

Supporting Information

**Milliscale self-integration of megamolecule biopolymers
on a drying gas-aqueous liquid crystalline interface**

Kosuke Okeyoshi*, Maiko K. Okajima, Tatsuo Kaneko*

Japan Advanced Institute of Science and Technology
1-1 Asahidai, Nomi, Ishikawa 923-1292, Japan

*Correspondence and requests for materials should be addressed to

K. O. (Email: okeyoshi@jaist.ac.jp)

T. K. (Email: kaneko@jaist.ac.jp)

Supplementary Movie S1.

Drying process of *sacran* solution and pure water during drying at 60°C. Scale bar: 5 mm.
see **Fig. 2A and Fig. S3.**

Supplementary Movie S2.

Drying process of *xanthan gum* solution during drying at 25°C, 40°C, and 60°C. Scale bar: 5 mm.
see **Fig. 4A.**

Supplementary Movie S3.

Drying process of *sacran*, MTs, and DNA solutions at 37°C. Scale bar: 5 mm.
see **Fig. 5A.**

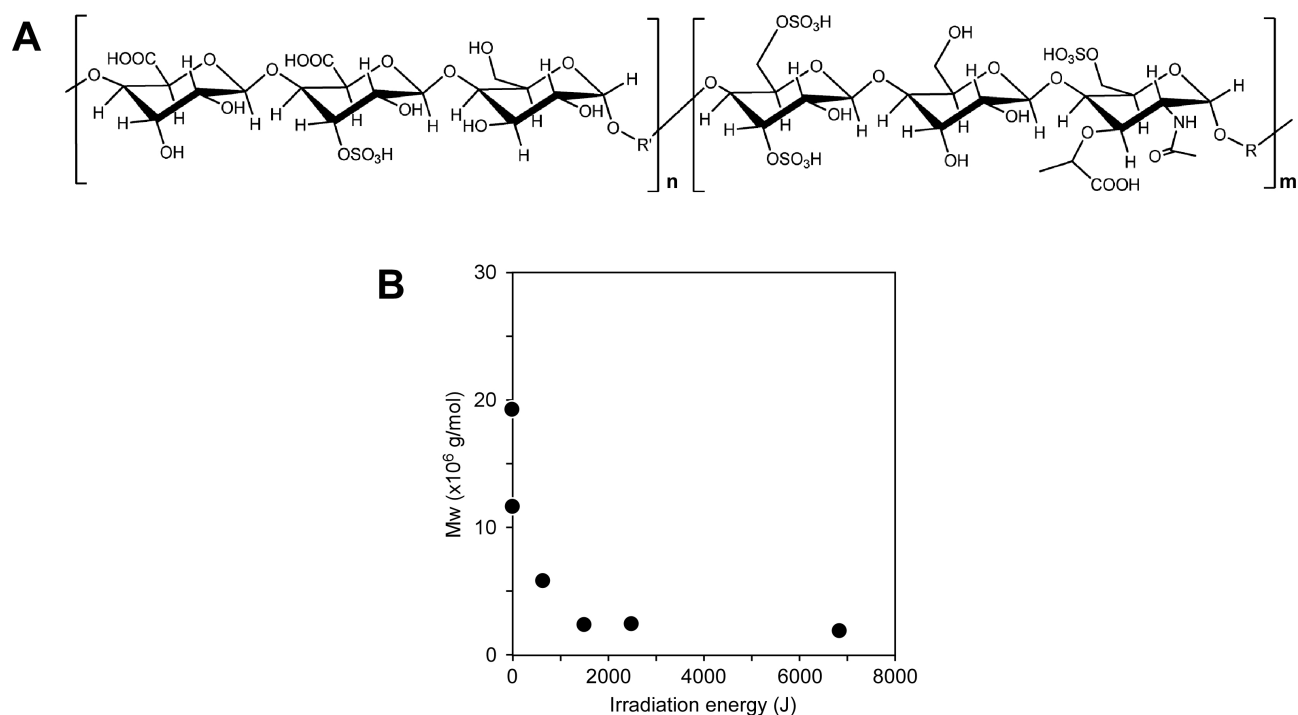


Figure S1. A. Chemical structure of *sacran*. **B.** Effect of irradiation energy during ultrasonication on molecular weight. Irradiation energy was calculated from irradiation time. The average molecular weight, M_w , was determined using the Berry model with the SEC-MALLS system.

