Supplemental Information:

Dispersion Quantification:

The micron-millimeter scale dispersion of CNCs in the various samples was evaluated using optical microscopy (OM). The ability of this technique to resolve CNC aggregates smaller than 2 μm was limited by the pixel resolution of the imaging camera at the chosen magnification. The OM images were binarized using the Otsu thresholding algorithm and further denoised to remove single pixel objects. Other noisy features recognized to not be CNCs were manually removed. The areas of the remaining objects were then measured using ImageJ. This process was repeated for 2 more optical images per sample.

CNC aggregates smaller than 2 μm and individually dispersed CNCs were imaged using transmission electron microscopy (pixel resolution: 3.9 nm). Here, a background subtraction was performed to remove low frequency variations in image intensity. The images were then binarized using the Otsu algorithm and denoised. Following this, the areas of the CNC objects were measured using ImageJ. This process was repeated for 2 more TEM images per sample.

Assuming that the areas in the optical images, where CNCs are not resolvable, i.e., the bright regions, present the dispersion morphology seen the TEM images (Figure S1), a rough quantitative estimate of the overall CNC dispersion may be obtained using the optical and TEM imaging data as follows.

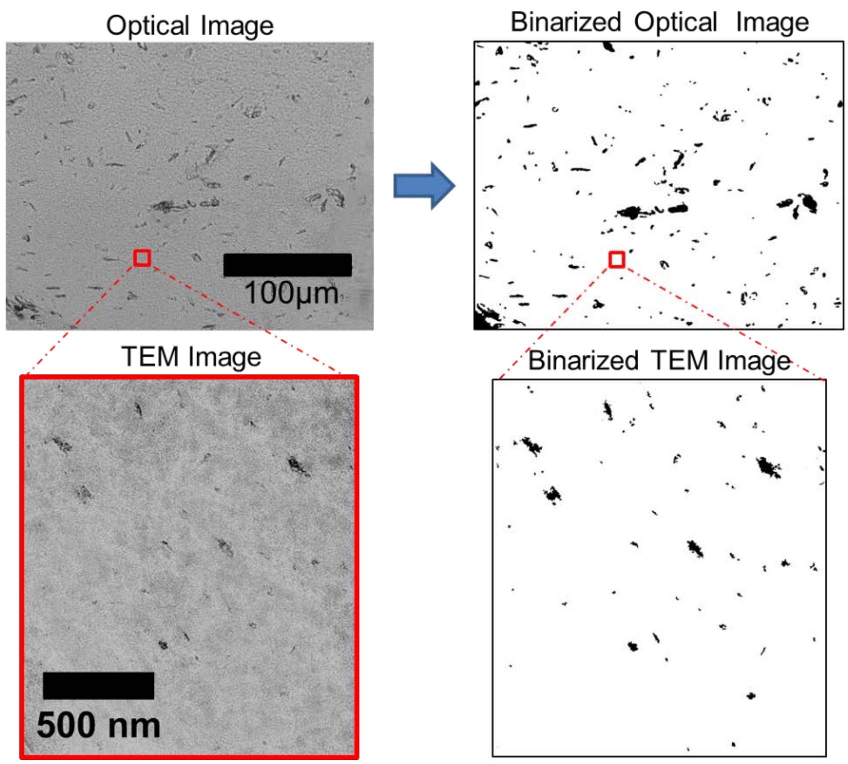

Figure S1. Schematic illustrating the size and generalized location of a 5 % modified TEM image relative to a typical optical image. The denoised, binarized version of the same images are shown on the right.

For a given image area (A), the average area (size) of a CNC aggregate (A_{agg}) is the total CNC area in the sample (A_{CNC}) divided by the total number of aggregates (N_{agg}), which is

$$A_{agg} = \frac{A_{CNC}}{N_{agg}} = \frac{A_{CNC-TEM}+A_{CNC-OM}}{N_{CNC-TEM}+N_{CNC-OM}} \tag{1}$$

Here,

$$A_{CNC-OM} = a_o A \tag{2}$$

$$N_{CNC-OM} = a_o A/A_o \tag{3}$$

where, a_o is the area fraction of the particles in the optical image and A_o is the average area of an optically observed particle.

