

1 Supporting Information

2 **Polyethyleneimine-modified graphitic carbon nitride nanosheets: a label-free Raman**
3 **traceable siRNA delivery system**

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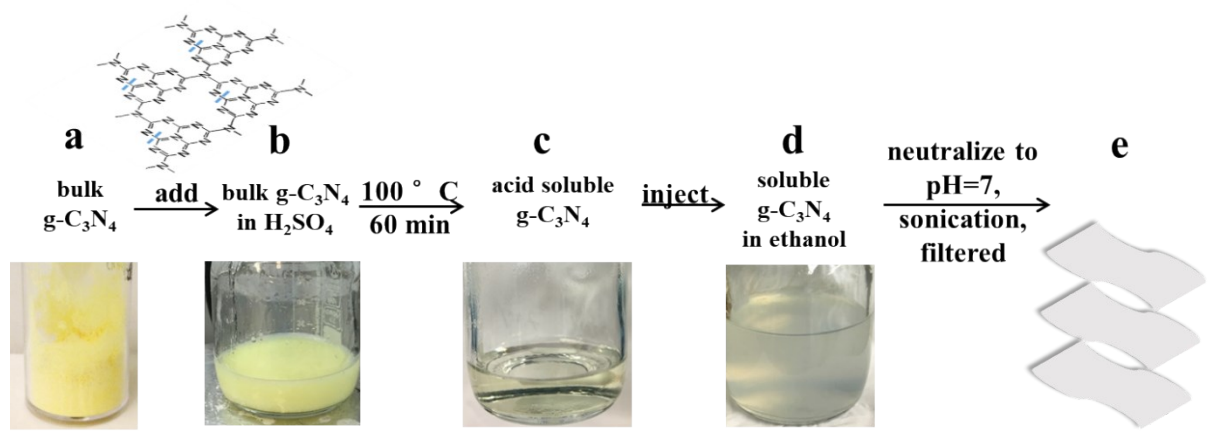
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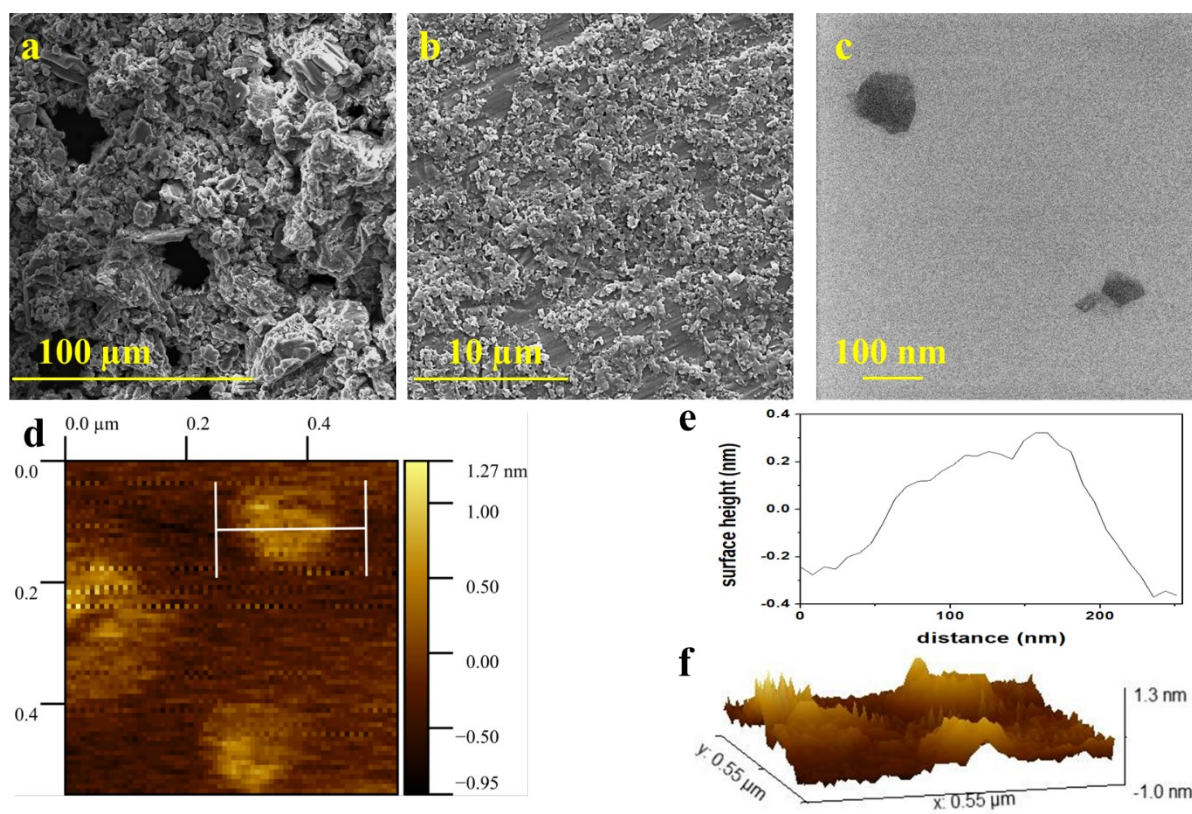
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2 **Fig. S1** Synthesis process of acidified layered $g\text{-C}_3\text{N}_4$ nanosheets and photographs of (a) bulk
 3 $g\text{-C}_3\text{N}_4$ powder, (b) $g\text{-C}_3\text{N}_4$ powder in H_2SO_4 before reaction, (c) acidified $g\text{-C}_3\text{N}_4$ nanosheets
 4 soluble in H_2SO_4 after reaction, (d) the suspension of acid-soluble $g\text{-C}_3\text{N}_4$ nanosheets in
 5 ethanol and (e) layered $g\text{-C}_3\text{N}_4$ nanosheets.