Elemental analyses and chromatographic and spectroscopic studies of *sacran* revealed the following sugar residues: Glc, Gal, Man, Fuc, Rha, Xyl, Rib, methylated hexose, uronic acids, and trace muramic acid. The carboxylate composition of *sacran* was 11 mol%, and substitution of sulphate groups was favoured when the sulphate composition was 22 mol% with sugar residues.^[1-5]

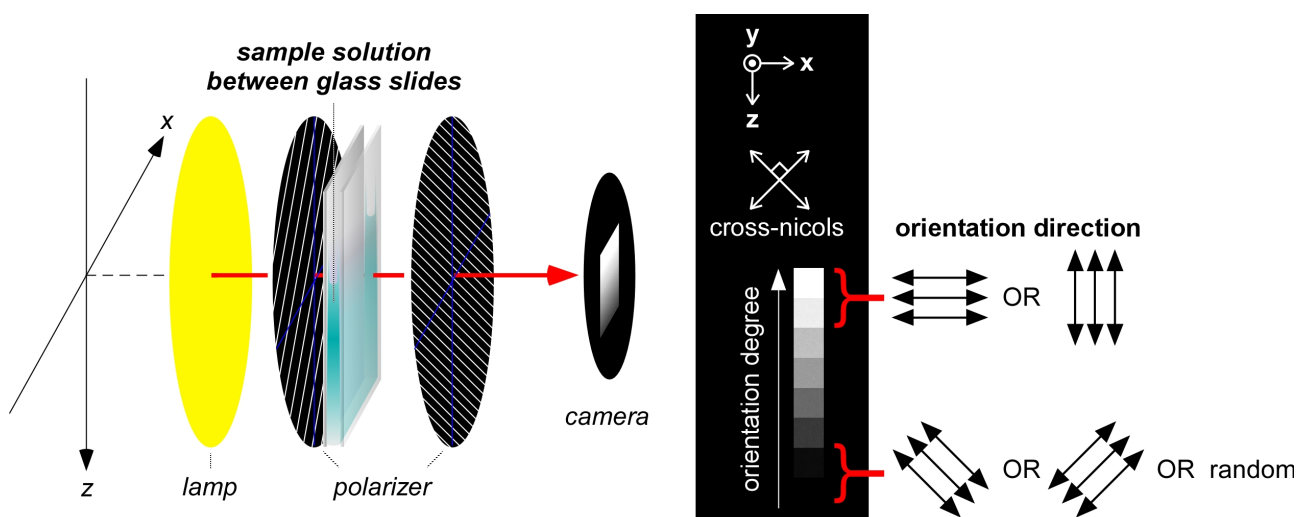


Figure S2. Schematic illustration of the experimental setup used for observations under cross-polarized light. The polarizers were normally adjusted to 45° and 135° .

