Supporting Information

of

Selective Separation and Complexation of Trivalent Actinides and Lanthanides by An Unsymmetric Pyridine-derived Triazinyl and Amide Extractant

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Contents

^{1}H·NMR·spectra·of synthetic intermediates·and·ligand

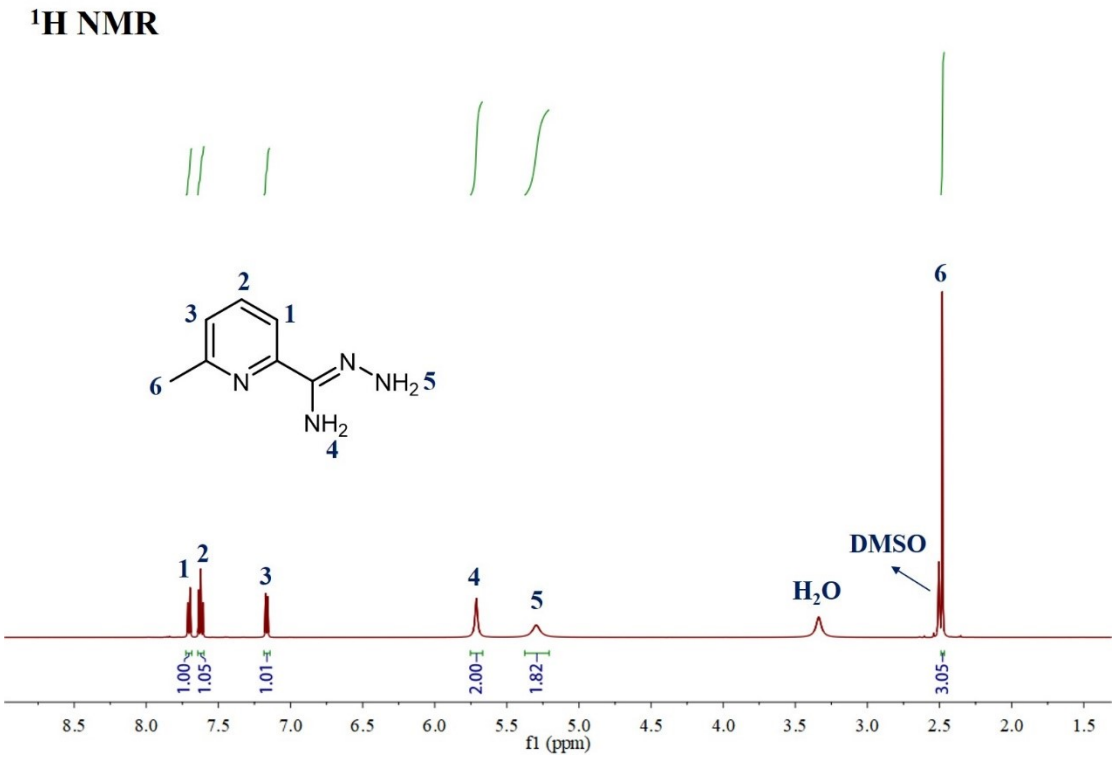

