*Supplementary Information for:*

# DFT Calculation of $^{225}Ac^{3+}$, $^{213}Bi^{3+}$ and $^{177}Lu^{3+}$ with Hybrid Chelator 3p-*C*-DEPA to Predict the Stability Constants and Reactivities: Challenges and Perspectives

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**Table S1.** Calculated energies from the optimized structures of complex

| Complex | Functional/ Basis Sets[a] | Electronic Energy (Hartree) | Zero-point Energy (Hartree) | Thermal Free Energy Correction (kcal/mol) | Total Entropy (cal/mol/K) | ΔG*solv (COSMO) (kcal/mol) | ΔG*solv (SMD) (kcal/mol) |
|---|---|---|---|---|---|---|---|
| $[Ac(H_2O)_9]^{3+}$ | M06-HF/ 6-311G(d) | -1050.824 | -1050.594 | -1050.650 | 175.375 | -1885.643 | -1885.693 |
| | B3LYP/ 6-311G(d) | - | - | - | - | - | - |
| $[Ac(DOTA)(H_2O)]^{-}$ | M06-HF/ 6-311G(d) | -1885.515 | -1885.053 | -1885.109 | 183.086 | -1885.643 | -1885.693 |
| | B3LYP/ 6-311G(d) | - | - | - | - | - | - |
| [Ac(3p-*C*-DEPA)]$^{2-}$ | M06-HF/ 6-311G(d) | -2723.788 | -2723.070 | -2723.147 | 254.254 | -2724.068 | -2724.104 |
| | B3LYP/ 6-311G(d) | - | - | - | - | - | - |
| $[Bi(H_2O)_9]^{3+}$ | M06-HF/ 6-311G(d) | -691.871 | -691.640 | -691.694 | 168.023 | -692.412 | -692.486 |
| | B3LYP/ 6-311G(d) | -692.094 | -691.869 | -691.926 | 178.930 | -692.685 | -692.705 |
| $[Bi(DOTA)(H_2O)]^{-}$ | M06-HF/ 6-311G(d) | -1526.597 | -1526.134 | -1526.192 | 182.735 | -1526.726 | -1526.732 |
| | B3LYP/ 6-311G(d) | -1526.929 | -1526.480 | -1526.539 | 189.387 | -1527.015 | -1527.051 |
| [Bi(3p-*C*-DEPA)]$^{2-}$ | M06-HF/ 6-311G(d) | -2364.860 | -2364.144 | -2364.221 | 254.532 | -2365.136 | -2365.135 |
| | B3LYP/ 6-311G(d) | -2365.409 | -2364.711 | -2364.788 | 260.718 | -2365.624 | -2365.658 |
| $[Lu(H_2O)_9]^{3+}$ | M06-HF/ 6-311G(d) | -691.871 | -691.640 | -691.694 | 178.930 | -725.209 | -725.822 |
| | B3LYP/ 6-311G(d) | -725.209 | -725.328 | -725.378 | 158.875 | -726.126 | -726.189 |
| $[Lu(DOTA)(H_2O)]^{-}$ | M06-HF/ 6-311G(d) | -1559.923 | -1559.459 | -1559.514 | 176.523 | -1560.060 | -1560.068 |
| | B3LYP/ 6-311G(d) | -1560.377 | -1559.926 | -1559.983 | 183.437 | -1560.482 | -1560.512 |
| [Lu(3p-*C*-DEPA)]$^{2-}$ | M06-HF/ 6-311G(d) | -2398.161 | -2397.442 | -2397.518 | 251.566 | -2398.447 | -2398.450 |
| | B3LYP/ 6-311G(d) | -2398.831 | -2398.133 | -2398.212 | 262.653 | -2399.090 | -2399.096 |

[a]6-311G(d)basis set was used for all atoms except Ac, Bi, and Lu.

`**Table S2.** Compiled $\Delta G°_g$, $\Delta G_{aq}$, and log $K_1$ calculated in this work

