Supporting information for:
Mineralization of Magnetic Nano-Tape in Self-organized Nanospace Composed of Nucleopeptide and Peptide

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S1. Synthesis strategy of the nucleopeptide.

Figure S1. Synthesis strategy of the nucleopeptide.
S2. $^1$HNMR spectra of the spacer peptide.

Intensity ratio of attributed protons.

<table>
<thead>
<tr>
<th>Spacer peptide</th>
<th>Intensity ratio</th>
<th>Experimental value</th>
<th>Calculated value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ac-(VE)$_3$-CONH$_2$</td>
<td>abeg : d</td>
<td>8 : 9.5</td>
<td>8 : 9</td>
</tr>
</tbody>
</table>

Figure S2. $^1$HNMR spectrum of the spacer peptide, Ac-(VE)$_3$-CONH$_2$. 
S3. $^1$HNMR spectrum of the peptide main chain of the nucleopeptide.

<table>
<thead>
<tr>
<th>peptide main chain of the nucleopeptide</th>
<th>Intensity ratio</th>
<th>Experimental value</th>
<th>Calculated value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ac-VEVS-(VE)$_7$-CONH$_2$</td>
<td>abg : d : h</td>
<td>32.9 : 56 : 2</td>
<td>35 : 54 : 2</td>
</tr>
</tbody>
</table>

Figure S3. $^1$HNMR spectrum of the peptide main chain of the nucleopeptide, Ac-VEVS-(VE)$_7$-CONH$_2$. 
S4. $^1$HNMR spectrum of the nucleopeptide.

Intensity ratio of attributed proton.

<table>
<thead>
<tr>
<th>Nucleopeptide</th>
<th>Intensity ratio</th>
<th>Experimental value</th>
<th>Calculated value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ac-VEVS(g(GC)$_3$(VE)$_7$-CONH$_2$</td>
<td>abegkq : lr</td>
<td>59.5 : 6</td>
<td>56 : 6</td>
</tr>
</tbody>
</table>

Figure S4. $^1$HNMR spectrum of the nucleopeptide, Ac-VEVS(g(GC)$_3$(VE)$_7$-CONH$_2$.
S5. Arrangement of the nucleotide chains in the nanosheet at each molar ratio, and growth mechanism of the nanosheets.

Figure S5. (a); Ideal arrangements nucleopeptide and spacer peptide, which take β-sheet conformation. (b); Schematic pictures of the 3D nucleopeptide/spacer peptide nanosheet, whose molar ratio is 1:5. (c); Schematic diagram for the growth process of the nucleopeptide/spacer peptide nanosheet, whose molar ratio is 1:6. We formed the assemblies on mica substrate (Area; 2.0 × 10⁻⁴ m²) and STEM grid (Area; 5.6 × 10⁻⁵ m²). We prepared the

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assemble by incubating each substrate in 5.0 mL (mica system) or 1.5 mL (STEM grid system) of solutions (0.1 mM). The number of molecules of the peptide chains in the solution at the mica and STEM grid systems are $3.0 \times 10^{17}$ and $9.0 \times 10^{16}$, respectively, and the molecular area of the peptide chains is $2.9 \text{ nm}^2$ per molecule. The total areas of the self-assembled peptide chains on mica substrate and STEM grid, assuming that the peptide chains lay on the surface as double layers, are $0.43 \text{ m}^2$ and $0.13 \text{ m}^2$, respectively. These values are large enough compared with the area of each substrate. Hence, the used amounts of the solution in this study do not affect assembly formation on mica substrate and STEM grid.
S6. Deconvolutions of UV-Vis spectra to absorption and scattering components.

a) UV-Vis spectra

Figure S6. UV-vis spectra of the self-assembly composed of the nucleopeptide and spacer peptide in aqueous solutions (a) were separated to absorption spectra based on the base paring (b) and scattering components (c) by peak deconvolution at various pH conditions (pH 6.5, pH 7.0, and pH 7.5), respectively. The molar ratio of the nucleopeptide and spacer peptide was fixed at 1:5.
S7. TEM images of the nucleopeptide/spacer peptide aggregates obtained in the aqueous solution at pH 6.5.

**Figure S7.** TEM images of the nucleopeptide/spacer peptide aggregates obtained in aqueous solution at the pH 6.5 after (a); 7 days and (b); 16 days incubation at room temperature, respectively. The molar ratio of the nucleopeptide and spacer peptide was fixed at 1:5. An aliquot sample solution that incubated each period was placed on STEM grid. After adsorption for 3 min, the excess solution was removed by absorption onto filter paper.
S8. Bright field- and dark field (DF)-TEM images and SAED pattern of the nucleopeptide/spacer peptide nanosheet.

Figure S8. Bright field- and dark field (DF)-TEM images and SAED pattern of the nucleopeptide/spacer peptide nanosheet formed on STEM grids after 10 days incubation at 15°C under the pH 7.0. Molar ratio of the nucleopeptide and spacer peptide was fixed at 1:5. DF-TEM images show the different domains attributed to each SAED spots (position 1-3).
Figure S9. AFM image of the nucleopeptide/spacer peptide nanosheet formed on mica for 7 days incubation under the pH 7.5 at 15°C. The molar ratio of the nucleopeptide and spacer peptide was fixed at 1:6.
**S10.** EDX mapping data of the magnetite-nucleopeptide/spacer peptide hybrid nanosheet after mineralization.

Figure S10. TEM and dark field STEM images, and EDX mapping of the magnetite-nucleopeptide/spacer peptide hybrid nanosheet after mineralization. The nanosheet formed on STEM grid for 10 days incubation under the pH 7.5. The molar ratio of the nucleopeptide and spacer peptide was fixed at 1:6. Mineralization was carried out for 7 days at 15°C.

**S11.** Section analysis profile of the magnetite-nucleopeptide/spacer peptide hybrid
nanosheet.

Figure S11. Section analysis profile of modulus and adhesion images at same AFM observation spot in Figure 9(h) and (i).