

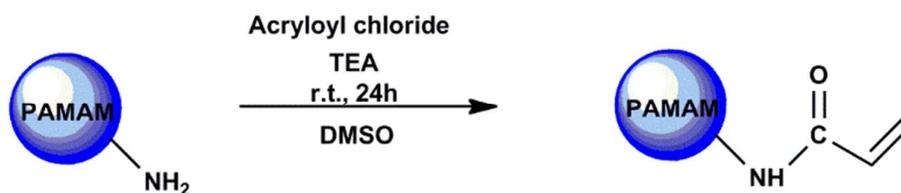
## Bioinspired from Salivary Acquired Pellicle: A Multifunctional Coating for Biominerals

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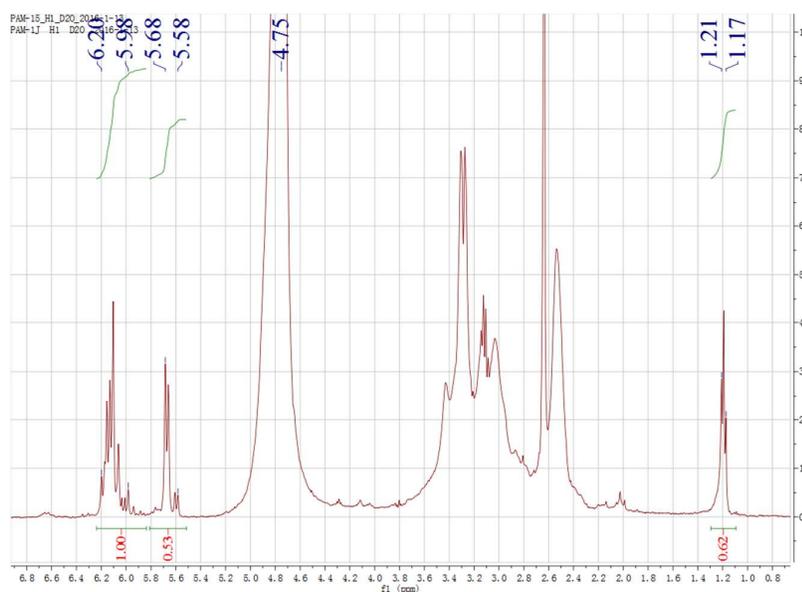
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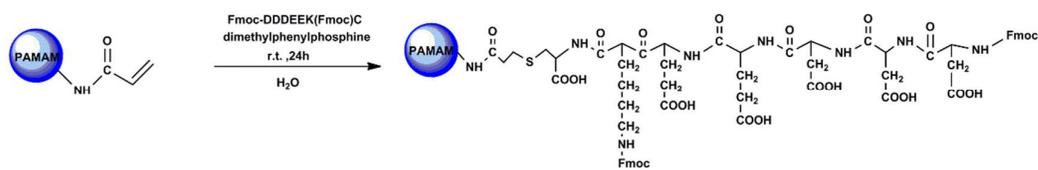
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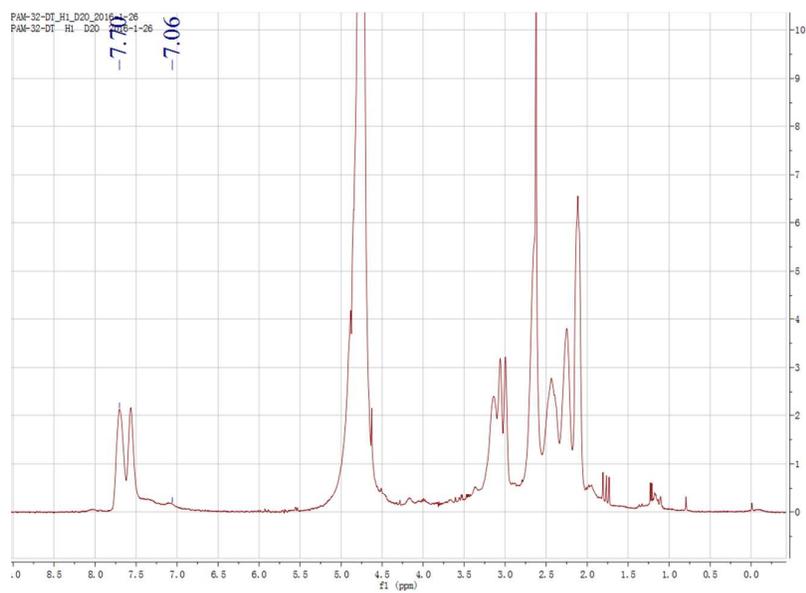
**Figure S1.** Synthetic route of acryloyl chloride grafted amine-terminated PAMAM (acryloyl-PAMAM).



