Peptoid Synthesis
All peptoids were synthesized on a commercial Aapptec Apex 396 robotic synthesizer on using a solid-phase submonomer cycle as described previously. All amine submonomers and other reagents used for our peptoid synthesis are obtained from commercial sources and used without further purification. Rink amide resin (0.60 mmol/g, Novabiochem, Cat No. 01-64-0013) was used to generate C-terminal amide peptoids. In this method, the Fmoc group on the resin was deprotected by adding 2 mL of 20% (v/v) piperidine/N,N-dimethylformamide (DMF), agitating for 20 min, draining, and washing with DMF. All DMF washes consisted of the addition of 1 mL of DMF, followed by agitation for 1 min (repeated five times). An acylation reaction was then performed on the amino resin by the addition of 1.0 mL of 1.2M bromoacetic acid in DMF, followed by 0.18 mL of N,N-diisopropylcarbodiimide (DIC, 1.15 mmol, neat). The mixture was agitated for 20 min at room temperature, drained, and washed with DMF. Nucleophilic displacement of the bromide with various primary amines occurred by a 1.0 mL addition of the primary amine monomer as a 1.0 - 1.5 M solution in N-methyl-2-pyrrolidone (NMP), followed by agitation for 60 min at room temperature. The monomer solution was drained from the resin, and the resin was washed with DMF as described above. The acylation and displacement steps were repeated until a polypeptoid of the desired length was synthesized. All reactions were performed at room temperature. All polypeptoids were acetylated on the resin after synthesis using a mixture (2.0 mL per 100 mg of resin) of 0.4 M acetic anhydride and 0.4 M pyridine in DMF for 30 min, followed by washing with DMF. Peptoid chains were cleaved from the resin by addition of 4.0 mL 95% (v/v) trifluoroacetic acid (TFA) in water for 20 min, which was then evaporated off under a stream of nitrogen gas. Following cleavage, peptoids were dissolved in 4.0 mL mixture (v/v = 1:1) of water and acetonitrile for further purification.

All peptoids were purified by reverse-phase HPLC on a Vydac C4 column (10 µm, 22 mm × 250 mm), using a gradient of 5-95% acetonitrile in H₂O with 0.5% TFA over 60 min. All final products were analyzed by analytical reverse-phase HPLC (5 - 95% gradient at 1 mL/min over 30 minutes at 60°C with a C4, 5 µm, 4.6 × 150 mm column), and electrospray mass spectrometry (Agilent 1100 series LC/MSD trap system, Agilent Technologies, Santa Clara,CA) or matrix-assisted laser desorption/ionization mass spectrometry (Applied Biosystem/MDS SCIEX 4800 MALDI TOF/TOF Analyzer). The final peptoid products were lyophilized at least twice from their solution in mixture (v/v = 1:1) of water and acetonitrile. All lyophilized peptoids were finally divided into small portions (3.0 × 10⁻⁶ mol) and stored at -80°C.

Peptoid Sequences
Structures of the synthesized peptoids and molecular weight of each peptoid as determined by
mass spectrometry are show below. The following monomer abbreviations were used to name the sequences:

**Nce:** N-(2-carboxyethyl)glycine
**NXpe:** N-[2-(X-phenethyl)]glycine

\[(\text{NXpe})_4(\text{Nce})_8 \ (X = 4-H)\] Molecular weight: 1694.7 (Calculated), 1694.4 (Found).

\[(\text{NXpe})_4(\text{Nce})_8 \ (X = 4-\text{Cl})\] Molecular weight: 1832.5 (Calculated), 1831.6 (Found).

\[(\text{NXpe})_4(\text{Nce})_8 \ (X = 4-\text{OMe})\] Molecular weight: 1814.8 (Calculated), 1813.5 (Found).

\[(\text{NXpe})_4(\text{Nce})_8 \ (X = 2,4-\text{Dichloro})\] Molecular weight: 1970.3 (Calculated), 1969.0 (Found).

\[(\text{Nce})_4(\text{NXpe})_4(\text{Nce})_4 \ (X = 4-\text{Cl})\] Molecular weight: 1832.5 (Calculated), 1832.5 (Found).
(NceNXpe)$_6$ ($X = 4$-H) Molecular weight: 1758.9 (Calculated), 1757.9 (Found).

(NXpe)$_4$(Nce)$_{12}$ ($X = 4$-Cl) Molecular weight: 2349.0 (Calculated), 2348.2 (Found).

Preparation of peptoid stock solution
Lyophilized peptoids ($3.0 \times 10^{-6}$ mol) were mixed with 1.5 mL in deionized ($\geq 18$ MΩ) and filtered water (0.2 μm) water in glass vial, and 10 μl saturated (NH$_4$)$_2$CO$_3$ solutions were used to facilitate dissolution.

