# Supporting Information

# NaClO-oxidized Cellulose Nanofibers/Chitosan Composite Films with Improved Water Resistance and High Mechanical Strength

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| Number of pages | 8 (page S2-S7) |
| Number of figures | 11 (figure S1 – S11) |
| Number of tables | 4(table S1 – S4) |

**Table S1**. The yield and the carboxylate content of the OCNFs oxidized in different conditions.

| NaClO% (w/w) | 18 | 15 | 13 | 13 |
|---|---|---|---|---|
| Time (h) | 2 | 2 | 2 | 3 |
| COONa (mmol/g) | 0.55 | 0.47 | 0.43 | 0.61 |
| Yield (%) | 57.7 | 69.6 | 83.4 | 62.8 |

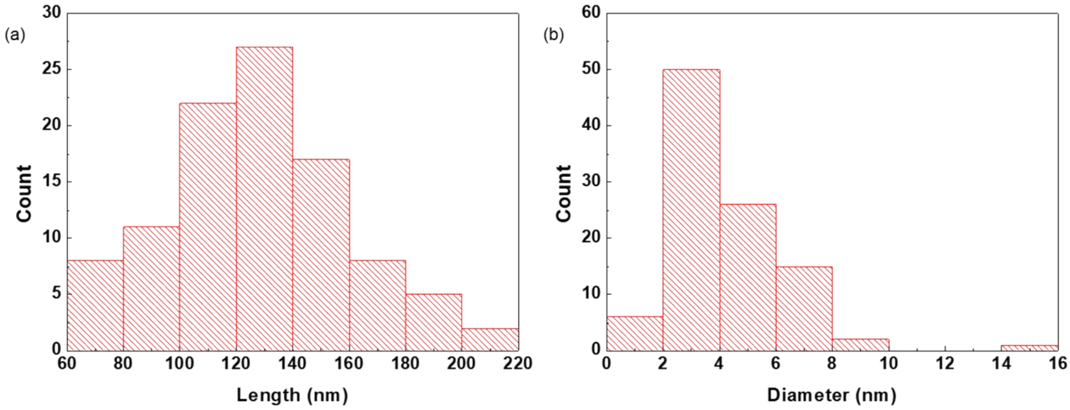


**Figure S1.** Distribution graphs of the OCNFs (a) length, (b) diameter, calculated from TEM images using the ImageJ.

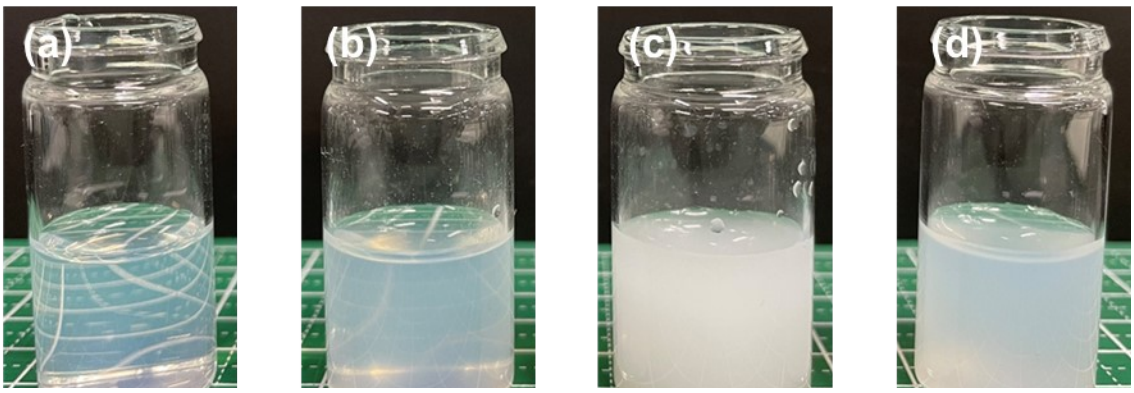


**Figure S2.** Images of OCNF dispersions oxidized using (a) 18% (w/w), (b) 15% (w/w), and (c) 13% (w/w) NaClO $_{(aq)}$. for 2 h, and (d) 13% (w/w) NaClO $_{(aq)}$ for 3 h.

