Supporting Information for Local Vibrational Mode Analysis of Ion-Solvent and Solvent-Solvent Interactions for Hydrated Mg^{2+} Clusters

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Atomic Cartesian coordinates (in Å) of hydrated divalent magnesium clusters **1**-**17** using ωB97X-D/6-311++G(d,p) level of theory.

$[Mg(H_2O)]^{2+}$, **1**

O	0.00000	0.00000	-1.01353
H	0.00000	0.77563	-1.60341
H	0.00000	-0.77563	-1.60341
Mg	0.00000	0.00000	0.94292

$[Mg(H_2O)_2]^{2+}$, **2**

O	0.00000	0.00000	-1.97219
H	0.54551	0.54902	-2.56017
H	-0.54551	-0.54902	-2.56017
O	0.00000	0.00000	1.97219
H	0.54902	-0.54551	2.56017
H	-0.54902	0.54551	2.56017
Mg	0.00000	0.00000	0.00000

$[Mg(H_2O)_3]^{2+}$, **3**

O	1.82696	0.80451	-0.00015
H	2.12017	1.59129	0.48370
H	2.60440	0.48834	-0.48464
O	-1.61021	1.17995	-0.00015
H	-2.43818	1.04048	0.48370
H	-1.72511	2.01131	-0.48464
O	-0.21676	-1.98445	-0.00015
H	-0.87929	-2.49965	-0.48464
H	0.31801	-2.63177	0.48370
Mg	0.00000	0.00000	0.00054

$[Mg(H_2O)_4]^{2+}$, **4**

O	1.82696	0.80451	-0.00015
H	2.12017	1.59129	0.48370
H	2.60440	0.48834	-0.48464
O	-1.61021	1.17995	-0.00015
H	-2.43818	1.04048	0.48370
H	-1.72511	2.01131	-0.48464
O	-0.21676	-1.98445	-0.00015
H	-0.87929	-2.49965	-0.48464
H	0.31801	-2.63177	0.48370
Mg	0.00000	0.00000	0.00054

$[Mg(H_2O)_5]^{2+}$, **5**

O	0.00000	2.08555	0.32924
H	0.00000	2.72391	-0.39362
H	0.00000	2.60175	1.14396
O	-1.98116	0.00000	0.62902
H	-2.53550	0.77375	0.78173
H	-2.53550	-0.77375	0.78173
O	0.00000	-2.08555	0.32924
H	0.00000	-2.60175	1.14396
H	0.00000	-2.72391	-0.39362
O	1.98116	0.00000	0.62902
H	2.53550	0.77375	0.78173
H	2.53550	-0.77375	0.78173
O	0.00000	0.00000	-1.97799
H	0.77262	0.00000	-2.55563
H	-0.77262	0.00000	-2.55563
Mg	0.00000	0.00000	0.08129

$[Mg(H_2O)_6]^{2+}$, **6**

O	1.82696	0.80451	-0.00015
H	2.12017	1.59129	0.48370
H	2.60440	0.48834	-0.48464
O	-1.61021	1.17995	-0.00015
H	-2.43818	1.04048	0.48370
H	-1.72511	2.01131	-0.48464
O	-0.21676	-1.98445	-0.00015
H	-0.87929	-2.49965	-0.48464
H	0.31801	-2.63177	0.48370
Mg	0.00000	0.00000	0.00054

$[Mg(H_2O)_5\text{-}(H_2O)]^{2+}$, **7**

O	1.08122	-1.31585	0.00531
H	2.00140	-0.97836	0.00395
H	1.11562	-2.27685	0.00942
O	3.30998	0.26807	-0.00091
H	3.89151	0.32500	0.76660
H	3.89130	0.31929	-0.76899
O	0.85131	1.45339	-0.00503
H	1.81540	1.27823	-0.00445
H	0.71719	2.40544	-0.00823
O	-2.10813	1.36172	-0.00545
H	-2.49313	1.79073	-0.77773
H	-2.49446	1.79499	0.76378
O	-1.31366	-0.85950	1.72910
H	-0.78878	-1.28265	2.41774
H	-2.23637	-0.96672	1.98452
Mg	-0.53878	-0.03422	0.00010
O	-1.31230	-0.87242	-1.72314

H	-2.23476	-0.97960	-1.97949
H	-0.78703	-1.30223	-2.40735

$[Mg(H_2O)_5\text{-}2(H_2O)]^{2+}$, **8**

Mg	0.00006	0.31220	0.00014
O	-1.65891	0.31928	-1.24938
H	-2.49778	-0.10104	-0.97032
H	-1.77746	0.65472	-2.14216
O	1.65893	0.31865	1.24953
H	2.49779	-0.10156	0.97025
H	1.77764	0.65405	2.14231
O	-1.18861	-0.68139	1.32925
H	-2.12475	-0.89314	1.13873
H	-0.95414	-1.08025	2.17138
O	0.00047	2.37780	-0.00015
H	0.72711	2.95029	0.26573
H	-0.72605	2.95040	-0.26617
O	1.18807	-0.68206	-1.32905
H	0.95326	-1.08118	-2.17095
H	2.12429	-0.89375	-1.13884
O	3.64097	-0.96208	-0.14688
H	4.41373	-0.48376	-0.46921
H	3.96599	-1.83086	0.11655
O	-3.64101	-0.96211	0.14657
H	-3.96562	-1.83104	-0.11687
H	-4.41404	-0.48408	0.46869

$[Mg(H_2O)_5\text{-}3(H_2O)]^{2+}$, **9**

Mg	0.00127	0.31220	0.00136
O	-1.65768	0.33060	-1.24807

H	-2.49817	-0.08754	-0.97062
H	-1.77494	0.66998	-2.13954
O	1.66018	0.30732	1.25071
H	2.49740	-0.11506	0.96976
H	1.78021	0.63877	2.14479
O	-1.19123	-0.68194	1.32662
H	-2.12819	-0.88929	1.13531
H	-0.95830	-1.08498	2.16718
O	0.00972	2.37776	0.00912
H	0.73859	2.94638	0.27721
H	-0.71456	2.95422	-0.25464
O	1.18539	-0.68149	-1.33173
H	0.94900	-1.07642	-2.17518
H	2.12077	-0.89756	-1.14239
O	3.63720	-0.97566	-0.15075
H	4.41181	-0.49911	-0.47123
H	3.95884	-1.84672	0.10928
O	-3.64473	-0.94849	0.14294
H	-3.97271	-1.81512	-0.12388
H	-4.41588	-0.46872	0.46695

