## **Supporting Information**

The Improvement of Shape Memory Function of Poly(ε-caprolactone)/ Nano-Crystalline Cellulose Nanocomposite via the recrystallization under a high-pressure environment

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Figure S1. (A) FTIR spectra and (B) XRD curves of MCC and NCC.



Figure S2. (A) DLS and (B) TEM of NCC.



Figure S3. (A) TGA and (B) DTG of MCC and NCC.



Figure S4. (A) TGA and (B) DTG of PCL and PCL-NCC nanocomposites with different NCC contents reprocessed at 100 kPa.



**Figure S5.** POM images of PCL and PCL-NCC nanocomposites with different NCC contents reprocessed at 100 kPa, (A) PCL; (B) PCL-NCC-3; (C) PCL-NCC-6; (D)PCL-NCC-9.



**Figure S6.** POM images of PCL-NCC-3 nanocomposites reprocessed at 200 MPa, different holding temperatures for 1 h, (A) 120°C; (B) 140°C; (C) 160°C.



**Figure S7.** Gel fractions of PCL-NCC-3 nanocomposites reprocessed at different holding temperatures, 200 MPa for 1 h.



**Figure S8.** Gel fractions of PCL-NCC nanocomposites with different NCC contents reprocessed at 500 MPa at 140°C for 1 h.



Figure S9. POM images of PCL-NCC-6 nanocomposites reprocessed at

different pressures, 140°C for 1 h, (A) 100 kPa; (B)200 MPa; (C) 500 MPa.



**Figure S10.** The characterizations of PCL reprocessed at 100 kPa, 200 MPa, 350 MPa and 500 MPa, 140°C for 1 h, (A) XRD curves; (B) the magnified parts of XRD curves at 21°-24°; (C) the degrees of crystallinity; (D) DSC curves.



**Figure S11.** Gel fractions of PCL-NCC-6 nanocomposites and PCL reprocessed at different pressures, 140°C for 1 h.



**Figure S12.** (A) Digital photos showing the shape memory processes of PCL samples reprocessed at 100 kPa, 200 MPa, 350 MPa and 500 MPa, 140°C for 1 h at 55°C; (B) shape memory properties of these PCL samples reprocessed at different pressures.



**Figure S13.** (A) XRD curves and (B) the degrees of crystallinity of PCL-NCC-6 nanocomposite reprocessed at 100 kPa, 140°C for 1 h between the whole and the crosslinked part;(C) XRD curves and (D) the degrees of crystallinity of PCL-NCC-6 nanocomposite reprocessed at 200MPa, 140°C for 1 h; (E) XRD curves and (F) the degrees of crystallinity of PCL-NCC-6 nanocomposite reprocessed at 500 MPa, 140°C for 1 h.