

Electronic supplementary information

The extraordinary richness of the reaction between diazomethane and tetracyanoethylene:
can computational calculations shed light on old papers?

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Table S1. Electronic energy and optimized geometry of the neutral systems

1

Total Energy= -447.638103126 Hartree, NIMAG= 0

X

X,1,1.

C,1,r1,2,90.

C,1,r1,2,90.,3,180.,0

C,3,r2,1,a2,2,0.,0

C,3,r2,1,a2,2,180.,0

C,4,r2,1,a2,2,0.,0

C,4,r2,1,a2,2,180.,0

N,3,r3,1,a3,2,0.,0

N,3,r3,1,a3,2,180.,0

N,4,r3,1,a3,2,0.,0

N,4,r3,1,a3,2,180.,0

r1=0.68389344

r2=1.42639847

r3=2.58075543

a2=121.58921373

a3=122.09985279

2

Total Energy= -148.785746870 Hartree, NIMAG= 0

N

X,1,1.

N,1,r1,2,90.

C,1,r2,2,90.,3,180.,0

H,4,r3,1,a3,2,0.,0

H,4,r3,1,a3,2,180.,0

r1=1.1372511

r2=1.2943678

r3=1.07793034

a3=118.00492139

3

Total Energy= -596.434584780 Hartree, NIMAG= 0

N,-0.5250795625,0.4389775121,0.8243813819

N,0.6387811135,0.7823739608,0.7034125668

C,0.858650533,1.9939356368,-0.098601562

H,1.2185228747,1.6831672362,-1.0834493129

H,1.6118758696,2.6225976363,0.3734359539

C,-0.5480378065,2.6836358779,-0.1838817827

C,-1.4844298033,1.4214823123,0.038655549

C,-0.6912318477,3.645060449,0.917971787

N,-0.7541400714,4.3988712423,1.7860488304

C,-0.7584988467,3.3629111247,-1.4620586108

N,-0.8916939742,3.9054935258,-2.4691800646
C,-2.6830371052,1.6665330809,0.8289313215
N,-3.6269051887,1.8479120628,1.463067318
C,-1.8068975983,0.7410287776,-1.2147126735
N,-2.0182084661,0.1751034045,-2.195555722

4

Total Energy= -486.944586934 Hartree, NIMAG= 0
C,0.113226376,1.8566047589,0.3304821426
H,0.894621847,1.3358721074,-0.2068573065
H,0.2446544445,1.9572465415,1.3996537384
C,-0.5445848937,3.0206405177,-0.3852517694
C,-1.3056464854,1.6493232263,-0.1628470323
C,-0.9805429416,4.1534079754,0.3959033714
N,-1.3102213516,5.0596537462,1.0265999945
C,-0.1218523369,3.3322818515,-1.7297633358
N,0.2343869951,3.5828295811,-2.7965686092
C,-2.377525381,1.6368506287,0.8038097454
N,-3.2240253071,1.6124342423,1.5852050621
C,-1.5182471244,0.8154981208,-1.3218152686
N,-1.6782752808,0.1350890724,-2.2378797126

5a

Total Energy= -596.377934673 Hartree, NIMAG= 0
N,-0.6969809729,0.6644974856,0.6816025127
N,0.3844201313,0.3552791113,-0.2633023734
C,0.4921844789,1.5474409999,0.4779233673
H,0.1252249663,0.4472240557,-1.2479946824
H,1.2844445263,1.6831785615,1.2038005362
C,-0.3250629151,2.7390178413,-0.104191277
C,-1.593732996,1.7475269997,0.0078684117
C,-0.3892212754,3.8770653831,0.8050414313
N,-0.3892214916,4.7662199685,1.5371470261
C,0.064647552,3.1609296085,-1.4457115982
N,0.3881101141,3.444326477,-2.5147204102
C,-2.6245987772,2.1817092267,0.9372527576
N,-3.4424291562,2.5081313135,1.6794398591
C,-2.1671429026,1.3110901071,-1.2568360774
N,-2.5709711519,0.9354466906,-2.2688545035

5b

Total Energy= -596.383600680 Hartree, NIMAG= 0
N,-0.6864829209,0.6772344377,0.7096490814
N,0.3400596233,0.4282181292,-0.348763923
C,0.4872490067,1.5435797831,0.5250599223
H,0.8750174641,-0.3667989425,0.000444086
H,1.2734426148,1.6598498332,1.2609824641
C,-0.3005300219,2.7250007273,-0.0978989101
C,-1.575544847,1.7121202579,0.0172752643
C,-0.3899474517,3.8785350789,0.7900095163

N,-0.4174915385,4.7791206508,1.5073779691
C,0.1077997384,3.1268968204,-1.4385881714
N,0.4460192741,3.4455051347,-2.4923767225
C,-2.6136600789,2.1667972311,0.9345048055
N,-3.4334271479,2.5162178589,1.6636478273
C,-2.1543884452,1.2629779781,-1.2422112127
N,-2.6377968495,0.9144316911,-2.2275835166

6

Total Energy= -596.441099356 Hartree, NIMAG= 0

N,-0.9049037507,0.5363756134,0.5383907055
N,0.4716705048,0.5649849117,0.4791564248
C,0.8810298498,1.7305215885,0.1849203465
H,-1.2797835066,-0.3744885712,0.2976721191
H,1.9244683255,2.0094652648,0.1625349494
C,-0.276072362,2.7378834958,-0.0183747161
C,-1.4580703921,1.6783487719,-0.2330566437
C,-0.4568830091,3.5113853352,1.2208464526
N,-0.5496377888,4.1092891582,2.2007981048
C,-0.0774537097,3.6444003866,-1.1494247487
N,0.1169253266,4.3574487537,-2.0323868032
C,-2.7537910824,2.1101541547,0.2981926174
N,-3.7843196142,2.4279473679,0.7008787776
C,-1.599366127,1.3339497479,-1.6638977853
N,-1.714142554,0.991417851,-2.7577848107