Similarly,

$$A_{CNC-TEM} = a_{TEM}(1-a_o)A \tag{4}$$

$$N_{CNC-TEM} = a_{TEM}(1 - a_o)A/A_{TEM} \tag{5}$$

where, a_{TEM} is the area fraction of the particles in the TEM image and A_{TEM} is the average area of a TEM-observed particle. We note that the TEM images only represent the areas in the optical images where CNCs are not resolvable. Therefore, the area fraction of the TEM resolvable particles over the entire sample is calculated as $a_{TEM}(1 - a_o)$. The total area fraction of particles is $a_o + a_{TEM}(1 - a_o)$ (Table 1).

From the above equations A_{agg} may be obtained from the imaging data as,

$$\mathrm{A}_{agg} = \frac{a_o A + a_{TEM}(1-a_o)A}{a_o A/A_o + a_{TEM}(1-a_o)A/A_{TEM}} = \frac{a_o + a_{TEM}(1-a_o)}{a_o/A_o + a_{TEM}(1-a_o)/A_{TEM}} \tag{6}$$

Since the imaged aggregates are not spherical, using A_{agg} to obtain an accurate representation of the particle size in the length dimension is not straightforward. However, for comparative purposes, the average diameter of a sphere with a cross-sectional area = A_{agg}, is calculated as $\sqrt{4\mathrm{A}_{agg}/\pi}$. The measured values of A_{agg} and D_{agg} for the various samples are shown in Table 1.

