Supporting Information

Charge-reversible and pH-responsive biodegradable micelles and vesicles from linear-dendritic supramolecular amphiphiles for anticancer drug delivery

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**Scheme S1** Synthetic Routes of (A) Acetal Monomer AEEEMA and (B) Ada-PEG.

**A**

![Chemical reaction diagram]

**B**

![Chemical reaction diagram]

**Fig. S1** $^1$H NMR spectra of (A) VEA and (2) AEEEMA.
Fig. S2 MALDI-TOF MS of CD-G0.

Fig. S3 Detailed $^1$H NMR spectrum of CD-G1.
**Fig. S4** Detailed $^1$H NMR spectrum of CD-G2.

**Fig. S5** Detailed $^1$H NMR spectrum of CD-G3.
**Fig. S6** MALDI-TOF MS of CD-G1.

**Fig. S7** GPC traces of (A) the polyacetal dendrimers with a CD core and (B) the adamantane-terminated poly(sulfobetaine).
Fig. S8 $^1$H NMR spectra of (A) Ada-Br and (B) Ada-PEG.

Fig. S9 2D NOESY spectrum of Ada-PEG and CD-G1. The red rectangle showed the correlational peaks between the protons of adamantane groups and the internal protons of CDs.
Fig. S10 DLS profiles of (A) the CD-G1/Ada-PSB$_{40}$ micelles and (B) the CD-G2/Ada-PSB$_{40}$ vesicles after being incubated at pH=7.4 for 12 h.

Fig. S11 (A) TEM images and (B) DLS analysis of (a) DOX@CD-G1/Ada-PSB$_{40}$ and (b) DOX@CD-G2/Ada-PSB$_{40}$ nanoparticles.
**Table S1** MWs of and PDIs of the polyacetal dendrimers with a CD core and the adamantane-terminated poly(sulfobetaine)

<table>
<thead>
<tr>
<th>Sample</th>
<th>$M_n, \text{calc.}$</th>
<th>$^aM_n, \text{GPC}$</th>
<th>$^aM_w, \text{GPC}$</th>
<th>$^a$PDI</th>
</tr>
</thead>
<tbody>
<tr>
<td>CD-G1</td>
<td>5986</td>
<td>6420</td>
<td>6930</td>
<td>1.08</td>
</tr>
<tr>
<td>CD-G2</td>
<td>14736</td>
<td>15300</td>
<td>17100</td>
<td>1.12</td>
</tr>
<tr>
<td>CD-G3</td>
<td>32236</td>
<td>30600</td>
<td>35200</td>
<td>1.15</td>
</tr>
<tr>
<td>Ada-PSB$_{40}$</td>
<td>—</td>
<td>5170</td>
<td>6620</td>
<td>1.28</td>
</tr>
<tr>
<td>Ada-PSB$_{80}$</td>
<td>—</td>
<td>9620</td>
<td>12800</td>
<td>1.33</td>
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</tbody>
</table>

$^a$MWs and PDIs determined by GPC.

**Table S2** Characterizations of the DOX-loaded micelles and vesicles

<table>
<thead>
<tr>
<th>Nanoparticle</th>
<th>$^a$Size (nm)</th>
<th>$^a$PDI</th>
<th>DLC (wt%)</th>
<th>DLE (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>DOX@CD-G1/Ada-PSB$_{40}$</td>
<td>191</td>
<td>0.106</td>
<td>5.73</td>
<td>57.3</td>
</tr>
<tr>
<td>DOX@CD-G2/Ada-PSB$_{40}$</td>
<td>231</td>
<td>0.119</td>
<td>7.22</td>
<td>56.1</td>
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</tbody>
</table>

$^a$Sizes and PDIs determined by DLS.