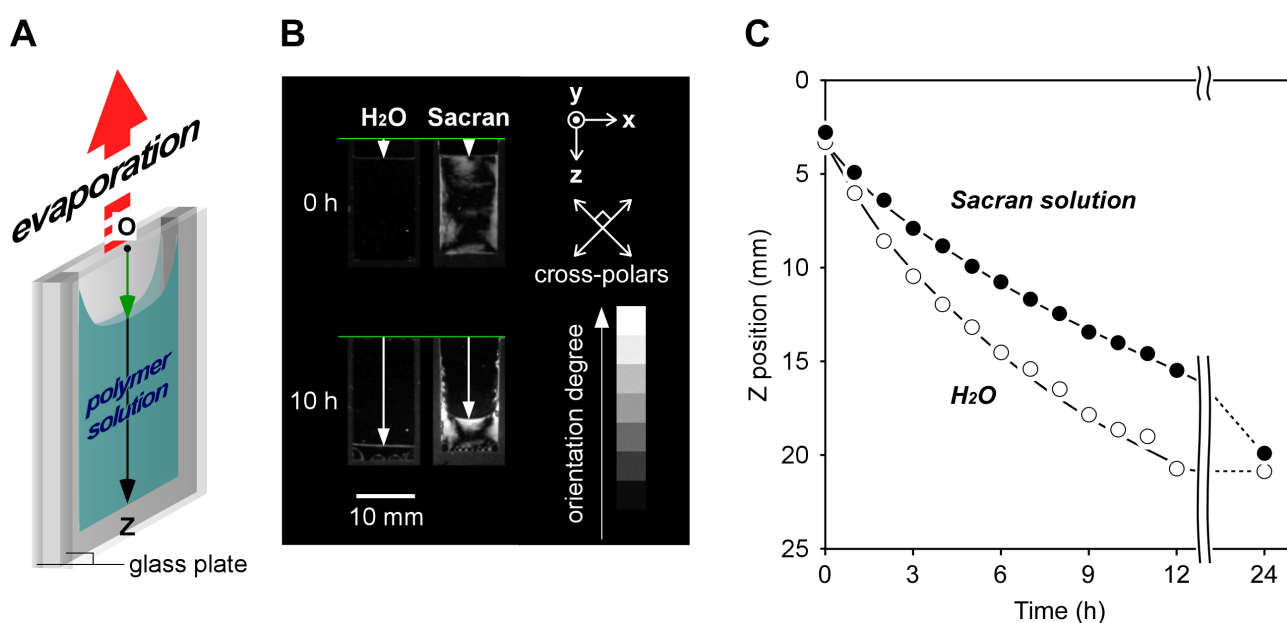


Figure S3. Comparison of gas-liquid interfacial Z-position between polymer solution and water during the drying process from a one-side open cell ($10 \text{ mm} \times 1 \text{ mm} \times \sim 20 \text{ mm}$).

The drying process for the polymer solution and pure water were compared. The descent of the gas-liquid interface of the polymer solution was slower than that of pure water. The oriented domains on the gas-liquid interface caused the slower evaporation rate, working like a skin layer. The evaporated volume of the polymer solution was approximately two-thirds of that of pure water.