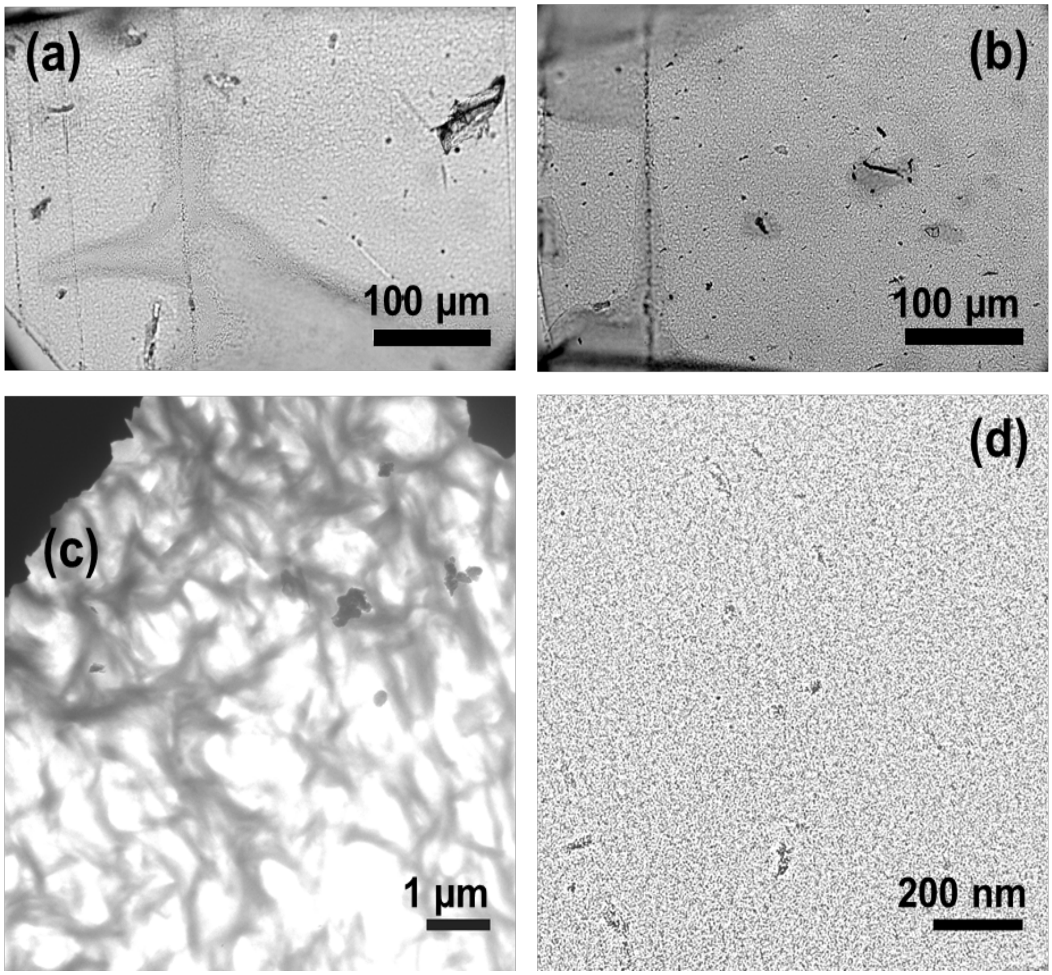

Figure S2. Optical images of a) 1% CNC unmodified and b) 1 % CNC modified samples. TEM images of c) 1% CNC unmodified and d) 1 % CNC modified samples.

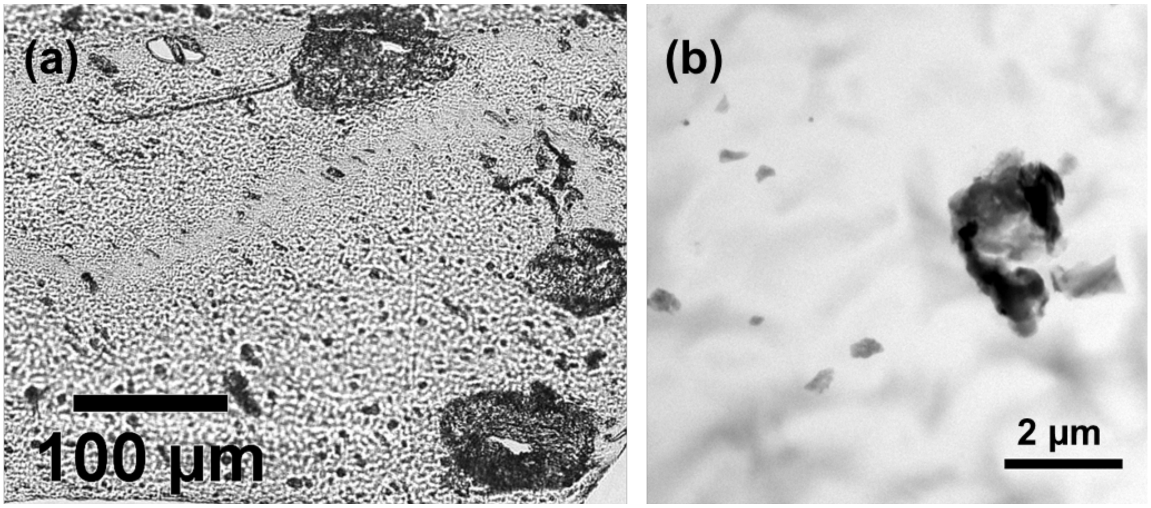

Figure S3. (a) Optical and (b) TEM images of 10% *modified* CNC sample.

