

## ***Supporting Information for***

### **Crystallographic interdigitation in oyster shell folia enhances material strength**

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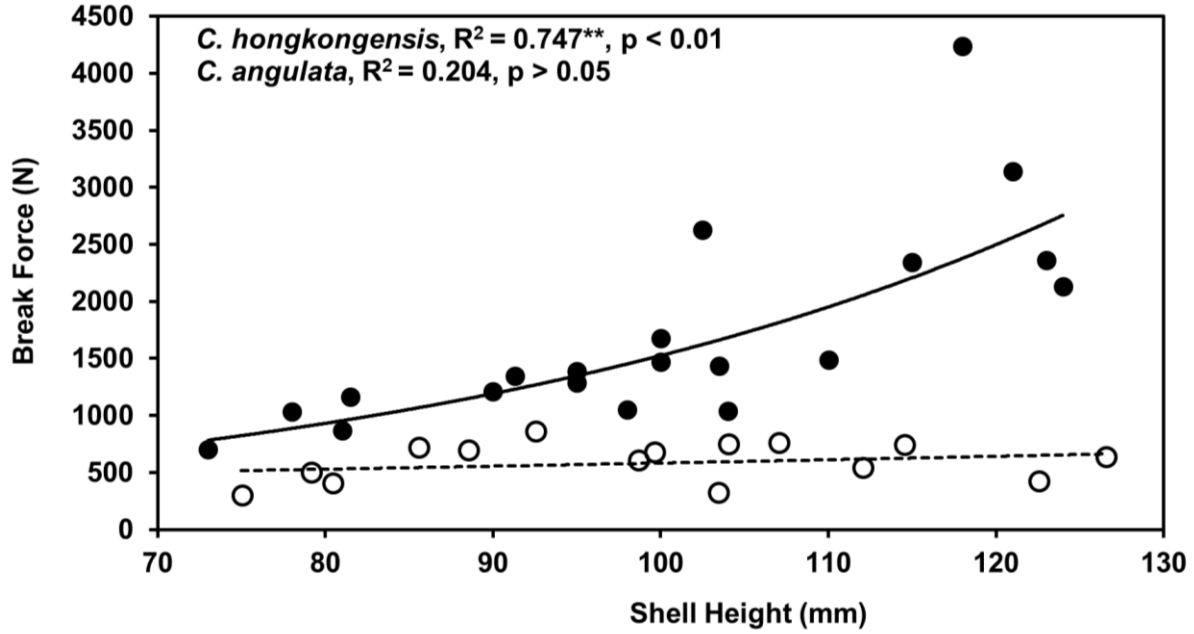
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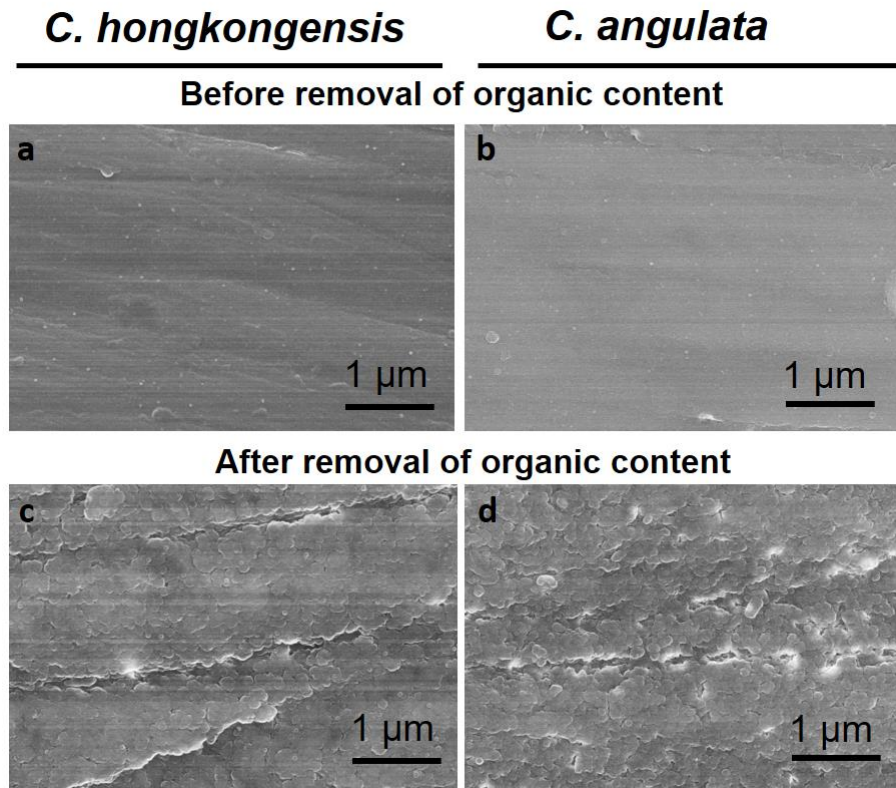
**Entire shell compression resistance.** Entire oyster shells of *C. hongkongensis* and *C. angulata* were prepared for compression testing by washing with water and brushing to remove the tissue and the commensal and then drying in an oven for 48h at 60°C. The shell height and the distance from the umbo to bill, was measured using calipers (mm). To measure compressive resistance as a function of height, the shells within the range of height of 70 mm to 130 mm were selected for compression testing. The compressive force (N) was measured using a load compression machine (MTS QTest/25) with flat load cell. In order to mimic live oysters in nature, both valves of each shell were realigned and placed between two flat compression anvils for compression testing. The force increased with the compressive displacement until reaching a peak load and then dropping when the shell cracked. This peak load was assessed as compressive resistance. Additionally, the cracks of all samples were further examined to identify those that went through all layers of at least one valve of the paired valves as a validation check. The relationship between shell height and compressive strength was also investigated using correlation analysis with 20 shells of *C. hongkongensis* and 15 shells of *C. angulata*. In Figure S1, a significant positive correlation between shell height and break force was observed in the shells of *C. hongkongensis*. However, there is no significant correlation between shell height and compressive strength in *C. angulata* shells. Importantly, *C. hongkongensis* shells are able to withstand a significantly higher compressive force ( $1700 \pm 872.65$  N) than *C. angulata* shells ( $607 \pm 171.71$  N).