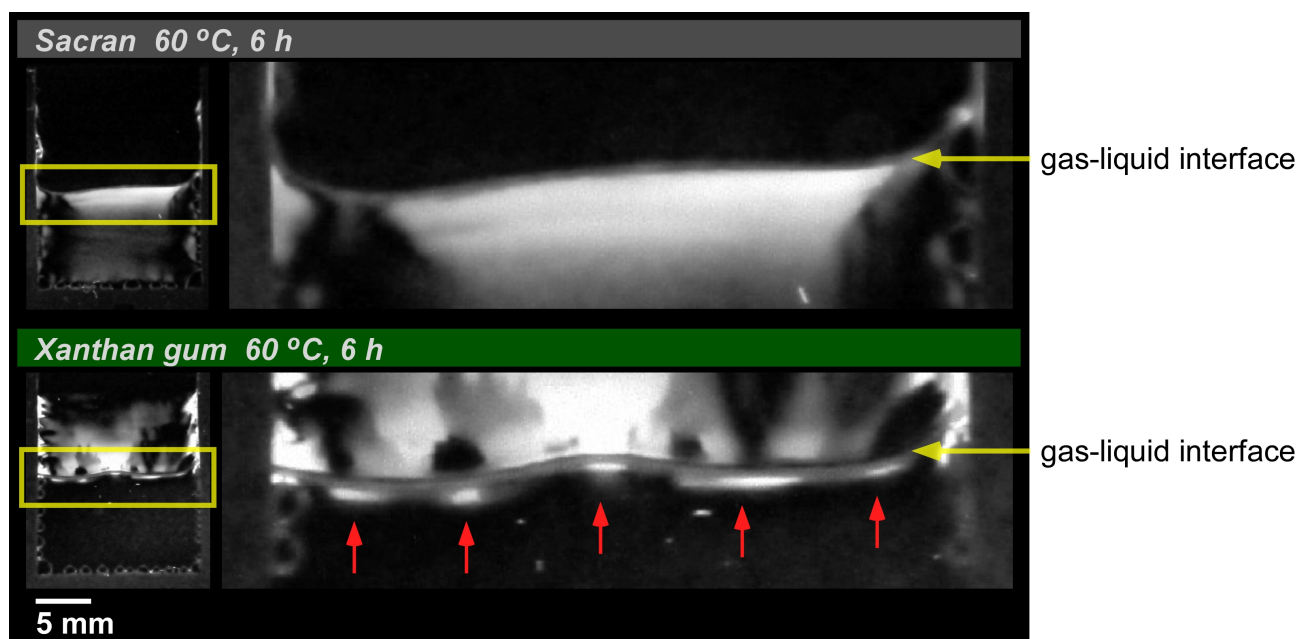


Figure S4. Side views of *sacran* and *xanthan gum* solutions after 6 hours drying at 60°C under cross-polarized light.

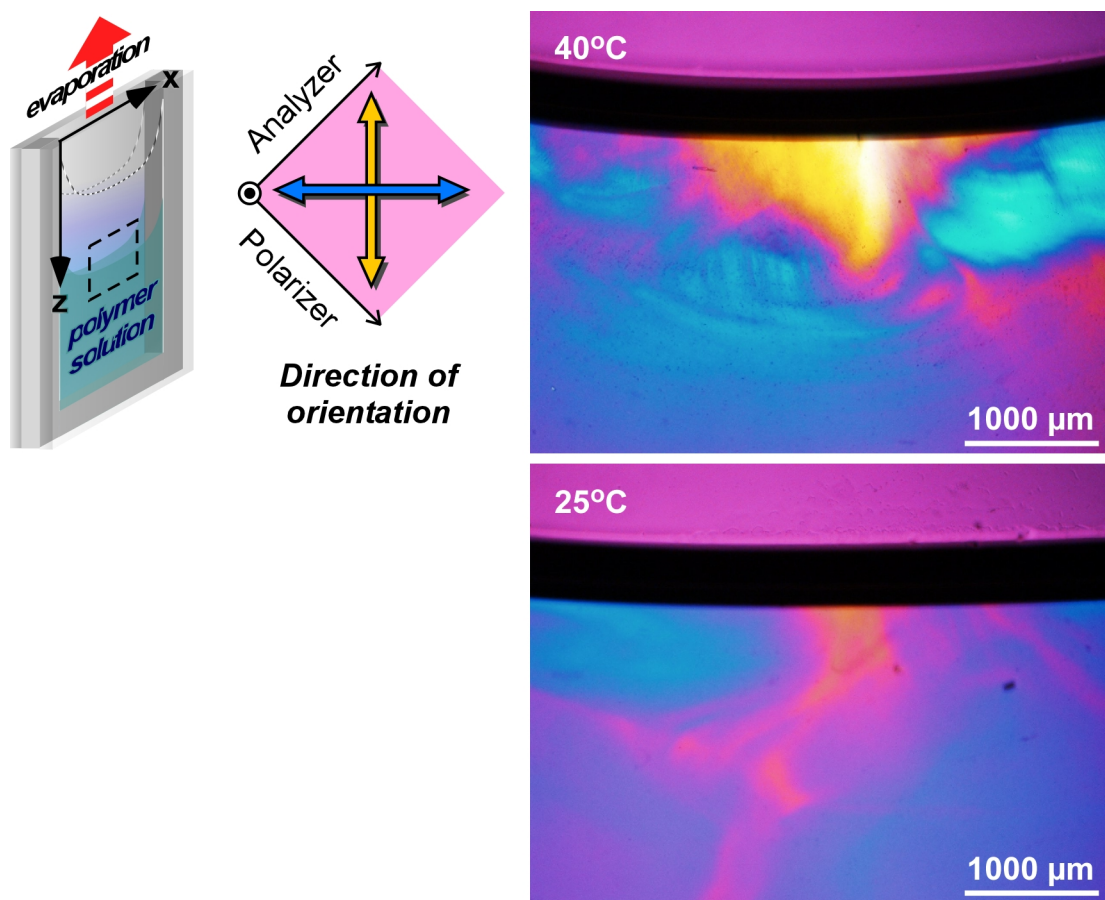


Figure S5 Microscopic images of the *xanthan gum* solutions in the glass cell observed through the cross-nicols with a first order retardation plate ($\lambda = 530$ nm) after 6 hours drying at 60°C. Initial concentration: 0.5 wt%.

