

Supporting Information

A Computational Examination on the Structure, Spin State Energetics and Spectroscopic Parameters of High-valent Fe^{IV}=NTs Species

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In order to find out the transition state for the formation of species $[(N_4Py)Fe^{IV}=NTs]^{2+}$ from its precursor complex $[Fe^{II}(N_4Py)(PhINTs)]^{2+}$, the relaxed potential energy surface scan was performed with the variation of the N-I bond distances from the 3.74 Å to 4.54 Å. There is no distinct minima. So the formation of $[(N_4Py)Fe^{IV}=NTs]^{2+}$ is likely to be a barrierless process.

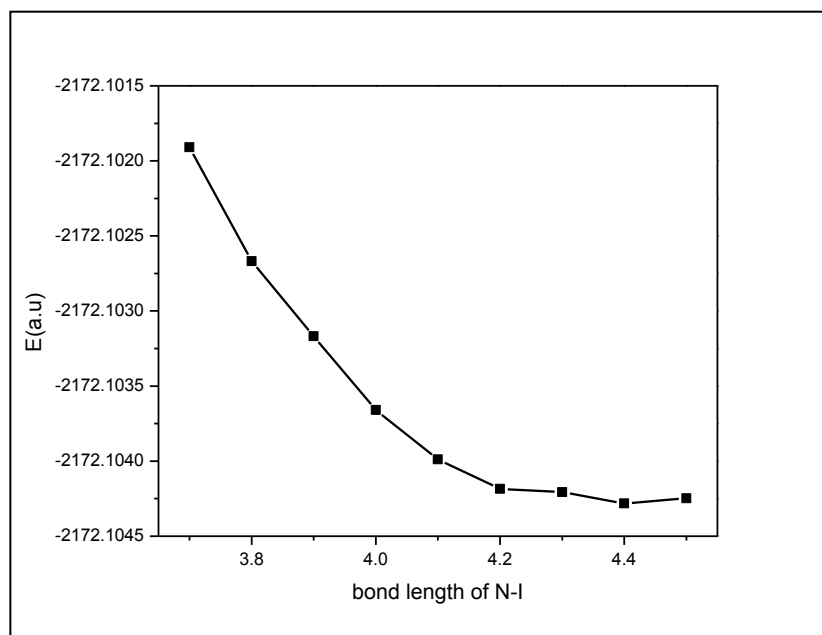


Figure S1. A relaxed scan of N-I bond distance performed for species **1**.

Table S1. B3LYP Optimized cartesian coordinates of species **1** in all the eight different spin state configurations

