Supporting Information

Bioinspired enamel-like oriented minerals on general surfaces: towards high mechanical property

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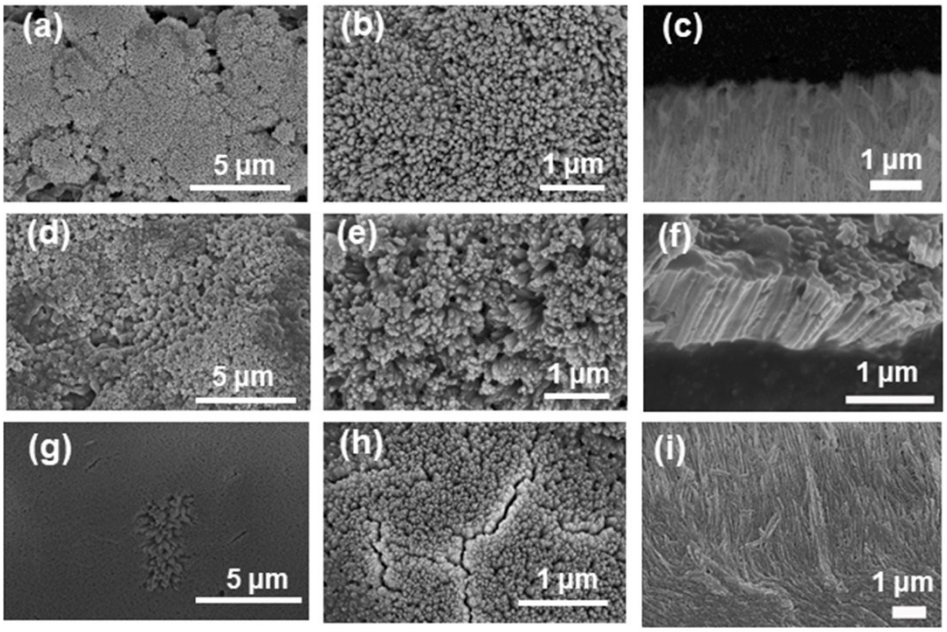

Fig. S1 SEM image of minerals formed on different substrates in AAO-assisted double-layer gel system (ADGS). (a), (b), (c) Minerals formed on cellulous fibre membrane, (d), (e), (f) glass sheet and (g), (h), (i) Silicon chips respectively. (b), (e), (h) are magnification of (a), (d), (g) and (c), (f), (i) are cross-section view of (a), (d), (g) correspondingly.

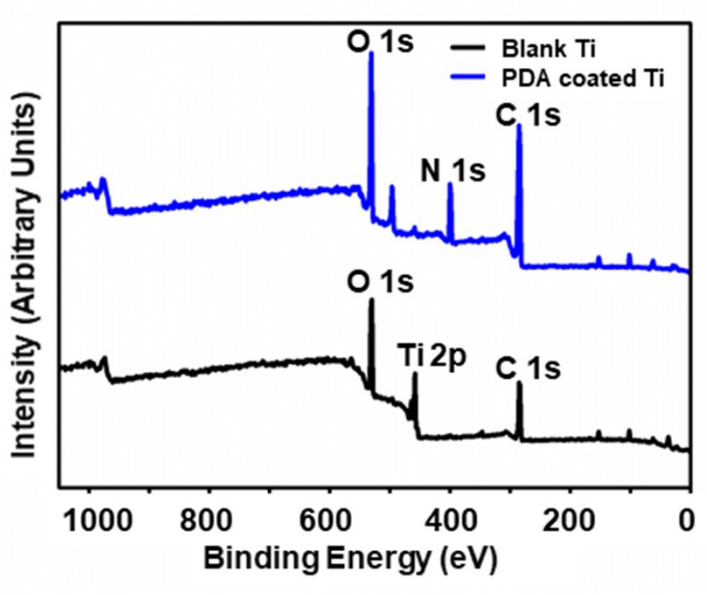

Fig. S2 XPS survey spectra of blank and PDA coated Ti sheet.

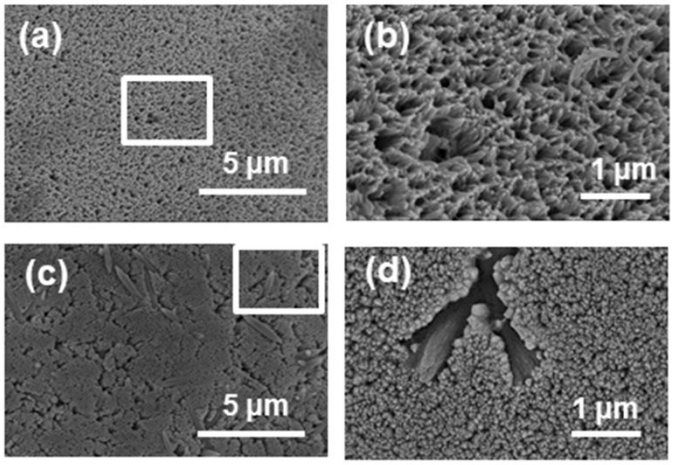

Fig. S3 SEM images of minerals formed on the sixth day of mineralization (Ti substrate) in DGS group (a)and ADGS group (c). (b) and (d) are magnification of (a) and (c), respectively.