Figure S1. ^{1}H NMR of (Z)-6-methylpicolinohydrazonamide

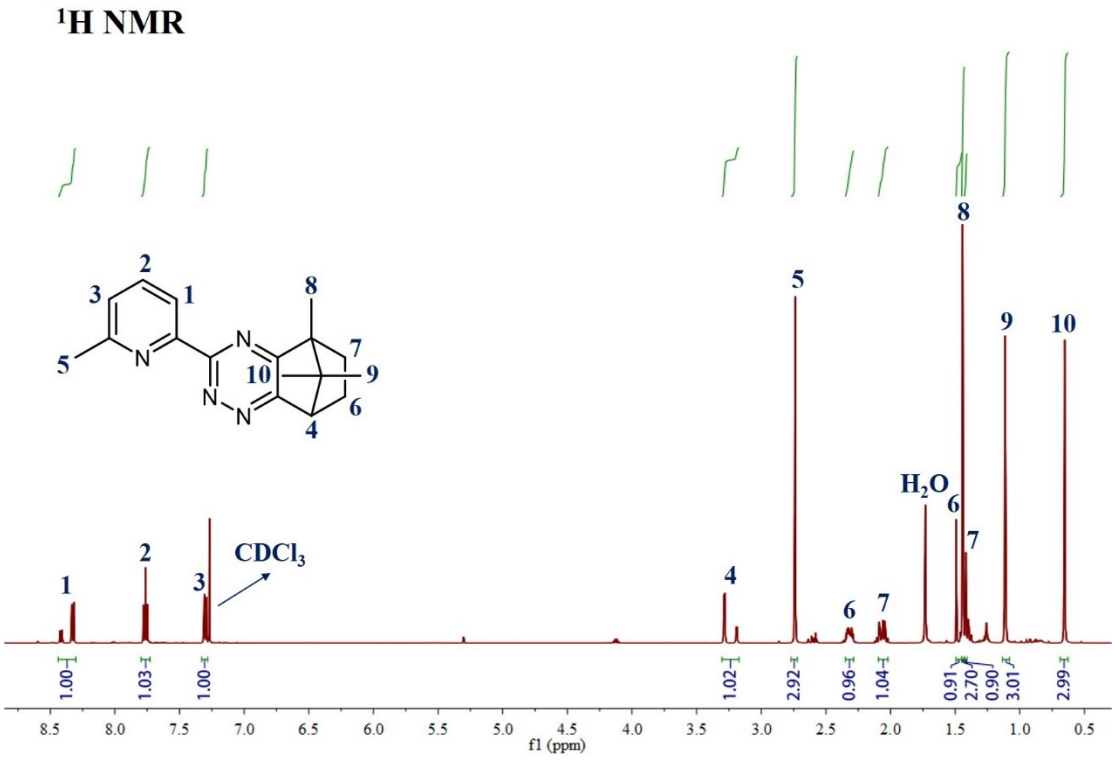

Figure S2. ^{1}H NMR of 5,9,9-trimethyl-3-(6-methylpyridin-2-yl)-5,6,7,8-tetrahydro-5,8-methanobenzo[e][1,2,4]triazine

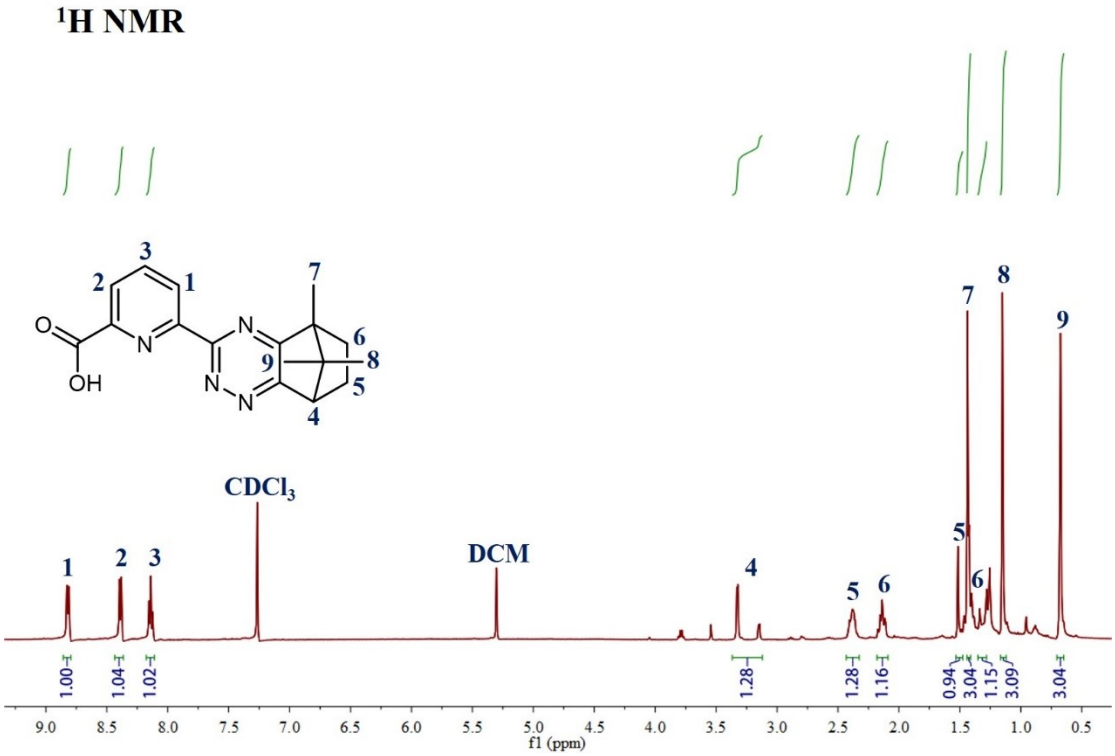

Figure S3. [1]H NMR of 6-(5,9,9-trimethyl-5,6,7,8-tetrahydro-5,8-methanobenzo[e][1,2,4]triazin-3-yl)picolinic acid

