Supporting Information

Self-Stripping of Free-Standing Microparticle Gel Membranes Driven by Asymmetric Swelling

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1. Supplementary Movies captions (Movie S1-5)

Supplementary Movie S1.

Swelling process of a microparticle gel/PET membrane. The microparticle gel/PET membrane was formed at 120°C. When water dropped on it, the membrane gradually became swelling with the increasing of size during about 1 min.

Supplementary Movie S2.

Self-sloughing of a microparticle gel membrane from PET substrate. The microparticle gel/PET membrane was formed at 60°C. While we put it into water, the microparticle gel membrane immediately started to escape from the PET substrate.

Supplementary Movie S3.

Self-sloughing of a microparticle gel membrane prepared at 100°C from PET substrate.

Supplementary Movie S4.

Self-sloughing of a microparticle gel membrane from PET substrate in 25% saline solution.

Supplementary Movie S5.

Self-sloughing of a microparticle gel membrane from PP substrate in 0.9% saline solution.

2. Supplementary Figures

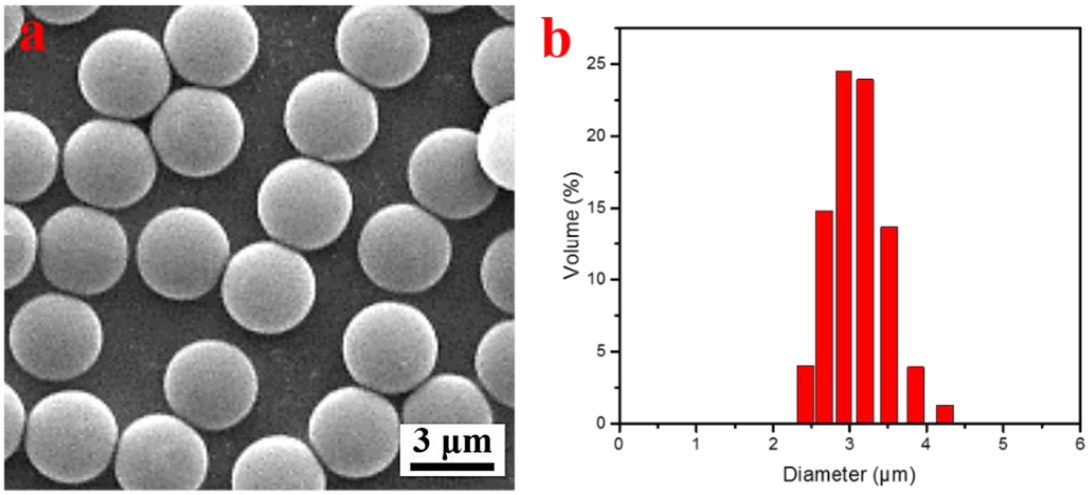

Figure S1. SEM image and DLS size distribution of PGMA seed particles (**a**, **b**).

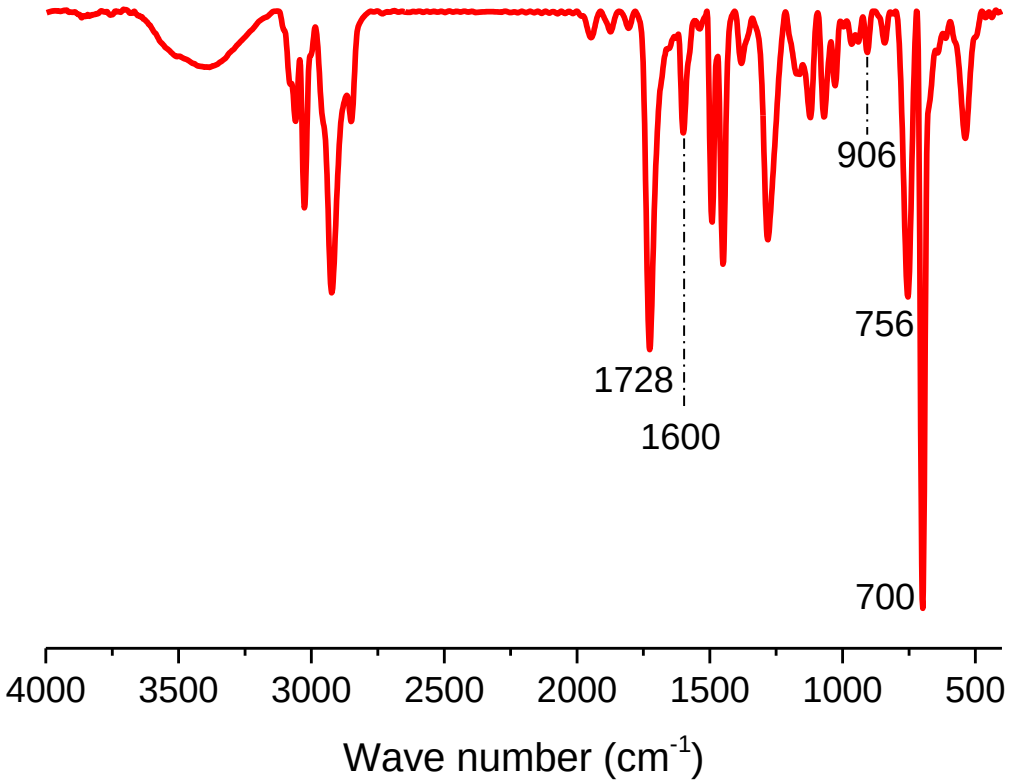

Figure S2. FT-IR spectrum of PGMA/PSt core-satellite superparticles.

