Electronic Supplementary Information

Bioinspired interconnected hydrogel capsules for enhanced catalysis

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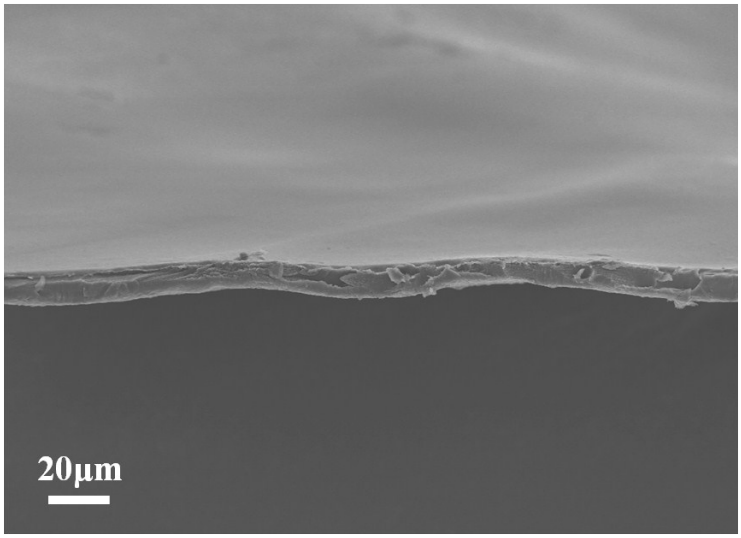

Fig. S1 SEM photograph of the CS-CMCS membrane cross-section.

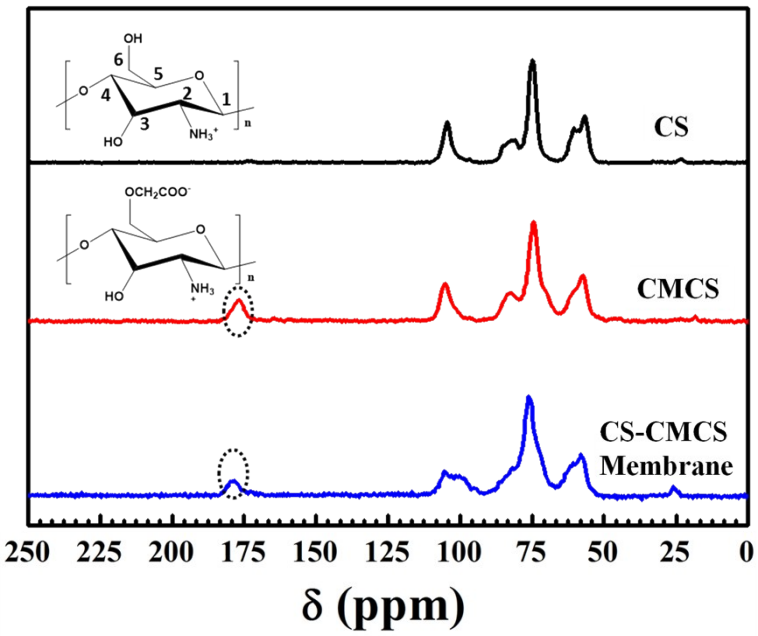

Fig. S2 Solid-state ^{13}C NMR spectra of CS, CMCS and CS-CMCS membrane.

The solid-state ^{13}C NMR spectra of CS, CMCS and CS-CMCS membrane were shown in **Fig. S2** (The frozen-dried CS and CMCS from their aqueous solutions were used as control groups.). The spectrum of CS showed five major peaks with chemical shift values at 56.6, 60.4, 74.8, 81.8 and 104.3 ppm, which were attributed to C-2, C-6, C-3/C-5, C-4 and C-1, respectively.[S1] While the spectrum of CMCS was similar to CS except the additional signal of COO^- at 176.9 ppm and the reduced signal of C-6. The spectrum of CS-CMCS retained all the carbon peaks presenting in CS as well as COO^- signal of CMCS at 179.1 ppm. These results indicated that both CS and CMCS were present in the CS-CMCS membrane of capsules.

