Supplementary Information

**Preparation, properties and mechanism of anionic and cationic CNCs/WPU composite films**

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**Fig. s1** XRD patterns of three kinds of CNCs: TOCNCs, CaCNCs, and SCNCs.

Fig. s1 shows the XRD patterns of three kinds of CNCs: TOCNCs, CaCNCs, and SCNCs. These three CNCs all appeared diffraction peaks at 15.0 o, 16.8 o and 22.9 o. This verified these three CNCs all reserved the cellulose I crystallinity. The small diffraction peak at 10.4 o may come from a few impurities or test deviation of equipment.