

## Supplementary Information

### Local strain calculation

The plastic FEA utilized a beam-solid element assembly (Figure S1a). Solid elements were used in the middle of the coiling loop in order to avoid the convergence difficulty caused by beam elements under large inelastic deformation due to torsional loading<sup>1</sup>. Since the solid element does not provide direct cross-sectional outputs for curvature, torsional strain and axial strain along the fiber like the beam element does, we extract nodes displacements from each load substep in the simulation and calculate local strains along the fiber during the unfolding process. Results at the unfolding percentage of 80% are shown here as an example. For the curvature, the trace of center nodes (Figure S1a) in solid elements is rebuilt in MATLAB. A circle is fitted to every three adjacent points along the trace<sup>2</sup>. The apparent curvature  $\kappa_a$  is calculated as the inverse of the radius of the circle. The bending curvature  $\kappa$  (Figure S1b, red markers) which correlates with the bending stress is calculated as  $\kappa = \kappa_a - \kappa_i$ , where  $\kappa_i$  is the corresponding initial curvature of the fiber center line. The as-calculated  $\kappa$  is equivalent to  $\sqrt{\kappa_Y^2 + \kappa_Z^2}$  (Figure S1b, blue markers), where  $\kappa_Y$  and  $\kappa_Z$  are element outputs for the bending curvature about the Y and Z centroid axis of the beam element. The shear strain  $\gamma$  is calculated based on the corner angle of the surface mesh (Figure S1a):  $\gamma = \theta_d - \theta_i$ , where  $\theta_d$  is the angle on the deformed mesh, and  $\theta_i$  is the angle on the initial mesh. The shear strain at each  $L_a$  is averaged by 40 values around the circumference (Figure S1c). The as-calculated  $\gamma$  is equivalent to  $r\gamma_{TE}$  (Figure S1c, blue markers), where,  $r$  is the fiber radius,  $\gamma_{TE}$  is the element output of torsional strain from the beam element. The axial strain is calculated as  $\varepsilon = (l_d - l_i)/l_i$ , where  $l_d$  is the length between two adjacent center nodes after deformation,  $l_i$  is the initial length. The as-calculated  $\varepsilon$  is equivalent to the beam element output of axial strain. Discontinuities exist in all three local strain results at the transition from beam element to solid element. This is caused by the rigid surface constraint which is necessary to assemble beam and solid element.