| Equilibrium | Functional/ Basis Sets[a] | $\Delta G°_g$ (kcal/mol) | $\Delta G_{aq}$ (COSMO) (kcal/mol) | $\Delta G_{aq}$ (SMD) (kcal/mol) | log $K_1$ | log $K_1$ (SMD) | log $K_1$ (COSMO) |
|---|---|---|---|---|---|---|---|
| $[Ac(H_2O)_9]^{3+}$ + $[DOTA]^{4-}$ ⇌ $[Ac(DOTA)(H_2O)]^{-}$ | M06-HF/ 6-311G(d) | -1126.46 | -38.44 | -31.04 | 815.83 | 22.75 | 28.17 |
| | B3LYP/ 6-311G(d) | - | - | - | - | - | - |
| $[Ac(H_2O)_9]^{3+}$ + [3p-*C*-DEPA]$^{5-}$ ⇌ [Ac(3p-*C*-DEPA]$^{2-}$ | M06-HF/ 6-311G(d) | -882.54 | -93.56 | -82.91 | 637.07 | 60.76 | 68.57 |
| | B3LYP/ 6-311G(d) | - | - | - | - | - | - |
| $[Bi(H_2O)_9]^{3+}$ + $[DOTA]^{4-}$ ⇌ $[Bi(DOTA)(H_2O)]^{-}$ | M06-HF/ 6-311G(d) | -1150.60 | -74.86 | -64.06 | 833.52 | 46.95 | 54.86 |
| | B3LYP/ 6-311G(d) | -847.57 | -94.88 | -71.71 | 611.44 | 52.55 | 69.54 |
| $[Bi(H_2O)_9]^{3+}$ + [3p-*C*-DEPA]$^{5-}$ ⇌ [Bi(3p-*C*-DEPA]$^{2-}$ | M06-HF/ 6-311G(d) | -901.42 | -65.03 | -80.37 | 650.910 | 40.51 | 47.66 |
| | B3LYP/ | -1258.34 | -80.24 | -61.42 | 912.486 | 45.01 | 58.81 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| | 6-311G(d) | | | | | | |
| $[Lu(H_2O)_9]^{3+}$ + $[DOTA]^{4-}$ ⇌ $[Lu(DOTA)(H_2O)]^{-}$ | M06-HF/ 6-311G(d) | -876.24 | -81.97 | -62.23 | 830.72 | 41.38 | 46.13 |
| | B3LYP/ 6-311G(d) | -888.91 | -92.76 | -74.44 | 607.82 | 45.44 | 56.04 |
| $[Lu(H_2O)_9]^{3+}$ + $[3p\text{-}C\text{-}DEPA]^{5-}$ ⇌ $[Lu(3p\text{-}C\text{-}DEPA)]^{2-}$ | M06-HF/ 6-311G(d) | -882.03 | -65.03 | -39.96 | 636.69 | 20.93 | 29.44 |
| | B3LYP/ 6-311G(d) | -785.70 | -54.41 | -43.91 | 485.44 | 24.72 | 29.79 |

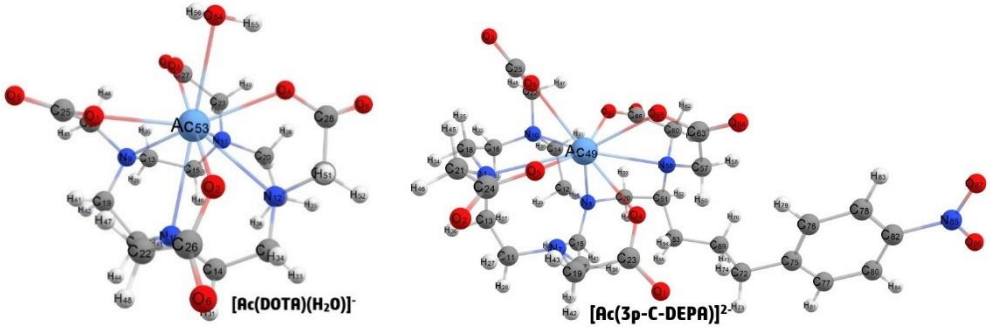


**Figure S1.** DFT-optimized structure and cartesian coordinate of $[Ac(DOTA)(H_2O)]^{-}$, and $[Ac(3p\text{-}C\text{-}DEPA]^{2-}$ complex.

## Cartesian coordinates of the most important optimized structures (Angstroms).

Cartesian coordinate of $[Ac(DOTA)(H_2O)]^{-}$ complex

| Atom | X | Y | Z |
|---|---|---|---|
| O | -1.68064 | 1.51603 | -1.19104 |
| O | 1.36684 | 1.67614 | -1.3471 |
| O | -1.7094 | -1.49417 | -1.00593 |
| O | 1.20431 | -1.78731 | -1.35848 |
| O | -3.86496 | 1.90424 | -1.45026 |
| O | 2.28492 | 3.69355 | -1.06288 |
| O | -3.05131 | -3.06954 | -0.1773 |
| O | 3.05819 | -3.03208 | -1.37481 |
| N | -1.99345 | 0.41115 | 1.18691 |
| N | 0.56713 | 1.89211 | 1.28443 |
| N | -0.05136 | -1.87944 | 1.21162 |
| N | 2.3573 | -0.26277 | 0.60049 |
| C | -2.08176 | -0.71396 | 2.13213 |
| C | 1.9205 | 1.82971 | 1.86998 |
| C | -0.73411 | -1.39349 | 2.41892 |
| C | 2.91805 | 1.02609 | 1.02381 |
| C | -0.43482 | 2.00299 | 2.35112 |
| C | 2.49634 | -1.18577 | 1.743 |
| C | -1.84284 | 1.71917 | 1.84357 |
| C | 1.30768 | -2.34006 | 1.55279 |
| C | -3.20137 | 0.50354 | 0.34294 |
| C | 0.48532 | 3.04672 | 0.35938 |
| C | -0.79088 | -2.98972 | 0.57647 |
| C | 3.16943 | -0.81601 | -0.49893 |
| C | -2.92338 | 1.39931 | -0.88338 |
| C | 1.47864 | 2.82472 | -0.80282 |
| C | -1.9821 | -2.49802 | -0.26773 |
| C | 2.44149 | -2.0097 | -1.1504 |
| H | -2.5041 | -0.38297 | 3.0881 |
| H | -2.76054 | -1.45701 | 1.71985 |
| H | 2.32083 | 2.8399 | 2.00108 |
| H | 1.84292 | 1.38181 | 2.85893 |
| H | 3.82331 | 0.87008 | 1.62523 |
| H | 3.19694 | 1.5874 | 0.1397 |
| H | 3.28753 | -1.61517 | 1.93099 |
| H | 2.0162 | -0.41739 | 2.62698 |
| H | 1.28356 | -2.92581 | 2.48028 |