7

Total Energy= -596.407469466 Hartree, NIMAG= 0

N,-0.5447206565,0.933427462,1.2384983883
N,0.4463188481,1.7941737851,1.270404007
C,0.5548582394,2.7724286054,0.4008282981
H,1.1177328082,1.6595724919,2.0240482075
H,1.3043168867,3.5433935621,0.4500221674
C,-0.7280134646,2.7669955006,-0.4132357362
C,-1.2231735813,1.2571857482,-0.0399710031
C,-1.6372698823,3.795606128,0.1296465241
N,-2.2997982369,4.6161046817,0.5937309521
C,-0.5375161452,2.9850088443,-1.8482375578
N,-0.3733937632,3.1886020957,-2.969660545
C,-2.6777384654,1.1476897089,0.0876268231
N,-3.8215589612,1.0594675922,0.1807482015
C,-0.7281464976,0.3072634094,-1.0609742179
N,-0.3122269982,-0.4578357757,-1.8150095194

8a

Total Energy= -596.451950044 Hartree, NIMAG= 0

N,1.6370488253,3.0268340157,1.528539449
N,1.1889791954,1.7629751968,1.3694431276
C,0.0404237024,1.242300957,0.8189645283
H,1.6957243331,1.1308268476,1.9764326475

H,0.1094732768,0.1713918648,0.6635126175
C,-1.1461636546,1.8538209964,0.539730119
C,1.5086057788,3.9152109613,0.592708315
C,-2.1895078701,1.0765979314,-0.0488083899
N,-3.0317120614,0.4394708636,-0.516782194
C,-1.4396274385,3.2032491428,0.8829657948
N,-1.6614364727,4.2916236242,1.2023781446
C,2.0275886059,5.2222597008,0.8683887619
N,2.4416299154,6.2825439148,1.0569772082
C,1.0288931057,3.6744484621,-0.7363937871
N,0.7188244085,3.4295264605,-1.8214938623

8b

Total Energy= -596.465372120 Hartree, NIMAG= 0
N,0.870500948,2.6770723933,2.7694338887
N,0.9901457874,1.8244092073,1.757573904
C,0.0554859463,1.1693768886,0.9950935203
H,1.9674560844,1.6544696536,1.5413038198
H,-0.9878279428,1.3388954118,1.2112593922
C,0.4115880399,0.3225287045,-0.0135573675
C,-0.2376560262,3.1041712405,3.2926216762
C,-0.5840697269,-0.3415548397,-0.7857267339
N,-1.3885100731,-0.8820954115,-1.414303002
C,1.7864376213,0.0879372502,-0.3116399635
N,2.9193158963,-0.0521110518,-0.4942132633
C,-0.1019634411,4.0265443002,4.3831446573
N,-0.0336118392,4.7702871075,5.2630766381
C,-1.5839685976,2.7751119643,2.9239386542
N,-2.6723778466,2.5160394612,2.6341563794

8c

Total Energy= -596.444874681 Hartree, NIMAG= 0
N,0.6472206369,2.6228866627,3.2611659119
N,1.4637708453,1.8881760561,2.4133564754
C,1.4059783781,2.3121783556,1.2150576787
H,1.5043388172,1.1223027315,-0.5379755898
H,0.7722056303,3.1480818923,0.914443784
C,2.2196082203,1.6368676328,0.1222980461
C,0.4663093343,2.061191065,4.4064899004
C,2.8960161808,2.6553359515,-0.6991095988
N,3.3892155386,3.4734010741,-1.3413967546
C,3.1658392654,0.6292806836,0.6177217142
N,3.9100991887,-0.1779544146,0.9608941276
C,-0.3472703633,2.7487347846,5.3696315965
N,-0.9986955987,3.2738594553,6.163535373
C,1.0039234136,0.7859878506,4.8049947137
N,1.3990837523,-0.232267171,5.1762477118

9

Total Energy= -1044.03080737 Hartree, NIMAG= 0

N,-0.1163498488,0.7877473993,-0.098860189
 N,1.0077152604,1.6621094436,0.0624478823
 C,0.4309963379,2.6352118029,0.9843549968
 H,1.3460622848,2.0104585236,-0.8311524048
 C,-0.8640162065,3.1301057461,0.2129677193
 C,-1.2312669502,1.72343772,-0.5548359796
 C,-1.9739343855,3.691169505,0.9801620996
 N,-2.8526130102,4.1967632308,1.526674847
 C,-0.4101135089,4.1648464126,-0.7249326446
 N,0.0053706876,4.9788059177,-1.4262998538
 C,-2.6083071819,1.2864443545,-0.3317740238
 N,-3.7221247732,1.0071292705,-0.2465381048
 C,-1.0441233478,1.8447250974,-2.0093140524
 N,-0.8815384373,1.9263395017,-3.1461859004
 H,1.0957080439,3.4532172255,1.2507219411
 C,-0.2009705359,0.2963107649,1.3145337305
 C,0.1553075448,1.6615323299,2.1906994381
 C,-0.8914542477,2.0232687093,3.1445347124
 N,-1.6731807771,2.2652862897,3.9547156747
 C,1.4077104356,1.5207411663,2.9419611917
 N,2.3905414644,1.4629128459,3.5390447731
 C,0.8523922505,-0.7194322302,1.4606986831
 N,1.6410504451,-1.5480458421,1.5869479292
 C,-1.4600239857,-0.3326256635,1.7139150371
 N,-2.384603248,-0.8909444414,2.1121591074

10

Total Energy= -148.747604729 Hartree, NIMAG= 0

N,-2.2784385074,-1.3873216746,-0.468363129
 N,-2.7080194479,-0.2327625026,-0.3121079623
 C,-2.9194563545,0.9335406076,-0.213235728
 H,-2.6367222436,-1.9879788665,0.2789310365
 H,-3.7070624465,1.5700124361,-0.5771732172

