

Electronic Supplementary Information

Confined encapsulation of living cells in self-assembly fiber macrospheres with micro/nanoporous polymer shell for transformation of contaminants to green energy

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Experimental

Materials and chemicals

Glass fiber was purchased from Hangzhou hi tech composite material Co., Ltd. (China). PVDF (average Mw 95000Da of 1010) and PSf pellets (Mw 120000Da of P-3500) were obtained from Solvay (USA). N, N-Dimethylformamide, sodium alginate, calcium chloride, sodium chloride and sodium hydroxide were bought from Sinopharm chemical Reagent Co. Ltd. (China). All chemicals were of analytical grade without further purification. The experiment solutions were prepared from deionized water prepared by a self-made RO-EDI system, in which ion concentration was analyzed by IRIS Intrepid ICP and Metrohm 861Compact IC and controlled to meet $\sigma \leq 0.5 \mu\text{S cm}^{-1}$.

Analytical methods

Standard analytical methods were used to determine COD and ammonia-nitrogen ($\text{NH}_3\text{-N}$). Total organic carbon (TOC) was measured by a TOC-V analyzer (Shimadzu, Japan). The light intensity was examined by using the digital lux meter (AS831). Hydrogen production was analyzed by gas chromatography (GC-2010, China), equipped with the thermal conductivity detector.

Table S1. Synthetic high-salinity organic wastewater characterization.

| Composition | Values |
|--|-----------|
| COD (mg/L) | 3056-3945 |
| NaCl (g/L) | 40.0-80.0 |
| pH | 5.0-9.0 |
| SS (mg/L) | 100-150 |
| NH ₃ -N (mg/L) | 42.5-48.7 |
| NaHCO ₃ (g/L) | 0.1 |
| CaCl ₂ (g/L) | 0.1 |
| MgSO ₄ ·7H ₂ O (g/L) | 0.1 |

Table S2. Comparison of encapsulated cell within different host matrix for various applications.

| Microorganism | Immobilizing matrix | Application | Performance | Ref. |
|--|---|------------------------------------|--|------|
| <i>Pseudomonas putida</i> | structural membrane constructed particle | phenol degradation | similar biodegradation rates were observed in encapsulated and free cell ($C_{\text{phenol}} = 800 \text{ g/L}$) | S1 |
| heterotrophic acteria | structural membrane constructed particle | olive mill wastewater treatment | biodegradation rate between encapsulated and free cells was 1:2 | S2 |
| <i>Klebsiella oxytoca</i> | Alginate | KCN (cyanide) degradation | removal efficiency of free cell (90 %) and encapsulated cell (91 %) | S3 |
| Ammonia-oxidizing bacteria | calcium alginate | ammonia degradation | around 1.8 times reduction of activity after encapsulation in the initial stage (6 days) | S4 |
| Rhodococcus erythropolis LSSE8-1 | Fe ₃ O ₄ nanoparticles (NPs) | dibenzothiophene (DBT) degradation | encapsulated and free cells exhibited similar desulfurizing activities | S5 |
| Strain XA05 and FG03 | polyvinyl alcohol (PVA) | phenol degradation | encapsulated cells showed lower degradation rate than free cells (C_{phenol} below 500 mg/l) | S6 |
| Chlorophyta <i>Dunaliella tertiolecta</i> | alginate-SiO ₂ -polycation@alginate-SiO ₂ | CO ₂ remediation | activity of cells inside a hybrid matrix is consistently higher than for those encapsulated within alginate beads | 25 |
| NIH-3T3 cells and human mesenchymal stem cells | CNT-gelatin methacrylate (GelMA) hybrid | tissue engineering | enhancer for mechanical properties while maintaining beneficial bioactive properties of GelMA material | S7 |

| | | | | |
|--|--|------------------------------------|--|-----------|
| <i>Botryococcus braunii</i> and <i>Chlorella vulgaris</i> | low-sodium silica gels | CO ₂ remediation | decrease in activity with time due to stresses exerted on cells isolated from natural environment or from oxygen saturation within gel | 17 |
| THP-1 cells | different types of alginate microsphere | cell culture | type of alginate affected cell proliferation, but there was no significant effect on viability of encapsulated cells | S8 |
| Photosynthetic bacteria cells | self-assembly fiber microspheres with porous polymer shell | high-salinity wastewater treatment | encapsulated cells exhibited 1.3 times higher than free cells | This work |

