

## Supporting Information

# **The effect of side-chain functionality and hydrophobicity on the gene delivery capabilities of cationic helical polypeptides**

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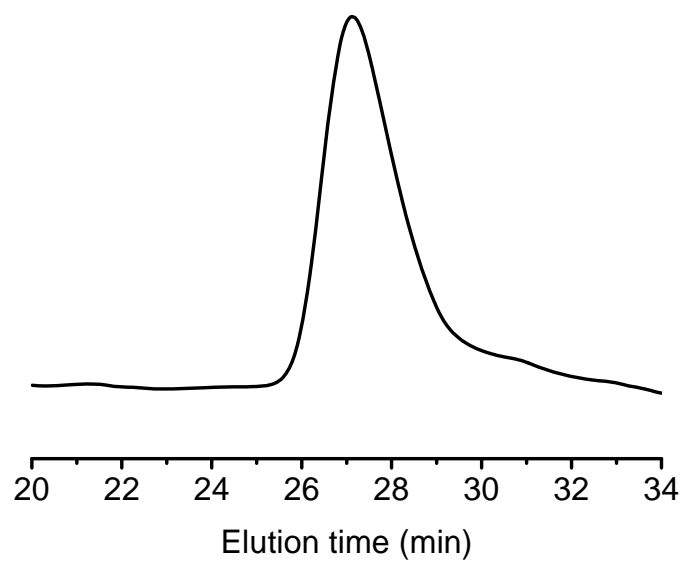


Fig. S1. GPC trace of poly( $\gamma$ -(4-propargyloxybenzyl)-L-glutamate) (PPOBLG).

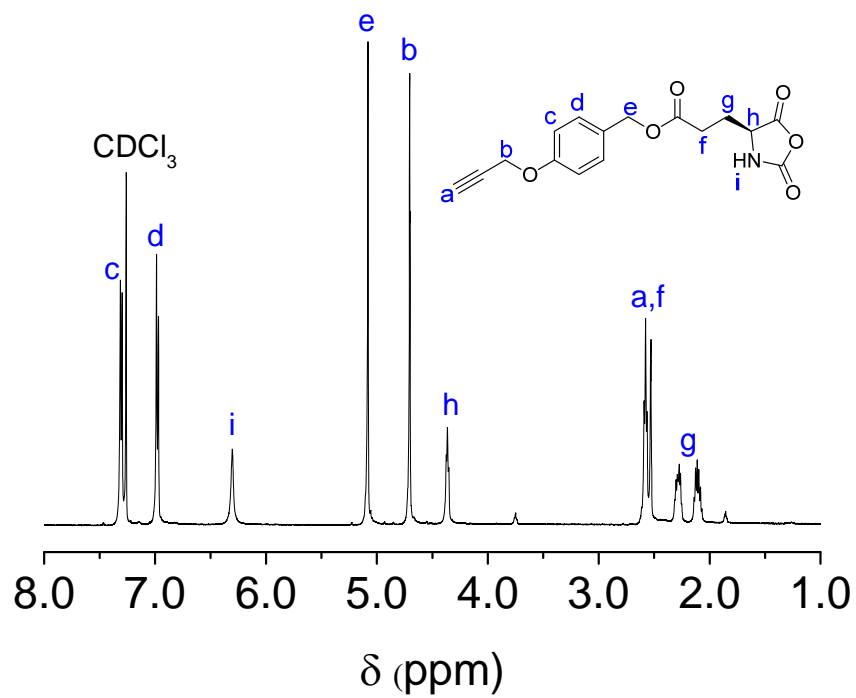


Fig. S2. <sup>1</sup>H NMR spectrum of POB-L-Glu-NCA in CDCl<sub>3</sub>.

