Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Strengthened, Recyclable Shape Memory Rubber Films with Rigid Filler Nano-Capillary Network

Wenchao Wu, Chuanhui Xu*, Zhongjie Zheng, Baofeng Lin, Lihua Fu

Guangxi Key Laboratory of Petrochemical Resource Processing and Process Intensification Technology, School of Chemistry and Chemical Engineering, Guangxi University, Nanning 530004, China

Corresponding Author: Chuanhui Xu xuhuiyee@gxu.edu.cn

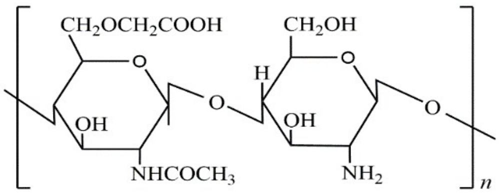

Figure S1. Chemical structure of 6-O-CM-chitosan (CMCS)

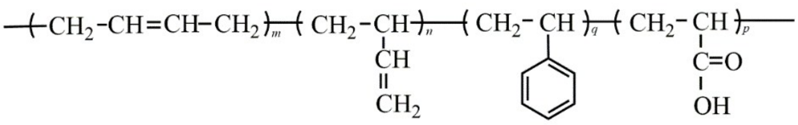

Figure S2. Chemical structure of carboxylated styrene butadiene rubber (XSBR)

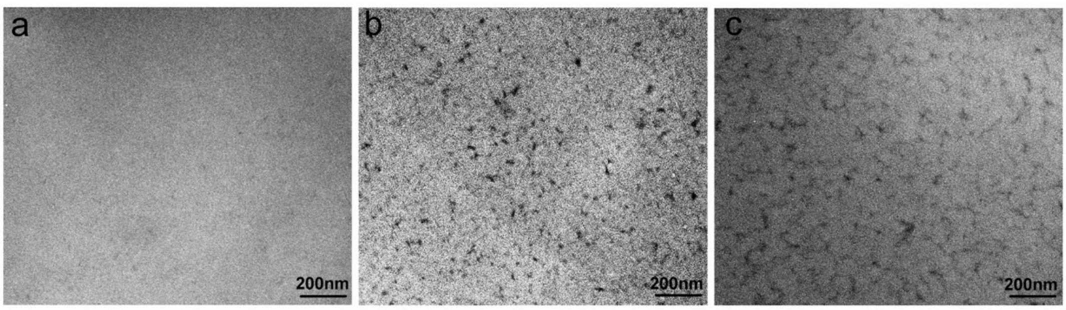

Figure S3.TEM images of (a) neat XSBR, (b) XSBR/CMCS film with 5 wt% CMCS and (c) XSBR/CMCS film with 10 wt% CMCS

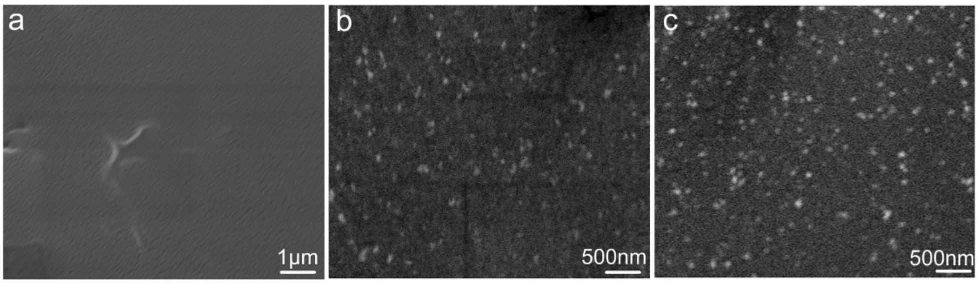

Figure S4. SEM images of (a) neat XSBR, (b) XSBR/CMCS film with 5 wt% CMCS and (c) XSBR/CMCS film with 10 wt% CMCS

