Supplementary Information

Synthesis of 4-ethoxy-9-Allyl-1, 8-Naphthalimide (EANI)

In a three-neck 250-mL round bottom flask, 4-bromo-1,8-naphthalic anhydride (5.00 g, 18 mmol) and allylamine (1.03 g, 18 mmol) in 80 mL ethanol were mixed and refluxed for 4 h. After cooling to room temperature, 100 mL ice water was poured into the mixture. 5.20 g yellowish-brown solid (4-bromo-N-allylnaphthalimide) was isolated by filtering and then vacuum dried, m.p. 129-131°C. 4-Bromo-N-allylnaphthalimide (1.60 g, 5 mmol) and KOH (0.30 g, 5.05 mmol) were refluxed in 50 mL ethanol for 4 h. 50 mL ice water was poured into the mixture and the precipitates were isolated by filtering. 1.25 g yellow solid (4-ethoxy-9-allyl-1, 8-naphthalimide) was obtained after recrystallization in ethanol, m.p. 126-128 °C.^{1}H NMR (CDCl$_3$, 400 MHz) δ: 1.60-1.65(t, 3H), 4.32-4.39 (q, 2H), 4.79-4.82(d, 2H), 5.18-5.23(d, 1H), 5.28-5.35(d, 1H), 5.95-6.08(m, 1H), 7.02-7.05(d, 1H), 7.68-7.74(t, 1H), 8.55-8.63(m, 3H). FTIR (cm^{-1}) v: 2983, 2891 (—CH$_3$), 1695(C=O), 1651(C=C).
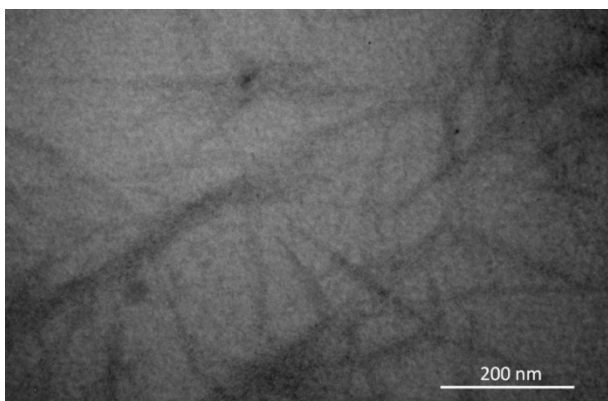
TEM

200 nm

Figure S1. TEM image of unmodified CNCs in DMF

Figure S1 showed the TEM images of CNC suspension in DMF. Independent and need-like morphology of CNCs was observed, indicating that CNCs dispersed well in DMF. Good dispersion of CNCs in DMF was propitious to improve the accessibility of surface OH groups of CNCs and ensure the homogeneous surface immobilization of initiator.
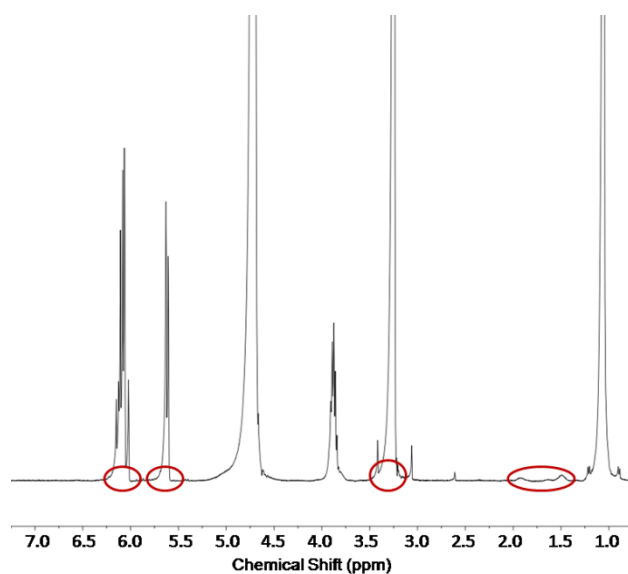
¹H NMR