$[Mg(H_2O)_5\text{-}4(H_2O)]^{2+}$, **10**

Mg	-0.19561	0.00845	-0.25076
O	-1.94959	0.18892	-1.35629
H	-2.12247	0.13250	-2.29956
H	-2.79718	0.36453	-0.90116
O	1.59995	-0.18863	0.73984
H	2.27735	0.50947	0.74167
H	2.02991	-1.04989	0.87971
O	-0.00603	-1.97305	-0.72753
H	-0.61689	-2.55188	-1.18985

H	0.73267	-2.51268	-0.38026
O	-1.34025	0.33908	1.40982
H	-1.05708	0.37550	2.32685
H	-2.30385	0.48884	1.37071
O	0.58164	1.72192	-1.05698
H	0.16639	2.37159	-1.62922
H	1.45926	2.05998	-0.78692
O	-3.91787	0.66831	0.50378
H	-4.36004	1.52328	0.55466
H	-4.60032	0.01339	0.68933
O	3.01057	2.14781	0.13255
H	3.12682	2.85915	0.77208
H	3.82913	2.12208	-0.37512
O	2.20738	-2.91358	0.58028
H	2.11784	-3.50561	1.33526
H	2.97950	-3.22791	0.09709

$[Mg(H_2O)_5\text{-}5(H_2O)]^{2+}$, **11**

O	0.43765	0.00277	1.98757
H	-0.19266	-0.01185	2.71100
H	1.34063	0.01629	2.39050
Mg	-0.13990	-0.00144	0.04564
O	-2.15768	-0.05312	0.47733
H	-2.68822	-0.86292	0.39108
H	-2.72747	0.73019	0.39723
O	1.68468	0.04705	-0.88980
H	2.06318	0.85569	-1.27209
H	2.10199	-0.73862	-1.27952
O	-0.41908	1.91861	-0.59367
H	-1.24481	2.41936	-0.49018
H	0.26666	2.45744	-1.02147

O	-0.32456	-1.92761	-0.61102
H	0.38632	-2.42834	-1.04403
H	-1.12468	-2.46904	-0.51175
O	-3.06120	2.55374	0.01304
H	-3.29121	3.13065	0.74876
H	-3.69408	2.76054	-0.68239
O	1.98992	2.69212	-1.75336
H	2.59615	3.29699	-1.31366
H	2.04132	2.91255	-2.68912
O	2.11822	-2.57174	-1.77857
H	2.75368	-3.15042	-1.34492
H	2.17950	-2.78049	-2.71642
O	-2.93312	-2.69632	-0.00911
H	-3.55560	-2.92734	-0.70631
H	-3.13449	-3.29027	0.72142
O	2.81209	0.03619	3.16002
H	3.22391	-0.72677	3.57267
H	3.20346	0.81013	3.57210

$[Mg(H_2O)_5\text{-}12(H_2O)]^{2+}$, **12**

Mg	-0.03857	-0.02635	-0.18159
O	-1.53434	-1.05634	0.77983
O	1.36918	-1.15071	0.80542
O	-1.43650	1.39729	-0.59605
O	1.46694	1.28605	-0.57891
O	-0.07186	-1.17614	-1.85624
H	-2.34178	-0.68576	1.18248
H	-1.59537	-2.02650	0.76338
H	1.39364	-2.12060	0.77314
H	2.20087	-0.81801	1.19931
H	-1.43591	2.35664	-0.41373

H	-2.31339	1.12482	-0.92082
H	2.33007	0.96572	-0.89570
H	1.50605	2.25236	-0.43646
H	-0.87999	-1.58850	-2.20771
H	0.72383	-1.61045	-2.21193
O	-3.85219	0.22674	1.45916
O	-1.46909	-3.70674	0.04040
O	2.96767	2.47350	2.21078
O	-2.35758	2.46038	2.44766
O	1.31045	-3.76799	-0.09927
O	3.71833	-0.00396	1.55522
H	-4.13035	0.24580	0.52590
H	-4.59949	-0.09058	1.97278
H	-0.51002	-3.86261	-0.06963
H	-1.83022	-4.48541	0.47209
H	2.43412	3.05634	1.64773
H	3.31621	3.00748	2.92661
H	-2.95552	1.74727	2.17314
H	-2.58936	2.69747	3.34741
H	1.76711	-3.44812	-0.90146
H	1.78812	-4.54442	0.20477
H	3.49591	0.91902	1.83011
H	4.33151	-0.34955	2.20854
O	-1.32663	3.89549	0.47233
O	-3.77654	0.11249	-1.30201
O	3.79126	-0.10579	-1.15071
O	1.40926	3.84679	0.28858
O	-2.55571	-2.31195	-2.09883
O	2.40366	-2.29849	-2.17924
H	-1.73434	3.53037	1.29310
H	-1.83196	4.67353	0.22384
H	-3.45581	-0.76289	-1.59317

H	-4.41920	0.40350	-1.95429
H	4.02307	-0.12934	-0.19890
H	4.55961	0.22989	-1.62038
H	0.45694	4.02494	0.43039
H	1.78312	4.62827	-0.12616
H	-2.33993	-2.95160	-1.39309
H	-2.93934	-2.81314	-2.82370
H	2.98266	-1.58576	-1.83880
H	2.82126	-2.63911	-2.97483

$[Mg(H_2O)_6\text{-}H_2O]^{2+}$, **13**

Mg	0.45918	-0.01700	0.02694
O	1.98773	-0.12610	1.49384
H	1.95709	-0.17027	2.45431
H	2.91916	-0.12099	1.24983
O	0.63237	2.09891	0.06250
H	0.04517	2.70936	-0.39307
H	1.11335	2.61507	0.71639
O	-1.07888	0.10527	-1.37830
H	-1.02128	0.16482	-2.33535
H	-2.02296	0.11436	-1.12586
O	0.60800	-2.11748	-0.23717
H	1.11079	-2.72402	0.31482
H	-0.00915	-2.65343	-0.74419
O	-1.07850	-0.11143	1.44016
H	-1.08148	-0.26217	2.38809
H	-2.01020	-0.04382	1.15479
O	-3.44330	0.09057	0.02079
H	-4.05364	-0.65438	-0.02277
H	-4.00085	0.87443	0.08395
O	1.97795	0.07787	-1.44198

