## **Supplementary Information**

## Fabrication of Self-Healing Hydrogels with Surface Functionalized Microcapsules from Stellate Mesoporous Silica

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A certain amount of EGDA and BIE in ampoules were showed in Figure S3 (a) and S3 (b) without UV light. It can be seen that EGDA cannot be polymerized without UV light condition for 2 hours. As shown in S3 (c) and S3 (d), it was observed that EGDA and BIE had polymerized about 30 minutes under irradiation of a UV lamp (375 nm). Figure S5 are TGA curves of STMS microcapsules loaded with or without healing agent and further functionalized. From Figure S5 (a), the STMS has a slight weight loss about 5.8%. As can be seen from the figure, the weight loss percentage of STMS-EGDA/BIE (Figure S5 (c)) is about 33.8%, which is basically consistent with the mass ratio of 1:3 from the initial addition of STMS and EGDA/BIE. Then, the obtained STMS-EGDA/BIE particles were encapsulated by PDA, and the surface PDA is pyrolyzed to produce weightlessness. As can be shown in Figure S4 (b), the PDA@STMS-EGDA/BIE has a slight weight loss about 38.0 %. Figure S4 (d) shown that P(MAUPy-co-nBA) was attached to the exterior of the microcapsules by modification of the surface PDA and has a weight loss of 44.0%. These results showed functional microcapsules were successfully modified.



**Figure S1.** <sup>1</sup>H NMR spectrum of 2,5-Dioxopyrrolidin-1-yl pent-4-ynoate (400 MHz, CDCl<sub>3</sub>, 298 K).



Figure S2. <sup>1</sup>H NMR spectrum of MAUPy (400 MHz, CDCl<sub>3</sub>, 298 K).



**Figure S3.** Effect of ultraviolet light on polymerization of EGDA in the presence of photoinitiator (BIE). Healing agents (3.0 mL) and photoinitiators (10 mg) without UV light condition for 2 hours (a and b). Healing agents (3.0 mL) and photoinitiators (10 mg)with UV light condition for 30 min (c and d).



Figure S4. SEM image of microcapsules after ultrasonication (the working distance was 7.5 mm

and accelerating voltage was 10.0 KV).



**Figure S5.** Thermogravimetric analysis (TGA) of curves (a) STMS, (b) PDA@STMS-EGDA/BIE, (c) STMS-EGDA/BIE, and (d) P(MAUPy-*co*-nBA)@STMS-EGDA/BIE.



**Figure S6.** Rheological characterization of prepared hydrogel (blank hydrogel): (a) storage modulus (G') and loss modulus (G'') values are investigated through dynamic strain sweep; (b) an immediate recovery after the 1000% strain deformation (t = 200 s,  $\gamma = 1\%$ ); (c) dynamic strain amplitude cyclic test ( $\gamma = 0.5\%$  or 400%).



**Figure S7.** Rheological characterization of prepared hydrogel (5 mg of microcapsules): (a) storage modulus (G') and loss modulus (G'') values are investigated through dynamic strain sweep; (b) an immediate recovery after the 1000% strain deformation (t = 200 s,  $\gamma = 1\%$ ); (c) dynamic strain amplitude cyclic test ( $\gamma = 0.5\%$  or 400%).



**Figure S8.** Rheological characterization of prepared hydrogel (10 mg of microcapsules): (a) storage modulus (G') and loss modulus (G'') values are investigated through dynamic strain sweep; (b) an immediate recovery after the 1000% strain deformation (t = 200 s,  $\gamma = 1\%$ ); (c) dynamic strain amplitude cyclic test ( $\gamma = 0.5\%$  or 400%).



Figure S9. The stretch process of self-healing hydrogel.



**Figure S10.** The self-healing process of hydrogels (5mg microcapsule) were observed by optical microscope at ambient temperature, (a)  $t = 0 \min$ , (b)  $t = 30 \min$ , (c) t = 1 h, (d) t = 2 h, (e) t = 3 h, (f) t = 4 h.



**Figure S11.** The self-healing process of hydrogels (without microcapsule) were observed by optical microscope at ambient temperature, (a)  $t = 0 \min$ , (b)  $t = 30 \min$ , (c) t = 1 h, (d) t = 2 h, (e) t = 3 h, (f) t = 4 h.