0.44566	-1.57272	-2.35880
H	-1.46907	-0.99473	-2.72803
H	-0.95973	0.98460	1.31324
B	-0.31715	-0.65707	2.57680
H	0.44566	-1.57272	2.35880
H	0.07879	0.12572	3.40904
H	-1.46907	-0.99473	2.72803

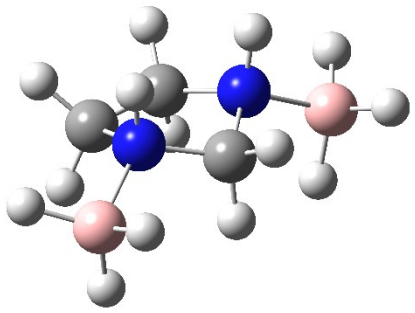

Figure S3: The optimized structure for the *cis* enveloped **IMBB** (C) at M06-2X/6-31G* level

Table S4: Calculated coordinates at M06-2X/6-31G* level for the *trans* enveloped **IMBB** (D)

Atom	x	y	z
C	-0.83860	1.23090	0.40602
C	0.47707	1.20540	-0.39344
H	-1.64754	1.75563	-0.10088
H	-0.68353	1.65419	1.40226
H	0.34055	1.50090	-1.43264
H	1.24857	1.82350	0.06492
C	0.11448	-0.87926	0.66042
H	0.03089	-1.94636	0.46346
H	0.56062	-0.68619	1.64010
H	-1.77353	-0.37599	1.34710
N	-1.19797	-0.20943	0.52112
N	0.94387	-0.21455	-0.35155
B	-2.04690	-0.74540	-0.78752
H	-1.37201	-0.44666	-1.75336
H	-2.15359	-1.93808	-0.62232
H	-3.09030	-0.13850	-0.74546
H	0.71848	-0.64416	-1.25070
B	2.55334	-0.39003	-0.06050
H	3.10119	0.16279	-0.98479
H	2.71290	0.16722	1.00697
H	2.73605	-1.58559	-0.01960

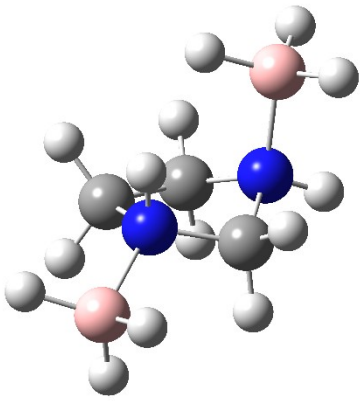

Figure S4: The optimized structure for the *trans* enveloped **IMBB** (D) at M06-2X/6-31G* level

Table S5: Calculated coordinates at M06-2X/6-31G* level for the *cis* twisted **IMBB** (E)

Atom	x	y	z
C	-0.61729	1.18065	0.35761
C	0.81829	1.26775	-0.15978
C	-0.05312	-0.96503	-0.26345
H	-0.65074	0.96177	1.42752
H	-1.23254	2.05552	0.14920
H	1.51724	1.67458	0.56996
H	0.88021	1.84474	-1.08609
H	-0.14336	-1.74547	-1.01712
H	-0.04574	-1.40150	0.73638
N	-1.16979	-0.01106	-0.32751
N	1.18807	-0.16095	-0.43423
H	-1.36361	0.22436	-1.30354
B	-2.57548	-0.59160	0.30584
B	2.40370	-0.71435	0.53821
H	1.52041	-0.24491	-1.39379
H	3.37102	-0.05995	0.22519
H	-3.36679	0.31111	0.15900
H	1.99232	-0.48941	1.65696
H	-2.82347	-1.57325	-0.35699
H	2.49270	-1.89270	0.27754
H	-2.30396	-0.83135	1.46136

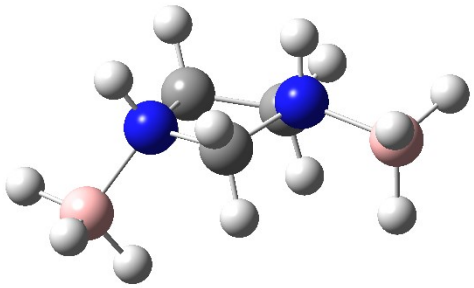

Figure S5: The optimized structure for the *cis* twisted **IMBB** (E) at M06-2X/6-31G* level

Table S6: Calculated coordinates at M06-

2X/6-31G* level for the *trans* twisted **IMBB** (F)

Atom	x	y	z
C	-0.83827	-1.23078	-0.40651
C	0.47694	-1.20522	0.39365
C	0.11443	0.87956	-0.66009
H	-0.68261	-1.65360	-1.40286
H	-1.64724	-1.75604	0.09977
H	1.24860	-1.82358	-0.06409
H	0.33967	-1.50047	1.43283
H	0.03058	1.94658	-0.46282
H	0.56062	0.68688	-1.63981
N	-1.19795	0.20947	-0.52113
N	0.94388	0.21468	0.35173
H	-1.77338	0.37620	-1.34716
B	-2.04720	0.74490	0.78749
H	-2.15407	1.93760	0.62256
H	-3.09048	0.13782	0.74508
H	-1.37241	0.44599	1.75335
H	0.71875	0.64428	1.25094
B	2.55350	0.38978	0.06031
H	2.73641	1.58529	0.01927
H	3.10133	-0.16311	0.98457
H	2.71252	-0.16765	-1.00713

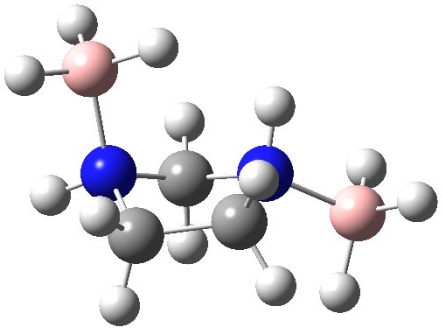

Figure S6: The optimized structure for the *trans* twisted **IMBB** (F) at M06-2X/6-31G* level

***Ab initio* Electronic Structure Calculations for *Trans* Conformer of IMBB (B) [This conformer will be referred to as IMBB from this point]**

Table S7: Calculated coordinates at MP2/6-31G* level for **IMBB**

Atom	x	y	z
C	-0.47574	1.20517	-0.39914
C	0.83112	1.23479	0.40867
C	-0.11283	-0.87746	0.66624
H	-1.24939	1.83091	0.04542
H	-0.33099	1.49087	-1.44056
H	0.66776	1.64931	1.40819
H	1.64016	1.77174	-0.08609
H	-0.03043	-1.94618	0.47551
H	-0.55258	-0.68127	1.64811
N	-0.94608	-0.21711	-0.34832
H	-0.72083	-0.65158	-1.24762
N	1.19992	-0.20627	0.51591
B	-2.56026	-0.39309	-0.06253
H	-3.10265	0.15268	-0.99504
H	-2.74059	-1.5895	-0.01943
H	-2.73125	0.16772	1.00028
H	1.77958	-0.36886	1.34438
B	2.05625	-0.74672	-0.78739
H	1.38576	-0.47681	-1.76271
H	3.09253	-0.12611	-0.74769
H	2.18081	-1.93527	-0.60100