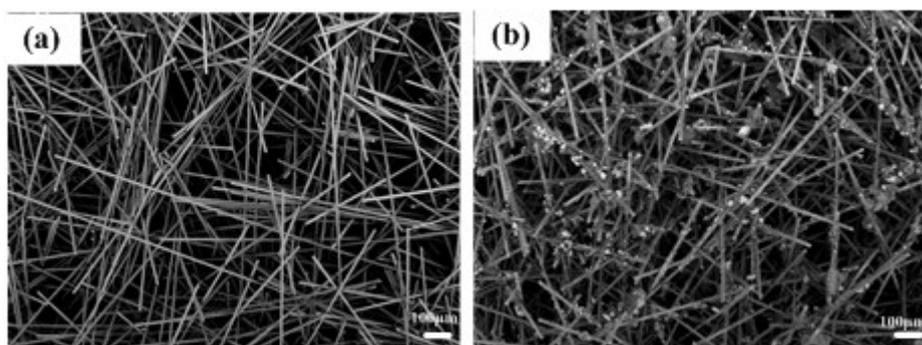


Figure S1. SEM images of interior morphology for sample MGFS@Polymer and PSB/MGFS@Polymer.

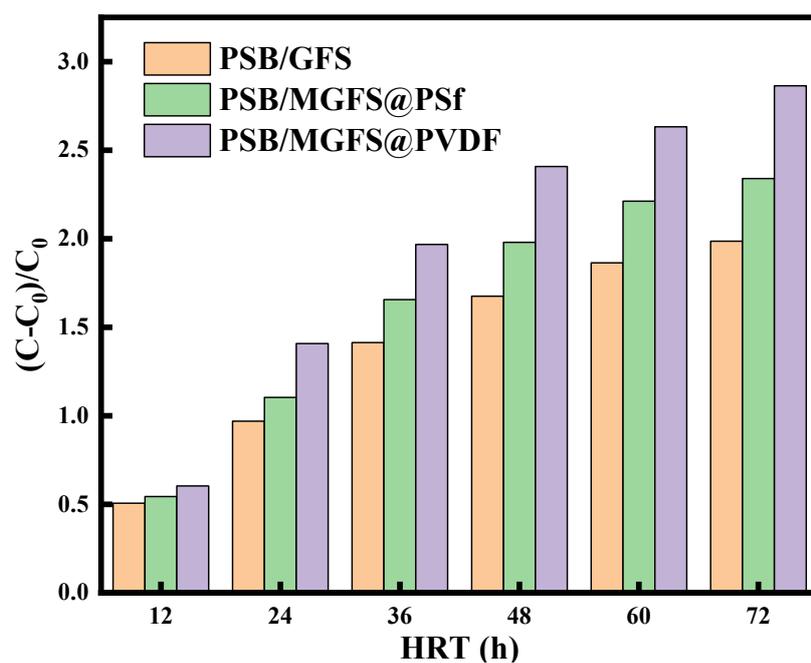


Figure S2 Encapsulation yields of PSB cells into GFS, MGFS@PSf, and MGFS@PVDF based on the initial concentration with different HRT.