11

Total Energy= -502.967470999 Hartree, NIMAG= 0

N,1.4919783947,-0.0339669998,0.
 H,1.9523689367,0.8756131185,0.
 C,0.248798039,-0.0071497344,-0.77515856
 C,0.248798039,-0.0071497344,0.77515856
 C,-0.1555399567,1.2106076003,-1.4444567533
 N,-0.4426479589,2.1824244269,-1.993315656
 C,-0.0924361179,-1.226393716,-1.4784630172
 N,-0.3813207044,-2.1867280755,-2.0445911153
 C,-0.1555399567,1.2106076003,1.4444567533
 N,-0.4426479589,2.1824244269,1.993315656
 C,-0.0924361179,-1.226393716,1.4784630172
 N,-0.3813207044,-2.1867280755,2.0445911153

12a

Total Energy= -596.415260974 Hartree, NIMAG= 0
C,-1.0075988703,-0.4053682729,0.
H,-1.9809176098,0.0768618615,0.
C,0.1078345687,0.2848510882,0.7864644836
C,0.1078345687,0.2848510882,-0.7864644836
C,-0.2258440484,1.5196329363,1.4544975932
N,-0.5039718395,2.4996936038,1.9928341229
C,1.1026784593,-0.5197711487,1.4562819658
N,1.8989259584,-1.1283623232,2.0239895104
C,-0.2258440484,1.5196329363,-1.4544975932
N,-0.5039718395,2.4996936038,-1.9928341229
C,1.1026784593,-0.5197711487,-1.4562819658
N,1.8989259584,-1.1283623232,-2.0239895104
N,-0.9723564422,-1.8415174512,0.
N,-2.1086527646,-2.3334118918,0.
H,-1.9890975409,-3.3579453451,0.

12b, Total Energy= -596.396611420 Hartree, NIMAG= 0
N,0.8388581153,-0.544022782,0.
H,2.4437790579,-2.3495318245,0.
C,-0.0925363762,0.322684782,-0.7690406663
C,-0.0925363762,0.322684782,0.7690406663
C,0.5414209777,1.4124747155,-1.4773013941
N,1.0359179278,2.2838381381,-2.0455045176
C,-1.2324840607,-0.2542300673,-1.4493222286
N,-2.1241844395,-0.6727182322,-2.0462248864
C,0.5414209777,1.4124747155,1.4773013941
N,1.0359179278,2.2838381381,2.0455045176
C,-1.2324840607,-0.2542300673,1.4493222286
N,-2.1241844395,-0.6727182322,2.0462248864
N,0.4125008542,-1.918933877,0.
C,1.4118768476,-2.7006206718,0.
H,1.1992322209,-3.7655558666,0.

13a
Total Energy= -745.237968398 Hartree, NIMAG= 0
N,-1.7652920397,1.7277513342,0.6908824944
N,-0.8680443272,0.731136102,1.1693295779
C,0.490481648,0.9955235182,0.696423331
H,-2.5242319126,1.2806670348,0.1864582831
C,0.4689714201,2.5456411306,0.3576415692
C,-1.0244652019,2.6541975489,-0.1964411418
C,0.6428060865,3.3834709209,1.5500315502
N,0.820657082,4.0346736283,2.4828448388
C,1.4832768912,2.9036280751,-0.6333064134
N,2.3113468433,3.1425924073,-1.3972735694
C,-1.5794224625,4.0126267298,-0.1125641804
N,-2.0310150095,5.0708252731,-0.0705886776
C,-1.0653083171,2.2028023739,-1.6064366181
N,-1.1494029679,1.7908281022,-2.6791846952

H,0.7088421511,0.4531550973,-0.2218794068
N,-0.8087121004,0.7159583586,2.6213918307
N,0.3399254285,0.5437388828,3.0171963417
C,1.3150145208,0.5277083023,1.8922379609
H,2.1703182704,1.151663863,2.1518548449
H,1.6613961968,-0.4993768333,1.75586996

13b

Total Energy= -745.241295540 Hartree, NIMAG= 0

N,-0.8256465489,1.0882068247,1.2094247201
N,0.6178945802,0.8228941452,1.0802835621
C,1.1914973502,2.048158813,0.5060013847
H,-1.2891482853,0.1811875805,1.2014298893
C,-0.0155489433,2.922557882,-0.0062746202
C,-1.158459455,1.8079421231,-0.0235331276
C,-0.3740534185,3.9774199512,0.952764096
N,-0.6744206048,4.8090891955,1.6895928951
C,0.2353001941,3.5182336663,-1.3193720277
N,0.4547414199,3.9801833681,-2.3508630734
C,-2.5203092579,2.3495857333,0.0520359468
N,-3.6002307066,2.7454025346,0.0938241974
C,-1.035257741,0.9706518149,-1.2424006586
N,-0.9585252658,0.2674621501,-2.1515830212
H,1.8891460196,1.8436189448,-0.3068141541
N,1.9501613286,2.7256879628,1.6037383612
N,1.9675057196,2.024521635,2.6147536412
C,1.2444537068,0.7426817129,2.4067425002
H,1.9885448325,-0.0569371829,2.4439509495
H,0.5094972756,0.6006629948,3.2007864291

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Total Energy= -635.724822385 Hartree, NIMAG= 0

N,-0.6387813165,0.6933959957,0.5281541919
N,0.7440656314,1.0155487785,0.719768924
C,0.8954494918,2.4165821868,0.3470391107
H,-0.7448710636,-0.2858187454,0.2805118835
H,1.4615249768,3.066437987,1.0030019829
C,-0.4747464269,2.9706726728,-0.1102919367
C,-1.2934845099,1.6180411014,-0.42183879
C,-1.0868771748,3.6981970989,1.0120194027
N,-1.5170475599,4.2845636638,1.9044404574
C,-0.3700224628,3.8592901325,-1.2717559802
N,-0.2522833382,4.5639070054,-2.1748975954
C,-2.7251737625,1.719595358,-0.102532142
N,-3.8520151619,1.7700432204,0.1279464755
C,-1.1897381928,1.2231646499,-1.8446890557
N,-1.1159739299,0.8183438017,-2.921871704
C,1.5363801574,1.3748821224,-0.4951163535
H,2.597057781,1.1987031848,-0.366163536
H,1.1493167613,1.1572323953,-1.4816672052

