Supporting Information

TODGA-Inspired Complexant for Minor Actinide Extraction

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General Information

All reagents were purchased from U.S. chemical suppliers, stored according to published protocols, and used as received. All synthesis experiments were performed in oven-dried glassware. Reaction progress was monitored using thin-layer chromatography on glass-backed, normal phase, silica gel plates. R_f values for compounds which resulted in a concentrically observed spot are reported using the conditions listed. Melting point data listed is for a single, uncorrected experiment from an automated melting point apparatus with camera. All reported yields listed are for compounds afforded from automated flash-column chromatography and corrected for residual solvent, if applicable, from ^{1}H NMR spectroscopy. Infrared spectral data was acquired from the (form) listed. All ^{1}H- and ^{13}C NMR data was acquired from a 500 MHz multinuclear spectrometer with broad-band N_2 cryoprobe. Chemical shifts are reported using the δ scale and are referenced to the residual solvent signal: $CDCl_3$ (δ 7.26) for ^{1}H NMR and $CDCl_3$ (δ 77.16) for ^{13}C NMR.[i] Splittings are reported as follows: (s) = singlet, (d) = doublet, (t) = triplet, sextet, and (m) = multiplet. ^{13}C NMR spectra were corrected for ring down using linear back prediction. High resolution mass spectrometry (HRMS) data was obtained QToF in positive ion mode. The following HRMS acquisition parameters were utilized: Capillary voltage 0.5–1.5 KV, Sampling cone 40 V, Source offset 60, Source temperature 80 °C, Desolvation 350 °C, Cone gas 30 L/h, Desolvation gas 600 L/h, lock spray capillary voltage 0.2 KV, lock mass: 556.2771 in positive mode (leucine enkephalin).

Counting Statistics

For each solvent extraction experiment (**Tables S1, S2, S4, S5, S7,** and **S8**), a critical limit (L_c, Equation S1) was determined to assess whether the net count was statistically significant at the 95% confidence level.

$$L_C = 1.645\sqrt{2B} \qquad \textit{Equation S1}$$

If the net counts per minute (cpm) were below the calculated L_c , a footnote was included in the corresponding data table (**Tables S5 and S8**) to indicate that the activity was not detected. In such cases, an upper limit (L_U , Equation S2) was reported at the 95% confidence level.

$$L_U = cpm_{net} + 1.645\sqrt{(cpm_{net} + 2B)} \qquad \textit{Equation S2}$$

The uncertainty with each distribution value (D_M, where M = $^{241}Am^{3+}$ or $^{154\text{-}152}Eu^{3+}$) was determined based on the counting error (2σ) as defined by *Equation S3*.

$$\sigma_{D_M} = D_M \times \sqrt{\left(\frac{\left(\sqrt{{\sigma_{org}}^2+{\sigma_{blank}}^2}\right)}{Net\ cpm_{org}}\right)^2 + \left(\frac{\left(\sqrt{{\sigma_{aq}}^2+{\sigma_{blank}}^2}\right)}{Net\ cpm_{aq}}\right)^2} \qquad \textit{Equation S3}$$

The uncertainty in the average D_M was determined using Equation S4 and is presented in the summary tables (**Table S3, S6** and **S9**).

$$\sigma_{Avg(D_M)} = \frac{\sqrt{{\sigma_{D_1}}^2+{\sigma_{D_2}}^2}}{2} \qquad \textit{Equation S4}$$

D_1 and D_2 represents the individual D_M values obtained from duplicate samples corresponding to each data point.

Procedure for Synthesis of 1

2,2'-((oxalylbis(3,1-phenylene))bis(oxy))bis(N,N-dipropylacetamide) (**1**). To a 20 mL vial equipped with a magnetic stir bar was added, 3,3′-dihydroxybenzil (1.0000 g, 4.13 mmol, 1.00 equiv) followed by dissolution in acetonitrile (4.13 mL, 1 M). Potassium carbonate (K_2CO_3, 1.2563 g, 9.09 mmol, 2.20 equiv) and N,N-diisopropylacetamide (1.49 ml, 8.43 mmol, 2.04 equiv) were then charged successively and the reaction mixture was heated to 82 °C and continued for 12 hours. The progress of the reaction was monitored by TLC and 1H NMR until completion. After completion, the reaction mixture was cooled to room temperature, diluted with ethyl acetate, and transferred to a separatory funnel. The crude mixture was washed with water (3 × 5 mL) and the organic layer was separated. The aqueous phase was back-extracted with ethyl acetate (3 × 5 mL). The combined organic extracts were washed with 5 mL of a saturated aqueous brine solution, dried over anhydrous sodium sulfate (Na_2SO_4), and concentrated under reduced pressure at ambient temperature using a rotary evaporator. The crude product was adsorbed onto silica gel and purified by automated flash chromatography on silica gel using an isopranol:dichloromethane gradient mobile phase. R_f = 0.80, 10% isopropanol: dichloromethane on normal phase silica gel TLC plate; isolated yield 2.0571 g, 95%; yellow solid; melting point 54.0-54.2 °C. ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.50 – 7.45 (m, 4H), 7.39 (t, *J* = 8.0 Hz, 2H), 7.29 – 7.26 (m, 2H), 4.74 (s, 4H), 3.30 (t, *J* = 7.5 Hz, 4H), 3.24 (t, *J* = 7.7 Hz, 4H), 1.64 (sextet, *J* = 7.5 Hz, 4H), 1.56 (sextet, *J* = 7.5 Hz, 4H), 0.94 (t, *J* = 7.3 Hz, 6H), 0.86 (t, *J* = 7.3 Hz, 6H); ^{13}C NMR (126 MHz, $CDCl_3$) δ 194.1, 166.7, 158.8, 134.3, 130.3, 123.9, 122.4, 114.0, 66.9, 48.9, 47.1, 22.3, 20.8, 11.42, 11.36; IR (ATR-solid): ῡmax = 2965, 2934, 2875, 1645, 1582, 1429, 1299, 1233, 1216, 1073, 777, 741, 646, 599, 476 cm^{-1}; HRMS (QToF): m/z: $[M+Na]^+$ Calcd for $C_{30}H_{40}N_2O_6Na^+$ $[M+Na]^+$ 547.2782, found 547.2784.

