

Water-responsive *Bombyx Mori* Silk for High-efficiency Actuators

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Water-responsive (WR) material that mechanically swells and shrinks in response to changes in relative humidity (RH) has shown a potential to serve as high-energy actuating components for various engineering applications. Nature has developed remarkable WR materials that contain both stiff nanocrystalline domains and soft amorphous domains that can be crucial to their WR behaviors. This work reports that regenerated *Bombyx (B.) mori* silk can be processed to increase β -sheet crystallinity, which dramatically increases the WR energy density to 1.6 MJ m⁻³, surpassing that of all known natural muscles. When the silk's β -sheet crystallinity increases from 19.7% to 57.6%, silk's WR energy density shows an eightfold increase, suggesting that high crystallinity of silk dramatically reduces energy dissipation during the hydration/dehydration processes. Moreover, the availability and processability of *B. mori* silk opens up possibilities for simple and scalable modification and production of powerful WR actuators.