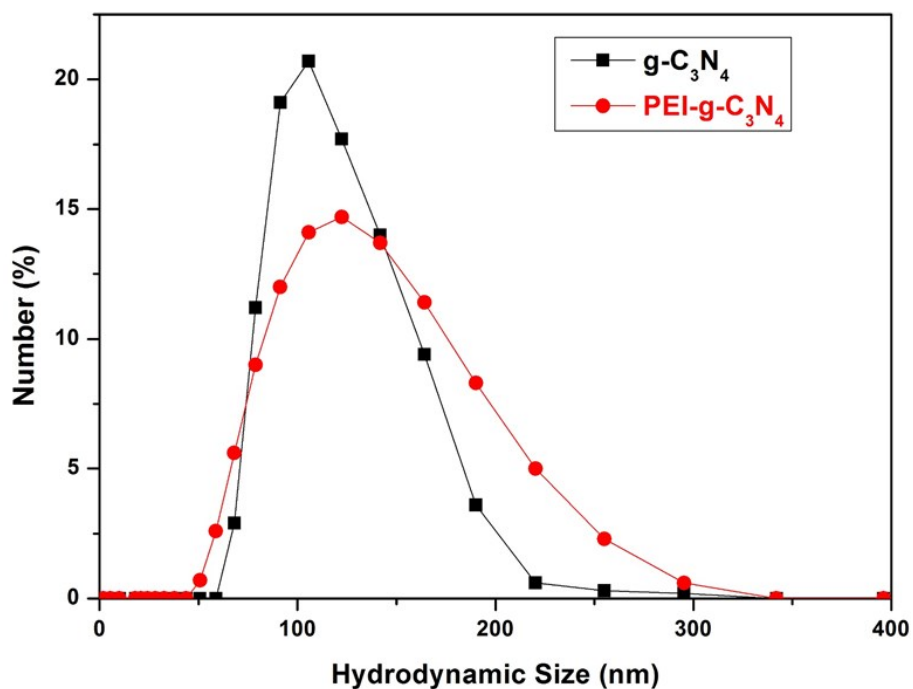
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8 **Fig. S2.** SEM images of a) bulk $g\text{-C}_3\text{N}_4$ powder, b) layered $g\text{-C}_3\text{N}_4$ nanosheets and c) the
 9 TEM image of layered $g\text{-C}_3\text{N}_4$ nanosheets, (d) the AFM image of $g\text{-C}_3\text{N}_4$ nanosheets with a
 10 randomly selected line, (e) the surface height curve of the selected line from the AFM of $g\text{-C}_3\text{N}_4$
 11 nanosheets, (f) the 3D pattern of $g\text{-C}_3\text{N}_4$ nanosheets.

