Electronic Supplementary Information

Ultrasensitive detection of disease biomarkers using an immuno-wall device with enzymatic amplification

Keine Nishiyama,^a Toshihiro Kasama,^{b,c} Seiya Nakamata,^a Koya Ishikawa,^d Daisuke Onoshima,^{c,e} Hiroshi Yukawa,^{c,d,e} Masatoshi Maeki,^f Akihiko Ishida,^f Hirofumi Tani,^f Yoshinobu Baba^{c,d,e} and Manabu Tokeshi^{*,c,e,f,g}

^aGraduate School of Chemical Sciences and Engineering, Hokkaido University, Kita 13 Nishi 8, Kitaku, Sapporo 060-8628, Japan

^bDepartment of Bioengineering, School of Engineering, The University of Tokyo, Shinkawasaki, Saiwai-ku, Kawasaki-shi, Kanagawa, 212-0032, Japan

^cImPACT Research Center for Advanced Nanobiodevices, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, Japan

^dDepartment of Biomolecular Engineering, Graduate School of Engineering, Nagoya University, Furocho, Chikusa-ku, Nagoya 464-8603, Japan

^eInstitute of Nano-Life-Systems, Institutes of Innovation for Future Society, Nagoya University, Furocho, Chikusa-ku, Nagoya 464-8603, Japan

^fDivision of Applied Chemistry, Faculty of Engineering, Hokkaido University, Kita 13 Nishi 8, Kitaku, Sapporo 060-8628, Japan

^gInnovative Research Centre for Preventive Medical Engineering, Nagoya University, Furo-cho Chikusa-ku, Nagoya 464-8603, Japan

Corresponding author: (M. Tokeshi) Phone: +81-11-706-6744; Fax: +81-11-706-6745; E-mail: tokeshi@eng.hokudai.ac.jp

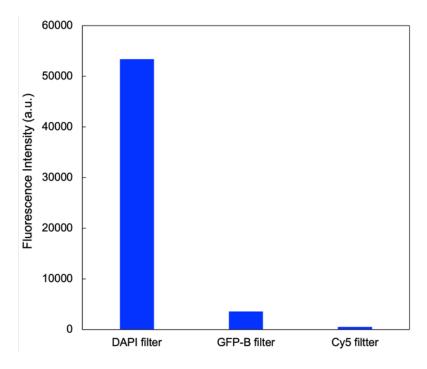


Fig. S1 Background signal of the immuno-wall obtained using various filters. Exposure time of the

CCD camera was 1 s for all filters.

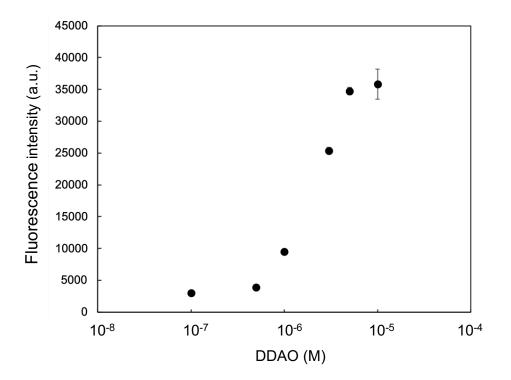


Fig. S2 Calibration curves of 9H-(1,3-dichloro-9,9-dimethylacridin-2-one-7-yl) (DDAO) in a microchannel without the immuno-wall. Exposure time of the CCD camera was 5 s.

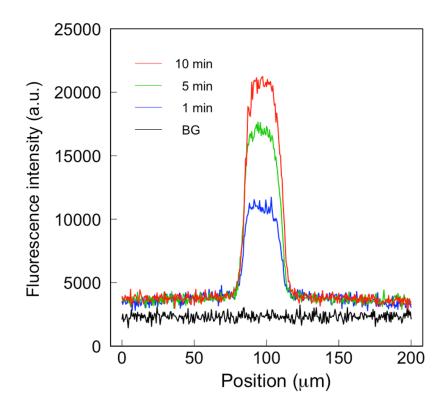


Fig. S3 Fluorescence intensity profile as a function of the position with injection of 0.5 μ M DDAO. Exposure time of the CCD camera was 5 s.

Method	LOD ^a	Assay time (min)	Sample volume (µL)	Ref.
Immunoassay on a power-free microchip	17 pg/mL	23	0.5	(38)
Surface plasmon resonance-based immunoassay	1.2 ng/mL	5	50	(39)
Label-free immunoassay based on electron transfer	40 pg/mL	50	10	(40)
Paper-based device	5.0 μg/mL	22	0.5	(41)
Nanoribon sensors using a miniature bead-based enzyme-linked immunosorbent assay	50 pg/mL	60	10	(42)
Graphene/polyethylene glycol hybrids for single-step immunoassay microdevice	400 ng/mL	2	-	(43)
Immuno-wall device (Fluorescence-label)	1.7 ng/mL	16	0.25	This work
Immuno-wall device (Enzyme-label)	2.5 pg/ mL	46	0.25	This work

Table S1 Analytical performance of correlative devices for C-reactive protein (CRP) detection

^a LOD, limit of detection