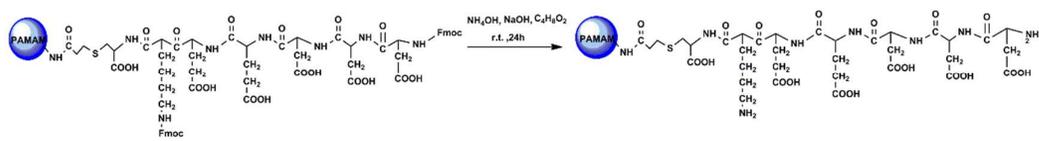
**Figure S2.** <sup>1</sup>H NMR spectrum of acryloyl-PAMAM: 400 Hz; Deuterium oxide;  $\delta$ =5.98–6.20 (m, CHCH<sub>2</sub>); 5.58–5.68 (m, CHCH<sub>2</sub>); 1.17–1.21(m, CH<sub>2</sub>NH<sub>2</sub>).



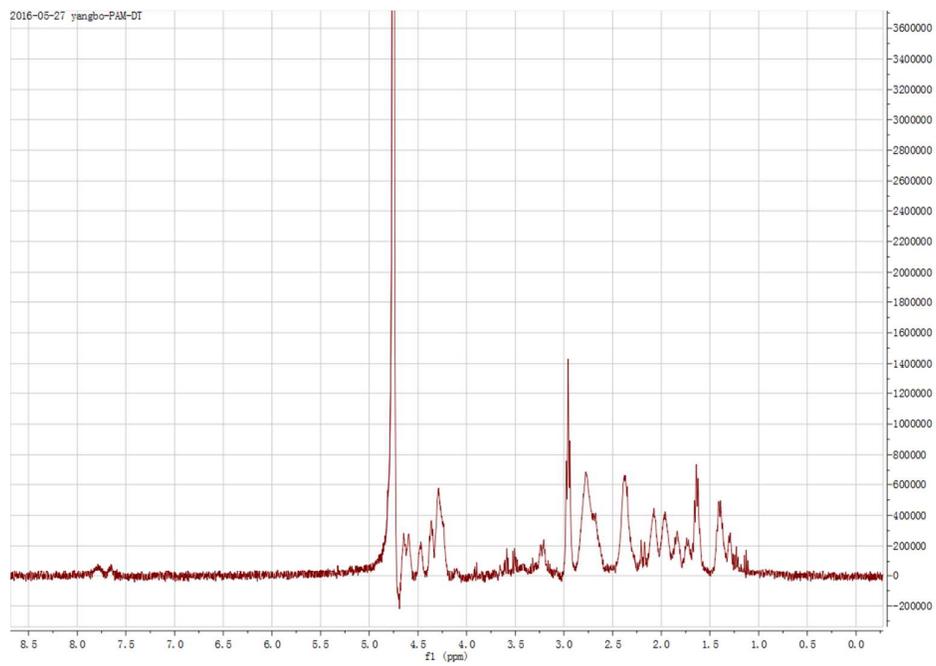
**Figure S3.** Synthetic route of Fmoc protected DDDEEK-PAMAM.



**Figure S4.**  $^1\text{H}$  NMR spectrum of Fmoc protected DDDEEK-PAMAM: 400 HZ; Deuterium oxide;  $\delta=7.06\text{--}7.70$  (m, CHarom in Fmoc).