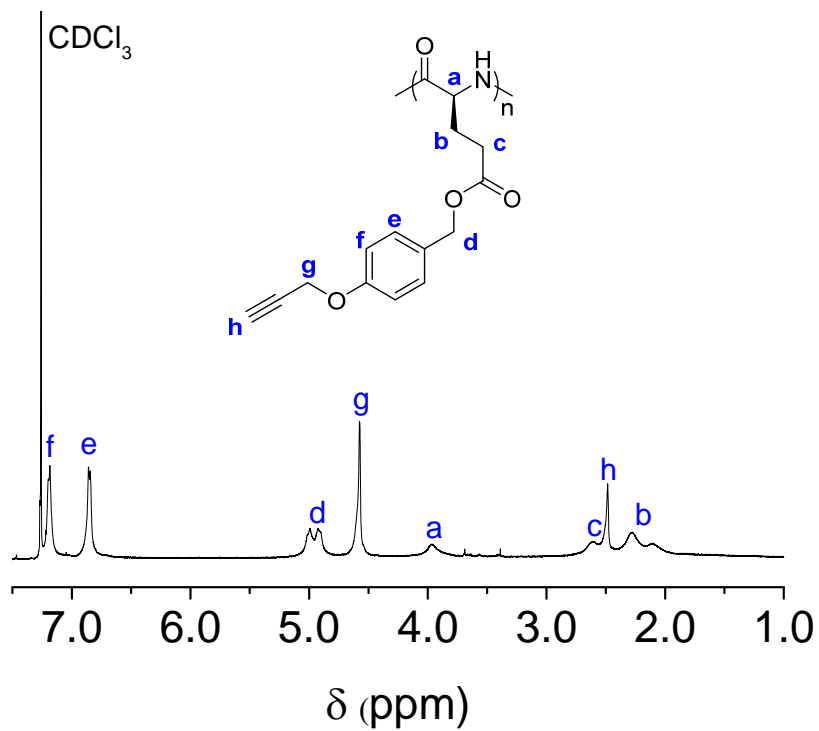


Fig. S3.  $^1\text{H}$  NMR spectrum of PPOBLG in  $\text{CDCl}_3$ .

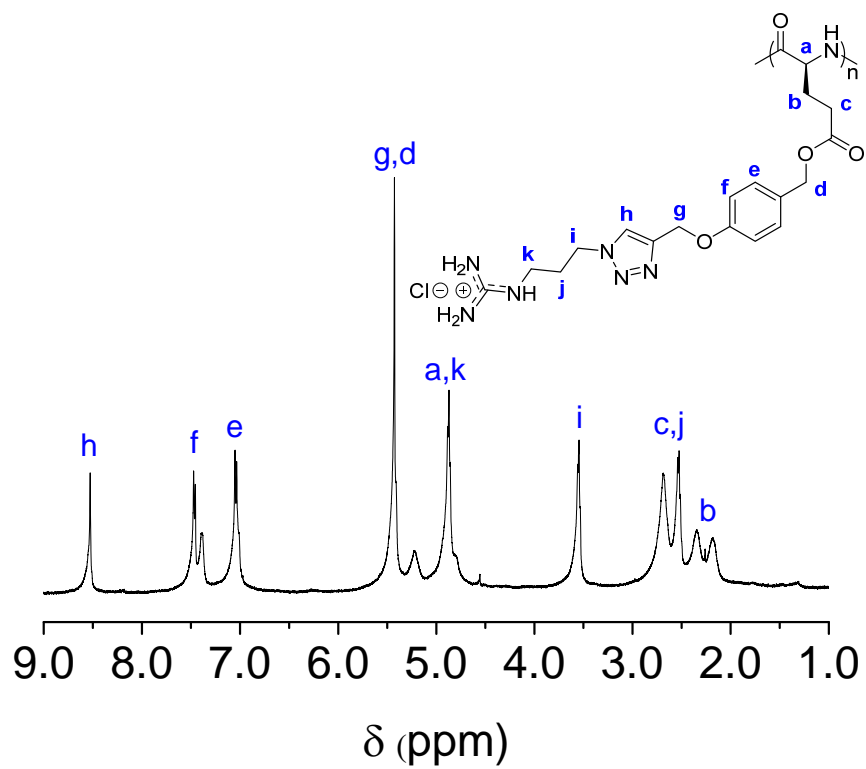


Fig. S4.  $^1\text{H}$  NMR spectrum of polypeptide G3 in  $\text{TFA-d}$ .

