Electronic Supplementary Material

Regulating the micro-nano structure of cellulose nanofibers reinforced polyvinyl alcohol composites for enhanced mechanical and barrier properties via one-pot wet milling

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Table S1 Comparison of mechanical properties of PC_{-15} composite film and PVA/CNFs(10wt%) films in other researches

PC composite films	Tensile strength [MPa]	Elongation at break [%]	Young's modulus [MPa]	References
PC_{-15}	49.3	125.1	1337.12	
PVA/CNFs (10wt%)	44.2	134.8	476.6	(Liu et al. 2013)
	44.3	89.2	1473.86	(Miao et al. 2016)
	14.89	/	517	(Schoeler et al. 2020)
	53.5	25.2	1033	(Qiu and Netravali. 2012)

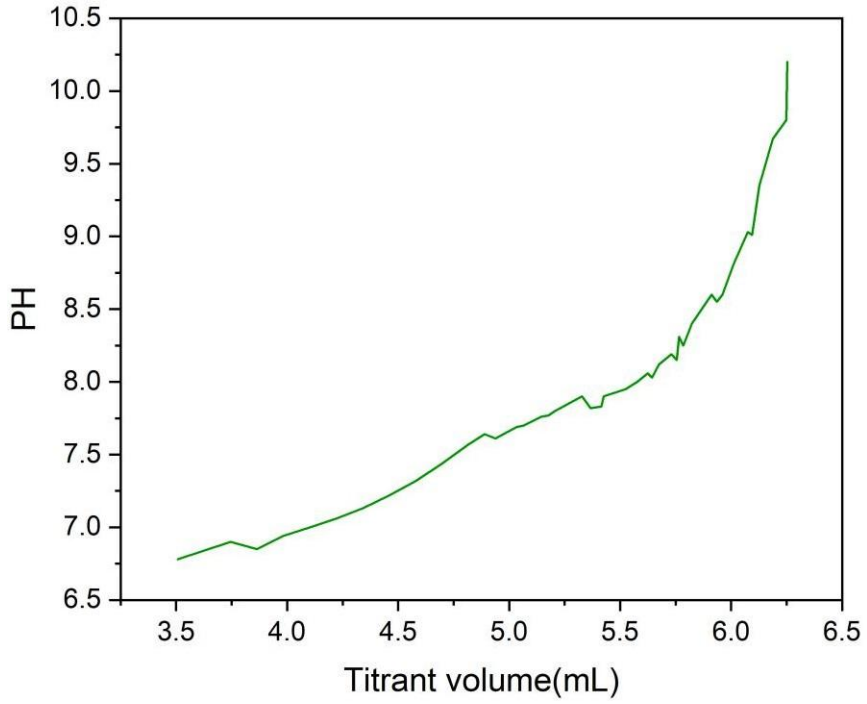

Fig.S1 The pH titration curve of the carboxylate content measurement to determine endpoint for PH

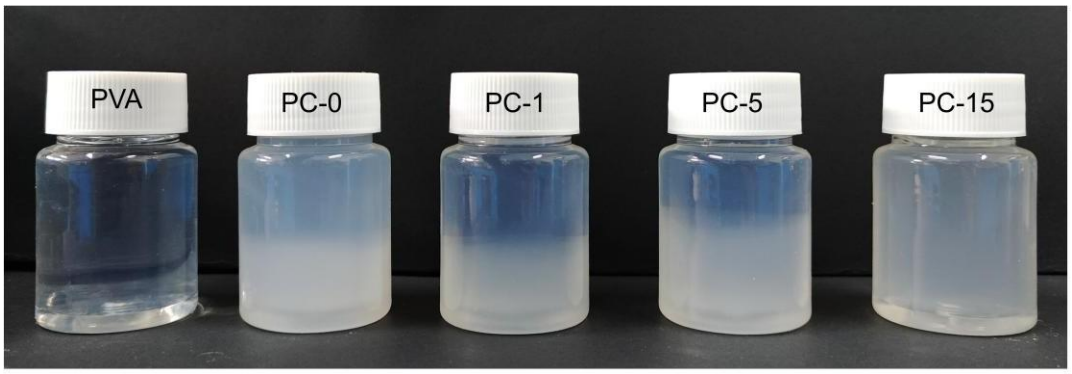

Fig.S2 PVA aqueous solution and PC series suspension after being sat still for several days