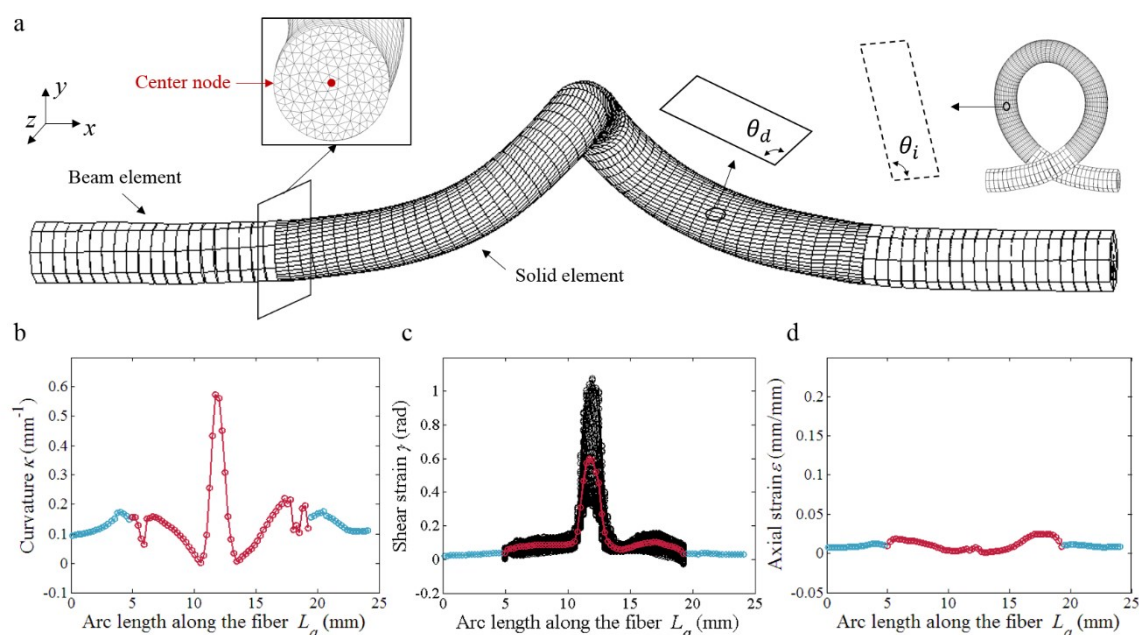


Figure S1. Local strain results calculated from beam and solid elements at the unfolding percentage of 80%: a) deformed meshes of beam and solid elements; b) curvature, c) shear strain, and d) axial strain

calculated from beam (blue markers) and solid (red markers) elements. The shear strains in solid elements are averaged over 40 values (black markers) around the circumference at each  $L_a$ .

### Multilinear plastic FEA

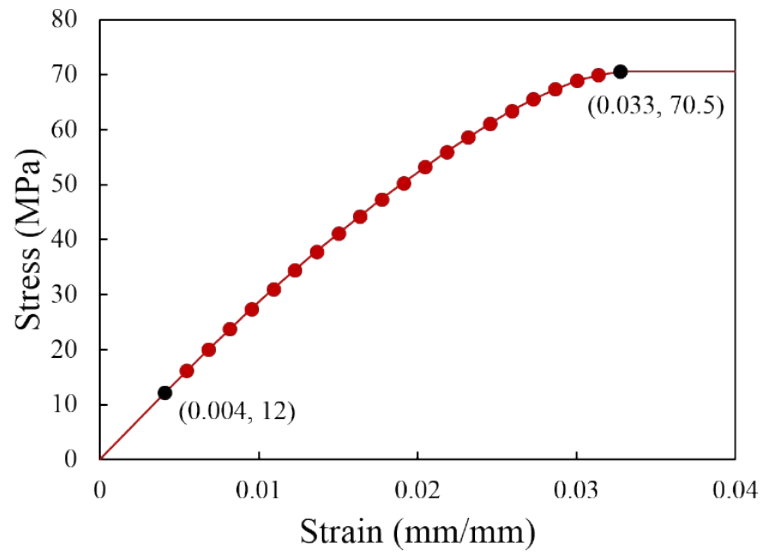


Figure S2 Piece-wise linear stress-strain curve for multilinear isotropic hardening in the plastic FEA. The first stress-strain point defines the yield stress (12 MPa). Subsequent points define the multilinear isotropic hardening behavior of the material. The last point corresponds to the fracture of the fiber in tensile test, while in the plastic FEA, the fiber behaves perfectly plastically after the material reaches the tensile strength (70.5 MPa). The data points are collected from one of the uniaxial tensile tests of straight PLA fiber with the diameter of 1.20 mm (gauge length = 250 mm, strain rate = 0.01 mm/mm/s). The tensile strength with a standard deviation for 7 specimens is  $71.3 \pm 0.95$  MPa.