| | | | |
|---|---|---|---|
| H | 1.66143 | -2.99231 | 0.7625 |
| H | -0.92043 | -2.23183 | 3.10266 |
| H | -0.06744 | -0.69692 | 2.9202 |
| H | -2.11502 | 2.48245 | 1.11931 |
| H | -2.54346 | 1.80003 | 2.68359 |
| H | -0.18633 | 1.29557 | 3.13922 |
| H | -0.41796 | 3.0068 | 2.79379 |
| H | -4.0429 | 0.9287 | 0.89614 |
| H | -3.46926 | -0.4767 | -0.03987 |
| H | -0.5085 | 3.08837 | -0.08052 |
| H | 0.73502 | 3.9757 | 0.87638 |
| H | -0.11603 | -3.46238 | -0.13408 |
| H | -1.14315 | -3.71003 | 1.31834 |
| H | 3.2566 | -0.03974 | -1.25782 |
| H | 4.15471 | -1.13385 | -0.15058 |
| Ac | 0.00263 | 0.00654 | -0.37675 |
| O | -0.3407 | -0.28744 | -2.95167 |
| H | 0.41409 | -0.36032 | -3.54038 |
| H | -1.23183 | -0.33131 | -3.30603 |

Cartesian coordinate of [Ac(3p-*C*-DEPA)]$^{2-}$complex

| **Atom** | **X** | **Y** | **Z** |
|---|---|---|---|
| O | 1.91169 | 2.96331 | -0.7639 |
| O | -3.52888 | 4.13434 | -1.88834 |
| O | -6.04251 | -1.93069 | -1.24094 |
| O | 0.4563 | 1.31509 | -0.99115 |
| O | -2.68296 | 2.08946 | -1.81913 |
| O | -4.175 | -0.92992 | -1.41062 |
| N | -1.08923 | 2.47916 | 0.94305 |
| N | -0.95274 | -0.44633 | 2.27234 |
| N | -4.09991 | 1.69751 | 0.6138 |
| N | -3.88652 | -1.24455 | 1.38838 |
| C | -2.25039 | 3.22373 | 1.39256 |
| C | -2.03857 | -1.06193 | 3.08253 |
| C | -3.51367 | 2.44992 | 1.71284 |
| C | -3.07887 | -1.93975 | 2.38449 |
| C | -0.61252 | 0.85559 | 2.88697 |
| C | -4.80179 | -0.2878 | 1.99571 |
| C | -0.19672 | 2.03869 | 2.01116 |
| C | -5.20444 | 0.87286 | 1.06274 |
| C | -0.29956 | 3.29293 | 0.00571 |
| C | 0.17142 | -1.42232 | 2.25569 |
| C | -4.56077 | 2.5641 | -0.47186 |
| C | -4.62442 | -2.19071 | 0.54239 |
| C | 0.80621 | 2.46753 | -0.65539 |
| C | -3.48684 | 2.99116 | -1.48647 |
| C | -5.04301 | -1.63325 | -0.82985 |
| H | -2.00278 | 3.82391 | 2.28792 |
| H | -2.48882 | 3.97075 | 0.63148 |
| H | -1.59808 | -1.6624 | 3.88116 |
| H | -2.5648 | -0.27042 | 3.58876 |
| H | -3.71062 | -2.33859 | 3.19006 |
| H | -2.61386 | -2.78514 | 1.91316 |
| H | -5.72394 | -0.78886 | 2.30417 |
| H | -4.3716 | 0.09849 | 2.90719 |
| H | -5.92938 | 1.48314 | 1.64811 |
| H | -5.72906 | 0.48512 | 0.23596 |
| H | -4.24255 | 3.17068 | 2.10724 |
| H | -3.30374 | 1.76628 | 2.51956 |
| H | 0.75628 | 1.83079 | 1.56825 |
| H | -0.01683 | 2.86322 | 2.7127 |
| H | -1.46069 | 1.18256 | 3.46702 |
| H | 0.19068 | 0.72266 | 3.61547 |
| H | 0.14737 | 4.15028 | 0.51074 |
| H | -0.94852 | 3.65598 | -0.77721 |
| H | 0.66594 | -1.38424 | 3.23139 |
| H | -5.28721 | 2.0106 | -1.05301 |
| H | -5.05739 | 3.455 | -0.08587 |
| H | -3.97932 | -3.02838 | 0.33342 |
| H | -5.51456 | -2.55613 | 1.05401 |
| Ac | -1.78158 | -0.15115 | -0.87891 |
| H | -0.28431 | -2.39729 | 2.20492 |
| C | 1.32082 | -1.43388 | 1.20457 |