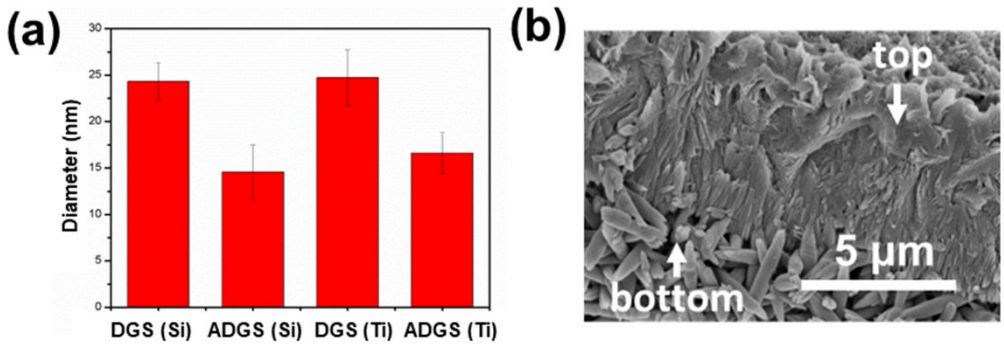

Fig. S4 (a) Statistic diameter of nanocrystals obtained from DGS and ADGS groups after 8 days of mineralization. (b) SEM image of the cross-section structure of minerals in ADGS group on eighth day of mineralization.

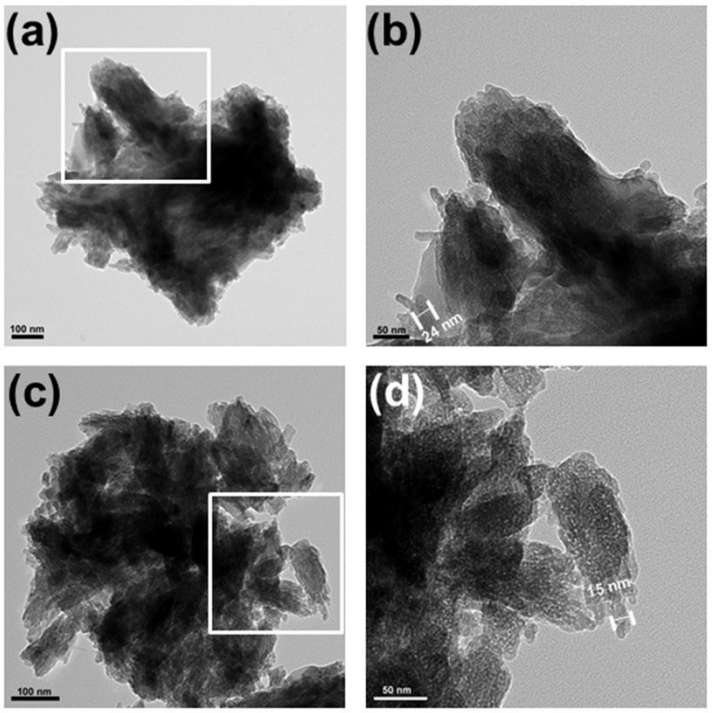

Fig. S5 TEM images of crystal bundles formed in (a) DGS group and (c) ADGS group after 8 days of mineralization (Ti substrate). (b) and (d) are magnification of (a) and (c), respectively.