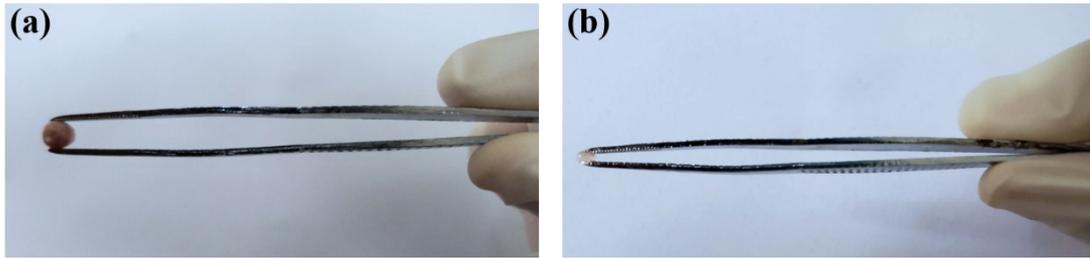


Figure S3. Comparison of mechanical strength between the polymeric coated GFS macrospheres (a) and alginate beads (b). Compared with the traditional sodium alginate entrapped beads, the compression tests showed that the prepared MGFS@Polyme macrocapsules maintained the original structure even if under the strong squeezing.

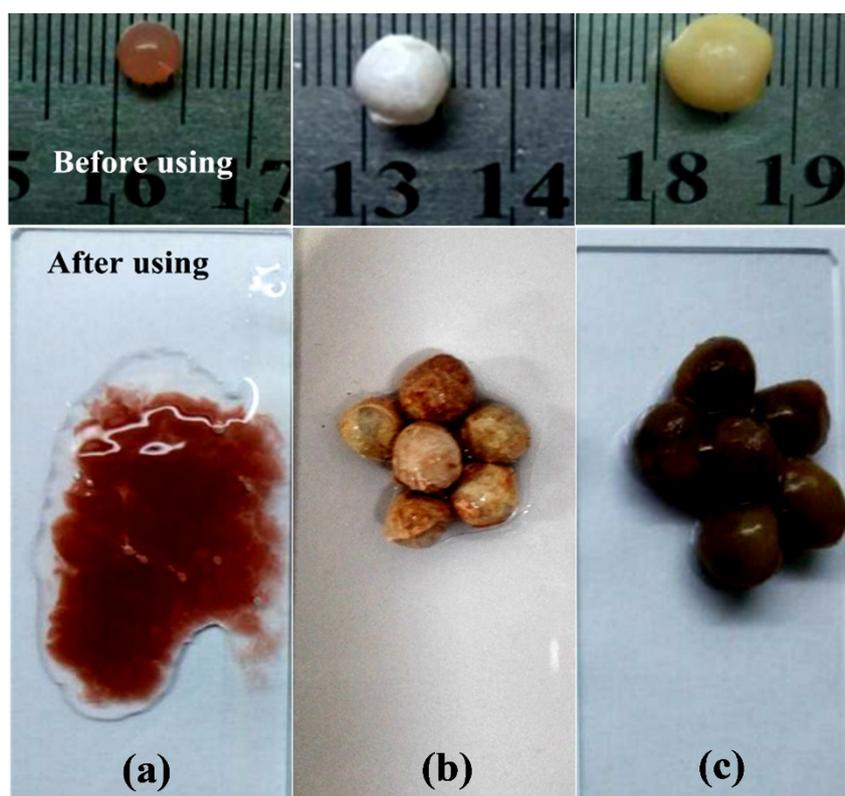


Figure S4. Photograph of the sodium alginate entrapped beads (a), PSB/MGFS@PSf (b) and PSB/MGFS@PVDF (c) before and after operation. After the high-salinity wastewater treatment, it was found that the sodium alginate (SA) entrapped beads was broken and became colloidal liquid, while all the core-shell structured macrospheres contained a large number of PSB and still kept their spherical forms.