| | | | |
|---|---|---|---|
| H | 1.91767 | -2.27551 | 1.57409 |
| C | 2.24413 | -0.20869 | 1.28618 |
| H | 1.91366 | 0.54432 | 0.59523 |
| H | 2.16171 | 0.21317 | 2.28418 |
| N | 0.8441 | -1.77421 | -0.14939 |
| C | 1.69965 | -1.4282 | -1.28788 |
| H | 2.55988 | -2.09113 | -1.36499 |
| H | 2.04552 | -0.41317 | -1.20481 |
| C | 0.50303 | -3.19744 | -0.23628 |
| H | 1.20046 | -3.80908 | 0.33346 |
| H | 0.5824 | -3.51019 | -1.26913 |
| C | 0.95994 | -1.47726 | -2.64321 |
| O | 1.62849 | -1.76668 | -3.60744 |
| O | -0.26488 | -1.18097 | -2.61969 |
| C | -0.92581 | -3.53207 | 0.17714 |
| O | -1.77893 | -2.69632 | -0.19352 |
| O | -1.12471 | -4.54961 | 0.81421 |
| C | 3.43104 | -0.48648 | 1.04185 |
| H | 3.87593 | -1.04256 | 0.12579 |
| H | 4.13489 | -1.10578 | 1.84231 |
| C | 4.51593 | 0.83367 | 0.95706 |
| H | 4.43127 | 1.36763 | 1.89988 |
| H | 4.06127 | 1.46724 | 0.20473 |
| C | 5.97513 | 0.62188 | 0.63608 |
| C | 6.40513 | 0.55085 | -0.68783 |
| C | 6.92093 | 0.46941 | 1.64749 |
| C | 7.7331 | 0.33566 | -0.99649 |
| H | 5.69034 | 0.66872 | -1.48159 |
| C | 8.25323 | 0.25453 | 1.35933 |
| H | 6.60915 | 0.52419 | 2.57531 |
| C | 8.64487 | 0.19121 | 0.03478 |
| H | 8.06094 | 0.28325 | -2.01483 |
| H | 8.979 | 0.14074 | 2.13878 |
| N | 10.05122 | -0.02657 | -0.27835 |
| O | 10.81281 | -0.1518 | 0.62912 |
| O | 10.37014 | -0.06704 | -1.4227 |

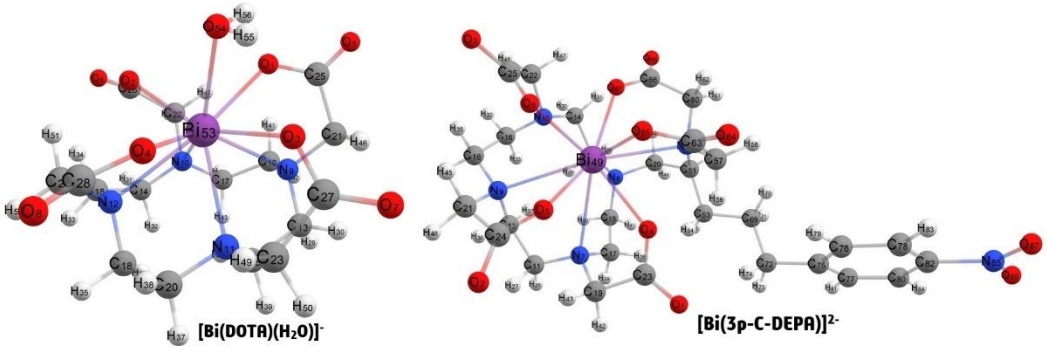


**Figure S2.** DFT-optimized structure and cartesian coordinate of $[Bi(DOTA)(H_2O)]^-$, and $[Bi(3p\text{-}C\text{-}DEPA]^{2-}$ complex.

Cartesian coordinate of $[Bi(DOTA)(H_2O)]^-$ complex

| Atom | X | Y | Z |
|---|---|---|---|
| O | -1.95557 | 1.48216 | -1.37571 |
| O | 1.57145 | 1.80982 | -1.42933 |
| O | -1.84193 | -1.83925 | -0.73787 |
| O | 1.35152 | -1.97195 | -1.251 |
| O | -4.14113 | 1.96405 | -1.36698 |
| O | 2.53059 | 3.77595 | -0.96917 |
| O | -3.16521 | -3.15769 | 0.48567 |
| O | 3.42563 | -3.09277 | -0.77942 |
| N | -2.0572 | 0.59615 | 1.1513 |
| N | 0.55363 | 2.07843 | 1.20971 |
| N | -0.08172 | -1.76752 | 1.24825 |
| N | 2.40216 | -0.18642 | 0.76689 |
| C | -2.07277 | -0.4052 | 2.23105 |
| C | 1.86331 | 1.99753 | 1.88173 |