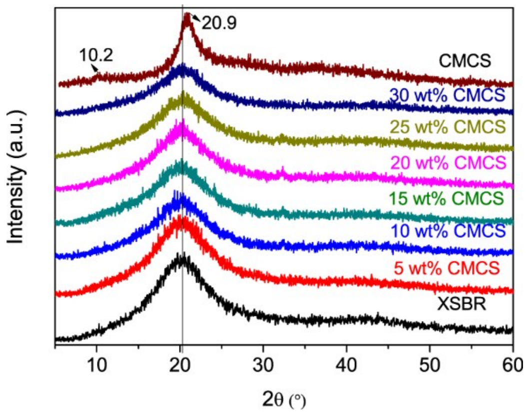

Figure S5. XRD of CMCS and XSBR/CMCS films. Disappearance of the crystallization peak of CMCS at $2\theta = 10.2^o$ and 20.9^o [1] in the films suggested that the orderly arrangement of chains in regenerated CMCS was disorganized by entangling/interaction between XSBR and CMCS chains.

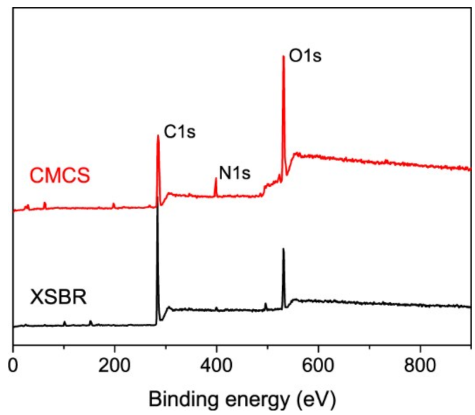

Figure S6. Low-resolution XPS spectra of CMCS and XSBR used in our films

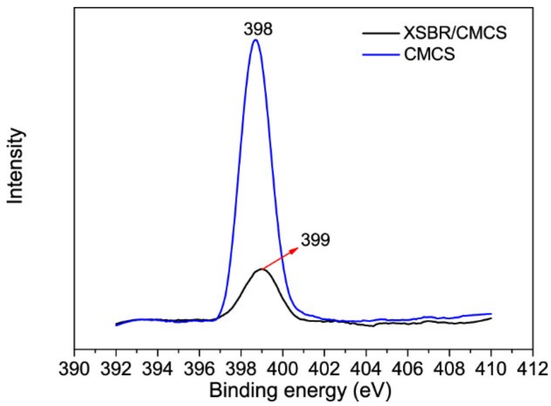

Figure S7. High-resolution N1s spectra of CMCS and XSBR/CMCS film (20 wt%)

Figure S8. Photograph of the CMCS powder

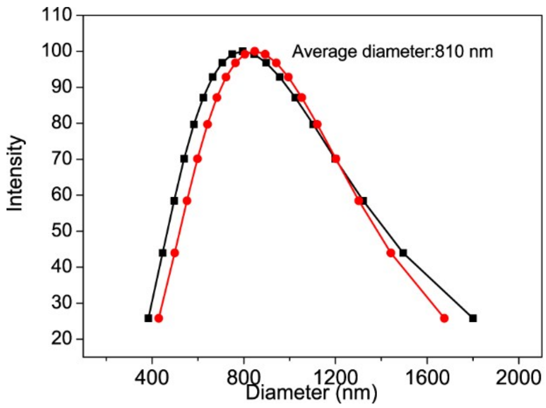

Figure S9. Particle size of the CMCS tested for twice.

