

Supporting Information

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1. Water contact angle test

The video in the supporting information file recorded the whole process of testing the water contact angle on PM-9 scaffolds. The water drop infiltrated completely into the scaffold immediately after contacting to it. This result demonstrated the effectiveness of MBG layer in improving the hydrophilicity of PLLA scaffolds.

2. FTIR spectra of the different scaffolds before and after GS loading

After up-taking GS molecules, all the scaffolds showed the absorption bands of amino groups at 620 cm^{-1} (out-of-plane bending vibration) and 1500 cm^{-1} (formation vibration), which indicated that GS drugs were successfully loaded. In the spectra of MBG-coated scaffolds, as it can be observed in Fig.1S (b, c, d), the Si–O–H band (964 cm^{-1}) present in PM scaffolds mostly disappeared after GS adsorption, which illustrated that GS molecules had interacted with Si–OH groups by hydrogen bonding.

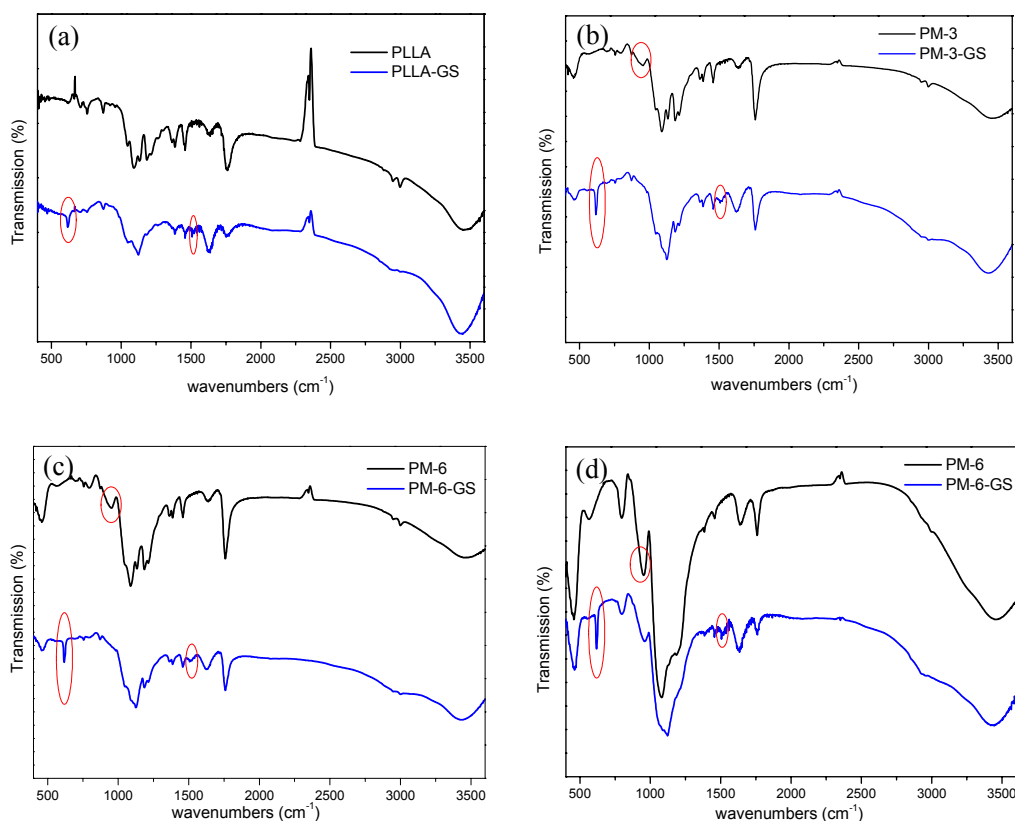


Fig.1S FT-IR spectrograms of various scaffolds before and after GS loading, red circles point out changes of absorption peaks.

3. ICP-AES analysis of the extracted products

Measurements of Si, Ca, P concentrations in the extracted products were carried out through Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES), and results were showed in Table.1S. Compared with phosphorus and silicon, the calcium component was prone to be washed away from the Si-O-Si network due to the unmatched ionic radii of Ca²⁺ and Si⁴⁺. The percentages of Si, Ca and P species losses from the whole MBG coating to the extract solution were theoretically calculated according to the values of MBG fraction of the composites, ICP-AES results and other parameters of extraction. It was

observed that part of Ca and P components still remained in the coating. From PM-3 to PM-9, higher MBG fractions in the composites corresponded to less losses of calcium and phosphorus, because much thicker MBG layers prevented the inner MBG from the quick loss of Ca and P components.

Table.1S Molar ratios and percentages lost in the extract solutions of Si, Ca, P components for composite scaffolds during extracting process.

Samples	Si/Ca/P molar ratio	Percentages of Si, Ca and P lost (%)		
		Si	Ca	P
PM-3	1/2.30/0.79	5.50	67.8	35.1
PM-6	1/3.63/1.29	2.77	54.5	29.1
PM-9	1/4.75/1.45	1.37	36.3	16.1

4. TG-DTA analysis of degraded composites

After 21 days *in vitro* treatment in SBF, the contributions to weight changes of the composites, including the two competing processes of polymer/MBG degradation and hydroxyapatite deposition, were evaluated through thermogravimetric analysis. The degraded polymer mass (Δm_{PLLA}) and decreased mass of the whole scaffold (Δm) were calculated following the equations below.

$$\Delta m_{\text{PLLA}} = m_1 * W_{p1}\% - m_2 * W_{p2}\% \quad S1$$

$$\Delta m = m_1 - m_2 = m_1 * (1 - WL\%) \quad S2$$

In Equation S1, m_1 and m_2 are the mass of the composite before or after treatment in SBF respectively; $W_{p1}\%$ and $W_{p2}\%$ are the PLLA percentage of the composite before or after treatment in SBF respectively.