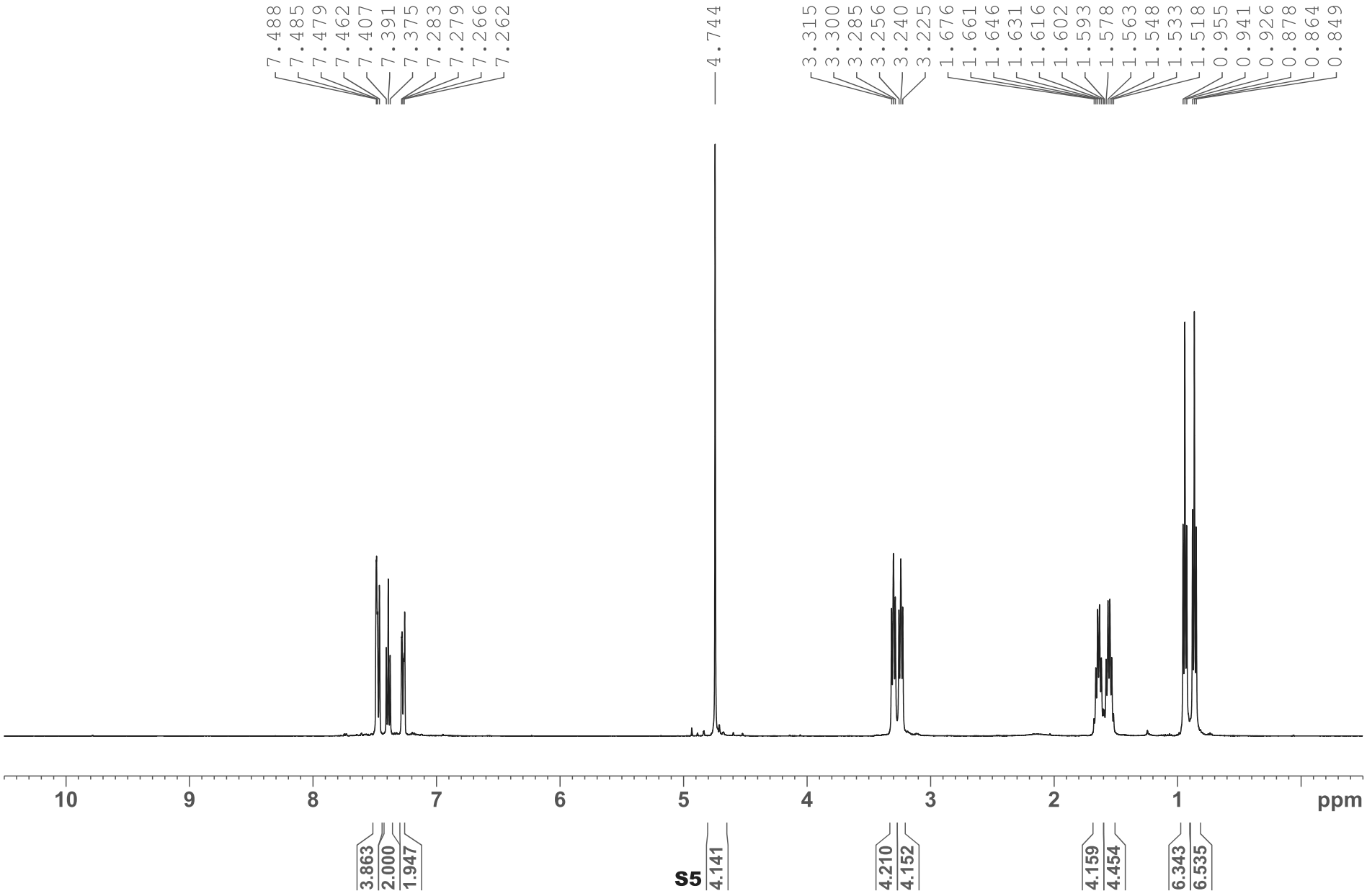
Procedure for Synthesis of 3

2,2',2'',2'''-(((pyridine-2,6-diylbis(1,2,4-triazine-3,5,6-triyl))tetrakis(benzene-3,1-diyl)) tetrakis (oxy))tetrakis(N,N-dipropylacetamide) (**3**). To a 20 mL reaction vial equipped with a magnetic stir bar at ambient temperature was charged (*2Z*,*6Z*)-pyridine-2,6-bis(carbohydrazonamide) (**2**) (0.2530 g, 1.31 mmol, 1.00 equiv) and 2,2'-((oxalylbis(3,1-phenylene))bis(oxy))bis(N,N-dipropylacetamide (**1**) (1.3736 g, 2.62 mmol, 2.00 equiv). The mixture was dissolved in anhydrous THF (5.5 mL, 0.24M) and subsequently heated to 66 °C and continued for 18 hours. Reaction progress was monitored by TLC and ^{1}H NMR. The crude product was then adsorbed onto silica gel and purified by automated flash chromatography on silica gel using an isopropanol:dichloromethane gradient mobile phase. R_f = 0.75, 10% isopropanol: dichloromethane on normal phase silica gel TLC plate; isolated yield 1.3639 g, 89%; yellow oil; ^{1}H NMR (500 MHz, $CDCl_3$) δ 8.87 (d, *J* = 7.8 Hz, 2H), 8.20 (t, *J* = 7.7 Hz, 1H), 7.39 (d, *J* = 7.5 Hz, 2H), 7.33 – 7.27 (m, 8H), 7.19 (d, *J* = 7.5 Hz, 2H), 7.07 (d, *J* = 8.1 Hz, 4H), 4.65 (s, 4H), 4.59 (s, 4H), 3.33 – 3.16 (m, 16H), 0.96 – 0.78 (m, 24H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 167.1, 167.0, 160.4, 158.6, 158.4, 156.3, 153.5, 138.6, 136.7, 136.6, 130.0, 129.9x (overlaps with 130.0), 126.0, 123.4, 123.0, 118.2, 117.0, 115.9, 115.4, 67.3, 67.2x (overlaps with 67.3), 49.0, 48.9, 47.64, 47.62, 22.3, 22.2, 20.76, 20.7x (overlaps with 20.76), 11.5, 11.4x (overlaps with 11.5), 11.39, 11.36; IR (ATR-neat): ῡmax = 2962, 2932, 2874, 1644, 1581, 1430, 1372, 1221, 1144, 1071, 874, 787, 699, 481 cm^{-1}; HRMS (QToF): m/z: $[M+Na]^+$ Calcd for $C_{67}H_{83}N_{11}O_8Na^+$ $[M+Na]^+$ 1192.6287, found 1192.6324.

2,2'-((oxalylbis(3,1-phenylene))bis(oxy))bis(N,N-dipropylacetamide) (1)