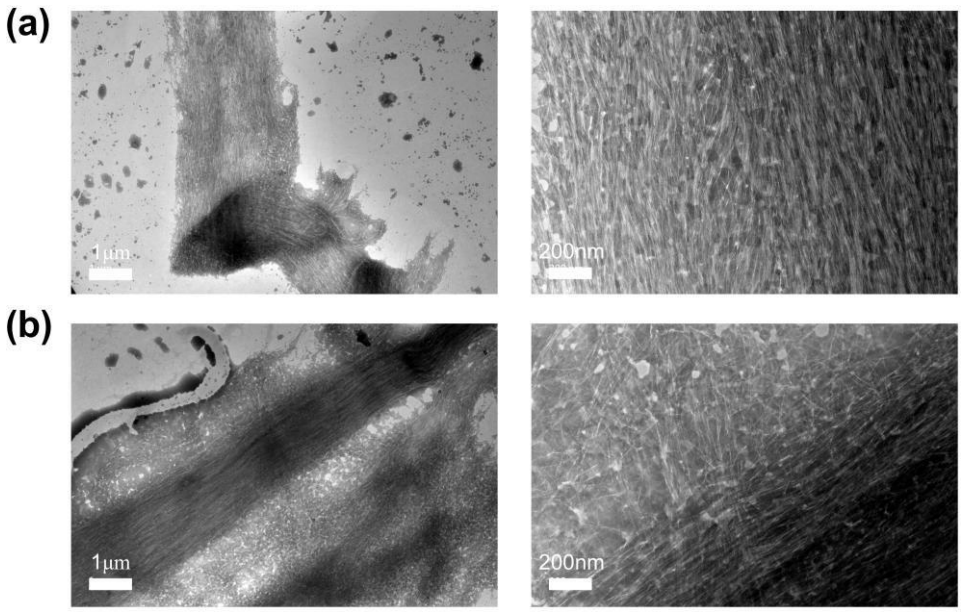

Fig.S3 TEM images of (a) PC_{-1} and (b) PC_{-5} composites in different scale: the dark areas represent penetrated dye, while the overexposed areas may indicate a continuous and impenetrable phase.

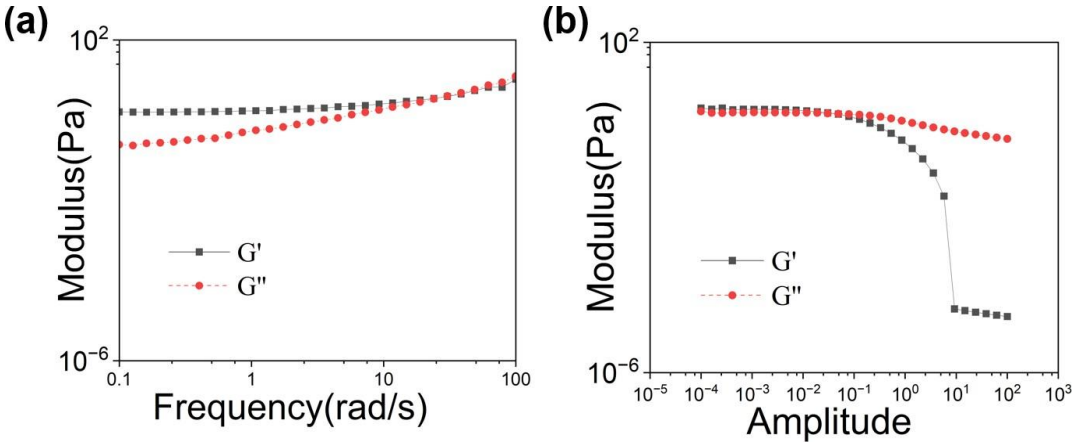

Fig.S4 Rheological behavior of PC_{-5} suspension: (a) G´ and G´´ as a function of frequency. (b) G′and G″as a function of amplitude