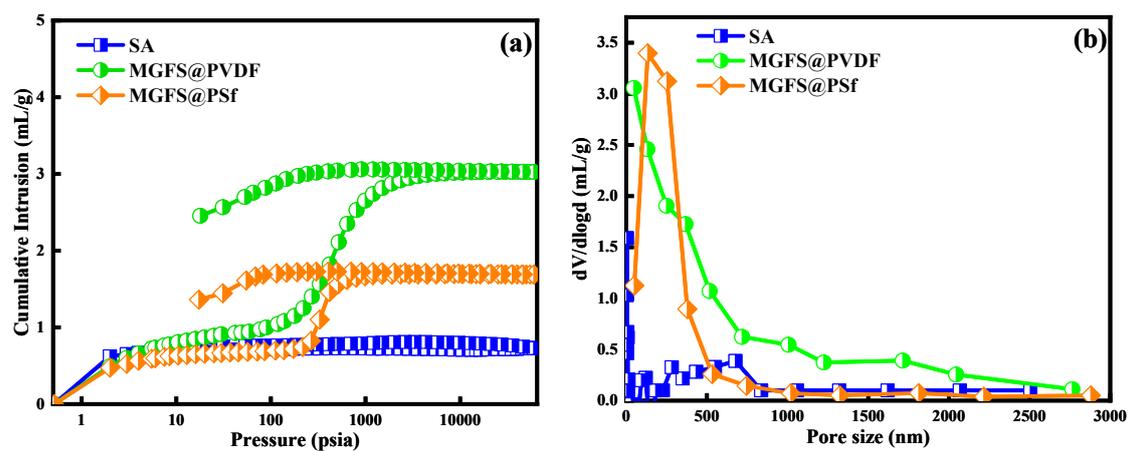


Figure S5. Cumulative intrusion/extrusion vs pressure curve (a) and pore size distribution (b) for different samples.