Table S8: Calculated coordinates at MP2/6-311G* level for **IMBB**

Atom	**x**	**y**	**z**
C	-0.45534	1.19228	-0.42441
C	0.78815	1.21545	0.47235
C	-0.10877	-0.91624	0.61489
H	-1.24573	1.84294	-0.05411
H	-0.22095	1.44714	-1.45648
H	0.53942	1.56100	1.47982
H	1.60541	1.80889	0.06477
H	-0.00627	-1.97183	0.37129
H	-0.56121	-0.77864	1.59961
N	-0.94931	-0.22327	-0.37483
H	-0.76960	-0.65114	-1.28070
N	1.19304	-0.21715	0.52212
B	-2.54789	-0.36572	-0.03756
H	-3.12021	0.17780	-0.95662
H	-2.75761	-1.55887	0.03226
H	-2.68298	0.21061	1.02471
H	1.75250	-0.40252	1.35342
B	2.07929	-0.68165	-0.77816
H	1.40957	-0.42164	-1.75807
H	3.09318	-0.02180	-0.72137
H	2.25725	-1.87109	-0.62800

Table S9: Calculated coordinates at MP2/6-311+G* level for **IMBB**

Atom	x	y	z
C	-0.46604	1.19639	-0.41776
C	0.79561	1.22343	0.45509
C	-0.10682	-0.90850	0.62248
H	-1.25431	1.84043	-0.03035
H	-0.25427	1.45745	-1.45365
H	0.57042	1.58568	1.46249
H	1.60873	1.80559	0.02323
H	-0.00556	-1.96563	0.38409
H	-0.55481	-0.76470	1.60885
N	-0.95137	-0.22162	-0.36736
H	-0.76374	-0.64831	-1.27353
N	1.19587	-0.21113	0.51809
B	-2.55065	-0.37781	-0.04132
H	-3.12191	0.16771	-0.96130
H	-2.75429	-1.57330	0.02127
H	-2.69423	0.19355	1.02455
H	1.75520	-0.38725	1.35244
B	2.08219	-0.69778	-0.77444
H	1.40034	-0.46192	-1.75511
H	3.09098	-0.02728	-0.73823
H	2.27177	-1.88276	-0.59992

Table S10: Calculated coordinates at M06-2X/6-31G* level for **IMBB**

Atom	x	y	z
C	-0.47715	1.20537	-0.39326
C	0.83904	1.23099	0.40542
C	-0.11451	-0.87868	0.66129
H	-1.24843	1.82342	0.06555
H	-0.34136	1.50087	-1.43256
H	0.68470	1.65481	1.40156
H	1.64778	1.75535	-0.10221
H	-0.03135	-1.94601	0.46533
H	-0.56038	-0.68458	1.64090
N	-0.94374	-0.21466	-0.35113
H	-0.71767	-0.64448	-1.25002
N	1.19809	-0.20933	0.52099
B	-2.55349	-0.39013	-0.06093
H	-3.10060	0.16303	-0.98547
H	-2.73635	-1.58565	-0.02041
H	-2.71348	0.16703	1.00649
H	1.77415	-0.37580	1.34664
B	2.04635	-0.74601	-0.78780
H	1.37165	-0.44671	-1.75358
H	3.09024	-0.13997	-0.74578
H	2.15212	-1.93878	-0.62255

Table S11: Calculated coordinates at M06-2X/cc-pVQZ level for **IMBB**

Atom	**x**	**y**	**z**
C	-0.47186	1.19805	-0.40241
C	0.81066	1.22040	0.43772
C	-0.10891	-0.90192	0.62644
H	-1.24769	1.82905	0.01691
H	-0.29142	1.47791	-1.43381
H	0.61378	1.60300	1.43741
H	1.61830	1.77896	-0.02177
H	-0.01180	-1.95617	0.39624
H	-0.55391	-0.75033	1.60819
N	-0.94778	-0.21678	-0.36213
H	-0.74723	-0.63525	-1.26555
N	1.19231	-0.21286	0.51740
B	-2.53998	-0.38022	-0.04671
H	-3.10560	0.17550	-0.95258
H	-2.73848	-1.56867	-0.00220
H	-2.68435	0.17153	1.02028
H	1.75272	-0.38843	1.34481
B	2.06334	-0.70661	-0.77561
H	1.38732	-0.43870	-1.74259
H	3.08089	-0.06586	-0.73282
H	2.21967	-1.89008	-0.61834

***Ab initio* Electronic Structure Calculations for the MDIMBB and DDIMBB by the Removal of Hydrogen on the BH_3 Above the Ring and Hydrogen on Corresponding Nitrogen Followed by a Hydrogen on the BH_3 Below the Ring and Hydrogen on the Respective Nitrogen**

Table S12: Calculated coordinates at MP2/6-31G* level for the **MDIMBB**

Atom	x	y	z
C	-0.62115	1.25174	-0.09321
C	0.89789	1.22191	0.20016
C	0.02648	-0.93913	0.37579
H	-1.20249	1.57540	0.77121
H	-0.89891	1.86605	-0.95076
H	1.10251	1.61291	1.20257
H	1.48938	1.79048	-0.51961
H	0.07654	-1.96867	0.02315
H	-0.26127	-0.91456	1.43287
N	-1.00285	-0.16675	-0.35781
H	-0.86368	-0.35518	-1.35506
N	1.24684	-0.20319	0.11528
B	-2.55591	-0.54509	0.01523
H	-3.22932	0.19260	-0.66917
H	-2.66982	-1.71589	-0.27491
H	-2.64645	-0.32268	1.20427
B	2.47844	-0.74401	-0.26050
H	2.58334	-1.93159	-0.33073
H	3.38027	-0.00090	-0.50619

Table S13: Calculated coordinates at MP2/6-311G* level for the **MDIMBB**

Atom	x	y	z
C	-0.61507	1.24971	-0.13180
C	0.88916	1.21576	0.23537
C	0.02773	-0.93832	0.38185
H	-1.23277	1.61758	0.68706
H	-0.83936	1.82907	-1.02773
H	1.03910	1.56411	1.26211
H	1.51187	1.81259	-0.43128
H	0.07930	-1.96987	0.03834
H	-0.25807	-0.90210	1.43810
N	-1.00155	-0.17532	-0.36184
H	-0.87267	-0.38634	-1.35044
N	1.24695	-0.20324	0.11994
B	-2.54613	-0.53620	0.02607
H	-3.22792	0.19152	-0.66539
H	-2.67908	-1.71359	-0.23887
H	-2.63884	-0.29268	1.21268
B	2.47425	-0.73508	-0.27754
H	2.57683	-1.92347	-0.36723
H	3.37241	0.01662	-0.51915

Table S14: Calculated coordinates at MP2/6-311+G* level for the **MDIMBB**

Atom	**x**	**y**	**z**
C	-0.60894	1.25090	-0.15293
C	0.88438	1.21254	0.25443
C	0.02715	-0.94225	0.37962
H	-1.24686	1.64584	0.63774
H	-0.79893	1.81086	-1.06920
H	1.00485	1.54332	1.29133
H	1.52516	1.82071	-0.38442
H	0.08287	-1.97192	0.03018
H	-0.26229	-0.91207	1.43555
N	-1.00194	-0.17696	-0.36378
H	-0.88089	-0.39921	-1.35202
N	1.24682	-0.20388	0.12771
B	-2.54560	-0.53154	0.03452
H	-3.23243	0.17765	-0.67295
H	-2.67717	-1.71653	-0.20052
H	-2.63293	-0.25734	1.21639
B	2.47396	-0.73049	-0.28547
H	2.57681	-1.91782	-0.38998
H	3.37033	0.02536	-0.52157

