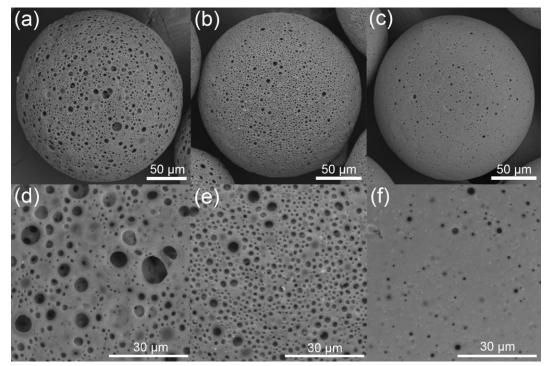


Fig. S1. The SEM images of PLGA/HA microspheres prepared without GDL in the aqueous phase. The size of HA was 4.60 µm.

Compared to the hPH prepared with GDL, this PLGA/HA microspheres had HA particles embedded on the surface. Moreover, the majority of the macropores were closed. This meant that GDL not only removed the exposed HA particles on the surface, but also help the macropores become open. In Figure S1, there were HA particles located near under the surface. These particles were accompanied with cavities, which resulted from the adsorbed water by HA.



**Fig. S2.** Morphologies of small microspheres obtained from the same formulation of hPH (a and d), mPH (b and e) and lPH (c and f). The size of the three kinds of microsphere is around 200  $\mu$ m. And they are designated as hPH-s, mPH-s and lPH-s.

As to the smaller microspheres, the macroporous features were enhanced, especially for hPH-s and mPH-s. This was probably due to the higher specific surface area of smaller microspheres, which facilitated water attraction. The number of pores counted by Image J in d, e and f were 509, 1314 and 164, respectively. Unlike hPH, hPH-s did not have the most pores. This was due to the confluence of water pockets in hPH-s, which reduced the whole number of pores but increased the portion of large pores. The results indicated that morphologies of microspheres also depended on the size.