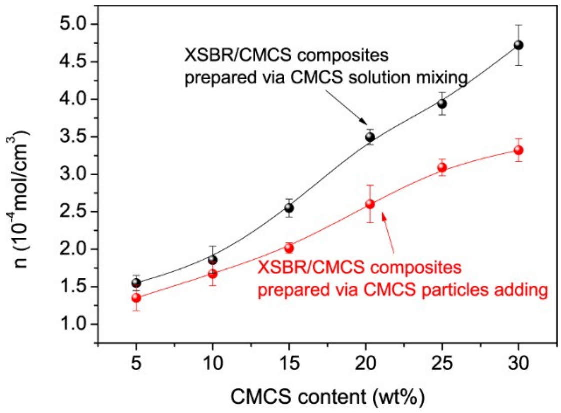

Figure S10. Crosslink density "n" of the XSBR/CMCS composites prepared through two methods: the film with rigid CMCS capillary network shows a higher crosslink density than the one with CMCS particles.

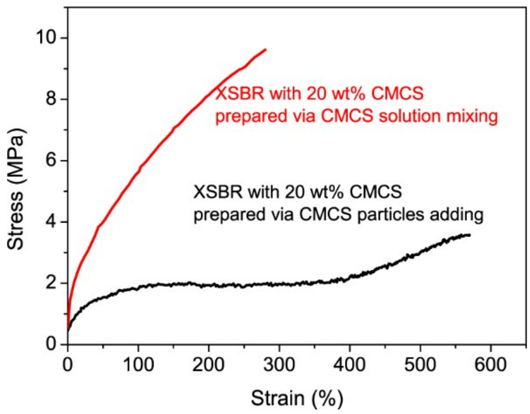

Figure S11. Typical stress-strain curve of XSBR/CMCS composites prepared through two methods: the film with rigid CMCS capillary network shows a significant better strength than the one with CMCS particles.