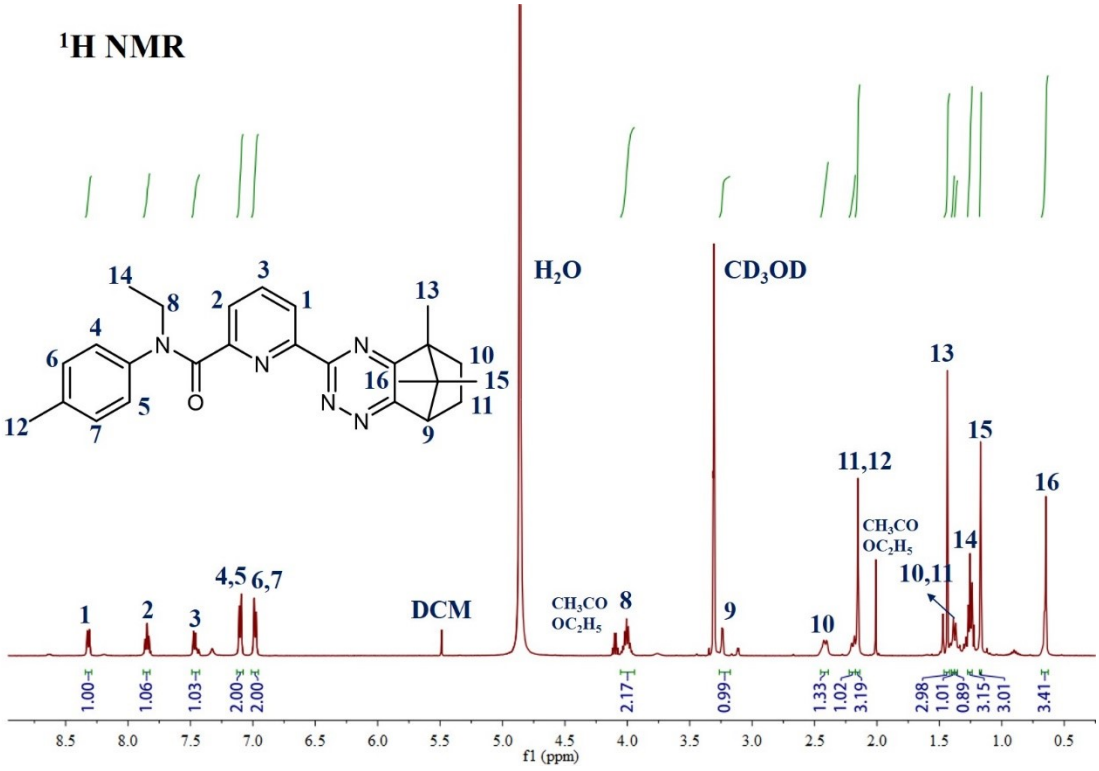

Figure S4. [1]H NMR of Et-Tol-CA-ATP ligand

HR-ESI-MS

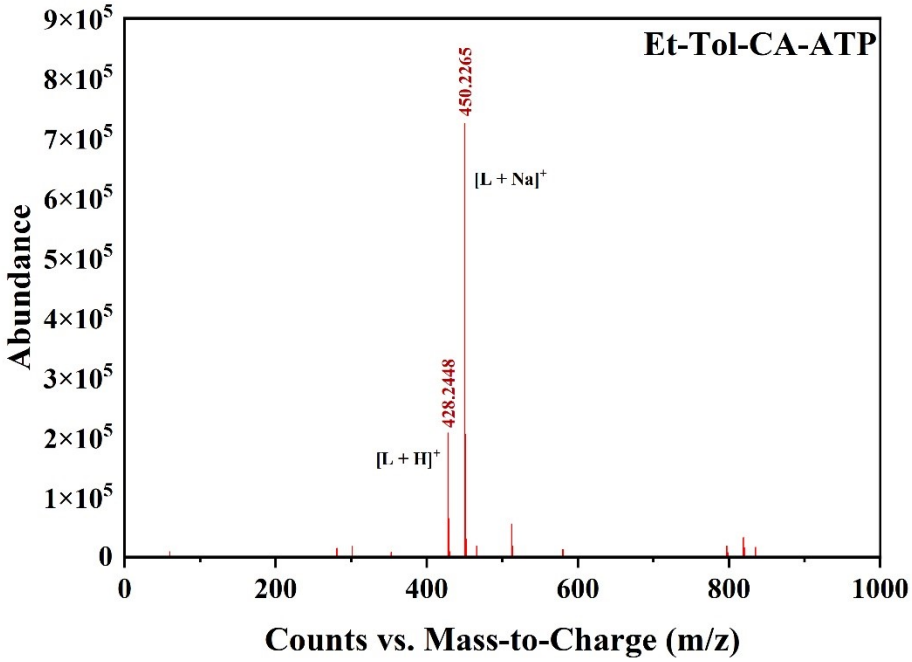

Figure S5. HR-ESI-MS of Et-Tol-CA-ATP ligand

Table S1. The simulated and experimental m/z values of HR-ESI-MS for the samples of Et-Tol-CA-ATP complexation with Eu(III) in the acetonitrile with M/L = 1:1 and 1:3.

Complex	Stimulated m/z	Measured m/z
$[L + H]^+$	428.2450	428.2516
$[L + Na]^+$	450.2270	450.2286
$[2L + Na]^+$	877.4642	877.4671
$[Eu(L)_2(NO_3)_2]^+$	1131.3708	1131.3718

Molar absorptivity and ligand absorption in UV-Vis titration

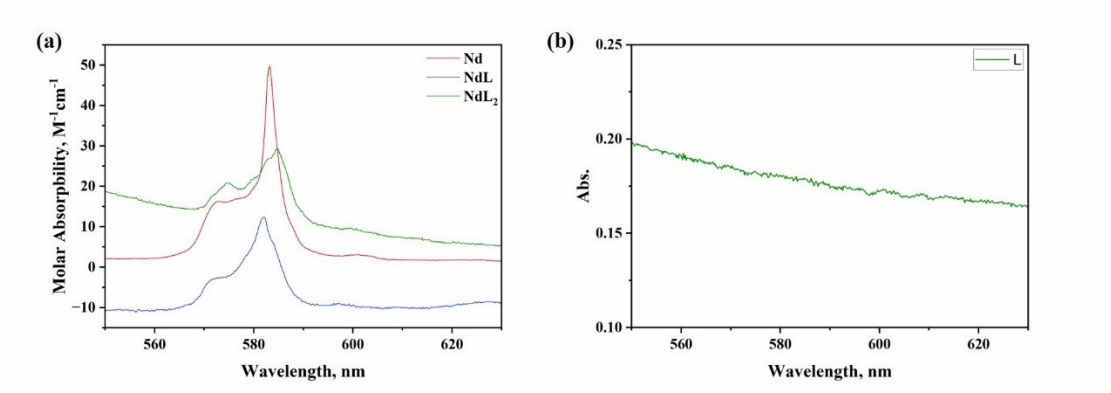

Figure S6. (a) fitted molar absorptivity during Nd(III) titrated by Et-Tol-CA-ATP and (b) the absorption spectra of free Et-Tol-CA-ATP ligand (50 mM)

