# Supplementary Information for
# Fabricating Nanopores with Diameters of Sub-1 nm to 3 nm Using Multilevel Pulse-voltage Injection

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The Supplementary Information includes:

SI-1. Detailed procedure and flowchart of MPVI

SI-2. Magnified TEM images of nanopores fabricated by MPVI

SI-3. Time trace of ionic current through a nanopore with $\phi_M$ = 0.88 nm

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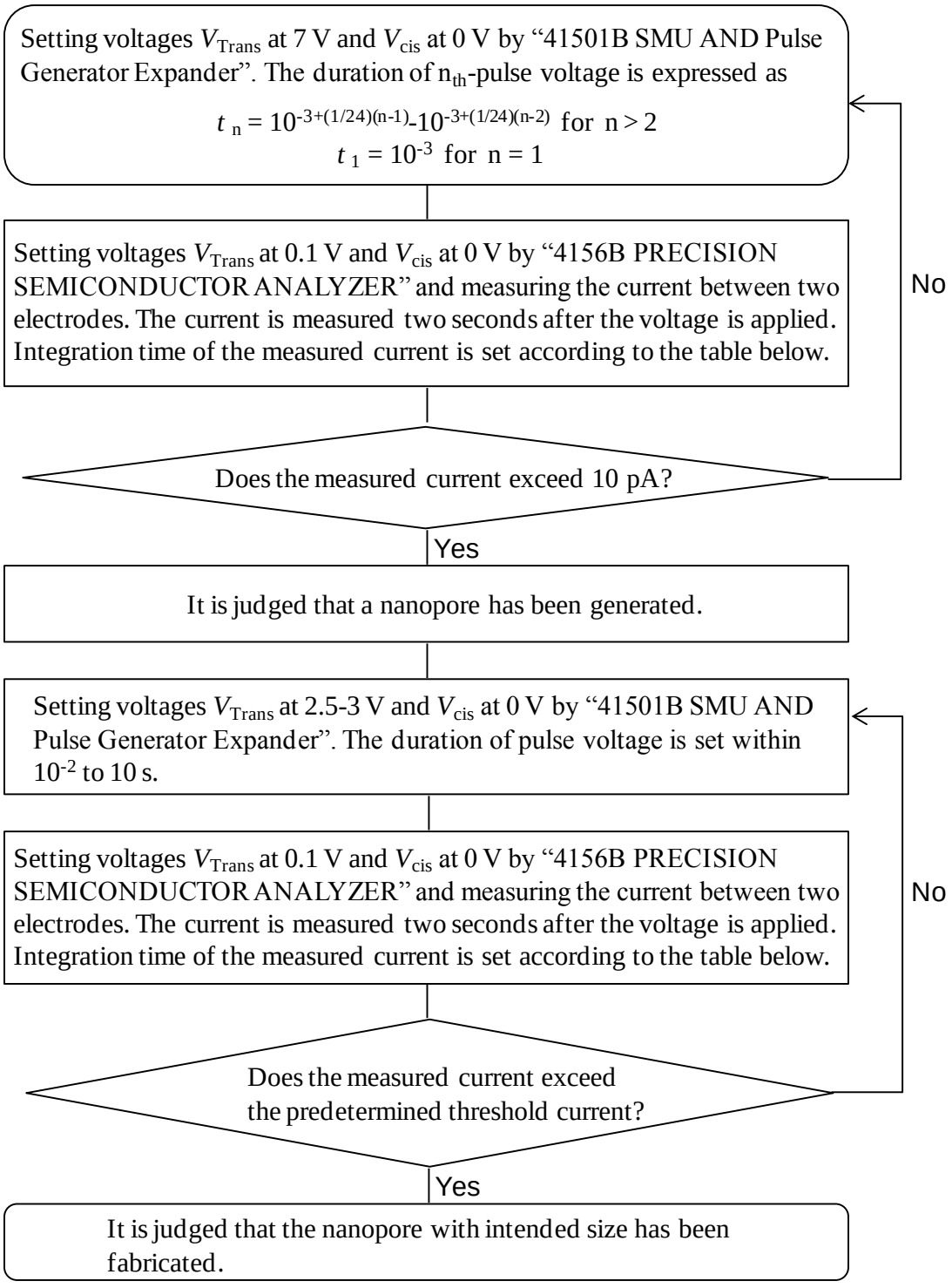
## SI-1. Detailed procedure and flowchart of MPVI