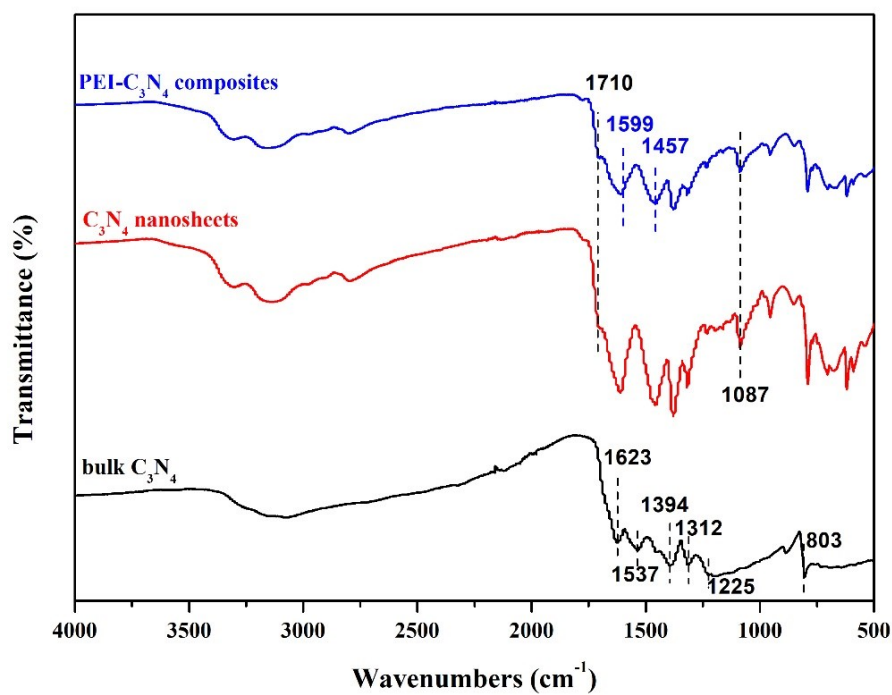
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3 **Fig. S3.** Hydrodynamic size distributions of g-C₃N₄ nanosheets and PEI-g-C₃N₄ composites
4 in water at pH 7.

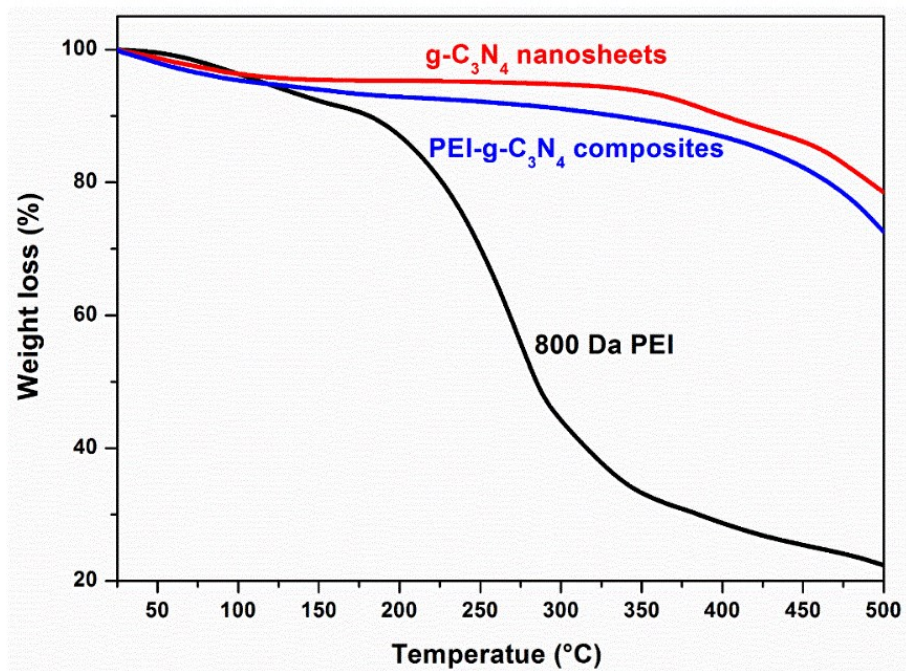
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7 **Fig. S4.** FT-IR spectra of bulk g-C₃N₄ powder, g-C₃N₄ nanosheets and PEI-g-C₃N₄ composites.