Table S15: Calculated coordinates at M06-2X/6-31G* level for the **MDIMBB**

Atom	x	y	z
C	-0.63219	1.24881	-0.05449
C	0.89953	1.22301	0.16508
C	0.02711	-0.94495	0.34099
H	-1.17316	1.52042	0.85359
H	-0.96132	1.89856	-0.86516
H	1.15236	1.64557	1.14252
H	1.45287	1.77065	-0.59980
H	0.07689	-1.96329	-0.04182
H	-0.26151	-0.95383	1.39846
N	-1.00856	-0.15745	-0.36130
H	-0.88433	-0.31427	-1.36336
N	1.24606	-0.20223	0.10124
B	-2.54969	-0.55162	0.02421
H	-3.23820	0.20344	-0.62458
H	-2.66637	-1.71403	-0.29479
H	-2.61717	-0.36235	1.22119
B	2.48911	-0.73939	-0.22297
H	2.59997	-1.92647	-0.27786
H	3.39363	0.00722	-0.44361

Table S16: Calculated coordinates at M06-2X/cc-pVQZ level for the **MDIMBB**

Atom	**x**	**y**	**z**
C	-0.62868	1.24637	-0.07416
C	0.89328	1.21825	0.18254
C	0.02764	-0.94490	0.34330
H	-1.18630	1.54712	0.80796
H	-0.92703	1.87623	-0.90516
H	1.11781	1.61939	1.17009
H	1.46160	1.77943	-0.55299
H	0.07986	-1.95950	-0.03464
H	-0.25538	-0.95163	1.39721
N	-1.00681	-0.16177	-0.35940
H	-0.89019	-0.33073	-1.35464
N	1.24333	-0.20229	0.10265
B	-2.53613	-0.54598	0.02848
H	-3.22839	0.19604	-0.62337
H	-2.66326	-1.70768	-0.27170
H	-2.61053	-0.34423	1.21761
B	2.48028	-0.73277	-0.23188
H	2.59292	-1.91384	-0.29660
H	3.37905	0.01322	-0.44970

Table S17: Calculated coordinates at MP2/6-31G* level for the **DDIMBB**

Atom	**x**	**y**	**z**
C	-0.65629	1.17204	0.38608
C	0.65621	1.17207	-0.38599
C	0.00002	-1.07641	0.00021
H	-0.46337	1.30593	1.45809
H	-1.37841	1.91908	0.05233
H	1.37830	1.91914	-0.05222
H	0.46328	1.30600	-1.45800
H	-0.10984	-1.71163	-0.88431
H	0.10992	-1.71122	0.88503
N	-1.15508	-0.17569	0.10460
N	1.15507	-0.17565	-0.10456
B	-2.47849	-0.54047	-0.13633
H	-2.71789	-1.67632	-0.42409
H	-3.33231	0.29147	-0.05336
B	2.47856	-0.54042	0.13596
H	2.71804	-1.67626	0.42367
H	3.33236	0.29151	0.05269

Table S18: Calculated coordinates at MP2/6-311G* level for the **DDIMBB**

Atom	**x**	**y**	**z**
C	-0.65442	1.17066	0.38985
C	0.65433	1.17070	-0.38976
C	0.00002	-1.07653	0.00023
H	-0.45333	1.29591	1.46057
H	-1.37666	1.91880	0.06220
H	1.37655	1.91886	-0.06209
H	0.45323	1.29599	-1.46047
H	-0.10894	-1.70841	-0.88516
H	0.10902	-1.70796	0.88594
N	-1.15595	-0.17596	0.10563
N	1.15594	-0.17591	-0.10560
B	-2.47797	-0.53830	-0.13742
H	-2.71563	-1.67574	-0.42870
H	-3.33055	0.29751	-0.05211
B	2.47804	-0.53824	0.13702
H	2.71579	-1.67568	0.42825
H	3.33060	0.29755	0.05140

Table S19: Calculated coordinates at MP2/6-311+G* level for the **DDIMBB**

Atom	**x**	**y**	**z**
C	-0.65645	1.17351	0.38726
C	0.65636	1.17355	-0.38716
C	0.00002	-1.07685	0.00025
H	-0.46012	1.30581	1.45825
H	-1.37973	1.91790	0.05260
H	1.37961	1.91796	-0.05247
H	0.46002	1.30590	-1.45814
H	-0.11443	-1.70848	-0.88487
H	0.11452	-1.70798	0.88572
N	-1.15652	-0.17619	0.11115
N	1.15651	-0.17614	-0.11112
B	-2.47917	-0.54133	-0.13869
H	-2.71328	-1.67979	-0.42931
H	-3.33346	0.29326	-0.05619
B	2.47925	-0.54128	0.13825
H	2.71346	-1.67972	0.42882
H	3.33351	0.29330	0.05543

Table S20: Calculated coordinates at M06-2X/6-31G* level for the **DDIMBB**

Atom	**x**	**y**	**z**
C	-0.66865	1.17619	0.36771
C	0.66858	1.17622	-0.36763
C	0.00002	-1.06802	0.00021
H	-0.50634	1.32580	1.44270
H	-1.38141	1.91736	0.00431
H	1.38130	1.91742	-0.00421
H	0.50625	1.32587	-1.44261
H	-0.10288	-1.70517	-0.88394
H	0.10295	-1.70477	0.88465
N	-1.15652	-0.17419	0.09525
N	1.15651	-0.17415	-0.09522
B	-2.47404	-0.55124	-0.13139
H	-2.70335	-1.69273	-0.40184
H	-3.33158	0.27655	-0.05507
B	2.47410	-0.55119	0.13103
H	2.70349	-1.69268	0.40144
H	3.33162	0.27659	0.05444

Table S21: Calculated coordinates at M06-2X/cc-pVQZ level for the **DDIMBB**

Atom	**x**	**y**	**z**
C	-0.66894	1.17514	0.36244
C	0.66887	1.17517	-0.36235
C	0.00002	-1.06666	0.00023
H	-0.51325	1.32662	1.43284
H	-1.37580	1.91249	-0.00445
H	1.37569	1.91254	0.00457
H	0.51316	1.32671	-1.43274
H	-0.09861	-1.70038	-0.88026
H	0.09869	-1.69993	0.88104
N	-1.15562	-0.17411	0.09190
N	1.15562	-0.17406	-0.09187
B	-2.46822	-0.55106	-0.12961
H	-2.69709	-1.68840	-0.39482
H	-3.32302	0.27214	-0.05405
B	2.46829	-0.55101	0.12922
H	2.69723	-1.68835	0.39439
H	3.32307	0.27218	0.05337

***Ab initio* Electronic Structure Calculations for the MDIMBB and DDIMBB by the Removal of Hydrogen on the BH_3 Below the Ring and Hydrogen on Corresponding Nitrogen Followed by Hydrogen on the BH_3 Above the Ring and Hydrogen on the Respective Nitrogen**

Table S22: Calculated coordinates at MP2/6-31G* level for the **MDIMBB**

Atom	x	y	z
C	0.89795	1.22186	0.20009
C	-0.62123	1.25171	-0.09309
C	0.02652	-0.93908	0.37580
H	1.48938	1.79044	-0.51971
H	1.10269	1.61280	1.20250
H	-0.89916	1.86605	-0.95056
H	-1.20245	1.57528	0.77145
H	-0.26108	-0.91450	1.43294
H	0.07644	-1.96864	0.02321
N	1.24681	-0.20321	0.11505
N	-1.00294	-0.16673	-0.35773
B	2.47853	-0.74394	-0.26042
H	3.38043	-0.00088	-0.50602
H	2.58377	-1.93151	-0.33051
H	-0.86367	-0.35507	-1.35499
B	-2.55596	-0.54512	0.01517
H	-2.64655	-0.32306	1.20427
H	-3.22932	0.19281	-0.66903
H	-2.66987	-1.71584	-0.27531

