## **Supplementary Information**

# Fast and effective mitochondrial delivery of ω-Rhodamine-Bpolysulfobetaine-PEG copolymers

Nobuyuki Morimoto,<sup>\*,†</sup> Riho Takei,<sup>†</sup> Masaru Wakamura,<sup>†</sup> Yoshifumi Oishi,<sup>†</sup> Masafumi Nakayama,<sup>‡</sup> Makoto Suzuki,<sup>†</sup> Masaya Yamamoto,<sup>†</sup> Françoise M. Winnik.<sup>\*,§</sup>

<sup>†</sup> Department of Materials Processing, Graduate School of Engineering, Tohoku University, 6-6-02 Aramaki-aza Aoba, Aoba-ku, Sendai, 980-8579, Japan

<sup>‡</sup>Frontier Research Institute for Interdisciplinary Sciences (FRIS), Tohoku University, Aramaki aza Aoba 6-3, Aoba-ku, Sendai 980-8578, Japan

<sup>§</sup>Department of Chemistry and Faculty of Pharmacy, University of Montreal, CP6128 Succursale Center Ville, Montreal, QC, H3C 3J7, Canada



**Supplementary Figure S1.** Gel permeation chromatography (GPC) chromatograms of P(DMAPS-ran-PEGMA). GPC measurements were performed with a JASCO GPC system equipped with TSKgel G3000PW<sub>XL</sub> and G4000PW<sub>XL</sub> columns (Tosoh Co. Tokyo, Japan) eluted with aqueous NaNO<sub>3</sub> (100 mM). The molecular weights of the polymers were determined by calibration with PEG standards.



**Supplementary Figure S2.** <sup>1</sup>H-NMR spectrum of P(DMAPS-ran-PEGMA) in D<sub>2</sub>O containing 1 M NaCI.



Supplementary Figure S3. UV-Vis spectra of P(DMAPS-ran-PEGMA) 18k before and after aminolysis reaction. The aminolysis reaction was applied to P(DMAPS-ran-PEGMA) in MES buffer (pH6.0) by addition of a large excess of *n*-butylamine (100 equiv.) for 2 h at r.t. in order to convert the  $\omega$ -trithiocarbonate group to a thiol. The absorbance derived from trithiocarbonate group at 310 nm was disappeared by the conversion.



#### Supplementary Figure S4. Synthetic scheme of sRhoP



 $\alpha$ RhoP-74k

Supplementary Figure S5. Localization RhoPs in HeLa cells. The images were obtained by CLSM after 1 h co-incubation with each polymer at 37  $^{\circ}$ C, 5% CO<sub>2</sub> in the presence of 10 % serum.



# Supplementary Figure S6. Effects of the concentration of the endocytosis inhibitors

## on the uptake of (A) $\alpha$ RhoP-18k, (B) $\omega$ RhoP-21k, (C) sRhoP-18k and (D) $\alpha$ RhoH-30k.

The fluorescence intensity of HeLa cells was evaluated by flow cytometry. CytoD:

Cytocharacine D, M $\beta$ CD: methyl- $\beta$ -cyclodextrin.



**Supplementary Figure S7.** (A) Fluorescence intensity of  $\alpha$ RhoP in HeLa cells as a function of the potassium concentration during polymer uptake. (inset) Plot of the cellular uptake as a function of potassium Nernst potential. (B) Correlation between  $\alpha$ RhoP concentration and the amount of cellular uptake. The concentration of  $\alpha$ RhoP was ranged from 0.001 mg/mL to 1.0 mg/mL.

[K <sup>⁺</sup> ] (mM)	0	1.5	4.2	14.5	43.8	141.2
[NaCl]	139.7	136.7	137	126.6	97.4	0
[KCI]	0	0	2.7	13.1	42.3	139.7
[Na <sub>2</sub> HPO <sub>4</sub> ]	9.6	8.1	8.1	8.1	8.1	8.1
[KH <sub>2</sub> PO <sub>4</sub> ]	0	1.5	1.5	1.5	1.5	1.5

Supplementary Table S1. Buffer compositions with various potassium concentrations.