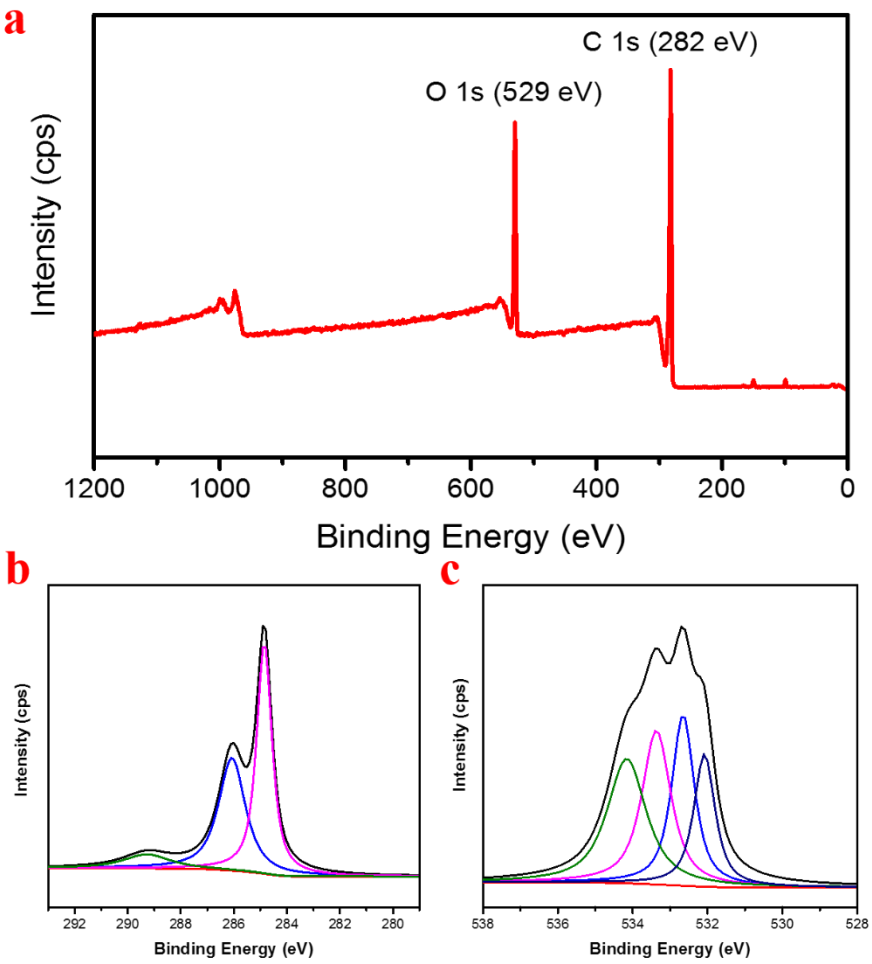

Figure S3. (a) XPS spectra of PGMA/PSt core-satellite superparticles. (**b** and **c**) C 1s and O 1s XPS of core-satellite superparticles.

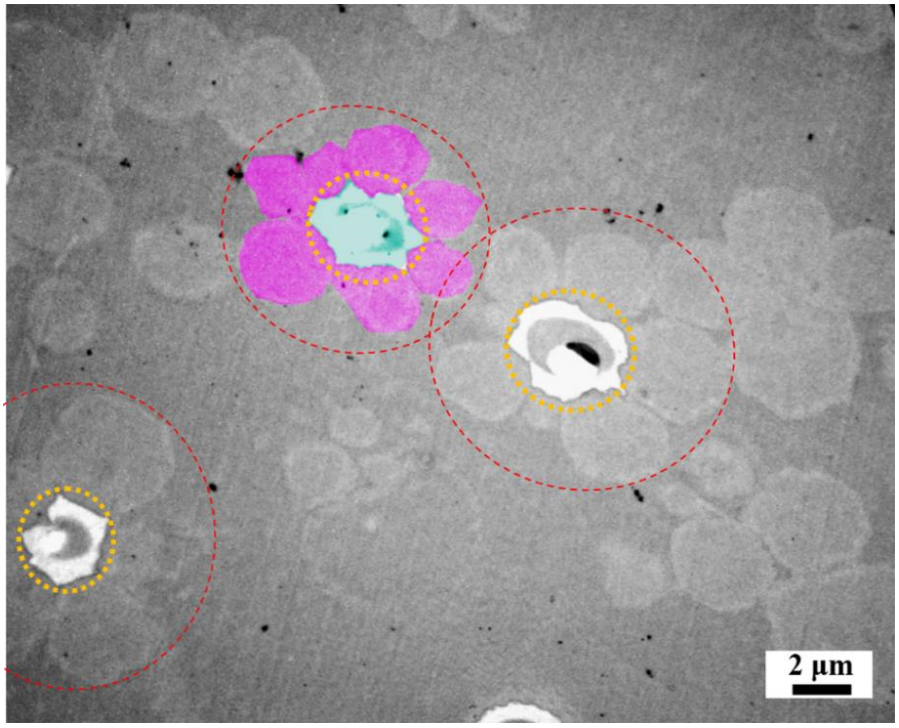

Figure S4. TEM image of ultrathin section of core-satellite superparticles.

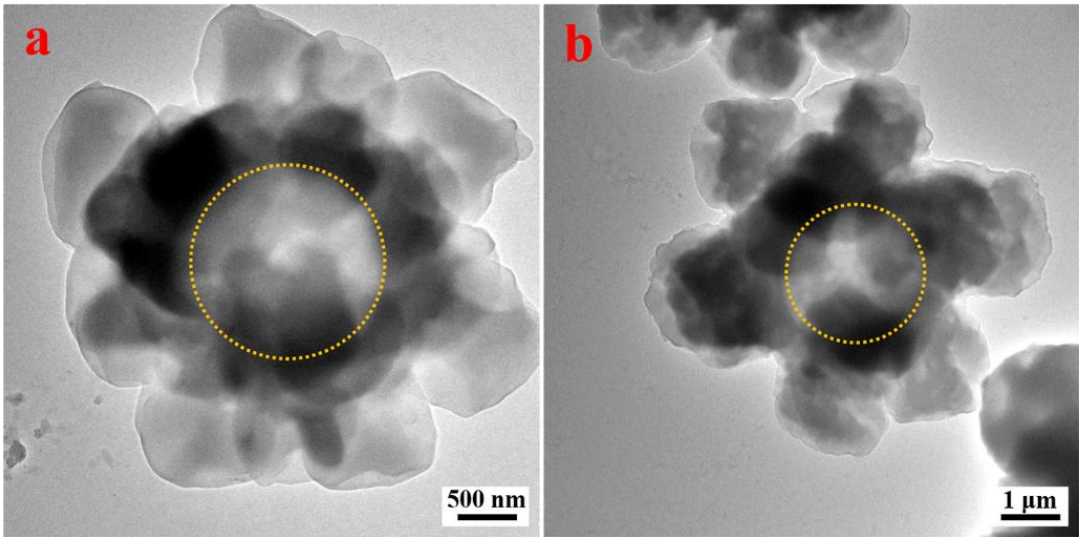

Figure S5. TEM images of core-satellite superparticles with hollow structure.