500 MHz ^{1}H NMR Spectrum in $CDCl_3$

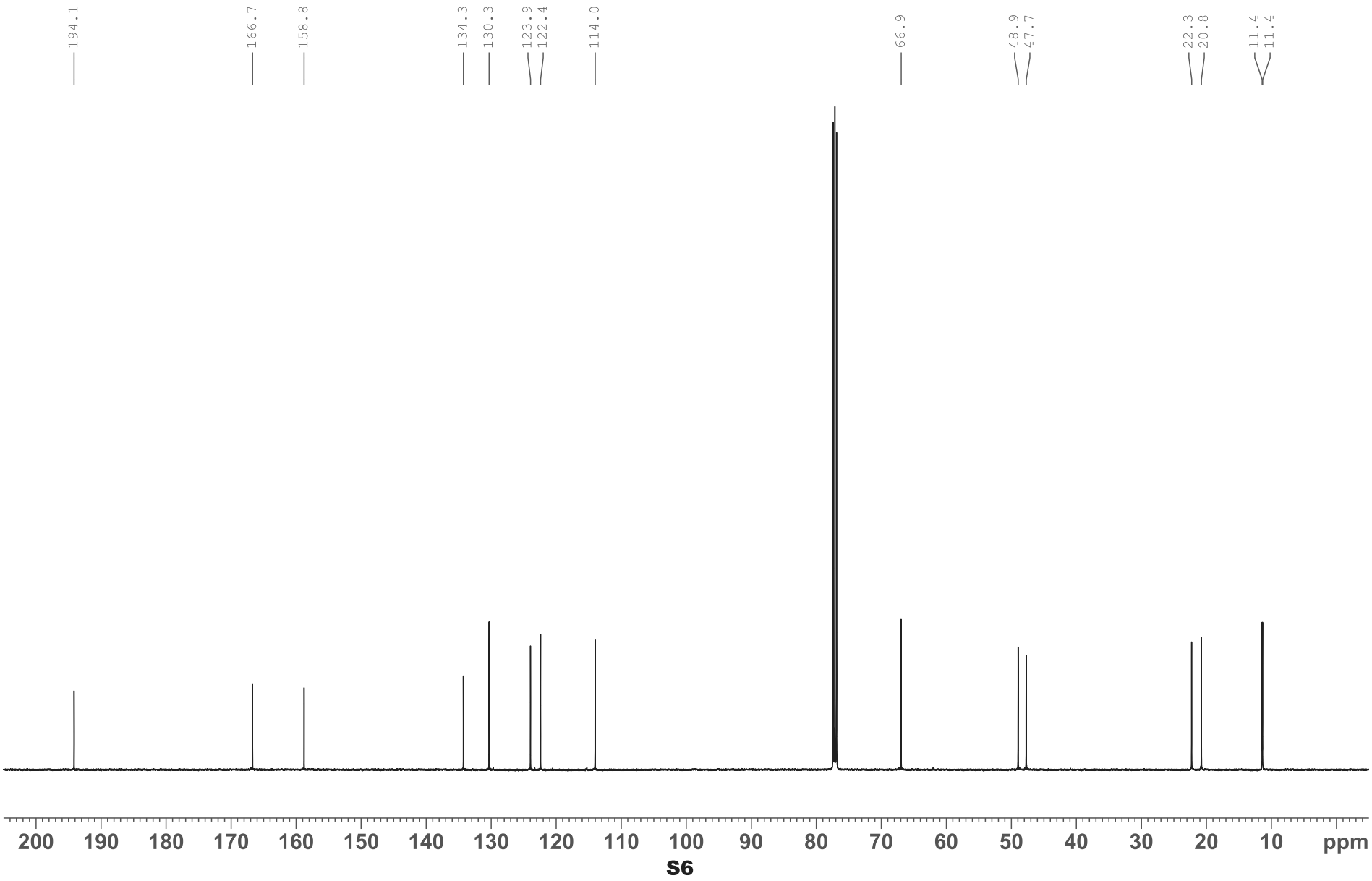
KG-A-159



2,2'-((oxalylbis(3,1-phenylene))bis(oxy))bis(N,N-dipropylacetamide) (1)

125 MHz $^{13}C\{^{1}H\}$ NMR Spectrum in $CDCl_3$

KG-A-159

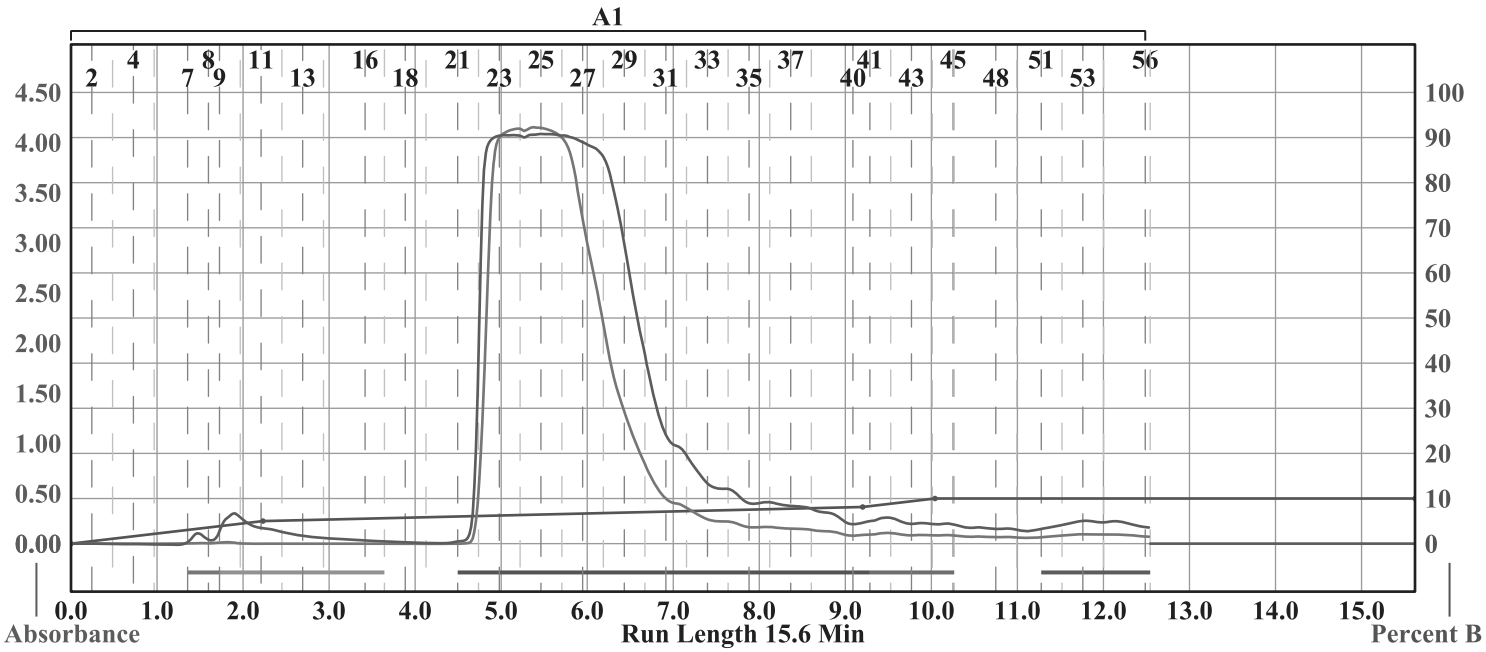


Sample: KG-A-63 | CombiFlash NEXTGEN : 222G00894 | Monday 25 November 2024 10:38AM

Column: C18 40-50g
Flow Rate: 60 ml/min
Equilibration Volume: 236.6 ml
Initial Waste: 0.0 ml
Air Purge: 0.0 min
Solvent A: Dichloromethane
Solvent B: Iso Propyl Alcohol

Peak Tube Volume: Max.
Non-Peak Tube Volume: Max.
Loading Type: Solid
Wavelength 1 (red): 214nm
Peak Width: 2 min
Threshold: 0.20 AU
Wavelength 2 (purple): 254nm

Run Notes: 2,2'-((oxalylbis(3,1-phenylene))bis(oxy))bis(N,N-dipropylacetamide) (1)



Rack A

71	72	73	74	75
70	69	68	67	66
61	62	63	64	65
60	59	58	57	56
51	52	53	54	55
50	49	48	47	46
41	42	43	44	45
40	39	38	37	36
31	32	33	34	35
30	29	28	27	26
21	22	23	24	25
20	19	18	17	16
11	12	13	14	15
10	9	8	7	6
1	2	3	4	5

