

Supporting Information for:

***In situ* AFM Study of Amelogenin Assembly and Disassembly Dynamics on Charged Surfaces Provides Insights on Matrix Protein Self-Assembly**

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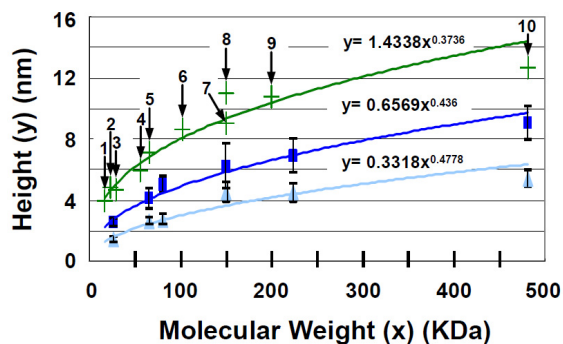
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Table S1 and Fig. S1 to S4

Calibration curves to show the correlation between protein molecular weight and height from *ex-situ* AFM (Light blue curve), *in situ* AFM (Blue curve) and hydrodynamic diameters (green curve) (modified from reference <sup>1</sup>)



Molecular Weight	<i>Ex situ</i> AFM Height	<i>In situ</i> AFM Height	Hydrodynamic Diameter
20 kDa	1.4 nm	2.4 nm	4.4 nm
40 kDa	1.9 nm	3.3 nm	5.7 nm
60 kDa	2.3 nm	3.9 nm	6.6 nm
80 kDa	2.7 nm	4.4 nm	7.4 nm
100 kDa	3.0 nm	4.9 nm	8.0 nm
120 kDa	3.3 nm	5.3 nm	8.6 nm
140 kDa	3.5 nm	5.7 nm	9.1 nm
160 kDa	3.7 nm	6.0 nm	9.5 nm
180 kDa	4.0 nm	6.3 nm	10.0 nm
200 kDa	4.2 nm	6.6 nm	10.4 nm
240 kDa	4.6 nm	7.2 nm	11.1 nm
420 kDa	5.9 nm	9.1 nm	13.7 nm
440 kDa	6.1 nm	9.3 nm	13.9 nm
800 kDa	8.1 nm	12.1 nm	17.4 nm
2340 kDa	13.5 nm	19.3 nm	26.0 nm

Table S1. Amelogenin rP172 parameters generated from the previously established protein calibration curves.<sup>1</sup>