As mentioned above, patchy particles with different structures were prepared with various DBP/S ratios (Supplementary Figure S6). From Figure S6, we conclude that the increasing of S facilitates the transformation of patchy particles to Janus particles which compose of a smooth hemisphere and a raspberry-like hemisphere (Supplementary Figure S6 and S7). When increasing DBP, these patchy particles tend to irregular, and the smooth hemisphere is disappeared. Interestingly, when we dispersed the prepared particles (as shown in Supplementary Figure S7) in dimethyl formamide (DMF) at 40°C for 6 h, the special patchy particles partially dissolved although the crosslinking agent was not used in the whole procedure, and single-hole particles like the swollen PGMA seeds but with many dimples on the surface (Supplementary Figure S7b). The results indicate that the swollen PGMA seeds performed the self-crosslinking reaction in the process of double-speed swelling polymerization.

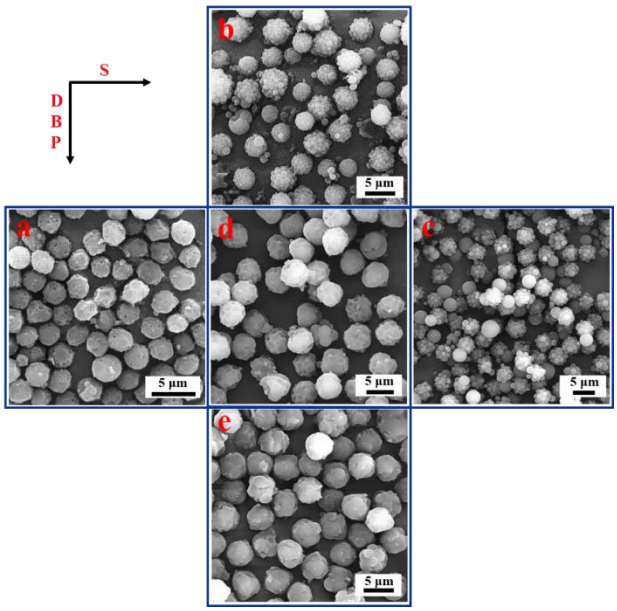

Figure S6. Patchy particles were synthetized with various DBP/S ratios. (a) 1.0/2.5, (b) 1.0/5.0, (c) 1.0/7.5, (d) 2.0/5.0 and (e) 3.0/5.0.

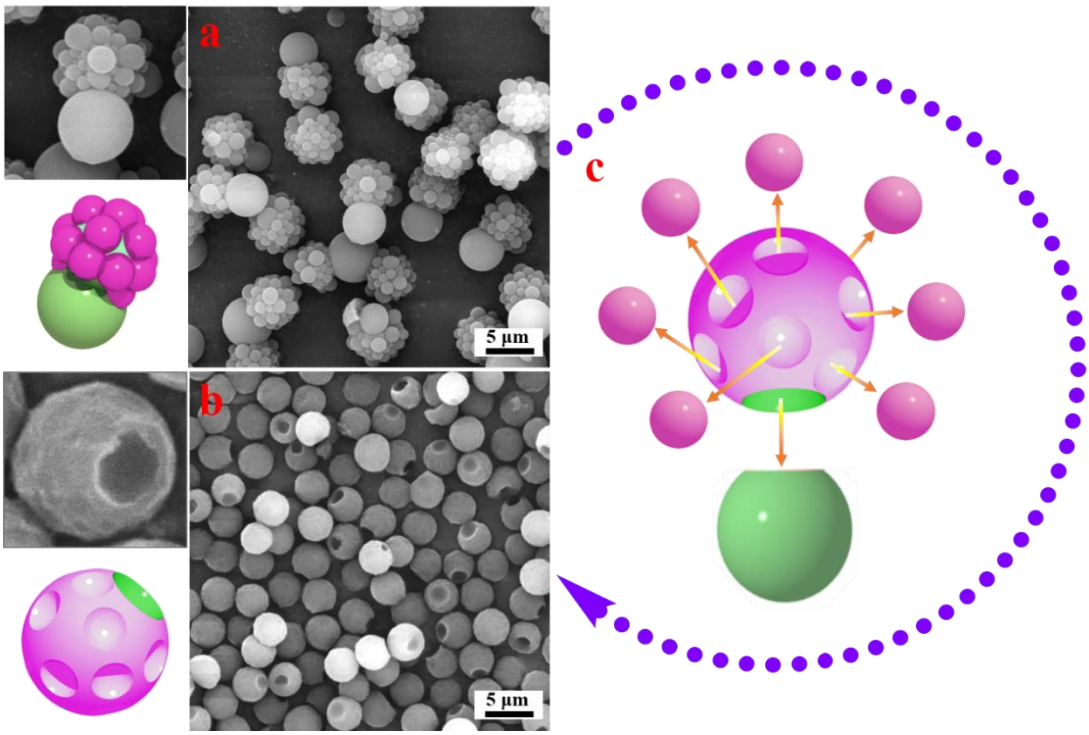

Figure S7. The dissolution experiment of patchy particles synthetized with DBP/S=1.0/7.5. SEM images of patchy particles before (a) and after (b) dissolved by DMF. (c) Schematic diagram of the dissolution process.