${}^7\mathbf{1}_{(\text{HS,T})}$				${}^3\mathbf{1}_{(\text{HS,T})}$			
26	0.103687000	0.291604000	-0.000254000	26	-0.121542000	0.030713000	0.261255000
7	-0.365026000	2.206262000	-0.001887000	7	-0.024749000	-0.350090000	2.081165000
7	0.704911000	-1.885240000	0.001610000	7	-0.366387000	0.775904000	-1.671257000
6	-0.667335000	-2.490354000	0.002089000	6	1.087931000	0.914233000	-2.066170000
6	-1.414529000	-1.978840000	-1.224061000	6	1.724978000	-0.460415000	-1.921136000
6	-2.225149000	-2.775869000	-2.022191000	6	2.638798000	-0.977452000	-2.829535000
6	-2.931027000	-2.174053000	-3.070636000	6	3.193682000	-2.237536000	-2.578654000
6	-2.802246000	-0.802458000	-3.277352000	6	2.798249000	-2.933256000	-1.440083000
6	-1.963663000	-0.068387000	-2.438145000	6	1.870157000	-2.351677000	-0.575354000
7	-1.281935000	-0.649937000	-1.435970000	7	1.353103000	-1.131397000	-0.801832000
6	1.482997000	-2.164087000	1.240659000	6	-1.092207000	2.087572000	-1.587644000
6	2.180300000	-0.935911000	1.794901000	6	-2.043345000	2.125525000	-0.413157000
6	3.278737000	-1.049929000	2.645279000	6	-3.158877000	2.957622000	-0.383161000
6	3.824982000	0.103385000	3.209557000	6	-3.930681000	3.010936000	0.779961000
6	3.262568000	1.343878000	2.902048000	6	-3.567128000	2.228082000	1.876427000
6	2.179253000	1.383974000	2.033134000	6	-2.447351000	1.409067000	1.769648000
7	1.648334000	0.267385000	1.493575000	7	-1.705129000	1.361018000	0.650558000
6	-1.414597000	-1.976751000	1.227322000	6	1.766560000	1.845044000	-1.073128000
6	-2.225257000	-2.772418000	2.026768000	6	2.724590000	2.792454000	-1.411137000
6	-2.931204000	-2.168813000	3.074137000	6	3.334704000	3.515598000	-0.379427000
6	-2.802463000	-0.796861000	3.278499000	6	2.963216000	3.266863000	0.941058000
6	-1.963829000	-0.064224000	2.438093000	6	1.986077000	2.304930000	1.194710000
7	-1.282022000	-0.647489000	1.436968000	7	1.402320000	1.611909000	0.205397000
6	1.483048000	-2.166192000	-1.236930000	6	-1.104153000	-0.209612000	-2.534982000
6	2.180382000	-0.938961000	-1.793233000	6	-1.969505000	-1.160961000	-1.736093000
6	3.278858000	-1.054430000	-2.643364000	6	-3.027146000	-1.855581000	-2.318243000
6	3.825147000	0.097923000	-3.209562000	6	-3.707074000	-2.807693000	-1.556268000
6	3.262755000	1.338944000	-2.904150000	6	-3.308958000	-3.036661000	-0.238109000
6	2.179404000	1.380523000	-2.035352000	6	-2.254685000	-2.292081000	0.280465000
7	1.648429000	0.264851000	-1.493954000	7	-1.604416000	-1.369821000	-0.455306000
1	-0.634880000	-3.586536000	0.003021000	1	1.186544000	1.288253000	-3.091380000
1	-2.308969000	-3.841295000	-1.831901000	1	2.911617000	-0.414391000	-3.716553000
1	-3.573057000	-2.772028000	-3.710126000	1	3.913542000	-2.665528000	-3.269489000
1	-3.342546000	-0.297282000	-4.070532000	1	3.195122000	-3.916487000	-1.212040000
1	-1.854754000	1.004869000	-2.542312000	1	1.528911000	-2.863932000	0.315807000
1	0.794660000	-2.535403000	2.006793000	1	-0.350027000	2.880958000	-1.459255000
1	2.210478000	-2.964516000	1.065599000	1	-1.614715000	2.292584000	-2.527467000
1	3.695985000	-2.027417000	2.867162000	1	-3.416853000	3.558908000	-1.249442000
1	4.678382000	0.034058000	3.877147000	1	-4.803631000	3.654842000	0.826237000
1	3.657451000	2.263478000	3.319934000	1	-4.140170000	2.244734000	2.797084000
1	1.712481000	2.320073000	1.745255000	1	-2.111592000	0.772265000	2.581510000
1	-2.309061000	-3.838169000	1.838294000	1	2.993674000	2.963966000	-2.448629000
1	-3.573275000	-2.765696000	3.714606000	1	4.088991000	4.261896000	-0.608978000
1	-3.342829000	-0.290329000	4.070770000	1	3.418214000	3.805120000	1.765360000
1	-1.854941000	1.009212000	2.540423000	1	1.661895000	2.076025000	2.203670000
1	0.794739000	-2.538813000	-2.002455000	1	-0.364610000	-0.798872000	-3.085817000
1	2.210517000	-2.966327000	-1.060478000	1	-1.697946000	0.325235000	-3.282655000
1	3.696102000	-2.032297000	-2.863577000	1	-3.307925000	-1.664367000	-3.349480000
1	4.678578000	0.027457000	-3.876993000	1	-4.532947000	-3.364309000	-1.988477000
1	3.657678000	2.257833000	-3.323559000	1	-3.804888000	-3.775748000	0.381648000
1	1.712641000	2.317113000	-1.749062000	1	-1.900737000	-2.434169000	1.295527000
16	-1.699520000	3.152005000	-0.002657000	16	1.100023000	-1.424156000	2.705494000
8	-2.385666000	2.874469000	-1.278618000	8	0.596071000	-2.748547000	2.279428000
8	-2.385572000	2.876637000	1.273824000	8	2.448722000	-0.987616000	2.321841000
6	-1.142548000	4.855653000	-0.004127000	6	0.907736000	-1.276784000	4.483766000
1	-0.557469000	5.036219000	-0.907791000	1	-0.119495000	-1.526877000	4.753682000
1	-2.040522000	5.480150000	-0.004618000	1	1.604547000	-1.993910000	4.927510000
1	-0.557391000	5.037751000	0.899179000	1	1.167260000	-0.260905000	4.787341000