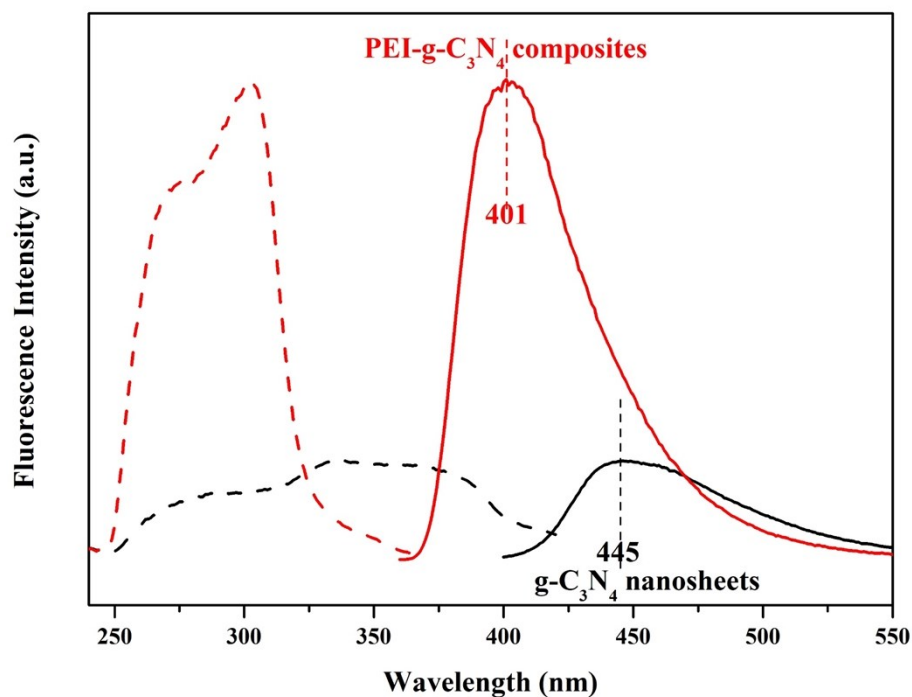
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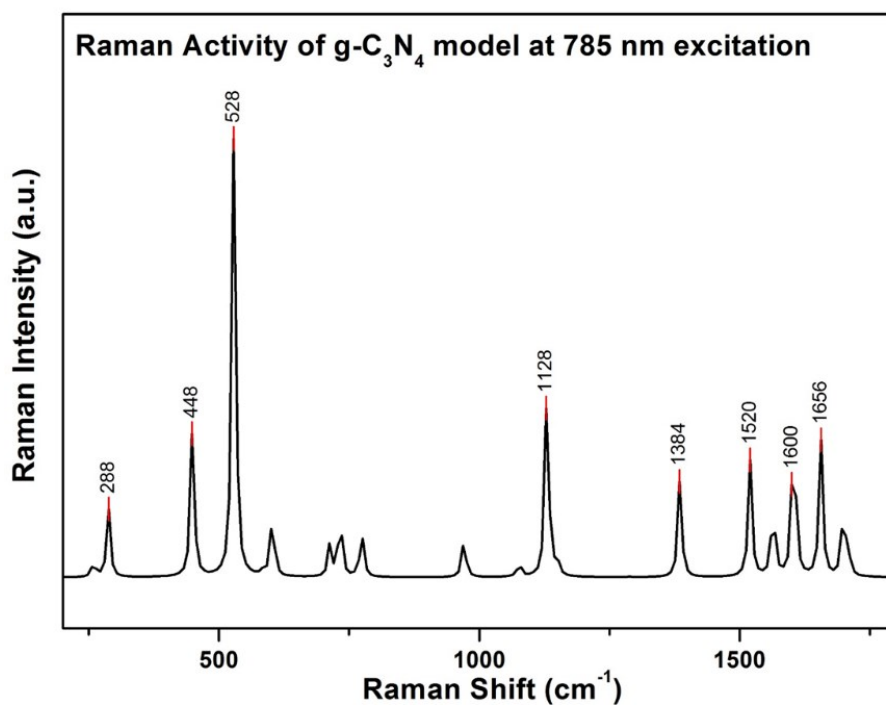
2 **Fig. S5.** TGA curves of the calcination processes of 800 Da of branched PEI, g-C₃N₄
 3 nanosheets and PEI-g-C₃N₄ nanocomposites.