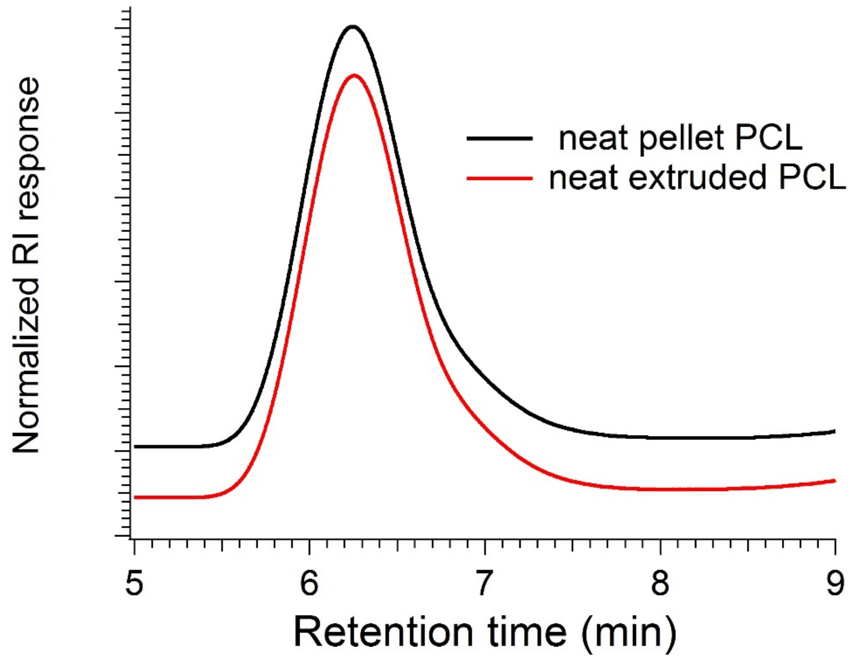

Figure S4. GPC result with vertical offset added due to clarity: For the neat pellet, PCL Mn = 74.1 kDa,, Mw = 140.6 kDa, PDI = 1.90. For neat extruded, PCL Mn = 75.3 kDa, Mw = 140.9 kDa, PDI = 1. 87.

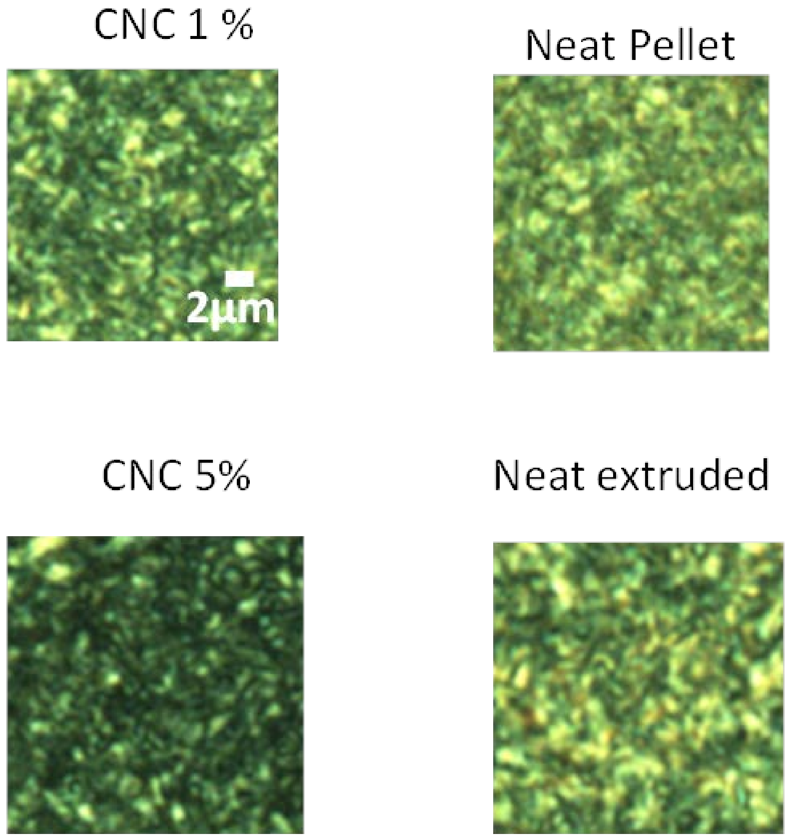

Figure S5. Optical graphs using transmission optical microscopy for samples at crystallization temp, 42 °C, scale bar is 2μm