

Supporting Information for

## Super-strong and Intrinsically Fluorescent Silkworm Silk from Carbon Nanodots Feeding

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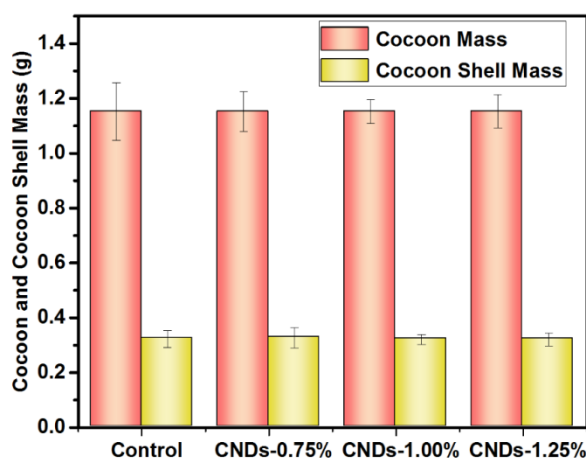
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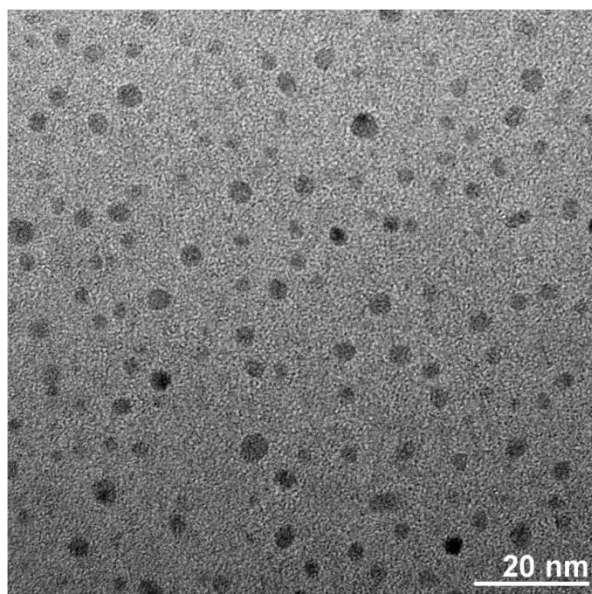
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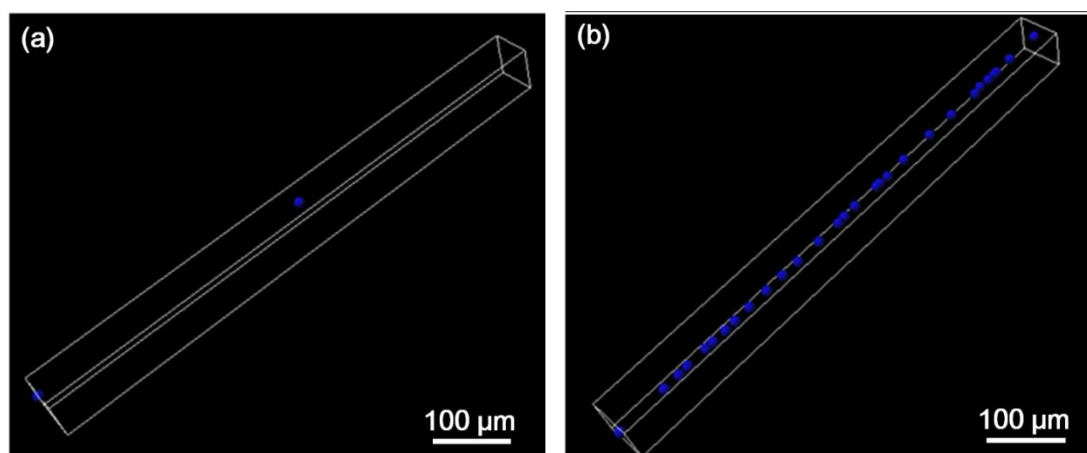
### Supplementary Figures



