Organic/Inorganic Template-Mediated Mineralization Process

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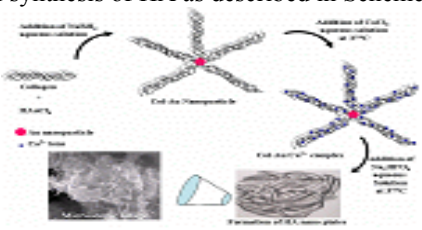
Statement of Purpose: Among the nanometer scaled materials, metallic nanostructure with various shape such as; spherical, plates, hallow, rods, wires etc. have received extensive attention due to their potential application in catalysis, biological labeling, information storage, photonics, and surface enhance Raman scattering (SERS)[1]. However, the utility of nanomaterials for any application is strongly dependent upon their physiochemical characteristics and their interaction with surface modifiers. Selecting a suitable modifier that mimic physiological condition is essential to shield them from aggregation and coalescence. In this work, wet chemical approach has been put forth to grow biomimetic HA using collagen (Col) immobilized gold nanoparticles thereby providing biomimetic environment.

During the mineralization process of bone tissue, Col is first synthesized, extruded form the cell, and then self assembled in the extracellular space. For this reason, bone is the typical example of an "organic matrix-mediated" mineralization process. Herein, inclusion of novel metal, we defined it as "Organic/inorganic template-mediated" mineralization process.

Methods: Col from bovine Achilles tendon, type I, hydrogen tetrachloroaurate ($HAuCl_4$), NaOH, $NaBH_4$, $CaCl_2$, and Na_2HPO_4 were purchased from Sigma Aldrich, USA, and use without further purification. Acetic acid used was purchased from Wako pure chemical industries Ltd. Japan. Experiment was preformed in triple distilled water.Col was immobilized on gold nanoparticles in situ chemical reduction process[2]. The product was centrifuged at 24000 rpm for 45 min. and pellet was recovered and purified by repeatedly washing with triply distilled water to make free from any acid impurity in order to meet physiologically friendly condition for wet chemical synthesis of HA as described in Scheme 1.



Scheme 1. Schematic representation of immobilization of collagen on gold nanoparticles and formation of hydroxyapatite on nano-matrix.

Results and Discussion: The UV-visible absorption spectra of Col-Au nanoparticles exhibit a surface plasmon band at 527 nm that broadens and decreases in intensity after the addition of $CaCl_2$ solution, which on addition of Na_2HPO_4 becomes flattened resulting the precipitation of Col-Au-HA nanocomposite. The formation of Col-Au and Col-Au-HA was characterized by FT-IR spectroscopic analysis. The infrared-active modes of the peptide backbone are expressed in the amide bands. The IR spectra of Col, after the treatment with $NaBH_4$, Col-Au, and Col-Au-HA(Figure 1) shows that the polypeptide back bone and side chain functional groups remain intact.

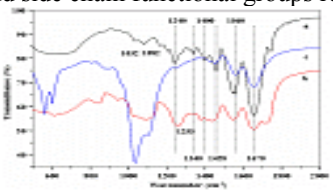


Figure 1. FT-IR spectra of (a) Col, (b) Col-Au nanoparticles, and (c) Col-Au-HA composite.

To provide further evidence of the mineralization, TEM (Figure 2 I) and AFM (Figure 2 II) analysis was performed under ambient conditions. The micrographs provided crucial insight into the morphology of mineral phase and absorption of gold nanoparticles.

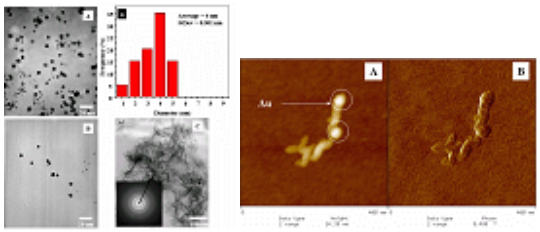


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Figure 2. (I) TEM images of (A) Col-Au nanoparticles recorded form the large aggregated area and its size distribution histogram. (B) Col-AU nanoparticles recorded form the least aggregated area. (C) Hydroxyapatite grown on Col-Au nano-matrix, the inset is the SAED pattern at this stage. (II) Tapping mode AFM images of Col-Au-HA after 2 h of reaction (A) Height image (B) Phase image (C) Three dimensional phase mode.

Conclusion: It is evident from this study that Col controlled nanoscale platelet of hydroxyapatite that are randomly aggregated in the form of quarter moon structure when engineered with gold nanoparticles. With the use of Col immobilized gold nanoparticles, it is expected that ceramics of biological interest may be produced in very economically friendly conditions with the morphology mimicking fibrous Col.

References

[1] B. C. Gates, Chem. Rev. 1995; 95: 511-522.
[2] S. Aryal, R. B. K.C., S. R. Bhattarai, P. Prabu, H. Y. Kim, J. Mater. Chem. DOI: 10.1039/b608300e