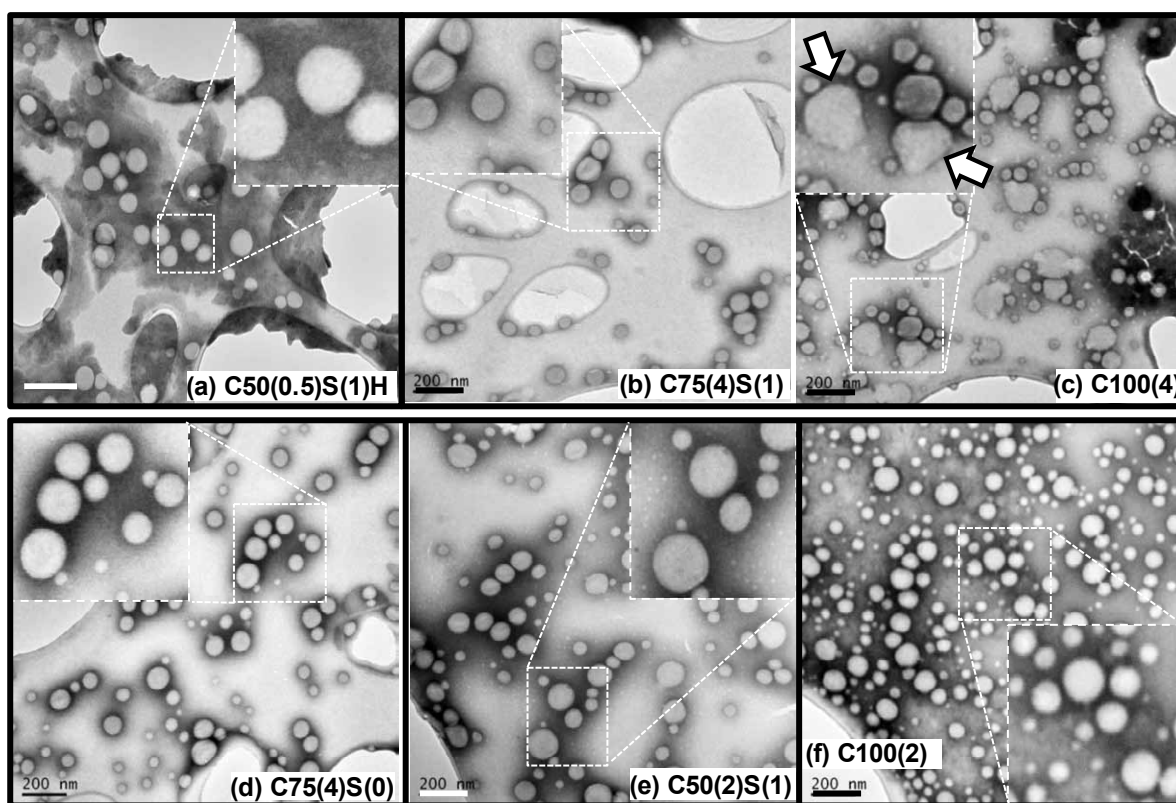


## SUPPORTING INFORMATION

### Tuning the mechanical properties of nanostructured ionomer films by controlling the extents of covalent crosslinking in core-shell nanoparticles

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**Fig. S1.** TEM images of deposited nanoparticles. Their identities are shown in each figure. The arrows in (c) show coalesced nanoparticles. The scale bars are 200 nm.