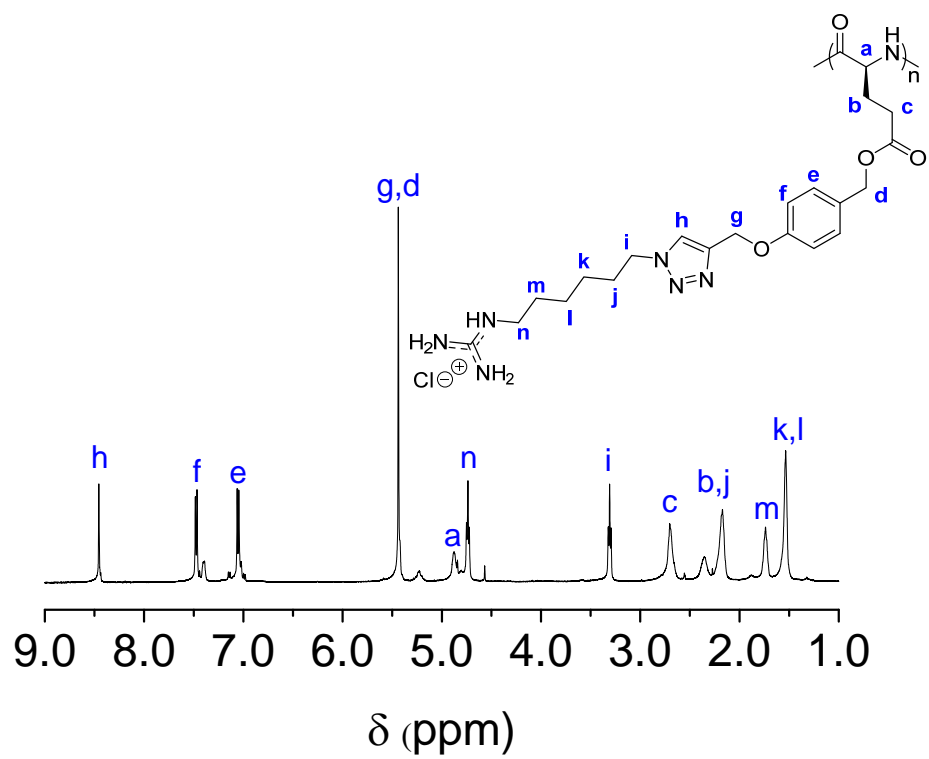


Fig. S5.  $^1\text{H}$  NMR spectrum of polypeptide G6 in  $\text{TFA-d}$ .