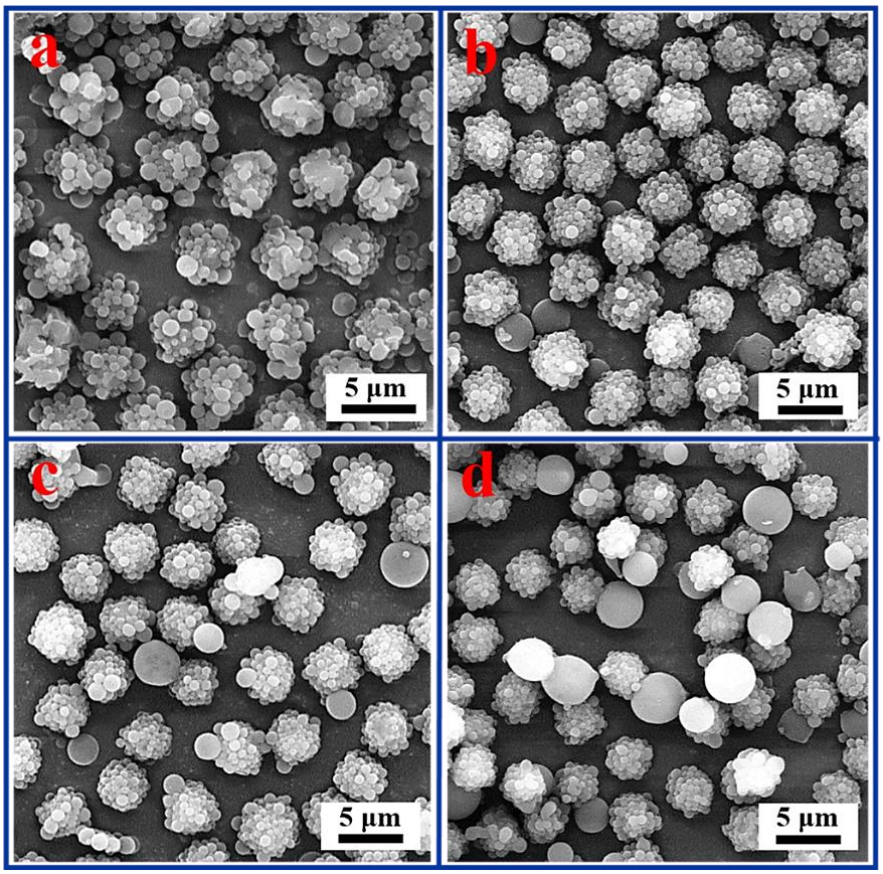

Figure S8. Patchy particles were fabricated with different Sp at Ss=10/6/1: Sp=1 (a), Sp=2 (b), Sp=4 (c) and Sp=6 (d).

When divinylbenzene (DVB) was used as crosslinking agent, the patchy particles with raspberry shape were formed with Sc=1.6% (Supplementary Figure S11). Then, as Sc was increased, the patchy particles become smooth and the protrusions clearly decreased in size. Ultrathin sections of the patchy particles (Supplementary Figure S12) were observed by TEM. Irregular shell was clearly appeared.

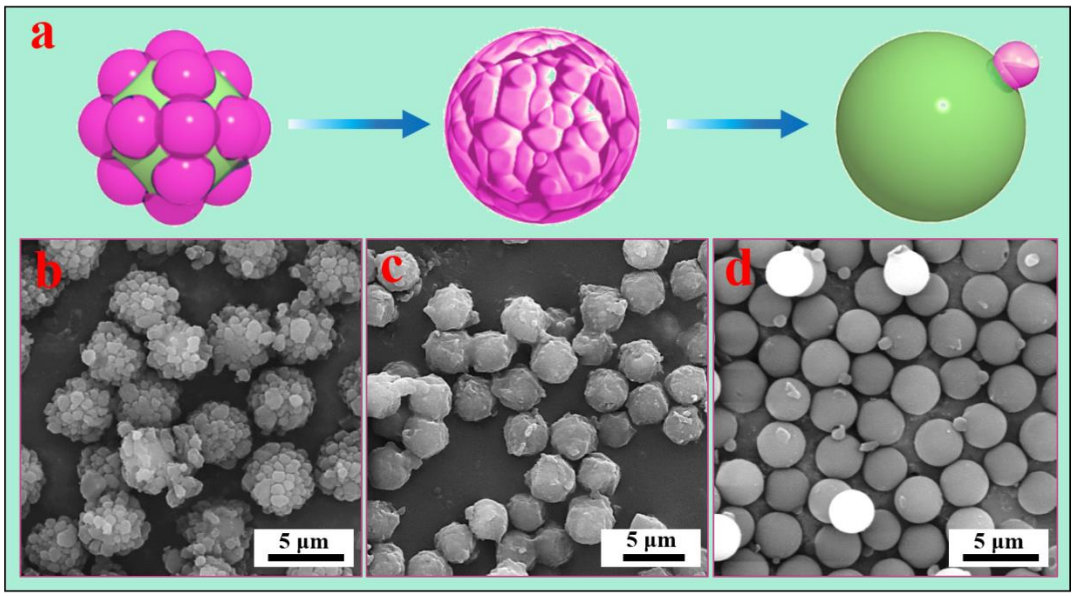

Figure S9. Preparation of the crosslinked patchy particles by double-speed swelling polymerization with DVB as crosslinking agent. (a) Schematic diagram of the morphology evolution of the crosslinked patchy particles synthetized with different ratios of DVB to the swelling monomer Sc: (b) 1.6%, (c) 5% and (d) 16%.

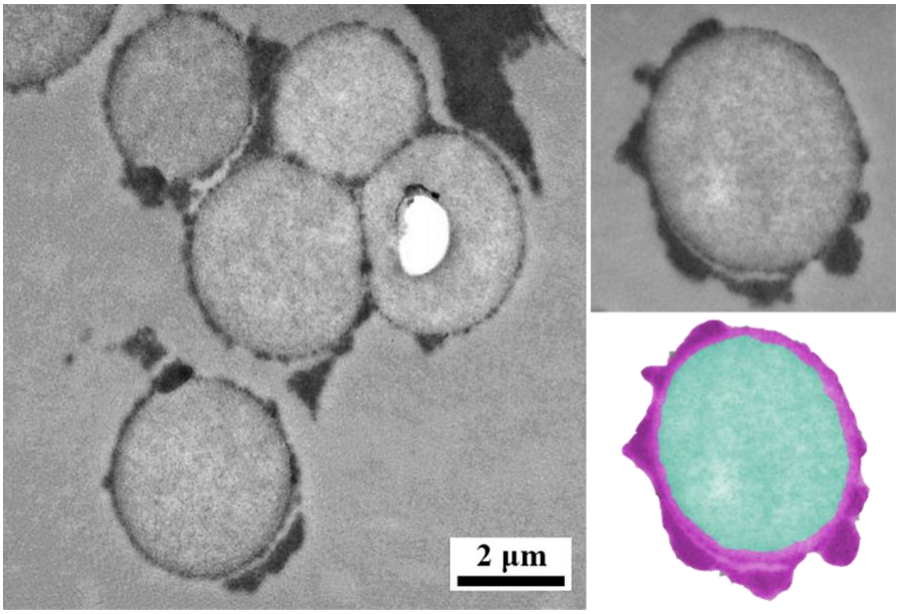

Figure S10. TEM image of ultrathin section of the crosslinked particles prepared with DVB (Sc=5%) and Sp=0.

