Supplementary information

Tailor-Made Spider-Eggcase-Silk Spheres for Efficient Lysosomal Drug Delivery

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Figure S1. Gene recombinant of spider eggcase silk proteins. A, complete gene sequence; B, plasmid design; C, verification of targeted plasmid; D, verification of tubuliform spidroins; E, Similarity comparison between the wild type and engineered one.
**Figure S2.** Preparation process in different lubricant substrates. A) High-molecule-weight silicon oil; B) Low-molecule-weight silicon oil.

**Figure S3.** Selected ATR-FTIR spectra of original silk spheres deduced after Fourier self-deconvolution. Within amide I band, the contributions of β-sheets, random coil/α-helix and β-turn are represented.
**Figure S4.** Nanoindentation curve of a single silk sphere.

**Figure S5.** Cytotoxicity of pure eTuSp1 spheres.
Figure S6. The standard concentration plot of the Dox. The absorbance of each sample is measured at the wavelength of 490 nm by UV-Vis spectrometry.

Table S1. Physicochemical properties of eTuSp1 spheres.

<table>
<thead>
<tr>
<th>Size (nm)</th>
<th>Polydispersity index</th>
<th>Zeta potential (mV)</th>
<th>Mobility (cm²/Vs ± SD)</th>
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<tbody>
<tr>
<td>eTuSp1</td>
<td>183</td>
<td>0.214</td>
<td>-33.5 ± 4.2</td>
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