Extraction acidity experiments in n-octanol

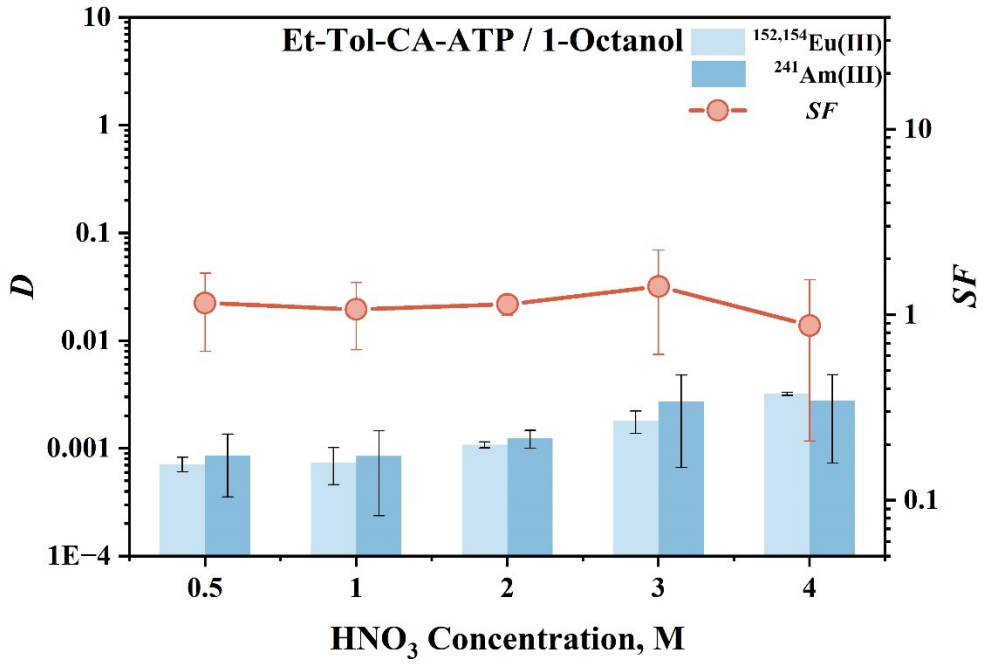

Figure S7. Distribution ratios and separation factors of Am(III) and Eu(III) by Et-Tol-CA-ATP in n-octanol (C_L = 10 mM)

Extraction kinetic measurements

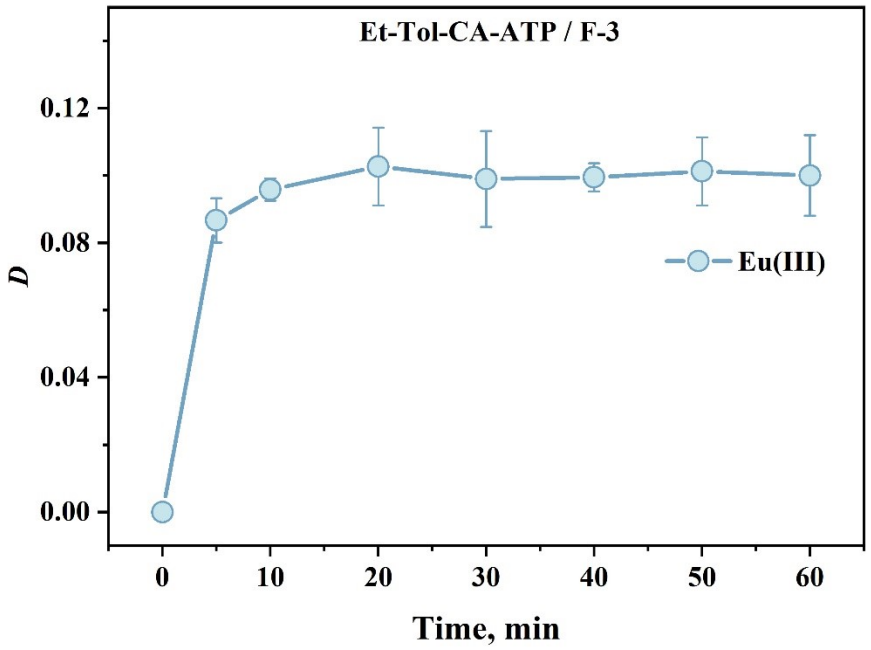

Figure S8. The influence of contact time on Eu(III) extraction by the ligands in F-3 ([Ligand] = 15 mM, $[HNO_3]$ = 3.0 M, T = 298 ± 1 K)