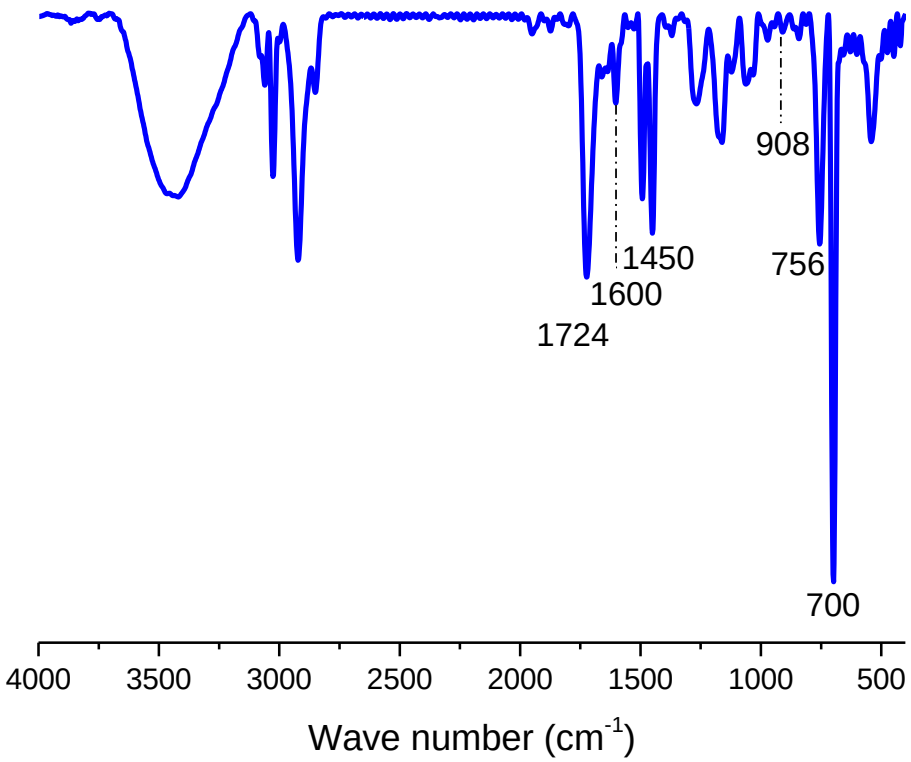

Figure S11. FTIR spectra of yolk–shell crosslinked particles.

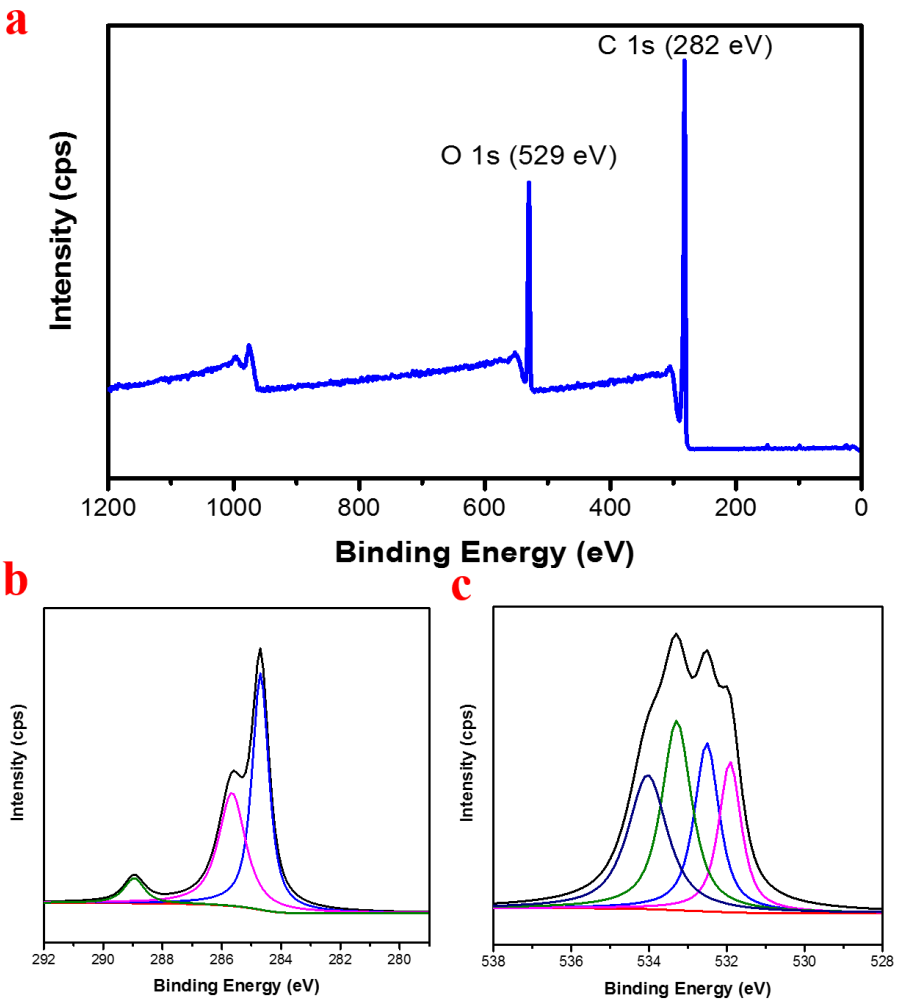

Figure S12. XPS spectra of yolk–shell crosslinked particles.

