**Supplementary Figure 1. Stereoscopic reconstruction of a fractured section of a C. pyramidata shell.** (a) One of the two stereoscopic input SEM images that were used for stereoscopic reconstruction. (b) Reconstructed 3D model with greyscale from SEM images. (c) Reconstructed 3D model with height information.

**Supplementary Figure 2. Geometric analysis of fibrous building block cross sections.** (a) Artificially colored TEM image showing the mosaic interlocking cross sections (Figure 3d). (b) Traced individual cross sections. (c) Each pair shows the overlay of the traced cross sections of the entire set (top), left branched (τ-shaped, bottom left), and right branched (γ-shaped, bottom right). The left and right picture in each pair are an overlay of traced boundaries and semi-transparent geometries of the cross sections, respectively.
Supplementary Figure 3. X-ray diffraction pattern of *C. pyramidata* shell (referenced to standard aragonite peaks, PDF #00-041-1475).

Supplementary Figure 4. SEM image of a fractured surface of a *C. pyramidata* shell showing the variation in cross-sectional geometries along the axial direction of fibers (white arrows).
Supplementary Figure 5. (a) TEM images showing the morphology of organic inclusions under conditions of overfocus (left), underfocus (middle), and nearly in-focus (right). (b) TEM image of type ii inclusions which have a much more elongated and thin geometry as compared to type i inclusions.

Supplementary Figure 6. Dimensions and geometries of type i organic inclusions. (a) TEM image of a cross-section of the building blocks showing two types (i and ii) of intracrystalline organic inclusions (Figure 6d). (b) High resolution TEM image showing the crystal lattice of aragonite and type i inclusions. (c) Overlay of traced type i inclusions based on TEM images with a zone axis of [-1-10]. (d) Average inclusion size and geometry derived from the data shown in (c).
Supplementary Figure 7. (a) TEM images of fragments from isolated individual building blocks and (b) cross-sectional sample prepared using focused ion beam milling showing multiple twinning boundaries.

Supplementary Figure 8. SEM image of a fractured section which shows that the peripheral regions towards the outer surface of shell (positive N) have rougher fractured surfaces compared to those from the inner side of each building blocks. This fracture behavior might result from the high abundance of organic inclusions in the outer region of each building block.
**Supplementary Figure 9.** Microscopic optical mappings at wavelength of 600 nm for transmission (a) and reflection (b). A modified Leica DMRX microscope was used to carry out parallel optical imaging and spectroscopic spatial optical mapping with step size of 1 µm. (c) Representative transmission and reflection spectra from a *C. pyramidata* shell taken at the point indicated by the crosses in (a) and (b).