SUPPLEMENTARY INFORMATION

Table S1. Statistics of the refined structures for the seven gadolinium derivatives obtained by cocrystallization. * Data from CNS program

Refinement	Gd-HPDO3A	Gd-DOTMA	Gd-DTPA	Gd-DO3A	Gd-DTPA-BMA	Gd-DOTA-BOM	Gd-DOTA
Resolution range (Å)	46.13-2.23	45.64-2.23	45.175-2.23	45.92-2.29	45.14-2.98	45.81-2.23	45.91-2.30
R (%)	20.65	20.95	21.57	21.78	23.55	22.14	21.39
R_{free} *(%)*	22.44	22.96	23.42	23.56	25.91	25.28	24.33
Site occupancies*	0.87, 0.66, 0.77, 0.60	0.65	0.99	0.83, 0.66, 0.63	0.34, 0.28	0.95	0.80
B-factors of Gd-sites ($Å^2$)*	33, 36, 31, 46	44	37	41, 41, 50	62, 62	38	53
Model							
Protein molecules by asymmetric unit	1	1	1	1	1	1	1
Amino acids	298	298	298	298	298	298	298
Choline molecules	3	6	6	4	5	6	6
Water molecules	296	297	289	293	215	295	269
No. of Gd complex molecules	4	1	1	3	0	0	1
Water molecules	296	297	289	293	215	295	269
RMS deviations from ideal:							
Bond lengths (Å)	0.014	0.013	0.013	0.015	0.023	0.014	0.013
Bond angles (deg.)	1.344	1.339	1.361	1.348	2.141	1.403	1.297
Observations	Loops C147-C150 and C169-C177 are not well defined						

Table S2. Statistics of the refined structures for the gadolinium derivatives obtained by soaking. * Data from CNS program

Refinement	Gd-HPDO3A	Gd-DOTMA	Gd-DTPA
Resolution range (Å)	45.36-2.70	45.14-2.70	46.94-2.70
R (%)	22.95	23.15	23.52
R_{free} *(%)*	25.04	25.36	26.21
Site occupancies*	0.56, 0.41, 0.37, 0.34	0.51, 0.39	0.52, 0.26, 0.21
B-factors of Gd-sites (Å^2)*	41, 44, 40, 42	45, 49	42, 48, 51
Model			
Protein molecules by asymmetric unit	1	1	1
Amino acids	298	298	298
Choline molecules	3	5	4
Water molecules	296	297	289
No. of Gd complex molecules	4	2	3
RMS deviations from ideal:			
Bond lengths (Å)	0.014	0.013	0.013
Bond angles (deg.)	1.344	1.339	1.361
Observations	Loops C147-C150 and C169-C177 are not well defined		

Table S3: Statistics of the refined structure for the Gd-HPDO3A derivative obtained with rotating anode.

Refinement	Gd-HPDO3A
Resolution range (Å)	45.36-2.39
R (%)	22.28
R_{free} *(%)*	25.16
Site occupancies*	0.71, 0.63, 0.44, 0.37
B-factors of Gd-sites (Å^2)*	39, 42,37, 49
Model	
Protein molecules by asymmetric unit	1
Amino acids	298
Choline molecules	3
Water molecules	296
No. of Gd complex molecules	4
RMS deviations from ideal:	
Bond lengths (Å)	0.019
Bond angles (deg.)	1.511
Observations	Loops C147-C150 and C169-C177 are not well defined