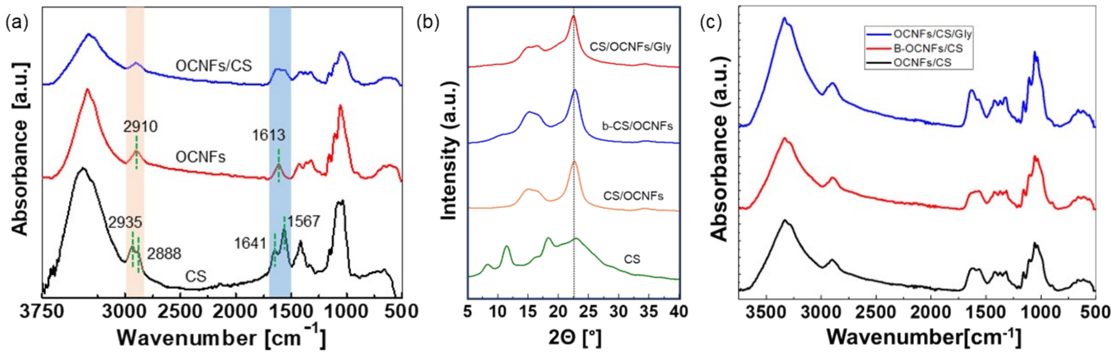


**Figure S3**. (a) FTIR spectra of CS, OCNFs, and CS/OCNFs film (b) XRD patterns of CS and composites (c) FTIR spectra of OCNFs/CS, b-OCNFs/CS, and OCNFs/CS/Gly films.

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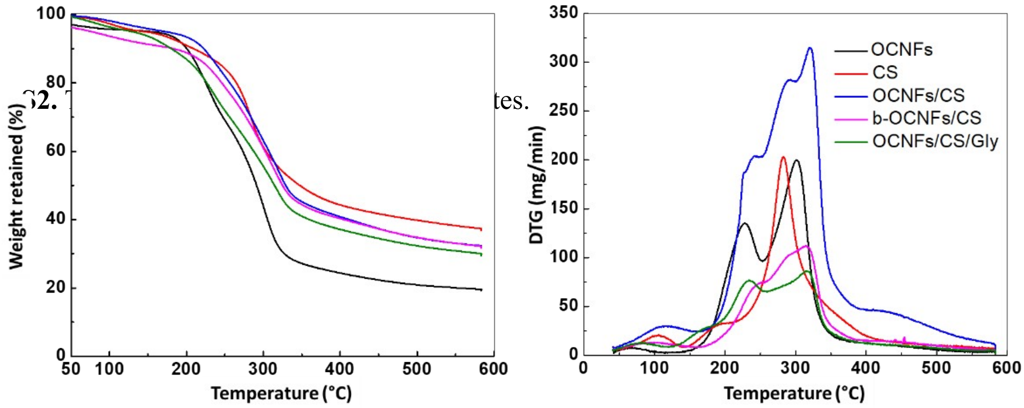


**Figure S4**. Weight retained percentage and DTG curves at different temperatures of CS, OCNFs, OCNF/CS, b-OCNFs/CS, and OCNFs/CS/Gly composite films.

**Table S3.** Mechanical properties of bio-based composite materials fabricated using different methods.

| Sample | CS | OCNFs | OCNF/CS | b-OCNFs/CS | OCNFs/CS/Gly |
|---|---|---|---|---|---|
| $T_{onset}$ (°C) | 145 | 154 | 172 | 76 | 129 |
| $T_{peak}$ (°C) | 282 | 300 | 319 | 314 | 314 |
| $T_{50\%}$ (°C) | 347 | 291 | 329 | 325 | 314 |

**Table S3.** Mechanical strength properties of materials fabricated by PLA and PHBV using different methods.

| | | | Strain at break (%) |
|---|---|---|---|
| | | | ~3 |

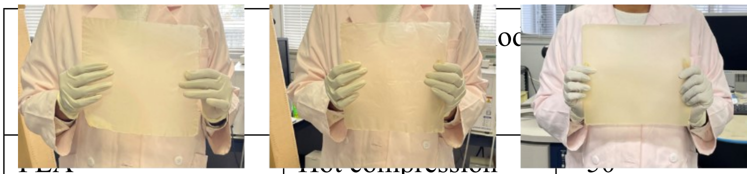


**Figure S5**. Images of prepared larger-scaled films of OCNF/CS-1, b-OCNF/CS-1, and OCNF/CS/Gly-10%.

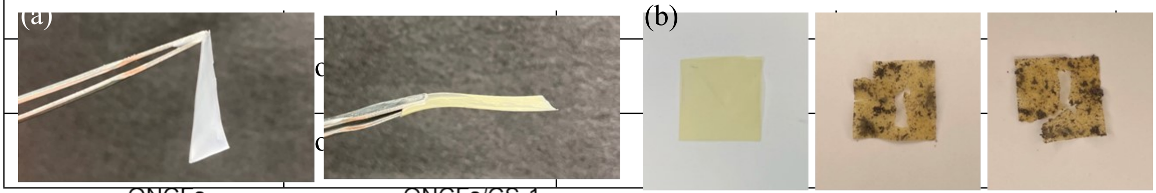


**Figure S6**. (a) Images of the films immersed in DI water. (b) Images of the film, which was buried in soil under natural weather conditions.