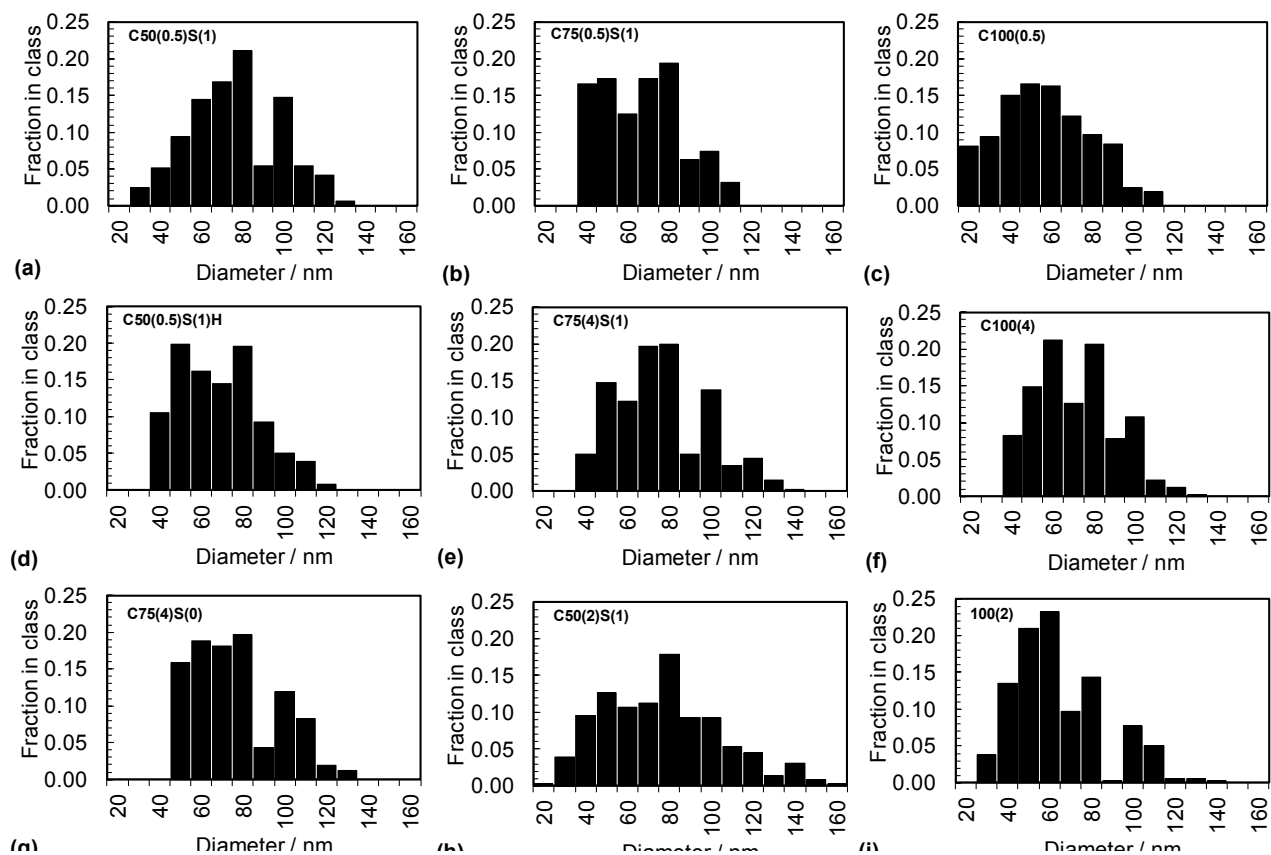


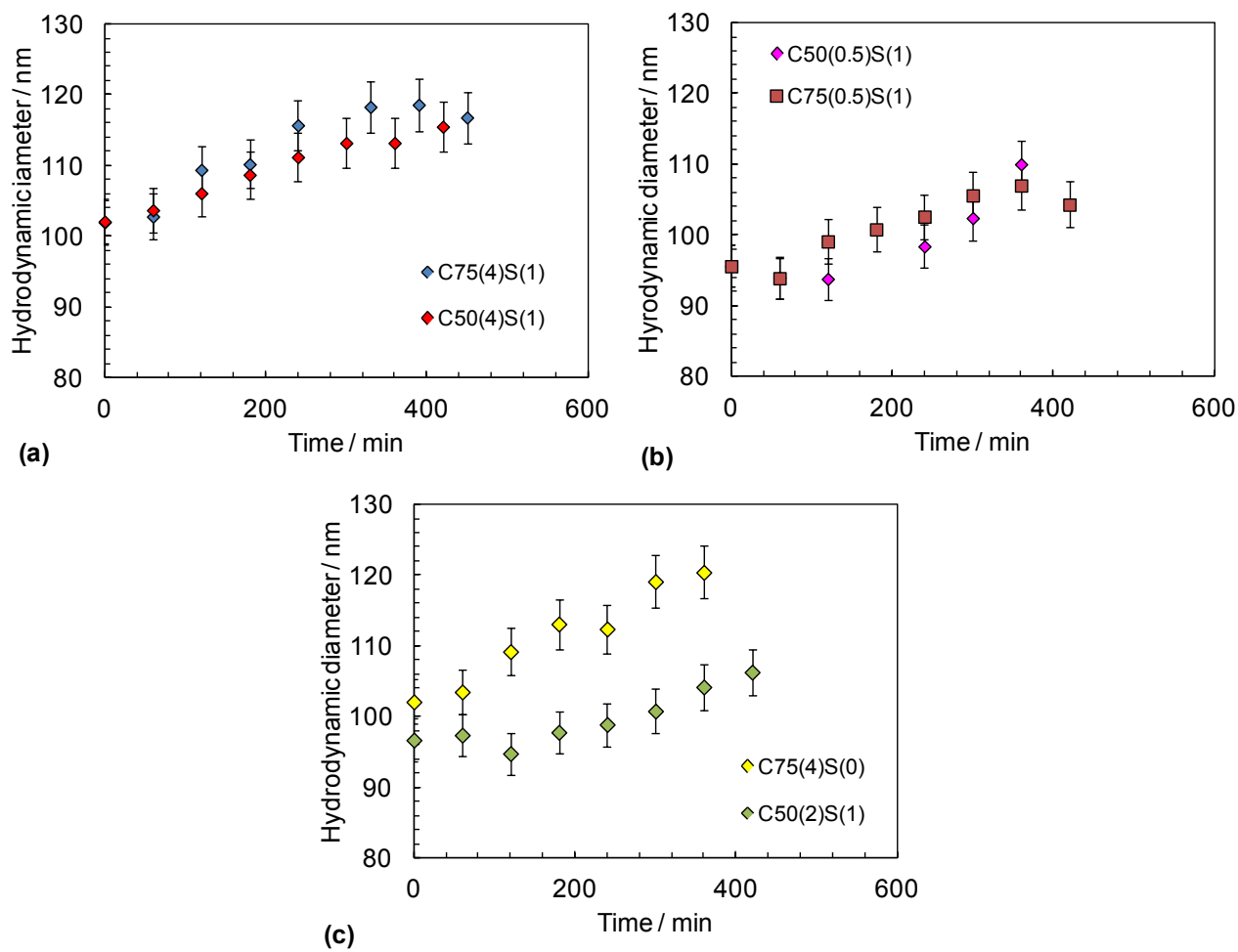
Fig. S2. Particle size distributions for Cx(y)S(z) particles from TEM data.

Table S1: Hydrodynamic diameters for core-shell Cx(y)S(z) and core C100(y) nanoparticles.