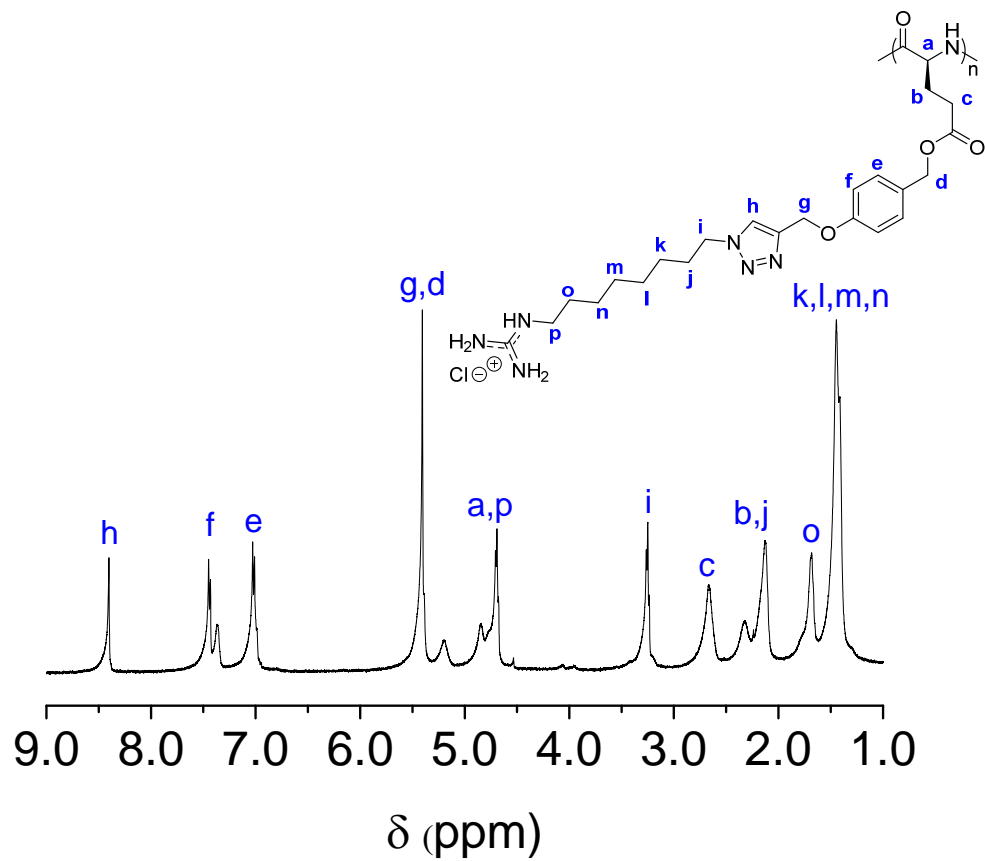


Fig. S6.  $^1\text{H}$  NMR spectrum of polypeptide G8 in  $\text{TFA-d}$ .

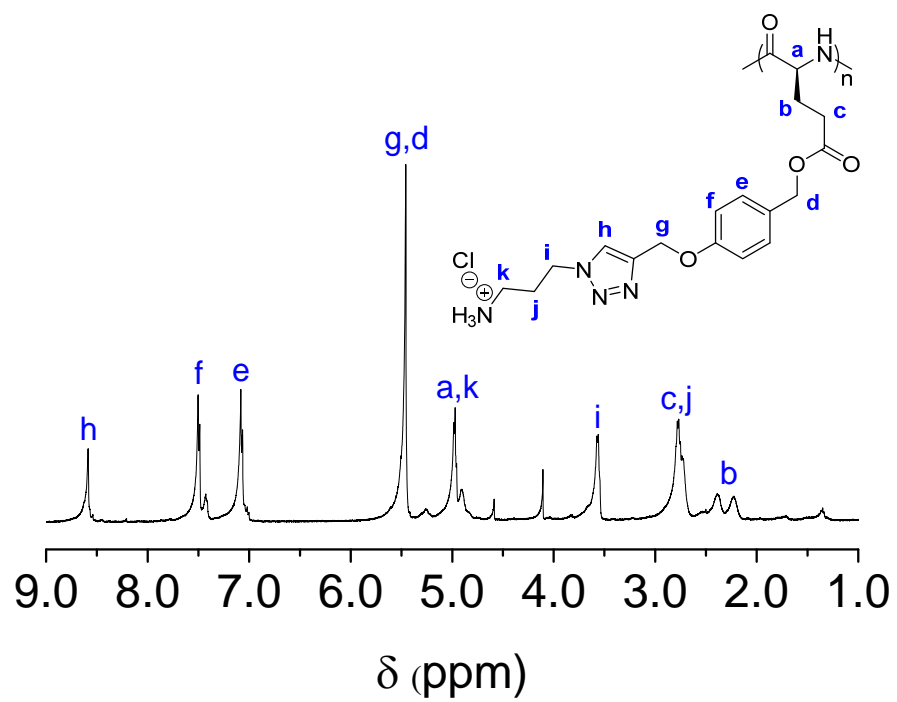


Fig. S7.  $^1\text{H}$  NMR spectrum of polypeptide P3 in  $\text{TFA-d}$ .