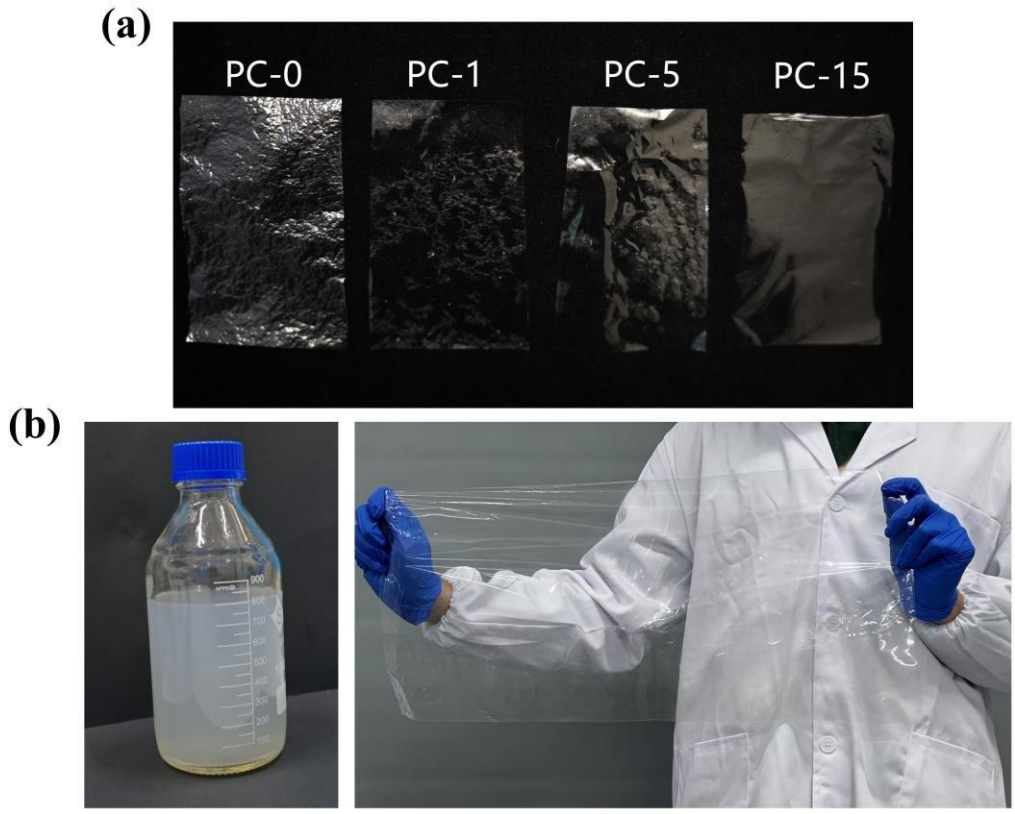

Fig.S5 (a) photographs of PC_{-0}, PC_{-1}, PC_{-5} and PC_{-15} films. (b) grinding PC_{-15} slurries in a substantial volumes and casting in a 70×30cm mold to produce a large-sized PC-15 film