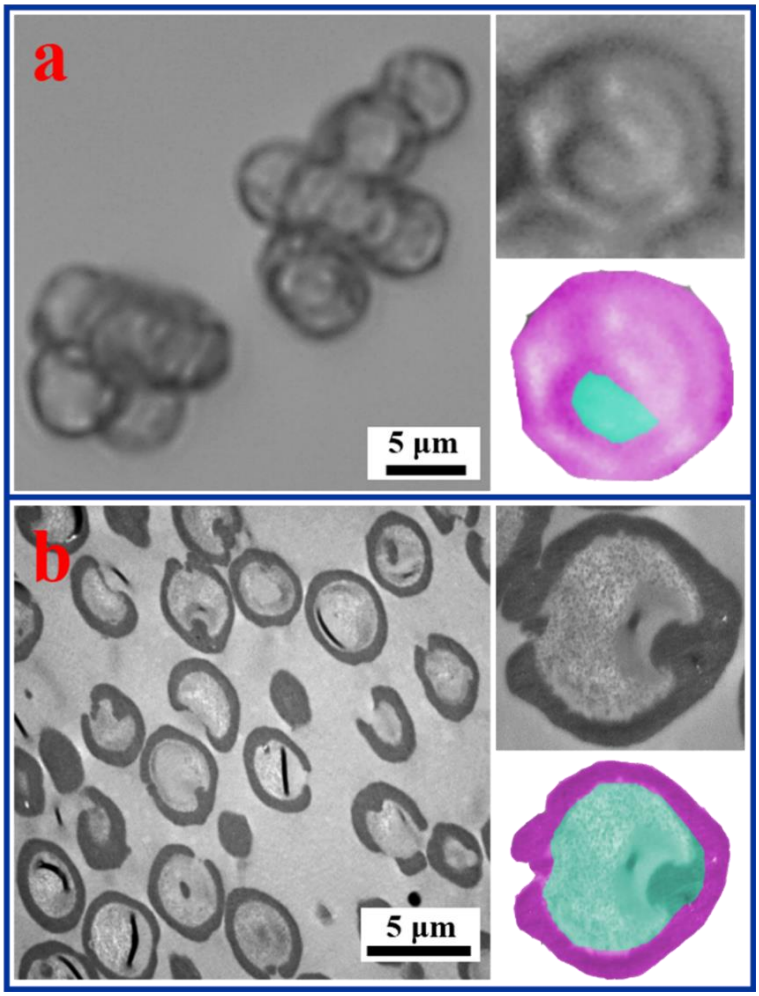

Figure S13. (a) OM picture of yolk–shell crosslinked particles prepared with Sp=4, Ss=10/6/1 and Sc=8.3% of EGDA. (b) TEM image of ultrathin section of yolk–shell crosslinked particles prepared with Sp=6, Ss=10/6/1 and Sc=8.3% of EGDA.

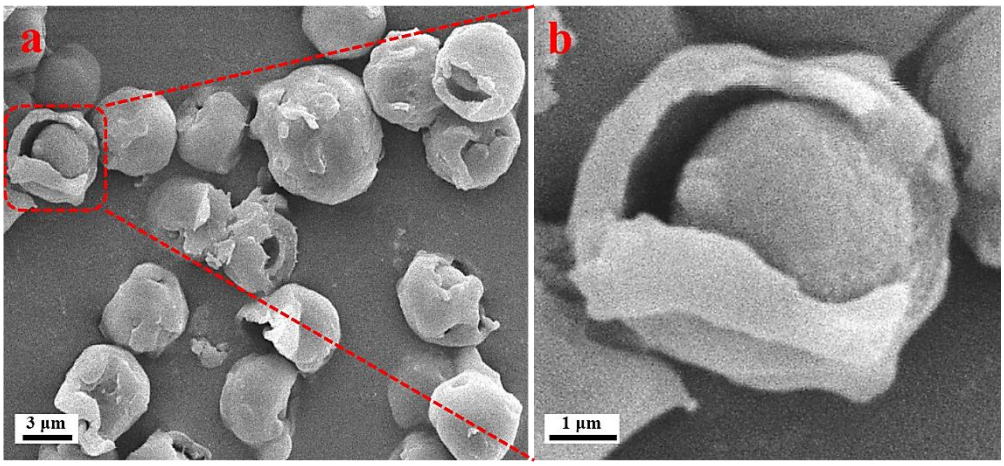

Figure S14. SEM images of yolk–shell crosslinked particles repeatedly prepared with Sp=6.

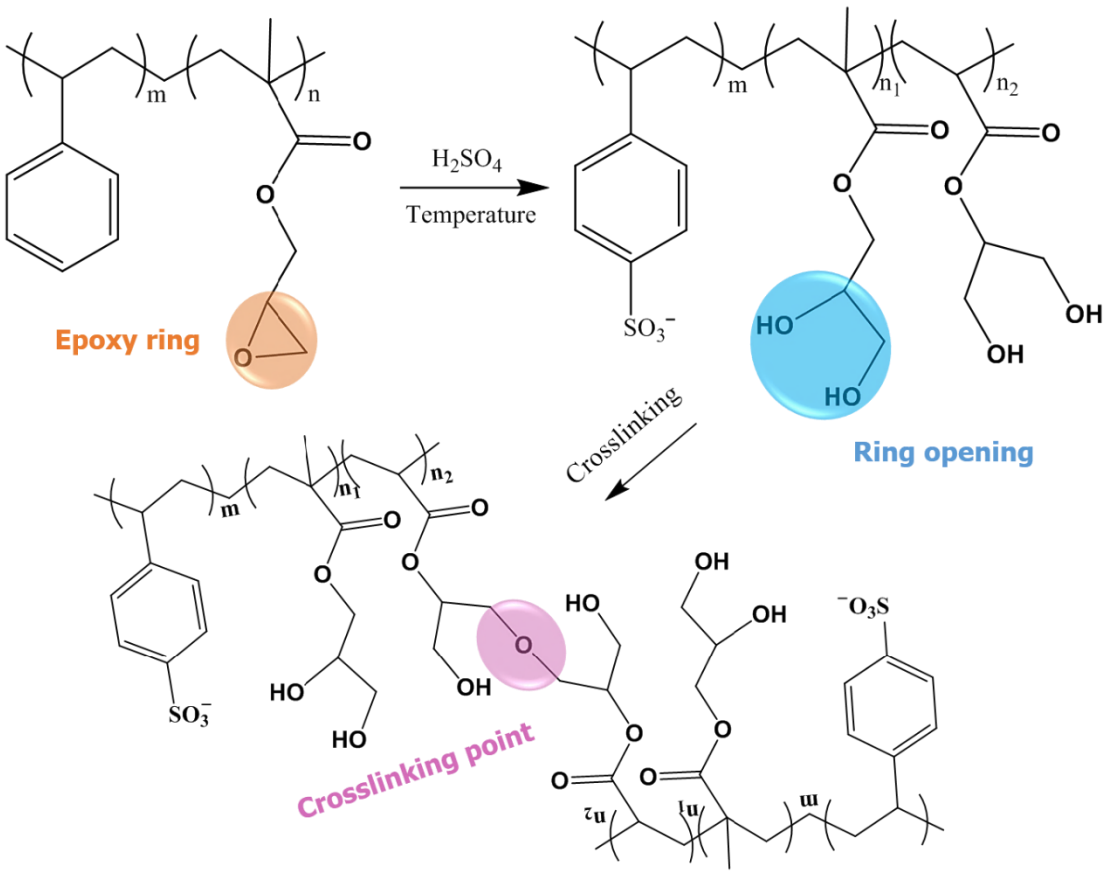

Figure S15. Synthesis of microparticle gels. Microparticle gels were synthetized by one-step acidification process by concentrated sulfuric acid. Simultaneously, PSt was sulfonated to enhance its hydrophilicity.

