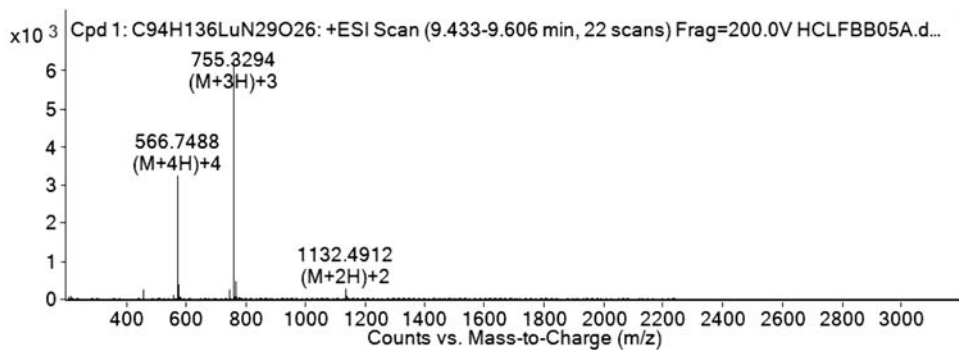
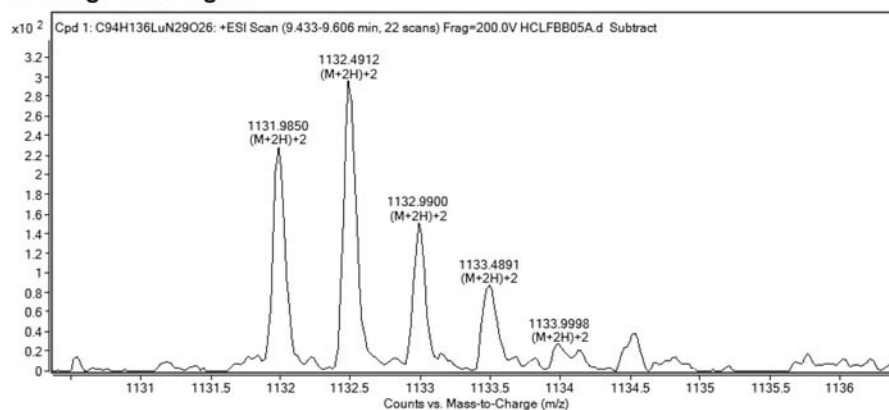


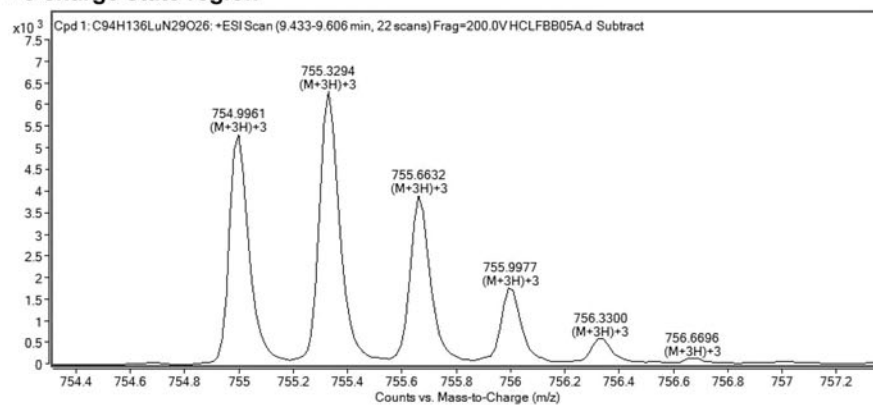
Supplementary Data



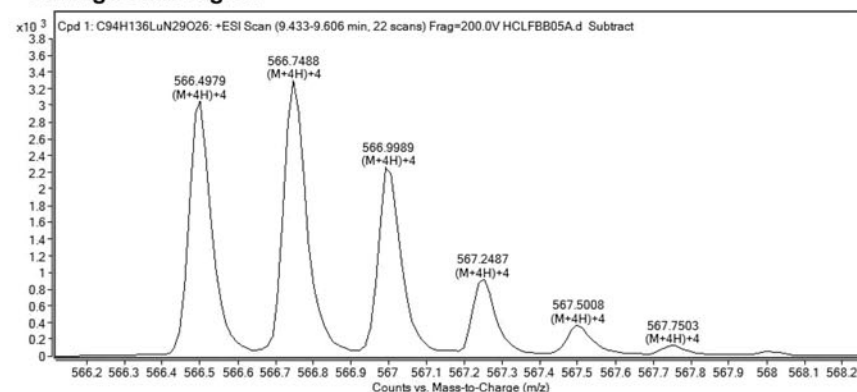
+2 charge state region



+3 charge state region



+4 charge state region



SUPPLEMENTARY FIG. S1. Electrospray mass spectral data of ^{nat}Lu-DO3A-4B4 (molecular formula: C₉₄H₁₃₆LuN₂₉O₂₆; exact mass=2261.9619).