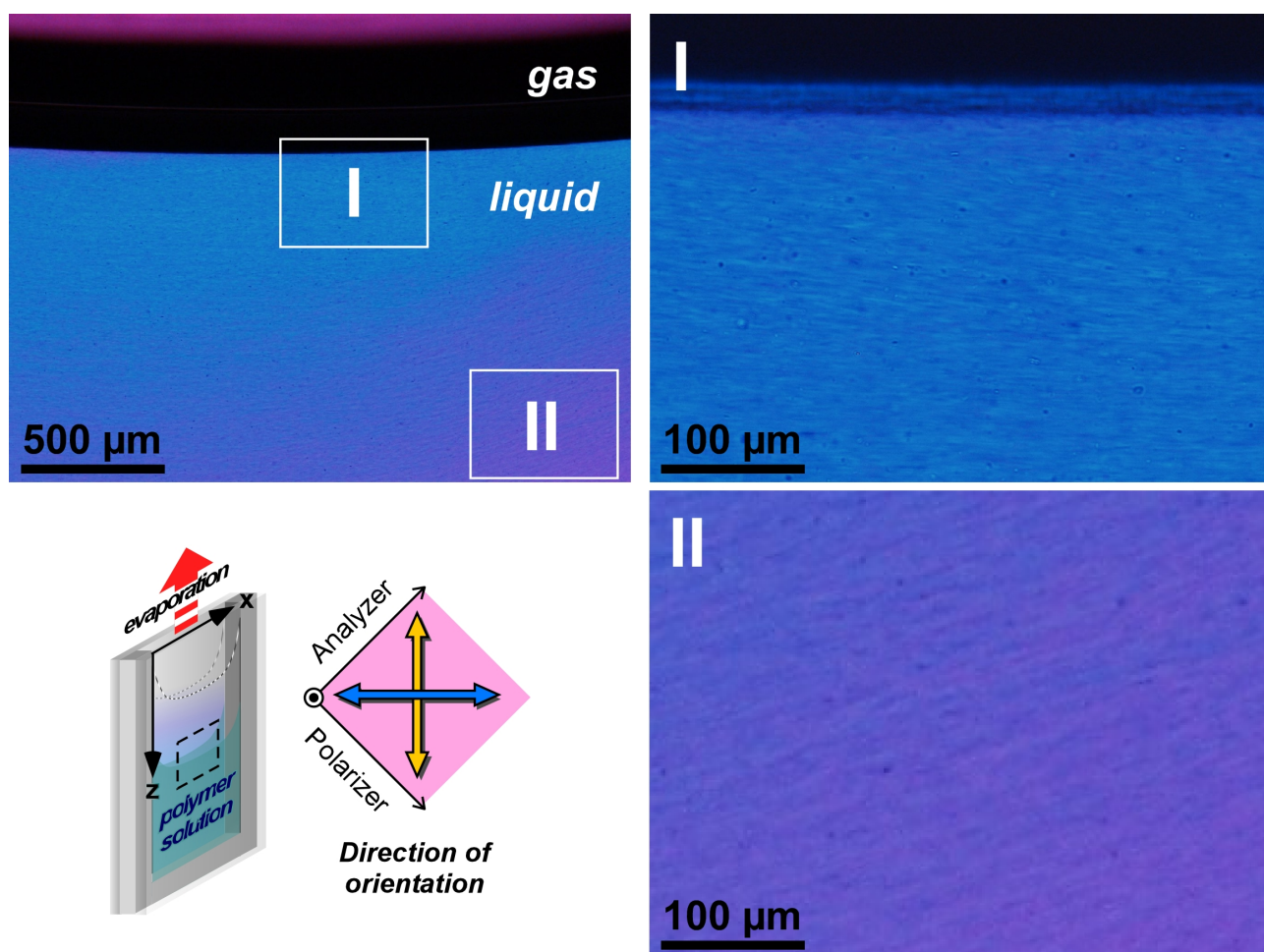


Figure S6. Microscopic images of the *sacran* solutions in the glass cell observed through the cross-nicols with a first order retardation plate ($\lambda = 530 \text{ nm}$) after 6 hours drying at 60°C . Initial concentration: 0.5 wt%.

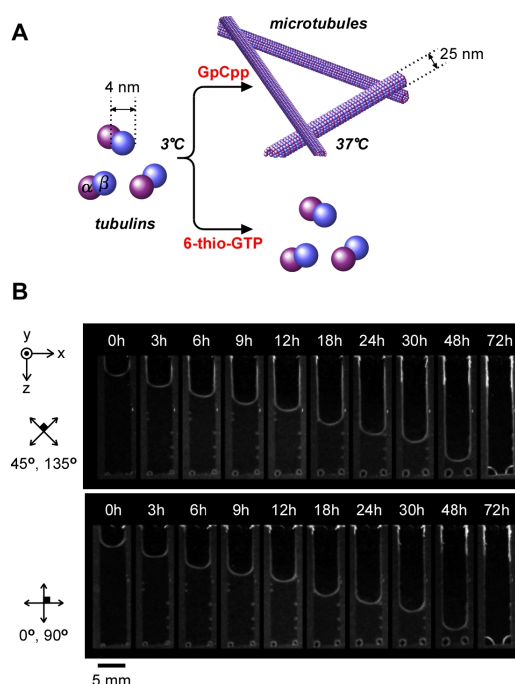


Figure S7. **A.** Control of tubulin/MT polymerization by GpCcp and 6-thio-GTP. **B.** Drying process of tubulin solution including 6-thio-GTP. Initial concentration of tubulin: 0.5 wt%. Drying temperature: 37°C.

Control of the tubulin/microtubules polymerization^[6-8] was carried out to clarify the effect of the polymerization on the orientation.

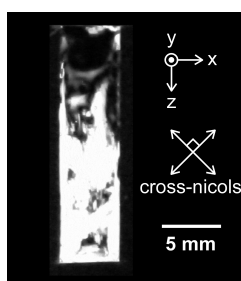


Figure S8. DNA solution in TE buffer at ~25°C. Initial concentration: 4 wt%.

References