H	2.36052	0.88025	-1.81014
H	2.34027	-0.66610	-1.93283

$[Mg(H_2O)_6\text{-}2(H_2O)]^{2+}$, **14**

Mg	-0.00000	0.62040	-0.00060
O	2.08272	0.46502	-0.00206
H	2.52402	-0.40749	-0.00170
H	2.75683	1.14864	-0.00323
O	0.00163	0.85926	2.11881
H	-0.76994	0.77815	2.68646
H	0.77406	0.77802	2.68526
O	-2.08273	0.46500	0.00126
H	-2.75684	1.14861	0.00109
H	-2.52401	-0.40752	0.00257
O	-0.00164	0.85503	-2.12048
H	0.76993	0.77280	-2.68797
H	-0.77407	0.77265	-2.68676
O	0.00001	-1.45124	0.00140
H	0.79570	-2.00669	0.00118
H	-0.79568	-2.00669	0.00270
O	-2.69727	-2.21496	0.00468
H	-3.10364	-2.63632	-0.76047
H	-3.10222	-2.63468	0.77150
O	2.69729	-2.21494	-0.00039
H	3.10366	-2.63483	0.76558
H	3.10227	-2.63610	-0.76639
O	-0.00002	2.72863	-0.00273
H	-0.00065	3.29530	-0.77949
H	0.00060	3.29687	0.77288

$[Mg(H_2O)_6\text{-}3(H_2O)]^{2+}$, **15**

O	-0.35638	1.99973	0.54014
H	0.17797	2.52540	1.13919
H	-0.95123	2.59613	0.05132
O	1.57328	-0.05545	1.39283
H	2.51412	0.01553	1.15146
H	1.51342	-0.36676	2.29850
O	-1.37316	0.43083	-1.53180
H	-1.81736	1.29126	-1.63582
H	-1.82304	-0.21325	-2.08272
Mg	0.00032	0.00027	-0.00026
O	1.56377	0.26158	-1.37985
H	1.47081	0.56340	-2.28594
H	2.50397	0.31331	-1.13115
O	-0.08741	-2.02887	-0.54137
H	-0.60272	-2.69816	-0.05664
H	0.51535	-2.47991	-1.13631
O	-1.31897	-0.60681	1.51887
H	-1.85328	-0.02739	2.06613
H	-1.64678	-1.51819	1.62087
O	-2.18766	3.06938	-1.25000
H	-1.94170	3.73449	-1.90177
H	-3.07678	3.30559	-0.96477
O	3.96268	0.25920	0.01548
H	4.59443	-0.46019	-0.08972
H	4.49481	1.05451	0.12501
O	-1.77634	-3.32977	1.23580
H	-1.44997	-3.95618	1.89061
H	-2.62427	-3.68135	0.94411

$[Mg(H_2O)_6\text{-}4(H_2O)]^{2+}$, **16**

O	0.57066	1.75829	1.05995

H	-0.02629	2.38142	1.47976
H	1.46857	2.06580	1.20154
O	-0.15074	0.92303	-1.85223
H	0.62479	1.13681	-2.39437
H	-0.91599	1.46234	-2.10645
Mg	0.00058	0.00028	0.00008
O	2.04168	-0.25618	-0.27172
H	2.56479	-0.88080	0.25485
H	2.48982	-0.03734	-1.10379
O	-0.57107	-1.75772	-1.06034
H	-0.75773	-1.81385	-1.99994
H	-0.68491	-2.63232	-0.68222
O	0.04328	-1.25377	1.65273
H	-0.67178	-1.27956	2.30779
H	0.86913	-1.60529	2.02081
O	-1.93371	0.58710	0.47154
H	-2.51779	1.02352	-0.16835
H	-2.44270	0.17968	1.18988
O	2.49245	0.88063	-2.79927
H	2.69102	0.36723	-3.58906
H	3.05140	1.66204	-2.86026
O	2.74779	-1.98900	1.82194
H	3.34758	-1.66849	2.50333
H	2.98305	-2.91377	1.69463
O	-2.49286	-0.88131	2.79864
H	-2.64371	-0.40296	3.62034
H	-3.09994	-1.62791	2.82724
O	-2.74807	1.98876	-1.82128
H	-3.39171	1.65267	-2.45337
H	-2.93983	2.92890	-1.74297

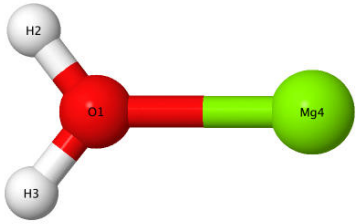
$[Mg(H_2O)_6\text{-}12(H_2O)]^{2+}$, **17**

Mg	-0.00000	0.00000	-0.00001
O	-1.05205	1.39169	-1.14798
O	-0.68354	-1.60586	-1.14556
O	1.72727	0.21183	-1.15420
O	-1.72728	-0.21183	1.15417
O	1.05204	-1.39168	1.14798
O	0.68355	1.60587	1.14551
H	-1.98927	1.33219	-1.39035
H	-0.69698	2.25442	-1.41696
H	-0.16353	-2.38738	-1.38951
H	-1.60878	-1.72998	-1.41258
H	2.14553	1.05308	-1.39522
H	2.29849	-0.52710	-1.41963
H	-2.14552	-1.05308	1.39522
H	-2.29850	0.52711	1.41961
H	1.98926	-1.33218	1.39034
H	0.69697	-2.25442	1.41694
H	0.16353	2.38737	1.38950
H	1.60878	1.72998	1.41257
O	-3.80951	1.04225	-1.14117
O	0.23779	3.83730	-1.31975
O	0.99907	-3.81833	-1.14337
O	-3.44625	-1.71281	-1.31258
O	2.80602	2.77391	-1.14929
O	3.20384	-2.12654	-1.31905
H	-3.81516	1.46844	-0.26185
H	-4.48657	1.47449	-1.66804
H	1.17670	3.57184	-1.24871
H	0.18888	4.50806	-2.00547
H	0.63489	-4.03658	-0.26327
H	0.96234	-4.62076	-1.67024
H	-3.68637	-0.76707	-1.24157