| | | | |
|---|---|---|---|
| C | -0.69796 | -1.01892 | 2.55268 |
| C | 2.90685 | 1.14248 | 1.1446 |
| C | -0.51445 | 2.29791 | 2.1959 |
| C | 2.29048 | -1.05071 | 1.95228 |
| C | -1.90101 | 1.97334 | 1.64673 |
| C | 1.27964 | -2.19648 | 1.81646 |
| C | -3.30529 | 0.56202 | 0.35988 |
| C | 0.5516 | 3.17471 | 0.21354 |
| C | -0.87112 | -2.96218 | 1.10184 |
| C | 3.31754 | -0.77379 | -0.22914 |
| C | -3.15471 | 1.42763 | -0.91826 |
| C | 1.66386 | 2.93792 | -0.83503 |
| C | -2.09771 | -2.6423 | 0.22148 |
| C | 2.74867 | -2.08763 | -0.80451 |
| H | -2.4552 | 0.04243 | 3.1567 |
| H | -2.76088 | -1.20167 | 1.95726 |
| H | 2.28631 | 2.99978 | 2.01119 |
| H | 1.40441 | 1.58395 | 2.87645 |
| H | 3.77672 | 1.0419 | 1.80831 |
| H | 3.23661 | 1.64981 | 0.24514 |
| H | 3.26463 | -1.50278 | 2.1744 |
| H | 2.02478 | -0.43332 | 2.80682 |
| H | 1.25553 | -2.7293 | 2.77724 |
| H | 1.62545 | -2.89398 | 1.06121 |
| H | -0.82638 | -1.68208 | 3.21971 |
| H | -0.00952 | -0.22868 | 2.83435 |
| H | -2.12467 | 2.64649 | 0.82319 |
| H | -2.63665 | 2.16707 | 2.43808 |
| H | -0.32274 | 1.68209 | 3.07151 |
| H | -0.51585 | 3.34225 | 2.53305 |
| H | -4.15297 | 0.94369 | 0.93457 |
| H | -3.5011 | -0.45861 | 0.03782 |
| H | -0.40386 | 3.17972 | -0.31183 |
| H | 0.72825 | 4.13649 | 0.7009 |
| H | -0.23901 | -3.60904 | 0.49588 |
| H | -1.20006 | -3.49695 | 1.9968 |
| H | 3.40401 | -0.0635 | -1.05108 |
| H | 4.29951 | -0.97466 | 0.20628 |
| Bi | -0.04468 | -0.00608 | -0.78182 |
| O | -0.39929 | -1.57068 | -2.98631 |
| H | 0.35606 | -2.12535 | -2.73026 |
| H | -1.20499 | -1.97397 | -2.63167 |

Cartesian coordinate of $[Bi(3p\text{-}C\text{-}DEPA)]^{2-}$ complex

| **Atom** | **X** | **Y** | **Z** |
|---|---|---|---|
| O | 1.79868 | 3.24656 | 0.30833 |
| O | -3.08623 | 4.03309 | -1.9986 |
| O | -5.20953 | -2.22282 | -2.4701 |
| O | 0.64525 | 1.44678 | -0.31077 |
| O | -2.04833 | 2.05968 | -1.89276 |
| O | -3.37625 | -1.02394 | -2.03287 |
| N | -1.47895 | 2.38458 | 1.04895 |
| N | -1.40406 | -0.49184 | 2.32263 |
| N | -4.09683 | 1.25769 | -0.10979 |
| N | -3.76033 | -1.59093 | 0.66997 |
| C | -2.80727 | 2.98984 | 1.19617 |
| C | -2.59166 | -1.2446 | 2.81978 |
| C | -3.97247 | 2.00002 | 1.14371 |
| C | -3.24593 | -2.21717 | 1.86013 |
| C | -1.36074 | 0.79827 | 3.05378 |
| C | -4.98446 | -0.80793 | 0.98594 |
| C | -0.83972 | 2.04037 | 2.32269 |
| C | -5.22656 | 0.32917 | -0.00734 |
| C | -0.58519 | 3.33913 | 0.35995 |
| C | -0.22514 | -1.36153 | 2.59945 |
| C | -4.32461 | 2.1362 | -1.26381 |
| C | -4.12273 | -2.56641 | -0.38114 |
| C | 0.75433 | 2.64445 | 0.07661 |
| C | -3.0407 | 2.82924 | -1.7696 |
| C | -4.26708 | -1.88949 | -1.76603 |
| H | -2.87407 | 3.53128 | 2.15103 |
| H | -2.93926 | 3.72321 | 0.4036 |

| | | | |
|---|---|---|---|
| H | -2.30032 | -1.77967 | 3.73935 |
| H | -3.34954 | -0.52034 | 3.10159 |
| H | -4.04843 | -2.74175 | 2.4317 |
| H | -2.54163 | -3.00959 | 1.54383 |
| H | -5.87309 | -1.43358 | 0.97472 |
| H | -4.91041 | -0.40419 | 1.99266 |
| H | -6.13251 | 0.87121 | 0.30864 |
| H | -5.41827 | -0.08729 | -0.99018 |
| H | -4.89608 | 2.57155 | 1.33407 |
| H | -3.85636 | 1.27962 | 1.94836 |
| H | 0.21982 | 1.92998 | 2.14001 |
| H | -0.94837 | 2.87705 | 3.03136 |
| H | -2.36807 | 1.03384 | 3.38754 |
| H | -0.75663 | 0.68758 | 3.96364 |
| H | -0.39739 | 4.21981 | 0.98291 |
| H | -1.02902 | 3.62756 | -0.58984 |
| H | -0.01776 | -1.31768 | 3.67937 |
| H | -4.65388 | 1.50345 | -2.08642 |
| H | -5.08079 | 2.89886 | -1.04628 |
| H | -3.30028 | -3.26983 | -0.45942 |
| H | -5.05935 | -3.04609 | -0.1555 |
| Bi | -1.42306 | -0.10858 | -0.68997 |
| H | -0.54174 | -2.38011 | 2.38647 |
| C | 1.14856 | -1.18014 | 1.89434 |
| H | 1.73477 | -1.99164 | 2.35541 |
| C | 1.88868 | 0.12504 | 2.2389 |
| H | 1.57667 | 0.88146 | 1.52809 |
| H | 1.60203 | 0.44407 | 3.24286 |
| N | 1.06308 | -1.42822 | 0.44155 |
| C | 2.1467 | -0.89378 | -0.38607 |
| H | 3.04337 | -1.52112 | -0.34632 |
| H | 2.37729 | 0.12269 | -0.09839 |
| C | 0.9598 | -2.86493 | 0.15874 |
| H | 1.65242 | -3.44796 | 0.77357 |
| H | 1.18943 | -3.01343 | -0.89467 |
| C | 1.75542 | -0.75984 | -1.88051 |
| O | 2.66765 | -0.59105 | -2.64375 |
| O | 0.52114 | -0.82054 | -2.15396 |
| C | -0.46772 | -3.37914 | 0.33984 |
| O | -1.33308 | -2.63699 | -0.20383 |
| O | -0.68234 | -4.40471 | 0.98147 |
| C | 3.42966 | -0.01939 | 2.13938 |
| H | 3.67928 | -0.89085 | 1.55962 |
| H | 3.84886 | -0.19519 | 3.15187 |
| C | 4.10459 | 1.23054 | 1.52478 |
| H | 4.32185 | 1.96724 | 2.29801 |
| H | 3.43167 | 1.71343 | 0.81539 |
| C | 5.38229 | 0.83347 | 0.82072 |
| C | 5.35069 | 0.43729 | -0.52256 |
| C | 6.59783 | 0.80183 | 1.50957 |
| C | 6.50846 | 0.01357 | -1.15938 |
| H | 4.4238 | 0.44268 | -1.08424 |
| C | 7.76604 | 0.38814 | 0.88938 |
| H | 6.62477 | 1.11039 | 2.54552 |
| C | 7.69357 | 0.00127 | -0.43897 |
| H | 6.47106 | -0.30058 | -2.1906 |
| H | 8.71296 | 0.36124 | 1.4035 |
| N | 8.92789 | -0.43442 | -1.10097 |
| O | 9.94544 | -0.4525 | -0.4481 |
| O | 8.87033 | -0.75138 | -2.26034 |