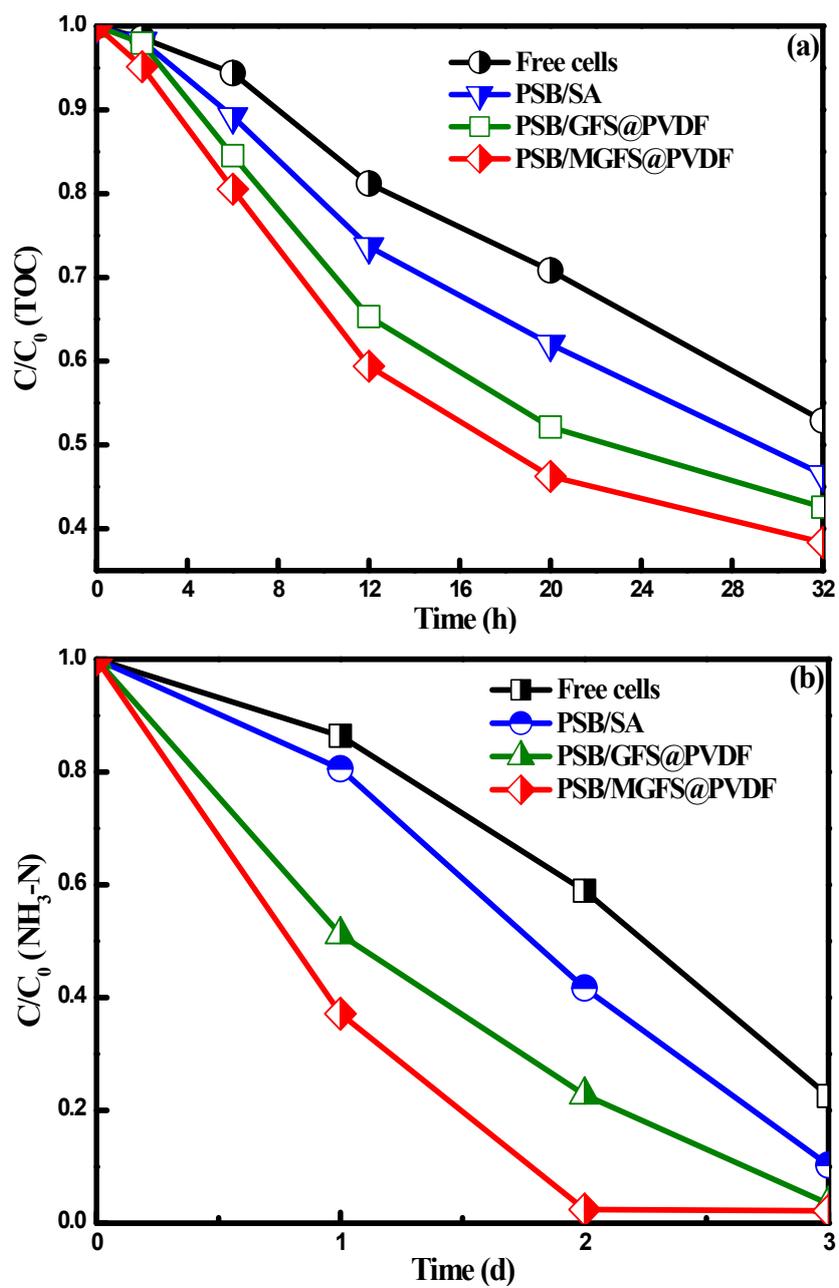


Figure S6. TOC (a) and $\text{NH}_3\text{-N}$ (b) removal efficiency of the different operation systems.

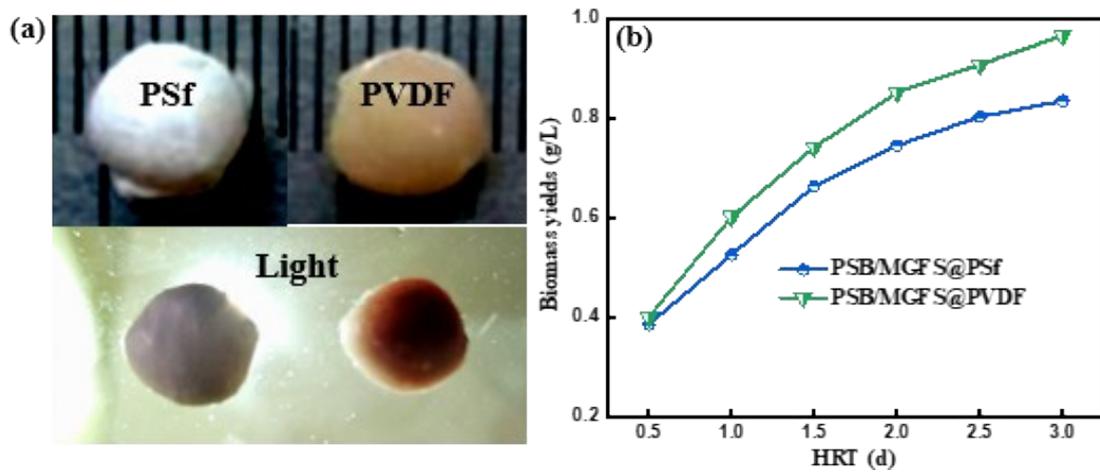


Figure S7. (a) Photograph of PSB/MGFS@PSf and PSB/MGFS@PVDF; (b) Comparison of biomass harvesting between MGFS@PSf and MGFS@PVDF with different HRT.