H	-4.00337	-2.09125	-1.99737
H	3.17936	2.56782	-0.27011
H	3.51803	3.14293	-1.67816
H	2.50456	-2.80682	-1.24607
H	3.80822	-2.42052	-2.00522
O	-2.80600	-2.77392	1.14930
O	-3.20385	2.12654	1.31906
O	3.80949	-1.04226	1.14118
O	-0.23778	-3.83731	1.31977
O	-0.99907	3.81833	1.14339
O	3.44626	1.71280	1.31260
H	-3.17933	-2.56783	0.27011
H	-3.51802	-3.14293	1.67816
H	-2.50456	2.80682	1.24608
H	-3.80822	2.42051	2.00523
H	3.81515	-1.46844	0.26186
H	4.48655	-1.47450	1.66806
H	-1.17669	-3.57186	1.24875
H	-0.18884	-4.50806	2.00549
H	-0.63489	4.03658	0.26329
H	-0.96235	4.62076	1.67026
H	3.68638	0.76707	1.24160
H	4.00337	2.09124	1.99739

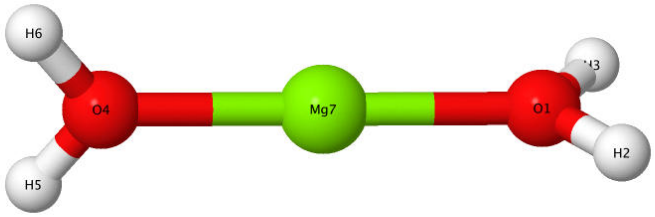
TABLE S1. Cumulative k^a values and two differently averaged (weighted) BE values for clusters **1-17**

#	$k^a_{cum.,explicit}$ (mdyn/Å)	BE (kcal/mol)	BE ÷ of 2nd shell H_2O (kcal/mol)	BE ÷ total water (kcal/mol)
1	1.748	-75.595	-75.595	-75.595
2	3.272	-143.372	-143.372	-71.686
3	4.026	-198.088	-198.088	-66.029
4	4.776	-243.485	-243.485	-60.871
5	4.377	-275.652	-275.652	-55.130
6	4.332	-305.297	-305.297	-50.883
7	4.617	-300.834	-300.834	-50.139
8	5.388	-324.944	-162.472	-46.421
9	5.669	-345.949	-115.316	-43.244
10	5.944	-365.531	-91.383	-40.615
11	6.316	-382.337	-76.467	-38.234
12	9.788	-478.727	-39.894	-28.160
13	4.644	-326.223	-326.223	-46.603
14	5.191	-347.678	-173.839	-43.460
15	5.492	-368.393	-122.798	-40.933
16	5.616	-368.631	-92.158	-36.863
17	9.941	-504.86	-42.072	-28.048



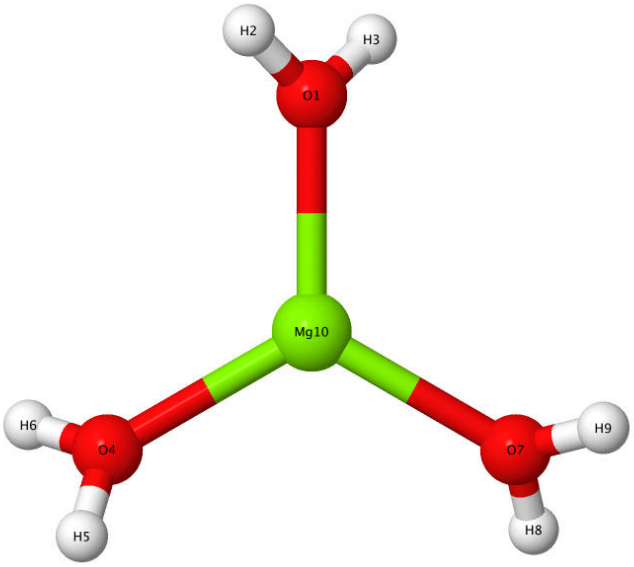
$[Mg(H_2O)]^{2+}$ (C_{2v})
1

Atom	#	Charge (e)
O	1	-0.60025
H	2	0.36906
H	3	0.36906
Mg	4	1.86213



$[Mg(H_2O)_2]^{2+}$ (D_{2d})
2

Atom	#	Charge (e)
O	1	-0.57770
H	2	0.35527
H	3	0.35527
O	4	-0.57770
H	5	0.35527
H	6	0.35527
Mg	7	1.73432

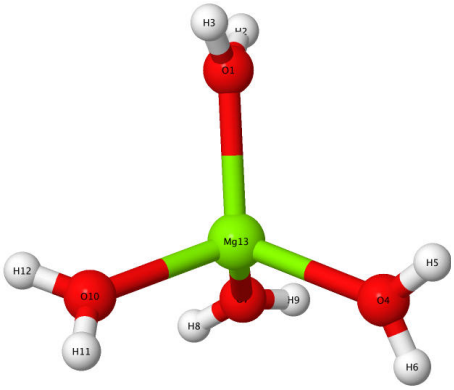


$[Mg(H_2O)_3]^{2+}$ (D_3)
3

Atom	#	Charge (e)
O	1	-0.56077
H	2	0.33737
H	3	0.33727
O	4	-0.56077
H	5	0.33737
H	6	0.33727
O	7	-0.56077
H	8	0.33727
H	9	0.33737
Mg	10	1.65840

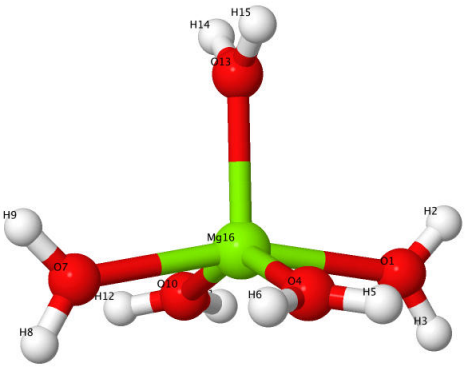
FIG. S1. NBO charges for $[Mg(H_2O)_n]^{2+}$ (n = 1-3) clusters (**1**-**3**).

