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Ultra-Low Noise Measurements of Ionic Transport Within Individual Single-Walled Carbon Nanotubes

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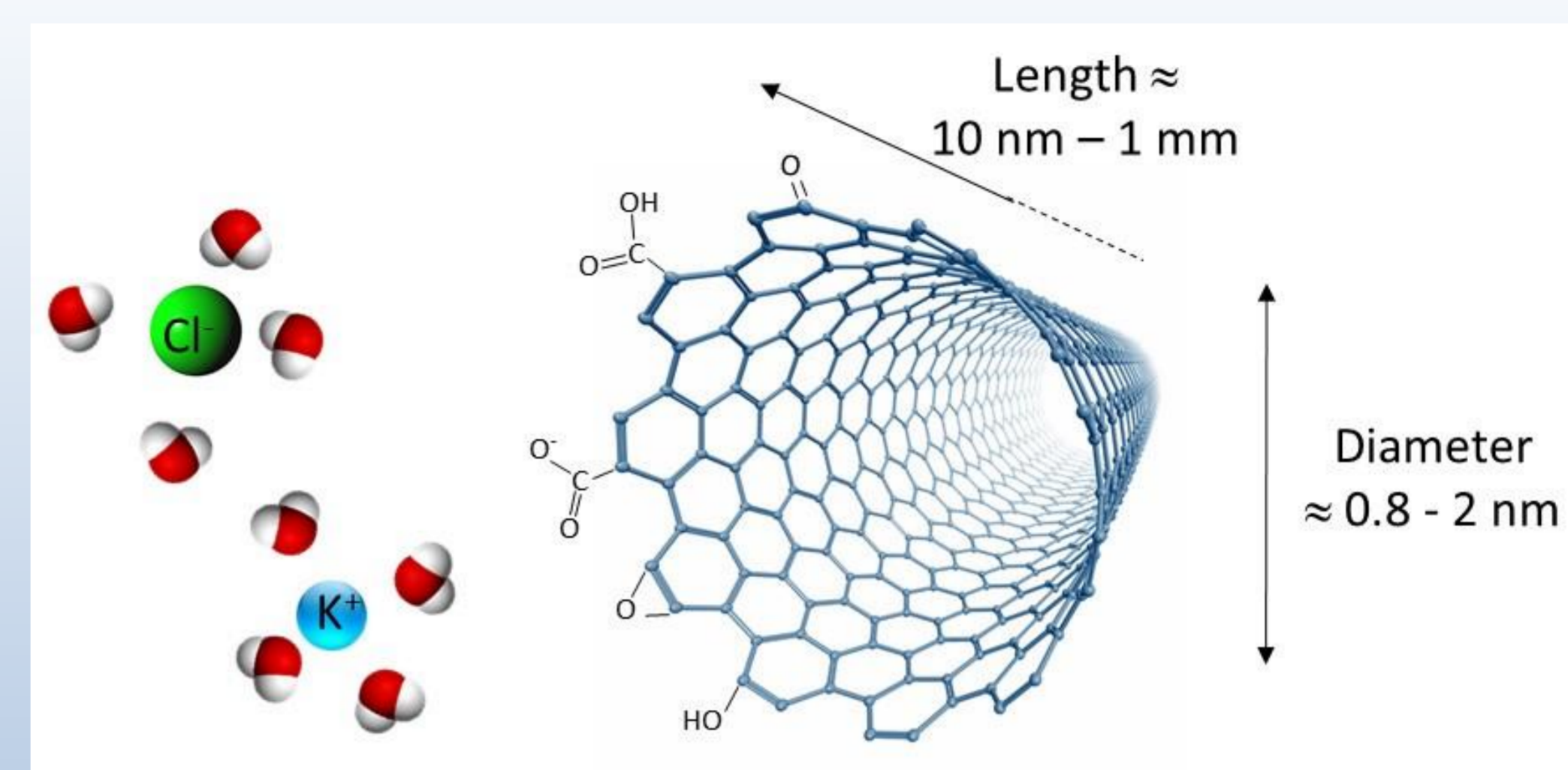
Abstract