Table S23: Calculated coordinates at MP2/6-311G* level for the **MDIMBB**

Atom	**x**	**y**	**z**
C	0.88930	1.21582	0.23484
C	-0.61525	1.24970	-0.13119
C	0.02776	-0.93828	0.38179
H	1.51153	1.81233	-0.43256
H	1.04005	1.56464	1.26131
H	-0.84042	1.82957	-1.02658
H	-1.23239	1.61690	0.68841
H	-0.25797	-0.90210	1.43809
H	0.07921	-1.96985	0.03826
N	1.24694	-0.20326	0.11972
N	-1.00163	-0.17523	-0.36177
B	2.47438	-0.73513	-0.27730
H	3.37260	0.01653	-0.51885
H	2.57725	-1.92353	-0.36654
H	-0.87257	-0.38592	-1.35043
B	-2.54623	-0.53634	0.02587
H	-2.63900	-0.29339	1.21260
H	-3.22795	0.19175	-0.66526
H	-2.67911	-1.71360	-0.23963

Table S24: Calculated coordinates at MP2/6-311+G* level for the **MDIMBB**

Atom	**x**	**y**	**z**
C	0.54520	1.22826	0.35208
C	-0.83210	1.22834	-0.31005
C	0.15209	-0.81652	-0.82596
H	1.15873	2.06338	0.00637
H	0.45885	1.26753	1.43867
H	-0.78955	1.66540	-1.31343
H	-1.60296	1.72242	0.28015
H	0.17386	-1.87707	-0.58046
H	0.29212	-0.66712	-1.90512
N	1.12788	-0.06525	-0.05215
N	-1.14689	-0.22437	-0.41555
B	2.37377	-0.55361	0.34569
H	3.07922	0.15418	1.00376
H	2.69462	-1.65462	0.00314
H	-1.83987	-0.37942	-1.14667
B	-1.77107	-0.86979	0.95923
H	-0.93386	-0.71268	1.82155
H	-2.79222	-0.24072	1.15546
H	-1.97057	-2.03736	0.68946

Table S25: Calculated coordinates at M06-2X/6-31G* level for the **MDIMBB**

Atom	**x**	**y**	**z**
C	0.89954	1.22300	0.16509
C	-0.63223	1.24874	-0.05447
C	0.02720	-0.94493	0.34125
H	1.45294	1.77073	-0.59967
H	1.15229	1.64546	1.14260
H	-0.96143	1.89832	-0.86526
H	-1.17327	1.52051	0.85353
H	-0.26134	-0.95356	1.39877
H	0.07683	-1.96336	-0.04135
N	1.24607	-0.20225	0.10115
N	-1.00858	-0.15754	-0.36110
B	2.48915	-0.73930	-0.22314
H	3.39358	0.00736	-0.44397
H	2.60025	-1.92637	-0.27792
H	-0.88417	-0.31443	-1.36314
B	-2.54975	-0.55157	0.02397
H	-2.61763	-0.36256	1.22096
H	-3.23796	0.20371	-0.62490
H	-2.66652	-1.71389	-0.29532

Table S26: Calculated coordinates at M06-2X/cc-pVQZ level for the **MDIMBB**

Atom	**x**	**y**	**z**
C	0.89327	1.21821	0.18260
C	-0.62869	1.24638	-0.07419
C	0.02767	-0.94492	0.34324
H	1.46165	1.77951	-0.55279
H	1.11774	1.61917	1.17025
H	-0.92693	1.87621	-0.90526
H	-1.18635	1.54720	0.80789
H	-0.25532	-0.95165	1.39718
H	0.07979	-1.95953	-0.03470
N	1.24331	-0.20232	0.10251
N	-1.00686	-0.16176	-0.35940
B	2.48035	-0.73270	-0.23181
H	3.37908	0.01336	-0.44954
H	2.59315	-1.91376	-0.29655
H	-0.89017	-0.33069	-1.35467
B	-2.53618	-0.54598	0.02855
H	-2.61041	-0.34430	1.21769
H	-3.22841	0.19613	-0.62321
H	-2.66329	-1.70765	-0.27172

Table S27: Calculated coordinates at MP2/6-31G* level for the **DDIMBB**

Atom	**x**	**y**	**z**
C	-0.65626	-1.17205	0.38604
C	0.65624	-1.17206	-0.38603
C	0.00000	1.07641	0.00004
H	-1.37837	-1.91910	0.05228
H	-0.46333	-1.30595	1.45806
H	0.46331	-1.30597	-1.45804
H	1.37834	-1.91911	-0.05226
H	0.10989	1.71138	0.88475
H	-0.10987	1.71147	-0.88460
N	-1.15507	0.17568	0.10459
N	1.15507	0.17567	-0.10458
B	-2.47852	0.54045	-0.13619
H	-3.33233	-0.29149	-0.05309
H	-2.71795	1.67630	-0.42392
B	2.47853	0.54044	0.13611
H	2.71798	1.67629	0.42384
H	3.33234	-0.29150	0.05296

Table S28: Calculated coordinates at MP2/6-311G* level for the **DDIMBB**

Atom	**x**	**y**	**z**
C	-0.65438	-1.17068	0.38982
C	0.65436	-1.17069	-0.38980
C	0.00000	1.07653	0.00005
H	-1.37662	-1.91883	0.06216
H	-0.45329	-1.29594	1.46053
H	0.45327	-1.29596	-1.46051
H	1.37659	-1.91884	-0.06213
H	0.10898	1.70814	0.88563
H	-0.10897	1.70823	-0.88547
N	-1.15595	0.17594	0.10562
N	1.15595	0.17593	-0.10561
B	-2.47800	0.53828	-0.13726
H	-3.33058	-0.29753	-0.05183
H	-2.71570	1.67571	-0.42852
B	2.47801	0.53827	0.13718
H	2.71573	1.67570	0.42843
H	3.33059	-0.29753	0.05168

Table S29: Calculated coordinates at MP2/6-311+G* level for the **DDIMBB**

Atom	**x**	**y**	**z**
C	-0.65641	-1.17352	0.38722
C	0.65639	-1.17353	-0.38720
C	0.00000	1.07685	0.00005
H	-1.37968	-1.91792	0.05255
H	-0.46008	-1.30584	1.45820
H	0.46006	-1.30586	-1.45818
H	1.37966	-1.91793	-0.05252
H	0.11448	1.70818	0.88538
H	-0.11446	1.70828	-0.88521
N	-1.15652	0.17617	0.11114
N	1.15652	0.17616	-0.11113
B	-2.47920	0.54131	-0.13851
H	-3.33348	-0.29328	-0.05589
H	-2.71336	1.67976	-0.42911
B	2.47921	0.54130	0.13843
H	2.71339	1.67975	0.42902
H	3.33349	-0.29328	0.05573

Table S30: Calculated coordinates at M06-2X/6-31G* level for the **DDIMBB**

Atom	**x**	**y**	**z**
C	-0.66863	-1.17620	0.36768
C	0.66861	-1.17621	-0.36766
C	0.00000	1.06802	0.00004
H	-1.38137	-1.91738	0.00426
H	-0.50631	-1.32583	1.44266
H	0.50629	-1.32584	-1.44264
H	1.38134	-1.91739	-0.00424
H	0.10292	1.70493	0.88437
H	-0.10290	1.70501	-0.88423
N	-1.15652	0.17417	0.09524
N	1.15652	0.17416	-0.09523
B	-2.47407	0.55123	-0.13124
H	-3.33160	-0.27656	-0.05482
H	-2.70340	1.69271	-0.40167
B	2.47408	0.55122	0.13117
H	2.70343	1.69270	0.40159
H	3.33161	-0.27657	0.05468