The coordinates of optimized structures of complexes by DFT method

Table S2. $Am(L)(NO_3)_3$

Am	1.81129800	-0.13872500	-0.02126200
N	4.20120300	-0.39816900	-1.78212400
O	5.22055400	-0.43303000	-2.43846500
O	4.02916800	-1.12485600	-0.74708300
O	3.23678300	0.39902300	-2.06345300
N	1.55296000	-2.98626300	-0.77223400
O	1.78072500	-2.59881300	0.43815100
O	1.55684200	-4.15246900	-1.08186000
O	1.30176500	-2.04884300	-1.60447700
N	0.74682900	0.70775400	2.60954300
O	1.53244500	-0.29109700	2.44227500
O	0.22306700	0.94785000	3.67268400
O	0.55275600	1.43342100	1.57245800
C	-4.77299500	-0.21543200	-0.94558100
C	-4.43192000	0.40941400	-2.14914700
C	-3.38257900	1.32287400	-2.21488500
C	-2.65831200	1.62400200	-1.06602900
C	-2.97194600	1.00630600	0.14364600
C	-4.02157700	0.09430200	0.19471600
C	-5.92865200	-1.18263300	-0.87231400
N	-1.57761200	2.58789900	-1.11468900
C	-1.90769700	3.94307600	-0.62079800
C	-0.32571000	2.10326700	-1.27316000
O	-0.09922600	0.89059000	-1.38553700
N	1.92790700	2.48585500	-0.69032000
C	3.08114500	3.15649300	-0.63933400
C	3.24445900	4.39376800	-1.26667100
C	2.16592500	4.92894400	-1.95880200
C	0.95239300	4.24617100	-1.97199100
C	0.87333600	3.02498100	-1.30292200
N	4.82568900	0.72942200	1.46881800
C	6.01904200	1.26451200	1.46830300
C	6.31727100	2.45983400	0.76134400
N	5.40109100	3.11196400	0.08897500
C	4.18562400	2.51042700	0.11444900
N	3.88882000	1.37963000	0.74977300
C	7.79658600	2.71156100	0.90300300
C	8.41678100	1.56132500	0.02274200
C	8.09606400	0.25262000	0.80264100

C	7.30841500	0.76699900	2.05171500
C	8.27992500	4.11056100	0.57436000
C	8.02137700	2.13135200	2.36619500
C	9.49521600	1.98344700	2.76160900
C	7.31802500	2.94077400	3.46690900
H	-4.98792300	0.17502400	-3.05185100
H	-3.11542700	1.78727000	-3.15791300
H	-2.37211300	1.20902100	1.02419500
H	-4.24981100	-0.39266400	1.13793000
H	-5.75935900	-1.95479400	-0.11852700
H	-6.09939400	-1.67949400	-1.82977300
H	-6.85641400	-0.66607200	-0.60333500
H	4.20226200	4.89381500	-1.21905500
H	2.26677200	5.87209300	-2.48446700
H	0.10069700	4.64251000	-2.50954200
H	9.49104700	1.72686100	-0.07995400
H	7.99651000	1.56183600	-0.98466300
H	9.00106200	-0.27097900	1.11412200
H	7.50067900	-0.44894000	0.21707300
H	7.20991400	0.05313200	2.86837900
H	8.04059200	4.36865300	-0.46021000
H	9.36325700	4.18913400	0.69397100
H	7.81132300	4.86177800	1.21526700
H	10.09947100	1.44276200	2.03415500
H	9.57426200	1.44974600	3.71317800
H	9.95432800	2.96528900	2.90633000
H	7.40001100	2.42541200	4.42774100
H	6.25422000	3.10267100	3.27905700
H	7.78359800	3.92302700	3.58541400
C	-2.73920600	4.76218700	-1.60354000
H	-0.98297500	4.45468700	-0.36261100
H	-2.45734000	3.81177200	0.31483300
H	-3.68699400	4.26984300	-1.82887300
H	-2.21247000	4.92373500	-2.54785900
H	-2.96630400	5.73999900	-1.17096200
O	-0.55763500	-1.10133000	0.59964000
H	-1.12837000	-1.12990900	-0.17694100
H	-0.37215200	-2.00453800	0.88721900

Table S3. $Eu(L)(NO_3)_3$

Eu	1.80442100	-0.13176400	-0.02866100
N	4.17410500	-0.38577200	-1.74050100
O	5.20592500	-0.42021200	-2.37832600
O	3.98746200	-1.10962200	-0.70624900
O	3.21402000	0.40515500	-2.04052000
N	1.51384200	-2.92042600	-0.82446400
O	1.75447200	-2.58424200	0.39439200
O	1.50035200	-4.07323000	-1.18301000
O	1.26862500	-1.94818200	-1.61957400
N	0.86909800	0.70847200	2.60247800
O	1.62925000	-0.30245400	2.39206000
O	0.41719700	0.95802900	3.69641100
O	0.62295000	1.43245300	1.57863700
C	-4.75944200	-0.23276300	-0.92116800
C	-4.42166100	0.37542200	-2.13407000
C	-3.37724200	1.29349000	-2.21390100
C	-2.65492600	1.61666000	-1.06983000
C	-2.96565700	1.01578500	0.14919700
C	-4.00984300	0.09855500	0.21428200
C	-5.90926500	-1.20563500	-0.83299600
N	-1.57977900	2.58568300	-1.13321000
C	-1.92242100	3.95049400	-0.67624100
C	-0.32322500	2.10473300	-1.27076100
O	-0.09072500	0.89142400	-1.35236800
N	1.92503100	2.49440700	-0.69395300
C	3.07757900	3.16364700	-0.64300500
C	3.24406300	4.39846600	-1.27468500
C	2.16746000	4.93052600	-1.97248200
C	0.95354100	4.24803200	-1.98650200
C	0.87270000	3.03014000	-1.31086400
N	4.80569400	0.72974600	1.46789700
C	5.99978500	1.26328700	1.47629000
C	6.30484200	2.45913900	0.77403400
N	5.39358900	3.11501300	0.09854100
C	4.17711700	2.51498500	0.11542200
N	3.87433700	1.38239300	0.74452200
C	7.78478100	2.70481600	0.92102100
C	8.40241600	1.55465400	0.03882700
C	8.07343300	0.24482600	0.81353200
C	7.28542100	0.75911000	2.06252500
C	8.27476000	4.10278200	0.59789100
C	8.00339400	2.11919500	2.38285700
C	9.47559600	1.96385700	2.78153600

