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## Supplementary information for Synthesis of comb-like poly(ethyleneimine)s and their application in biomimetic silicification

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Fig. S1 <sup>1</sup>H NMR spectrum of crude polymerization product of CMS monomer before precipitation.

In detail, the conversion was calculated according to the relative integral area (S) by following formula:

%Conv. =  $S_{\text{poly.}}/(S_a + S_{\text{poly.}}) *100\% = 63.7\%$ , here,  $S_{\text{poly.}} = [(S_4 + S_3 + S_b) - 0.5S_a]/2$ .

Combined with the initial molar ratio of CMS monomer and RAFT agent CDB (100:1), the polymeric degree of PCMS was denoted as 64.



**Fig. S2** <sup>1</sup>H NMR spectrum of crude polymerization product of MOZ (red line). In comparison, we also gave <sup>1</sup>H NMR data of pure MOZ monomer (black line). The proton signal disappeared in dash line part (red line), which indicated that the MOZ monomer reacted completely in this polymerization system.



**Fig. S3** <sup>1</sup>H NMR spectra of PCMS initiator and P(CMS-*g*-PEI<sub>69</sub>)<sub>64</sub> comb polymer. The proton signal b of initiating group (-CH<sub>2</sub>Cl at 4.5 ppm) nearly completely disappeared after polymerization reaction for MOZ monomer.