### Tensile curves of straight fibers

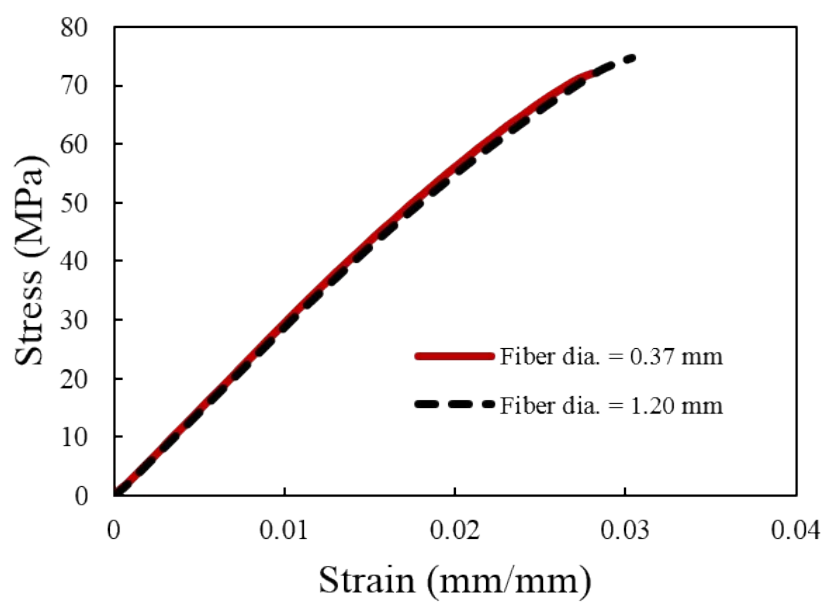


Figure S3 Stress-strain curves of straight fibers under uniaxial tension. Gauge length is 100 mm and crosshead rate is 500 mm/min for both fibers.

## Strain rate estimation

Von Mises strain  $\epsilon_{VM}$  (Figure S4a, blue solid line) at the top surface in the middle of the unfolding loop is extracted from the plastic FEA. In order to have the von Mises strain rate  $\dot{\epsilon}_{VM}$  (Figure S4a, red dashed line), we first calculate the equivalent elapsed time  $t$  after the breaking of sacrificial bond in the simulation, based on the pulling end displacement in the simulation and actual crosshead speed (8.33 mm/s with an initial acceleration of 52 mm/s<sup>2</sup>) in the uniaxial tensile test. Then the local strain rate  $\dot{\epsilon}_{VM}$  is obtained by differentiating  $\epsilon_{VM}$  over the elapsed time  $t$ . The strain rate reaches the peak of 0.33 mm/mm/s at  $p = 35\%$ , around which the bending crack initiates at the top middle of the loop (Figure 4a). This peak strain rate is used for both the large and small fibers in the three-point bending test. We approximate the strain rate in dynamic failure by the same method. In order to calculate the instantaneous speed after the breaking of sacrificial bond in dynamic failure, we track the adjacent bond in the traction direction from the high speed camera captures (Figure S4b) and calculate the average pulling speed of the unfolding loop between each frame. The acceleration ( $1.327 \times 10^5$  m/s<sup>2</sup>) was obtained by a linear fit (Figure S4c), based on which, we calculate the equivalent elapsed time  $t_d$  after the breaking of sacrificial bond in the simulation. The von Mises strain rate (Figure S4d, orange dashed line) in dynamic failure is obtained by differentiating the von Mises strain (Figure S4d, blue solid line) over the elapsed time  $t_d$ . The fiber backbone breaks in the middle of the loop within 99  $\mu$ s after the breaking of sacrificial bond (Figure S4b). The strain rate at the top middle of the loop is 274 mm/mm/s (Figure S4d) at  $t_d = 99$   $\mu$ s. With the acceleration unchanged, the strain rate at the top middle of the loop would reach the peak of 1492 mm/mm/s at  $t_d = 235$   $\mu$ s if the fiber backbone did not break.