**Figure S5.** Synthetic route of DDDEKC-PAMAM.



**Figure S6.**  $^1\text{H}$  NMR spectrum of DDDEKC-PAMAM.

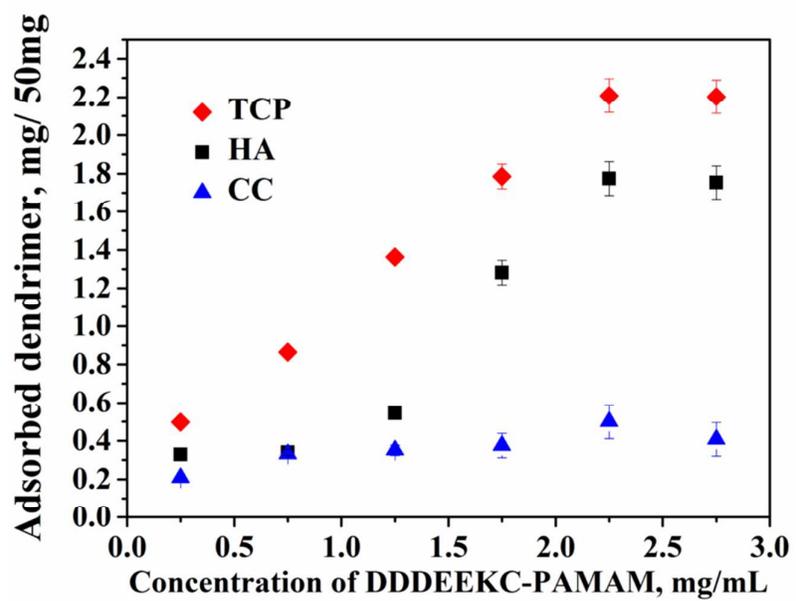
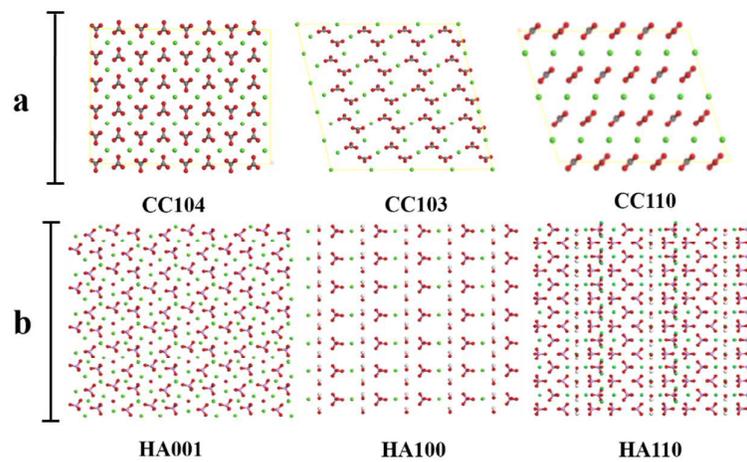
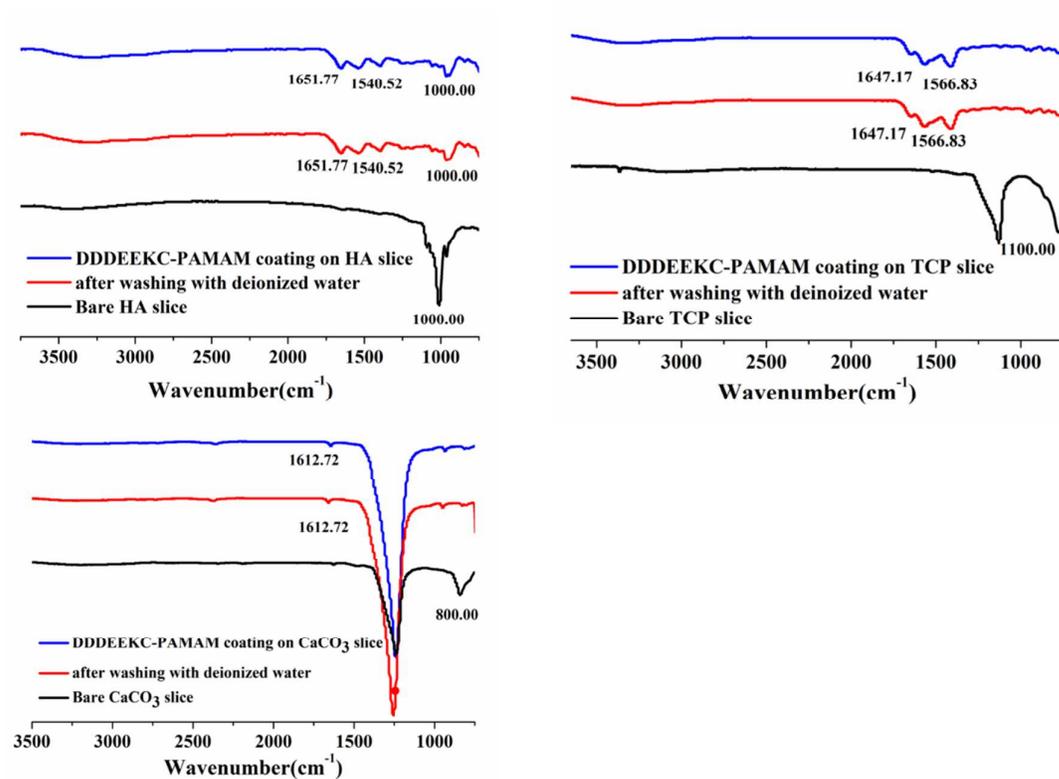