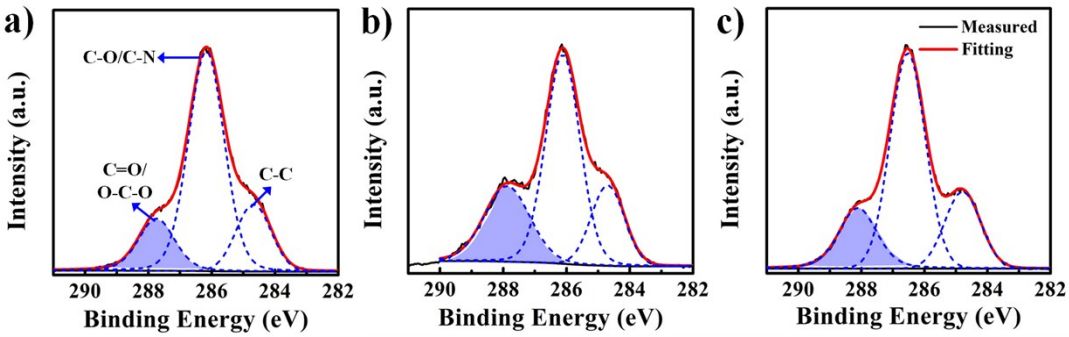

Fig. S3 XPS $C1_S$ narrow scanned spectra of a) CS, b) CMCS and c) CS-CMCS membrane.

XPS $C1_S$ spectra were recorded as shown in **Fig. S3** and the corresponding binding energies were listed in **Table S1**. Based on the XPS spectrum of the CS-CMCS membrane, the deconvolution of the overlapped C 1s signals included three peaks centered at 284.79, 286.53 and 288.11 eV, which were ascribed to the C-C, C-O/C-N, and C=O/O-C-O groups, respectively.[S2] In addition, the C=O/O-C-O peak area value of CS-CMCS membrane was about 20 %, which

suggested that the carbon content of C=O/O-C-O in CS-CMCS PEM was fell in between CS (16.5 %) and CMCS (26.3 %). As a result, the CS-CMCS membrane was assuredly composed of CS and CMCS.

Table S1. Assignments of $C1_S$ spectral bands based on binding energies (BE), and peak area (PA) of surface species.

BE (eV) {(PA (%)}

Element	CS	CMCS	CS-CMCS PEM	Assignments[S2]
$C1_S$	284.68{18.4}	284.70{19.4}	284.79{21.1}	C-C
$C1_S$	286.17{65.1}	286.12{54.3}	286.53{58.9}	C-O or C-N
$C1_S$	287.74{16.5}	287.87{26.3}	288.11{20.0}	C=O/O-C-O

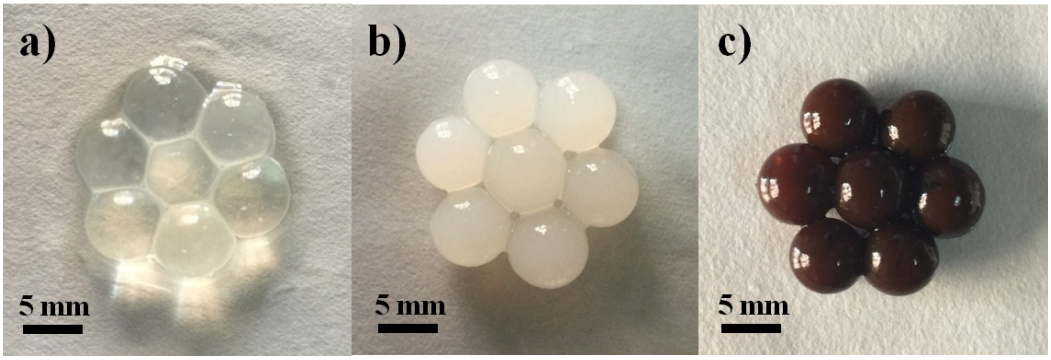

Fig. S4 Photographs of a) hollow capsules, b) hydrogel capsules with interconnected inner membranes and c) Ag NPs loaded hydrogel capsules with interconnected inner membranes.