$^5\mathbf{1}_{(IS,T)}$

26	-0.176502000	0.022108000	0.276964000
7	0.044106000	-0.570983000	1.929512000
7	-0.486047000	0.832052000	-1.619659000
6	0.910293000	1.337895000	-1.916938000
6	1.855052000	0.147689000	-1.958435000
6	2.866226000	-0.011795000	-2.897224000
6	3.704826000	-1.127153000	-2.780280000
6	3.489545000	-2.030937000	-1.743007000
6	2.442563000	-1.801242000	-0.846632000
7	1.646629000	-0.725570000	-0.952812000
6	-1.473598000	1.961864000	-1.565476000
6	-2.512617000	1.751013000	-0.488511000
6	-3.782167000	2.320484000	-0.541108000
6	-4.633933000	2.158508000	0.554214000
6	-4.192971000	1.430264000	1.660777000
6	-2.915451000	0.879201000	1.630640000
7	-2.095546000	1.040797000	0.576990000
6	1.325878000	2.209760000	-0.740293000
6	2.082743000	3.368116000	-0.851197000
6	2.482322000	4.023122000	0.319611000
6	2.111203000	3.499223000	1.556901000
6	1.346688000	2.335422000	1.593422000
7	0.961738000	1.710329000	0.468258000
6	-0.952226000	-0.270951000	-2.528256000
6	-1.527492000	-1.447861000	-1.769257000
6	-2.316011000	-2.401900000	-2.406430000
6	-2.746831000	-3.519198000	-1.689089000
6	-2.379002000	-3.651169000	-0.349621000
6	-1.603199000	-2.656053000	0.231895000
7	-1.189930000	-1.575857000	-0.462189000
1	0.936512000	1.899203000	-2.857491000
1	3.001330000	0.708196000	-3.698483000
1	4.508436000	-1.283191000	-3.493364000
1	4.117458000	-2.906721000	-1.619511000
1	2.235347000	-2.478352000	-0.024870000
1	-0.922645000	2.879787000	-1.337420000
1	-1.934374000	2.105316000	-2.548408000
1	-4.097460000	2.884407000	-1.413604000
1	-5.628185000	2.594415000	0.541409000
1	-4.826241000	1.285691000	2.529397000
1	-2.520085000	0.293437000	2.453924000
1	2.359641000	3.752495000	-1.827657000
1	3.076183000	4.930035000	0.260440000
1	2.408514000	3.976284000	2.484234000
1	1.034097000	1.882034000	2.525478000
1	-0.098868000	-0.619541000	-3.117478000
1	-1.685762000	0.118050000	-3.241082000
1	-2.582486000	-2.276774000	-3.451412000
1	-3.359103000	-4.274893000	-2.171537000
1	-2.685491000	-4.507951000	0.240313000
1	-1.283465000	-2.710151000	1.264177000
16	1.313195000	-1.446870000	2.589109000
8	1.258182000	-2.787936000	1.981154000
8	2.524269000	-0.627745000	2.459663000
6	0.868057000	-1.582858000	4.324206000
1	-0.081660000	-2.113114000	4.411804000
1	1.668572000	-2.158427000	4.798129000
1	0.812012000	-0.583731000	4.759502000