C	7.30063700	2.92828200	3.48416900
H	-4.97601200	0.12384000	-3.03314900
H	-3.11210600	1.74409600	-3.16413800
H	-2.36657400	1.23482900	1.02639800
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H	-6.08153600	-1.71288700	-1.78468500
H	-6.83867700	-0.69169100	-0.56470200
H	4.20187600	4.89859100	-1.22649900
H	2.27004800	5.87111500	-2.50242300
H	0.10458000	4.64258200	-2.52960300
H	9.47773700	1.71555700	-0.06033000
H	7.98479400	1.56032700	-0.96967200
H	8.97509900	-0.28473000	1.12470300
H	7.47500000	-0.45108800	0.22424400
H	7.18163600	0.04315800	2.87670900
H	8.03864300	4.36516400	-0.43634600
H	9.35816400	4.17656000	0.72007600
H	7.80783200	4.85391300	1.24012700
H	10.07907600	1.42184800	2.05439500
H	9.54997200	1.42785700	3.73220100
H	9.93886600	2.94330300	2.92932600
H	7.37762000	2.40954100	4.44361100
H	6.23812600	3.09536900	3.29372800
H	7.77020500	3.90810700	3.60709100
C	-2.74027700	4.74472400	-1.69052400
H	-1.00376900	4.47099800	-0.41408000
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H	-2.19983300	4.88456800	-2.63052000
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O	-0.52197200	-1.05497600	0.61595600
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H	-0.33454700	-1.96597000	0.87687700

Table S4. $[Am(L)_2(NO_3)]^{2+}$

Am	2.49851900	-0.35442500	-0.41602800
N	3.79095300	0.15412300	-2.96138500
O	4.33273300	-0.56994800	-2.03438200
O	2.68216300	0.71163100	-2.60955700
O	4.28513600	0.29940000	-4.04876900
C	-4.30882800	-1.18837100	-1.38096300
C	-3.53148400	-0.66512100	-2.42505200
C	-2.59669900	0.34356300	-2.19816100
C	-2.42124100	0.84148700	-0.90570300
C	-3.17449500	0.32938100	0.15186900
C	-4.10785200	-0.67783900	-0.09037600
C	-5.35485400	-2.24456800	-1.64453700
N	-1.49207700	1.93363600	-0.67540800
C	-2.11810100	3.22944600	-0.30888100
C	-0.17582500	1.65301100	-0.69220700
O	0.22395300	0.46216800	-0.74529400
N	2.06575100	2.25987100	-0.18214100
C	3.12011500	3.08393400	-0.08654100
C	3.05819200	4.42626300	-0.47751400
C	1.87054100	4.89323000	-1.03042600
C	0.77114300	4.03755400	-1.12138400
C	0.89418700	2.72728200	-0.64804200
N	5.54941900	0.55774200	0.99474100
C	6.54620900	1.33504700	1.34458100
C	6.47079500	2.75799700	1.20480300
N	5.39458500	3.35015100	0.73944000
C	4.38713700	2.49489200	0.42588000
N	4.43449400	1.16809500	0.52392200
C	7.79558800	3.32506500	1.63542000
C	8.76076400	2.86044600	0.47609100
C	8.85509100	1.31418200	0.63460900
C	7.91953900	1.01962600	1.85561500
C	7.82979700	4.81136000	1.94955700
C	8.16331600	2.27541800	2.77704600
C	9.60396300	2.39219500	3.29608400
C	7.22020200	2.32751000	3.99298200
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H	-2.01024300	0.74630800	-3.01862300
H	-3.03372200	0.71147200	1.15946700
H	-4.69441000	-1.06900000	0.73678200
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H	-6.31911900	-1.78383300	-1.89468700
H	-5.51683000	-2.87914800	-0.76760600