Figure S2. Typical ¹H NMR spectra of the supernatant after SI-AGET-ATRP

The residual signal at about 6.1 and 5.6 ppm associated with double bond was used to calculate the monomer conversion by comparing to the signal at 3.3 ppm of CH_3OH. The mass of produced free polymers was analogously obtained from the appearing signal at 1.4 and 2.0 ppm that was associated with free polymers. The calculations were based on the assumption that the amount of CH_3OH remained unchanged before and after polymerization.
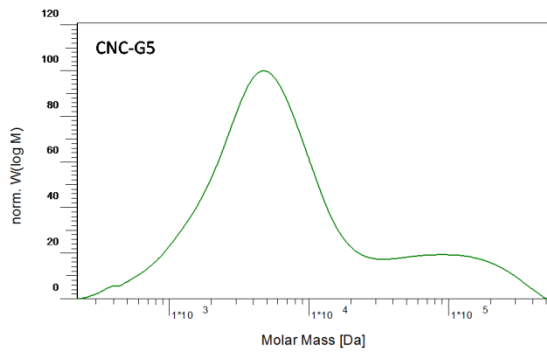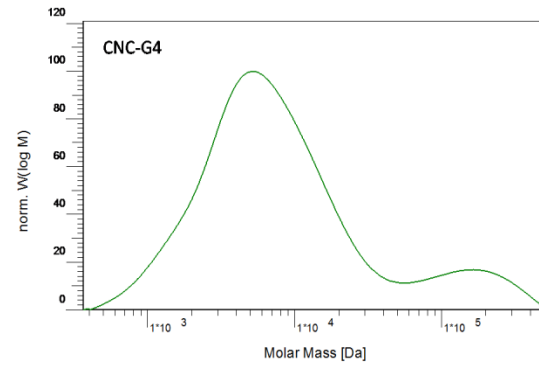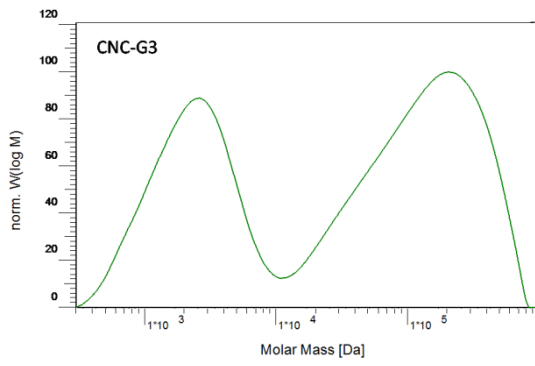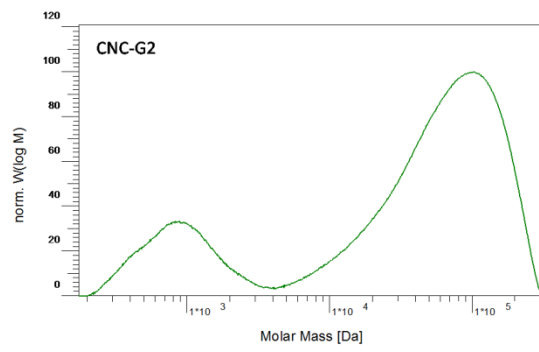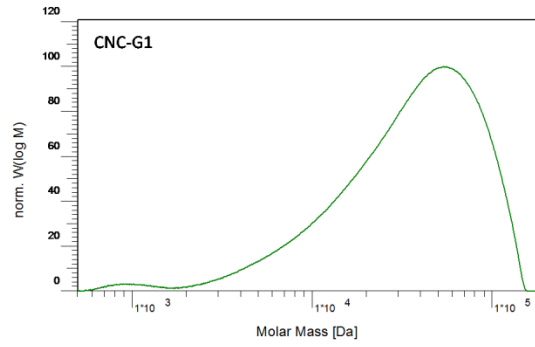
GPC Spectra

Figure S3 Molecular weight distributions of free polymer by AGET ATRP

As seen in Figure S3, two distinct peaks with various intensity ratios were observed for this series of surface-grafted CNCs.