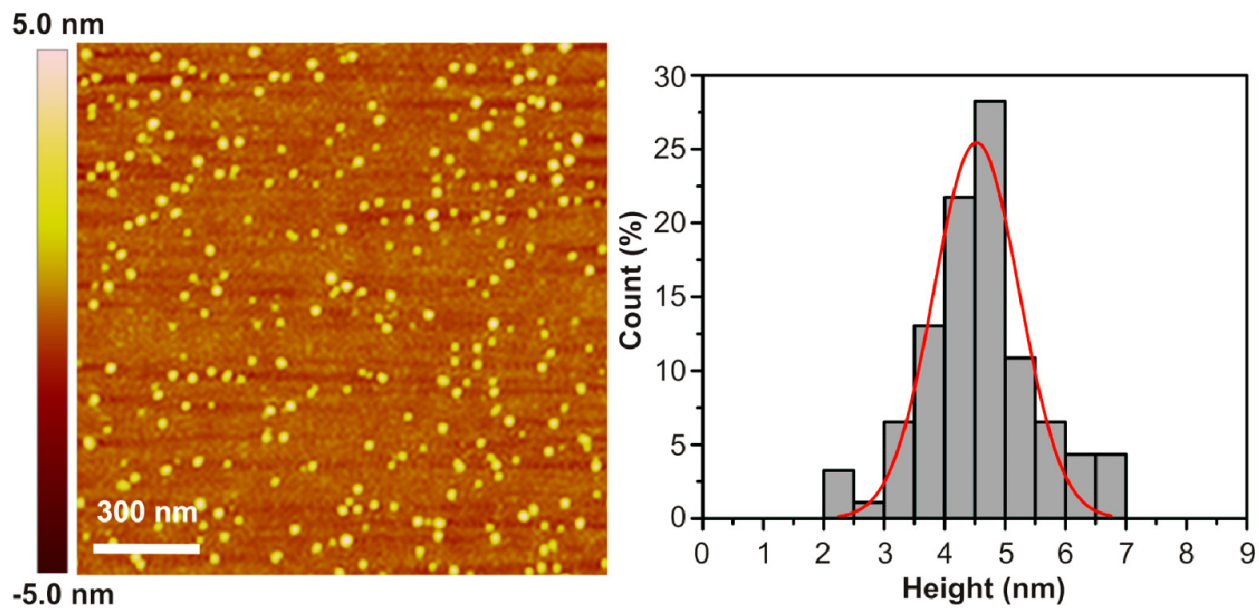


Figure S1. (a) Tapping mode *ex situ* AFM height image of uniform amelogenin particles on mica surface at pH 8.0, height =  $4.5 \pm 0.9$  nm, based on 92 counts.