H	3.93280800	5.05686000	-0.37459000
H	1.79783300	5.91302900	-1.39500500
H	-0.14446100	4.38510700	-1.58009800
H	9.73084400	3.34623700	0.61062500
H	8.38491100	3.16687900	-0.50453900
H	9.87218300	0.98716200	0.86282500
H	8.53650500	0.77483100	-0.26181700
H	8.04370300	0.03533700	2.31043300
H	7.54871300	5.40104100	1.07071000
H	8.83631900	5.11878100	2.24922000
H	7.13748500	5.07011300	2.75758800
H	10.36370400	2.39769700	2.51230800
H	9.82671300	1.55454400	3.96662800
H	9.72277300	3.31239400	3.87797500
H	7.45700000	1.51361600	4.68739200
H	6.15875900	2.23978100	3.73297300
H	7.34951700	3.26854800	4.53814300
C	-2.83252600	3.91095200	-1.47564800
H	-1.35880100	3.87058200	0.13955500
H	-2.84262900	3.00420500	0.47965100
H	-3.61647200	3.26625100	-1.88275900
H	-2.14856700	4.16265600	-2.29359400
H	-3.30308400	4.83504900	-1.12519300
O	4.39194900	-1.91667900	0.37788100
H	4.85760600	-2.21249400	-0.42469200
H	5.01067200	-1.26001700	0.78586100
C	2.37373400	2.79537300	5.70399800
C	3.16213500	1.63539100	5.68701900
C	2.71678400	0.46488600	5.07366400
C	1.46534800	0.44348400	4.45622200
C	0.66683800	1.58859400	4.45060500
C	1.12358200	2.75155100	5.06917100
C	2.83888200	4.04295400	6.41575300
N	0.97197700	-0.79000000	3.87060600
C	-0.14513900	-1.43776800	4.60513100
C	1.45629300	-1.15554800	2.66916000
O	2.20216100	-0.38322300	2.01399300
N	1.25529000	-2.46210400	0.69553800
C	1.05481100	-3.58536600	-0.00991900
C	0.74721000	-4.80775800	0.60153900
C	0.69834400	-4.85664300	1.98958800
C	0.90908800	-3.68792700	2.72433200
C	1.15309300	-2.49609000	2.03661000

N	1.96449100	-2.21790900	-3.29249900
C	1.59956400	-3.22437300	-4.05121600
C	1.04062500	-4.42362900	-3.51167100
N	0.85923000	-4.57747800	-2.21783200
C	1.22456400	-3.49612300	-1.48468800
N	1.75034900	-2.36695600	-1.96160900
C	0.79614400	-5.36685800	-4.65753700
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H	3.93106300	4.10411300	6.44966800
H	2.48072300	4.05578400	7.45311000
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H	0.58640200	-5.68512400	-0.01284600
H	0.50750600	-5.79502700	2.50082300
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H	2.20135500	-6.57424100	-5.82571800
H	2.83376200	-6.15714800	-4.24045100
H	3.12046300	-4.58552400	-6.74696900
H	3.77753800	-4.14333900	-5.17722000
H	1.82251000	-2.51502400	-6.12485000
H	0.23841400	-7.16588500	-3.58772600
H	-0.24737000	-7.15370200	-5.28810100
H	-1.14128600	-6.18676900	-4.09991800
H	1.22795000	-5.48159300	-7.49223200
H	0.23863400	-4.08999700	-7.93672200
H	-0.52469100	-5.54504900	-7.29645600
H	-1.02112000	-2.75711700	-6.26759800
H	-0.93361200	-3.08290500	-4.53352100
H	-1.74235700	-4.22823500	-5.61074300
C	0.28336700	-2.07651600	5.92567800
H	-0.62902100	-2.15469500	3.94148400
H	-0.87712300	-0.64832800	4.80259100
H	0.72999800	-1.33412900	6.59305000
H	1.01361300	-2.88035600	5.78146800

H	-0.59361700	-2.49728800	6.42769500

Table S5. $[Eu(L)_2(NO_3)]^{2+}$

Eu	2.47239700	-0.35066800	-0.40606200
N	3.69107800	0.20708000	-2.95136100
O	4.22270200	-0.57494100	-2.06815400
O	2.62350800	0.80212600	-2.55410300
O	4.16794800	0.36726300	-4.04643700
C	-4.16146100	-1.32486300	-1.59386500
C	-3.35879200	-0.75211700	-2.59176200
C	-2.46469300	0.27723400	-2.30178600
C	-2.35746400	0.74617500	-0.99125400
C	-3.13619400	0.18453700	0.02172700
C	-4.02796400	-0.84280500	-0.28362300
C	-5.16441300	-2.40325600	-1.92594100
N	-1.47291000	1.85998900	-0.69577500
C	-2.15204400	3.12708300	-0.32314900
C	-0.14996600	1.62403800	-0.67518900
O	0.28752000	0.44561200	-0.74737500
N	2.06462300	2.24856400	-0.11154700
C	3.10779500	3.08010600	0.01420600
C	3.02976000	4.43724100	-0.31967700
C	1.83618000	4.91273800	-0.85152000
C	0.74997200	4.04590800	-0.98609900
C	0.89120100	2.71845400	-0.56860000
N	5.56124900	0.53152400	0.96233700
C	6.55748200	1.29727900	1.33802100
C	6.47230500	2.72416500	1.26069700
N	5.38727100	3.32856300	0.83349800
C	4.38296200	2.48050500	0.48969700
N	4.43907300	1.15164800	0.52416500
C	7.79717400	3.28045400	1.70539500
C	8.75682500	2.87170400	0.52090900
C	8.86144700	1.32066600	0.61264500
C	7.93600100	0.96837500	1.82607700
C	7.82509000	4.75207800	2.08278100
C	8.17903300	2.18528300	2.79862500
C	9.62253700	2.28844500	3.31251700
C	7.24417500	2.18002000	4.02202500
H	-3.44544200	-1.10524200	-3.61606600
H	-1.85758500	0.71755200	-3.08708600
H	-3.04706200	0.54338600	1.04361600