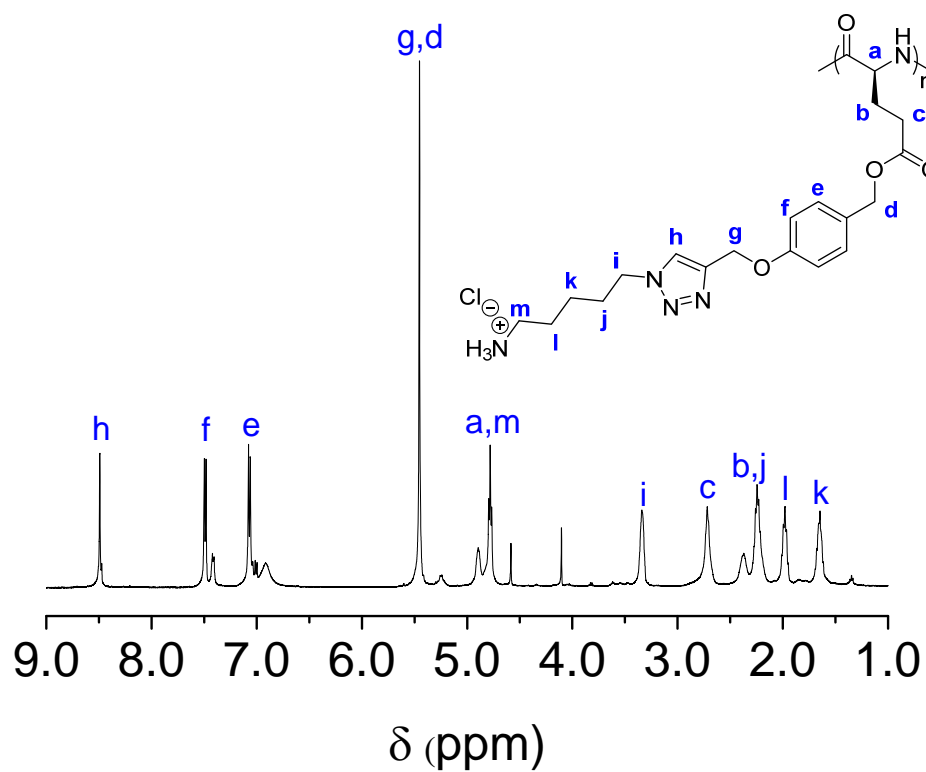


Fig. S8.  $^1\text{H}$  NMR spectrum of polypeptide P5 in  $\text{TFA-d}$ .

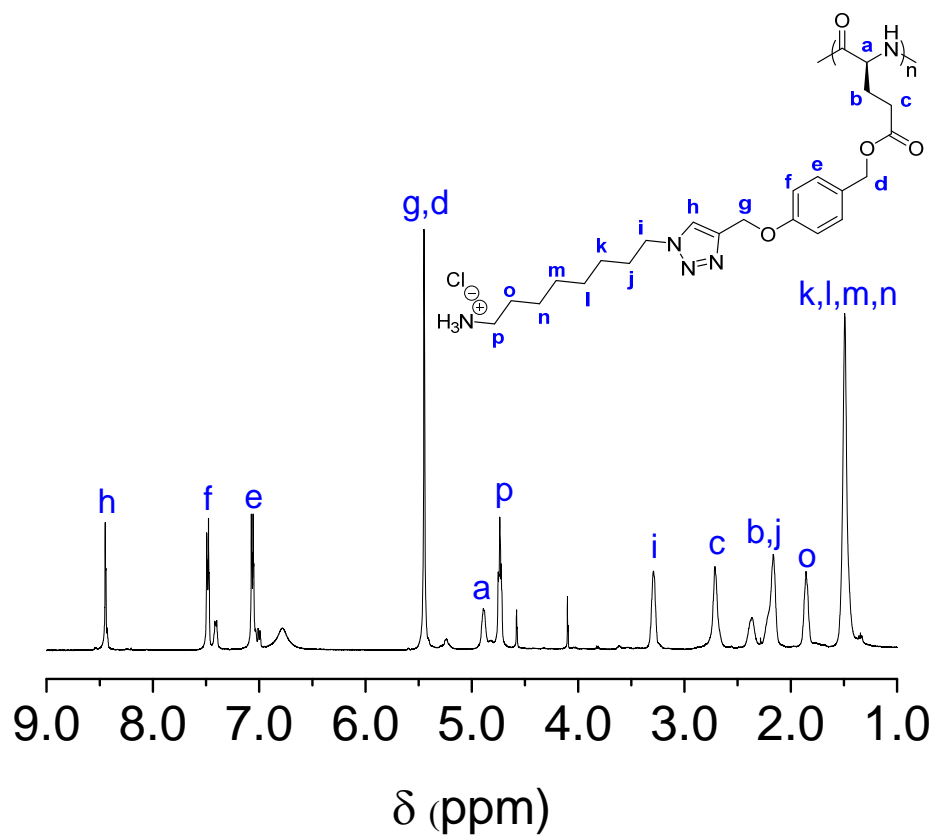


Fig. S9.  $^1\text{H}$  NMR spectrum of polypeptide P8 in  $\text{TFA-d}$ .

