

## Supporting Information

# Fabrication of robust multilayer films by triggering the coupling reaction between phenol groups and primary amine groups with visible light irradiation

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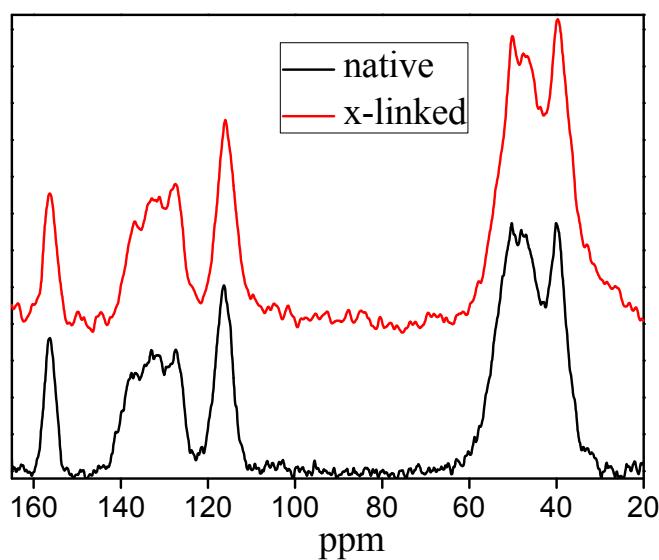
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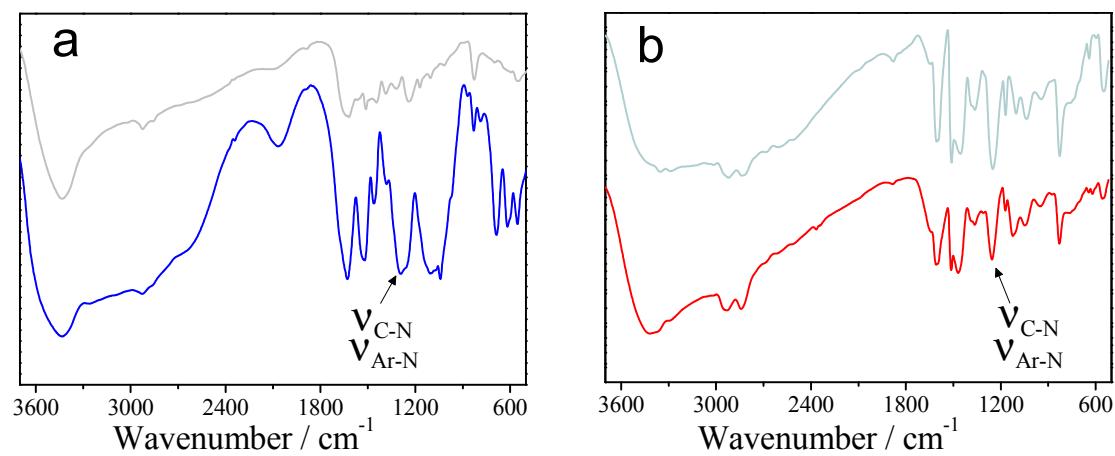
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### Preparation of the bulk hydrogels by HRP-H<sub>2</sub>O<sub>2</sub> method

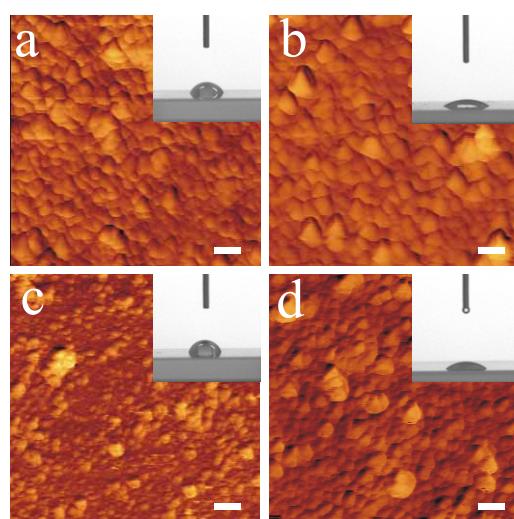
In this study, the alginate with the tyramine residuals was used as the phenol derivatives to prepare bulk hydrogels. The modified alginate was prepared according the literature and the grafting ratio is about 7%.<sup>1</sup> When adding the H<sub>2</sub>O<sub>2</sub> solutions to the homogenous PBS solution of alginate and HRP (1.5 wt.% and 5.6 units/mL, respectively) to a final concentration of 0.32 M, the coupling reaction was promptly taken place and the bulk hydrogels can be obtained in one minute. This result indicates that the x-linking reaction can be carried out in high concentrations of HRP and H<sub>2</sub>O<sub>2</sub>. Importantly, the inactivity of HRP from high ratio of [H<sub>2</sub>O<sub>2</sub>]/[HRP] was not observed in such condition.<sup>2,3</sup>



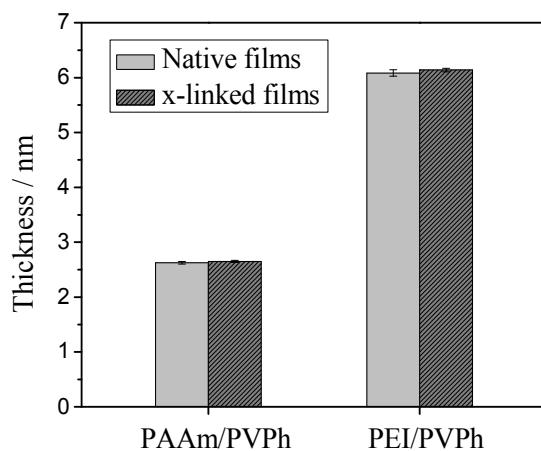
**Fig. S1** The solid-state  $^{13}\text{C}$ -NMR spectra of the native and x-linked PEI/PVPh complexes.



**Fig. S2** The full FT-IR spectra of the native (upper) and x-linked (lower) complexes of PAAm/PVPh (a) and PEI/PVPh (b).



**Fig. S3** The AFM height images of the native and x-linked films of (PAAm/PVPh)<sub>5</sub> (a, b) and (PEI/PVPh)<sub>5</sub> (c, d). AFM was operated in tapping mode using Si cantilevers with a applied force of ~5 nN, scan rate of 0.6 Hz and tapping frequency of 273 kHz. Insets: the images showing static contact angles of the corresponding films.



**Fig. S4** The average bilayer thickness of native and x-linked multilayer films of PAAm/PVPh and PEI/PVPh (n=3).

## References

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2. J. H. Ndez-ruiz, M. B. Arnao, A. N. Hiner, F. G. Canovas and M. Acosta, *Biochem. J.*, 2001, **354**, 107-114.
3. M. Kurisawa, J. E. Chung, Y. Y. Yang, S. J. Gao and H. Uyama, *Chem. Commun.*, 2005, 4312-4314.