16 mm x 100 mm Tubes

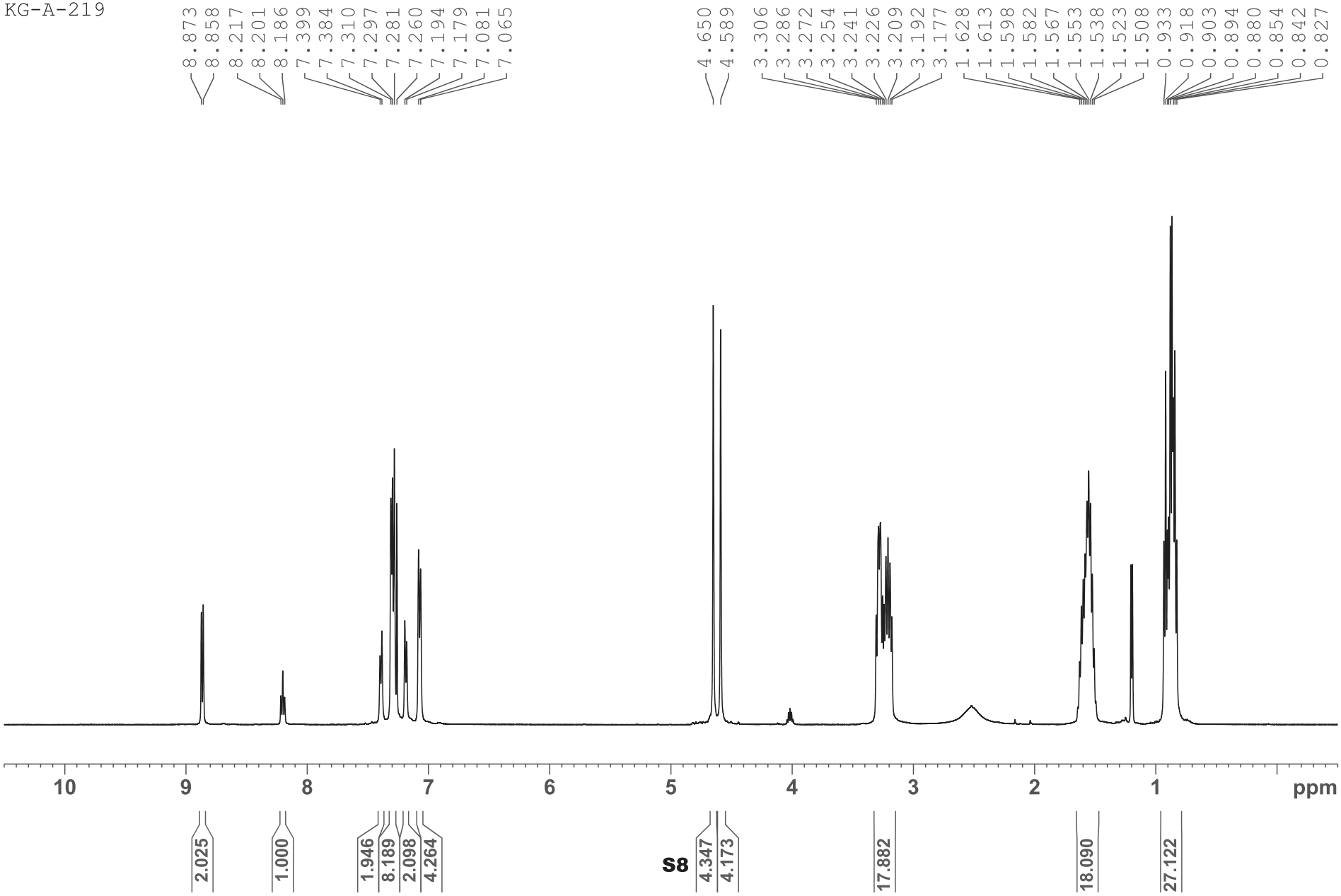
Peak #	Start Tube	End Tube
1	A:7	A:8
2	A:9	A:16
3	A:21	A:40
4	A:41	A:45
5	A:51	A:56

Duration	%B	Solvent A	Solvent B
0.0	0.0	Dichloromethane	Iso Propyl Alcohol
2.2	5.0	Dichloromethane	Iso Propyl Alcohol
7.0	8.1	Dichloromethane	Iso Propyl Alcohol
0.8	10.0	Dichloromethane	Iso Propyl Alcohol
5.6	10.0	Dichloromethane	Iso Propyl Alcohol

2,2',2'',2'''-(((pyridine-2,6-diylbis(1,2,4-triazine-3,5,6-triyl))tetrakis (benzene-3,1-diyl)) tetrakis(oxy))tetrakis(N,N-dipropylacetamide) (3)