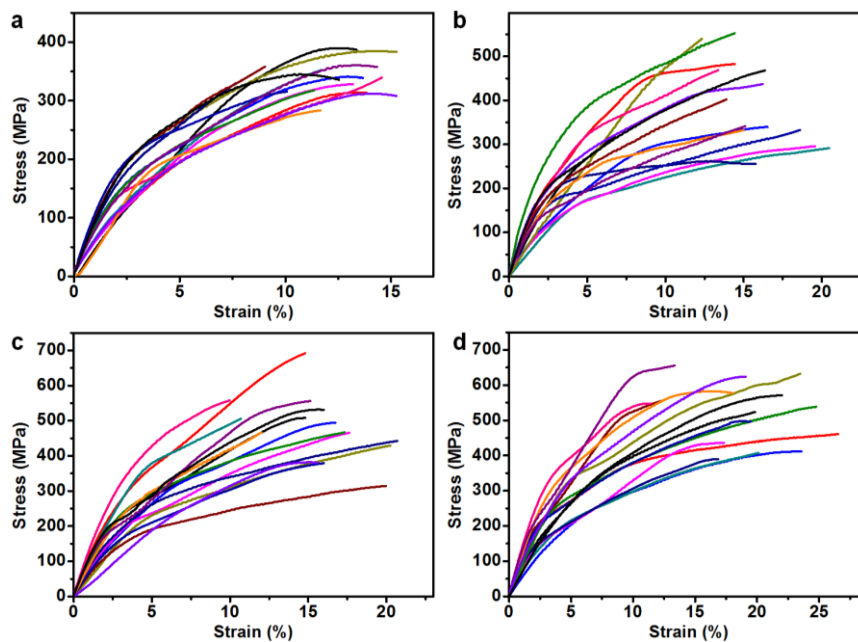
**Fig. S1** The mass of single cocoon and cocoon shell. All the cocoons have the similar mass, indicating that the addition of CNDs in the diets is safe for silkworm.



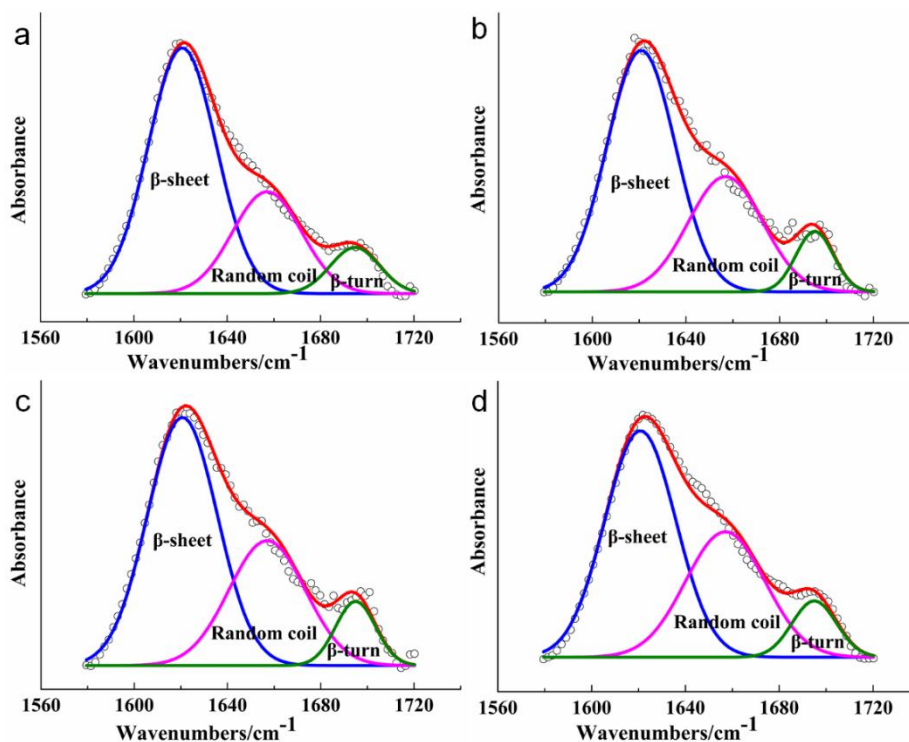
**Fig. S2** TEM image of CNDs. The CNDs have diameters ranging from 1 to 5 nm, and separated from each other, illustrating good dispersibility in aqueous solution