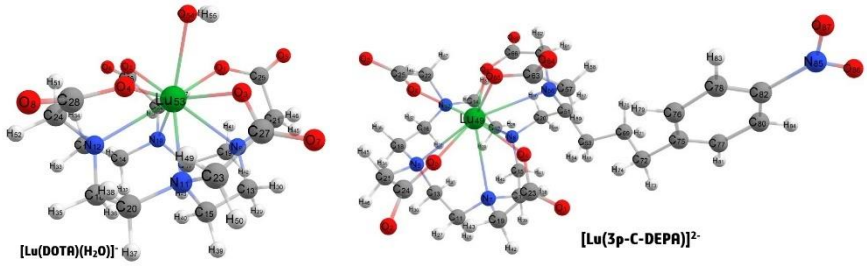


**Figure S3.** DFT-optimized structure and cartesian coordinate of $[Lu(DOTA)(H_2O)]^-$, and $[Lu(3p\text{-}C\text{-}DEPA)(H_2O)]^{2-}$ complex.

Cartesian coordinate of $[Lu(DOTA)(H_2O)]^-$ complex

| Atom | X | Y | Z |
|---|---|---|---|
| O | -2.05878 | 1.23606 | -1.27348 |
| O | 1.34118 | 2.40801 | -0.96505 |
| O | -1.45468 | -1.88311 | -0.87494 |
| O | 2.13565 | -1.4288 | -1.31244 |
| O | -4.24121 | 1.8056 | -1.18798 |
| O | 2.05406 | 4.50508 | -0.49721 |
| O | -2.61107 | -3.72968 | -0.2768 |
| O | 4.12719 | -2.49963 | -1.20008 |
| N | -1.97221 | 0.23238 | 1.17576 |
| N | 0.43545 | 1.96319 | 1.50633 |
| N | 0.10791 | -1.90235 | 1.32226 |
| N | 2.44073 | -0.11092 | 0.9889 |
| C | -2.0622 | -0.90293 | 2.15145 |
| C | 1.69625 | 1.86671 | 2.32103 |
| C | -0.70549 | -1.52672 | 2.53113 |
| C | 2.87222 | 1.19414 | 1.61241 |
| C | -0.66781 | 1.93211 | 2.52703 |
| C | 2.35382 | -1.07711 | 2.13865 |
| C | -2.02494 | 1.51517 | 1.9571 |
| C | 1.46392 | -2.29137 | 1.83634 |
| C | -3.31616 | 0.23367 | 0.45457 |
| C | 0.45137 | 3.39315 | 0.98427 |
| C | -0.4947 | -3.20639 | 0.83018 |
| C | 3.69963 | -0.55641 | 0.21939 |
| C | -3.23728 | 1.18609 | -0.76352 |
| C | 1.37719 | 3.4971 | -0.2592 |
| C | -1.64016 | -2.96464 | -0.18416 |
| C | 3.33422 | -1.61359 | -0.85501 |
| H | -2.56554 | -0.59325 | 3.10865 |
| H | -2.68147 | -1.72803 | 1.68817 |
| H | 2.05987 | 2.88553 | 2.65067 |
| H | 1.49661 | 1.3018 | 3.28545 |
| H | 3.70676 | 1.05891 | 2.35347 |
| H | 3.23518 | 1.886 | 0.79578 |
| H | 3.36126 | -1.4781 | 2.43581 |
| H | 1.93573 | -0.54801 | 3.03875 |
| H | 1.38818 | -2.91053 | 2.77123 |
| H | 1.97976 | -2.90619 | 1.04038 |
| H | -0.91755 | -2.41963 | 3.18337 |
| H | -0.1129 | -0.79837 | 3.15038 |
| H | -2.39093 | 2.32682 | 1.26091 |
| H | -2.75193 | 1.45427 | 2.81431 |
| H | -0.41419 | 1.20231 | 3.34788 |
| H | -0.79737 | 2.93988 | 3.01352 |
| H | -4.15136 | 0.50924 | 1.14595 |
| H | -3.49752 | -0.79817 | 0.03723 |
| H | -0.58801 | 3.66018 | 0.64996 |
| H | 0.76419 | 4.12864 | 1.76966 |
| H | 0.28966 | -3.78134 | 0.26386 |
| H | -0.85527 | -3.84329 | 1.67863 |
| H | 4.11269 | 0.32551 | -0.34399 |
| H | 4.50549 | -0.94422 | 0.89403 |
| Lu | 0.18332 | 0.11636 | -0.57983 |
| O | -0.80491 | -0.13632 | -2.93235 |
| H | -1.46856 | -0.78859 | -2.67364 |
| H | -1.28929 | 0.69278 | -3.00676 |

