Supporting Information

Rapid determination of the distribution of cellulose nanomaterial aggregates in composites enabled by multi-channel spectral confocal microscopy

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There is increased interest in the use of cellulose nanomaterials for the mechanical reinforcement of composites due to their high stiffness and strength. However, challenges remain in accurately determining their distribution within composite microstructures. We report the use of a range of techniques used to image aggregates of cellulose nanocrystals (CNC) within a thermoplastic polymer. Whilst Raman imaging accurately determines CNC aggregate size, the limited observable area results in poor reproducibility. In contrast, staining the CNCs with a fluorophore enables high reproducibility, but overestimates the aggregate size as CNC content increases. Spectral confocal microscopy is presented as an alternative technique that combines the accuracy of Raman with the reproducibility of conventional confocal microscopy, enabling the rapid determination of CNC aggregate distribution within composites.

Keywords: cellulose, nanomaterials, composites, confocal microscopy, spectral imaging, confocal Raman spectroscopy



Figure S1. Confocal Raman spectroscopy mapping of CNC-MAPE composite (laser wavelength: 785 nm, laser power: 41 mW, lateral resolution: 684 nm) A) Map acquired at 1096 cm-1, indicating intensity of band associated with cellulose; B) Map acquired at 1295 cm-1, indicating intensity of band associated with polyethylene; C) Chemical map indicating areas where cellulose (blue) and polyethylene (red) are dominant.



Figure S2. Representative confocal microscopy images of A) calcofluor white stained sample using conventional imaging method; B) calcofluor white stained sample using spectral imaging method; C) unstained sample using conventional imaging method; and D) unstained samples using spectral imaging method. Samples imaged using argon laser intensity of 0.2 %. Orange and white arrows indicate example CNC aggregates that can be identified in all the presented images.



Figure S3. Normalised emission spectra for pure polyethylene polymer.



Figure S4. Histogram of CNC aggregate distribution detected using A) SCM and B) CCM for CNC content of 0.625 (blue bars); 1.25 (red bars with rising diagonal lines); 2.5 (yellow bars with falling diagonal lines); and 5 wt.% (green bars with horizontal lines). Bin size: 500 for SCM, 1000 for CCM.



Figure S5. Average number of CNC aggregates identified at each wt.% using A) SCM and B) CCM that are attributed to the small (blue squares); medium (red circles); large (green triangles); and outlier (yellow inverted triangles) categories within each sample. Lines of best fit plotted for each category to guide the eye. Error ± SE.