

Supporting Information for

Metal-Tunable Self-Assembly of Hierarchical Structure in Mussel-Inspired Peptide Films

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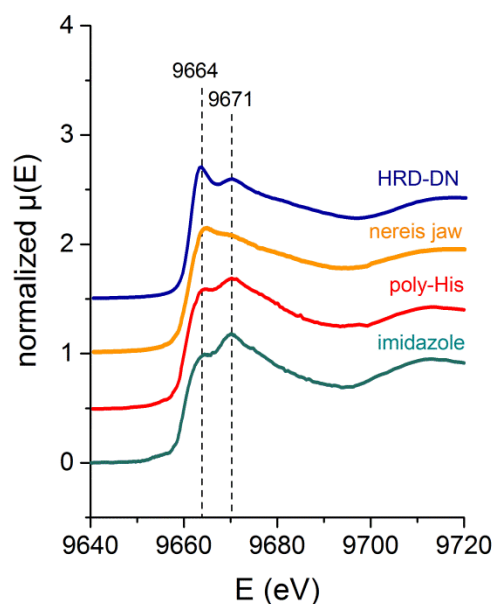


Figure S1. XANES spectra of HRD-based peptide film and Zn references. Normalized Zn K-edge XANES spectrum of HRD-based film (blue) containing Zn in a ratio 3:1 and references nereis jaw (orange), polyhistidine:Zn mixture (2:1 His:Zn) (red) and imidazole (green). The vertical dashed lines are intended for easy comparison of the double peak feature among various curves.

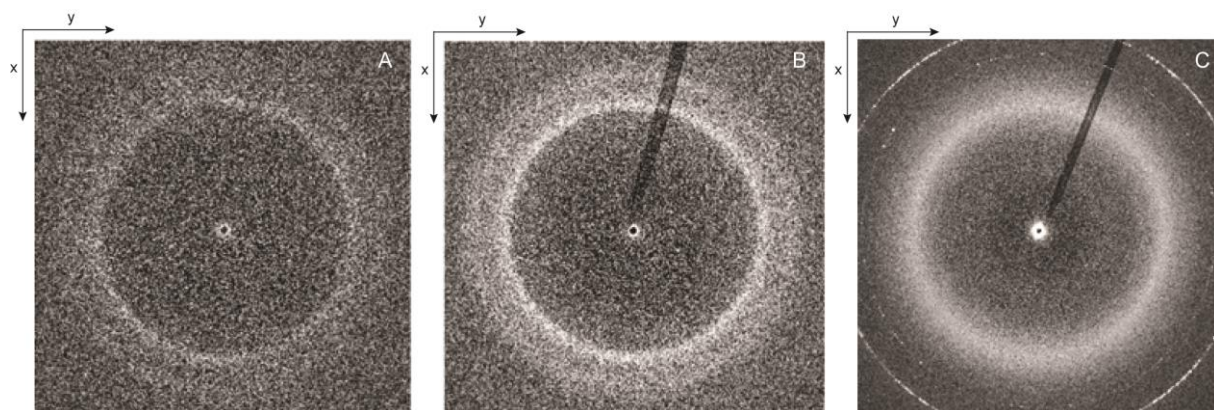


Figure S2. XRD patterns of HRD-based peptide films in the absence (A) and presence of metals (B, C). Films were exposed to the incident beam perpendicular to the film plane. XRD patterns of all three peptide films show diffuse rings with nearly equal intensity at all azimuthal angles (A, B, C). However, in the presence of Zn^{2+} (B) and Cu^{2+} (C), the ring at $d = 0.46$ nm indicating β -sheet structure appears somewhat stronger than in the absence of metal ions.

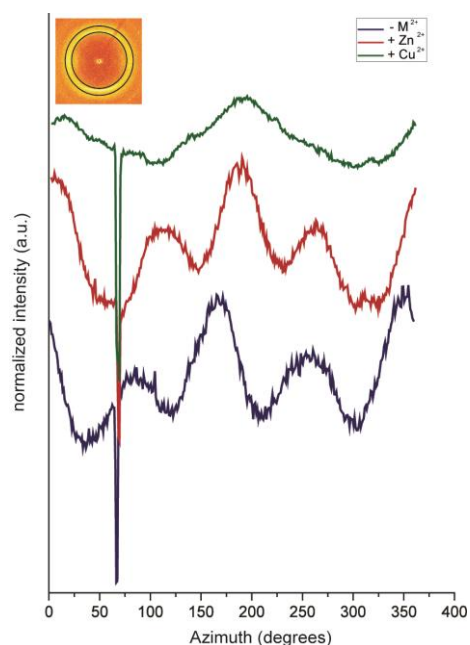


Figure S3. Azimuthal integration of HRD-based peptide film WAXD pattern exhibits four distinct peaks suggesting a distinct alignment of structures in films containing no metal ions or Zn. Films formed in the presence of Cu only show two broad peaks indicating little alignment of structures.

Table S4. Applied methods for the characterization of peptide structure and hierarchical organization within HRD-based peptide films in the absence and presence of metal ions.

	Film plane oriented perpendicular to incident beam			Film plane oriented parallel to incident beam	
	FTIR Spec.	pol. Raman Spec.	XRD	pol. Raman Spec.	XRD
HRD-DN – M^{2+}	crystalline β -sheet structure	β -sheet structure, no orientation	β -sheet structure, no orientation	β -sheet structures oriented with backbones parallel to film plane	β -sheet crystallites stacked in direction of film's y-axis
HRD-DN + Zn^{2+}	crystalline β -sheet structure, less prominent compared to $-M^{2+}$	β -sheet structure, no orientation	β -sheet structure, no orientation	β -sheet structures oriented with backbones parallel to film plane	β -sheet crystallites stacked in direction of film's y-axis, highly ordered
HRD-DN + Cu^{2+}	crystalline β -sheet structure, less prominent compared to $-M^{2+}$	β -sheet structure, slightly oriented	β -sheet structure, no orientation	β -sheet structures, less prominent polarization dependency	less aligned β -sheet crystallites stacked in direction of film's y-axis