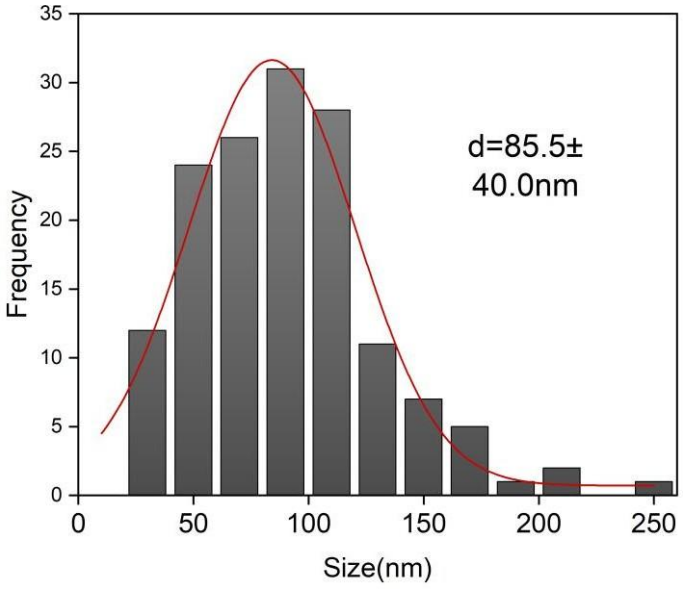

Fig.S6 Size distribution of CNFs discontinuous phase in the cross-section SEM image of PC_{-15} film

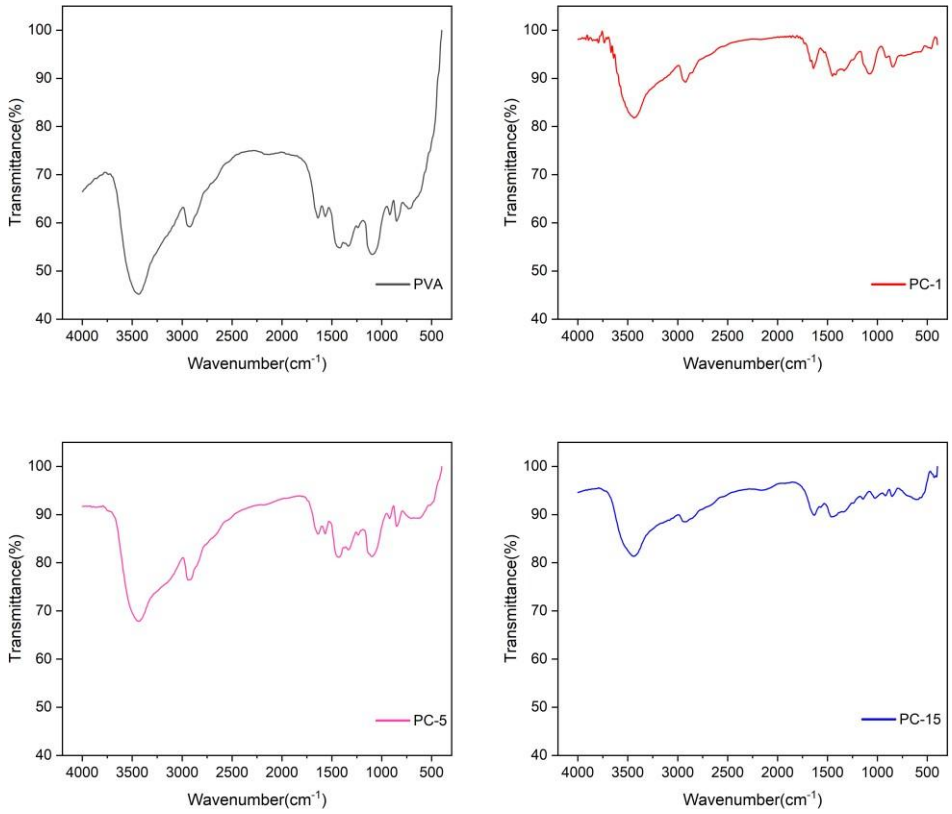

Fig.S7 The original FTIR spectra of PVA, PC_{-1}, PC_{-5} and PC_{-15} composite films