Table S31: Calculated coordinates at M06-2X/cc-pVQZ level for the **DDIMBB**

Atom	**x**	**y**	**z**
C	-0.66892	-1.17515	0.36240
C	0.66890	-1.17516	-0.36238
C	0.00000	1.06666	0.00005
H	-1.37576	-1.91251	-0.00452
H	-0.51323	-1.32666	1.43279
H	0.51321	-1.32668	-1.43277
H	1.37573	-1.91252	0.00454
H	0.09865	1.70011	0.88073
H	-0.09863	1.70020	-0.88057
N	-1.15562	0.17409	0.09187
N	1.15562	0.17408	-0.09187
B	-2.46825	0.55104	-0.12945
H	-3.32305	-0.27215	-0.05377
H	-2.69714	1.68840	-0.39462
B	2.46827	0.55103	0.12937
H	2.69718	1.68838	0.39453
H	3.32305	-0.27216	0.05363

Bond Dissociation Energies and Thermochemical Study of the Dehydrogenation of IMBB

Table S32: Free energies (*E*+*G*) and enthalpies (*E*+*G*) at the CBS-QB3 level for all the molecules involved in the calculation of the thermochemical properties for the removal of 1st and 2nd hydrogen from **IMBB** at 298.15 K. All the energies are in Hartrees.

Molecule	***E+H***	***E+G***
IMBB	-281.331169	-281.372897
MDIMBB	-280.175157	-280.216134
DDIMBB	-279.017891	-279.058145
H_2	-1.162778	-1.177574

Table S33: Energies [at CCSD(T)/CBS] and corrections for enthalpy (*H*), Gibbs free energy (*G*) and Zero Point Energy (ZPE) [at the MP2/cc-pVTZ level] for all the molecules involved in the calculation of the $(B\text{-}N)_{BDES}$ of **IMBB** at 298.15 K. All the energies are in Hartrees.

Molecule	E_{CBS}	***H***	***G***	***ZPE***
IMBB	-281.55300044	0.197972	0.156862	0.188944
IMB	-254.94514563	0.161751	0.124153	0.154387
IM	-228.3338855	0.124487	0.092557	0.119458
BH_3	-26.54900366	0.030623	0.009262	0.026799

Table S34: Energies [at CCSD(T)/CBS] and corrections for enthalpy (*H*), Gibbs free energy (*G*) and Zero Point Energy (ZPE) [at the MP2/cc-pVTZ level] for all the molecules involved in the calculation of the thermochemical properties for the removal of 1st and 2nd hydrogen from **IMBB** at 298.15 K. All the energies are in Hartrees.

Molecule	E_{CBS}	***H***	***G***	***ZPE***
IMBB	-281.55300044	0.197972	0.156862	0.188944
MDIMBB	-280.3743475	0.175029	0.134151	0.166353
DDIMBB	-279.19413684	0.152076	0.110785	0.14394
H_2	-1.1744711	0.013617	-0.00116	0.010313

Table S35: Free energies (*E*+*G*) and enthalpies (*E*+*G*) at the CBS-QB3 level for all the molecules involved in the calculation of the thermochemical properties for the removal of 1st and 2nd hydrogen from **IMBB** at 653 K. All the energies are in Hartrees

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Molecule	***E+H***	***E+G***
IMBB	-281.301223	-281.433932
MDIMBB	-280.147530	-280.275441
DDIMBB	-278.992632	-279.115729
H_2	-1.158844	-1.196924

Table S36: Free energies (*E*+*G*) and enthalpies (*E*+*G*) at the CBS-QB3 level for all the molecules involved in the calculation of the thermochemical properties for the removal of 1st and 2nd hydrogen from **IMBB** at 823 K. All the energies are in Hartrees.

Molecule	***E+H***	***E+G***
IMBB	-281.281019	-281.470846
MDIMBB	-280.129056	-280.310904
DDIMBB	-278.975896	-279.149738
H_2	-1.156952	-1.207065

Reaction Energy Pathway for Hydrogen Release from IMBB

Table S37: Calculated coordinates at the CBS-QB3 level for TS1 of uncatalyzed dehydrogenation.

Atom	x	y	z
C	-0.73302	1.26483	-0.38522
C	0.64939	1.23193	0.32169
C	0.05272	-0.90471	-0.53871
H	-1.45882	1.88952	0.13276
H	-0.62197	1.63539	-1.40888
H	0.61294	1.61721	1.33963
H	1.41498	1.77047	-0.23274
H	0.46188	-0.83791	-1.54966
H	-0.06454	-1.94742	-0.25453
N	-1.16813	-0.13945	-0.39104
H	-2.42806	-0.58386	-0.74693
N	1.03944	-0.22161	0.36195
B	-2.28969	-0.63242	0.63352
H	-2.77626	0.24247	1.28159
H	-3.24250	-0.91787	-0.28490
H	-2.05328	-1.68862	1.13685
H	0.86698	-0.56389	1.30478
B	2.61240	-0.53686	0.01718
H	2.78422	-0.10031	-1.09976
H	3.25361	0.04671	0.86021
H	2.72350	-1.74028	0.08517

Table S38: Calculated coordinates at the CBS-QB3 level for TS2 of uncatalyzed dehydrogenation.

Atom	x	y	z
C	0.90369	1.18061	0.31520
C	-0.53160	1.27647	-0.23705
C	0.04076	-0.98055	0.34283
H	1.61227	1.83888	-0.18466
H	0.91059	1.40818	1.38789
H	-0.53393	1.64461	-1.26894
H	-1.15584	1.94583	0.36217
H	-0.09861	-1.16285	1.42069
H	0.07515	-1.94398	-0.16831
N	1.26177	-0.22417	0.09901
N	-1.02212	-0.11235	-0.17163
H	-1.91314	-0.59806	-1.13904
B	-2.52745	-0.46112	0.09367
H	-3.23602	0.48550	0.25934
H	-2.85998	-0.87132	-1.18009
H	-2.69158	-1.45267	0.73978
B	2.48009	-0.72512	-0.33592
H	3.38050	0.03308	-0.52604
H	2.59270	-1.89953	-0.50912

Table S39: Calculated coordinates at the CBS-QB3 level for reactant of uncatalyzed dehydrogenation.

Atom	x	y	z
C	0.52100	1.22481	0.36271
C	-0.84692	1.25921	-0.34919
C	0.10282	-0.86556	-0.66989
H	1.27014	1.81382	-0.16192
H	0.46812	1.55422	1.39757
H	-0.76501	1.72552	-1.33318
H	-1.62496	1.75523	0.22563
H	0.01564	-1.93103	-0.47915
H	0.52695	-0.67686	-1.65751
N	0.96089	-0.21177	0.33287
H	0.71988	-0.62217	1.23392
N	-1.21192	-0.18750	-0.50542
B	2.57309	-0.43772	0.07390
H	3.12366	0.12390	0.98963
H	2.73405	-1.63524	0.07675
H	2.78283	0.07680	-1.00305
H	-1.77492	-0.31787	-1.34282
B	-2.09247	-0.78212	0.76098
H	-1.41221	-0.60724	1.74919
H	-3.10560	-0.12735	0.77452
H	-2.26592	-1.94838	0.50206

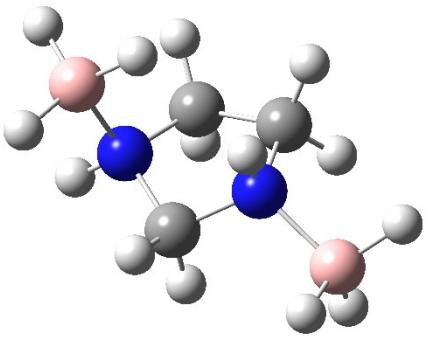

Figure S7: The structure of the reactant for the uncatalyzed dehydrogenation of **IMBB** at CBS-QB3 level.

