## Supporting Information

## Biomineralization-Inspired Sandwich Dentin Desensitization Strategy Based on Multifunctional Nanocomposite with Yolk-Shell Structure

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**Figure S1.** The low magnification of intermediate products in the synthesis process of PSTZ. (A) SiO<sub>2</sub>, (B) core-shell SiO<sub>2</sub>@TiZrO<sub>2</sub>, (C) yolk-shell SiO<sub>2</sub>@TiZrO<sub>2</sub>, (D) yolk-shell SiO<sub>2</sub>@mTiZrO<sub>2</sub> (STZ), (E) PAH-ACP@yolk-shell SiO<sub>2</sub>@mTiZrO<sub>2</sub> (PSTZ).



Figure S2. XRD pattern of STZ and PSTZ.



**Figure S3.** The low magnification of representative TEM images of stained unmineralized collagen fibrils (A) and PSTZ-mediated intrafibrillar mineralization after 1 d (B), 4 d (C), 7 d (D) incubation.



**Figure S4.** Representative FESEM images of PSTZ-mediated remineralization of DDM on 4 d (A), 14 d (B). Cross images were shown in  $A_1$ - $B_1$  and  $A_2$ - $B_2$ , longitudinal section images were shown in  $A_3$ - $B_3$  and  $A_4$ - $B_4$ . High-magnification images ( $A_2$ - $B_2$  and  $A_4$ - $B_4$ ) were magnified from the selected area (yellow square) in the low-power image ( $A_1$ - $B_1$  and  $A_3$ - $B_3$ ). Scale bar in ( $A_1$ - $B_1$  and  $A_3$ - $B_3$ ), 1 µm, in ( $A_2$ - $B_2$  and  $A_4$ - $B_4$ ), 100 nm.



**Figure S5.** Representative cross-section FESEM images of DDM remineralized with PSTZ-containing solution after 0 d (A<sub>1</sub>), 1 d (B<sub>1</sub>), 4 d (C<sub>1</sub>), 7 d (D<sub>1</sub>), 14 d (E<sub>1</sub>), 21 d (F<sub>1</sub>). Corresponding elemental distribution (A<sub>2</sub>-F<sub>2</sub>, A<sub>3</sub>-F<sub>3</sub>) and contents of Ca and P (A<sub>4</sub>-F<sub>4</sub>). Scale bar, 10  $\mu$ m.



**Figure S6.** Effect of STZ and PSTZ on the hemolysis of erythrocytes. Data are presented as mean  $\pm$  SD (N = 3). Inset: micro-images of haemolysis results.