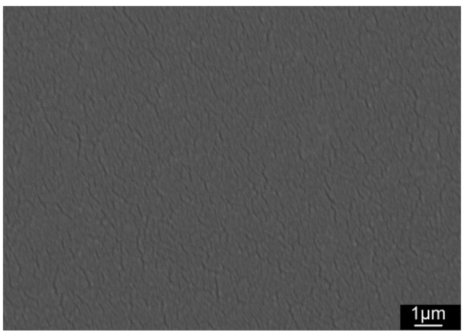

Figure S12. SEM image of the tensile fracture surface of XSBR

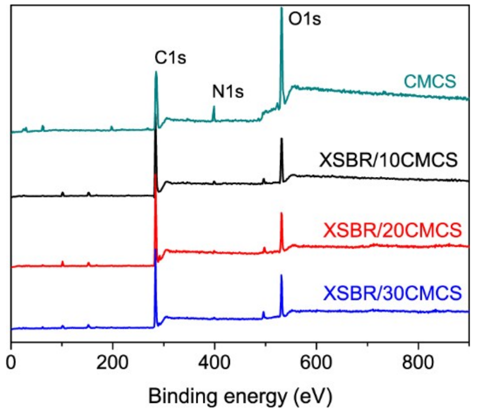

Figure S13. Low-resolution XPS spectra of the recycled XSBR

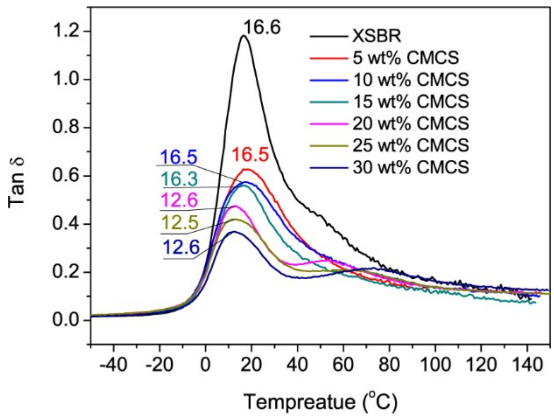

Figure S14. Tan δ curves of XSBR/CMCS films

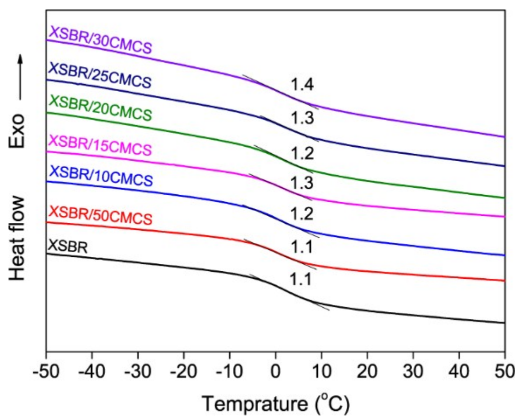

Figure S15. DSC curves of XSBR/CMCS films

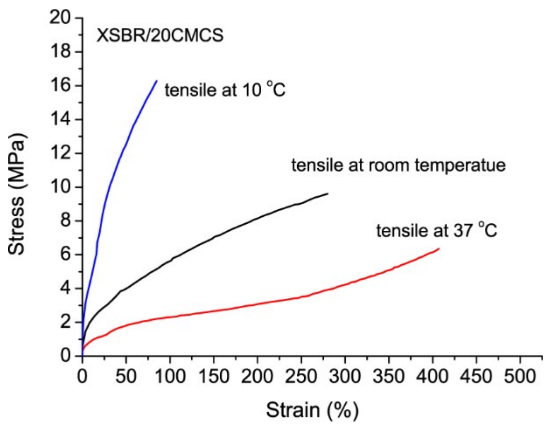

Figure S16. The stress-strain curves of the XSBR/CMCS film (20 wt% CMCS) programmed at 10 °C and 37 °C

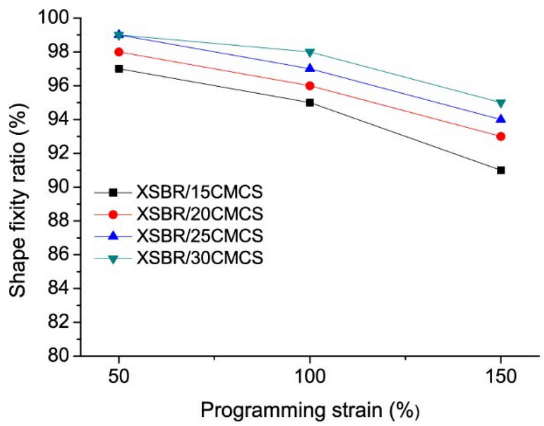

Figure S17. Changes of SF of the XSBR/CMCS films at three programming strains (50%, 100% and 150%) after 24h at 10 °C

Table S1 Detail residual strains for every tensile cycle

samples	1st cycle at ε=50%	2nd cycle at ε=100%	3rd cycle at ε=150%
XSBR	11%	24%	40%
Film with 5 wt% CMCS	7%	10%	13%
Film with 10 wt% CMCS	23%	48%	70%
Film with 15 wt% CMCS	27%	56%	83%
Film with 20 wt% CMCS	29%	58%	87%
Film with 25 wt% CMCS	30%	64%	98%
Film with 30 wt% CMCS	30%	63%	-

Movie S1. The film with 30 wt% CMC had a 100% SF in spiral shape at the maximum temperature of 13.8 °C. This temperature is more comfortable to human body.

Movie S2. Shape fixing of rectangular strip of XSBR/CMCS film (20 wt% CMC): the marked 1 cm was elongated the to 2 cm at room temperature (~23°C) and then immersed in water (10 °C) under loading. About 8s later, unloading, the elongated rectangular strip showed excellent 100% shape fixing.

Movie S3. Shape recovery of rectangular strip of XSBR/CMCS film (20 wt% CMC): the fixed rectangular strip was immersed in water (40 °C), about 10s later, the elongated rectangular strip showed full shape recovery.

Reference

1. L.H. Fan, Y.M. Du, B.Z. Zhang, J.H. Yang, J.P. Zhou, J. F. Kennedy. *Carbohydr. Polym.* 2006, **65**, 447.