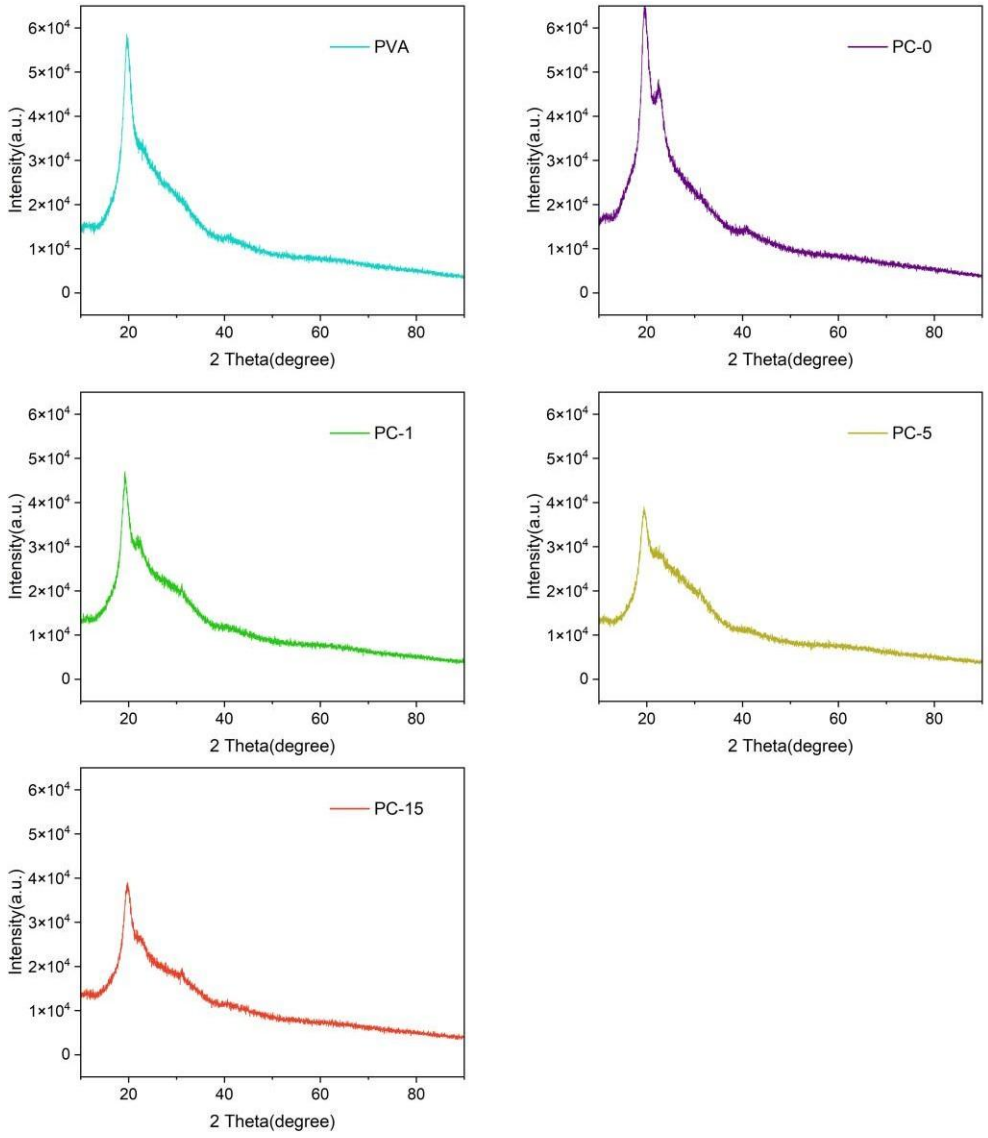

Fig.S8 The original XRD patterns of PVA and PC films without background correction and peak deconvolution