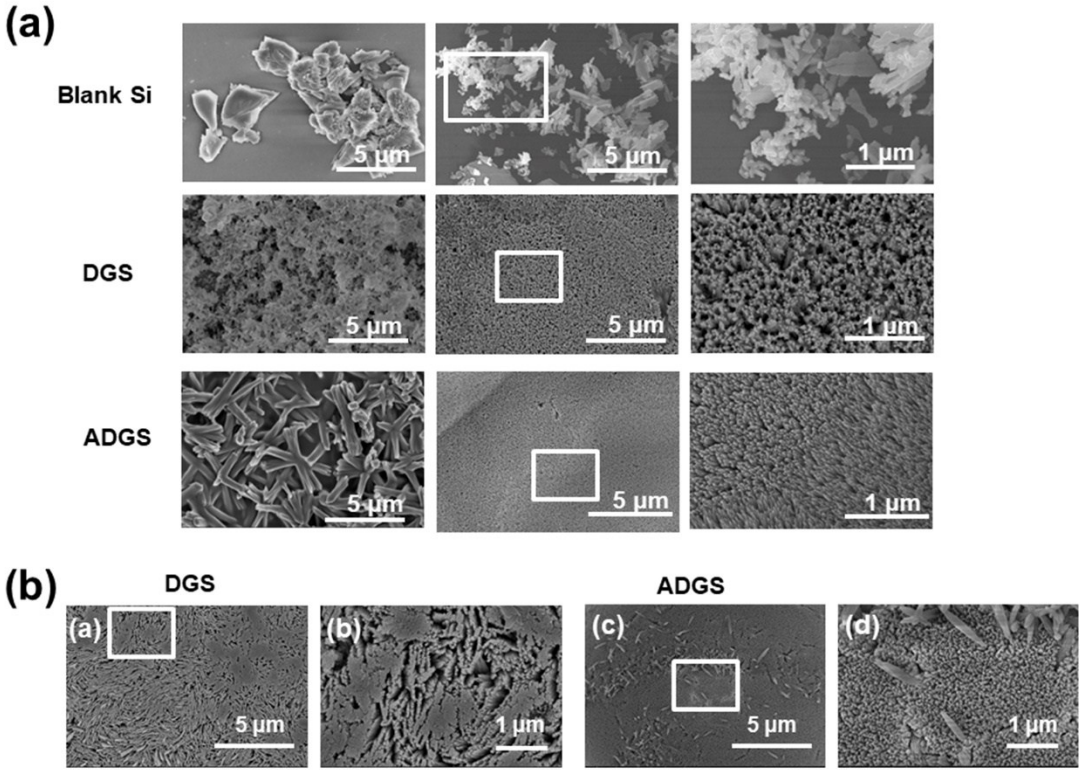

Fig. S6 SEM image of minerals formed on Si sheet. (a) Minerals formed on bare Si, DGS group and ADGS group after 4 and 8 days of mineralization. (b) Minerals formed on sixth day of mineralization in DGS and ADGS groups.

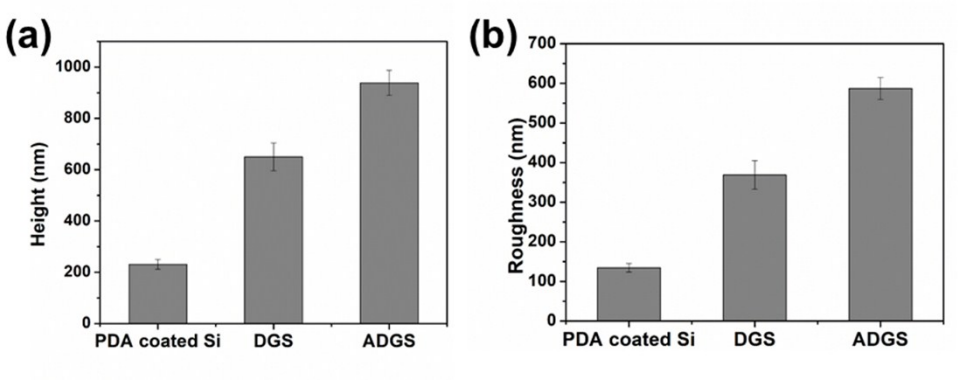

Fig. S7 (a) Statistic height and (b) roughness of different surfaces analysed from AFM results. (n=15)

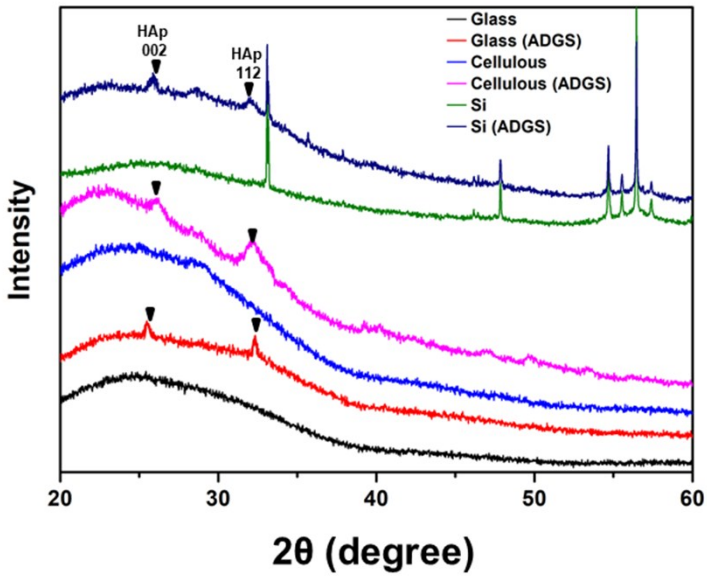

Fig. S8 XRD pattern of different substrates mineralized for 8 days in ADGS group.