The MPVI procedure is controlled by a programme written in Excel VBA (Visual Basic for Applications). The detailed flowchart of the programme is described below.

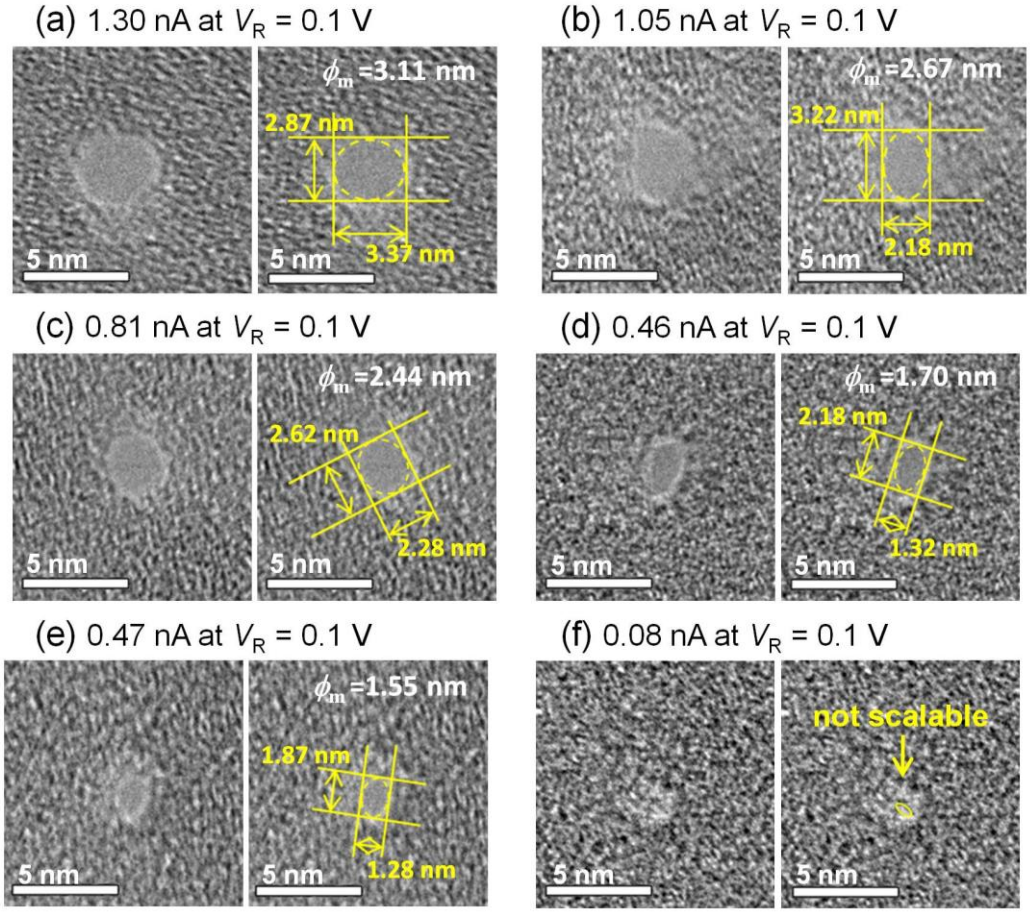


The integration time of the currents measured by a 4156B PRECISION SEMICONDUCTOR ANALYZER is listed in the table below. We allowed the integration time to be automatically changed according to the following table by setting it to “medium”.