**Figure S4.** DFT-optimized structure and cartesian coordinate of $[Lu(3p\text{-}C\text{-}DEPA)]^{2-}$ complex.

Cartesian coordinate of $[Lu(3p\text{-}C\text{-}DEPA)]^{2-}$ complex

| Atom | X | Y | Z |
|---|---|---|---|
| O | -2.05878 | 1.23606 | -1.27348 |
| O | 1.34118 | 2.40801 | -0.96505 |
| O | -1.45468 | -1.88311 | -0.87494 |
| O | 2.13565 | -1.4288 | -1.31244 |
| O | -4.24121 | 1.8056 | -1.18798 |

| | | | |
|---|---|---|---|
| O | 2.05406 | 4.50508 | -0.49721 |
| O | -2.61107 | -3.72968 | -0.2768 |
| O | 4.12719 | -2.49963 | -1.20008 |
| N | -1.97221 | 0.23238 | 1.17576 |
| N | 0.43545 | 1.96319 | 1.50633 |
| N | 0.10791 | -1.90235 | 1.32226 |
| N | 2.47073 | -0.11092 | 0.9889 |
| C | -2.0622 | -0.90293 | 2.15145 |
| C | 1.69625 | 1.86671 | 2.33103 |
| C | -0.70549 | -1.52672 | 2.53113 |
| C | 2.83222 | 1.19414 | 1.61241 |
| C | -0.66781 | 1.93211 | 2.52703 |
| C | 2.35382 | -1.07711 | 2.13865 |
| C | -2.02494 | 1.51517 | 1.9171 |
| C | 1.46392 | -2.29137 | 1.83634 |
| C | -3.31616 | 0.23367 | 0.45457 |
| C | 0.45137 | 3.39315 | 0.98427 |
| C | -0.4947 | -3.20639 | 0.83018 |
| C | 3.69963 | -0.55641 | 0.21939 |
| C | -3.25728 | 1.18609 | -0.76352 |
| C | 1.37719 | 3.4971 | -0.2592 |
| C | -1.64016 | -2.96464 | -0.18416 |
| C | 3.33422 | -1.61359 | -0.85501 |
| H | -2.56554 | -0.59325 | 3.10865 |
| H | -2.68147 | -1.72803 | 1.68817 |
| H | 2.05987 | 2.88553 | 2.65067 |
| H | 1.49661 | 1.3018 | 3.28545 |
| H | 3.70676 | 1.05891 | 2.35347 |
| H | 3.23518 | 1.886 | 0.79578 |
| H | 3.36126 | -1.4781 | 2.44581 |
| H | 1.93573 | -0.54801 | 3.03875 |
| H | 1.38818 | -2.91053 | 2.77123 |
| H | 1.97976 | -2.90619 | 1.04038 |
| H | -0.91755 | -2.41963 | 3.18337 |
| H | -0.1129 | -0.79837 | 3.15038 |
| H | -2.39093 | 2.32682 | 1.26091 |
| H | -2.55193 | 1.45427 | 2.81431 |
| H | -0.41419 | 1.20231 | 3.34788 |
| H | -0.79737 | 2.93988 | 3.01352 |
| H | -4.15136 | 0.50924 | 1.14595 |
| H | -3.49752 | -0.79817 | 0.03723 |
| H | -0.58801 | 3.67018 | 0.64996 |
| H | 0.76419 | 4.12864 | 1.76966 |
| H | 0.28966 | -3.78134 | 0.26386 |
| H | -0.85527 | -3.84329 | 1.64863 |
| H | 4.11269 | 0.32551 | -0.34399 |
| H | 4.50549 | -0.94422 | 0.89403 |
| Lu | 0.18332 | 0.11636 | -0.57983 |
| O | -0.80491 | -0.13632 | -2.93235 |
| H | -1.46856 | -0.78859 | -2.67364 |
| H | -1.28929 | 0.69278 | -3.00676 |