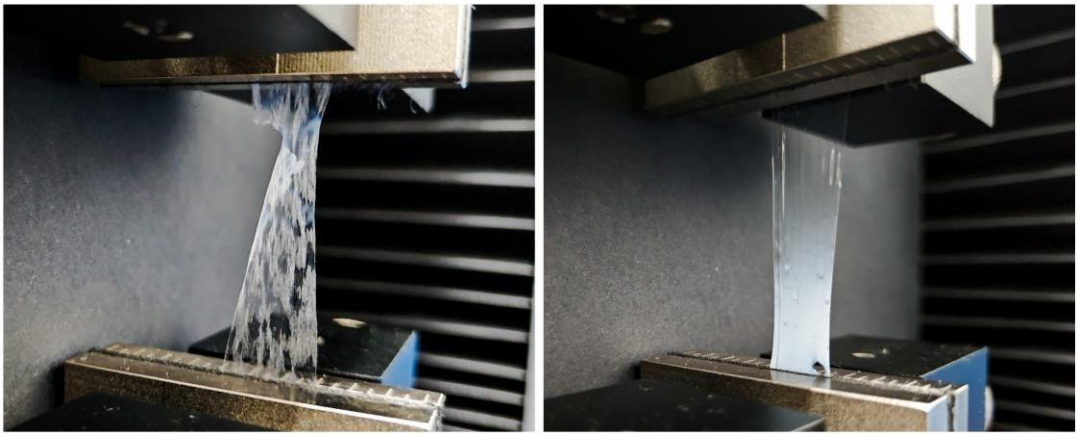

Fig.S9 Photographs of PC_{-0} and PC_{-15} films during tensile strength test manifesting their respective breaking mechanisms