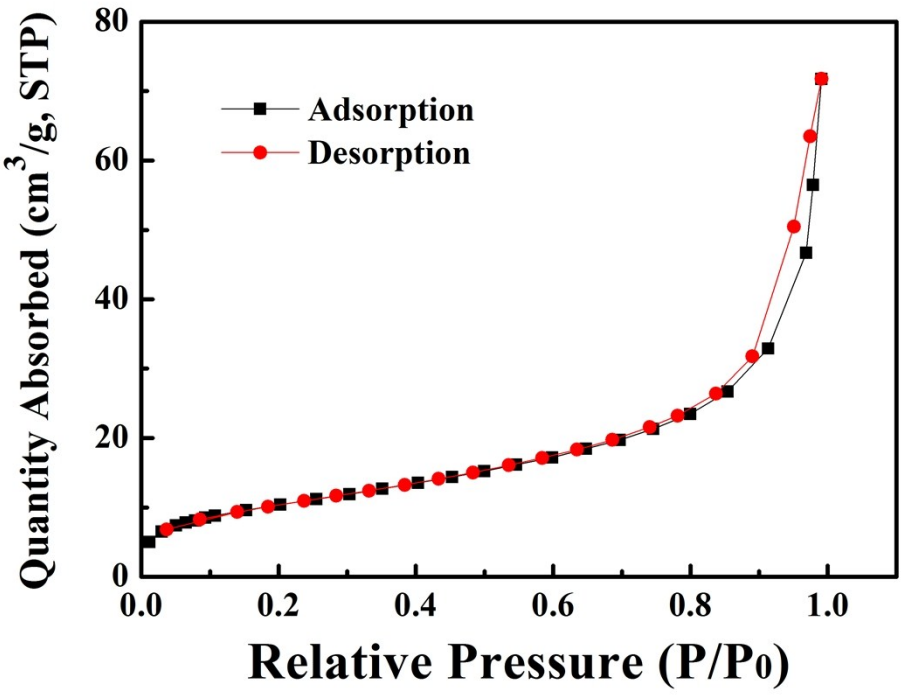

Fig. S5 N_2 adsorption-desorption isotherms of hydrogel capsules with interconnected inner membranes.

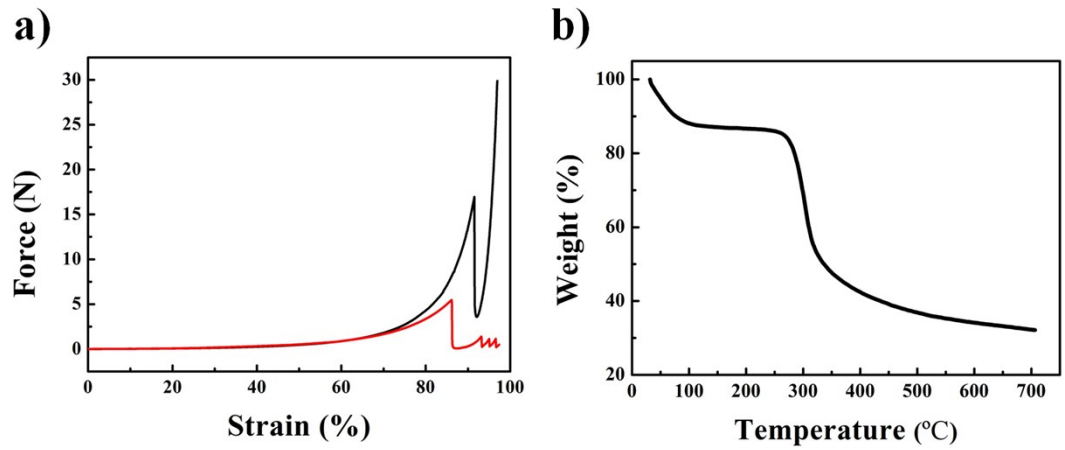

Fig. S6 Physical properties of interconnected hydrogel capsules. a) Force-Strain curves of interconnected hydrogel capsule (black line) and hollow capsule (red line). b) TGA curves of interconnected hydrogel capsules.