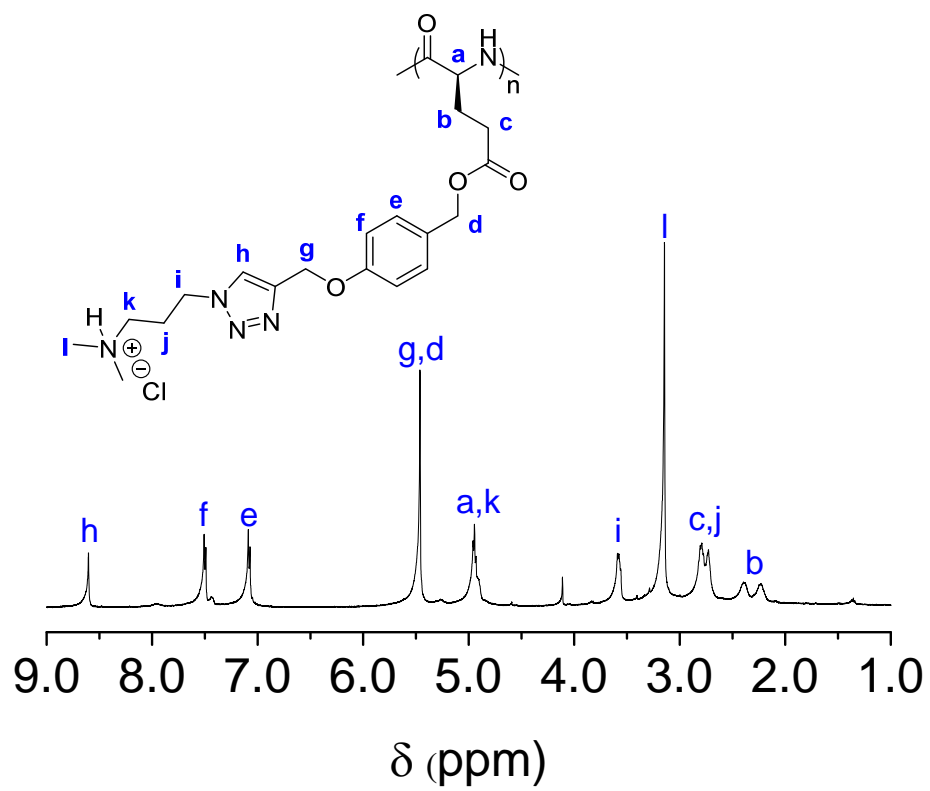


Fig. S10.  $^1\text{H}$  NMR spectrum of polypeptide T3 in TFA-*d*.

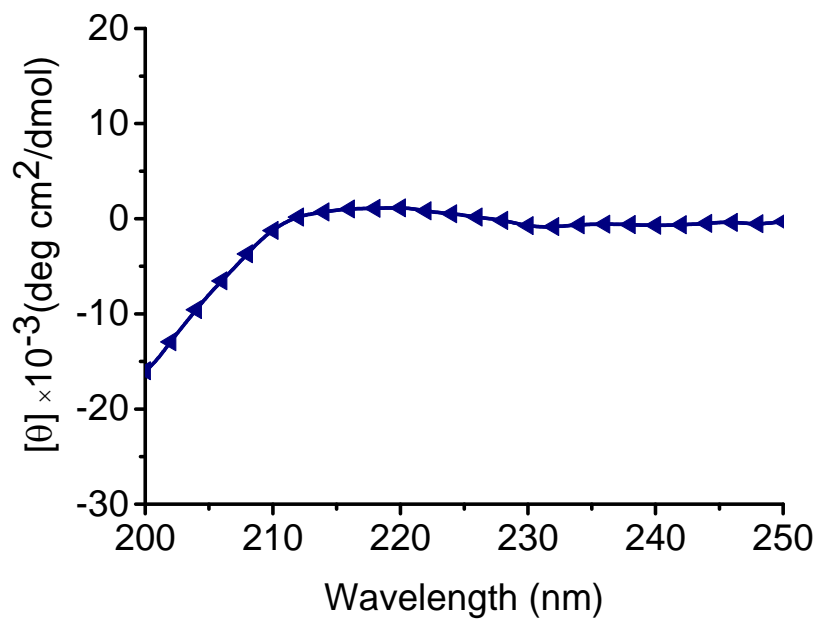


Fig. S11. CD spectrum of PLR in water (0.1 mg/mL) at pH 7.