Figure S7. Adsorption isotherm of DDDEEKC-PAMAM on 50 mg powder of HA, TCP and CC, respectively.

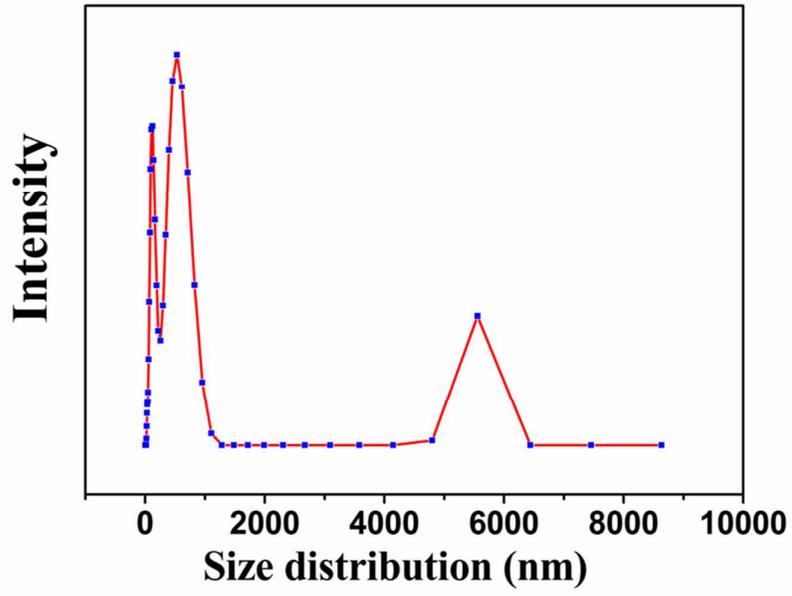


**Figure S8.** Schematic representation of CC planes (104), (103) and (110). HA planes (001), (100) and (110). Color codes: hydrogen atom, white; calcium atom, green; oxygen atom, red; phosphorous atom, purple. The density of Ca ion= the number of Ca ion / $a^2$  or  $b^2$ . The schematic representation of CC and HA planes are capture from software materials studio.