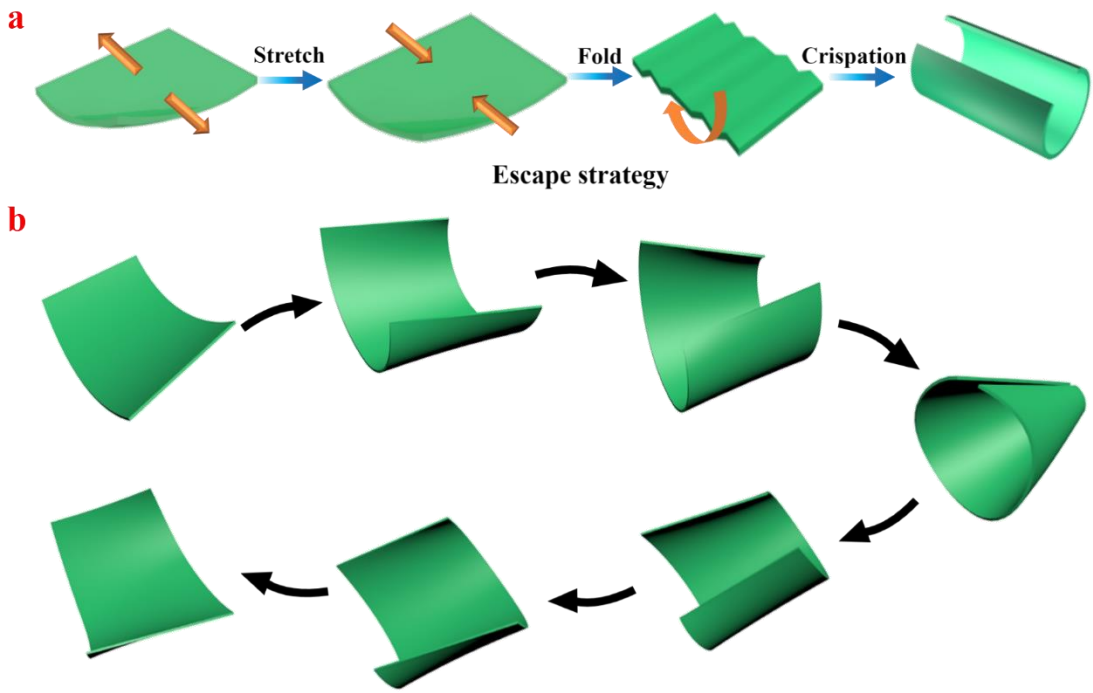

Figure S16. The process of self-stripping of a microparticle gel membrane from PET (a) in water and PP substrate in 0.9% saline solution (b).

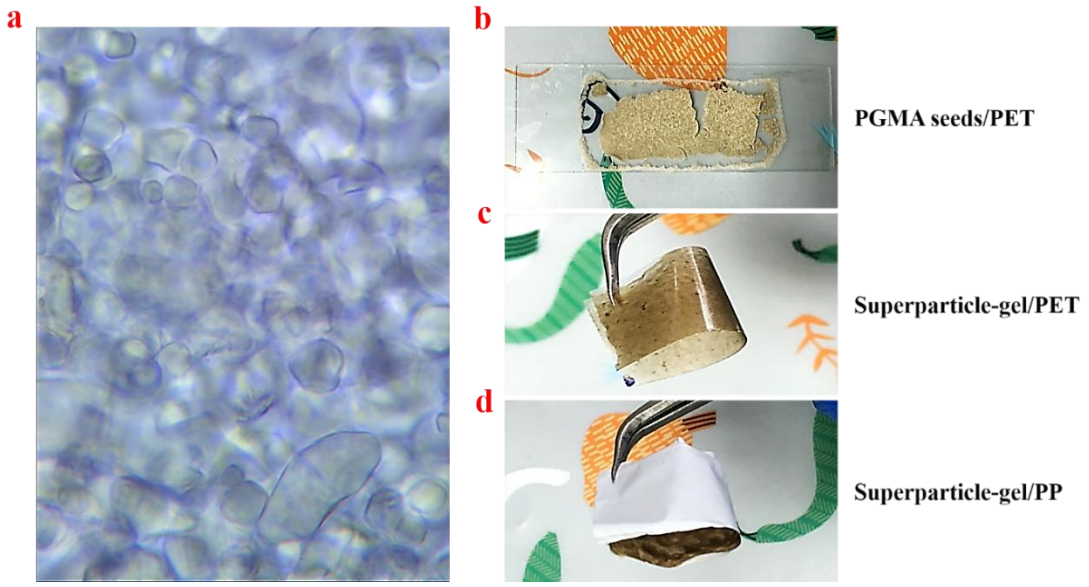

Figure S17. The specificity of our prepared patchy particles compared with smooth spherical particles. (a) OM picture of PGMA seeds treated with the same acidification process. (b) PGMA microparticle gels cannot self-assemble into membranes. (c) and (d) Photographs of microparticle-gel/PET and microparticle gel/PP membranes.