15

Total Energy= -502.968113724 Hartree, NIMAG= 0
N,-0.1430508335,0.5205025789,0.6909832533
N,0.5850745023,1.4690817961,1.4791579164
C,0.578947225,2.6035888211,0.9010398475
H,1.0656207416,3.4825612727,1.3051819414
C,-0.1915413216,2.5847176491,-0.4266130267
C,-0.5868695282,1.0938607267,-0.3657820928
C,-1.3573617038,3.4738637051,-0.3861410672
N,-2.2716724051,4.1713867935,-0.3248786961
C,0.6855985086,2.8841255447,-1.5634313456
N,1.3976741275,3.112236897,-2.4393130664
C,-1.3519044496,0.4350836827,-1.3648452972
N,-1.9758526931,-0.0613434576,-2.1991134066

16

Total Energy= -1044.07647993 Hartree, NIMAG= 0
N,-0.7502609519,1.4948688033,1.2477545415
N,0.335799982,1.4244146334,0.693964854
C,0.4302133867,2.1090167931,-0.6042064782
H,0.3838812037,1.3456440159,-1.386276147
H,1.3812273401,2.6349341428,-0.6771166419
C,-0.8096188384,3.0670321222,-0.6416087496
C,-1.7730425515,2.3091989268,0.3700751936
C,-0.4318819751,4.3857569818,-0.114461829
N,-0.0922236704,5.4065649605,0.2957532078
C,-1.3628473662,3.2133122426,-1.9879674321
N,-1.7711046305,3.3276594539,-3.0587039267
C,-2.5733153526,3.1704685571,1.2313654129
N,-3.2010590109,3.8412893169,1.9253956121
C,-2.6085411743,1.3100003211,-0.2959702216
N,-3.2316311459,0.487413731,-0.8077189254
C,2.8781965945,-1.35811667,1.39725539
C,3.5645790673,-0.2920509585,1.9103502511
C,2.7355957439,-2.5794755481,2.1179339297
N,2.6112163158,-3.5777746326,2.684316793
C,2.2706853284,-1.2874735722,0.107979847
N,1.7757020366,-1.2229129786,-0.9326680835
C,3.6956253366,0.9212240946,1.1707009021
N,3.796636903,1.9016847275,0.5702833663
C,4.1772952318,-0.3392853309,3.1963608779
N,4.6824742174,-0.3580369634,4.234297796

18b

Total Energy= -1044.08684634 Hartree, NIMAG= 0
N,1.7425839444,-1.3420897981,0.7927542066
N,1.9513611906,-2.1256008488,-0.3181859535
C,2.7341092043,-1.5513774639,-1.1371881059
H,0.8260659283,-1.4945768522,1.210343691

H,3.1020433232,-2.0155950556,-2.0405085423
 C,3.204908546,-0.1635326162,-0.6386619036
 C,2.0751129552,0.0670326062,0.4725714852
 C,4.5412642967,-0.2986410305,-0.0367973947
 N,5.5954563142,-0.4434460062,0.4038178403
 C,3.2276129549,0.86271876,-1.6813489557
 N,3.2493218284,1.6456092163,-2.5255441929
 C,2.5368146822,0.8000831262,1.6546104964
 N,2.8726983322,1.3832399611,2.5885966143
 C,0.9049351915,0.7676569972,-0.1072488853
 N,-0.0632712927,1.2501940898,-0.5023327313
 C,-3.3200570013,0.1005283115,0.6067366828
 C,-3.6083738344,0.1124673337,-0.7299205281
 C,-2.226063679,-0.6614859599,1.1160636749
 N,-1.3286991231,-1.2569034027,1.5284131021
 C,-4.0810881166,0.848132642,1.5517184659
 N,-4.6874036008,1.4408662442,2.3352576957
 C,-2.8489877684,-0.6519766291,-1.664162348
 N,-2.2527084058,-1.2780415726,-2.4289912161
 C,-4.6892766187,0.8816951235,-1.2522583711
 N,-5.5627162516,1.4937048242,-1.6942048267

19

Total Energy= -1044.05408079 Hartree, NIMAG= 0

N,0.2323248798,0.6532670478,0.6155052646
 N,0.5732046277,1.0984482884,1.8853831182
 C,0.7362401064,2.3543324708,1.8917039695
 H,-1.6181579782,-1.2404896548,0.4685958828
 H,0.9458915036,2.9153314814,2.7918110041
 C,0.4916810901,3.0010449512,0.5151839014
 C,0.6412189064,1.6842860617,-0.3991003007
 C,-0.8829913192,3.5266687544,0.4883181537
 N,-1.9588546433,3.9359118664,0.5232733019
 C,1.4423314772,4.0611440949,0.1795048373
 N,2.1882145993,4.9085338348,-0.0462602682
 C,-0.2562113221,1.7144937073,-1.5558452389
 N,-0.9695607486,1.7687271021,-2.4575466247
 C,2.043119469,1.5214810359,-0.8288239793
 N,3.1420891847,1.3657463622,-1.1359672802
 C,0.498101328,-0.7973372995,0.4413171361
 C,-0.753539435,-1.5933183461,1.0421527198
 C,1.7514783175,-1.2201496976,1.1003917209
 N,2.7213802894,-1.5518867322,1.6239021425
 C,0.5718715778,-1.1447030258,-0.9879394253
 N,0.6078146207,-1.4538230903,-2.0959183067
 C,-0.6127361629,-3.0384937873,0.8283803663
 N,-0.5108417454,-4.1706126582,0.6483733684
 C,-1.0051452978,-1.309194774,2.458933151
 N,-1.2620085251,-1.1181728534,3.5641887256

Table S2. Electronic energy and optimized geometry of the protonated systems

3aH⁺

Total Energy= -596.710382887 Hartree, NIMAG= 0

N,-0.5106647401,0.5010001028,0.7407663709
 N,0.6815069302,0.7794707956,0.6712478018
 C,0.8748722514,2.00022341,-0.0859053718
 H,1.2837098335,1.6955927292,-1.0608578809
 H,1.6311065867,2.6102202637,0.4144981406
 C,-0.5371306445,2.6876321514,-0.1935832278
 C,-1.5090488833,1.4208929408,0.0079130537
 C,-0.7009155297,3.6301549543,0.9188992888
 N,-0.7862827665,4.3484185746,1.8152060516
 C,-0.7398257214,3.347692031,-1.4810908661
 N,-0.8704926671,3.8671684487,-2.5001738811
 C,-2.6780683235,1.6430293395,0.8467041983
 N,-3.6018993335,1.785837805,1.5200047718
 C,-1.8479903296,0.7455536812,-1.2444350576
 N,-2.0844823891,0.1817709814,-2.2213217628
 H,-0.8084443735,-0.3569652092,1.2291943806