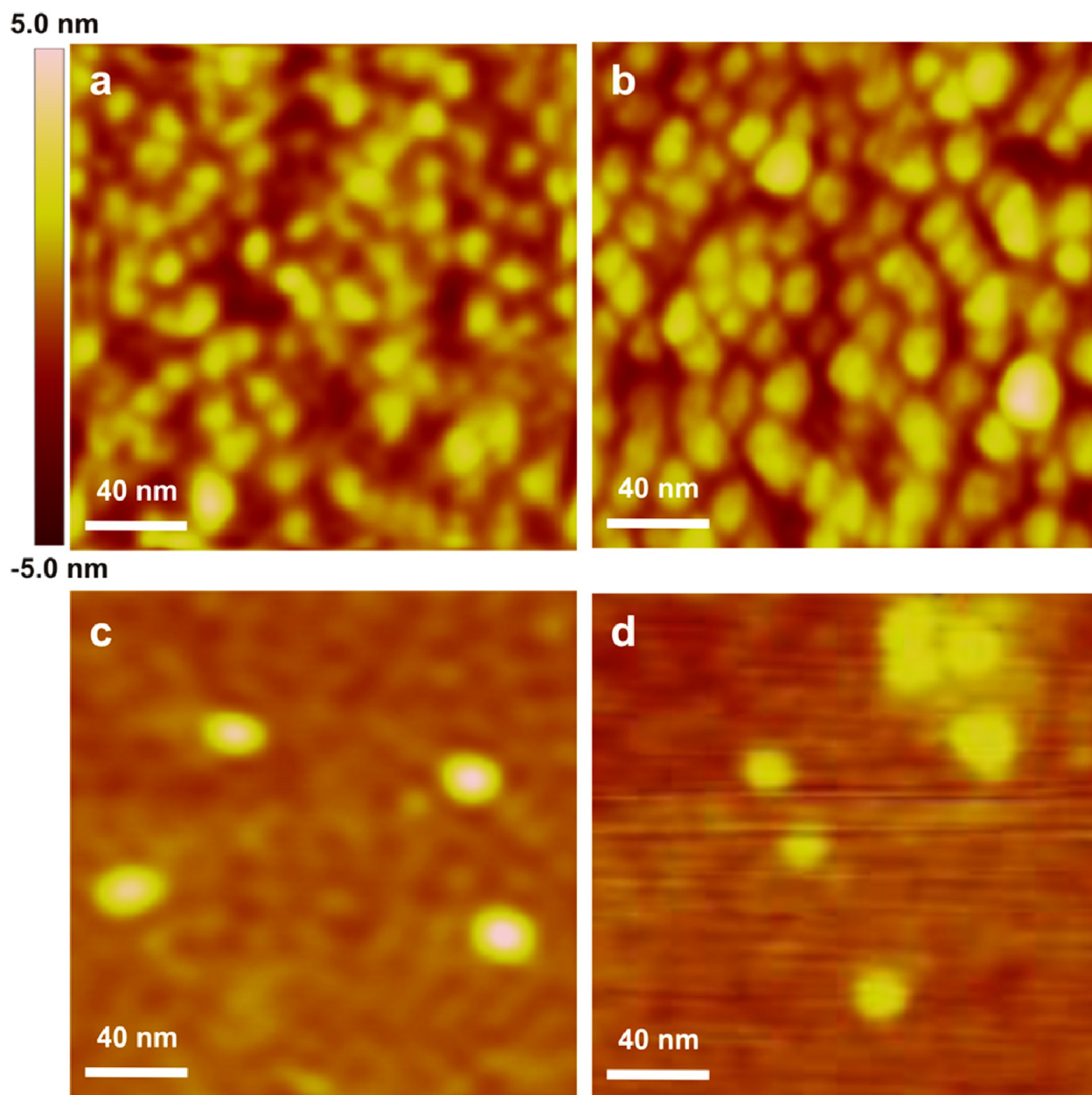
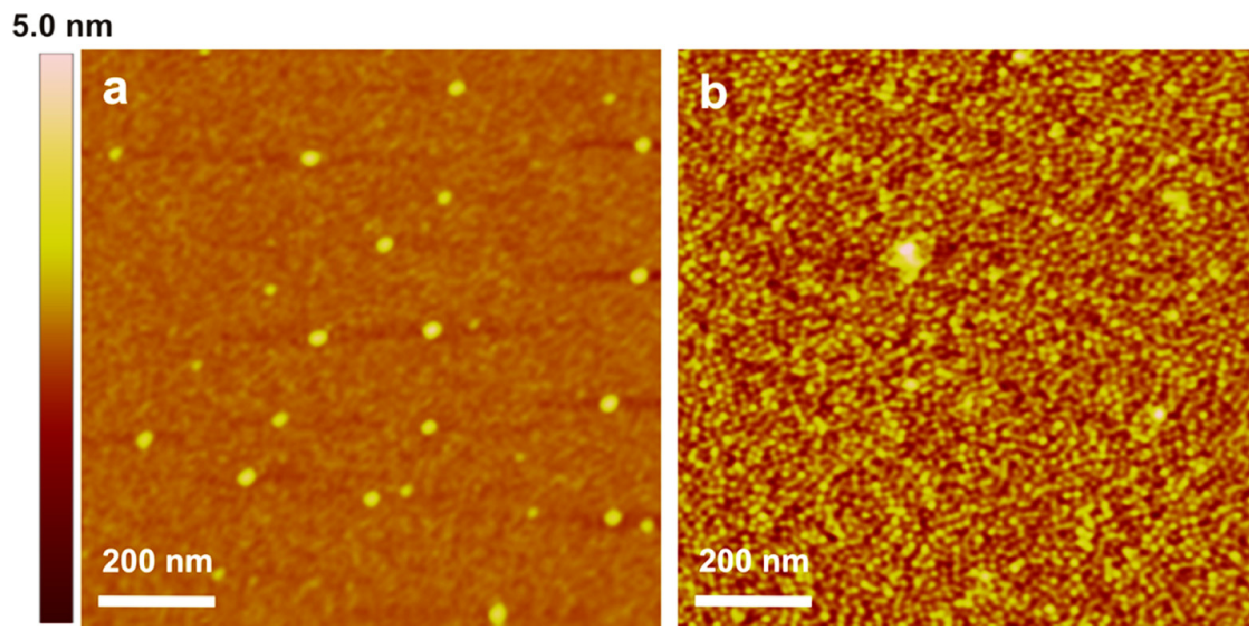
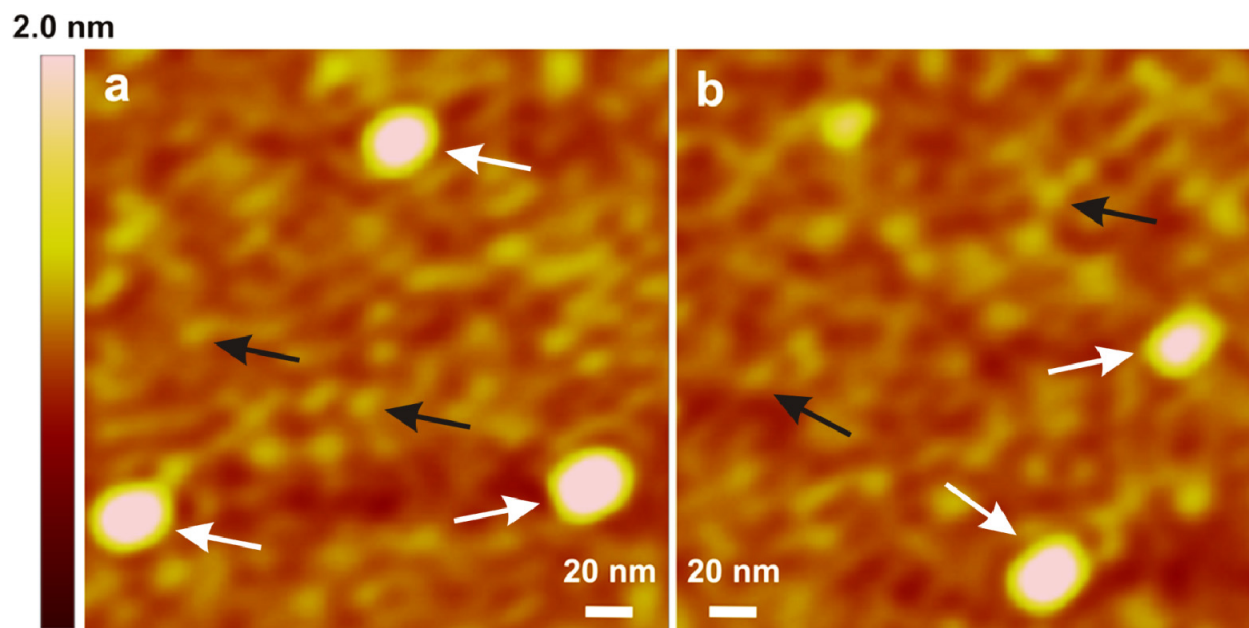