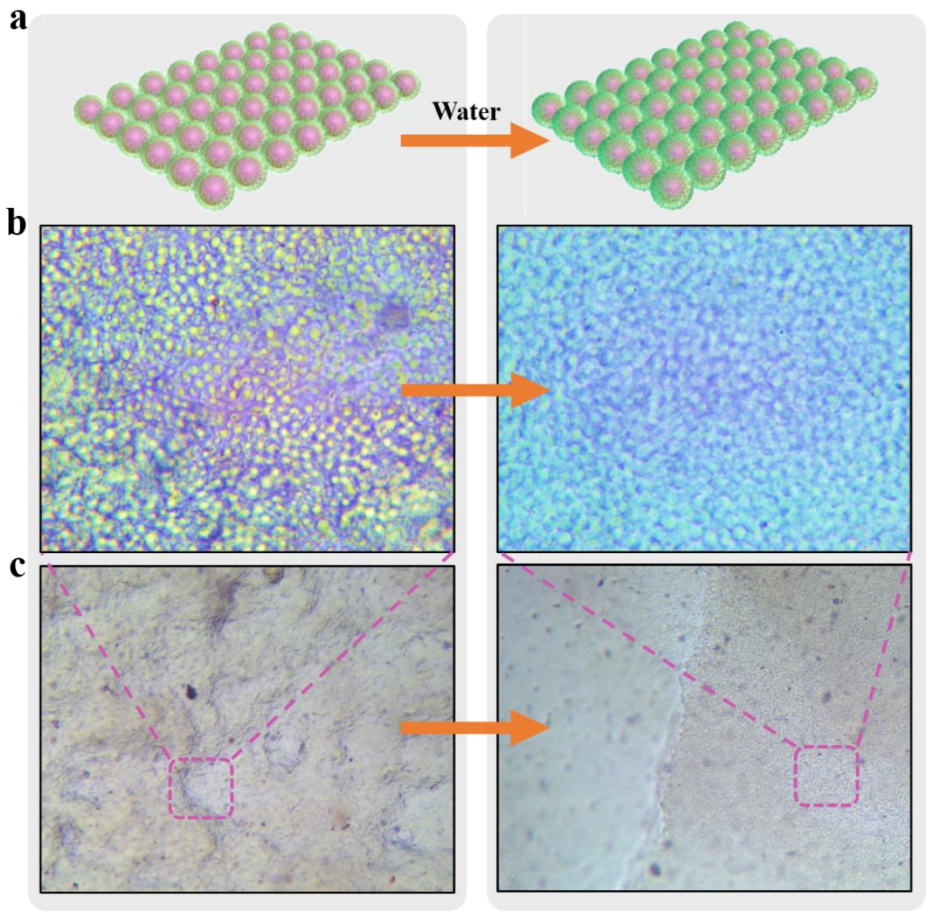

Figure S18. The structure alterations of microparticle gel/PET membranes before and after swelling.

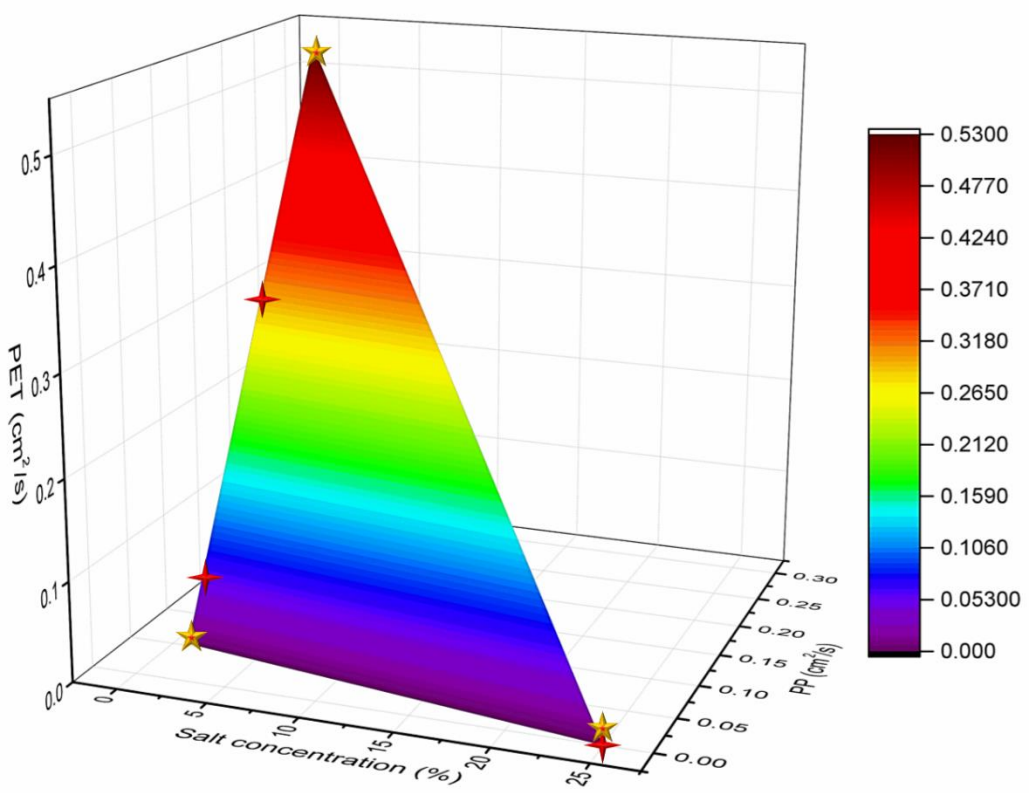

Figure S19. Stripping factor of microparticle gel membranes stripping from PET and PP substrate in saline solution with different salt concentrations.

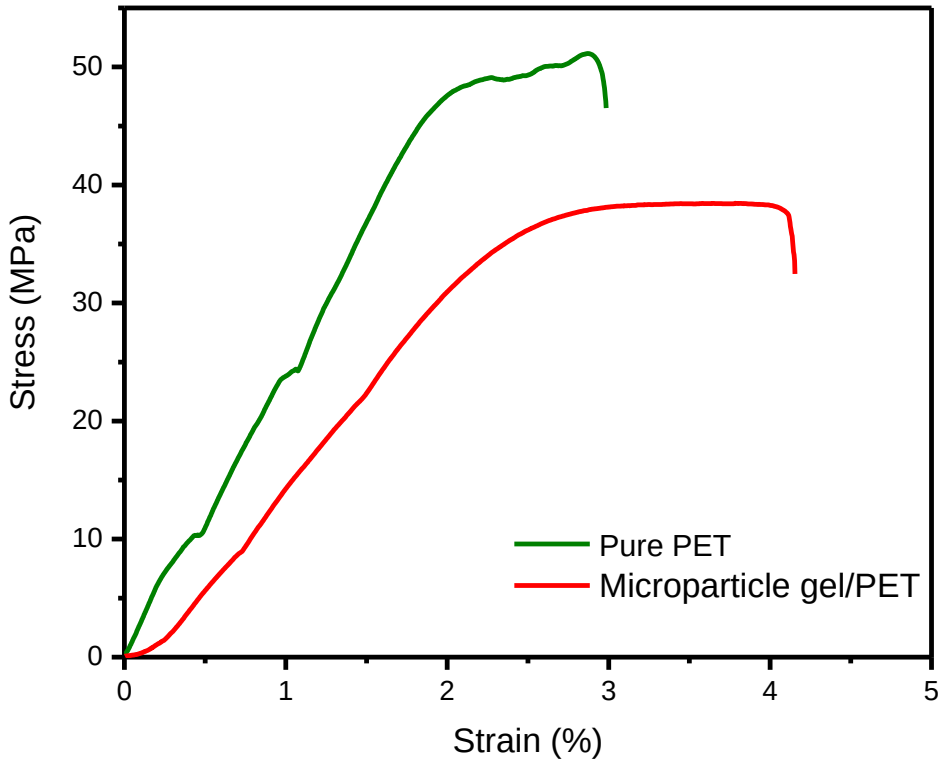

Figure S20. Stress-strain curves of pure PET and microparticle gel/PET membranes.

3. Supplementary Table

Table S1. Stripping factor of microparticle gel double-layered membranes prepared with different substrate membranes.

Substrates	Salt concentration/%		
	0	0.9	25
PET	0.53	0.002	0
PP	0.28	0.05	0