Calculations of bromine content of CNC-Br (C_{Br}) the molecular weight ($M_{n,\,w}$) from the increased weight and molecular weight ($M_{n,\,s}$) of CNC-G3 from solid-state ^{13}C NMR

$$C_{Br} = \frac{A_{172}/A_{105}}{(M_{gu} + A_{172}/A_{105} \bullet M_{initiator})} \qquad (1)$$

$$M_{n,w} = \frac{\Delta m}{C_{Br} \bullet m_0} + M_{initiator} \qquad (2)$$

$$M_{n,\,s} = (\frac{A'_{175}/A'_{105}}{A_{172}/A_{105}} - 1) \bullet M_{NIPAAM} + M_{initiator} \qquad (3)$$

Where A_{172} and A_{105} are the integral at 172 and 105 ppm for CNC-Br (Figure 2), respectively; M_{gu} is the molecular weight of glucose unit; $M_{initiator}$ is the molecular weight of immobilized initiator on CNCs; Δm is the increased weight of CNCs after grafting; m_0 is the initial mass of CNC-Br; M_{NIPAAM} is the molecular weight of NIPAAM; A'_{175} and A'_{105} are the integral at 175 and 105 ppm for CNC-G3 (Figure 2), respectively. The calculations of molecular weight took no consideration of encapsulated dye groups.
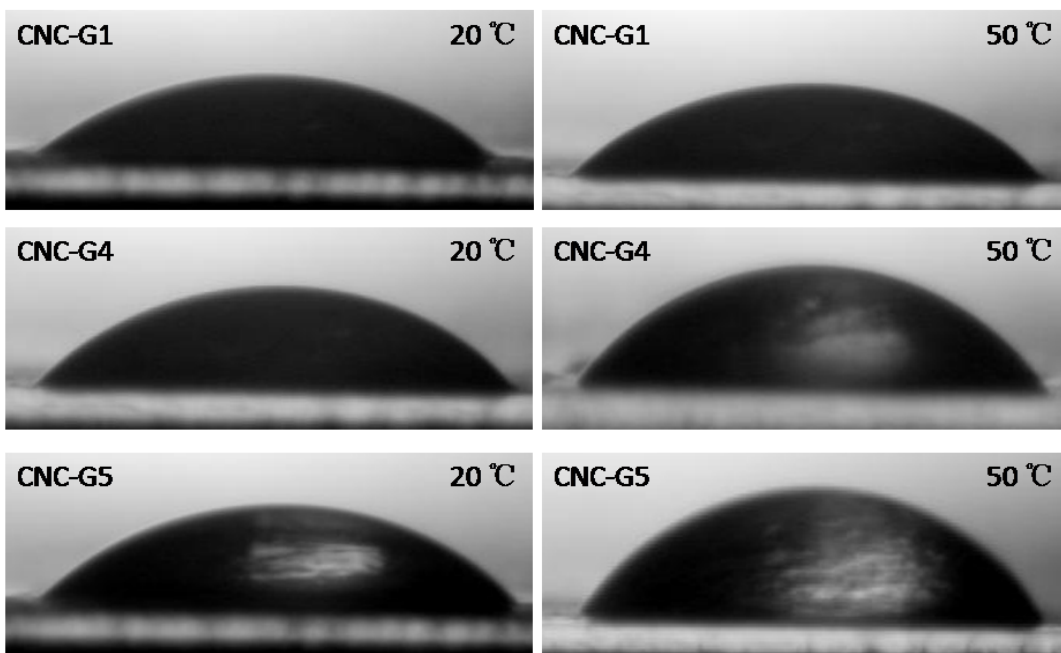
Contact angles

Figure S4 Contact angle pictures for surface-grafted CNCs at 20 and 50 °C.

There was an increase in the contact angle of surface-grafted CNCs when increasing the temperature from 20 to 50 °C.