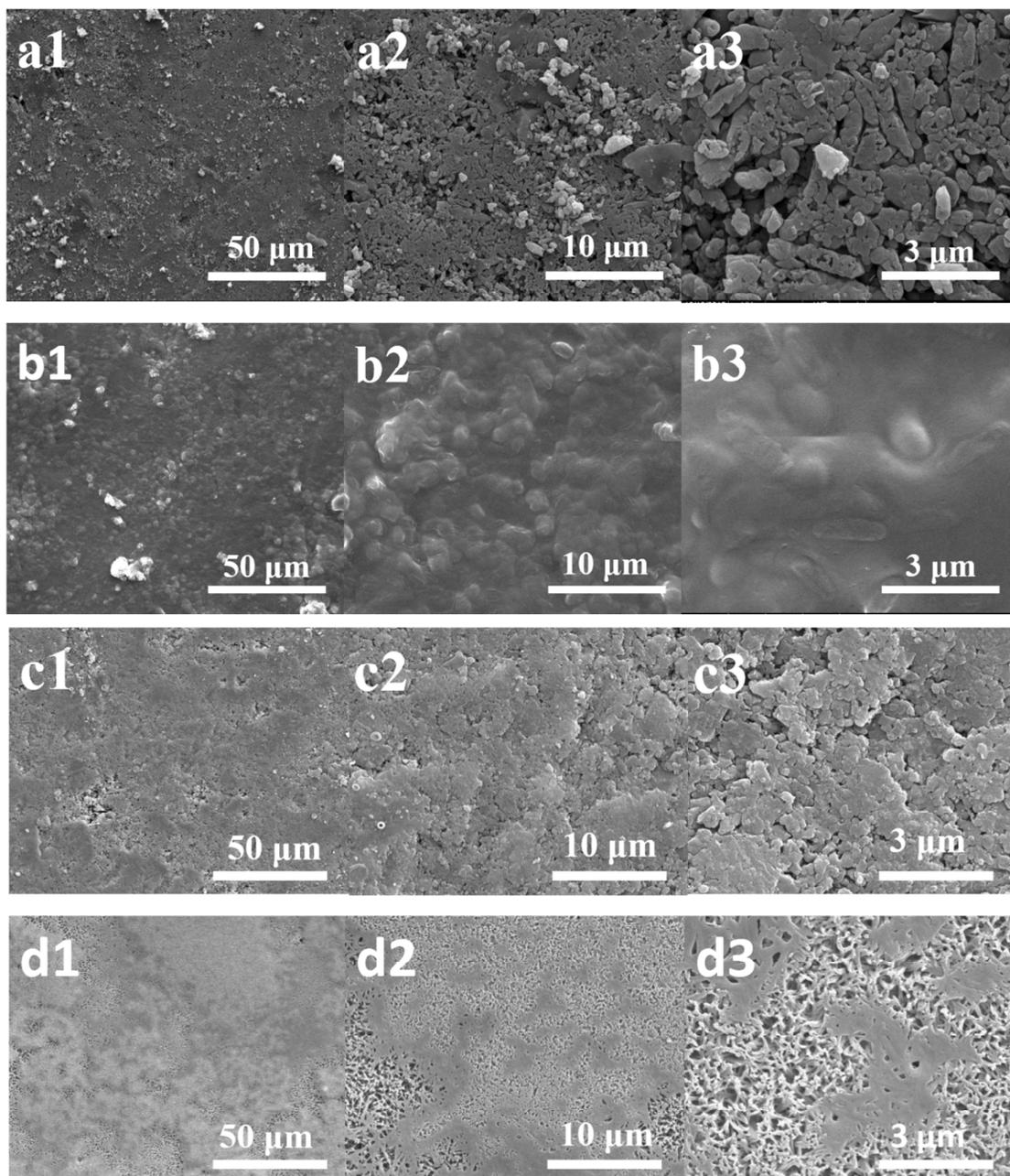
The density of Ca ion	CC (104)	CC (103)	CC (110)
	0.044	0.044	0.023
	HA (001)	HA (100)	HA (110)
	0.059	0.046	0.035



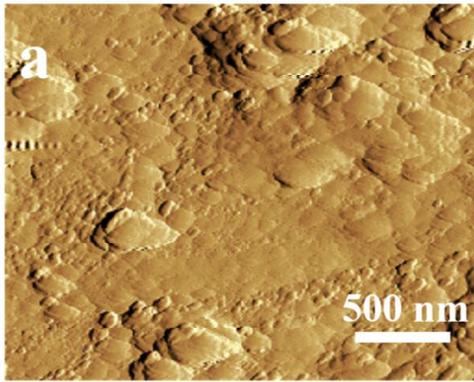
**Figure S9.** The ATR-IR spectra of bare, DDDEEK-PAMAM coated, and washed slices of HA, TCP and CC, respectively.



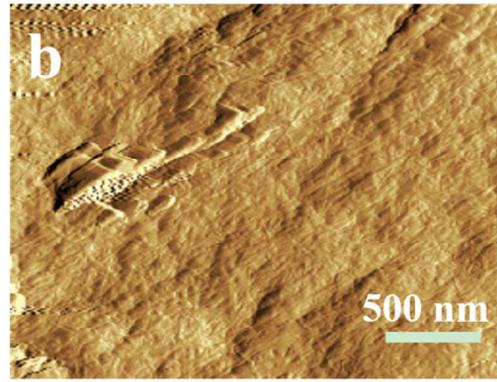
**Figure S10.** The size distribution of DDDEKC-PAMAM by DLS, indicating its self aggregation due to noncovalent intermolecular interaction among each other.



**Figure S11.** The SEM spectra of bare (a group) and DDDEKC-PAMAM coated (b group) CC, and bare (c group) and DDDEKC-PAMAM coated (d group) TCP.