**Figure S1.** Compression test results for shell height versus the break force for entire paired valve oyster shells of *C. hongkongensis* (filled circles) and *C. angulata* (open circles).

**The removal of organic content.** Five folia cubes of each species were examined, before and after immersing in sodium hypochlorite for 2 hours, under scanning electron microscopy (SEM) to confirm the removal of organic content (Figure S2) according to the previous study.<sup>1</sup> Folia cubes were glued to scanning electron microscopy stubs and carbon-coated. Secondary electron images were collected with a beam voltage of 20kV at a working distance of 9 mm in high vacuum mode on a scanning electron microscope (SEM; Hitachi S-3400N VP SEM, Hitachi, Japan). In Figure S2, the organic content was removed from the folia cubes as indicated by the appearance of pores of a fixed size within the surface of the folia cube, consistent with the results of a previous study that characterized the folia after removal of organic content by thermal treatment for Pacific oyster (*C. gigas*).<sup>1</sup> The pores of folia of *C. angulata* are larger and more numerous than those of *C.*

*hongkongensis* (Figure S2c and d) which is coincident with the higher organic content determined by the weight loss percentage of the folia after ignition (Figure 4a).



**Figure S2.** Ultrastructure of folia before and after removal of organic content in *Crassostrea hongkongensis* and *Crassostrea angulata*. Secondary electron images of the surface of folia before removal of organic content in (a) *C. hongkongensis* and (b) *C. angulata*. Secondary electron image of the surface of folia after removal of organic content in (c) *C. hongkongensis* and (d) *C. angulata*.

## ■ REFERENCES

- (1) Lee, S. W.; Kim, Y. M.; Kim, R. H.; Choi, C. S. Nano-structured biogenic calcite: A thermal and chemical approach to folia in oyster shell. *Micron* **2008**, 39 (4), 380-6.