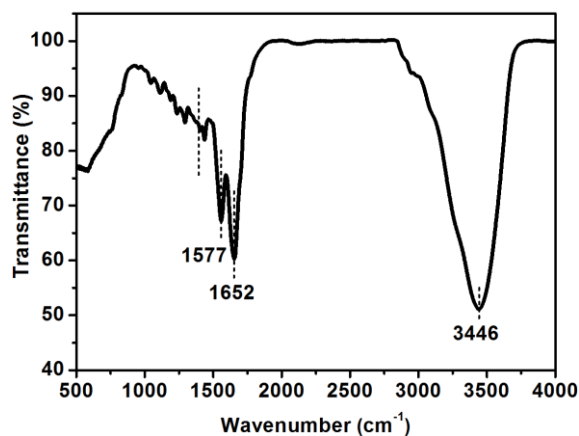
**Fig. S3** 3D reconstructed CLSM images of **a** control and **b** modified degummed silk. These results were consistent with 2D CLSM images (Fig. 2) and suggested the fluorescence uniformity of modified silk.



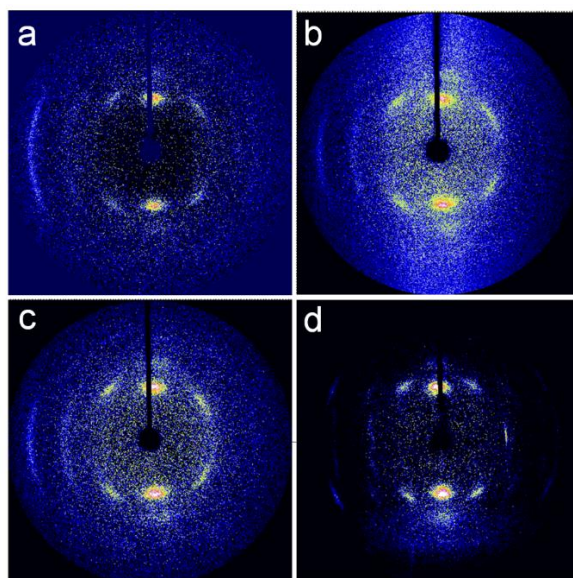
**Fig. S4** Stress-strain curves of different silk fibers: **a** Control, **b** CNDs-0.75%, **c** CNDs-1.00%, **d** CNDs-1.25%. Compared to control silks, the breaking strength and elongation of CNDs modified silks increased significantly. In addition, the mechanical properties reinforced gradually with increasing the addition of CNDs.



**Fig. S5** The deconvolution of FTIR spectra in amide I band of different silk fibers. **a** Control, **b** CNDs-0.75%, **c** CNDs-1.00%, **d** CNDs-1.25%



**Fig. S6** FTIR spectra of CNDs. The absorption peaks at 1577, 1652, and 3446  $\text{cm}^{-1}$  were ascribed to the bending vibrations of N-H, asymmetric stretching vibration of C=O, and stretching vibration of C-OH [S1, S2]. This indicated that there were abundant carboxyl and hydroxyl on the surface of CNDs.



**Fig. S7** Two-dimensional SR-WAXD patterns of different silk fibers. **a** Control, **b** CNDs-0.75%, **c** CNDs-1.00%, **d** CNDs-1.25%

## Supplementary References

[S1] S. Zhu, Q. Meng, L. Wang, J. Zhang, Y. Song et al., Highly photoluminescent carbon dots for multicolor patterning, sensors, and bioimaging. *Angew. Chem. Int. Ed.* **52**, 3953-3957 (2013). <https://doi.org/10.1002/anie.201300519>

[S2] S. Lu, L. Sui, J. Liu, S. Zhu, A. Chen, M. Jin, B. Yang, Near-infrared photoluminescent polymer-carbon nanodots with two-photon fluorescence. *Adv. Mater.* **29**, 1603443 (2017). <https://doi.org/10.1002/adma.201603443>