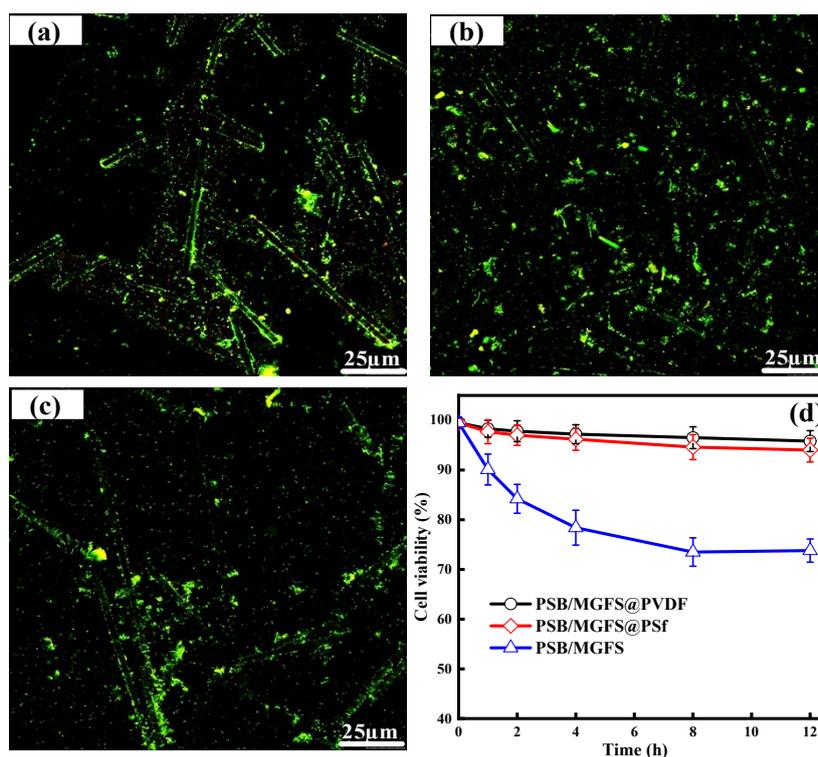


Figure S8. (a-c) Live/dead staining of encapsulated PSB cells in MGFS, MGFS@PSf, and MGFS@PVDF and (d) Viability of encapsulated cells after incubation for different time.

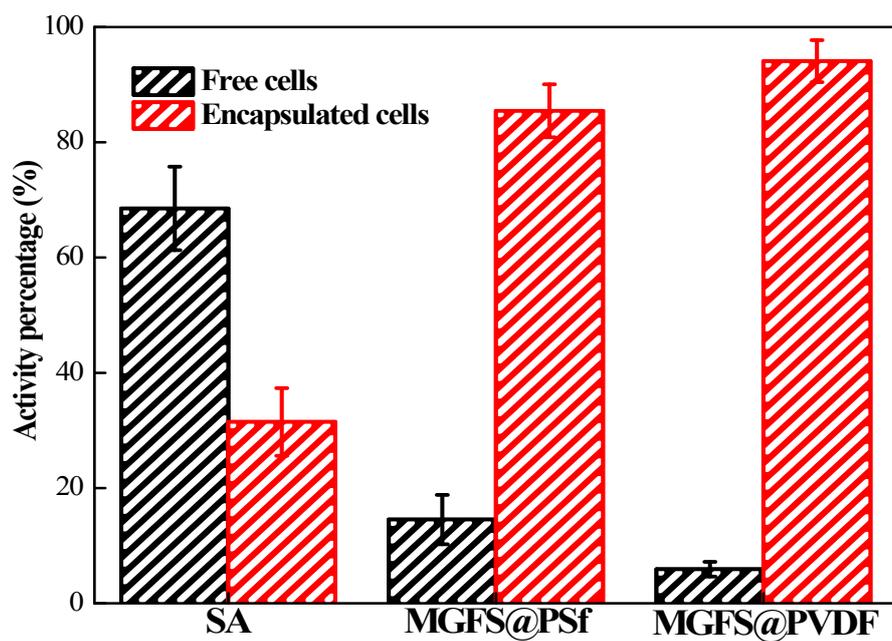


Figure S9. Activity percentage of leached and encapsulated cells into different colloids.