| measurement range | integration time |
|---|---|
| 10 pA | 1 s |
| 100 pA | 200 ms |
| 1 nA | 100 ms |

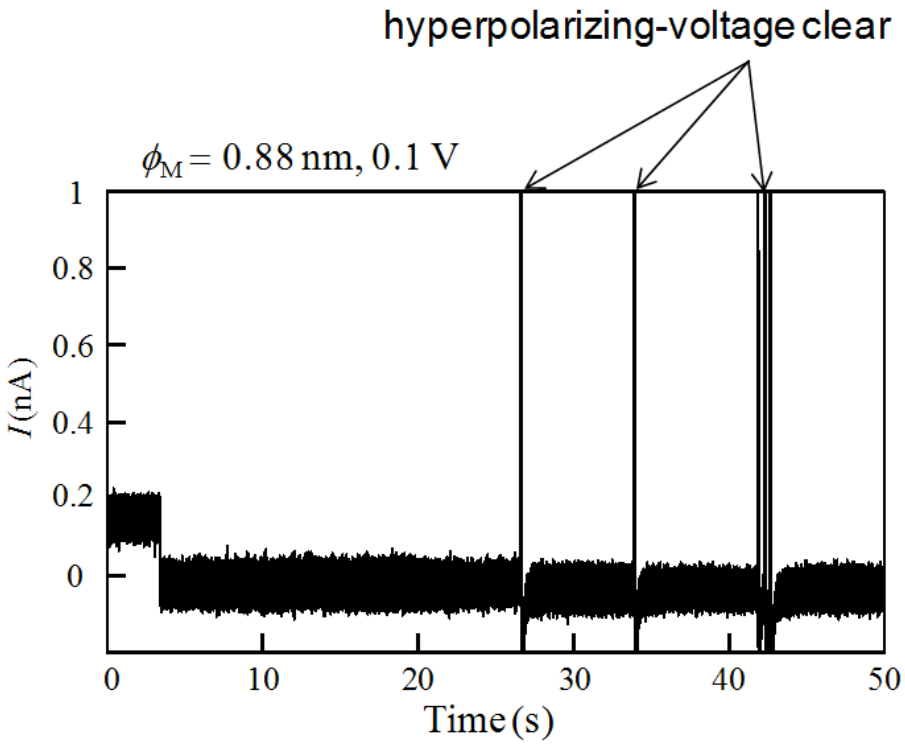
## SI-2. Magnified TEM images of nanopores fabricated by MPVI

Magnified views of the nanopores. Each left image shows the raw image of each right image. The ionic currents through the nanopore ($I_{Tot.}$ at $V_R$ = 0.1 V) were (a) 1.30 nA, (b) 1.05 nA, (c) 0.81 nA, (d) 0.46 nA, (e) 0.47 nA, and (f) 0.08 nA. The nanopore with $I_{Tot.}$ = 0.08 nA at $V_R$ = 0.1 V was too small to measure.