In Equation.S2, WL% is the mass loss percentage of the composite.

Comparing Δm_{PLLA} and Δm , we can conclude that the deposition of hydroxyapatite could not compensate for the loss of the composite scaffolds due to the degradation of polymer and MBG.

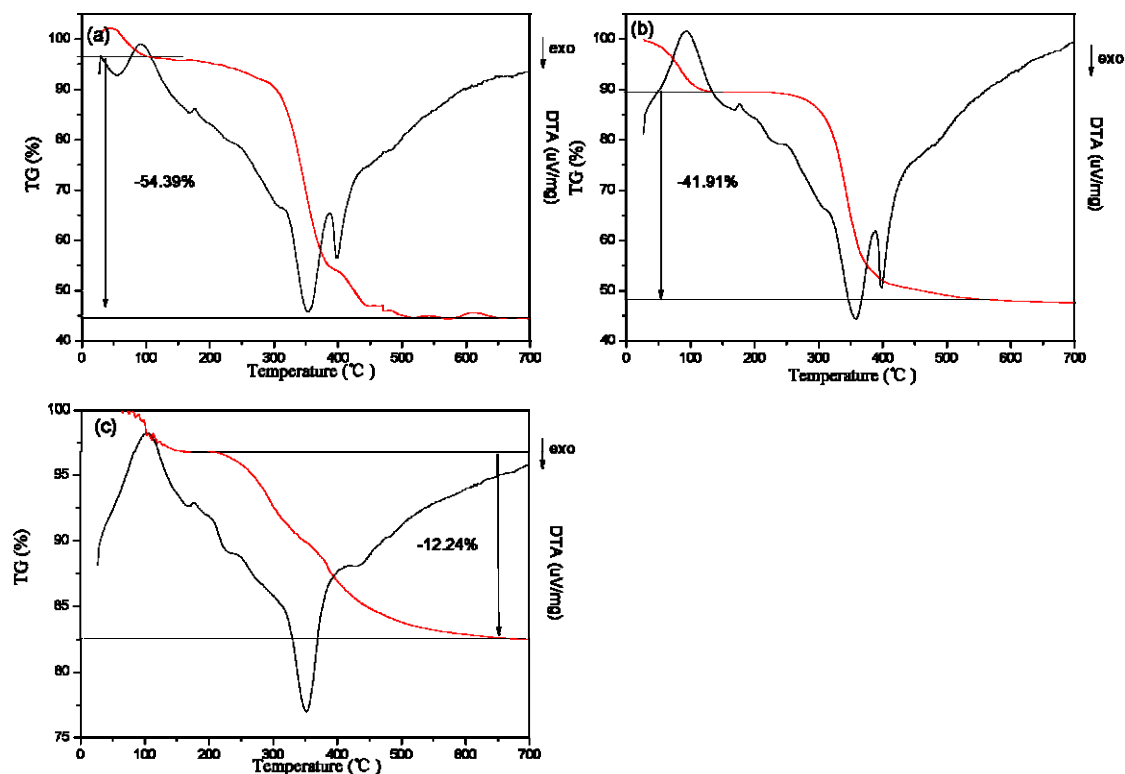


Fig.2S TG-DTA curves of the composites after 3 week treatments in SBF, (a) PM-3, (b) PM-6, (c) PM-9 scaffolds.

Table.2S Parameters in Equation S1, S2 for the three composite scaffolds during SBF treatment.

Samples	PM-3	PM-6	PM-9
m_1	1 g	1 g	1 g
WL%	10.1%	11.7%	12.0%
m_2	0.899 g	0.883 g	0.88 g
$W_{p1}\%$	65.5%	55.2%	27.8%
$W_{p2}\%$	54.4%	41.9%	12.2%

Δm_{PLLA}	0.166 g	0.182 g	0.171 g
Δm	0.101 g	0.117 g	0.120 g

5. Gel Permeation Chromatography

The M_w of PLLA remained in the composites after extraction were estimated by Gel Permeation Chromatography (Spectra Physics SP-8430, waters 2414 detector). The hybrid scaffolds were dissolved using CHCl_3 and after the centrifugation the supernatant were subjected to be examined. Chromatographic separations were carried out with a polystyrene gel column (Waters Styragel; bead size = 5 μm ; column size = 7.8 mm internal diameter = 300 mm) using CHCl_3 as a solvent (flow rate: 1.0 ml/min). The temperature of column was maintained at 30°C.

Table.3S give the molecular weight of PLLA in the composite scaffolds, which revealed the degradation of PLLA during preparation, especially in the P123 extraction using EtOH and HCl. The M_w of PLLA in different PM scaffolds was almost the same at around 42,000 after treated in EtOH and HCl. The values were a little lower than the original 50,000 we used to prepare these scaffolds which demonstrated some degradations.

Table.3S Results obtained from GPC to evaluate the M_w changes of PLLA in the composites

Samples	M_w	M_n	PD (M_w/M_n)
PM-3	41053	24506	1.675
PM-6	41811	29369	1.424
PM-9	41629	27658	1.505