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## **Supporting Information**

## Design of Silk-Vaterite Microsphere Systems as Drug Carriers with pHresponsive Release Behavior

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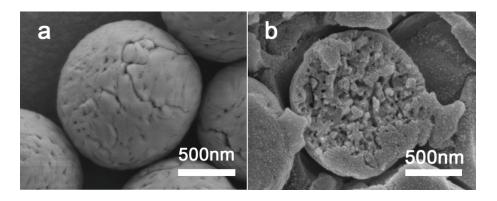
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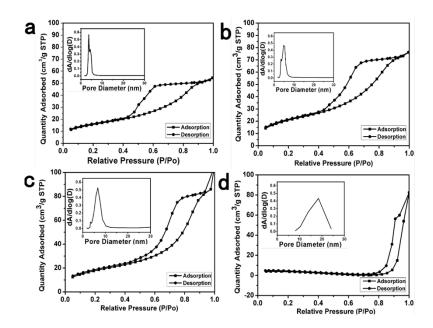
#The author has the same contribution with the first author

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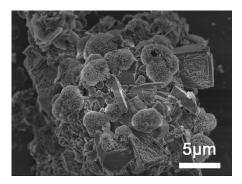
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**Figure S1.** Morphological changes of silk-vaterite microspheres after treatment at 350°C for 6 h: (a) Surface images of the treated microspheres and (b) Cross-section images of the treated microspheres. Significant fusion of vaterite nanoparticles was observed after the treatment.



**Figure S2.** Nitrogen adsorption-desorption of silk-vaterite microspheres after the different thermal treatments. The samples were as follows: (a) M-0; (b) M-300; (c) M-305; and (d) M-310. The pore size increased while the surface area of the microspheres increased and then decreased following the increase of treatment temperature and time.



**Figure S3.** Typical microstructures of DOX-loaded silk-vaterite microspheres when the loaded DOX released in PBS solution for 8 days. The vateritecalcite/hydroxyapatite transition appeared in the drug-release process.