500 MHz ^{1}H NMR Spectrum in $CDCl_3$

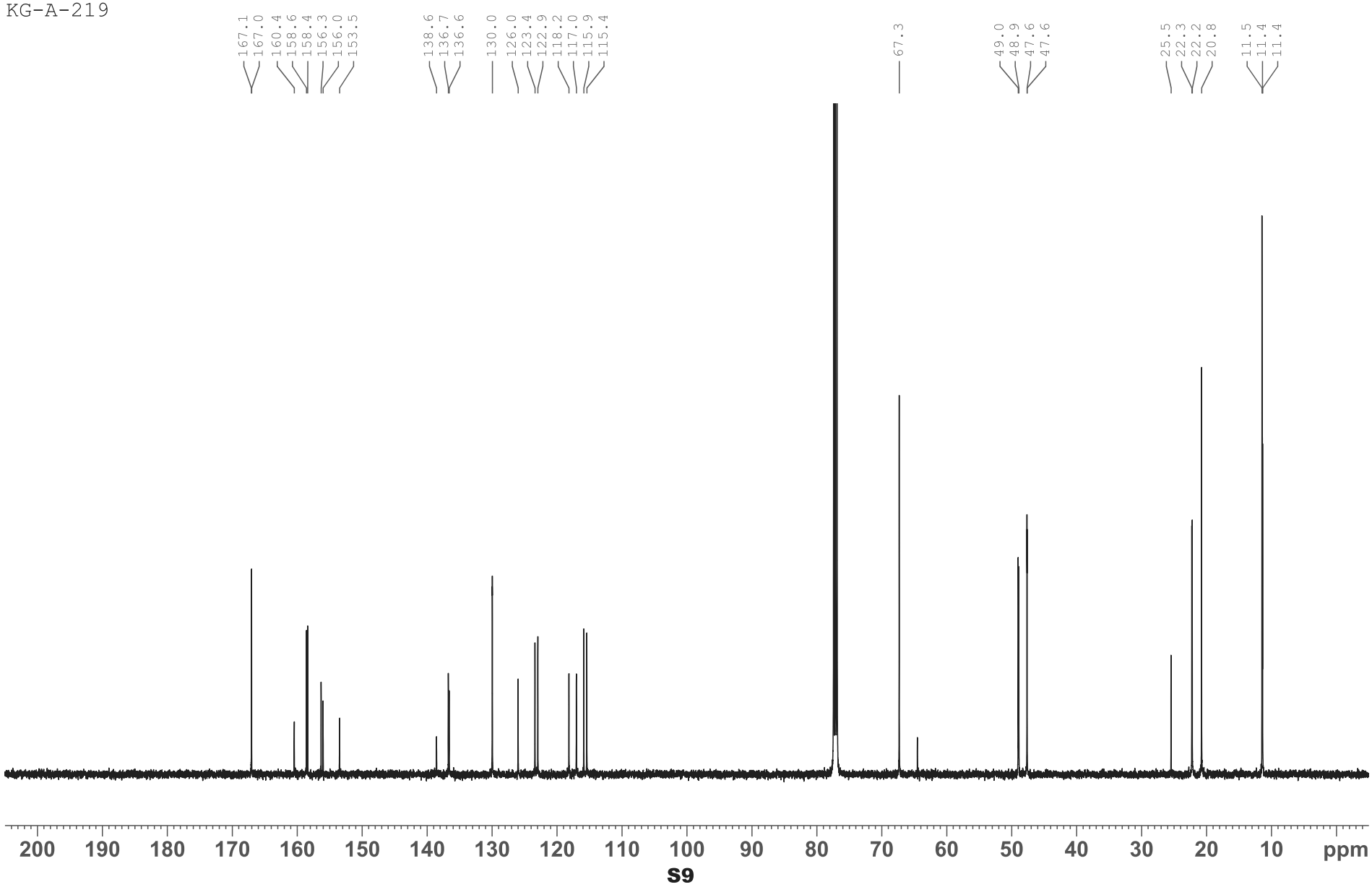
KG-A-219



2,2',2'',2'''-(((pyridine-2,6-diylbis(1,2,4-triazine-3,5,6-triyl))tetrakis (benzene-3,1-diyl)) tetrakis(oxy))tetrakis(N,N-dipropylacetamide) (3)

125 MHz $^{13}C\{^{1}H\}$ NMR Spectrum in $CDCl_3$

KG-A-219



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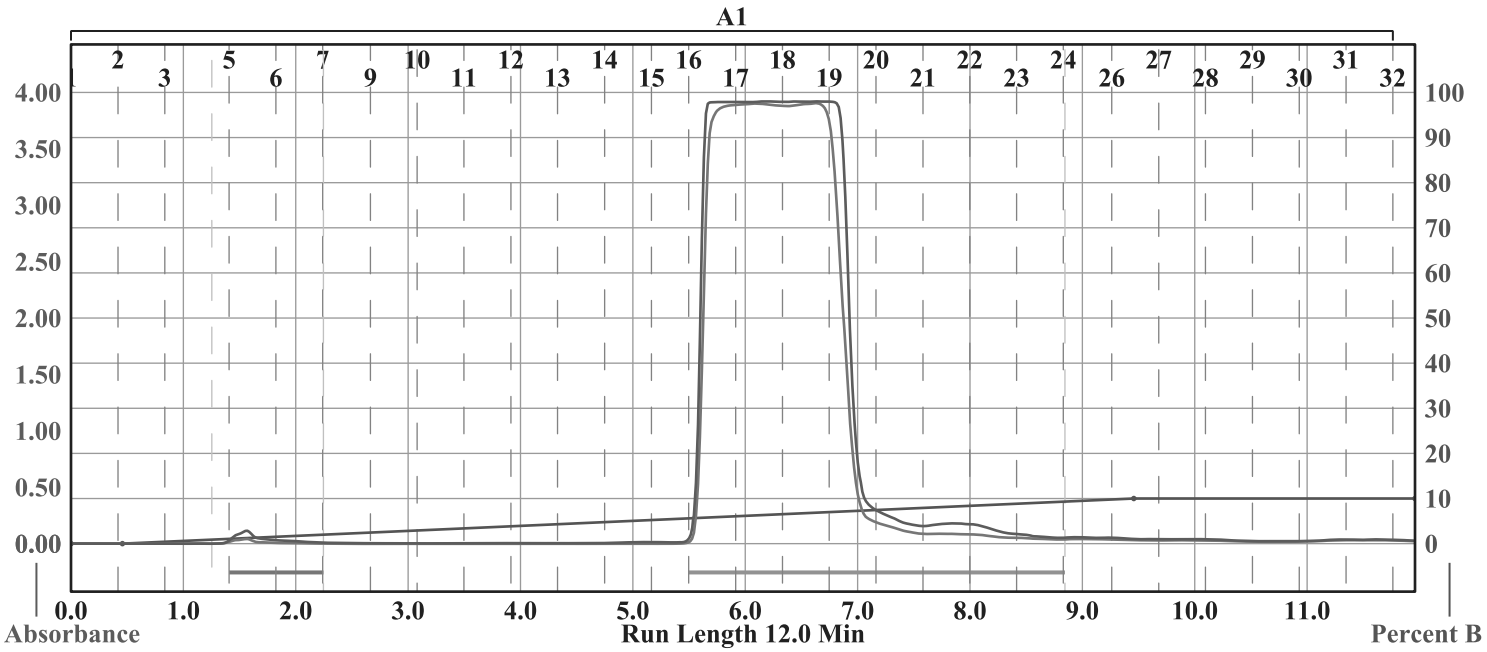
Column: Silica 40g Gold
SN: E0040108585151DC Lot: 331818203Z
Flow Rate: 60 ml/min
Equilibration Volume: 274.6 ml
Initial Waste: 0.0 ml
Air Purge: 1.0 min
Solvent A: Dichloromethane
Solvent B: Iso Propyl Alcohol

Peak Tube Volume: Max.
Non-Peak Tube Volume: Max.
Loading Type: Solid
Wavelength 1 (red): 254nm
Peak Width: 2 min
Threshold: 0.20 AU
Wavelength 2 (purple): 280nm

Run Notes: 2,2',2",2"'-(((pyridine-2,6-diylbis(1,2,4-triazine-3,5,6-triyl))tetrakis(benzene-3,1-diyl)) tetrakis (oxy))tetrakis(N,N-dipropylacetamide) (3)



Rack A

70	69	68	67	66
61	62	63	64	65
60	59	58	57	56
51	52	53	54	55
50	49	48	47	46
41	42	43	44	45
40	39	38	37	36
31	32	33	34	35
30	29	28	27	26
21	22	23	24	25
20	19	18	17	16
11	12	13	14	15
10	9	8	7	6
1	2	3	4	5

18 mm x 150 mm Tubes

Peak #	Start Tube	End Tube
1	A:5	A:7
2	A:16	A:24

Duration	%B	Solvent A	Solvent B
0.0	0.0	Dichloromethane	Iso Propyl Alcohol
0.5	0.0	Dichloromethane	Iso Propyl Alcohol
9.0	10.0	Dichloromethane	Iso Propyl Alcohol
2.5	10.0	Dichloromethane	Iso Propyl Alcohol

Table S1. Raw Data to Accompany Figure 2(a). $^{241}Am^{3+}$ Distribution (*D*)-values, at 1M $HNO_{3(aq)}$ as *f*(t) in 1-octanol