$[Mg(H_2O)_4]^{2+}$ (S_4)
4



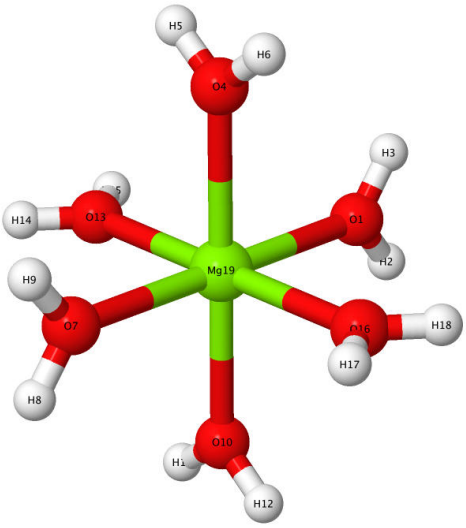
Atom	#	Charge (e)
O	1	-0.54545
H	2	0.32159
H	3	0.32146
O	4	-0.54545
H	5	0.32159
H	6	0.32146
O	7	-0.54545
H	8	0.32159
H	9	0.32146
O	10	-0.54545
H	11	0.32159
H	12	0.32146
Mg	13	1.60958

$[Mg(H_2O)_5]^{2+}$ (C_{2v})
5



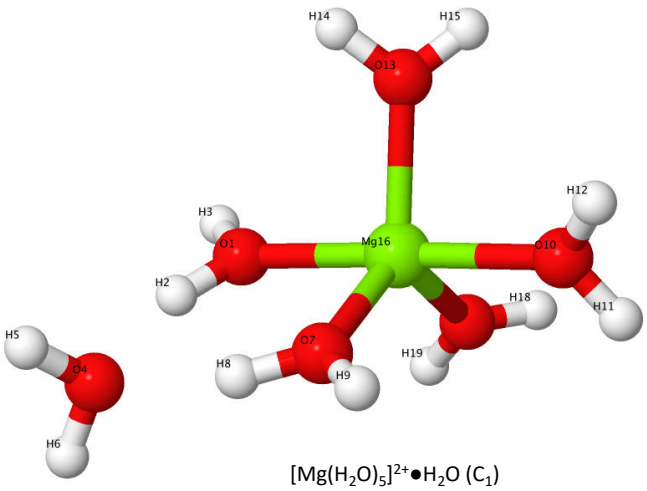
Atom	#	Charge (e)
O	1	-0.52578
H	2	0.29987
H	3	0.30239
O	4	-0.52990
H	5	0.30661
H	6	0.30661
O	7	-0.52578
H	8	0.30239
H	9	0.29987
O	10	-0.52990
H	11	0.30661
H	12	0.30661
O	13	-0.53301
H	14	0.30801
H	15	0.30801
Mg	16	1.59741

$[Mg(H_2O)_6]^{2+}$ (O_h)
6



Atom	#	Charge (e)
O	1	-0.51598
H	2	0.29263
H	3	0.29263
O	4	-0.51598
H	5	0.29263
H	6	0.29263
O	7	-0.51598
H	8	0.29263
H	9	0.29263
O	10	-0.51598
H	11	0.29263
H	12	0.29263
O	13	-0.51598
H	14	0.29263
H	15	0.29263
O	16	-0.51598
H	17	0.29263
H	18	0.29263
Mg	19	1.58430

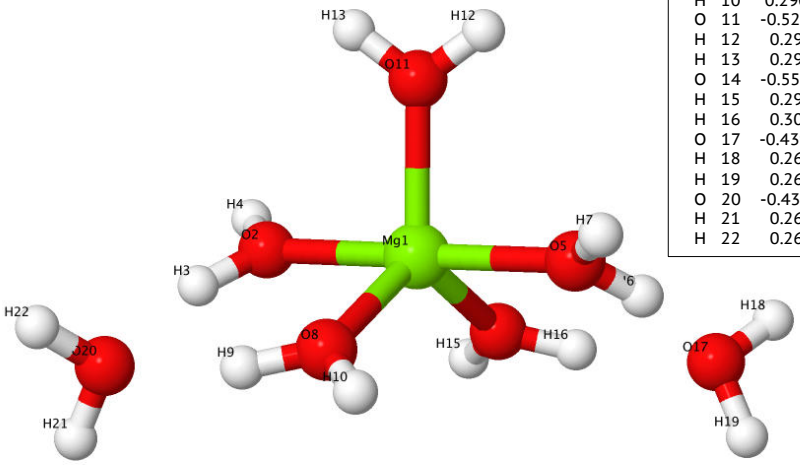
FIG. S2. NBO charges for $[Mg(H_2O)_n]^{2+}$ (n = 4-6) clusters (**4**-**6**).



Atom	#	Charge (e)
O	1	-0.55422
H	2	0.30679
H	3	0.28784
O	4	-0.44101
H	5	0.26504
H	6	0.26504
O	7	-0.55767
H	8	0.31175
H	9	0.29282
O	10	-0.52062
H	11	0.29730
H	12	0.29730
O	13	-0.52589
H	14	0.30263
H	15	0.30410
Mg	16	1.58794
O	17	-0.52590
H	18	0.30411
H	19	0.30264

$[Mg(H_2O)_5]^{2+}\bullet H_2O$ (C_1)

7

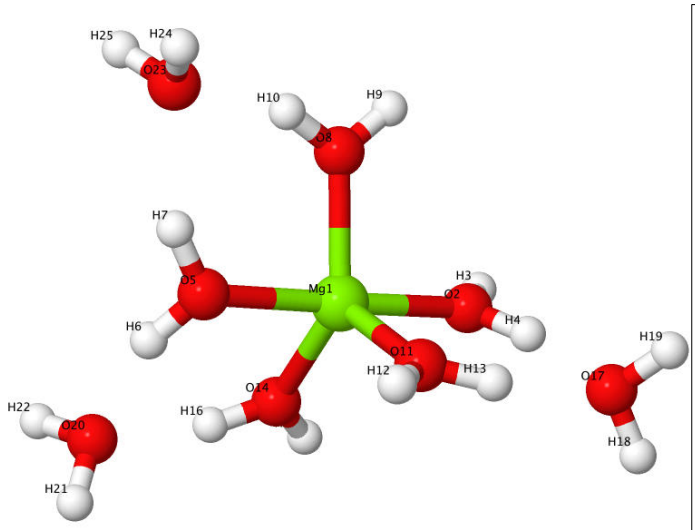


Atom	#	Charge (e)
Mg	1	1.58076
O	2	-0.54907
H	3	0.30295
H	4	0.28468
O	5	-0.54910
H	6	0.30297
H	7	0.28469
O	8	-0.55330
H	9	0.30883
H	10	0.29040
O	11	-0.52146
H	12	0.29999
H	13	0.29999
O	14	-0.55328
H	15	0.29041
H	16	0.30883
O	17	-0.43972
H	18	0.26297
H	19	0.26259
O	20	-0.43971
H	21	0.26259
H	22	0.26298