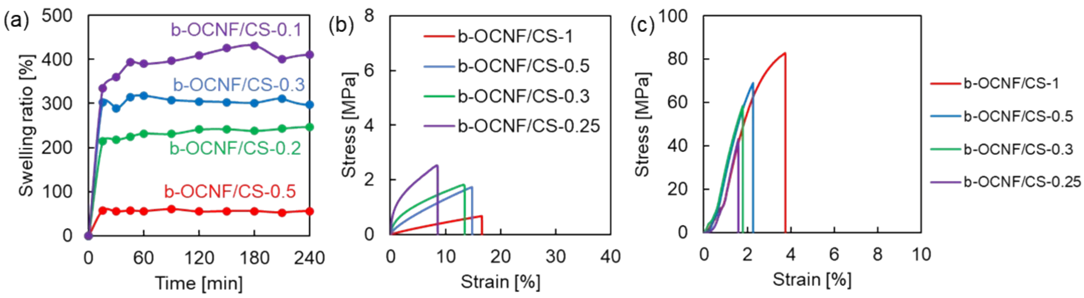


**Figure S7**. (a) Swelling ratio and stress-strain curves of b-OCNF/CS with increasing OCNFs content in (b) wet and (c) dry state.

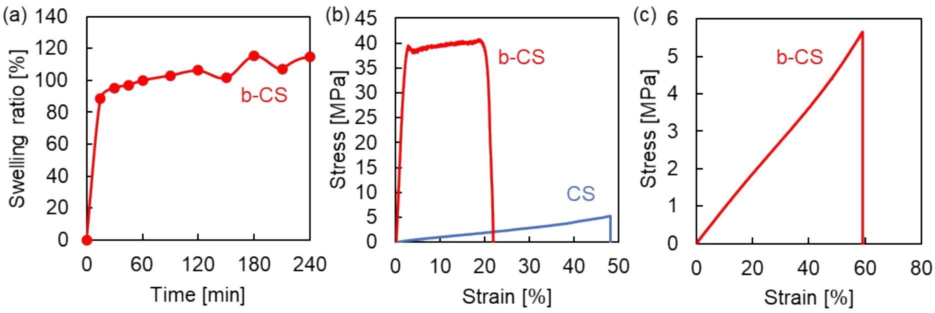


**Figure S8**. (a) Swelling ratio and stress-strain curves in (b) dry and (c) wet state of b-CS film.

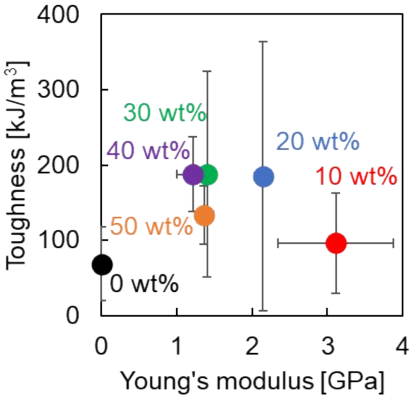


**Figure S9**. Young's modulus and toughness plots of OCNF/CS/Gly in the wet state.

**Table S4.** Swelling ratios of the OCNF/CS/Gly-10% sample in different time intervals for 24 hours.

| Weight (W) in different time intervals (g) | Sample 1 | Sample 2 |
|---|---|---|
| **Dry W** | 0.0165 | 0.0168 |
| **W 0.5 h** | 0.0251 | 0.0267 |
| **W 1 h** | 0.0265 | 0.0273 |
| **W 1.5 h** | 0.0273 | 0.027 |
| **W 2 h** | 0.0265 | 0.0274 |
| [illegible] | [illegible] | [illegible] |
| **W 3 h** | 0.0275 | 0.0271 |
| **W 3.5 h** | 0.0269 | 0.0272 |
| **W 4 h** | 0.0277 | 0.0271 |
| **W 4.5 h** | 0.0277 | 0.0277 |
| **W 5 h** | 0.0279 | 0.0274 |
| **W 24 h** | 0.0276 | 0.0279 |