Time (min.)	Sample ID	CPM	Error (%)	Net CPM	Recovery (%)	D_{Am} (org./aq.)	D_{Am} Avg.
5	org. - 1	2273	0.66	2253	97.9	1.6162	1.7061
	org. - 2	2493	0.63	2473	103.3	1.7959	
	aq. - 3	1414	0.84	1394			
	aq. - 4	1397	0.85	1377			
10	org. - 5	2525	0.63	2505	94.2	2.4950	2.5900
	org. - 6	2764	0.60	2744	101.1	2.6849	
	aq. - 7	1024	0.99	1004			
	aq. - 8	1042	0.98	1022			
15	org. - 9	2531	0.63	2511	93.9	2.5415	2.6052
	org.- 10	2745	1.00	2725	100.5	2.6690	
	aq. - 11	1008	1.00	988			
	aq. - 12	1041	1.00	1021			
30	org. -13	2537	1.00	2517	94.1	2.5476	2.6641
	org. -14	2770	1.00	2750	100.3	2.7806	
	aq. - 15	1008	1.00	988			
	aq. - 16	1009	1.00	989			
45	org. -17	2516	1.00	2496	92.1	2.6638	2.8357
	org. -18	2832	1.00	2812	100.6	3.0075	
	aq. - 19	957	1.02	937			
	aq. - 20	955	1.02	935			
60	org. -21	2593	1.00	2573	94.3	2.7314	2.7610
	org.-22	2699	1.00	2679	97.7	2.7906	
	aq. - 23	962	1.02	942			
	aq. - 24	980	1.01	960			
90	org. -25	2549	1.00	2529	92.4	2.7670	2.9065
	org. -26	2865	1.00	2845	101.4	3.0460	
	aq. - 27	934	1.04	914			
	aq. - 28	954	1.02	934			
120	org. -29	2563	1.00	2543	93.4	2.7111	2.8187
	org. -30	2876	1.00	2856	102.8	2.9262	
	aq. - 31	958	1.02	938			
	aq. - 32	996	1.00	976			
A_0	A_0 - 33	3802	1.00	3782			

A_0	A_0 - 34	3691	1.00	3671
A_0	Avg.	3746.5		3726.5
Blank	Avg.	20	6.99	

Table S2. Raw Data to Accompany Figure 2(a). $^{154\text{-}152}Eu^{3+}$ Distribution (D)-values, at 1M $HNO_{3(aq)}$ as f(t) in 1-octanol

Time (min.)	Sample ID	CPM	Error (%)	Net CPM	Recovery (%)	D_{Eu} (org./aq.)	D_{Eu} (Avg.)
	org. - 1	231	2.08	99	93.7	0.0260	0.0277
5	org. - 2	246	2.01	114		0.0294	
	aq. - 3	3941	0.50	3809			
	aq. - 4	4012	0.50	3880			
	org. - 5	245	2.02	113	92.8	0.0301	0.0286
10	org. - 6	235	2.07	103		0.0271	
	aq. - 7	3887	0.51	3755			
	aq. - 8	3939	0.50	3807			
	org. - 9	237	2.06	105	88.1	0.0294	0.0304
15	org. -10	246	2.02	114		0.0313	
	aq. - 11	3699	1.00	3567			
	aq. - 12	3775	1.00	3643			
	org. -13	248	2.01	116	91.9	0.0312	0.0312
30	org. -14	249	2.00	117		0.0312	
	aq. - 15	3846	1.00	3714			
	aq. - 16	3881	1.00	3749			
	org. -17	239	2.05	107	100.4	0.0262	0.0280
45	org. -18	254	1.98	122		0.0298	
	aq. - 19	4211	1.00	4079			
	aq. - 20	4221	1.00	4089			
	org. -21	252	1.99	120	102.8	0.0288	0.0295
60	org. -22	260	1.96	128		0.0301	
	aq. - 23	4296	1.00	4164			
	aq. - 24	4387	0.99	4255			
	org. -25	262	1.95	130	104.5	0.0308	0.0312
90	org. -26	268	1.93	136		0.0316	
	aq. - 27	4357	1.00	4225			
	aq. - 28	4440	1.00	4308			
	org. -29	251	1.99	119	103.5	0.0284	0.0293
120	org. -30	262	1.95	130		0.0302	
	aq. - 31	4329	1.00	4197			
	aq. - 32	4433	1.00	4301			
A_0	A_0 - 33	4293	1.00	4161			

A_0	A_0 - 34	4309	1.00	4177
A_0	Avg.	4301		4169
Blank	Avg.	132	2.76	

Table S3. Summary D_M for Tables S1 and S2

Time (min)	D_{Am} Avg.	D_{Am} Uncertainty (2σ)	D_{Eu} Avg.	D_{Eu} Uncertainty (2σ)	SF (Avg.) (D_{Am}/D_{Eu})	SF Uncertainty (2σ)
5	1.7061	0.2542	0.0277	0.0048	61.6	1.4971
10	2.5900	0.2686	0.0286	0.0043	90.6	23.0919
20	2.6052	0.0901	0.0304	0.0026	85.8	1.4834
30	2.6641	0.3295	0.0312	0.0000	85.3	10.6510
45	2.8357	0.4860	0.0280	0.0051	101.2	1.0589
60	2.7610	0.0837	0.0295	0.0018	93.8	2.8481
90	2.9065	0.3947	0.0312	0.0011	93.2	9.2795
120	2.8187	0.3043	0.0293	0.0026	96.2	1.6915

Table S4. Raw Data to Accompany Figure 2. (b) $^{241}Am^{3+}$ Distribution (D)-values, f($HNO_{3(aq)}$) with Initial Concentration in 1-octanol of 25 mM

$[HNO_3]$ (M)	Sample ID	CPM	Error (%)	Net CPM	Recovery (%)	D_{Am} (org./aq.)	D_{Am} Avg.
	org. - 1	1769	0.75	1751	98.7	0.9491	0.9890
0.25	org. - 2	1970	0.71	1952	105.6	1.0290	
	aq. - 3	1863	0.73	1845			
	aq. - 4	1915	0.72	1897			
	org. - 5	2335	0.65	2317	93.9	1.9686	2.0339
0.5	org. - 6	2491	0.63	2473	98.2	2.0993	
	aq. - 7	1195	0.91	1177			
	aq. - 8	1196	0.91	1178			
	org. - 9	2422	0.64	2404	91.3	2.5739	2.6643
0.75	org. -10	2668	1.00	2650	98.8	2.7547	
	aq. - 11	952	1.02	934			
	aq. - 12	980	1.01	962			
	org. -13	2549	1.00	2531	92.0	3.0239	3.1362
1	org. -14	2763	1.00	2745	98.1	3.2485	
	aq. - 15	855	1.08	837			
	aq. - 16	863	1.08	845			
	org. -17	2853	1.00	2835	92.5	4.3953	4.5108
2	org. -18	3025	1.00	3007	97.2	4.6262	
	aq. - 19	663	1.23	645			
	aq. - 20	668	1.22	650			
	org. -21	2976	1.00	2958	90.9	6.6027	6.6260
3	org.-22	3090	1.00	3072	94.3	6.6494	
	aq. - 23	466	1.46	448			
	aq. - 24	480	1.44	462			
	org. -25	3068	1.00	3050	92.9	7.5682	7.5611
4	org. -26	3168	1.00	3150	96.0	7.5540	
	aq. - 27	421	1.54	403			
	aq. - 28	435	1.52	417			
	org. -29	2872	1.00	2854	90.5	6.8937	7.0554
5	org. -30	3078	1.00	3060	96.5	7.2170	
	aq. - 31	432	1.52	414			
	aq.- 32	442	1.50	424			
0.25	A_0 - 33	3663	1.00	3645			
0.5	A_0 - 34	3737	1.00	3719			