3bH⁺

Total Energy= -596.709026631 Hartree, NIMAG= 0

N,-0.577810902,0.4679001009,0.8412459147
 N,0.5869192413,0.8295138634,0.688669834
 C,0.8786341873,2.025406691,-0.1425113406
 H,1.2120605214,1.6790719595,-1.1269266665
 H,1.6564057816,2.6261176145,0.3300897024
 C,-0.5255645954,2.701466587,-0.1790100519
 C,-1.4727215799,1.409855598,0.0568640049
 C,-0.652625936,3.6279441719,0.9525524099
 N,-0.7020166503,4.3295998823,1.864495387
 C,-0.7917056073,3.3724769599,-1.4488256087
 N,-0.9802122811,3.9019775305,-2.4535447268
 C,-2.6864350163,1.6739024708,0.8193201758
 N,-3.6457052879,1.8962269629,1.4161168524
 C,-1.7426550646,0.6883814342,-1.1910122829
 N,-1.9021435194,0.1008538215,-2.1696405395
 H,1.3228999186,0.2601092516,1.1320783957

5aH⁺

Total Energy= -596.661808069 Hartree, NIMAG= 0

N,-0.6749144352,0.6840407269,0.6066166856
 N,0.3984625799,0.4141595108,-0.3701060419
 C,0.517885705,1.5425479319,0.5076608774
 H,0.9338343164,-0.3947581425,-0.0352799442
 H,1.258558043,1.6359114584,1.295446203
 C,-0.2932442752,2.7240827981,-0.1103098335
 C,-1.6171020795,1.7274061979,-0.0285675124
 C,-0.3909392592,3.8576160644,0.7986231183

N,-0.4409098814,4.7313693053,1.547433553
C,0.1149988804,3.1092761762,-1.4511231938
N,0.4478692519,3.4051274922,-2.5131373931
C,-2.616055934,2.1341911901,0.9414853252
N,-3.4020555875,2.4282962793,1.7312699274
C,-2.1843288725,1.2867695844,-1.2863419572
N,-2.6431724215,0.9518089643,-2.2886844153
H,-0.9292243506,0.0103473724,1.3367016915

5bH⁺

Total Energy= -596.665671172 Hartree, NIMAG= 0

N,-0.6470223597,0.7107472034,0.6600044524
N,0.4052818604,0.4786482167,-0.3788897081
C,0.4928579103,1.6325180751,0.5066496614
H,0.9622187494,-0.3487289347,-0.1494264573
H,1.3234334039,1.7003631367,1.20148662
C,-0.316561512,2.8046819528,-0.0951021354
C,-1.5914092222,1.7603186551,0.0042595229
C,-0.4099003131,3.9298124142,0.8216738639
N,-0.4595185416,4.8077670681,1.565474106
C,0.0796824072,3.195619777,-1.4406596363
N,0.4087959942,3.4118835971,-2.5237512162
C,-2.6007250048,2.1307843135,0.9744382998
N,-3.3970831758,2.4303195375,1.7511559329
C,-2.1384045314,1.3138968815,-1.2636065788
N,-2.4695599356,0.9262475488,-2.2977743571
H,0.1642290708,0.5739727471,-1.3751090301

6aH⁺

Total Energy= -596.718597960 Hartree, NIMAG= 0

N,-0.9216995877,0.5026083158,0.5100497854
N,0.5445430645,0.5825510664,0.5369472146
C,0.8691123437,1.7682183328,0.2327403324
H,-1.1843384338,-0.3819127158,0.0530877232
H,1.9161816893,2.0583045694,0.2096175359
C,-0.2755904458,2.766272796,-0.03030044
C,-1.4918625356,1.7379730858,-0.2543437852
C,-0.4618290567,3.5536304979,1.1961750722
N,-0.5839848422,4.119154016,2.192516378
C,-0.0337990241,3.6224572164,-1.1903698089
N,0.1815733674,4.2909046127,-2.1027197313
C,-2.7619867695,2.1426274737,0.3317226938
N,-3.7687775252,2.4336707558,0.8102470645
C,-1.6356114956,1.3548234158,-1.6574601352
N,-1.7453146062,1.0086918783,-2.7506532456
H,-1.2635826724,0.466262253,1.4830207863

6bH⁺

Total Energy= -596.724436447 Hartree, NIMAG= 0

N,-0.9368818594,0.5175489678,0.5398308684

N,0.4378107292,0.6221998872,0.4541459439
C,0.8940805423,1.7841793227,0.1647227158
H,-1.2785260454,-0.4028347424,0.2622671364
H,1.9496681064,2.028352629,0.1666156293
C,-0.2743555655,2.7454128203,-0.0357387345
C,-1.476291836,1.6715620508,-0.2451597615
C,-0.4235049095,3.4769445224,1.2319706742
N,-0.4917665658,4.0194707703,2.2461750848
C,-0.094954857,3.651971847,-1.1675603935
N,0.0699608582,4.361935228,-2.0586620738
C,-2.7542098983,2.1187131859,0.2997803707
N,-3.770817174,2.4676447139,0.71184673
C,-1.5955491926,1.3077976511,-1.6661153727
N,-1.6557965105,0.957574509,-2.7621341713
H,1.006264818,-0.181789293,0.7312833541