Crystal growth experiments
Crystallization of CaCO$_3$ was performed by slow diffusion of (NH$_4$)$_2$CO$_3$ vapor into 96-well plates in which each well has 190 μL of 5.0 mM CaCl$_2$ solution and 5.0 μL of 2.0 mM peptoid stock solution (Final peptoid concentration = 51.3 μM). For control experiments similar volumes of water were added. The 96-well plate was placed in a closed desiccator cabinet. All crystallization experiments were repeated three to five times. Crystals morphology were studied by optical microscopy (OLYMPUS, CKX41)

In situ AFM study of the calcite step growth
The dependence of molecular step speed on peptide concentration was measured by using in situ AFM to image growth on the (104) face of calcite in solutions at a fixed supersaturation, $\sigma$, of 0.14. The supersaturation is defined as $\sigma = \ln(\alpha_{Ca}^{2+}\alpha_{CO3}^{2-}/K_{sp})$, where $\alpha$ denotes the species activity, and $K_{sp}$ denotes the equilibrium solubility constant at 25°C. This supersaturated ($\sigma = 0.14$) calcium carbonate solution was made by directing mixing 10.5 mM NaHCO$_3$ with 0.34 mM CaCl$_2$ at equal volumes.

Natural calcite crystals were cleaved to produce fresh (104) faces as substrates for calcite growth. Calcite samples were used immediately upon cleaving after a brief cleaning with a nitrogen jet to remove any debris. Growth solutions were prepared immediately before use from reagent grade calcium chloride (CaCl$_2$·H$_2$O), and sodium bicarbonate (NaHCO$_3$) dissolved in deionized ($\geq 18$ MΩ) and filtered water (0.2 μm).

During calcite growth, the steady-state morphology of atomic steps was imaged at constant supersaturation ($\sigma = 0.14$) for all peptoids at various concentrations. Using established methods,
calcite was overgrown onto the surface of a calcite seed crystal in an AFM flow-through cell (50 µl) that continuously supplied the input solution at a rate greater than 30 ml/h via a syringe pump. These flow conditions insured that calcite growth was reaction and not transport limited as demonstrated in previous studies.\textsuperscript{2} Measurements of step speeds were conducted at room temperature with a Digital Instruments Nanoscope III or V(Veeco, Santa Barbara, CA) operating in Contact Mode. The AFM images were collected by using scan rates of 5 - 20 Hz and a resolution of 512 × 512, while minimizing tip-surface force interactions during the flow-through of the growth solutions to minimize artifactual effects on step edge morphology and measured velocities.\textsuperscript{3} Figure S2 is a representative \textit{in situ} AFM image to show the positive and negative steps on calcite (104) surface.

Figure S1. CaCO\textsubscript{3} crystals morphology in the presence of peptoids: (a) peptoid-1, and (b) peptoid-6.

Figure S2. \textit{In situ} AFM image to show the positive and negative steps on calcite (104) surface.
**Figure S3.** Measured enhancement of calcite step speed vs. peptoids concentration. Normalized propagation rates are shown as step velocity in the presence of peptoids, \( v \), over the step velocity without peptoids, \( v_0 \).

<table>
<thead>
<tr>
<th>Peptoid or Peptide</th>
<th>Molecular weight, g/mol</th>
<th>Step acceleration ((v/v_0))</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Peptoid-1</td>
<td>1695</td>
<td>( v/v_0 ): 22.25 at 50 nM; ( \sigma = 0.14 ) ( v/v_0 ): 21.48 at 50 nM; ( \sigma = 0.14 )</td>
<td>This paper</td>
</tr>
<tr>
<td>Asp6</td>
<td>708</td>
<td>( v/v_0 ): 0.84 at 50 nM; ( \sigma = 0.14 ) ( v/v_0 ): 0.88 at 50 nM; ( \sigma = 0.14 )</td>
<td>This paper</td>
</tr>
<tr>
<td>Asp6</td>
<td>708</td>
<td>( v/v_0 ): 1.15 at 100 nM; ( \sigma = 0.92 ) ( v/v_0 ): 1.44 at 100 nM; ( \sigma = 0.92 )</td>
<td>Reference 4</td>
</tr>
<tr>
<td>(Asp3Ser)Asp3</td>
<td>2957</td>
<td>( v/v_0 ): 1.64 at 100 nM; ( \sigma = 0.92 )</td>
<td>Reference 4</td>
</tr>
<tr>
<td>(Asp3Gly)Asp3</td>
<td>2777</td>
<td>( v/v_0 ): 1.6 at 100 nM; ( \sigma = 0.92 )</td>
<td>Reference 4</td>
</tr>
</tbody>
</table>

Table S1. Experimental measurements of step acceleration \((v/v_0)\) at low peptoid or peptide concentration under different supersaturated \((\sigma = 0.14 \text{ or } 0.92 \)) calcium carbonate solution