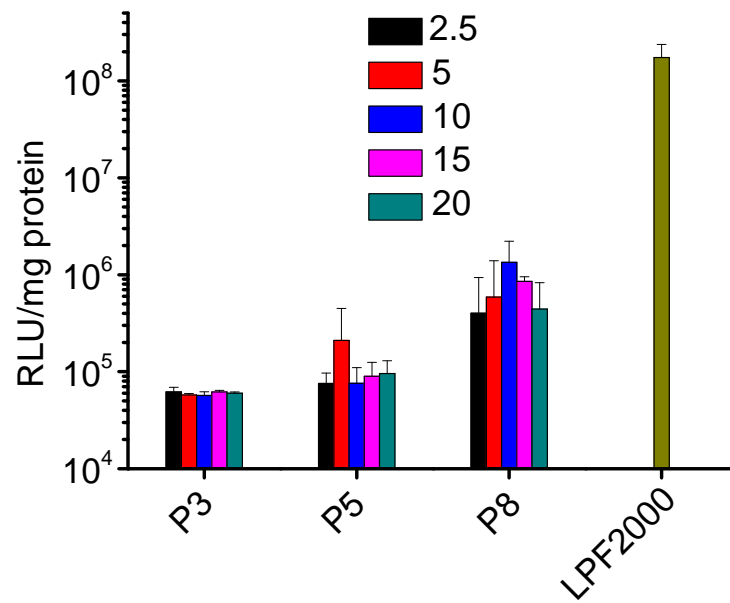


Fig. S12. *In vitro* transfection efficiencies of polyplexes at various N/P ratios in HeLa cells.