Despite 15 years of extensive investigation, the fabrication and study of nanofluidic devices incorporating a single carbon nanotube (CNT) still represent a remarkable experimental challenge [1-3]. In this study, we present the fabrication of nanofluidic devices that integrate an individual single-walled CNT (SWCNT), showcasing a notable reduction in noise by 1-3 orders of magnitude compared to conventional devices. This achievement was made possible by employing high dielectric constant materials for both the substrate and the CNT-covering layer. Furthermore, we provide a detailed account of the crucial factors contributing to the successful fabrication of SWCNT-based nanofluidic devices that are reliably leak-free, plug-free, and long-lived. Key considerations include the quality of the substrate-layer interface, the nanotube opening, and the effective removal of photoresist residues and trapped microbubbles. We demonstrate that these devices, characterized by a high signal-to-noise ratio, enable spectral noise analysis of ionic transport through an individual SWCNT, thus showing that SWCNTs obey Hooge's law in $1/f$ at low frequencies [4]. Beyond advancing our fundamental understanding of ion transport in SWCNTs, these ultra-low noise measurements open avenues for leveraging SWCNTs in nanopore sensing applications for single-molecule detection, offering high sensitivity and identification capabilities.

References

- [1] Manghi et al., *J. Phys. Chem. C* **2021**, 125(42):22943. doi: 10.1021/acs.jpcc.1c0820
- [2] Liu et al., *Science* **2010**, 327(5961):64–67. doi: DOI10.1126/science.1181799
- [3] Yazda et al., *Nanoscale* **2017**, 9(33):11976–11986. doi: 10.1039/C7NR02976D
- [4] BSAWMAII et al., submitted.

Motivations



- Diameter \approx molecule, Debye length \rightarrow Surface and confinement effects
- Smooth surface \rightarrow Low friction
- Functionalized ends \rightarrow Access selectivity
- High aspect ratio \rightarrow Access versus channel resistance
- Semiconducting or metallic \rightarrow Influence on transport mechanisms
- Atom-thin layer \rightarrow Outer environment effects