Table S40: Calculated coordinates at the CBS-QB3 level for products 1 of uncatalyzed dehydrogenation.

Atom	x	y	z
C	0.90136	1.22311	0.18730
C	-0.62529	1.25969	-0.07795
C	0.03185	-0.95529	0.33482
H	1.47675	1.79221	-0.54197
H	1.12505	1.61697	1.18278
H	-0.91326	1.88289	-0.92249
H	-1.18789	1.58032	0.79727
H	-0.24206	-0.98383	1.39354
H	0.08239	-1.96647	-0.05993
N	1.25371	-0.20614	0.09679
N	-1.01912	-0.16024	-0.35411
H	-0.91094	-0.32609	-1.35355
B	-2.56519	-0.54710	0.03578
H	-2.65114	-0.34784	1.22697
H	-3.25663	0.19423	-0.62334
H	-2.69221	-1.71083	-0.26946
B	2.49748	-0.73676	-0.22819
H	3.40422	0.00753	-0.43410
H	2.61456	-1.92019	-0.29746
H	0.00000	0.00000	0.37209
H	0.00000	0.00000	-0.37209

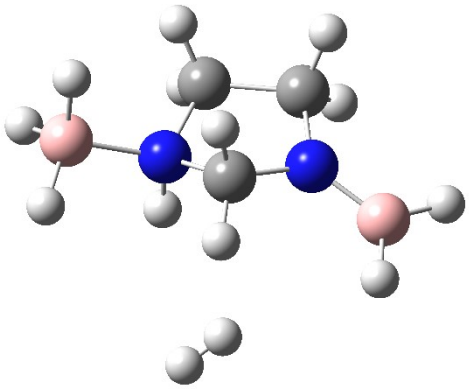

Figure S8: The structure of product 1 for the uncatalyzed dehydrogenation of **IMBB** at CBS-QB3 level.

Table S41: Calculated coordinates at the CBS-QB3 level for products 2 of uncatalyzed dehydrogenation.

Atom	x	y	z
C	-0.00001	-1.07218	0.00013
C	0.67692	1.18348	0.35373
C	-0.67687	1.18350	-0.35368
H	0.09167	-1.70876	-0.88365
H	-0.09171	-1.70851	0.88409
H	0.54140	1.34468	1.42982
H	1.37957	1.92215	-0.02932
H	-1.37951	1.92218	0.02938
H	-0.54135	1.34472	-1.42977
N	-1.16501	-0.17610	-0.08535
N	1.16501	-0.17612	0.08537
B	2.48234	-0.55599	-0.12665
H	3.34160	0.26705	-0.05309
H	2.71931	-1.69702	-0.38178
B	-2.48238	-0.55596	0.12643
H	-3.34162	0.26707	0.05270
H	-2.71939	-1.69699	0.38153
H	0.00000	0.00000	0.37209
H	0.00000	0.00000	-0.37209
H	0.00000	0.00000	0.37209
H	0.00000	0.00000	-0.37209

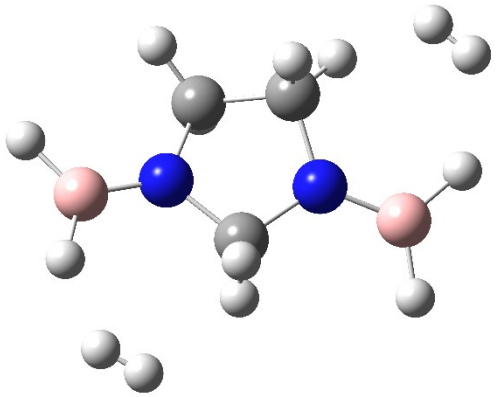

Figure S9: The structure of products 2 for the uncatalyzed dehydrogenation of **IMBB** at CBS-QB3 level.

Table S42: Calculated coordinates at the CBS-QB3 level for the first condensation product of catalysed dehydrogenation.

Atom	x	y	z
C	-0.48291	-1.36903	-0.57642
C	-1.72513	-0.97158	0.23181
C	-0.04428	0.55437	0.79620
H	-0.10383	-2.34905	-0.29621
H	-0.66171	-1.34009	-1.64809
H	-1.77967	-1.53048	1.16912
H	-2.65327	-1.08822	-0.32227
H	0.32350	1.57093	0.69732
H	0.19330	0.14353	1.77859
N	0.55916	-0.32645	-0.23770
H	0.71619	0.25448	-1.06295
N	-1.50128	0.47789	0.52066
B	1.97740	-0.96759	0.16575
H	2.40032	-1.59169	-0.75706
H	2.75065	-0.04773	0.56319
H	1.81558	-1.57795	1.19151
H	-2.02044	0.75610	1.35076
B	-1.99416	1.48035	-0.70409
H	-1.35347	1.15267	-1.67731
H	-3.17502	1.26233	-0.82141
H	-1.73475	2.59169	-0.31516
B	3.15309	1.03279	-0.19165
H	4.32630	0.80614	-0.22480
H	2.597575	1.048182	-1.25901
H	2.785907	1.868804	0.593445

Table S43: Calculated coordinates at the CBS-QB3 level for TS1 of catalysed dehydrogenation.

Atom	x	y	z
C	0.23831	1.35193	-0.18041
C	-1.22730	1.21806	0.24390
C	-0.41888	-0.95413	-0.00955
H	0.33869	1.82219	-1.16144
H	0.81968	1.93962	0.53352
H	-1.88726	1.97720	-0.17123
H	-1.34789	1.20552	1.32770
H	-0.46863	-1.29504	1.02822
H	-0.40493	-1.81669	-0.67129
N	0.72954	-0.05515	-0.24491
H	1.69392	-0.26018	0.90009
N	-1.63064	-0.13158	-0.24452
B	1.81802	-0.42964	-1.18444
H	2.31924	0.44019	-1.81978
H	3.09766	-0.74241	-0.22187
H	1.81835	-1.55602	-1.57234
H	-1.78013	-0.07320	-1.25155
B	-3.01320	-0.73888	0.40846
H	-2.80575	-0.77815	1.60010
H	-3.88042	0.04205	0.09470
H	-3.14963	-1.83013	-0.09369
B	3.32156	-0.23374	0.89324
H	3.46525	0.94918	0.86891
H	2.20639	-0.50653	1.59794
H	4.08844	-0.89445	1.52806

Table S44: Calculated coordinates at the CBS-QB3 level for the second condensation product of catalysed dehydrogenation.

Atom	x	y	z
C	1.74749	0.95444	-0.11415
C	0.32040	1.52108	-0.32266
C	0.21106	-0.62501	0.67478
H	2.39579	1.10447	-0.97583
H	2.21645	1.42334	0.75616
H	0.14986	1.92793	-1.31759
H	0.06840	2.28335	0.41280
H	0.16112	-0.30776	1.72071
H	-0.18508	-1.63034	0.56682
N	1.53991	-0.48292	0.11673
N	-0.60483	0.35278	-0.11207
H	-0.79867	-0.08333	-1.01630
B	-2.03127	0.76020	0.48610
H	-1.89506	1.01766	1.65287
H	-2.51443	1.61698	-0.18899
H	-2.76833	-0.27498	0.55354
B	2.41718	-1.51634	-0.20434
H	3.46718	-1.24190	-0.69348
H	2.08563	-2.63710	0.01844
B	-3.04207	-1.13431	-0.48125
H	-2.62652	-0.70013	-1.52530
H	-4.23341	-1.13913	-0.37361
H	-2.46140	-2.09891	-0.05325

Table S45: Calculated coordinates at the CBS-QB3 level for TS2 of catalysed dehydrogenation.