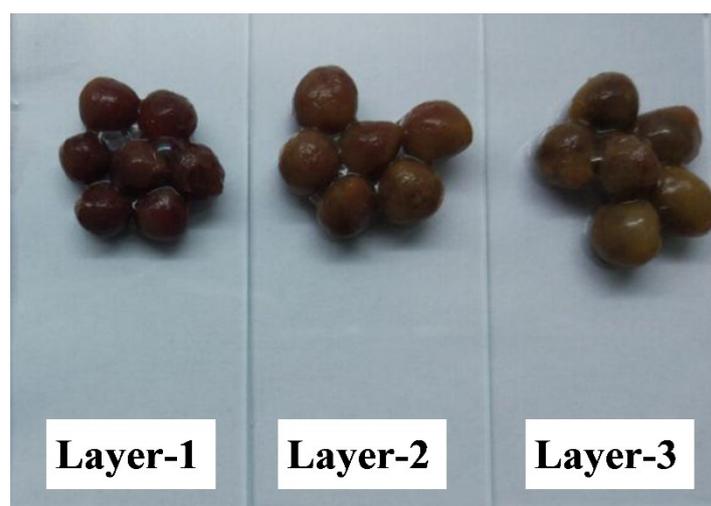


Figure S10. Photograph of the PSB/MGFS@PVDF microspheres coated with different layers after the long-term operation.

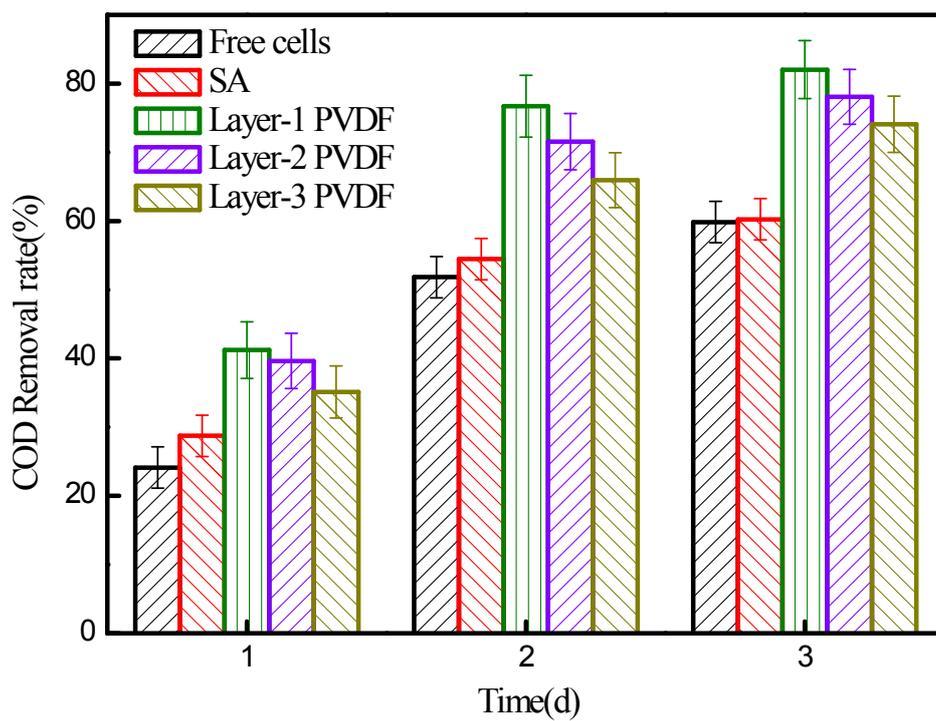


Figure S11. Performance of the prepared PSB/MGFS@PVDF microspheres coated with different layers on the COD removal.