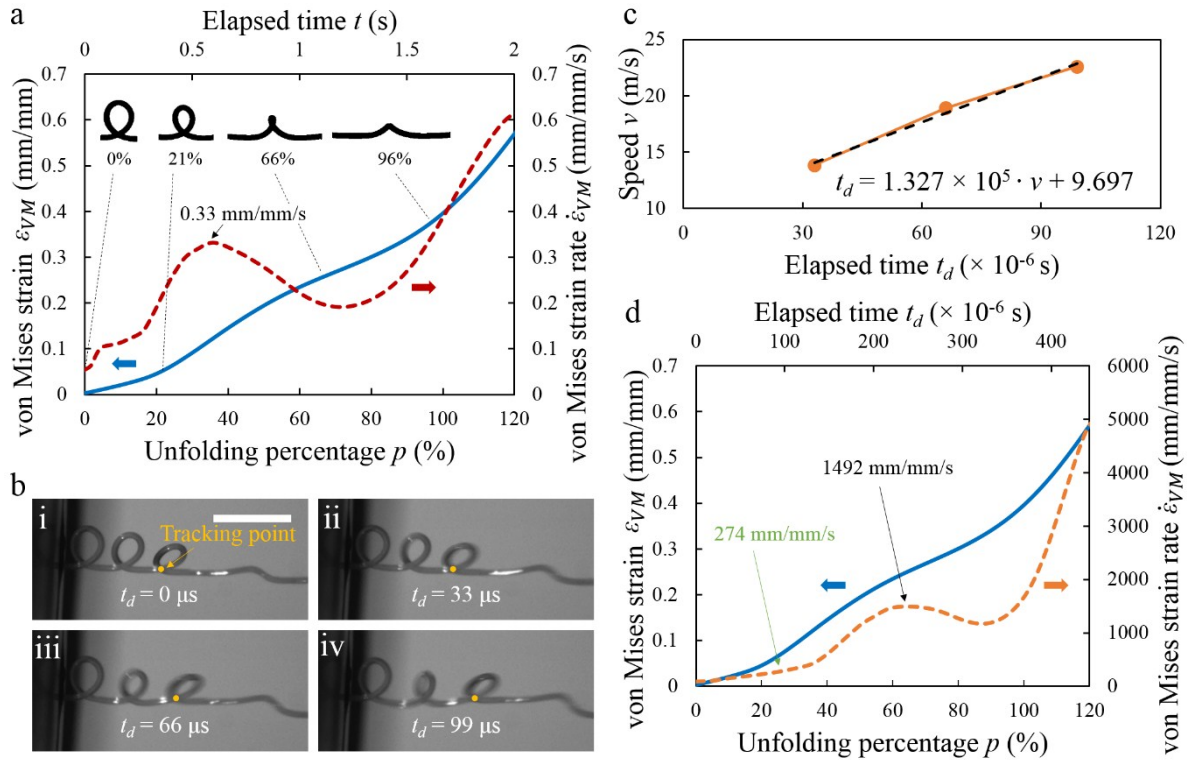


Figure S4. Estimation of the local strain rate at the top surface in the middle of the unfolding loop: a) von Mises strain (blue solid line, left Y axis) and von Mises strain rate (red dashed line, right Y axis) at the top surface in the middle of the unfolding loop, during the normal unfolding process of the loop without dynamic failure. The bottom and top X axis represent the unfolding percentage and elapsed time,

respectively. They correspond to each other and serve as different references for the unfolding process.

b) High speed imaging of the loop retraction after the breaking of sacrificial bond in dynamic failure. The speed of the loop retraction at  $t_d = 33 \mu\text{s}$ ,  $66 \mu\text{s}$  and  $99 \mu\text{s}$  is calculated by measuring the moving distance of the tracking point in ii – iv relative to its initial position at  $t_d = 0 \mu\text{s}$  in i. Scale bar is 5 mm.

c) Linear fit of the loop retraction speed over elapsed time. d) von Mises strain (blue solid line, left  $Y$  axis) and von Mises strain rate (orange dashed line, right  $Y$  axis) at the top surface in the middle of the unfolding loop, during the rapid unfolding process of the loop in dynamic failure. Simulation results, such as the von Mises strain and strain rate, are extracted from 101 load substeps and plotted as continuous lines here for the sake of visual demonstration.

### DSC test of large and small fibers

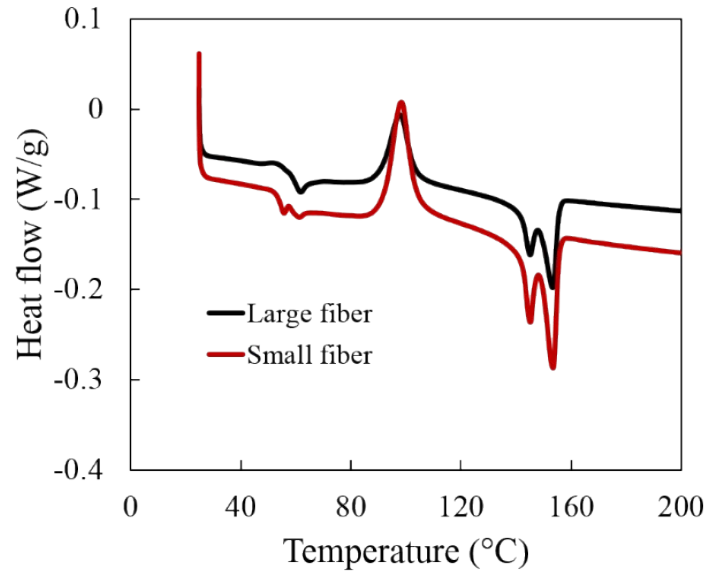


Figure S5. DSC results of FDM-extruded PLA fibers. The fibers were extruded at 230 °C on the conveyor belt in ambient air with two nozzles with different diameters (1 mm and 0.3 mm). The belt speed is equal to the extruding speed. The large and small fibers are 1.2 mm and 0.37 mm in diameter, respectively.