Atom	x	y	z
C	1.18916	1.23832	-0.21050
C	-0.04082	1.13894	0.68933
C	0.47887	-1.00958	-0.27237
H	0.90622	1.58288	-1.21305
H	1.96583	1.89418	0.17843
H	-0.77509	1.92671	0.51448
H	0.24491	1.15112	1.74480
H	0.65608	-1.91838	0.30123
H	0.20410	-1.29102	-1.29824
N	1.66060	-0.14950	-0.25351
N	-0.60948	-0.18310	0.31691
H	-1.54953	0.06362	-0.80011
B	-1.69330	-0.80091	1.09750
H	-2.19701	-0.15893	1.95919
H	-3.05921	-0.69044	-0.00667
H	-1.78001	-1.98702	1.06532
B	-3.18916	0.16469	-0.89186
H	-3.92794	-0.18314	-1.76693
H	-3.30049	1.27970	-0.47665
H	-2.03626	0.09617	-1.57403
B	2.97840	-0.58584	-0.19828
H	3.85096	0.22442	-0.16180
H	3.19663	-1.75745	-0.18532

Table S46: Calculated coordinates at the CBS-QB3 level for reactants of catalysed dehydrogenation.

Atom	x	y	z
C	0.52100	1.22481	0.36271
C	-0.84692	1.25921	-0.34919
C	0.10282	-0.86556	-0.66989
H	1.27014	1.81382	-0.16192
H	0.46812	1.55422	1.39757
H	-0.76501	1.72552	-1.33318
H	-1.62496	1.75523	0.22563
H	0.01564	-1.93103	-0.47915
H	0.52695	-0.67686	-1.65751
N	0.96089	-0.21177	0.33287
H	0.71988	-0.62217	1.23392
N	-1.21192	-0.18750	-0.50542
B	2.57309	-0.43772	0.07390
H	3.12366	0.12390	0.98963
H	2.73405	-1.63524	0.07675
H	2.78283	0.07680	-1.00305
H	-1.77492	-0.31787	-1.34282
B	-2.09247	-0.78212	0.76098
H	-1.41221	-0.60724	1.74919
H	-3.10560	-0.12735	0.77452
H	-2.26592	-1.94838	0.50206
B	0.00000	0.00000	0.00000
H	0.00000	1.18965	0.00000
H	1.03027	-0.59482	0.00000
H	-1.03027	-0.59482	0.00000

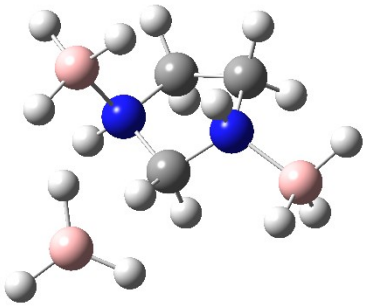

Figure S10: The structure of the reactants for the catalyzed dehydrogenation of **IMBB** at CBS-QB3 level.

Table S47: Calculated coordinates at the CBS-QB3 level for products 1 of catalysed dehydrogenation.

Atom	x	y	z
C	0.90136	1.22311	0.18730
C	-0.62529	1.25969	-0.07795
C	0.03185	-0.95529	0.33482
H	1.47675	1.79221	-0.54197
H	1.12505	1.61697	1.18278
H	-0.91326	1.88289	-0.92249
H	-1.18789	1.58032	0.79727
H	-0.24206	-0.98383	1.39354
H	0.08239	-1.96647	-0.05993
N	1.25371	-0.20614	0.09679
N	-1.01912	-0.16024	-0.35411
H	-0.91094	-0.32609	-1.35355
B	-2.56519	-0.54710	0.03578
H	-2.65114	-0.34784	1.22697
H	-3.25663	0.19423	-0.62334
H	-2.69221	-1.71083	-0.26946
B	2.49748	-0.73676	-0.22819
H	3.40422	0.00753	-0.43410
H	2.61456	-1.92019	-0.29746
B	0.00000	0.00000	0.00000
H	0.00000	1.18965	0.00000
H	1.03027	-0.59482	0.00000
H	-1.03027	-0.59482	0.00000
H	0.00000	0.00000	0.37209
H	0.00000	0.00000	-0.37209

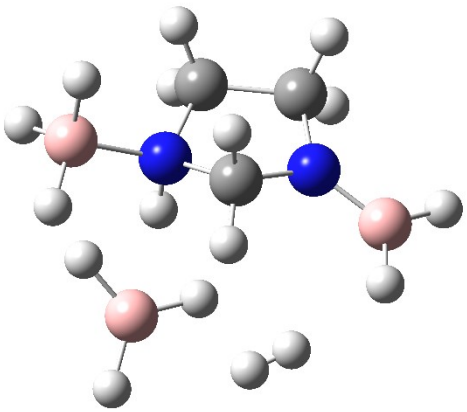

Figure S11: The structure of products 1 for the catalyzed dehydrogenation of **IMBB** at CBS-QB3 level.

Table S48: Calculated coordinates at the CBS-QB3 level for products 2 of catalysed dehydrogenation.

Atom	x	y	z
C	-0.00001	-1.07218	0.00013
C	0.67692	1.18348	0.35373
C	-0.67687	1.18350	-0.35368
H	0.09167	-1.70876	-0.88365
H	-0.09171	-1.70851	0.88409
H	0.54140	1.34468	1.42982
H	1.37957	1.92215	-0.02932
H	-1.37951	1.92218	0.02938
H	-0.54135	1.34472	-1.42977
N	-1.16501	-0.17610	-0.08535
N	1.16501	-0.17612	0.08537
B	2.48234	-0.55599	-0.12665
H	3.34160	0.26705	-0.05309
H	2.71931	-1.69702	-0.38178
B	-2.48238	-0.55596	0.12643
H	-3.34162	0.26707	0.05270
H	-2.71939	-1.69699	0.38153
B	0.00000	0.00000	0.00000
H	0.00000	1.18965	0.00000
H	1.03027	-0.59482	0.00000
H	-1.03027	-0.59482	0.00000
H	0.00000	0.00000	0.37209
H	0.00000	0.00000	-0.37209
H	0.00000	0.00000	0.37209
H	0.00000	0.00000	-0.37209

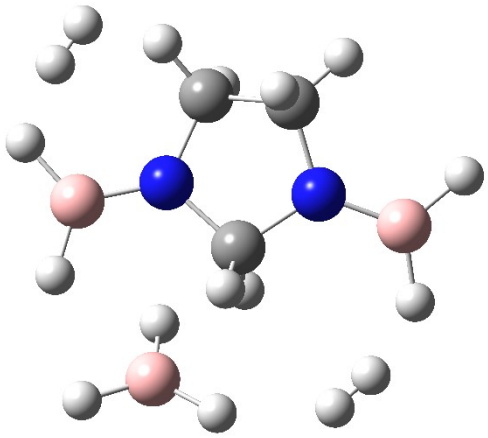

Figure S12: The structure of product 2 for the catalyzed dehydrogenation of **IMBB** at CBS-QB3 level.