6cH⁺

Total Energy= -596.718208767 Hartree, NIMAG= 0

N,-0.8913215182,0.5178683611,0.5265708766
N,0.4662416194,0.5626716033,0.5006617129
C,0.9017296079,1.7196099072,0.2073859314
H,-1.2692147138,-0.4059520847,0.3357934491
H,1.944454713,2.0022170635,0.1807839401
C,-0.2695949899,2.7156932402,-0.0585501243
C,-1.4648263853,1.6478804598,-0.2541004579
C,-0.4865691473,3.4896422619,1.1774887489
N,-0.5936412174,4.0795177119,2.1474909095
C,-0.0697401537,3.6565476389,-1.1538379534
N,0.1216841898,4.4126878707,-2.0016247743
C,-2.7280745902,2.1186295576,0.3194107846
N,-3.6836884708,2.5292451588,0.8140854738
C,-1.6152505287,1.3237556869,-1.6800914265
N,-1.7318942224,1.0190190464,-2.7850287461
H,-0.7207995924,4.6084914164,3.0037036858

6dH⁺

Total Energy= -596.716884663 Hartree, NIMAG= 0

N,-0.918328073,0.4900093938,0.4367468871
N,0.4547997087,0.575385055,0.5730601024
C,0.8760190901,1.7438982385,0.3405596329
H,-1.2081090615,-0.4153538449,0.0764035837
H,1.9114846408,2.0415468728,0.4426745743
C,-0.2426055335,2.7466922761,-0.0435105598
C,-1.4294154001,1.6647550784,-0.3091429873
C,-0.5752416917,3.5940954039,1.1100816755
N,-0.8858356547,4.2148081019,2.0299036043
C,0.0646350818,3.558457681,-1.2178305493
N,0.32710119,4.1926733222,-2.1426385392
C,-2.6854907294,2.0833556079,0.3465825247
N,-3.6643157209,2.3659094369,0.8567915169

C,-1.7041756703,1.4181138677,-1.727305245
N,-1.9420235629,1.1602532429,-2.825384521
H,-4.5183674834,2.6384983158,1.3321653896

8aH⁺

Total Energy= -596.735921511 Hartree, NIMAG= 0

N,1.5678189869,3.0181365937,1.4917277322
N,1.1653868512,1.7225506181,1.4252048114
C,0.0214678762,1.2067127003,0.844484676
H,1.8158035191,1.0769221156,1.8617361756
H,0.111656441,0.1484291973,0.6281739153
C,-1.1483858497,1.8564297666,0.5839550235
C,1.4833384939,3.9346780343,0.5347039871
C,-2.2232995489,1.1570465001,-0.0369180721
N,-3.0957030059,0.5899135458,-0.5370112562
C,-1.3285533876,3.2137053825,0.9650505144
N,-1.3335835502,4.316783886,1.3138570018
C,1.8383328824,5.2676249114,0.8524167058
N,2.1586904988,6.3498665021,1.1028314677
C,1.1360962066,3.5791990116,-0.7885865551
N,0.8914201811,3.259662251,-1.8724595315
H,1.946592115,3.3091798335,2.3957350542

8bH⁺

Total Energy= -596.747607507 Hartree, NIMAG= 0

N,1.7359365164,2.9604350491,1.4430799464
N,1.2426729038,1.7147772568,1.3602415239
C,1.3715969342,0.7068827284,2.298629633
H,0.721856123,1.5127214341,0.5098021958
H,0.8741769778,-0.1999297659,1.9770435897
C,2.01402193,0.7386208633,3.4974173157
C,1.5902378822,3.8993218958,0.523271479
C,2.0359116994,-0.4144824517,4.3316008459
N,2.0553229507,-1.3482842556,5.0102525387
C,2.6705271196,1.9295497519,3.9255008262
N,3.1414820887,2.971590145,4.1037240766
C,2.1665268494,5.1722115966,0.7435232021
N,2.6292170633,6.2195778171,0.9028862859
C,0.8628477316,3.610450556,-0.6581446319
N,0.2580278289,3.3033858782,-1.5946733369
H,2.2698264209,3.1771357909,2.3072346698

10H⁺

Total Energy= -149.091294207 Hartree, NIMAG= 0

N,0.07906863,1.14711922,0.
N,-0.01630297,-0.15238854,0.
C,-0.0027587,-1.29479095,0.
H,-0.21170684,1.58780029,-0.86961689
H,0.00060575,-2.3699656,0.
H,-0.21170684,1.58780029,0.86961689

15H⁺

Total Energy= -503.264475384 Hartree, NIMAG= 0

N,-0.1498744272,0.4954708896,0.6921877875
N,0.5491579049,1.4843524918,1.4097568121
C,0.5827487134,2.6539123887,0.8825279934
H,1.0809636769,3.515737088,1.3147468954
C,-0.190467197,2.5858766386,-0.4255530473
C,-0.5904442976,1.0819713269,-0.3666284794
C,-1.3635356912,3.4682331926,-0.3663250605
N,-2.2877924276,4.1504751772,-0.2840672191
C,0.7020697498,2.8722068466,-1.5567413788
N,1.429041934,3.0779309239,-2.4260299558
C,-1.3511687992,0.432490456,-1.361934042
N,-1.9760369464,-0.048991415,-2.2056953427
H,0.9740215773,1.193494895,2.2931287973

20a⁺

Total Energy= -596.702432532 Hartree, NIMAG= 0

N,0.0445939126,-0.0023273825,0.9161210985
N,0.9054440582,0.9844693952,0.640136325
C,0.8608137449,2.0936584913,-0.0202702808
H,0.2483866497,-0.5006856534,1.7788219714
H,0.0123741729,2.5838948954,-0.4835666185
C,2.3494814385,2.4123795783,0.0754760778
C,2.4072507909,0.9809568667,0.9034451465
C,3.052020833,2.3898930561,-1.2015543784
N,3.5779473183,2.3663777399,-2.2261358711
C,2.6597152693,3.588290691,0.8789463549
N,2.8725151722,4.5175987394,1.5255020869
C,3.0900748022,-0.1122534819,0.2370326075
N,3.6279889939,-0.9832617031,-0.2917233737
C,2.7125670858,1.0666277028,2.3206942056
N,2.9260889879,1.123353311,3.4516916442
H,-0.9332530402,0.2241872439,0.7565477742