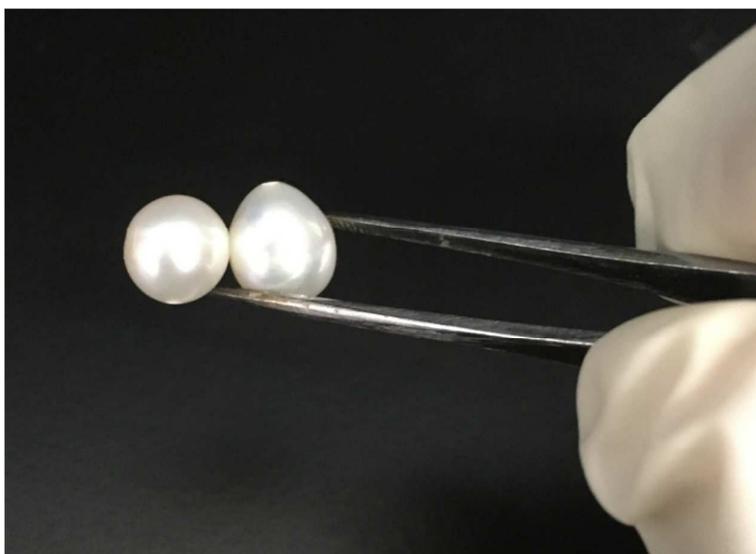


**(Ra: 277.557 Rz:200.316)**

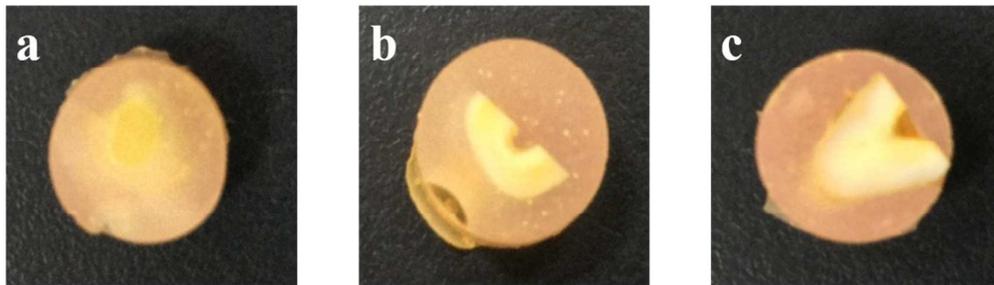


**(Ra: 62.98 Rz:2.185)**

**Figure S12.** Atomic force microscopy (AFM) images of (a) bare HA slice and (b) DDDEKC-PAMAM coated HA slice. The latter is much smoother in terms of Ra and Rz.



**Figure S13.** By using 200  $\mu\text{L}$  of DDDEKC-PAMAM solution (2.25 mg/mL), pearls (8 mm in diameter) were stucked. It indicates that the DDDEKC-PAMAM coating has a strong binding force on the surface of biominerals.



**Figure S14.** Dimethyl yellow loaded DDDEKC-PAMAM was dropped on the surface of enamel (a), dentin (b) and bone (c). After rinsing with deionized water, the coating was retained.