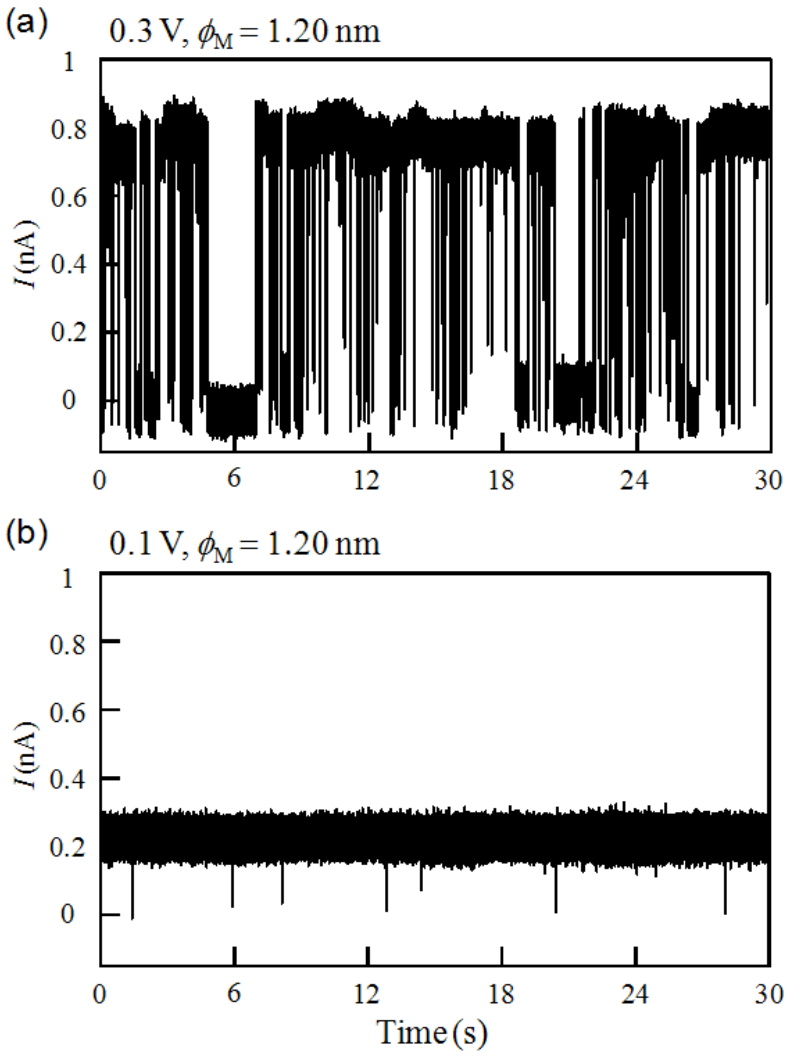
## SI-3. Time trace of ionic current through a nanopore with $\phi_M$ = 0.88 nm

The ionic current was measured at 0.1 V, and the measurement data were low-pass filtered at 5 kHz. The nanopore was closed a few seconds after the start of the measurement, indicating that the nanopore was clogged with ssDNA. Although hyperpolarizing voltages (±1.3-$V_{DC}$ pulse with a duration of 100 ms) were applied to remove ssDNA from the nanopore, the ionic current did not recover.




## SI-4. Dependence of frequency of ssDNA translocation events on applied voltage

The ionic current was measured at (a) 0.3 V and (b) 0.1 V using the same nanopore with $\phi_M$ = 1.20 nm. Each data set was low-pass filtered at 5 kHz. The event frequency significantly decreased as the voltage was decreased from 0.3 to 0.1 V; the statistical analysis of $\Delta I$ was difficult at 0.1 V.




## SI-5. Dependence of ionic-current-blockade histogram on applied voltage

The ionic currents were measured using the nanopores with $\phi_M$ = 2.33 nm (a-1 and a-2) and 2.55 nm (b-1 and b-2) at voltages of 0.3 V (a-1 and b-1) and 0.1 V (a-2 and b-2). Each data set was low-pass filtered at 10 kHz. $\Delta I_p$ was determined by the peak value of the Gaussian fit. In Figs. a-1 and a-2, a more discriminative peak in the histogram appears at 0.1 V. The ratio of $\Delta I_P$ at 0.3 V to $\Delta I_P$ at 0.1 V is approximately three. This result indicates that $\Delta I_P$ is proportional to the applied voltage. In Figs. b-1

and b-2, a discriminative peak in the histogram only appears at 0.1 V; it does not appear at 0.3 V. These results indicate that the translocation events cannot be measured accurately at 0.3 V when $\phi_M$ > 2.5 nm because the translocation speed of ssDNA through the nanopore increases with increasing $\phi_M$.