The compressive Force-Strain curves of interconnected hydrogel capsule and hollow capsule (d≈6 mm) were shown in **Fig. S6a**. With being compressed by fixture, the pressure of interconnected hydrogel capsule was presented to increase gradually and damaged at the force of 16.95 N, which exhibited favorable mechanical properties, while the CS-CMCS hollow capsule presented damaging compressive force of 5.47 N. Therefore, the existent of internal gel membrane

in interconnected hydrogel capsule produced much better mechanical properties compared with CS-CMCS hollow capsule without this structure. On the other hand, the interconnected hydrogel capsule showed considerable thermal performance with the thermal decomposition temperature about 300 °C. (**Fig. S6b**)

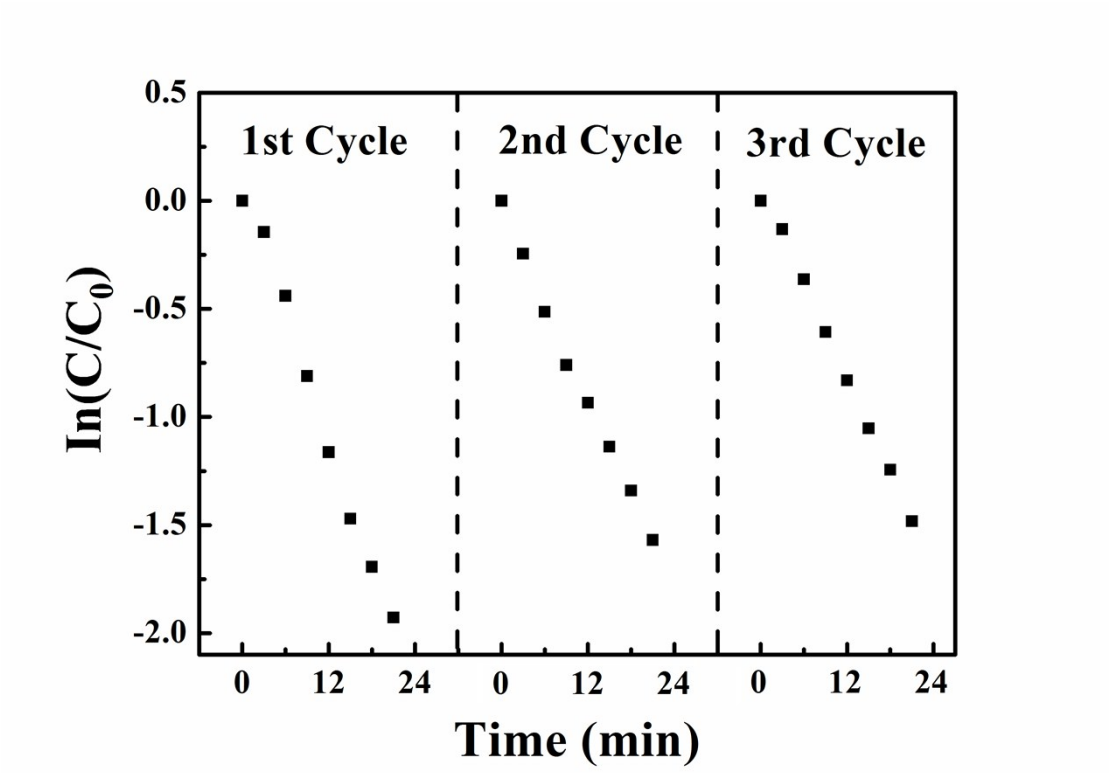

Fig. S7 The recyclability of Ag NPs loaded interconnected hydrogel capsules in representative plots of $\ln(C/C_0)$ against reaction time for the reduction of 4-NP by sodium borohydride.