- [1] M. K. Okajima, T. Bamba, Y. Kaneko, K. Hirata, E. Fukusaki, S. Kajiyama, T. Kaneko. *Macromolecules* **41**, 4061(2008).
- [2] M. K. Okajima, Q. T. Nguyen, S. Tateyama, H. Masuyama, T. Tanaka, T. Mitsumata, T. Kaneko. *Biomacromolecules* **13**, 4158 (2012).
- [3] A. C. S. Alcantara, M. Darder, P. Aranda, S. Tateyama, M. K. Okajima, T. Kaneko, M. Ogawa, E. Ruiz-Hitzky. *J. Mater. Chem. A* **2**, 1391 (2014).
- [4] M. K. Okajima, R. Mishima, K. Amornwachirabodee, T. Mitsumata, K. Okeyoshi, T. Kaneko. *RSC Advances* **5**, 86723 (2015).
- [5] K. Amornwachirabodee, M. K. Okajima, T. Kaneko. *Macromolecules* **48**, 8615 (2015).
- [6] K. Okeyoshi, R. Kawamura, R. Yoshida, Y. Osada. *J. Mater. Chem. B* **2**, 41 (2014).
- [7] K. Okeyoshi, R. Kawamura, R. Yoshida, Y. Osada. *Chem. Commun.* **51**, 11607 (2015).
- [8] K. Okeyoshi, R. Kawamura, R. Yoshida, Y. Osada. *Sci. Rep.* **5**, 9581 (2015).