$[Mg(H_2O)_5]^{2+}\bullet 2(H_2O)$ (C_1)

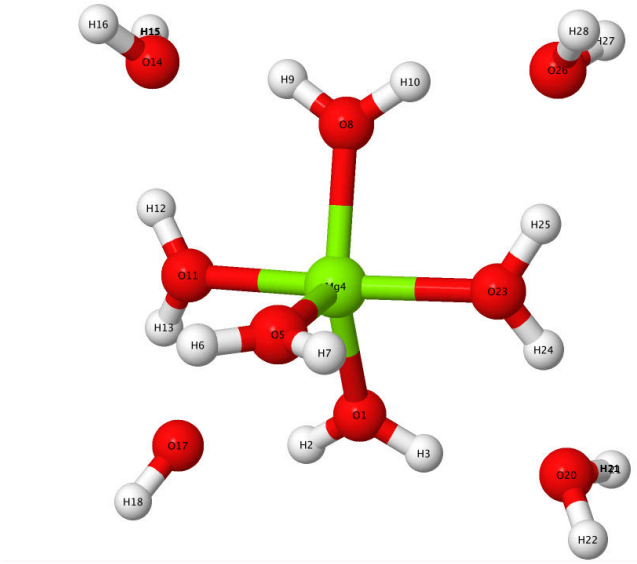
8

FIG. S3. NBO charges for $[Mg(H_2O)_n]^{2+}-m(H_2O)$ (n = 5, m = 1 and 2) clusters (**7** and **8**).



$[Mg(H_2O)_5]^2 \bullet 3(H_2O)$ (C_1)
9

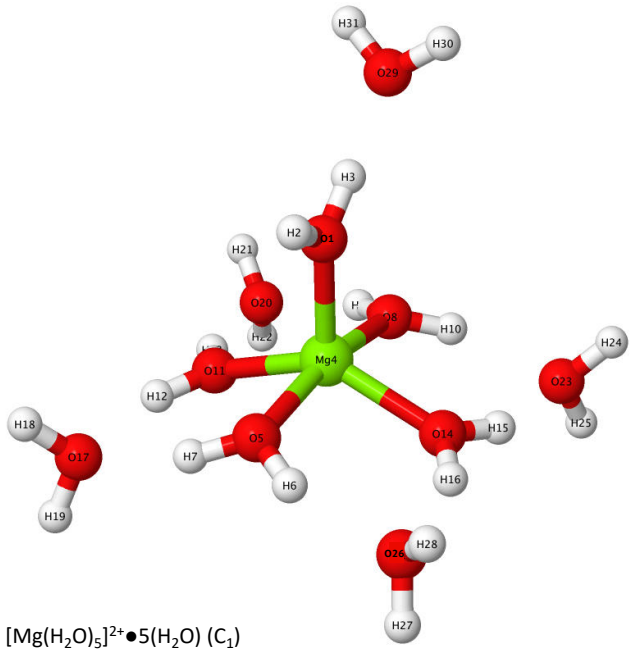
Atom	#	Charge (e)
Mg	1	1.57274
O	2	-0.54579
H	3	0.28340
H	4	0.30029
O	5	-0.56053
H	6	0.29271
H	7	0.29268
O	8	-0.55051
H	9	0.28624
H	10	0.30642
O	11	-0.54903
H	12	0.28969
H	13	0.30614
O	14	-0.55045
H	15	0.28624
H	16	0.30639
O	17	-0.43856
H	18	0.25963
H	19	0.25962
O	20	-0.43814
H	21	0.25747
H	22	0.25700
O	23	-0.43814
H	24	0.25749
H	25	0.25700



$[Mg(H_2O)_5]^2 \bullet 4(H_2O)$ (C_1)
10

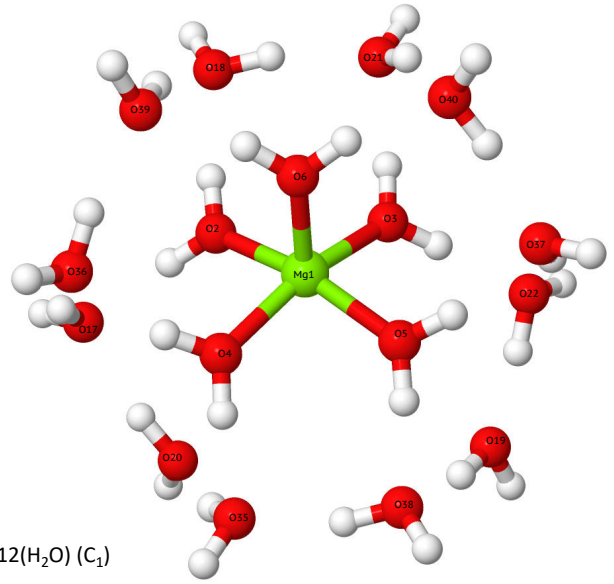
Atom	#	Charge (e)
O	1	-0.56946
H	2	0.29758
H	3	0.29754
Mg	4	1.56273
O	5	-0.51585
H	6	0.29535
H	7	0.29534
O	8	-0.56949
H	9	0.29755
H	10	0.29760
O	11	-0.56328
H	12	0.29339
H	13	0.29334
O	14	-0.43648
H	15	0.25085
H	16	0.25170
O	17	-0.43647
H	18	0.25168
H	19	0.25085
O	20	-0.43647
H	21	0.25085
H	22	0.25168
O	23	-0.56324
H	24	0.29337
H	25	0.29332
O	26	-0.43647
H	27	0.25084
H	28	0.25168

FIG. S4. NBO charges for $[Mg(H_2O)_n]^{2+} - m(H_2O)$ (n = 5, m = 3 and 4) clusters (**9** and **10**).