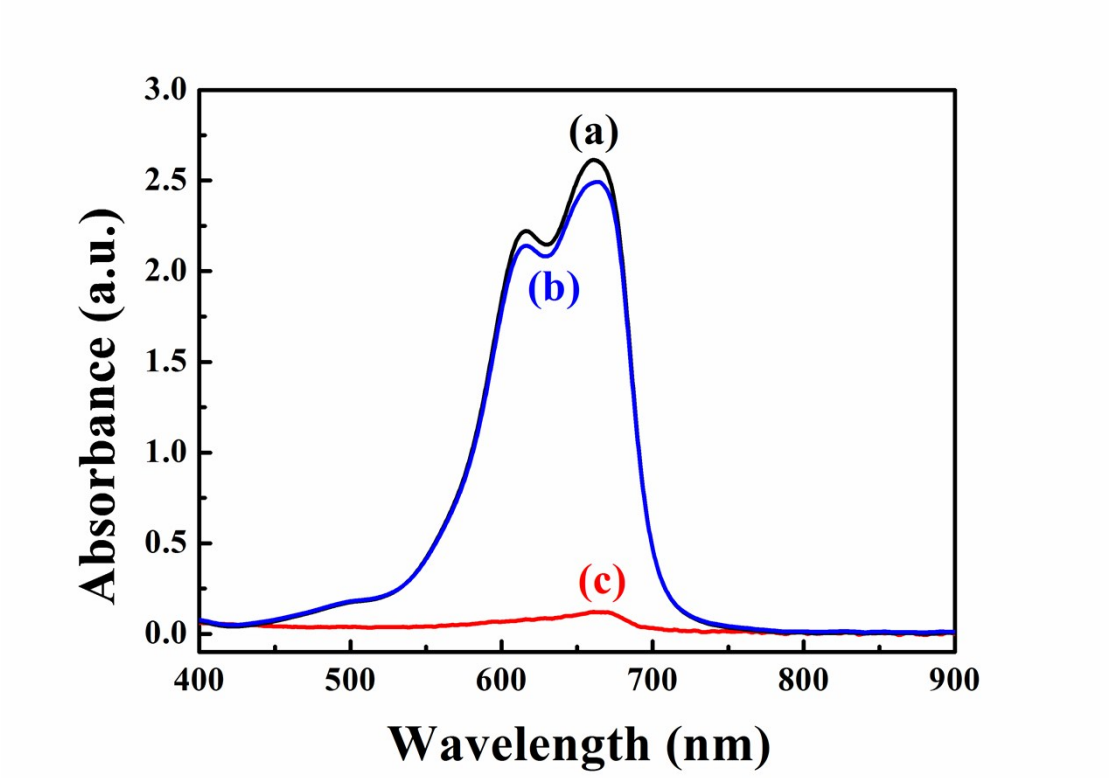

Fig. S8 UV-VIS spectra of (a) freshly prepared mixture of MB dye and $NaBH_4$ aqueous solution, (b) the same solution stored for 5 min, and (c) the same solution stored for 5 min in the presence of Ag NPs loaded interconnected hydrogel capsules.

Supplementary Reference:

S1 M. F. Cervera, J. Heinämäki, M. Räsänen, S. L. Maunu, M. Karjalainen, O. M. N. Acosta, A. I. Colarte, J. Yliruusi, *Carbohyd. Polym.*, 2005, **58**, 401.

S2 L. Dambies, C. Guimon, S. Yiacoumi, E. Guibal, *Colloid Surf. A-Physicochem. Eng. Asp.*, 2001, **177**, 203.