## **Supporting Information**

## A Controllable Aptamer-Based Self-Assembled DNA Dendrimer for High Affinity Targeting, Bioimaging and Drug Delivery

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Oligonucleotide	Sequence (from 5' to 3')
$\mathbf{Y}_{0a}$	GAC CGA TGG ATG ACC TGT CTG CCT AAT GTG CGT CGT AAG
$\mathbf{Y}_{0b}$	GAC CGA TGG ATG ACT TAC GAC GCA CAA GGA GAT CAT GAG
Y <sub>0c</sub>	GAC CGA TGG ATG ACT CAT GAT CTC CTT TAG GCA GAC AGG
Y <sub>1a</sub>	GAA GCC ACT CTG ACC TGT CTG CCT AAT GTG CGT CGT AAG
Y <sub>1b</sub>	GAA GCC ACT CTG ACT TAC GAC GCA CAA GGA GAT CAT GAG
Y <sub>1c</sub>	TCA TCC ATC GGT CCT CAT GAT CTC CTT TAG GCA GAC AGG
$\mathbf{Y}_{2a}$	GAC ACA CTG AGG TCC TGT CTG CCT AAT GTG CGT CGT AAG
Y <sub>2b</sub>	GAC ACA CTG AGG TCT TAC GAC GCA CAA GGA GAT CAT GAG

**Table S1** Sequences of Oligonucleotides Used in This Work

Y <sub>2c</sub>	TCA GAG TGG CTT CCT CAT GAT CTC CTT TAG GCA GAC AGG
$\mathbf{Y}_{3a}$	TAC TCG AGA CAT A CC TGT CTG CCT AAT GTG CGT CGT AAG
Y <sub>3b</sub>	TAC TCG AGA CAT A CT TAC GAC GCA CAA GGA GAT CAT GAG
Y <sub>3c</sub>	ACC TCA GTG TGT C CT CAT GAT CTC CTT TAG GCA GAC AGG
Sgc8-linker	TAT GTC TCG AGT ATT TTT ATC TAA CTG CTG CGC CGC CGG GAA
	AAT ACT GTA CGG TTA GA
RS-linker	TAT GTC TCG AGT ATT TTT NNN NNN NNN NNN NNN NNN NNN NN



**Figure S1**. Native PAGE analysis of  $Y_0, Y_1, Y_2, Y_{3.}$ 



**Figure S2**. AFM imaging for the last generation  $(G_{3-sgc8})$ .



**Figure S3**. Microscopy images of the colocalization of FITC- $G_{3-sgc8}$  and Lysotracker (lysosome marker), indicating that the internalized  $G_{3-sgc8}$  was localized in the lysosome.



**Figure S4**. Subcellular distribution of Dox (red) loaded  $G_{3-sgc8-cy5}$  (cyan). Hoechest (blue) and Lysotracker (green) were used to stain the cell nuclei and acidic organelles. Cells were imaged using a 60x oil-immersion objective. The merged pictures were used to indicate that drugs have started to diffuse into nuclei within 30min incubation. Scar bar: 20um.



**Figure S5**. Flow cytometeric analysis after incubating  $G_{3-sgc8}$  with CEM and Ramos cell lines for 2 hours, demonstrating that  $G_{3-sgc8}$  had strong selective cell recognition.



**Figure S6.** The fluorescence spectra of the Dox solution after adding different amounts of  $G_{3-sgc8}$ . Due to FRET between Dox and double-stranded DNA, the change of fluorescence intensity can be used to evaluate the max drug amount in  $G_{3-sgc8}$ .



Figure S7. Cumulative release of Dox loaded in  $G_{3-sgc8}$  in 60 hours.



**Figure S8.** MTS assay results verifying the biocompatibility of  $G_{3-sgc8}$  in CEM cells, HeLa cells and Ramos cells.