0.75	A_0 - 35	3673	1.00	3655
1	A_0 - 36	3677	1.00	3659
2	A_0 - 37	3780	0.99	3762
3	A_0 - 38	3766	1.00	3748
4	A_0 - 39	3733	1.00	3715
5	A_0 - 40	3628	1.00	3610
Blank	Avg.	18	7.36	

Table S5. Raw Data to Accompany Figure 2(b). $^{154\text{-}152}Eu^{3+}$ Distribution (D)-values, $f(HNO_{3(aq)})$ with Initial Concentration in 1-octanol of 25 mM

$[HNO_3]$ (M)	Sample ID	CPM	Error (%)	Net CPM	Recovery (%)	D_{Eu} (org./aq.)	D_{Eu} Avg.
0.25	org. - 1	165	2.46	16[a]	107.8	0.0042	0.0046
	org. - 2	168	2.44	19[b]	109.2	0.0049	
	aq. - 3	3963	0.71	3814			
	aq. - 4	4011	0.7	3862			
0.5	org. - 5	190	2.3	41	107.6	0.0103	0.0103
	org. - 6	190	2.29	41	108.7	0.0102	
	aq. - 7	4114	0.71	3965			
	aq. - 8	4161	0.71	4012			
0.75	org. - 9	216	2.15	67	106.3	0.0166	0.0149
	org. -10	204	2.22	55	108.1	0.0133	
	aq. - 11	4196	0.71	4047			
	aq. - 12	4282	0.71	4133			
1	org.- 13	218	2.14	69	107.2	0.0174	0.0166
	org.- 14	214	2.16	65	111.0	0.0157	
	aq. - 15	4118	0.71	3969			
	aq. - 16	4279	0.71	4130			
2	org. -17	242	2.03	93	108.2	0.0231	0.0252
	org. -18	261	1.96	112	110.5	0.0273	
	aq. - 19	4168	0.71	4019			
	aq. - 20	4245	0.71	4096			
3	org. -21	291	1.85	142	109.6	0.0347	0.0327
	org.-22	277	1.9	128	111.2	0.0307	
	aq. - 23	4239	0.71	4090			
	aq. - 24	4317	0.71	4168			
4	org. -25	287	1.87	138	107.8	0.0348	0.0340
	org.- 26	291	1.85	142	115.8	0.0331	
	aq. - 27	4114	0.71	3965			
	aq. - 28	4437	0.71	4288			
5	org.- 29	412	1.56	263	115.9	0.0623	0.0463
	org.- 30	281	0.89	132	116.0	0.0303	
	aq. - 31	4369	0.71	4220			
	aq. - 32	4507	0.71	4358			
0.25	A_0 - 33	3828	0.71	3679			

0.5	A_0 - 34	4001	0.71	3852
0.75	A_0 - 35	4150	0.7	4001
1	A_0 - 36	4046	0.71	3897
2	A_0 - 37	4076	0.71	3927
3	A_0 - 38	4132	0.71	3983
4	A_0 - 39	4083	0.71	3934
5	A_0 - 40	4126	0.71	3977
Blank	Avg.	149	2.59	

[a,b]Net cpm is below the critical limit (L_C) and therefore must be reported as not detected, L_C = 20 (95% confidence level).Upper limits (L_U) at the 95% confidence level: [a]L_U = 45, [b]L_U =48.

Table S6. Summary D_M for Tables S4 and S5

$[HNO_3]$ (M)	D_{Am} Avg.	D_{Am} Uncertainty (2σ)	D_{Eu} Avg.	D_{Eu} Uncertainty (2σ)	SF (Avg.) (D_{Am}/D_{Eu})	SF Uncertainty (2σ)
0.25	0.9890	0.1131	0.0046	0.0010	217.0	24.1
0.5	2.0339	0.1849	0.0103	0.0002	197.9	21.3
0.75	2.6643	0.2557	0.0149	0.0046	178.4	72.9
1	3.1362	0.3177	0.0166	0.0023	189.4	45.9
2	4.5108	0.3264	0.0252	0.0059	178.7	29.4
3	6.6260	0.0660	0.0327	0.0057	202.5	37.3
4	7.5611	0.0202	0.0340	0.0024	222.6	15.1
5	7.0554	0.4572	0.0463	0.0453	152.4	180.5

Table S7. Raw Data to Accompany Figure 2(c). Distribution of $^{241}Am^{3+}$ as a Function of Complexant Concentration in 1-Octanol Log-Log plot

[Ligand] (mM)	Sample ID	CPM	Error (%)	Net CPM	Recovery (%)	D_{Am} (org./aq.)	D_{Am} Avg.
6.25	org. - 1	722	1.18	689	103.1	0.1874	0.1895
	org. - 2	778	1.13	745	109.4	0.1917	
	aq. - 3	3710	1.00	3677			
	aq. - 4	3919	1.00	3886			
12.5	org. - 5	1916	1.00	1883	96.5	0.8547	0.8610
	org. - 6	2046	1.00	2013	102.3	0.8673	
	aq. - 7	2236	1.00	2203			
	aq. - 8	2354	1.00	2321			
25	org. - 9	3109	1.00	3076	93.4	3.4915	3.6202
	org. -10	3362	1.00	3329	99.6	3.7489	
	aq. -11	914	1.05	881			
	aq. -12	921	1.04	888			
37.5	org. -13	3268	1.00	3235	87.6	6.8105	7.2875
	org. -14	3659	1.00	3626	96.6	7.7645	
	aq. - 15	508	1.40	475			
	aq. - 16	500	1.41	467			
50	org. -17	3578	1.00	3545	90.5	12.3090	13.3527
	org. -18	4064	1.00	4031	101.8	14.3964	
	aq. -19	321	1.76	288			
	aq. - 20	313	1.79	280			
62.5	org. -21	3753	1.00	3720	92.3	19.7872	20.1905
	org.-22	3987	1.00	3954	97.9	20.5938	
	aq. - 23	221	2.13	188			
	aq. - 24	225	2.11	192			
75	org. -25	3718	1.00	3685	90.4	25.4138	26.1863
	org. -26	3969	1.00	3936	96.4	26.9589	
	aq. - 27	178	2.37	145			
	aq. - 28	179	2.36	146			
A_0	A_0 - 29	4293	1.00	4260			
A_0	A_0 - 30	4243	1.00	4210			
A_0	Avg.	4268		4235			
Blank	Avg.	33	5.52				

Table S8. Raw Data to Accompany Figure 2(c). Distribution of $^{154\text{-}152}Eu^{3+}$ as a Function of Complexant Concentration in 1-Octanol Log-Log plot