Thermal analysis of PLA fibers was performed in a DSC instrument (DSC Q2000) with the heating program found in the literature<sup>3</sup>. The temperature was first held at 25 °C for 3 min, and then increased to 200 °C with a rate of 2 °C/ min. The samples were cut from the same batch of straight fibers for the three-point bending test. The sample weight was 5.8 mg for the large fiber and 5.2 mg for the small fiber. The degree of crystallinity  $X_c$  of the specimen was calculated by the following equation:

$$X_c = \frac{\Delta H_m - \Delta H_c}{\Delta H_{100}} \times 100$$

where,  $\Delta H_m$  is the enthalpy of fusion,  $\Delta H_c$  is the enthalpy of cold crystallization,  $\Delta H_{100}$  is fusion enthalpy of 100% crystalline PLA, which is 93 J/g<sup>3</sup>.

The calculated crystallinity is approximately 2.5% for the large fiber and 1.1% for the small fiber, which are around the crystallinity of as-received PLA filament (2.4%) reported in the literature<sup>3</sup>.

## Toughness enhancement

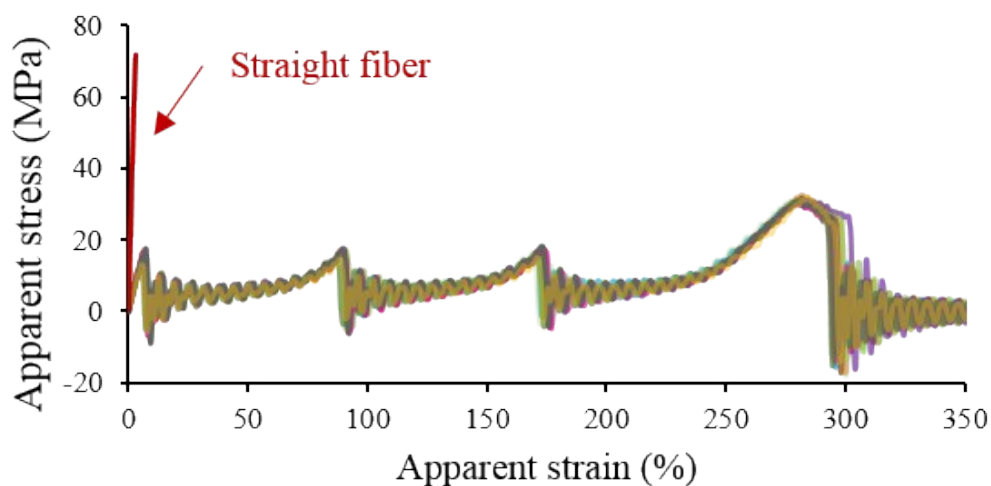


Figure S6 Apparent stress-strain curves of small fiber with three coiling loops. All ten test specimens are plotted to show the deviation. The stress-strain curve of the straight small fiber is also plotted as a benchmark to show the toughness enhancement in coiling fibers.

Table S1 Toughness values of the small fiber with three coiling loops. The benchmark toughness value of the straight fiber is 1.1 kJ/kg.

	Toughness (kJ/kg)		
	Average	Maximum	Minimum
3 coiling loops (small fiber)	5.51	5.86	5.15
%Benchmark	500%	533%	468%

## References

1. *ANSYS Mechanical APDL Element Reference (Release 15.0)*, ANSYS Inc., Canonsburg, PA, 2013.
2. J. Korsawe, *Circlefit3d - fit circle to three points in 3d space*, MATLAB Central File Exchange. Retrieved March 10, 2017.
3. M. Ivey, G. W. Melenka, J. P. Carey and C. Ayranci, *Advanced Manufacturing: Polymer & Composites Science*, 2017, **3**, 81-91.