Microdevice fabrication and filling

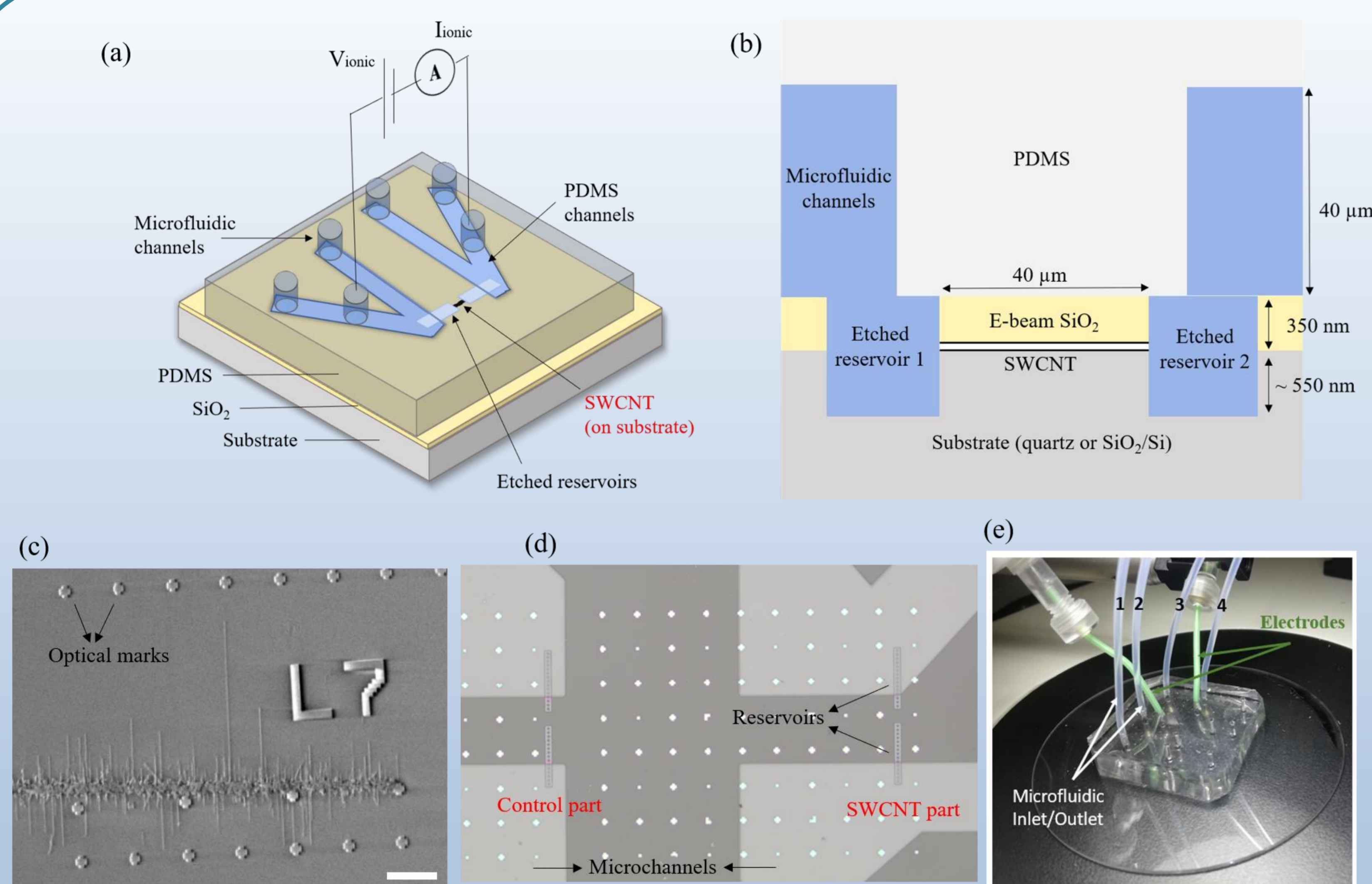


Fig. 1. (a) Schematic of the device made of an individual SWCNT on a substrate, covered with a thin SiO₂ layer and connecting two etched reservoirs fed by two microfluidic channels. (b) Side-view image of the nanofluidic device. (c) SEM image of SWCNTs transferred on a substrate with optical marks. (d) SEM image of the complete device (SWCNT and control parts). (e) Photograph of a fabricated device, on quartz substrate, connected to fluidic capillaries and Ag/AgCl electrodes. The scale bar in (c) is 40 μ m.

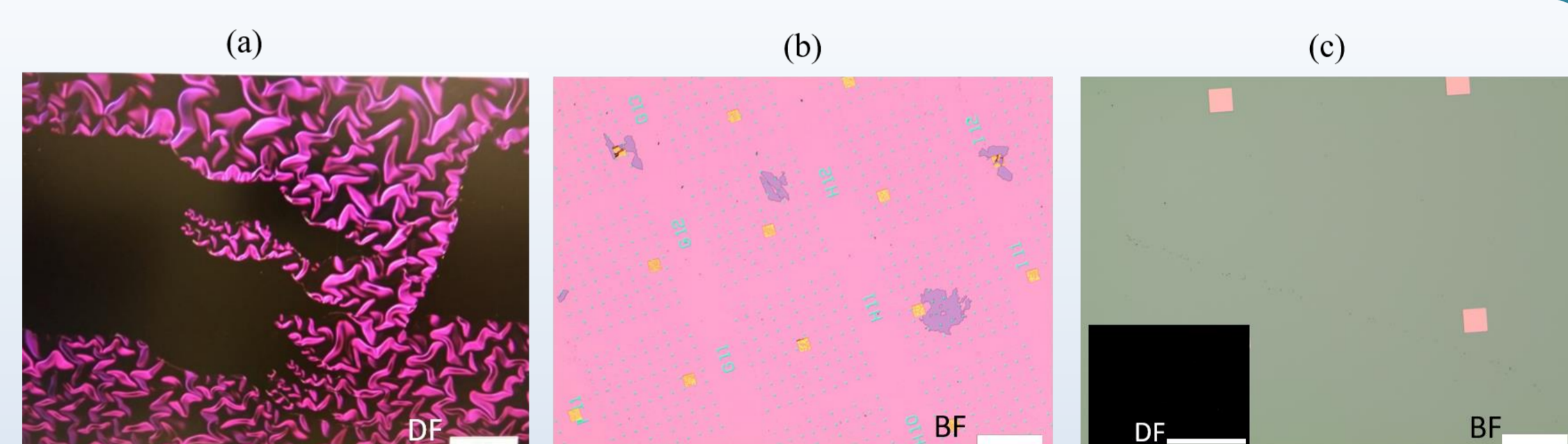


Fig. 2 SiO₂ layer adhesion. Optical microscopy images of the e-beam SiO₂ layer on the fused quartz substrate depending on the protocol of fabrication: (a) after a standard acetone lift-off (dark-field image), (b) with an additional exposure to remover PG (bright-field image), (c) with an additional annealing at 200°C after e-beam deposition (bright-field image); the inset in (c) shows a dark field image displaying null contrast. Scale bars in (a,b,c) are 500 μ m, and 100 μ m in the inset of (c).