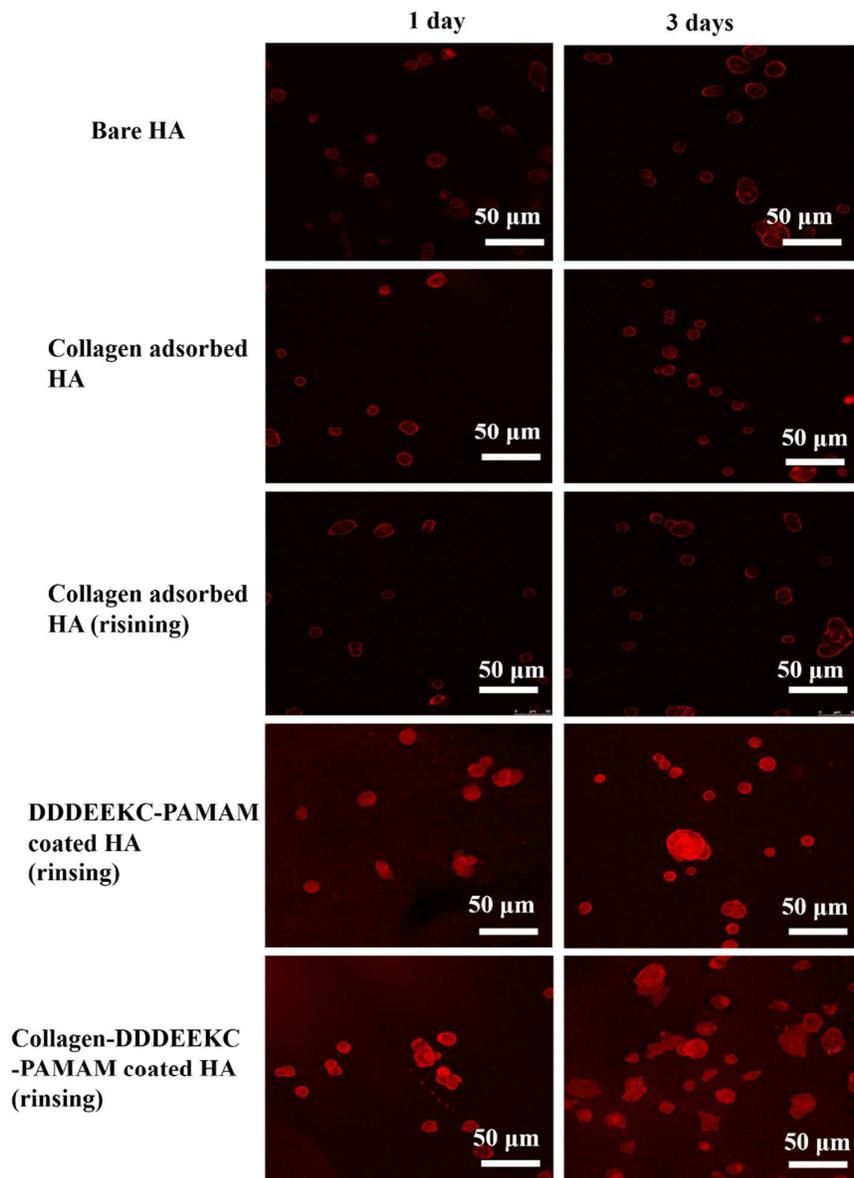
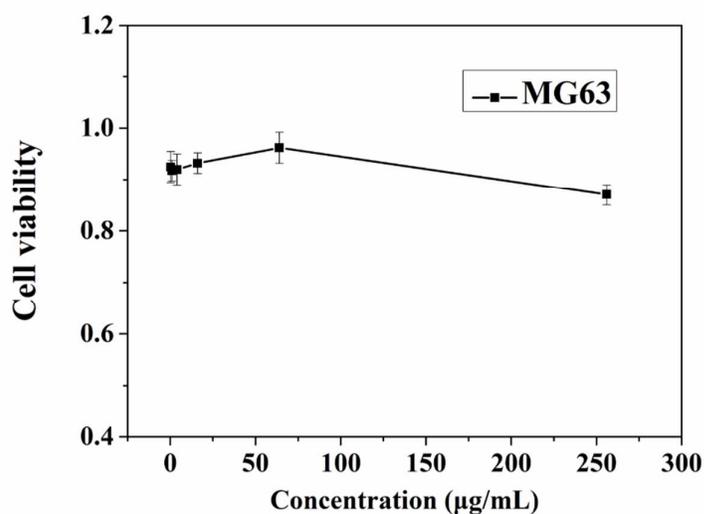


Figure S15. MG63 cell adhesion on bare and different HA surfaces after 1 day and 3 days culturing.



**Figure S16.** Cytotoxicity of DDDEEK-C-PAMAM at various concentrations using MG63 cells by MTT method.

**Table S1.** EDS elemental composition of the biomineral surfaces before and after coated with DDDEEK-C-PAMAM.

	C%	N%	O%	Ca%	P%
Bare HA	0%	0%	33.67%	45.45%	20.87%
Coated HA	28.3%	53.29%	8.48%	2.94%	6.99%
Bare TCP	0%	0%	39.63%	48.18	12.19%
Coated TCP	31.51%	52.12%	7.41%	3.41%	5.55%
Bare CC	27.87%	0%	39.57%	32.57%	0%
Coated CC	19.44%	31.78%	32.52%	16.26%	0%