**Table S3**. Atomic charge distribution of donor atoms in $[DOTA]^{4-}$ and $[3p\text{-}C\text{-}DEPA]^{5-}$

| **Chelators** | **Donor Atom** | **Natural Population Analysis (NPA)** | **Chelators** | **Donor Atom** | **Natural Population Analysis (NPA)** |
|---|---|---|---|---|---|
| $[DOTA]^{4-}$ | O1 | -0.88640 | $[3p\text{-}C\text{-}DEPA]^{5-}$ | O1 | -0.88879 |
| | O2 | -0.88863 | | O2 | -0.88015 |
| | O3 | -0.88892 | | O3 | -0.88636 |
| | O4 | -0.85891 | | O4 | -0.86616 |
| | O5 | -0.85908 | | O5 | -0.87407 |
| | O6 | -0.86149 | | O6 | -0.86812 |
| | O7 | -0.85854 | | O50 | -0.89235 |
| | O8 | -0.88575 | | O51 | -0.86811 |
| | Average O | -0.87347 | | O53 | -0.86789 |
| | N9 | -0.59168 | | O54 | -0.88691 |
| | N10 | -0.58181 | | Average O | -0.87789 |
| | N11 | -0.59308 | | N7 | -0.60188 |
| | N12 | -0.60405 | | N8 | -0.59975 |
| | Average N | -0.59266 | | N9 | -0.60094 |

| | | | | | |
|---|---|---|---|---|---|
| | | | | N10 | -0.59290 |
| | | | | Average N cyclen | -0.59887 |
| | | | | N45 | -0.59119 |

**Table S4.** Charge distribution of the ion center in the $[DOTA]^{4-}$ and $[3p\text{-}C\text{-}DEPA]^{5-}$ complexes

| **Chelators** | **Ions** | **Natural Population Analysis (NPA)** | **Chelators** | **Ions** | **Natural Population Analysis (NPA)** |
|---|---|---|---|---|---|
| $[DOTA]^{4-}$ | $Lu^{3+}$ | 1.60296 | $[3p\text{-}C\text{-}DEPA]^{5-}$ | $Lu^{3+}$ | 1.77991 |
| | $Bi^{3+}$ | 2.25461 | | $Bi^{3+}$ | 2.27544 |
| | $Ac^{3+}$ | 1.42212 | | $Ac^{3+}$ | 1.36652 |

**Table S5.** Bond order between the ion and donor atoms in the $[DOTA]^{4-}$ complex.

| **Chelators** | **Ions** | **Bond** | **Wiberg Bond Index (WBI)** |
|---|---|---|---|
| $[DOTA]^{4-}$ | $Lu^{3+}$ | O1-Lu | 0.2712 |
| | | O2-Lu | 0.2852 |
| | | O3-Lu | 0.2705 |
| | | O4-Lu | 0.2836 |
| | | N9-Lu | 0.1383 |
| | | N10-Lu | 0.1453 |
| | | N11-Lu | 0.1422 |
| | | N12-Lu | 0.1368 |
| | | O54-Lu | 0.2343 |
| | $Bi^{3+}$ | O1-Bi | 0.1512 |
| | | O2-Bi | 0.1539 |
| | | O3-Bi | 0.1860 |
| | | O4-Bi | 0.1971 |
| | | N9-Bi | 0.0962 |
| | | N10-Bi | 0.0814 |
| | | N11-Bi | 0.1133 |
| | | N12-Bi | 0.0921 |
| | | O54-Bi | 0.1448 |
| | $Ac^{3+}$ | O1-Ac | 0.3428 |
| | | O2-Ac | 0.3341 |
| | | O3-Ac | 0.3211 |
| | | O4-Ac | 0.3254 |
| | | N9-Ac | 0.1276 |
| | | N10-Ac | 0.1313 |
| | | N11-Ac | 0.1270 |
| | | N12-Ac | 0.1283 |
| | | O54-Ac | 0.2767 |

**Table S6.** Bond order between the ion and donor atoms in the $[3p\text{-}C\text{-}DEPA]^{5-}$ complex

| Chelators | Ions | Bond | Wiberg Bond Index (WBI) |
|---|---|---|---|
| $[3p\text{-}C\text{-}DEPA]^{5-}$ | $Lu^{3+}$ | O4-Lu | 0.1834 |
| | | O5-Lu | 0.2176 |
| | | O6-Lu | 0.2259 |
| | | O65-Lu | 0.2380 |
| | | O67-Lu | 0.1878 |
| | | N7-Lu | 0.0967 |
| | | N8-Lu | 0.1289 |
| | | N9-Lu | 0.0931 |
| | | N10-Lu | 0.0857 |
| | | N56-Lu | 0.0823 |
| | $Bi^{3+}$ | O4-Bi | 0.1344 |
| | | O5-Bi | 0.1919 |
| | | O6-Bi | 0.2060 |
| | | O65-Bi | 0.2274 |
| | | O67-Bi | 0.1510 |
| | | N7-Bi | 0.0542 |
| | | N8-Bi | 0.0615 |
| | | N9-Bi | 0.0629 |
| | | N10-Bi | 0.0527 |
| | | N56-Bi | 0.0715 |
| | $Ac^{3+}$ | O4-Ac | 0.2827 |
| | | O5-Ac | 0.3160 |
| | | O6-Ac | 0.3251 |
| | | O65-Ac | 0.3248 |
| | | O67-Ac | 0.2826 |
| | | N7-Ac | 0.1223 |
| | | N8-Ac | 0.1364 |
| | | N9-Ac | 0.1200 |
| | | N10-Ac | 0.1207 |
| | | N56-Ac | 0.1125 |