[Ligand] (mM)	Sample ID	CPM	Error (%)	Net CPM	Recovery (%)	D_{Eu} (org./aq.)	D_{Eu} Avg.
6.25	org. - 1	150	2.58	10[a]	61.0	0.0045	0.0059
	org. - 2	157	2.52	17[b]	63.3	0.0074	
	aq. - 3	2364	0.65	2224			
	aq. - 4	2440	0.64	2300			
12.5	org. - 5	163	2.48	23[c]	75.4	0.0084	0.0097
	org. - 6	171	2.42	31	77.3	0.0111	
	aq. - 7	2879	0.59	2739			
	aq. - 8	2939	0.58	2799			
25	org. - 9	307	1.80	167	91.1	0.0527	0.0531
	org. -10	310	1.79	170	91.5	0.0535	
	aq. - 11	3308	1.00	3168			
	aq. - 12	3319	1.00	3179			
37.5	org. -13	539	1.36	399	101.4	0.1203	0.1249
	org. -14	584	1.31	444	105.7	0.1295	
	aq. -15	3456	1.00	3316			
	aq. -16	3568	1.00	3428			
50	org. -17	794	1.12	654	106.7	0.2011	0.2080
	org. -18	854	1.08	714	110.2	0.2149	
	aq. - 19	3392	1.00	3252			
	aq. - 20	3462	1.00	3322			
62.5	org. -21	1116	1.00	976	109.9	0.3202	0.3148
	org.-22	1073	1.00	933	107.8	0.3095	
	aq. - 23	3188	1.00	3048			
	aq. - 24	3155	1.00	3015			
75	org. -25	1279	1.00	1139	107.7	0.4062	0.4075
	org. -26	1308	1.00	1168	109.9	0.4088	
	aq. - 27	2944	1.00	2804			
	aq. - 28	2997	1.00	2857			
A_0	A_0 - 29	3621	1.00	3481			
A_0	A_0 - 30	3983	1.00	3843			
A_0	Avg.	3802		3662			
Blank	Avg.	140	2.67				

[a,b,c] Net cpm is below the critical limit (L_C) and therefore must be reported as not detected, L_C = 28 (95% confidence level).Upper limits (L_U) at the 95% confidence level: $^{a}L_U$ = 38, $^{b}L_U$ = 45, $^{c}L_U$ = 51.

Table S9. Summary D_M for Tables S7 and S8

[Ligand] (mM)	D_{Am} Avg.	D_{Am} Uncertainty (2σ)	D_{Eu} Avg.	D_{Eu} Uncertainty (2σ)	SF (Avg.) (D_{Am}/D_{Eu})	SF Uncertainty (2σ)
6.25	0.189547	0.0061275	0.0059439	0.0040940	31.9	22.3
12.5	0.861021	0.0177555	0.0097363	0.0037875	88.4	33.2
25	3.62018	0.3640001	0.0530953	0.0010766	68.2	5.5
37.5	7.28749	1.3490574	0.1249236	0.0130050	58.3	4.7
50	13.35273	2.9520305	0.2080189	0.0195497	64.2	8.2
62.5	20.19049	1.1405858	0.3148314	0.0152130	64.1	6.7
75	26.18635	2.1851169	0.4075129	0.0036982	64.3	4.8

Table S10. Raw Data to Accompany Figure 3(a). $^{241}Am^{3+}$ Stripping in 1-Octanol

Initial Extraction

$[HNO_3]$ (M)	Sample ID	CPM	Error (%)	Net CPM	Recovery (%)	D_{Am}
3.0	org. - 1	3137	0.56	3119	92.8	5.0715
	aq. - 2	633	1.26	615		
3.0	org. - 3	3070	0.57	3052	91.8	4.7762
	aqg. - 4	657	1.23	639		
3.0	org. - 5	2977	0.58	2959	89.1	4.7344
	aqg. - 6	643	1.25	625		
A_o	Ao - 7	4040	0.50	4022		
Blank	Avg.	18	7.44			

Stripping

$[HNO_3]$ (M)	Sample ID	CPM	Error (%)	Net CPM	Recovery (%)	D_{Am}
0.1	org. - 1	2027	1.00	2009	96.7	1.9931
	aq. - 2	1026	1.00	1008		
0.01	org. - 3	1974	1.00	1956	98.6	1.8575
	aqg. - 4	1071	1.00	1053		
0.001	org. - 5	2048	1.00	2030	104.2	1.9297
	aqg. - 6	1070	1.00	1052		

A_o for stripping data is the net CPM of the matched org.- # sample ID in the initial extraction

Table S11. Raw Data to Accompany Figure 3(b). $^{241}Am^{3+}$ Stripping and Re-extraction in 1-Octanol

Initial Extraction

Trial	$[HNO_3]$ (M)	Sample ID	CPM	Error (%)	Net CPM	Recovery (%)	D_{Am}
1	**3.0**	org. - 1	2287	1.00	2249		4.74
		aq. - 2	512	1.40	474		
2	**3.0**	org. - 3	2461	1.00	2423		5.07
		aq. - 4	516	1.39	478		
	A_o	Ao - 5	3112	1.00	3074		
	Blank	Avg.	38	5.11			

Stripping

Stripping and Trial	$[HNO_3]$ (M)	Sample ID	CPM	Error (%)	Net CPM	Avg. CPM
S1/T1	**0.01**	aq. - 1	620	1.27	582	597
		aq. - 2	650	1.24	612	
S1/T2	**0.01**	aq. - 3	664	1.23	626	629.5
		aq. - 4	671	1.22	633	
S2/T1	**0.01**	aq. - 5	1389	1.00	1351	1356.5
		aq. - 6	1400	1.00	1362	
S2/T2	**0.01**	aq. - 5	1407	1.00	1369	1376
		aq. - 6	1421	1.00	1383	
S3/T1	**0.01**	aq. - 5	444	1.50	406	403.5
		aq. - 6	439	1.51	401	
S3/T2	**0.01**	aq. - 5	416	1.55	378	375.5
		aq. - 6	411	1.56	373	

Re-Extraction

Trial	$[HNO_3]$ (M)	Sample ID	CPM	Error (%)	Net CPM	Recovery (%)	D_{Am}	Adjusted D_{Am}
1	**3.0**	org. - 1	2780	1.00	2743	105.83	5.25	5.08
		aq. - 2	559	1.34	522			
2	**3.0**	org. - 3	2584	1.00	2547	98.67	5.12	5.02
		aq. - 4	534	1.37	497			
	A_o	Ao - 5	3122	1.00	3085			
		Avg.	37	5.18				

***D_{Am} was adjusted to factor in remaining counts in the organic phase before the second extraction**

[i] (a) Fulmer, G. R.; Miller, A. J. M.; Sherden, N. H.; Gottlieb, H. E.; Nudelman, A.; Stoltz, B. M.; Bercaw, J. E.; Goldberg, K. I. NMR Chemical Shifts of Trace Impurities: Common Laboratory Solvents, Organics, and Gases in Deuterated Solvents

Relevant to the Organometallic Chemist. *Organometallics* **2010**, *29*, 2176–2179. (b) Gottlieb, H. E.; Kotlyar, V.; Nudelman, A. NMR Chemical Shifts of Common Laboratory Solvents as Trace Impurities. *J. Org. Chem.* **1997**, *62,* 7512−7515.