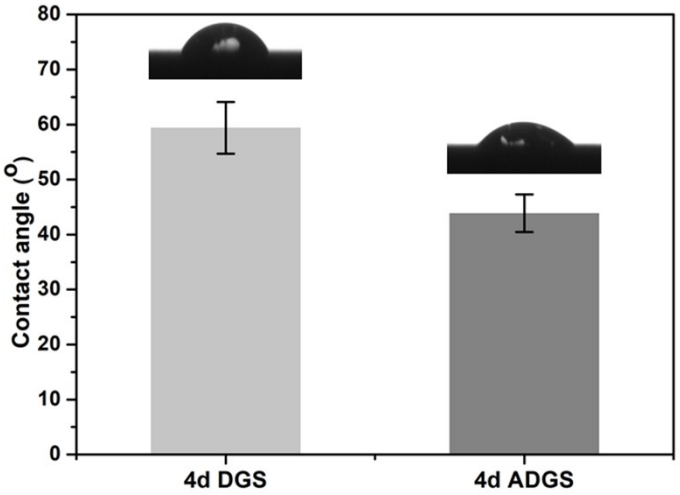

Fig. S9 Water contact angle of Ti substrate in DGS and ADGS group after 4 days of mineralization.

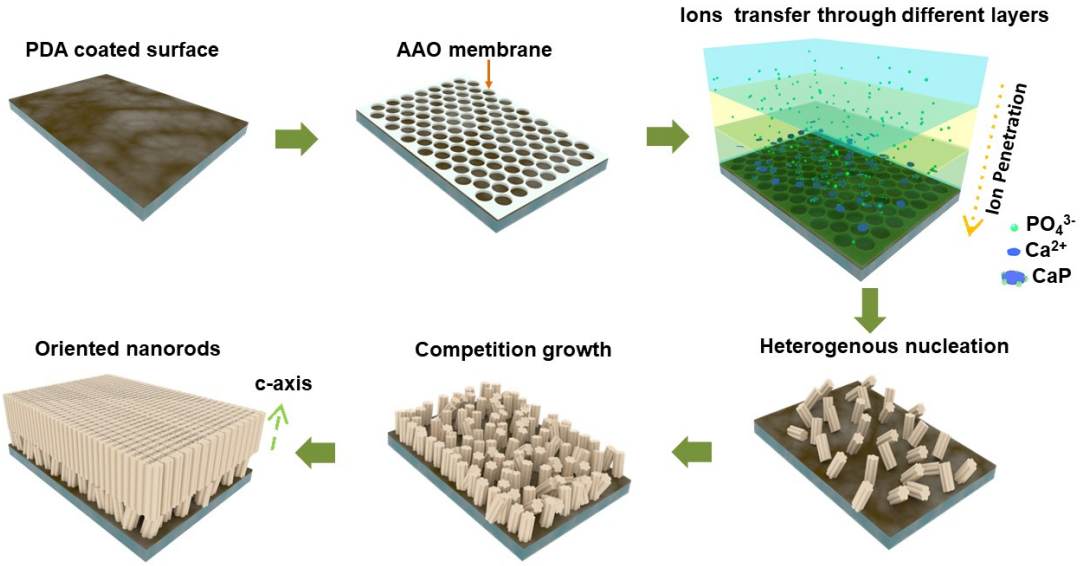

Fig. S10 The assumed mechanism of the ADGS mineralization. PDA serves as binding agent and absorbs calcium ion due to chelation effect, then phosphate ions from upper solution transfer through ion-free gel and reach the calcium-containing gel layer, where some of them combine with calcium ions to form CaPs, others continue going down and arrive the PDA layer, they finally combine with calcium ions which are bond to PDA molecules, becoming CaPs and mineral nucleuses. AAO was added here to help accelerate the transportation of ions and other molecules through the capillary force from its through-hole pores, favouring the fast forming of plenty nucleuses. The hydrophilicity of the substrate surface and AAO membrane help decrease the interfacial energy barrier, which allows the crystals to grow in a heterogenous way. As the mineralization continues, crystals compete for space to grow, resulting the selection of advantageous growth direction, crystals with favourable planes grow faster than other crystals, covering other crystals to form oriented arrays. Finally, only those with fastest growth plane remain, crystals are parallel to each other.