ELECTRONIC SUPPLEMENTARY INFORMATION

Title:"**Actinide selectivity of 1,10-phenanthroline-2,9-dicarboxamide and its derivatives: A theoretical prediction followed by experimental validation**"

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Table S1. Percentage electrostatic, Pauli, steric and orbital interactions in the total bonding energy as obtained from energy decomposition analysis using ADF software with a) frozen core approximation and BP86 exchange-correlation functional b) all electron level TZ2P basis sets with BP86 and B3LYP exchange-correlation functionals c) frozen core approximation and BP86 exchange-correlation functional alongwith solvation effect (COSMO model).

a) Small core pseudopotential

Complex Name	**% electrostatic interaction**	**% Pauli interaction**	**% steric interaction**	**% orbital interaction**
[Am-PDAM]$^{3+}$	77.40	-40.53	36.87	63.12
[Eu-PDAM]$^{3+}$	70.05	-31.00	39.05	63.95
[Am-THIOAM]$^{3+}$	65.84	-42.84	23.00	76.99
[Eu-THIOAM]$^{3+}$	65.97	-35.95	30.02	69.97

b) All-electron level calculations

Complex Name	**% electrostatic interaction**		**% Pauli interaction**		**% steric interaction**		**% orbital Interaction**	
	BP86	**B3LYP**	**BP86**	**B3LYP**	**BP86**	**B3LYP**	**BP86**	**B3LYP**
[Am-PDAM]$^{3+}$	77.89	63.30	-38.78	-12.83	39.11	50.47	60.89	49.53
[Eu-PDAM]$^{3+}$	71.98	54.33	-30.81	-0.19	41.17	54.14	58.83	45.86
[Am-THIOAM]$^{3+}$	74.65	61.19	-46.78	-19.32	27.87	41.87	72.13	58.13
[Eu-THIOAM]$^{3+}$	67.73	50.91	-36.62	-3.97	31.11	46.94	68.88	53.06

c) Calculations using solvation effect (COSMO model)

Complex Name	**% electrostatic interaction**	**% Pauli interaction**	**% steric interaction**	**% orbital interaction**
[Am-PDAM]$^{3+}$	61.34	-32.12	29.22	52.98
[Eu-PDAM]$^{3+}$	57.82	-25.59	32.23	50.27
[Am-THIOAM]$^{3+}$	53.99	-35.13	18.86	63.11
[Eu-THIOAM]$^{3+}$	51.44	-57.89	23.55	58.08

Table S2 The calculated BCP properties (electron density [ρ], Laplacian of the electron density [$\nabla^2\rho$]) of the M-N and M-O/M-S bonds in Am^{3+} and Eu^{3+} complexes using B3LYP/TZ2P method

Bond	**M-N**		**M-O/M-S**	
	ρ (e a_0^{-3})	**$(-1/4)\nabla^2\rho$ (e a_0^{-5})**	**ρ (e a_0^{-3})**	**$(-1/4)\nabla^2\rho$ (e a_0^{-5})**
$[Am\text{-}PDAM]^{3+}$	0.0310	-0.0246	0.0449	-0.0428
$[Eu\text{-}PDAM]^{3+}$	0.0233	-0.0187	0.0336	-0.0341
$[Am\text{-}THIOAM]^{3+}$	0.0284	-0.0231	0.0314	-0.0113
$[Eu\text{-}THIOAM]^{3+}$	0.0237	-0.0191	0.0227	-0.0094
$[Am\text{-}PHEN]^{3+}$	0.0349	-0.0286	---	---
$[Eu\text{-}PHEN]^{3+}$	0.0259	-0.0208	---	---

^{1}H and ^{13}C NMR studies of the ligand **7**

A typical example is depicted in Figure 4 giving a first order spectrum of **7** in which aromatic protons show two different types of signal. Protons of the central aromatic ring resonate as a singlet at 7.92 ppm (Figure a) whereas the other aromatic protons exhibit two doublets. Diagnostic signals are provided by the N-CH_2 and N-H protons at 3.53 (q) and 8.65 (t) ppm, respectively. The terminal methyl groups of the alkyl chains resonate as a triplet at 0.85 ppm. Other alkyl protons are found as multiplets at 1.22-1.48 and 1.64-1.74 ppm with the expected 24 and 8 protons integration for each signal respectively. The carbon spectra present the expected 7 signals for the alkyl atoms, the N-CH_2 signal at 39.5 ppm and the expected aromatic CH (6 signals). The amide signal is found at 163.6 ppm (Figure b).

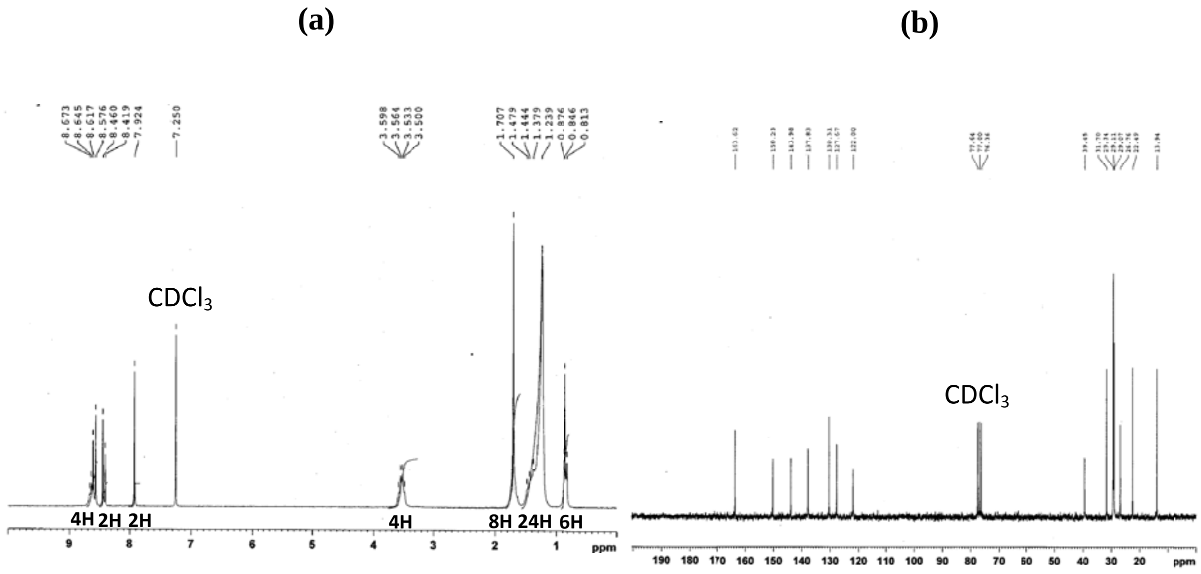

Figure S1:a) ^{1}H NMR spectra of **7** in $CDCl_3$ at 25 °C. b) ^{13}C NMR of **7** in $CDCl_3$.