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6 **Fig. S6.** Fluorescence excitation and emission spectra of g-C₃N₄ nanosheets and PEI-g-C₃N₄
 7 composites.



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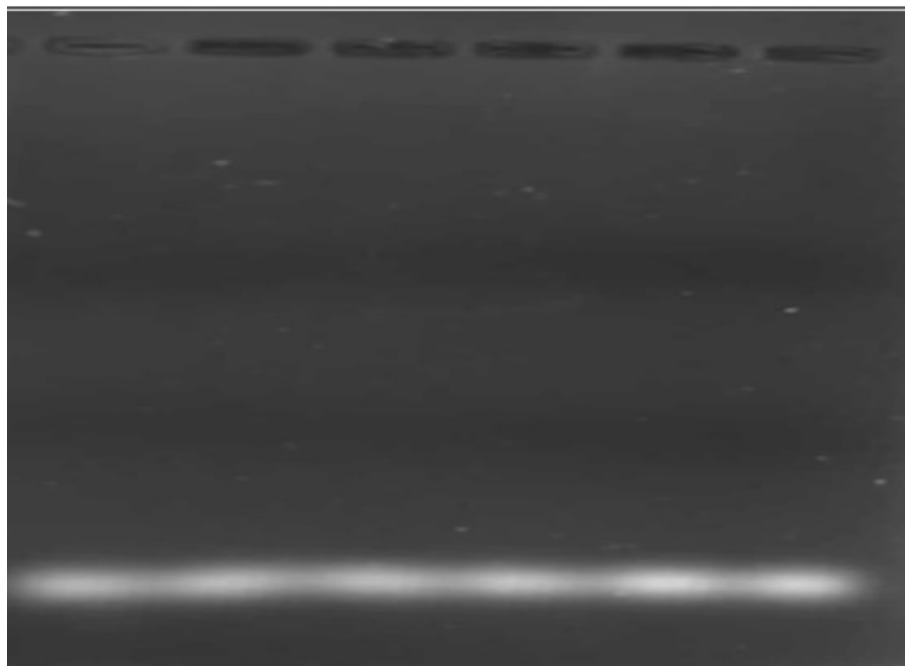
2 **Fig. S7.** The calculated Raman activity of g-C₃N₄ model under 785 nm excitation laser and
3 the vibration animations of g-C₃N₄ model at 448 and 528 cm⁻¹ Raman shift is in the followed
4 mp4 videos S1 and S2 at <https://doi.org/XXXXXXXX>