$^1\mathbf{1}_{(IS,T)}$

26	-0.141154000	0.032363000	0.135249000
7	0.104627000	-0.265251000	1.917873000
7	-0.495513000	0.627583000	-1.796906000
6	0.914178000	1.075173000	-2.121796000
6	1.805271000	-0.133256000	-1.903825000
6	2.851634000	-0.498474000	-2.737548000
6	3.599965000	-1.634932000	-2.409180000
6	3.250593000	-2.365853000	-1.278232000
6	2.179023000	-1.941479000	-0.490594000
7	1.481241000	-0.830839000	-0.786223000
6	-1.491402000	1.746066000	-1.800636000
6	-2.368267000	1.694618000	-0.571437000
6	-3.602754000	2.335499000	-0.510895000
6	-4.320723000	2.312146000	0.685532000
6	-3.780795000	1.643709000	1.784542000
6	-2.547826000	1.014101000	1.649686000
7	-1.855873000	1.040973000	0.495139000
6	1.291933000	2.111288000	-1.077562000
6	2.023417000	3.267134000	-1.314068000
6	2.353356000	4.079975000	-0.223619000
6	1.933019000	3.711689000	1.053537000
6	1.193068000	2.541236000	1.211438000
7	0.885575000	1.760400000	0.164537000
6	-0.962984000	-0.559503000	-2.583918000
6	-1.560295000	-1.617107000	-1.681248000
6	-2.378994000	-2.627551000	-2.176998000
6	-2.832935000	-3.622164000	-1.309758000
6	-2.455057000	-3.572869000	0.031576000
6	-1.646846000	-2.528151000	0.466553000
7	-1.209156000	-1.568708000	-0.374253000
1	0.998872000	1.468029000	-3.140619000
1	3.075287000	0.083200000	-3.626206000
1	4.429478000	-1.944625000	-3.037227000
1	3.793467000	-3.260555000	-0.993725000
1	1.873400000	-2.492171000	0.389982000
1	-0.947419000	2.695840000	-1.808883000
1	-2.090162000	1.720761000	-2.716281000
1	-3.992668000	2.847648000	-1.385147000
1	-5.285076000	2.805758000	0.755403000
1	-4.304359000	1.601630000	2.733467000
1	-2.076656000	0.475599000	2.463070000
1	2.330972000	3.528949000	-2.321424000
1	2.928260000	4.988321000	-0.374651000
1	2.171912000	4.316370000	1.921504000
1	0.837209000	2.209124000	2.179131000
1	-0.106789000	-0.987154000	-3.114546000
1	-1.681100000	-0.248465000	-3.348645000
1	-2.651197000	-2.637996000	-3.227953000
1	-3.469841000	-4.420612000	-1.677578000
1	-2.777492000	-4.328703000	0.739293000
1	-1.316113000	-2.454845000	1.493947000
16	1.307531000	-1.130307000	2.673251000
8	1.012905000	-2.546003000	2.350216000
8	2.616251000	-0.557554000	2.333786000
6	1.005531000	-0.882747000	4.426120000
1	0.005660000	-1.242928000	4.673124000
1	1.762669000	-1.468626000	4.954613000
1	1.118991000	0.176986000	4.661639000

³1_(LS,T)