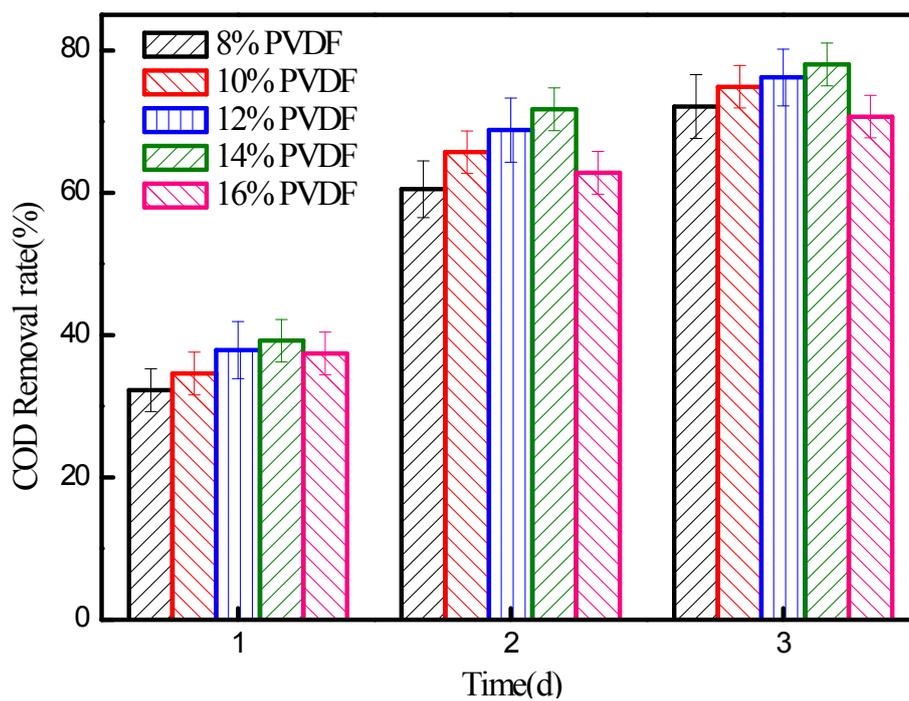


Figure S12. Performance of the prepared PSB/MGFS@PVDF macrospheres coated with different polymer concentration on the COD removal.

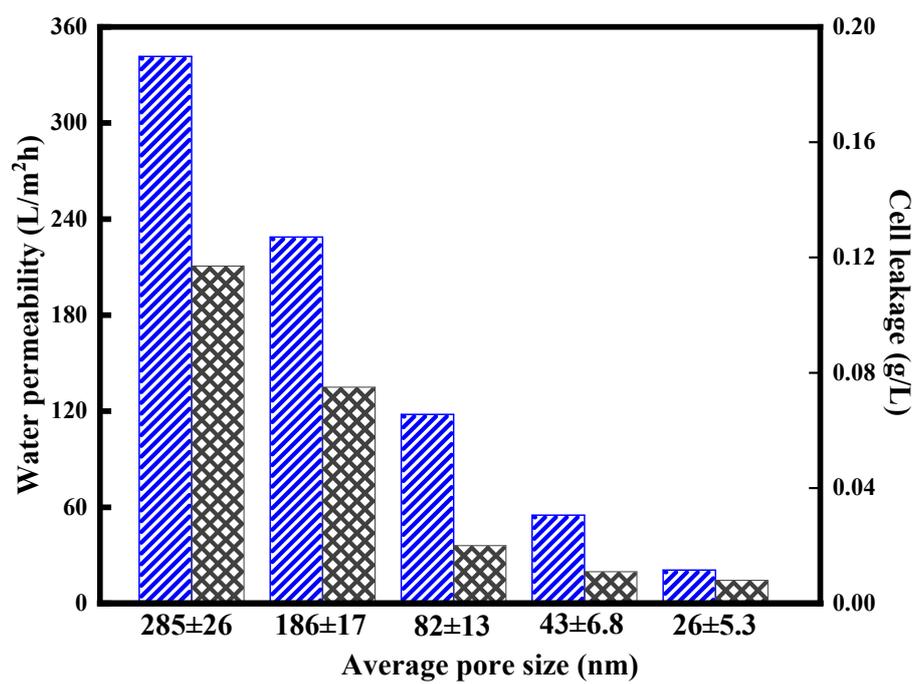


Figure S13. Variation on water permeability and living cells leakage of porous polymeric shells with different sizes.

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