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Free DNA

Weight ratio of
materials: DNA

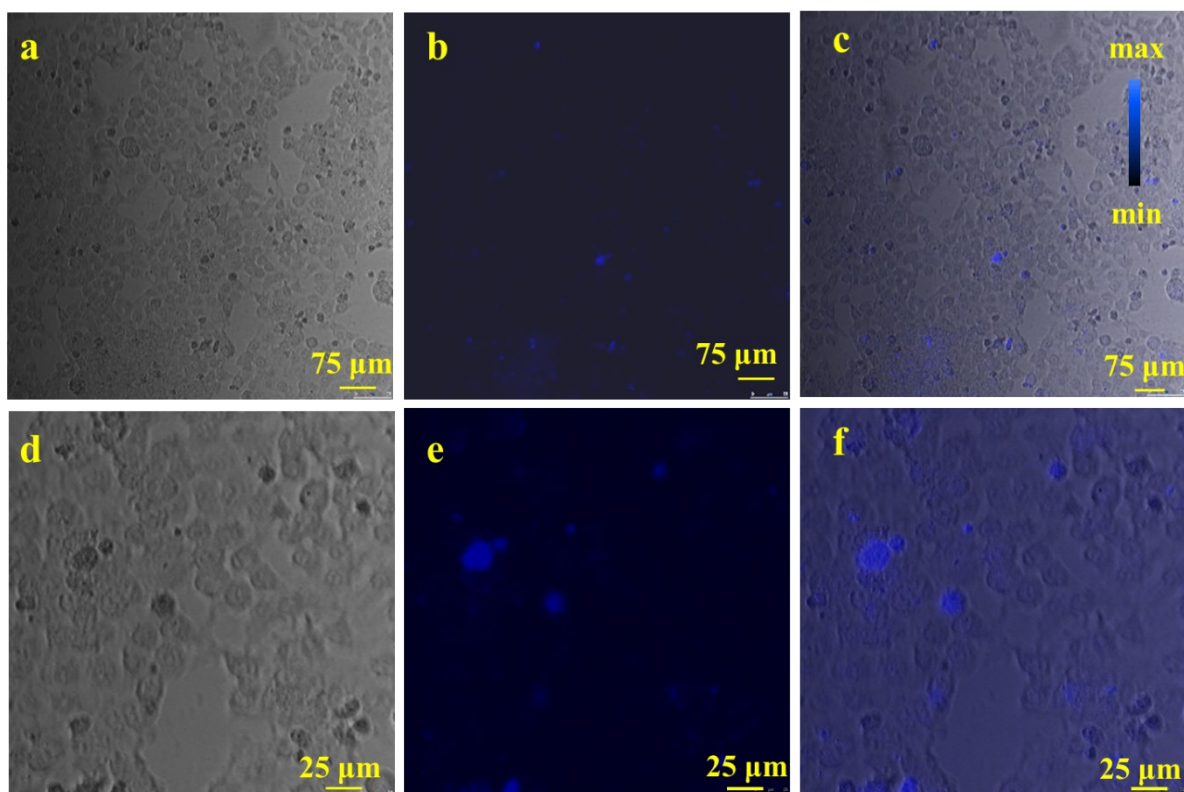
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2 **Fig. S8.** Agarose gel electrophoresis indicating the model DNA binding capabilities of g-
3 C₃N₄ nanosheets at different materials to DNA weight ratios from 10 to 200.

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2 **Fig. S9.** Fluorescence images of PEI-g-C₃N₄@siRNA composites with low magnitude (a-c)
3 and high magnitude (d-f), (a,d) optical images, (b,e) fluorescence images and (c,f) merged
4 images.

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2 **Table S1.** Hydrodynamic sizes and zeta potential of g-C₃N₄ nanosheets and PEI-g-C₃N₄
3 composites measured at pH 7.

| Sample name | PDI | Hydrodynamic size [nm] | Zeta potential [mV] ^a |
|-------------------------------------|------|------------------------|----------------------------------|
| g-C ₃ N ₄ | 0.25 | 118.4 ± 0.9 | -32.2 ± 1.5 |
| PEI-g-C ₃ N ₄ | 0.46 | 122.1 ± 0.5 | 42.5 ± 0.1 |

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