26	-0.131664000	0.040514000	0.129238000
7	0.152057000	-0.447381000	1.798117000
7	-0.476797000	0.755406000	-1.763036000
6	0.952844000	1.111147000	-2.107643000
6	1.749786000	-0.169759000	-1.961657000
6	2.751352000	-0.590159000	-2.823622000
6	3.412039000	-1.791458000	-2.541006000
6	3.025533000	-2.527882000	-1.424959000
6	2.001297000	-2.048204000	-0.607493000
7	1.390071000	-0.877639000	-0.862557000
6	-1.382708000	1.946377000	-1.693959000
6	-2.252120000	1.883016000	-0.461794000
6	-3.434904000	2.607358000	-0.338935000
6	-4.140322000	2.553867000	0.863606000
6	-3.641977000	1.770804000	1.905634000
6	-2.460624000	1.064559000	1.709441000
7	-1.778795000	1.121816000	0.549401000
6	1.438627000	2.058772000	-1.022978000
6	2.248405000	3.167348000	-1.228905000
6	2.681318000	3.890655000	-0.111664000
6	2.284589000	3.480912000	1.159978000
6	1.462306000	2.362004000	1.285012000
7	1.051261000	1.667528000	0.213590000
6	-1.062666000	-0.354136000	-2.584598000
6	-1.727426000	-1.400319000	-1.713691000
6	-2.637906000	-2.315372000	-2.235498000
6	-3.139922000	-3.320882000	-1.408271000
6	-2.713363000	-3.381115000	-0.081872000
6	-1.814068000	-2.425974000	0.379750000
7	-1.336620000	-1.450634000	-0.418574000
1	1.047043000	1.545392000	-3.108697000
1	3.008149000	-0.000331000	-3.697752000
1	4.205063000	-2.145773000	-3.192185000
1	3.503794000	-3.468673000	-1.175321000
1	1.669481000	-2.600668000	0.263207000
1	-0.767390000	2.850446000	-1.648639000
1	-1.984812000	2.022037000	-2.604822000
1	-3.794149000	3.206073000	-1.170210000
1	-5.063817000	3.111946000	0.982557000
1	-4.158768000	1.700168000	2.856560000
1	-2.027920000	0.435303000	2.477503000
1	2.537832000	3.460883000	-2.232941000
1	3.318291000	4.760568000	-0.237833000
1	2.605478000	4.013088000	2.048744000
1	1.124420000	2.001479000	2.248480000
1	-0.257531000	-0.828891000	-3.153992000
1	-1.767205000	0.048991000	-3.318422000
1	-2.941555000	-2.246900000	-3.275611000
1	-3.848448000	-4.046131000	-1.796204000
1	-3.067747000	-4.153183000	0.592307000
1	-1.443254000	-2.437085000	1.396650000
16	1.278715000	-1.343631000	2.642194000
8	0.990798000	-2.757027000	2.334060000
8	2.603112000	-0.782066000	2.348590000
6	0.876838000	-1.046286000	4.366793000
1	-0.141775000	-1.385605000	4.562896000
1	1.588827000	-1.636491000	4.951012000
1	0.998706000	0.015535000	4.587598000

⁵1_(HS,S)

26	-0.176004000	0.021388000	0.276554000
7	0.044046000	-0.574629000	1.928181000
7	-0.485205000	0.834822000	-1.618486000
6	0.911343000	1.340709000	-1.914827000
6	1.855777000	0.150310000	-1.957902000
6	2.867574000	-0.007730000	-2.896273000
6	3.705964000	-1.123360000	-2.780611000
6	3.489887000	-2.028874000	-1.745004000
6	2.442328000	-1.800580000	-0.848963000
7	1.646604000	-0.724586000	-0.953869000
6	-1.472436000	1.964858000	-1.562631000
6	-2.511321000	1.752985000	-0.485725000
6	-3.780564000	2.323253000	-0.537200000
6	-4.632159000	2.160164000	0.558091000
6	-4.191355000	1.430059000	1.663492000
6	-2.914157000	0.878313000	1.632239000
7	-2.094421000	1.040981000	0.578626000
6	1.327041000	2.210546000	-0.736708000
6	2.083959000	3.369055000	-0.845582000
6	2.483587000	4.021977000	0.326377000
6	2.112488000	3.495877000	1.562738000
6	1.347931000	2.332030000	1.597209000
7	0.962929000	1.708941000	0.470953000
6	-0.951720000	-0.266575000	-2.528918000
6	-1.528919000	-1.443729000	-1.771801000
6	-2.319057000	-2.395488000	-2.410339000
6	-2.751261000	-3.513412000	-1.694776000
6	-2.383185000	-3.648240000	-0.355672000
6	-1.605790000	-2.655219000	0.227326000
7	-1.191159000	-1.574451000	-0.465011000
1	0.937868000	1.903463000	-2.854507000
1	3.003290000	0.713584000	-3.696234000
1	4.510077000	-1.278267000	-3.493375000
1	4.117621000	-2.904934000	-1.622579000
1	2.234380000	-2.479138000	-0.028605000
1	-0.921155000	2.882327000	-1.333531000
1	-1.933370000	2.109661000	-2.545287000
1	-4.095753000	2.888630000	-1.408791000
1	-5.626163000	2.596665000	0.546153000
1	-4.824507000	1.284590000	2.532049000
1	-2.518917000	0.291109000	2.454556000
1	2.360867000	3.755133000	-1.821370000
1	3.077458000	4.928986000	0.268797000
1	2.409819000	3.971265000	2.490925000
1	1.035351000	1.877038000	2.528489000
1	-0.098109000	-0.615436000	-3.117594000
1	-1.684042000	0.124098000	-3.242077000
1	-2.585684000	-2.268194000	-3.455020000
1	-3.364802000	-4.267367000	-2.178332000
1	-2.690717000	-4.505596000	0.232883000
1	-1.285855000	-2.711514000	1.259424000
16	1.312033000	-1.453202000	2.586317000
8	1.255531000	-2.793206000	1.976140000
8	2.524101000	-0.635274000	2.458324000
6	0.866573000	-1.591570000	4.321137000
1	-0.083973000	-2.120565000	4.407725000
1	1.666181000	-2.169151000	4.794169000
1	0.811924000	-0.593083000	4.758137000

