Electronic Supplementary Information (ESI)

Multilayer composite microcapsule synthesized by Pickering emulsion

templates and its application in self-healing coating

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Fig. S1. (a) A chemical structure model of lignin, (b) size distribution of lignin particles obtained from dynamic light scanning (DLS), (c) photograph and transmission electron microscopy (TEM) images of (d) aggregated, (e) dispersed alkaline lignin.



Fig. S2. Photographs of samples: (a) the alkaline lignin at pH=7, (b) lignin dissolved at pH=11, (c) lignin particles suspension at pH=3, (d) After standing for 10minutes, lignin particles deposited in water.



Fig. S3. OM images of Pickering emulsions (sample 3) synthesized by hand shaken method for (a) three minutes, and (b) ten minutes. All scale bars are $200 \mu m$.



Fig. S4. OM images of Pickering emulsion (sample 3) standing in room temperature for (a) 0

h, (b) 3 h, (c) 6 h. All scale bars are 200 μ m.



Fig. S5. OM images of Pickering emulsions with different oil phase (same oil-water volume ratios): (a) mixture of 4 mL IPDI and 1 mL MDI, (b) 5 mL pure IPDI. All scale bars are 200 μ m.



Fig. S6. OM images (a,b,c) of Pickering emulsions with different IPDI-MDI volume ratios in oil phase (O/W volume ratios are 1:1) and SEM images (d,e,f) of microcapsules synthesized with different Pickering emulsions. (a,d) 4 mL IPDI and 1 mL MDI, (b,e) 2 mL IPDI and 3 mL MDI, (c,f) 0 mL IPDI and 5 mL MDI. All scale bars are 200 μm.



Fig. S7. TGA curves of microcapsule with different IPDI-MDI volume ratios in oil phase (O/W volume ratios are 1:1). Blue: 4 mL IPDI and 1 mL MDI; Orange: 2 mL IPDI and 3 mL MDI; Green: 0 mL IPDI and 5 mL MDI.



Fig. S8. OM images of Pickering emulsions with different oil-water volume ratios. (a) 2.0:1,(b) 1.6:1, (c) 1.2:1, (d) 0.8:1, (e) 0.4:1, (f) 0.1:1. All scale bars are 200 μm.



Fig. S9. OM images of scratched region on (a) common coating and (b) self-healing coating embedded with microcapsules which were immersed in water for seven days. The scale bars are $200 \mu m$.



Fig. S10. OM images of (a) microcapsules immersed in water for seven days and (b) crushed microcapsules.



Fig. S11. The original picture of steel plate after peeling off coating (B3 in Fig. 8).



Fig. S12. (a,b) Bode plots obtained on steel specimens coated with common coatings (red) and the self-healing coatings (blue) without scratch.



Fig. S13. Bode plots obtained on steel specimens coated with common coatings (red) and the self-healing coatings (blue) after immersed in 10 wt.% NaCl solution for 24 h (solid line) and for 72 h (dotted line).



Fig. S14. Nyquist plot obtained on steel specimens coated with common coatings (red) and the self-healing coatings (blue) after immersion in 10 wt.% NaCl solution for 72 h.