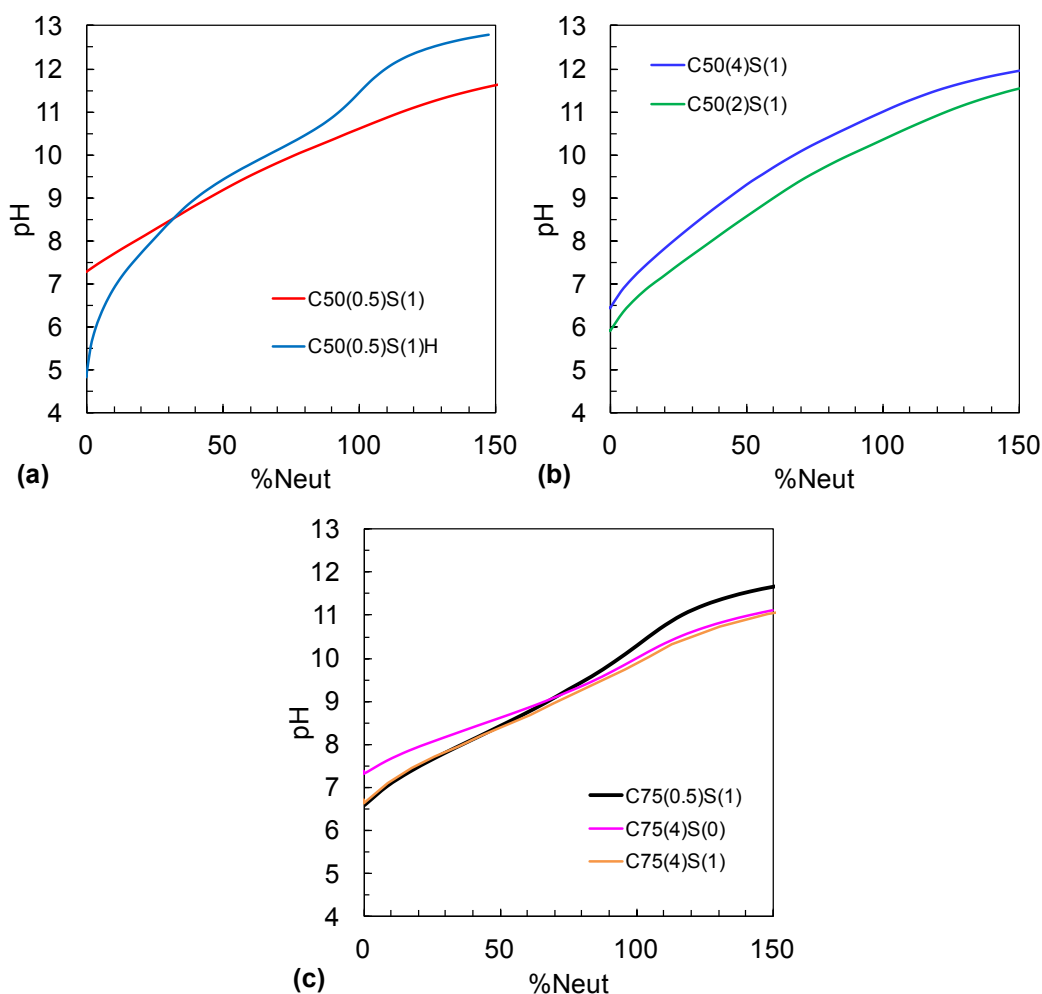
| Code          | $d_h^a$ / nm |
|---------------|--------------|
| C50(0.5)S(1)  | 110          |
| C50(2)S(1)    | 106          |
| C50(4)S(1)    | 116          |
| C50(0.5)S(1)H | 109          |
| C75(0.5)S(1)  | 104          |
| C75(4)S(0)    | 125          |
| C75(4)S(1)    | 117          |
|               |              |

