Supplementary Information

Preparation of hierarchical enamel like structures from nano- to macro-scale regulated by inorganic templates derived from enamel

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Figure S1 SEM images of enamel like HA layer. The apatite contains two structural components like natural enamel: prismatic enamel (P) and interprismatic enamel (IP).
**Figure S2** SEM images of the grown apatite layer on enamel after hydrothermal reaction at pH=12 and 120 °C for different times. (a-c) 1h; (d-f) 3h; (g-i) 6h. The part between the dotted line in the cross-section images (c), (f) and (i) is the newly formed apatite layer. The grown apatite crystallites possess cross-arranged structure at the beginning of the reaction. They were needle like at first and gradually transformed to regular nanorods. The length of the apatite crystals increased with the prolonged reaction time. P: prismatic enamel; IP: interprismatic enamel.
Figure S3 SEM images of the grown apatite layer on enamel after hydrothermal reaction at pH=12 and different temperatures for 24 h. (a, b) 90 °C; (c, d) 60 °C. P: prismatic enamel; IP: interprismatic enamel.

Figure S4 SEM images of the grown apatite layer on enamel after hydrothermal reaction at 120 °C and pH=6 for 24 h. P: prismatic enamel; IP: interprismatic enamel.
Figure S5 The SEM images of HA ceramic substrates with the smallest grain size (a), the medium grain size (d) and the largest grain size (g), and the morphology of apatite layer grown on the HA ceramic substrates with the smallest grain size (b, c), the medium grain size (e, f), and the largest grain size (h, i), where (c), (f), (i) are higher magnification images of (b), (e), (h), respectively.
**Figure S6** Low magnification (a) and high magnification (b) images of the growth apatite layer formed on enamel substrate at higher ion concentration (2X original solution). It can be seen from (b) that the interprismatic enamel (IP) is ordered and the prismatic enamel (P) is disordered.

![Figure S6 Images](image)

**Figure S7** Examination results of the FHA-gelatin composite layer. (a) Before penetration of gelatin and (b) after penetration of gelatin. The inset of (b) shows the EDS result of the FHA-gelatin composite layer and the C content is 11.2 wt. %. The samples were directly broken to expose the fresh section instead of embedded with epoxy resin to avoid contamination.

![Figure S7 Images](image)