Table S3. Electronic energy and optimized geometry of the TSs

1+2 → 3

Total Energy= -596.406493901 Hartree, NIMAG= 1

N,-0.464835767,0.1056663539,0.4825929377
 N,0.4282768571,0.8104350368,0.4241839242
 C,0.7941591647,2.0935360925,0.1505638958
 H,1.4077502495,2.1771707373,-0.7437173387
 H,1.1733149161,2.6207979758,1.0231785376
 C,-1.0514196856,2.8484291976,-0.2839198381
 C,-2.0197210032,1.7881203176,-0.1461578466
 C,-1.0818217825,3.9070564596,0.7045595223
 N,-1.0603204446,4.7472065122,1.4938500423
 C,-0.7727154034,3.3221488346,-1.6243022421
 N,-0.506268965,3.6987076876,-2.6809495575
 C,-2.7839049624,1.6700766486,1.0499166534
 N,-3.3905549171,1.5640362068,2.0282356502
 C,-2.4724527992,1.080805442,-1.2964795
 N,-2.8255731076,0.4950099072,-2.2284268505

1+10 → 6

Total Energy= -596.377882875 Hartree, NIMAG= 1

N,-1.9444419481,-1.4036141228,-0.1861474711
 N,-2.3851611432,-0.2351788893,-0.3177042547
 C,-2.0316697587,0.9032718202,-0.4901956531
 H,-2.2658800448,-1.826382645,0.6922354369
 H,-2.4437957443,1.8143207495,-0.8920375143
 C,0.0812329313,0.690204492,-0.1507372596
 C,0.2480483095,-0.7094801892,0.0644446494
 C,0.4848373904,1.2581382661,-1.4115728961
 N,0.7772861566,1.742058727,-2.4173474599
 C,0.1567446131,1.584947505,0.9756309616
 N,0.2027446513,2.317419597,1.8657680582
 C,0.7248497684,-1.545773433,-0.9923450317
 N,1.1265038491,-2.2218746075,-1.8373002791
 C,0.3712197518,-1.2175180339,1.3956819734
 N,0.4512454576,-1.6433406461,2.46652463

3 → 7

Total Energy= -596.330755146 Hartree, NIMAG= 1

N,-0.4850370926,0.3880093527,0.587992867
 N,0.679950921,0.9090567136,0.530881523
 C,0.8146521152,2.1532964092,-0.0160687201
 H,1.3813855293,0.8784052047,-0.4425478517
 H,1.7222553658,2.7181492031,0.1501053452
 C,-0.5863336585,2.7641965402,-0.1865198931
 C,-1.45861743,1.4046972799,0.0076384177
 C,-0.7913593435,3.7173156007,0.9137357549
 N,-0.8883361915,4.4604112471,1.788955539
 C,-0.7780894265,3.4341114181,-1.4722973899

N,-0.8974462062,3.9774476713,-2.4805692835
C,-2.6159181415,1.5751546476,0.8849749305
N,-3.5253157174,1.6996093379,1.579828683
C,-1.8748409854,0.8705686234,-1.2981482348
N,-2.157279618,0.4186545906,-2.3194967072

6 → 7

Total Energy= -596.335278788 Hartree, NIMAG= 1

N,-1.1565493535,0.4987445192,0.5564513528
N,0.2364928979,0.5581121272,0.5746055901
C,0.7825370987,1.6558763489,0.1812326832
H,-0.2451074489,-0.4226586471,0.1638386402
H,1.8461010544,1.8287483939,0.1296422733
C,-0.318207999,2.7146432931,-0.0120363754
C,-1.5718956859,1.721233967,-0.2130769546
C,-0.4418219911,3.5123089642,1.2210528981
N,-0.4928704546,4.1189148522,2.1988456039
C,-0.0771300634,3.5970808017,-1.1540412042
N,0.1452395959,4.2924432208,-2.0446040875
C,-2.8332269224,2.2547227824,0.2995615568
N,-3.8311002803,2.6703053251,0.695230035
C,-1.7074680694,1.3565672429,-1.6362687184
N,-1.7953222684,1.0120406386,-2.7319683035

6 → 10

Total Energy= -596.377882875 Hartree, NIMAG= 1

N,-1.9444419481,-1.4036141228,-0.1861474711
N,-2.3851611432,-0.2351788893,-0.3177042547
C,-2.0316697587,0.9032718202,-0.4901956531
H,-2.2658800448,-1.826382645,0.6922354369
H,-2.4437957443,1.8143207495,-0.8920375143
C,0.0812329313,0.690204492,-0.1507372596
C,0.2480483095,-0.7094801892,0.0644446494
C,0.4848373904,1.2581382661,-1.4115728961
N,0.7772861566,1.742058727,-2.4173474599
C,0.1567446131,1.584947505,0.9756309616
N,0.2027446513,2.317419597,1.8657680582
C,0.7248497684,-1.545773433,-0.9923450317
N,1.1265038491,-2.2218746075,-1.8373002791
C,0.3712197518,-1.2175180339,1.3956819734
N,0.4512454576,-1.6433406461,2.46652463

6 → 14a

Total Energy= -745.193557133 Hartree, NIMAG= 1

N,-1.8376245412,2.0341874061,0.2242995336
N,-1.0372923239,0.848525162,0.1426354105
C,0.2397799747,1.2421047336,0.1050923668
H,-2.7843768724,1.8037736477,-0.0637025711
C,0.3149606054,2.7931088191,-0.2460666551
C,-1.2066239932,3.002863224,-0.6856671508

C,0.6279009132,3.6432119688,0.9086560031
N,0.9014829346,4.2924981518,1.8202752477
C,1.2863746671,3.0535873499,-1.3080935082
N,2.0810575316,3.221058268,-2.1249686967
C,-1.7165655349,4.3609949817,-0.4614751645
N,-2.1434638968,5.4182096994,-0.3015120016
C,-1.3878166723,2.6491297677,-2.1146652906
N,-1.6050696314,2.3573400214,-3.2079686677
H,0.9339376565,0.6122670182,-0.4406650163
N,-1.1008994361,0.2059632416,2.4235785848
N,-0.0052939495,0.4775693837,2.5523606039
C,1.0992966738,0.9368320096,1.8816991214
H,1.5132465434,1.833355954,2.3370500736
H,1.8306641913,0.1410570616,1.7496995372