SUPPLEMENTARY TABLE S1. ELECTROSPRAY MASS SPECTROMETRY OBSERVED AND CALCULATED PEAK LIST FOR +4, +3, AND +2 CHARGE STATE REGIONS (DATA TABLE FROM AGILENT MASS HUNTER QUALITATIVE ANALYSIS COMPOUND REPORT)

<i>MS spectrum peak list</i>						
<i>m/z</i>	<i>Calc m/z</i>	<i>Diff (ppm)</i>	<i>z</i>	<i>Abund</i>	<i>Formula</i>	<i>Ion</i>
+ 4 charge region						
566.4979	566.4978	0.23	4	3088	C94 H140 Lu N29 O26	(M+4H)+4
566.7488	566.7485	0.55	4	3299	C94 H140 Lu N29 O26	(M+4H)+4
566.9989	566.9991	-0.39	4	2304	C94 H140 Lu N29 O26	(M+4H)+4
567.2487	567.2498	-1.92	4	930	C94 H140 Lu N29 O26	(M+4H)+4
567.5008	567.5004	0.65	4	372	C94 H140 Lu N29 O26	(M+4H)+4
567.7503	567.7511	-1.37	4	126	C94 H140 Lu N29 O26	(M+4H)+4
+ 3 charge region						
754.9961	754.9946	2.01		5343	C94 H139 Lu N29 O26	(M+3H)+3
755.3294	755.3288	0.75		6314	C94 H139 Lu N29 O26	(M+3H)+3
755.6632	755.6631	0.14		3914	C94 H139 Lu N29 O26	(M+3H)+3
755.9977	755.9973	0.6		1809	C94 H139 Lu N29 O26	(M+3H)+3
756.33	756.3315	-1.97		616	C94 H139 Lu N29 O26	(M+3H)+3
756.6696	756.6657	5.18		158	C94 H139 Lu N29 O26	(M+3H)+3
+ 2 charge region						
1131.985	1131.9882	-2.81	2	229	C94 H138 Lu N29 O26	(M+2H)+2
1132.4912	1132.4896	1.39	2	297	C94 H138 Lu N29 O26	(M+2H)+2
1132.99	1132.991	-0.86	2	153	C94 H138 Lu N29 O26	(M+2H)+2

SUPPLEMENTARY TABLE S2. DATA FOR OPTIMIZATION OF THE RADIOLABELING YIELD

<i>pH</i>	<i>% complex</i>	<i>Standard deviation</i>	<i>Time study (min)</i>	<i>% complex</i>	<i>Standard deviation</i>
4	56.99	3.24	15	25.73	4.23
5	70.32	2.98	30	50.83	3.69
6	95.38	2.39	45	63.61	3.64
7	89.42	2.56	60	94.98	2.34
8	83.58	4.23	90	99.42	1.81
<i>Temperature (°C)</i>	<i>% complex</i>	<i>Standard deviation</i>	<i>Ligand concentration (μM)</i>	<i>% complex</i>	<i>Standard deviation</i>
25	32.10	4.23	0.01	18.13	4.23
37	94.51	1.28	0.1	71.74	2.77
60	96.78	2.14	1	97.84	2.44
90	98.26	2.21	10	100.00	1.51

SUPPLEMENTARY TABLE S3. CELL BINDING ASSAY DATA

<i>¹⁷⁷Lu-DO3A-4B4 binding assay</i>									
<i>Cells (10⁶)</i>	<i>¹⁷⁷Lu-DO3A-4B4</i>		<i>Average</i>	<i>Preblocked</i>		<i>Average</i>	<i>¹⁷⁷Lu-DO3A-PA1</i>		<i>Average</i>
0	0	0	0	0	0	0	0	0	0
0.25	49.7	46	47.85	2.1	1.5	1.8	2.2	1.9	2.05
0.5	55.5	51.1	53.3	2.8	3.1	2.95	1.9	2.1	2
1	64.6	60	62.3	5.3	6.1	5.7	1.8	2.3	2.05
2	69.4	65.3	67.35	11	13	12	2.3	1.3	1.8
<i>¹⁵³Sm-DO3A-4B4 binding assay</i>									
<i>Cells (10⁶)</i>	<i>¹⁵³Sm-DO3A-4B4</i>		<i>Average</i>	<i>Preblocked</i>		<i>Average</i>	<i>¹⁵³Sm-DO3A-PA1</i>		<i>Average</i>
0	0	0	0	0	0	0	0	0	0
0.25	15.9	19.1	17.5	2	1.8	1.9	0.9	0.9	0.9
0.5	23.1	22.8	22.95	2.6	2.1	2.35	1.1	0.6	0.85
1	38.1	40.3	39.2	3.3	3.2	3.25	1	1	1
2	46.5	51	48.75	6.5	6	6.25	1.8	1.6	1.7
<i>¹⁶⁶Ho-DO3A-4B4 binding assay</i>									
<i>Cells (10⁶)</i>	<i>¹⁶⁶Ho-DO3A-4B4</i>		<i>Average</i>	<i>Preblocked</i>		<i>Average</i>	<i>¹⁶⁶Ho-DO3A-PA1</i>		<i>Average</i>
0	0	0	0	0	0	0	0	0	0
0.25	15	16.2	15.6	2.8	2.7	2.75	3.6	4.8	4.2
0.5	24.5	22.6	23.55	3.4	5.2	4.3	4.3	4.3	4.3
1	27	28.5	27.75	5.8	4.2	5	7.1	8.2	7.65
2	44.6	39.8	42.2	7.9	6.3	7.1	6.6	7.7	7.15

Data show % radioactivity bound to cells.