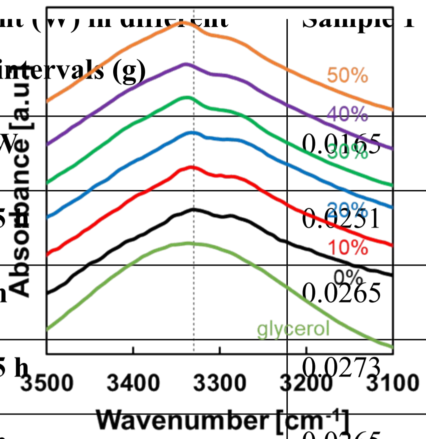


**Figure S10**. FTIR spectra of OCNFs/CS/Gly composite films at the varied amounts of glycerol.

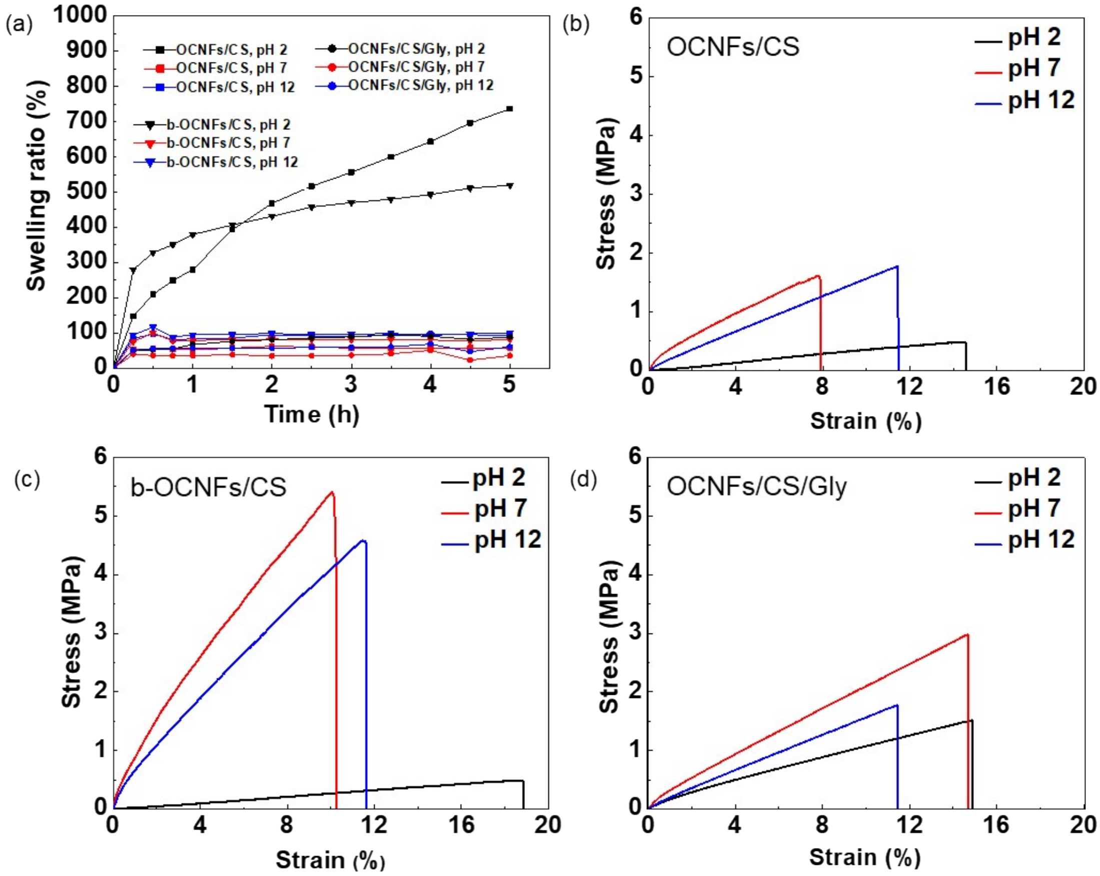


**Figure S11**. (a) Swelling behavior of the composite films at various pH conditions. Stress-strain curves of (b) OCNFs/CS, (c) b-OCNFs/CS, and (d) OCNFs/CS/Gly films in the wet state with different pH conditions.

## References


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