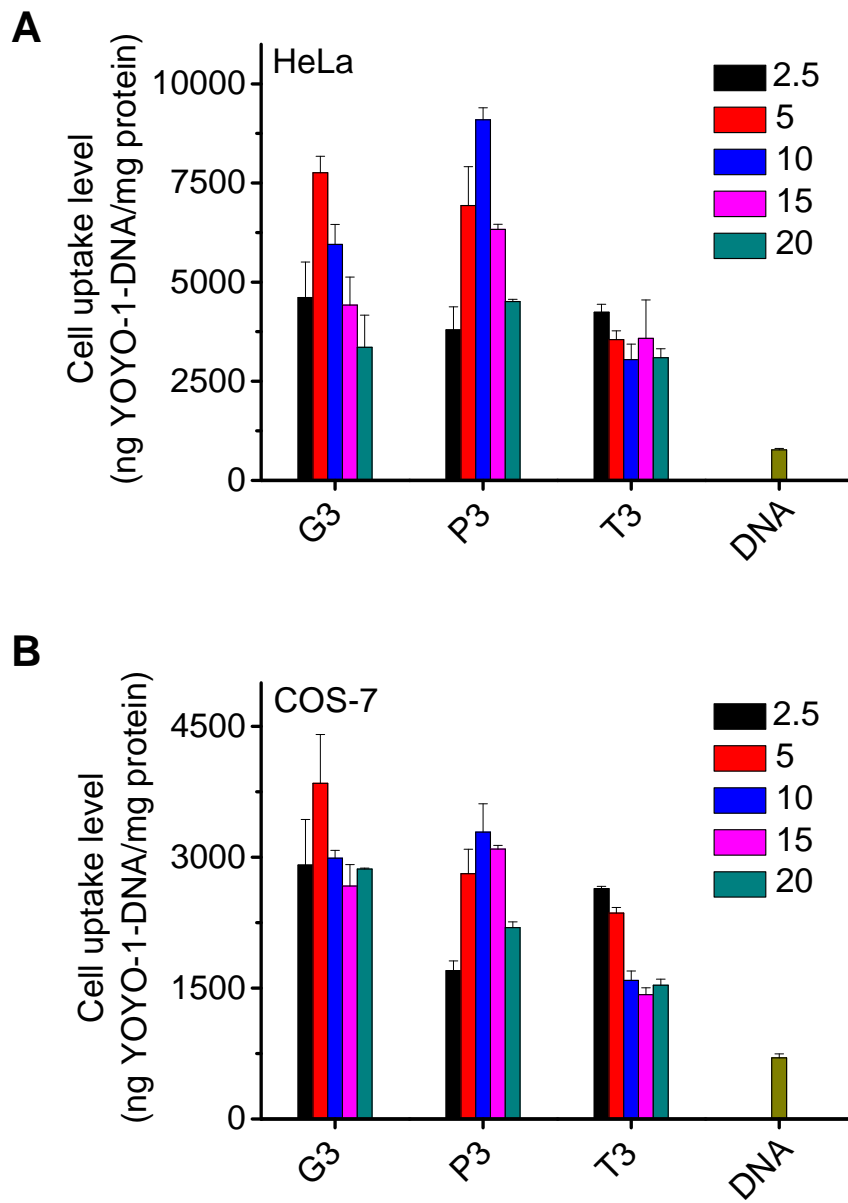


Fig. S13. Cellular uptake levels of polypeptide/YOYO-1-DNA polyplexes in HeLa (A) and COS-7 (B) cells at various N/P ratios.

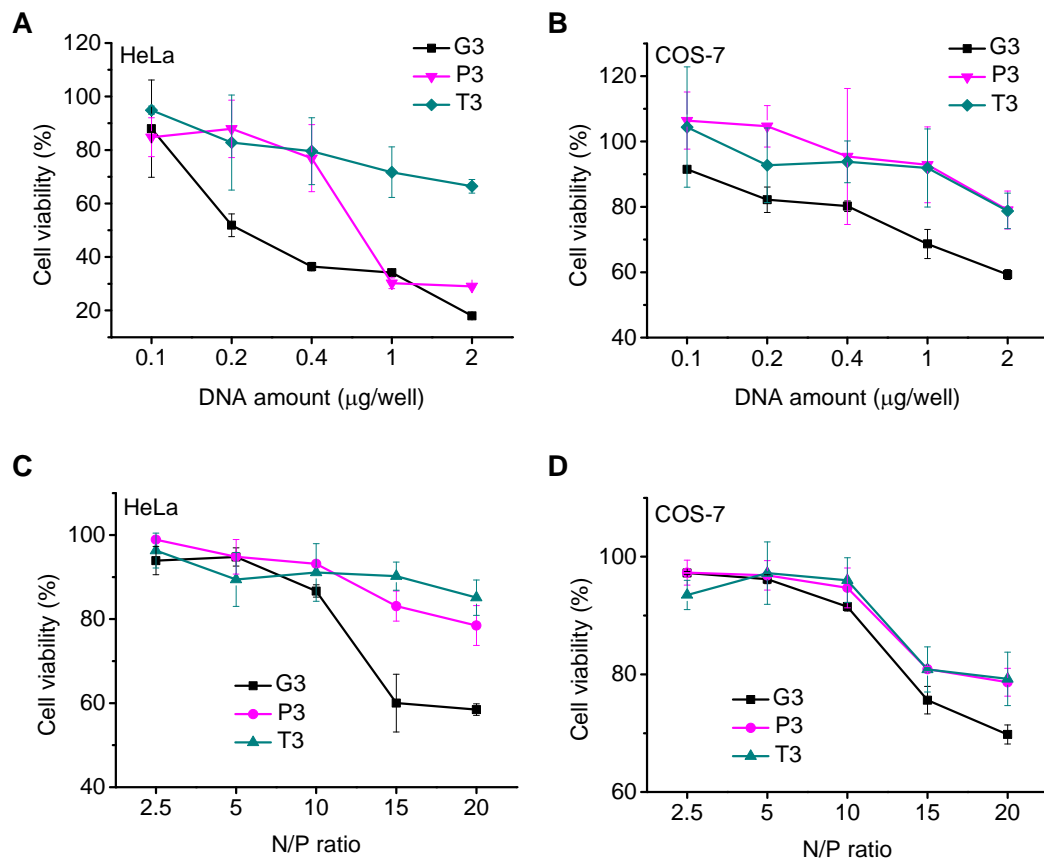


Fig. S14. Cytotoxicity of polypeptide/DNA polyplexes towards HeLa (A, C) and COS-7 (B, D) cells as determined by the MTT assay. The N/P ratio was maintained constant at 10 (A and B) while the DNA amount was maintained constant at 0.1 µg/well (C and D).