Electronic Supporting Information

**Title:** Hydrophobic Core Formation and Secondary Structure Elements in Uranyl(VI)–Binding Peptides

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**Fig. S1** FTIR-ATR spectra of c[DSDSSDS] in the presence of UO$_2^{2+}$ with different amount of NaOH.

**Fig. S2** FTIR-ATR spectra of c[DSDSSDS] in the absence of UO$_2^{2+}$ with different amount of NaOH.
Fig. S3 Structures of $\text{UO}_2^{2+}$–bound CaM–M2c (a,b,c) and CaM–M3c (d,e,f) from the MD trajectories. Waters and Na$^+$ ions are omitted for clarity. (a,d): Global view on the peptide along the O=U=O axis from the MD snapshot at $t = 200$ ns. Peptide in dark ribbon, uranium in pink ball, oxygens in the vicinity of uranium in red balls. (b,e): Superpositions of 100 snapshots from 200 ns MD trajectory (each 2 ns). Peptide in gray ribbon and $\text{UO}_2^{2+}$ in pink and red. (c,f): Global view on the peptide at $t = 200$ ns. Peptide in dark ribbon, hydrophobic sidechains in red, polar sidechains in blue spheres.

Fig. S4 Analysis of secondary structure elements of the $\text{UO}_2^{2+}$–bound CaM–M1c (top, left), CaM–M2c (top, right), CaM–M3c (bottom, left), and CaM-U2 (bottom, right) from the MD trajectories based on DSSP (hydrogen bond analysis algorithm).
Fig. S5 Time evolution of RMSD of backbone atoms from 200 ns trajectories of MD simulations of CaM-M1c, CaM-M2c, CaM-M3c, and CaM-U2 with UO$_2^{2+}$.