$^3\mathbf{1}_{(IS,S)}$

26	-0.131650000	0.040632000	0.129303000
7	0.152080000	-0.447725000	1.798041000
7	-0.476515000	0.755996000	-1.762861000
6	0.953179000	1.111510000	-2.107365000
6	1.749861000	-0.169616000	-1.961607000
6	2.751367000	-0.589982000	-2.823666000
6	3.411768000	-1.791514000	-2.541375000
6	3.025028000	-2.528161000	-1.425548000
6	2.000890000	-2.048463000	-0.607974000
7	1.389944000	-0.877682000	-0.862719000
6	-1.382212000	1.947119000	-1.693423000
6	-2.251843000	1.883422000	-0.461444000
6	-3.434750000	2.607549000	-0.338626000
6	-4.140324000	2.553778000	0.863818000
6	-3.641989000	1.770661000	1.905801000
6	-2.460519000	1.064589000	1.709650000
7	-1.778549000	1.122101000	0.549709000
6	1.439126000	2.058837000	-1.022509000
6	2.249176000	3.167267000	-1.228186000
6	2.682165000	3.890264000	-0.110784000
6	2.285230000	3.480416000	1.160766000
6	1.462684000	2.361674000	1.285546000
7	1.051591000	1.667481000	0.213950000
6	-1.062649000	-0.353210000	-2.584692000
6	-1.727558000	-1.399529000	-1.714056000
6	-2.638108000	-2.314365000	-2.236113000
6	-3.140324000	-3.319969000	-1.409126000
6	-2.713893000	-3.380512000	-0.082701000
6	-1.814509000	-2.425578000	0.379182000
7	-1.336869000	-1.450144000	-0.418914000
1	1.047493000	1.545921000	-3.108334000
1	3.008276000	0.000049000	-3.697625000
1	4.204731000	-2.145838000	-3.192621000
1	3.503028000	-3.469147000	-1.176149000
1	1.668926000	-2.601120000	0.262547000
1	-0.766698000	2.851029000	-1.647611000
1	-1.984136000	2.023316000	-2.604365000
1	-3.793964000	3.206345000	-1.169857000
1	-5.063916000	3.111706000	0.982721000
1	-4.158860000	1.699845000	2.856671000
1	-2.027831000	0.435289000	2.477685000
1	2.538736000	3.460941000	-2.232142000
1	3.319364000	4.760042000	-0.236756000
1	2.606154000	4.012404000	2.049632000
1	1.124605000	2.001070000	2.248917000
1	-0.257653000	-0.827941000	-3.154297000
1	-1.767185000	0.050274000	-3.318324000
1	-2.941650000	-2.245655000	-3.276243000
1	-3.848906000	-4.045053000	-1.797268000
1	-3.068421000	-4.152665000	0.591304000
1	-1.443745000	-2.436951000	1.396092000
16	1.278385000	-1.344529000	2.641988000
8	0.989973000	-2.757731000	2.333396000
8	2.603082000	-0.783449000	2.348792000
6	0.876463000	-1.047531000	4.366643000
1	-0.142233000	-1.386668000	4.562604000
1	1.588291000	-1.638056000	4.950728000
1	0.998573000	0.014192000	4.587765000