Figure S2. *Ex situ* AFM height images of amelogenin particles at pH8.0 under different conditions to show the tip convolution effects. (a) ScanAsyst mode, sharpened tip, diameter =  $\sim 9.0$  nm. (b) ScanAsyst mode, contaminated tip, diameter =  $\sim 15.0$  nm. (c) Tapping mode, contaminated tip, diameter =  $\sim 23$  nm. (d) Tapping mode, modified from reference<sup>2</sup>, diameter =  $\sim 20$  nm.



**-5.0 nm**  
 Figure S3. *Ex situ* AFM height images of amelogenin oligomeric particles at pH 8.0. (a) On mica surface, oligomer density =  $25 / \mu\text{m}^2$ . (b) On APS mica surface, oligomer density =  $2029 / \mu\text{m}^2$ .



**-2.0 nm**  
 Figure S4, *Ex situ* AFM height images of amelogenin particles at pH 8.0 on mica surface to show the co-existence of large amount of amelogenin monomers on mica surface along with amelogenin oligomers. White and black arrows indicate amelogenin oligomers and monomers respectively.

## References

- (1) Cho, K. R.; Huang, Y.; Yu, S. L.; Yin, S. M.; Plomp, M.; Qiu, S. R.; Lakshminarayanan, R.; Moradian-Oldak, J.; Sy, M. S.; De Yoreo, J. J. *J. Am. Chem. Soc.* **2011**, *133*, 8586.
- (2) Du, C.; Falini, G.; Fermani, S.; Abbott, C.; Moradian-Oldak, J. *Science* **2005**, *307*, 1450.