## Supporting Information

## **Collagen Functionalized With Graphene Oxide Enhanced Biomimetic Mineralization and In Situ Bone Defect Repair**

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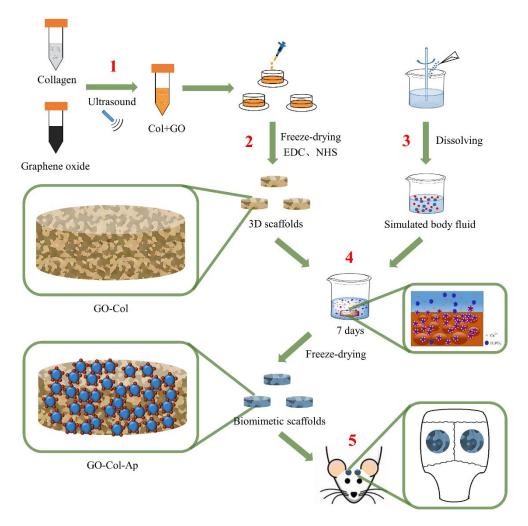
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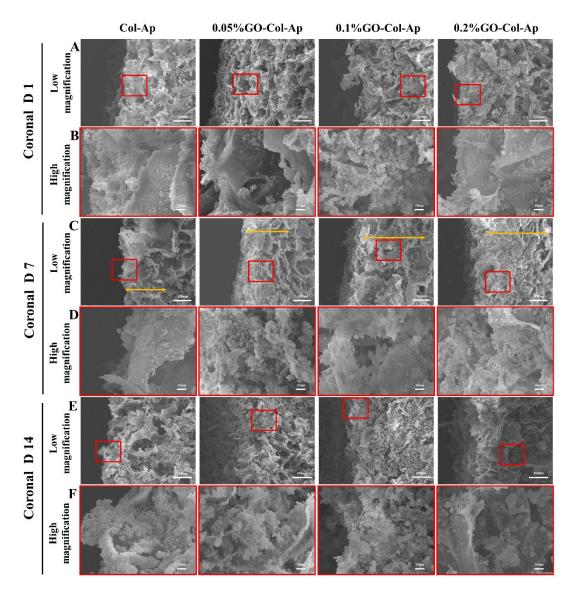
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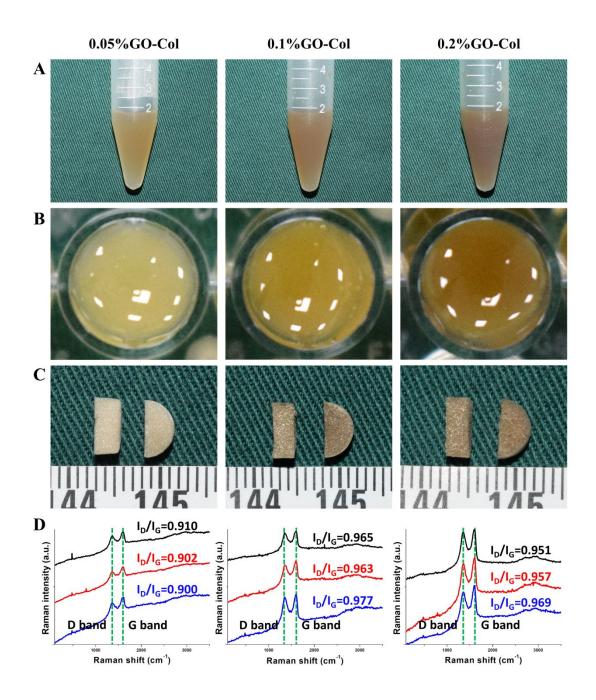
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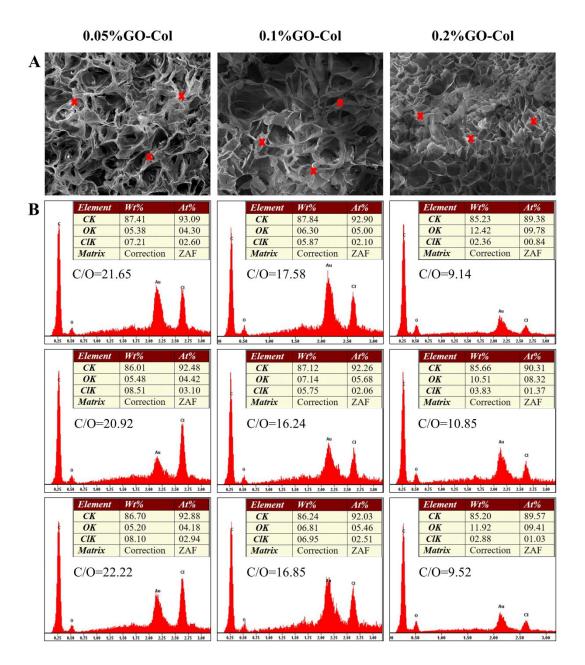
**Figure S1.** Schematic illustration of the preparation of GO-Col-Ap scaffolds and their applications in vivo. (1) The collagen solution and graphene oxide solution were mixed under ultrasound stirring for 30 min to obtain a homogeneous solution. (2) The solution was cast into molds (diameter: 5 mm, height: 1 mm), followed by freeze-drying, crosslinking, and freeze-drying again to form 3D porous scaffolds. (3) Simulated body fluid (SBF) was prepared in accordance with Kokubo's method with ion concentrations approximating those present in human blood plasma. (4) The 3D scaffolds were soaked into SBF solution. (5) The biomimetic scaffolds GO-Col-Ap were implanted into the critical-size bone defect area of rat skull to evaluate the orthotopic bone formation ability.



**Figure S2.** SEM images of the coronal section of the scaffolds. Apatite crystals were observed within the scaffolds after biomimetic apatite deposition at 1 day (A, B), 7 days (C, D), and 14 days (E, F). More apatite depositions were observed after 7 days and 14 days of soaking in SBF (B, D, F). After 7 days of soaking in SBF, the distances of apatite growth into the scaffolds were measured (yellow double-sided arrow in C). The results showed that Col-Ap group (230  $\mu$ m) and 0.05% GO-Col-Ap group (239  $\mu$ m) had significantly shorter distances than 0.1% GO-Col-Ap group (348  $\mu$ m) and 0.2% GO-Col-Ap group (333  $\mu$ m). (Scalar bar in A, C, and E: 100  $\mu$ m, in B, D, and F: 10  $\mu$ m.)



**Figure S3:** Characteristics of the GO-Col solutions and scaffolds. A: Mixture of the GO-Col solutions in tube. B: GO-Col solutions in molds. C: Surface and cross section images of GO-Col scaffolds. D: Raman spectrum of GO-Col scaffolds. (Black, red, and blue curves in D: three random points in the cross section of the GO-Col scaffolds).



**Figure S4:** SEM analysis and EDS evaluation of the GO-Col scaffolds. A: SEM images of the cross section of GO-Col scaffolds. B: EDS evaluation of the corresponding three random points (red cross) in SEM images.