

[Supplementary Materials]

## Bio-inspired nacre-like PEEK material with superior tensile strength and toughness

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### Relationship of processing parameters (e.g., temperatures and pressure) and compression ratios

**Table S1** Compression ratio of samples during PIF-processing as functions of temperature and pressure.

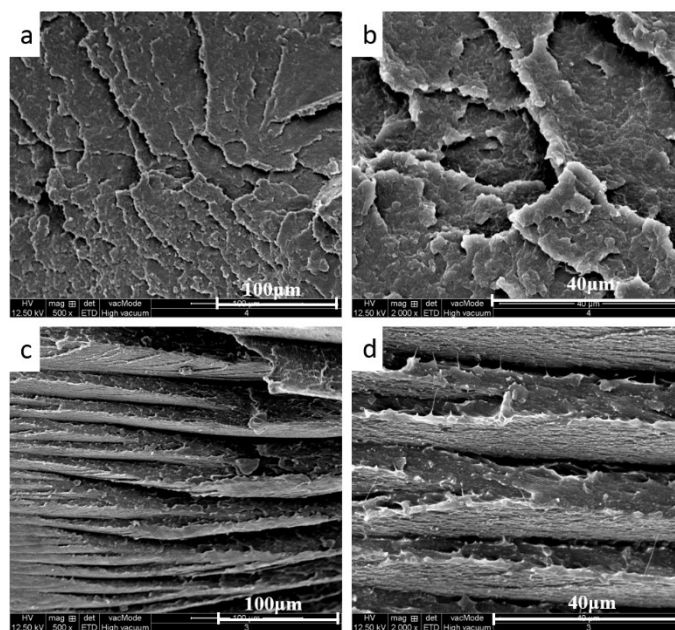
Temperature (°C)	Pressure (MPa)	compression ratios
280	0	1.0
280	50	1.4
280	100	1.6
280	200	1.8
280	350	2.2
280	650	2.6
220	500	1.7
240	340	1.7
260	230	1.7
280	150	1.7
300	100	1.7

Table S1 shows the macroscopic compression ratio of PEEK materials formed by PIF-processing at different temperatures and pressures. In general, higher temperature or pressure leads to greater compression ratio. However, they do not follow a simple linear relation. When the pressure was rather small, increasing the pressure is very effective

to improve the compression ratio. On the other hand, in order to obtain the same compression ratio, higher pressures are required at low temperatures.

### Fracture surface morphologies after impact tests for PEEK samples

A closer look at the fracture surface morphologies can show the PIF induced nacre-like structures more clearly. Figure S1 presents the SEM images of fractured surfaces after impact tests for PEEK samples. Discontinuities could be seen between neighboring spherulites in the same layer (Figure S1c), indicating the interlocking between these deformed spherulites. The discontinuities were similar to those found in nacre (Figure 3F), but not that regular. The spherulites deformed and aligned in the pressure-induced flow direction to form parallel layers no thicker than 5  $\mu\text{m}$  and show a nice orientation of cracks perpendicular to LD, and protrusions and grooves due to the pull-out of spherulites from opposite surfaces were clearly observed along FD (Figure S1d). We speculate that the zigzag arrangement of deformed spherulites contributed to the tortuous energy dissipating paths and resulted in improved toughness, while the alignment and stretching of the amorphous regions between lamellae and spherulites are responsible for the large elongation at break.



**Figure S1** SEM images of fractured surfaces after impact tests for PEEK samples: (a, b) without PIF-processing; (c, d) after PIF-processing.