Atom	#	Charge (e)
O	1	-0.56458
H	2	0.26746
H	3	0.31897
Mg	4	1.55978
O	5	-0.55830
H	6	0.28964
H	7	0.28958
O	8	-0.55925
H	9	0.29178
H	10	0.29179
O	11	-0.56546
H	12	0.29437
H	13	0.29435
O	14	-0.56533
H	15	0.29433
H	16	0.29426
O	17	-0.43498
H	18	0.25015
H	19	0.24930
O	20	-0.43481
H	21	0.24970
H	22	0.24919
O	23	-0.43480
H	24	0.24970
H	25	0.24919
O	26	-0.43498
H	27	0.24928
H	28	0.25014
O	29	-0.42981
H	30	0.24966
H	31	0.24967

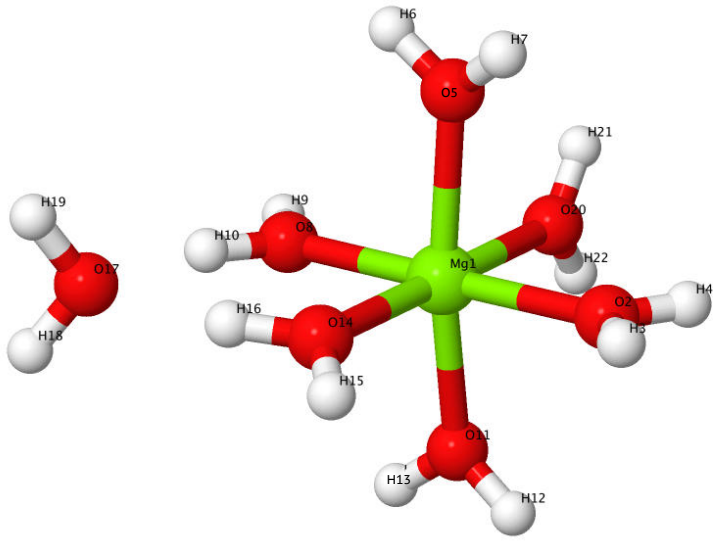
$[Mg(H_2O)_5]^{2+}\bullet 5(H_2O)$ (C_1)
11



Atom	#	Charge (e)
Mg	1	1.55654
O	2	-0.57207
O	3	-0.57273
O	4	-0.57726
O	5	-0.57936
O	6	-0.57388
O	35	-0.46829
O	36	-0.45309
O	37	-0.45935
O	38	-0.45794
O	39	-0.45455
O	40	-0.45777

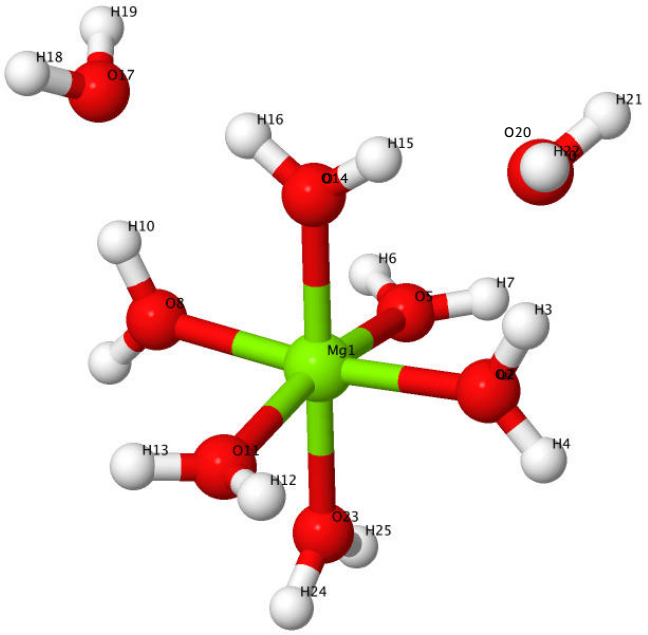
$[Mg(H_2O)_5]^{2+}\bullet 12(H_2O)$ (C_1)
12

FIG. S5. NBO charges for $[Mg(H_2O)_n]^{2+}-m(H_2O)$ (n = 5, m = 5 and 12) clusters (**11** and **12**).



Atom	#	Charge (e)
Mg	1	1.57950
O	2	-0.51176
H	3	0.29105
H	4	0.28742
O	5	-0.50795
H	6	0.28679
H	7	0.28800
O	8	-0.54463
H	9	0.28053
H	10	0.30000
O	11	-0.50830
H	12	0.28822
H	13	0.28716
O	14	-0.54147
H	15	0.28027
H	16	0.29785
O	17	-0.43880
H	18	0.25993
H	19	0.25994
O	20	-0.51191
H	21	0.28906
H	22	0.28912

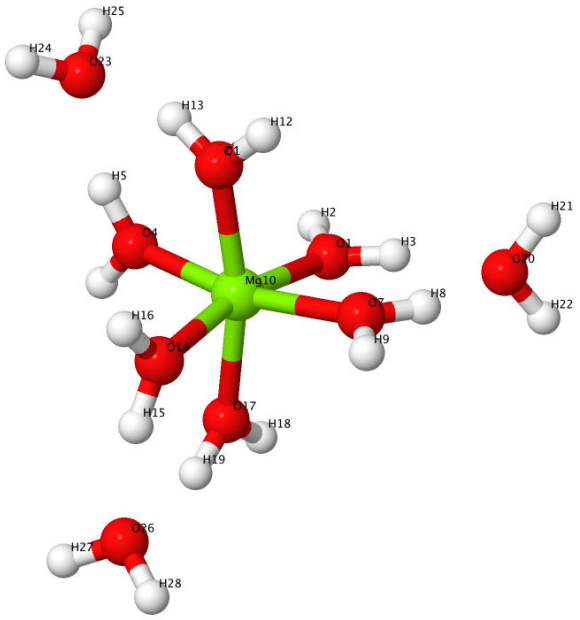
$[Mg(H_2O)_6]^{2+}\bullet H_2O$ (C_1)
13