$^1\mathbf{1}_{(LS,S)}$

26	-0.096114000	-0.000006000	0.150815000
7	-0.015400000	-0.000115000	1.876693000
7	-0.396668000	0.000038000	-1.902633000
6	1.056334000	-0.000003000	-2.317212000
6	1.676503000	-1.224580000	-1.679084000
6	2.569364000	-2.078360000	-2.309988000
6	3.049589000	-3.184433000	-1.599991000
6	2.592881000	-3.400711000	-0.303178000
6	1.691559000	-2.499470000	0.263964000
7	1.262303000	-1.412930000	-0.402141000
6	-1.137241000	1.241267000	-2.292480000
6	-1.954940000	1.769613000	-1.137424000
6	-3.012380000	2.656712000	-1.326935000
6	-3.682218000	3.164119000	-0.213984000
6	-3.279341000	2.757797000	1.058566000
6	-2.225597000	1.858909000	1.174010000
7	-1.570910000	1.378615000	0.098303000
6	1.676593000	1.224508000	-1.679045000
6	2.569507000	2.078241000	-2.309936000
6	3.049835000	3.184251000	-1.599912000
6	2.593172000	3.400514000	-0.303081000
6	1.691783000	2.499330000	0.264048000
7	1.262427000	1.412844000	-0.402087000
6	-1.137323000	-1.241123000	-2.292535000
6	-1.955082000	-1.769443000	-1.137512000
6	-3.012602000	-2.656437000	-1.327073000
6	-3.682516000	-3.163811000	-0.214154000
6	-3.279635000	-2.757556000	1.058417000
6	-2.225806000	-1.858774000	1.173909000
7	-1.571039000	-1.378520000	0.098234000
1	1.180736000	0.000010000	-3.405524000
1	2.875766000	-1.892137000	-3.334365000
1	3.753864000	-3.868957000	-2.062464000
1	2.926141000	-4.253079000	0.278614000
1	1.331278000	-2.627864000	1.273229000
1	-0.411841000	2.003323000	-2.593418000
1	-1.769491000	1.052046000	-3.165682000
1	-3.302252000	2.947386000	-2.332003000
1	-4.506670000	3.859078000	-0.339543000
1	-3.774746000	3.122590000	1.951829000
1	-1.883426000	1.490373000	2.133332000
1	2.875874000	1.892028000	-3.334326000
1	3.754157000	3.868734000	-2.062375000
1	2.926515000	4.252831000	0.278739000
1	1.331545000	2.627702000	1.273332000
1	-0.411973000	-2.003223000	-2.593485000
1	-1.769543000	-1.051828000	-3.165742000
1	-3.302480000	-2.947050000	-2.332157000
1	-4.507034000	-3.858685000	-0.339752000
1	-3.775104000	-3.122318000	1.951657000
1	-1.883632000	-1.490281000	2.133247000
16	1.283459000	0.000019000	2.965199000
8	1.999171000	-1.275460000	2.821247000
8	1.997373000	1.276665000	2.822618000
6	0.462367000	-0.001397000	4.563474000
1	-0.143297000	-0.904330000	4.657301000
1	1.257576000	-0.001262000	5.314214000
1	-0.144520000	0.900612000	4.658259000

Table S2. Optimized geometrical parameters of eight different electronic spin states of species **1** obtained from B3LYP* level (values in the parenthesis obtained from BP86 level). Bond lengths are given in Å and bond angles are given in degrees.

