Supplementary information

Natural shape-retaining microcapsules with shells made of chitosan-coated colloidal lignin particles

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Figure S1 | (A) UV-vis spectra of ciprofloxacin in dilute acetic acid solution at various concentrations with the maximum UV absorbance at 277 nm. (B) UV absorbance of ciprofloxacin at 277 nm as a function of ciprofloxacin concentration.



Figure S2 | **Apparent zeta potential of chitosan plotted against the concentration.** 1 wt-% chitosan was firstly dissolved in 0.1 M acetic acid and then diluted with pH 4.5 aqueous acetic acid.



Figure S3 | (A) Intensity-based particle diameter distributions of the chi-CLPs (0.2 wt-%) with the mass ratio of chitosan to CLP varying from 0 to 200 mg/g. (B) Number-based zeta potential distributions of the chi-CLPs (0.2 wt-%) with the mass ratio of chitosan to CLP varying from 0 to 200 mg/g.



Figure S4 | (A) Diameter distributions of the oil droplets stabilized by chi-CLPs (0.2 wt-%) with the mass ratio of chitosan to CLP varying from 0 to 200 mg/g. (B) Mean oil droplet diameter (d_{43}) and uniformity (according to Eq. 1) of the emulsion plotted against the concentration of chi-CLP (50 mg/g).



Figure S5 | Olive oil-in-water (volume ratio 1:1) Pickering emulsions formed by vigorous hand-shaking for 60s. (A) Photograph of the Pickering emulsions stabilized by chi-CLPs (0.2 wt-%) with the mass ratio of chitosan to CLP varying from 0 to 200 mg/g. (B) Optical microscopic images of the Pickering emulsions stabilized by chi-CLPs (0.2 wt-%) with the mass ratio of chitosan to CLP varying from 0 to 200 mg/g (Scale bars: 200 μ m).



Figure S6 | Olive oil-in-water emulsions stabilized by chitosan molecules at the concentration varying from 0.02 to 1 wt-%. Emulsions were formed by ultrasonication and the droplet diameter were measured 3 days after emulsion formation.



Figure S7 | Comparison of the stability of the olive oil-in-water (volume ratio 1 : 1) emulsions: 1 wt-% chitosan-stabilized emulsion on day 0 (1) and after six months (3); 1 wt-% chi-CLP (50 mg/g) stabilized emulsion on day 0 (2) and after six months (4).



Figure S8 | **Surface morphology of the oil droplet covered by STP cross-linked chi-CLPs** (**50 mg/g**), **imaged with E-SEM (oil was solidified at -20** °C). Images were captured after a prolonged exposure of the Pickering emulsion droplet to the electron beam.



Figure S9 | **Creaming behavior of the Pickering emulsion stabilized by 1 wt-% chi-CLP** (**50 mg/g**), **measured with Turbiscan.** Height was rescaled by starting from 0 at the intensity 0.5 (indicated by the dashed lines).



Figure S10 | Optical microscopic images of sodium triphosphate (STP) cross-linked and non-cross-linked olive oil-in-water (volume ratio 1:1) Pickering emulsions stabilized by 1 wt-% chi-CLP (50 mg/g). (A) STP cross-linked emulsion in wet state (oil droplets in water), in dry state (water evaporated at room temperature) and after rewetting with deionized water. (B) Non-cross-linked emulsion in wet state, dry state and after rewetting as the reference. Scale bars: 80 μm.