|           |     |
|-----------|-----|
| C100(0.5) | 94  |
| C100(2)   | 97  |
| C100(4)   | 102 |

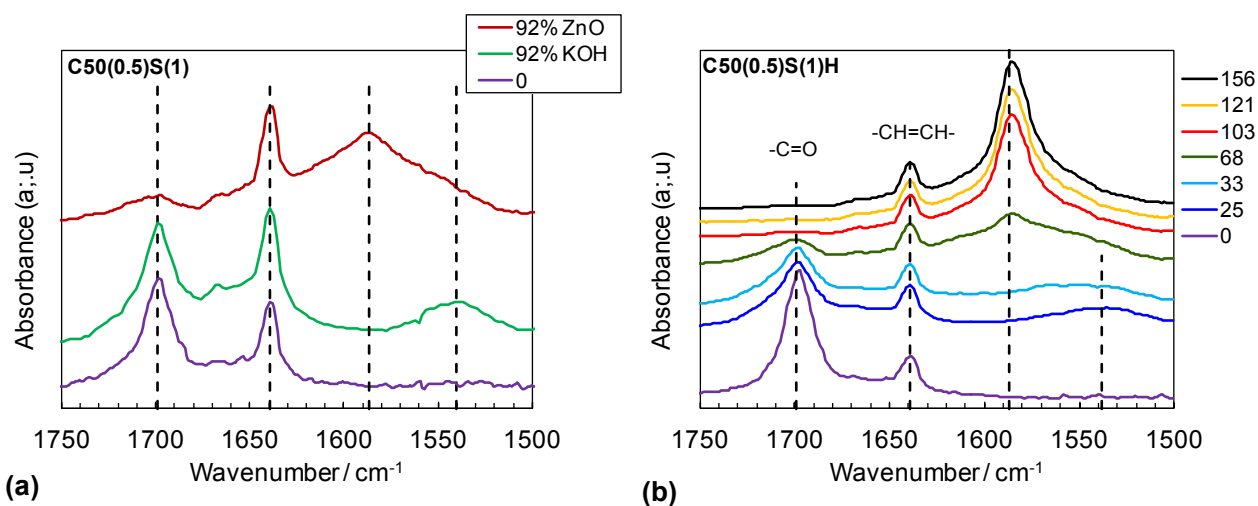
<sup>a</sup> Hydrodynamic diameter measured using PCS.



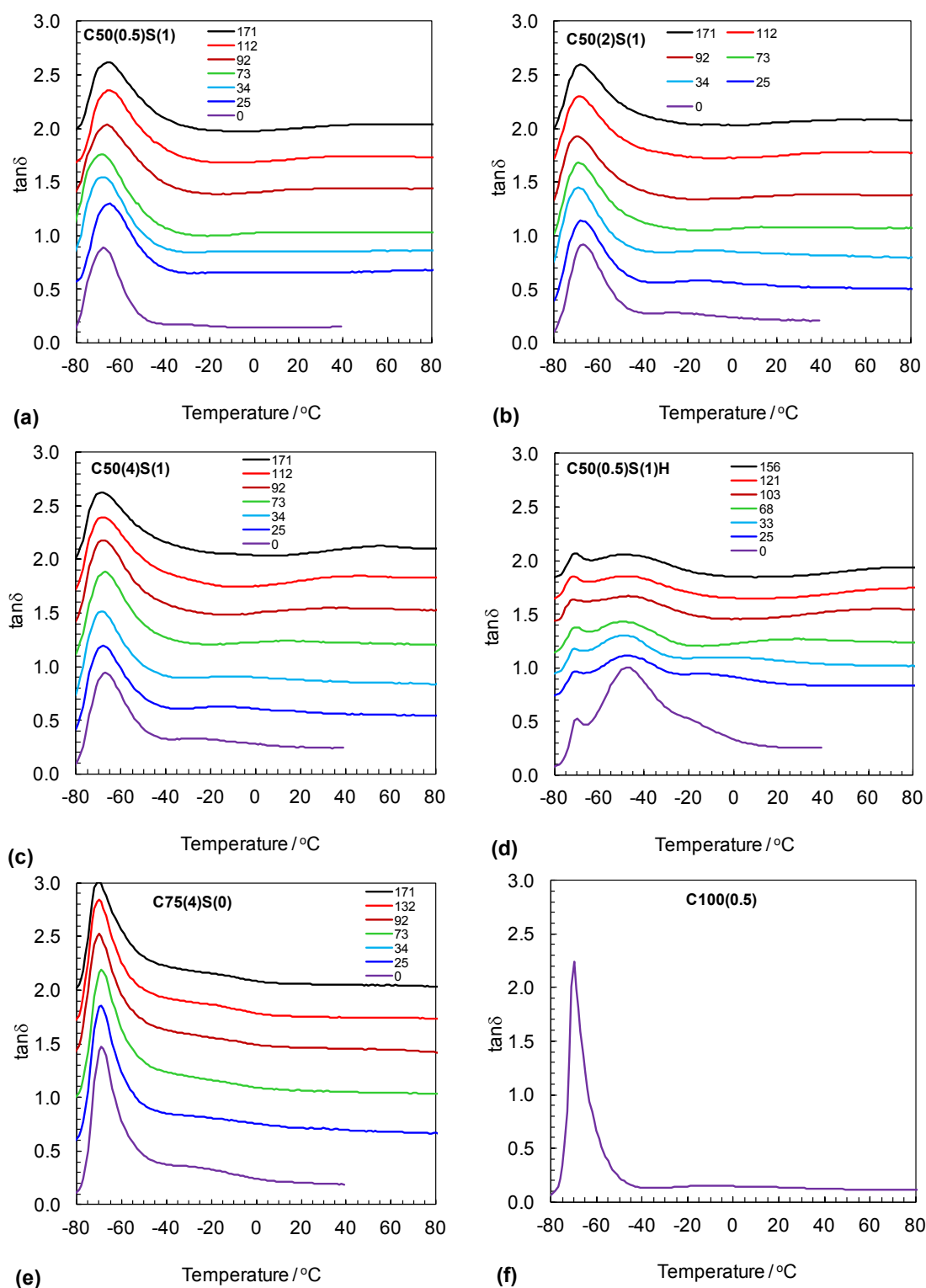
**Fig. S3.** Variation of hydrodynamic diameter with time for the core-shell systems during shell growth. The error bars are the uncertainty for the data.