H	-4.63481200	-1.27272300	0.50892100
H	-4.84192600	-3.00311200	-2.78275400
H	-6.13652500	-1.96434000	-2.18475100
H	-5.32844800	-3.07605100	-1.07831100
H	3.89733500	5.07305700	-0.19179000
H	1.74990500	5.94641600	-1.17120700
H	-0.16639100	4.39999500	-1.43782500
H	9.72502600	3.35724100	0.66917600
H	8.37222300	3.21725700	-0.44318500
H	9.88196700	0.99060500	0.82026500
H	8.54022400	0.81792200	-0.30382100
H	8.06922200	-0.03368000	2.23744300
H	7.53457500	5.37716000	1.23191900
H	8.83186500	5.05245900	2.38872100
H	7.13678700	4.97181100	2.90567000
H	10.37678100	2.33094100	2.52459200
H	9.85447300	1.42491600	3.94610800
H	9.74032600	3.18434500	3.93136200
H	7.48822300	1.33700100	4.67815700
H	6.18129400	2.10090600	3.76521600
H	7.37460200	3.09641600	4.60745700
C	-2.83031800	3.82227900	-1.50333800
H	-1.43269000	3.77439500	0.17905200
H	-2.90556700	2.85828100	0.42323300
H	-3.57614200	3.16859100	-1.96421800
H	-2.11638700	4.11540800	-2.28079900
H	-3.34255900	4.72286900	-1.15016200
O	4.33570600	-1.84594800	0.30691600
H	4.77397200	-2.11354500	-0.52169600
H	4.98033000	-1.21437400	0.72261600
C	2.37769200	2.89871100	5.64665500
C	3.18962900	1.75534100	5.61675000
C	2.75237200	0.57080700	5.02487800
C	1.48505500	0.51770100	4.44261600
C	0.66332300	1.64611500	4.44987900
C	1.11193900	2.82335700	5.04682500
C	2.83543500	4.16148200	6.33606300
N	1.00121400	-0.73129000	3.88285000
C	-0.08181100	-1.39327500	4.65422400
C	1.46205700	-1.09841600	2.67150900
O	2.17122700	-0.31753000	1.99031900
N	1.26338700	-2.42757700	0.71676200
C	1.06331400	-3.55571100	0.01988700

C	0.77110300	-4.77695800	0.64037900
C	0.73608300	-4.81701400	2.02924400
C	0.94257800	-3.64177900	2.75530000
C	1.17132600	-2.45194000	2.05845100
N	1.89035500	-2.16308000	-3.27030100
C	1.51982400	-3.16866000	-4.02786200
C	0.99398800	-4.38107600	-3.48468300
N	0.84337900	-4.54690500	-2.18806800
C	1.20967500	-3.46469000	-1.45693500
N	1.71187800	-2.32673700	-1.93640800
C	0.73729000	-5.31957000	-4.63171900
C	2.19698800	-5.69316600	-5.10271200
C	2.76641700	-4.37677400	-5.70988300
C	1.59711800	-3.35535200	-5.51429500
C	-0.17606100	-6.50189600	-4.35452800
C	0.32393300	-4.25495100	-5.74386500
C	0.22103700	-4.83699800	-7.16140900
C	-0.99767300	-3.52535100	-5.44040800
H	4.17219100	1.78522400	6.08064800
H	3.38640700	-0.31088600	5.02280400
H	-0.32120900	1.60675900	3.99152900
H	0.46453500	3.69638900	5.05369400
H	3.92663800	4.24516900	6.34108500
H	2.50392100	4.17592500	7.38221400
H	2.42503600	5.05498900	5.85487400
H	0.61046400	-5.65909500	0.03268400
H	0.55796900	-5.75361000	2.54832400
H	0.95111800	-3.66992300	3.83661400
H	2.13291700	-6.49348100	-5.84490600
H	2.79806100	-6.07733300	-4.27291500
H	2.99321100	-4.48200400	-6.77341100
H	3.68300600	-4.04047600	-5.21756100
H	1.67731000	-2.43903300	-6.10083300
H	0.23761400	-7.13651800	-3.56358500
H	-0.29150800	-7.11894200	-5.25092300
H	-1.17107700	-6.17718800	-4.03215400
H	1.09759000	-5.40628300	-7.47646400
H	0.07594800	-4.02723900	-7.88539500
H	-0.64831400	-5.49898600	-7.23770800
H	-1.16566800	-2.73122800	-6.17639600
H	-1.02681100	-3.06378300	-4.44656300
H	-1.84345700	-4.21807900	-5.50869800
C	0.39771000	-2.01984400	5.96312000

H	-0.57548100	-2.12002000	4.00870300
H	-0.81960300	-0.61470900	4.87198600
H	0.85290600	-1.26737300	6.61323000
H	1.13632600	-2.81217800	5.79902000
H	-0.45609600	-2.45253800	6.49413700