$M_{\text{Fe,N}}$	Fe-N1	Fe-N2	Fe-N3	Fe-N3'	Fe-N4	Fe-N4'	N2-Fe-N1
$^7\mathbf{1}_{\text{(HS,T)}}$	1.98(1.96)	2.28(2.28)	2.16(2.15)	2.20(2.21)	2.16(2.15)	2.20(2.21)	177.4(178.4)
$^3\mathbf{1}_{\text{(HS,T)}}$	1.85(1.74)	2.10(2.06)	2.18(1.97)	2.16(2.01)	2.12(1.99)	2.22(1.99)	169.9(175.9)
$^5\mathbf{1}_{\text{(IS,T)}}$	1.76(1.74)	2.09(2.09)	2.05(2.03)	2.34(2.34)	2.20(2.18)	2.06(2.04)	176.9(177.4)
$^1\mathbf{1}_{\text{(IS,T)}}$	1.80(1.73)	2.07(2.07)	2.00(1.98)	2.07(2.01)	2.03(2.01)	2.03(1.99)	172.6(173.4)
$^3\mathbf{1}_{\text{(LS,T)}}$	1.80(1.74)	2.06(2.06)	2.00(1.99)	2.05(1.99)	2.03(1.97)	2.03(2.01)	176.1(175.9)
$^5\mathbf{1}_{\text{(HS,S)}}$	1.76(1.74)	2.10(2.09)	2.05(2.18)	2.34(2.04)	2.20(2.03)	2.20(2.34)	176.7(177.4)
$^3\mathbf{1}_{\text{(IS,S)}}$	1.76(1.74)	2.06(2.06)	2.01(1.97)	2.05(2.07)	2.03(1.99)	2.03(1.99)	176.1(175.9)
$^1\mathbf{1}_{\text{(LS,S)}}$	1.73(1.72)	2.08(2.07)	2.00(2.02)	2.06(1.99)	2.05(1.97)	2.05(2.01)	172.2(166.9)

Table S3. Calculated Mössbauer isomer shift and G, A and D tensor values of species **1** along with experimental data in the parenthesis.

	g_x, g_y, g_z *	A_x, A_y, A_z *	ΔE_q ** (mm/s)	η **	δ ** (mm/s)	$D(\text{cm}^{-1})$ *	E/D *
$^7\mathbf{1}_{\text{(HS,T)}}$	2.008,2.012,2.012	-22.9,-21.9,-17.9	-0.15	0.47	-4.6	7.07	0.05
$^5\mathbf{1}_{\text{(HS,S)}}$	2.008,2.011,2.023	-24.2,-34.7,3.4	1.89	0.66	-4.8	5.09	0.02
$^3\mathbf{1}_{\text{(IS,S)}}$	2.005, 2.009,2.015	-9.6,-19.5,6.1	-0.19	0.43	-4.5	4.71	0.09
Exp	-	-20,-21,-3	+0.98	-0.7	0.02	29	0.23

* Computed using BP86 level; ** Computed using B3LYP level

Table S4. Solvent optimized geometrical parameters of eight different electronic spin states obtained from B3LYP level. Bond lengths are given in Å and bond angles are given in degrees.

$M_{\text{Fe,N}}$	Fe-N1	Fe-N2	Fe-N3	Fe-N3'	Fe-N4	Fe-N4'	N2-Fe-N1	ΔE^a	S(Fe)	S(N)
$^7\mathbf{1}_{\text{(HS,T)}}$	1.98	2.25	2.20	2.18	2.15	2.14	175.2	54.6	4.00	1.14
$^3\mathbf{1}_{\text{(HS,T)}}$	1.87	2.07	2.09	2.22	2.17	2.06	170.9	55.7	2.86	-0.89
$^5\mathbf{1}_{\text{(IS,T)}}$	1.77	2.07	2.30	2.03	2.02	2.18	177.1	34.8	2.81	0.84
$^1\mathbf{1}_{\text{(IS,T)}}$	1.82	2.05	2.06	2.01	1.99	2.02	172.1	25.5	0.93	-0.74
$^3\mathbf{1}_{\text{(LS,T)}}$	1.75	2.03	2.00	1.99	2.01	2.02	177.6	0	0.95	0.96
$^5\mathbf{1}_{\text{(HS,S)}}$	1.77	2.07	2.30	2.03	2.02	2.18	177.1	34.9	2.81	0.84
$^3\mathbf{1}_{\text{(IS,S)}}$	1.75	2.04	2.02	2.01	1.99	2.00	177.6	2.3	0.94	0.95
$^1\mathbf{1}_{\text{(LS,S)}}$	1.71	2.06	2.01	2.03	2.02	1.99	173.9	52.9	0	0

From the above table, the geometrical parameters, Mulliken spin densities and relative energies, it is apparent that the solvation barely affects the conclusion drawn from gas phase energetics.