6 → 14b

Total Energy= -745.186615510 Hartree, NIMAG= 1

N,-1.2542535691,0.9031341578,0.5901474999
N,0.17603878,0.7822453382,0.3836693268
C,0.587353037,1.9138073024,-0.1858107428
H,-1.6466309299,-0.0341048635,0.5544146958
C,-0.6014715419,2.817572804,-0.6016516666
C,-1.7616876319,1.7204759604,-0.5265010274
C,-0.8710290734,3.9030200047,0.3569798727
N,-1.0831292497,4.7504467697,1.1077735393
C,-0.4249150963,3.388271056,-1.9393033322
N,-0.2501300092,3.8384418323,-2.9846257107
C,-3.0928219615,2.2571611271,-0.2176109183
N,-4.150719661,2.6466642259,0.0157437023
C,-1.8315652052,0.9549592586,-1.7939127816
N,-1.9037675481,0.2932836541,-2.7342091106
H,1.5114458763,1.9433581977,-0.7437181027
N,1.5052960144,2.9438658188,1.5075775962
N,1.3708123221,2.0662197893,2.257466268
C,0.8787835203,0.8154731666,2.2801075068
H,1.6364173576,0.0388057478,2.3054114291
H,0.0032686894,0.6778533121,2.9075642261

7 → 8

Total Energy= -596.380076631 Hartree, NIMAG= 1

N,1.7994089566,2.9876395543,1.281522292
N,1.3082853446,1.7013510484,1.3306490511
C,0.049465454,1.3505387679,1.2388478509
H,-0.1174383544,0.3184903568,1.5572683953
C,-1.0210849601,2.2538600535,0.8343569256
C,1.4707607723,3.6692425674,0.2186612622
C,-1.653051356,2.0058001973,-0.3960672599
N,-2.1325381076,1.7790415336,-1.4298970009
C,-1.6538074484,2.9112339196,1.9083732842

N,-2.104529157,3.4531067151,2.8328006356
C,1.115423337,3.1332916945,-1.0589662335
N,0.9377187193,2.7059838352,-2.1188756679
C,1.7141638458,5.0825776382,0.2672191491
N,1.9047660943,6.2202639134,0.2761411701
H,1.9756000295,1.0442240847,1.7296152759

7 → 9

Total Energy= -1043.99553250 Hartree, NIMAG= 1

N,0.5497074384,0.3966424977,-0.0509132767
N,1.2171275761,1.5204486183,0.088639605
C,0.7793763431,2.3205507276,1.1125243604
H,1.6223472235,1.9125453573,-0.7612104856
C,-0.8352085544,3.0424422492,0.1791480903
C,-1.492339753,1.905672533,-0.4532731605
C,-1.5644823186,3.8043289513,1.1692641258
N,-2.1017908168,4.4848648436,1.9288316456
C,-0.0975972483,3.918974653,-0.7150581208
N,0.568988781,4.570372914,-1.3943380321
C,-2.703403972,1.3964519793,0.0848700224
N,-3.7133869124,1.0063658894,0.4908509232
C,-1.2365223582,1.6346953164,-1.827742149
N,-1.0118961649,1.4488699552,-2.9467408409
H,1.3600934175,3.199160804,1.3725715757
C,0.3418965987,-0.0687302558,1.3481804314
C,0.4175083955,1.3117815622,2.2424181425
C,-0.8078767325,1.5514940053,3.0045558863
N,-1.7409997124,1.6716546874,3.6678857681
C,1.5419783965,1.2966586127,3.1880514426
N,2.4315637166,1.3381600052,3.918625984
C,1.4671350573,-0.9690533865,1.6614703682
N,2.3519425694,-1.6792869013,1.8604884312
C,-0.899936375,-0.82663208,1.4964326903
N,-1.8350361352,-1.4823539384,1.6366638627

1+10H⁺ → 6aH⁺

Total Energy= -596.674474172 Hartree, E+ZPE=-596.581569 NIMAG= 1

N,-1.2430199832,1.6557052725,1.2418351546
N,-2.0711811746,0.5990951919,1.2176544066
C,-1.9655548604,-0.4930197062,0.7301261201
H,-1.6564099404,2.4660447828,0.769832156
H,-2.5762733332,-1.3880771758,0.6657333387
C,-0.2252813489,-0.7954213134,-0.2290355964
C,0.5296808482,0.4202375243,-0.1051194354
C,0.1943577555,-1.9331082613,0.5580783435
N,0.4771270388,-2.8443694725,1.2060421216
C,-0.8056338126,-1.1004809349,-1.5174910952
N,-1.317974351,-1.3496621051,-2.5202996772
C,1.5049040409,0.5591947507,0.9182396105
N,2.2971810296,0.6729265588,1.7527951191

C,0.5119724462,1.3862964793,-1.1463025299
N,0.4958256746,2.1734795116,-1.9931801894
H,-0.9681556194,1.8937337271,2.2000230628

6bH⁺ → 8aH⁺

Total Energy= -596.687636653 Hartree, NIMAG= 1

N,1.7012221051,2.9040340144,1.4384292435
N,1.3011190528,1.5512913046,1.4434850863
C,0.0656082836,1.2939714885,1.1020261709
H,1.4139355485,3.3901949435,2.293252523
H,-0.2150296735,0.2476297842,0.9931255748
C,-0.8284188512,2.3889690544,0.768565235
C,1.2181661121,3.5229330417,0.2795630151
C,-1.7697072951,2.1539147501,-0.2684189212
N,-2.4853167445,1.9565827869,-1.1555116773
C,-1.1243470428,3.3071472046,1.8154909735
N,-1.2598044622,4.0416519314,2.6993588457
C,1.5130109503,2.950500088,-0.9891608026
N,1.7268900982,2.4832612125,-2.0249775306
C,0.977329484,4.9161385001,0.3405259956
N,0.7584312061,6.0509829135,0.4010879097
H,2.0139899385,0.8476378314,1.6280600084