Table S49: Free energies (*E*+*G*) and enthalpies (*E*+*G*) at the CBS-QB3 level for reactants and activated complexes involved in the calculation of the activated thermochemical properties of uncatalyzed and catalyzed hydrogens release from **IMBB** at 298.15K. All the energies are in Hartrees.

Molecule	***E+H***	***E+G***
IMBB	-281.331168	-281.372901
TS1(uncatalyzed)	-281.269624	-281.312315
MDIMBB	-280.175157	-280.216135
TS2(uncatalyzed)	-280.113151	-280.154119
BH_3	-26.518657	-26.540036
TS1(catalyzed)	-307.837067	-307.884235
TS2(catalyzed)	-306.681662	-306.727704

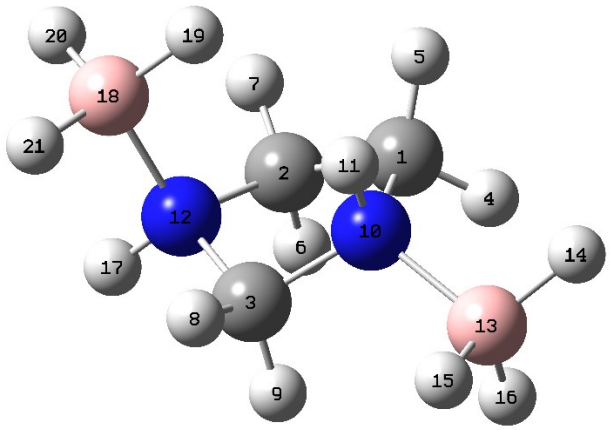

Figure S13: The optimized structure of **IMBB** at M06-2X/cc-pVQZ level with atom numbering

Table S50: The detailed optimized structural parameters for the **IMBB** at different levels of theories and basis sets. All distances are in pm; bond angles are in degrees.

Parameter	**MP2/6-31G***	**MP2/6-311G***	**MP2/6-311+G***	**M06-2X/6-31G***	**M06-2X/cc-pVQZ**
*r*B(18)-N(12)	165.0	164.1	164.1	164.9	163.5
*r*B(13)-N(10)	164.9	164.0	164.0	164.5	163.1
*r*N(12)-C(2)	149.1	149.0	149.1	148.9	148.5
*r*N(12)-C(3)	148.2	148.1	148.1	148.0	147.6
*r*N(10)-C(1)	149.9	150.0	150.0	149.5	149.3
*r*N(10)-C(3)	147.0	147.2	147.1	146.7	146.6
*r*B(18)-H(19)	121.4	121.5	121.7	121.6	121.0
*r*B(18)-H(20)	120.9	121.1	121.2	120.8	120.3
*r*B(18)-H(21)	121.0	121.2	121.3	120.9	120.4
*r*B(13)-H(14)	120.9	121.1	121.3	120.8	120.4
*r*B(13)-H(15)	121.1	121.3	121.4	121.0	120.6
*r*B(13)-H(16)	121.4	121.6	121.8	121.5	121.0
∠C(3)-N(12)-B(18)	113.0	113.0	113.0	112.6	112.8
∠C(2)-N(12)-B(18)	112.8	113.1	113.4	112.2	112.7
∠C(3)-N(10)-B(13)	112.8	112.2	112.4	112.5	112.4
∠C(1)-N(10)-B(13)	114.5	114.2	114.4	114.3	114.3
∠N(12)-B(18)-H(19)	106.0	106.3	105.9	105.4	105.5
∠N(12)-B(18)-H(20)	104.5	105.1	105.2	104.7	105.2
∠N(12)-B(18)-H(21)	104.7	105.0	105.1	104.9	105.2
∠N(10)-B(13)-H(14)	104.9	105.4	105.4	105.0	105.5
∠N(10)-B(13)-H(15)	104.9	105.4	105.5	105.0	105.5
∠N(10)-B(13)-H(16)	103.9	104.3	104.1	103.6	104.0

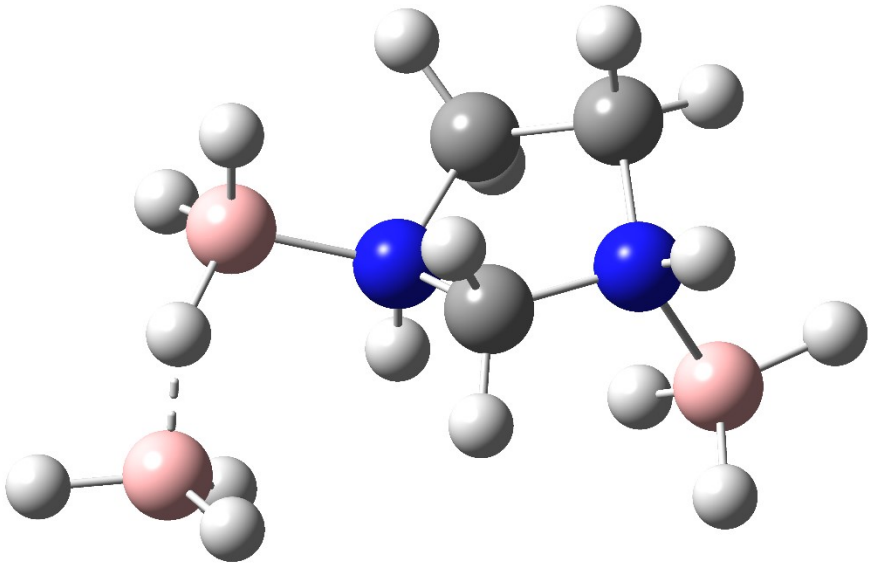

Figure S14: The optimized structure of the first condensation product formed between **IMBB** and BH_3 catalyst

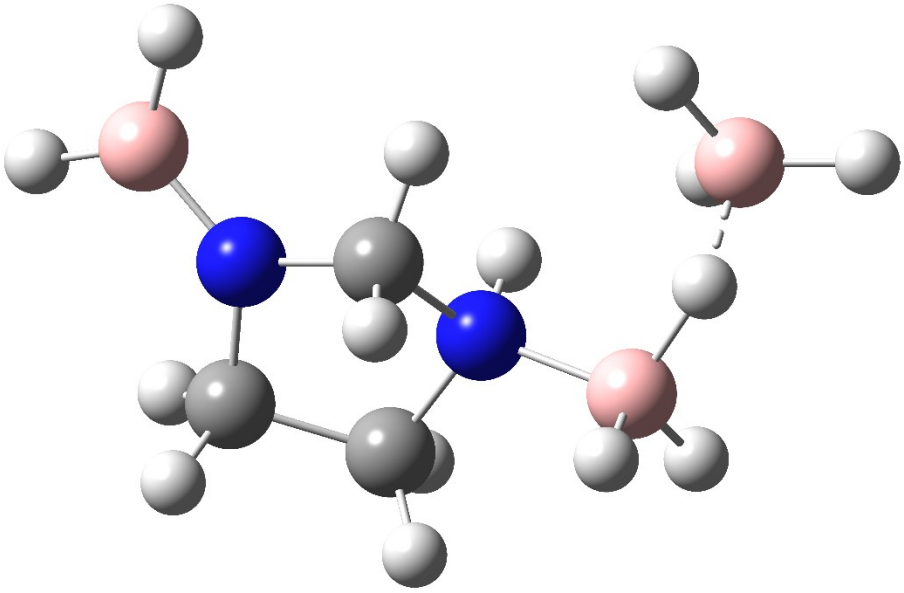

Figure S15: The optimized structure of the second condensation product formed between **MDIMBB** and BH_3 catalyst