Atom	#	Charge (e)
Mg	1	1.57331
O	2	-0.54309
H	3	0.29832
H	4	0.27765
O	5	-0.50330
H	6	0.28375
H	7	0.28375
O	8	-0.54309
H	9	0.27765
H	10	0.29832
O	11	-0.50330
H	12	0.28375
H	13	0.28375
O	14	-0.55878
H	15	0.29039
H	16	0.29039
O	17	-0.43677
H	18	0.25452
H	19	0.25451
O	20	-0.43677
H	21	0.25452
H	22	0.25451
O	23	-0.50962
H	24	0.28783
H	25	0.28783

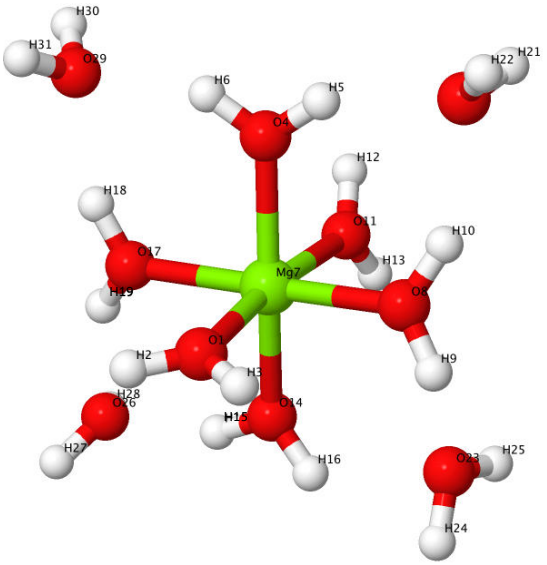
$[Mg(H_2O)_6]^{2+}\bullet 2(H_2O)$ (C_2)
14

FIG. S6. NBO charges for $[Mg(H_2O)_n]^{2+}-m(H_2O)$ (n = 6, m = 1 and 2) clusters (**13** and **14**).



Atom	#	Charge (e)
O	1	-0.53251
H	2	0.27350
H	3	0.29449
O	4	-0.53242
H	5	0.29445
H	6	0.27349
O	7	-0.53245
H	8	0.29447
H	9	0.27349
Mg	10	1.56840
O	11	-0.53254
H	12	0.27352
H	13	0.29452
O	14	-0.53248
H	15	0.29448
H	16	0.27347
O	17	-0.53250
H	18	0.27349
H	19	0.29450
O	20	-0.43614
H	21	0.25452
H	22	0.25449
O	23	-0.43614
H	24	0.25451
H	25	0.25451
O	26	-0.43614
H	27	0.25451
H	28	0.25451

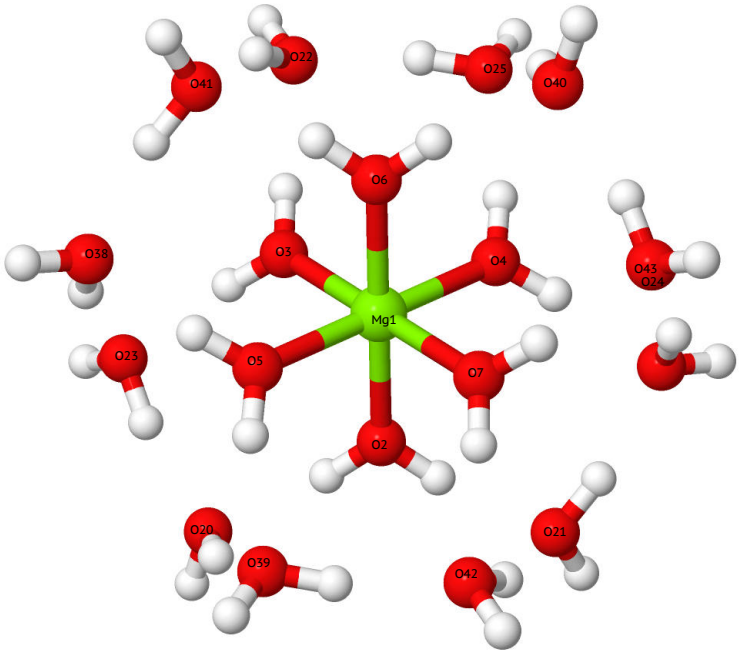
$[Mg(H_2O)_6]^{2+}\bullet 3(H_2O)$ (C_1)
15



Atom	#	Charge (e)
O	1	-0.49836
H	2	0.27899
H	3	0.27900
O	4	-0.55259
H	5	0.28785
H	6	0.28788
Mg	7	1.56068
O	8	-0.55258
H	9	0.28788
H	10	0.28788
O	11	-0.49827
H	12	0.27892
H	13	0.27891
O	14	-0.55260
H	15	0.28786
H	16	0.28786
O	17	-0.55247
H	18	0.28778
H	19	0.28779
O	20	-0.43406
H	21	0.24546
H	22	0.24545
O	23	-0.43405
H	24	0.24547
H	25	0.24548
O	26	-0.43405
H	27	0.24549
H	28	0.24548
O	29	-0.43405
H	30	0.24548
H	31	0.24549

$[Mg(H_2O)_6]^{2+}\bullet 4(H_2O)$ (C_1)
16

FIG. S7. NBO charges for $[Mg(H_2O)_n]^{2+}-m(H_2O)$ (n = 6, m = 3 and 4) clusters (**15** and **16**).



Atom	#	Charge (e)
Mg	1	1.55497
O	2	-0.55525
O	3	-0.55537
O	4	-0.55526
O	5	-0.55526
O	6	-0.55525
O	7	-0.55537
O	20	-0.45489
O	21	-0.45622
O	22	-0.45485
O	23	-0.45629
O	24	-0.45484
O	25	-0.45623
O	38	-0.45484
O	39	-0.45623
O	40	-0.45489
O	41	-0.45622
O	42	-0.45485
O	43	-0.45629

$[Mg(H_2O)_6]^{2+}\bullet 12(H_2O)$ (C_1)
17

FIG. S8. NBO charges for the $[Mg(H_2O)_6]^{2+} - 12\,(H_2O)$ cluster (**17**).