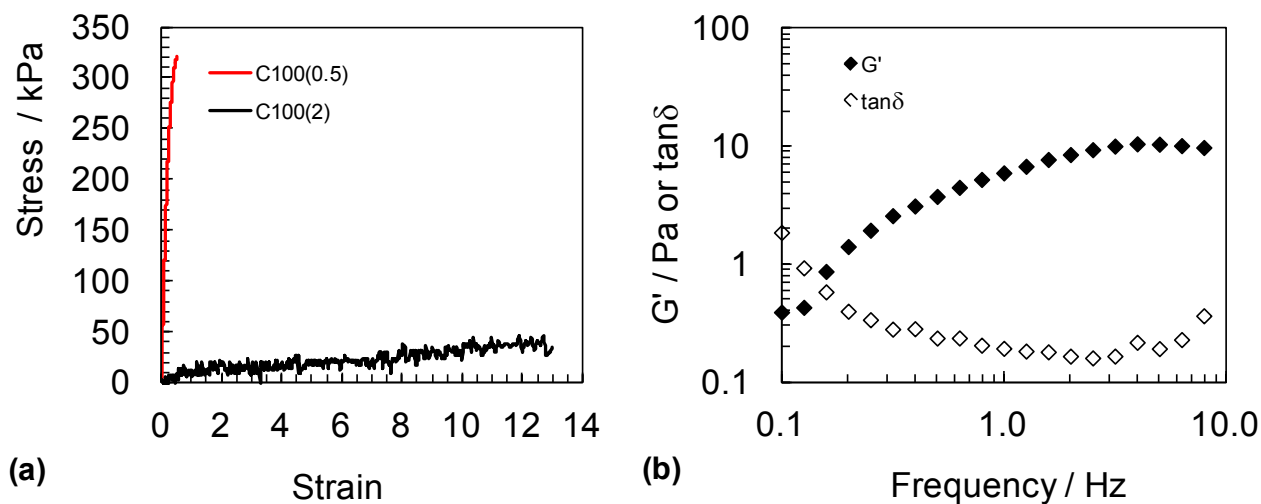


**Fig. S4.** Titration data for  $C_x(y)S(z)$  nanoparticles. The identities are shown in each figure.

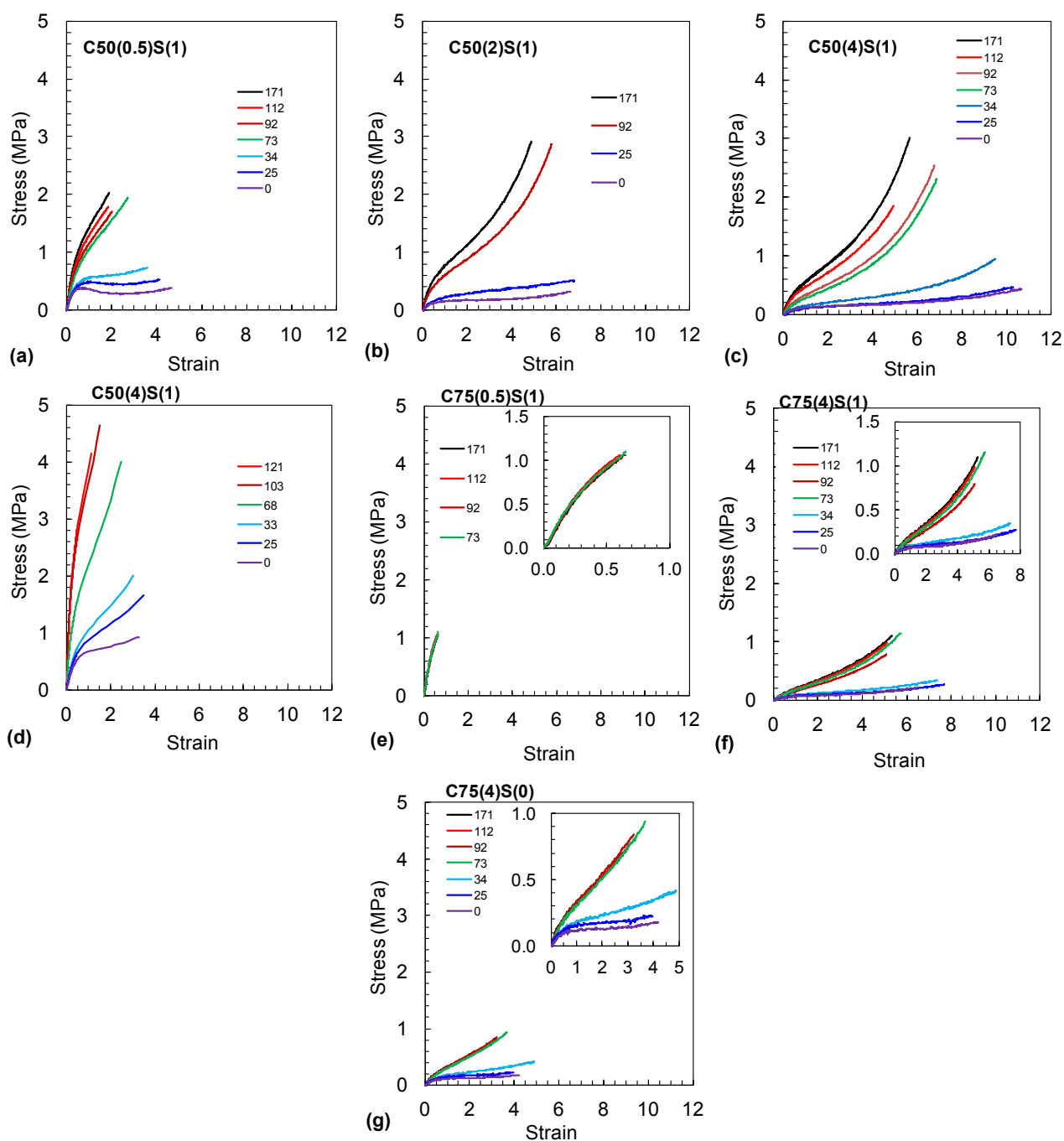


**Fig. S5.** FTIR spectra of (a) of  $C50(0.5)S(1)$  with nominal neutralisations of 92% using KOH or ZnO and (b)  $C50(0.5)S(1)H$  films at different nominal neutralisations. The non-neutralised spectrum is also shown for comparison in (a). The absorbance values have been shifted vertically for clarity.





**Fig. S7. Mechanical properties of core particle films.** (a) Tensile stress vs. strain data C100(0.5) and C100(2) films. (b) Frequency sweep data for C100(4) films.



**Fig. S8.** Stress strain curves for various  $C(x)S(y)$  films at different nominal neutralisations (shown in legends). The insets for (e) – (g) show expanded scales.