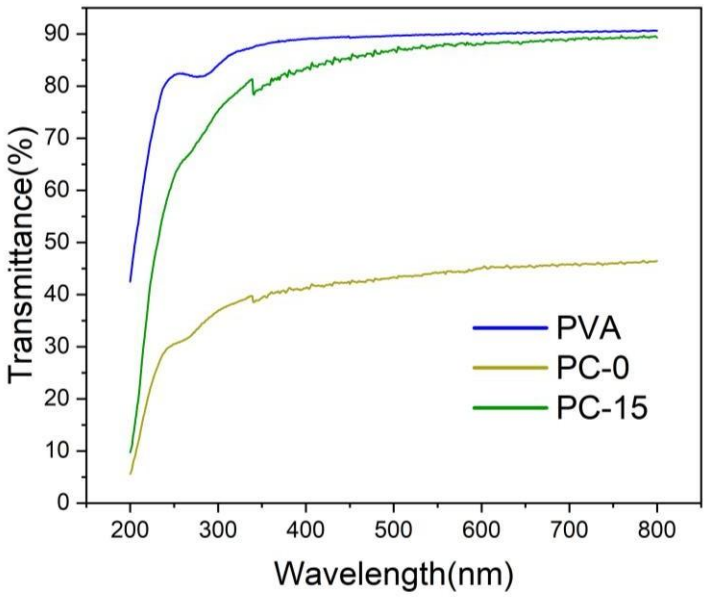

Fig.S10 UV-vis spectra of PVA, PC_{-0} and PC_{-15} films

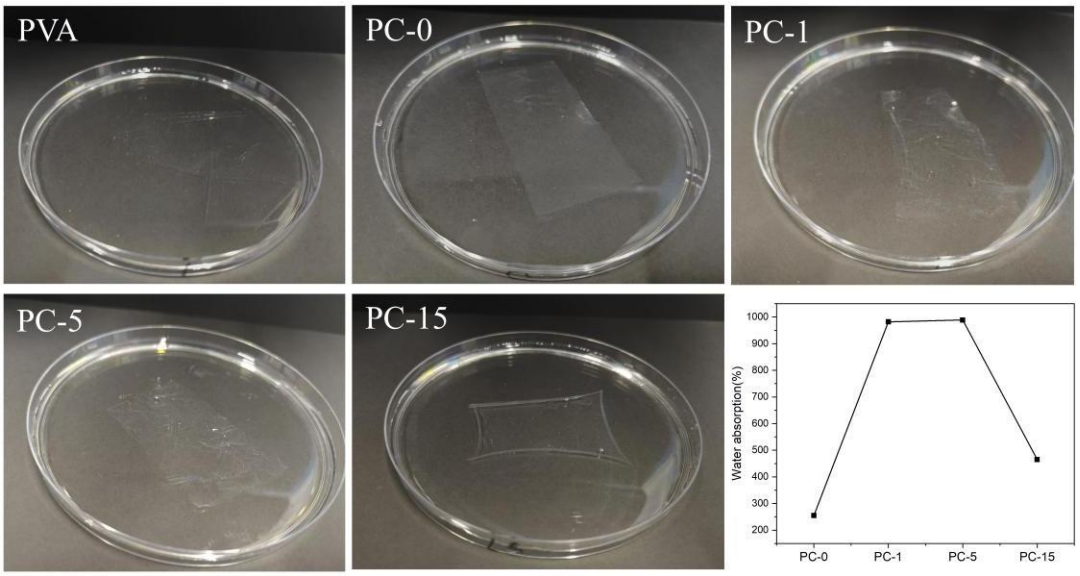

Fig.S11 Shape retention and water absorption of PVA and PC films under 2 hours of water swelling

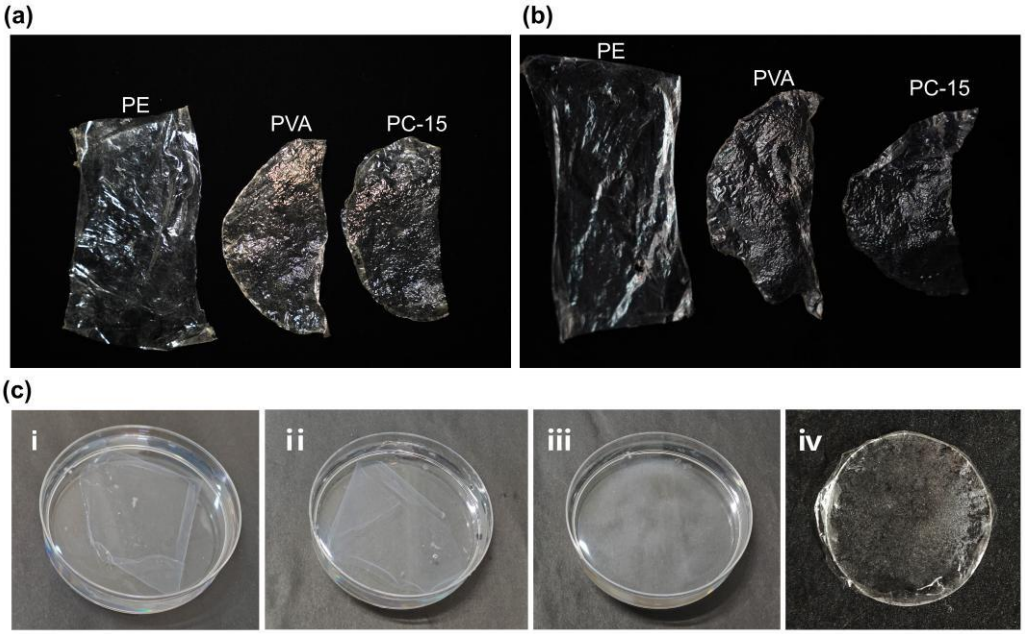

Fig.S12 Polyethylene (PE), PVA and PC-15 films after (a) 30 days and(b) 60 days of landfilling. (c) PC-15 films after (i) 2 hours and (ii) 24 hours of soaking in water under ambient condition, and (iii) after 6 hours in a 90 ℃ water bath