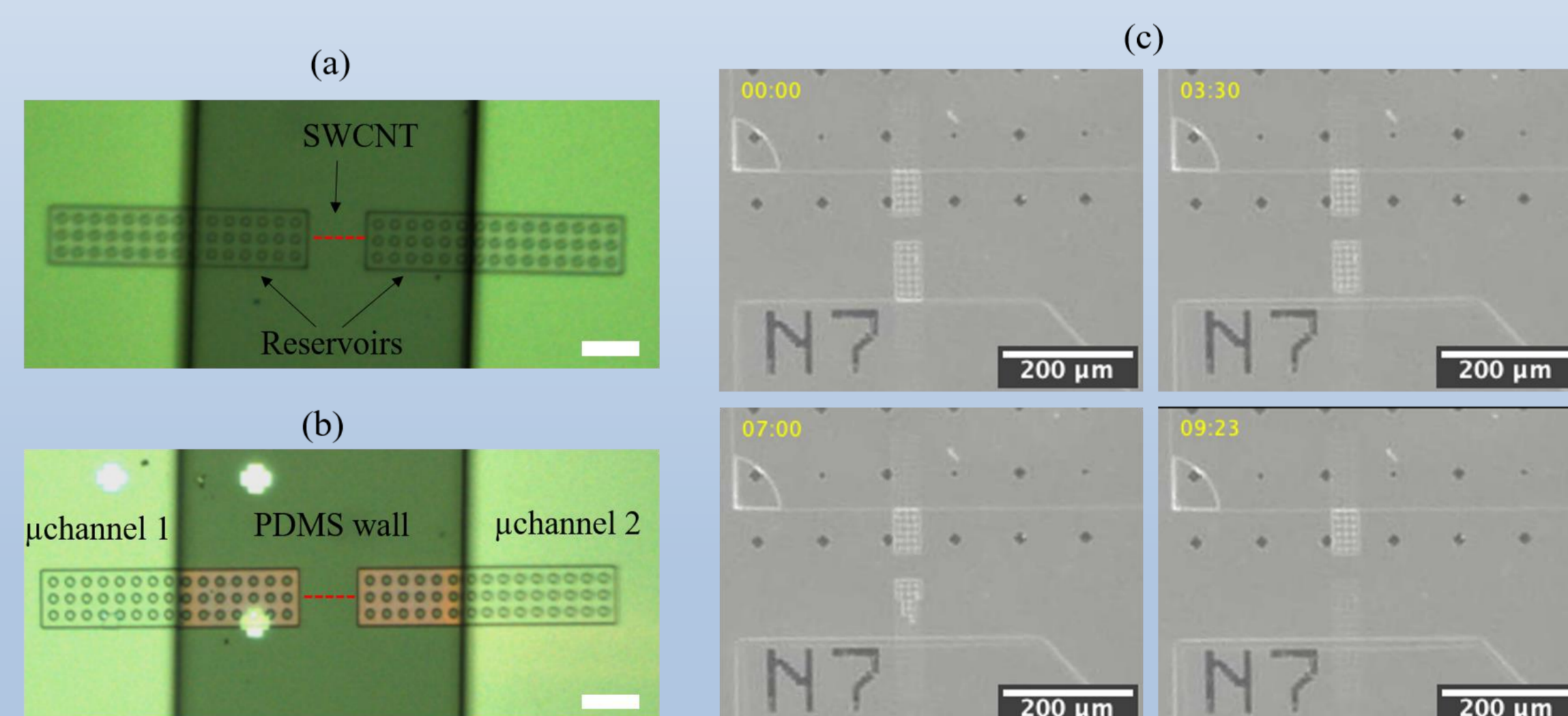


Fig. 3 Plug-free and bubble-free devices. (a,b) Post-fabrication optical microscopy images showing reservoirs (a) plugged and (b) not plugged by PDMS. The scale bars in (a,b) are 40 μ m. (c) Images taken with a stereomicroscope during water filling: on these images, a pressure of 200 mbar is applied in the lower channel and no pressure in the upper channel.

Ultra-low noise measurements of ionic transport

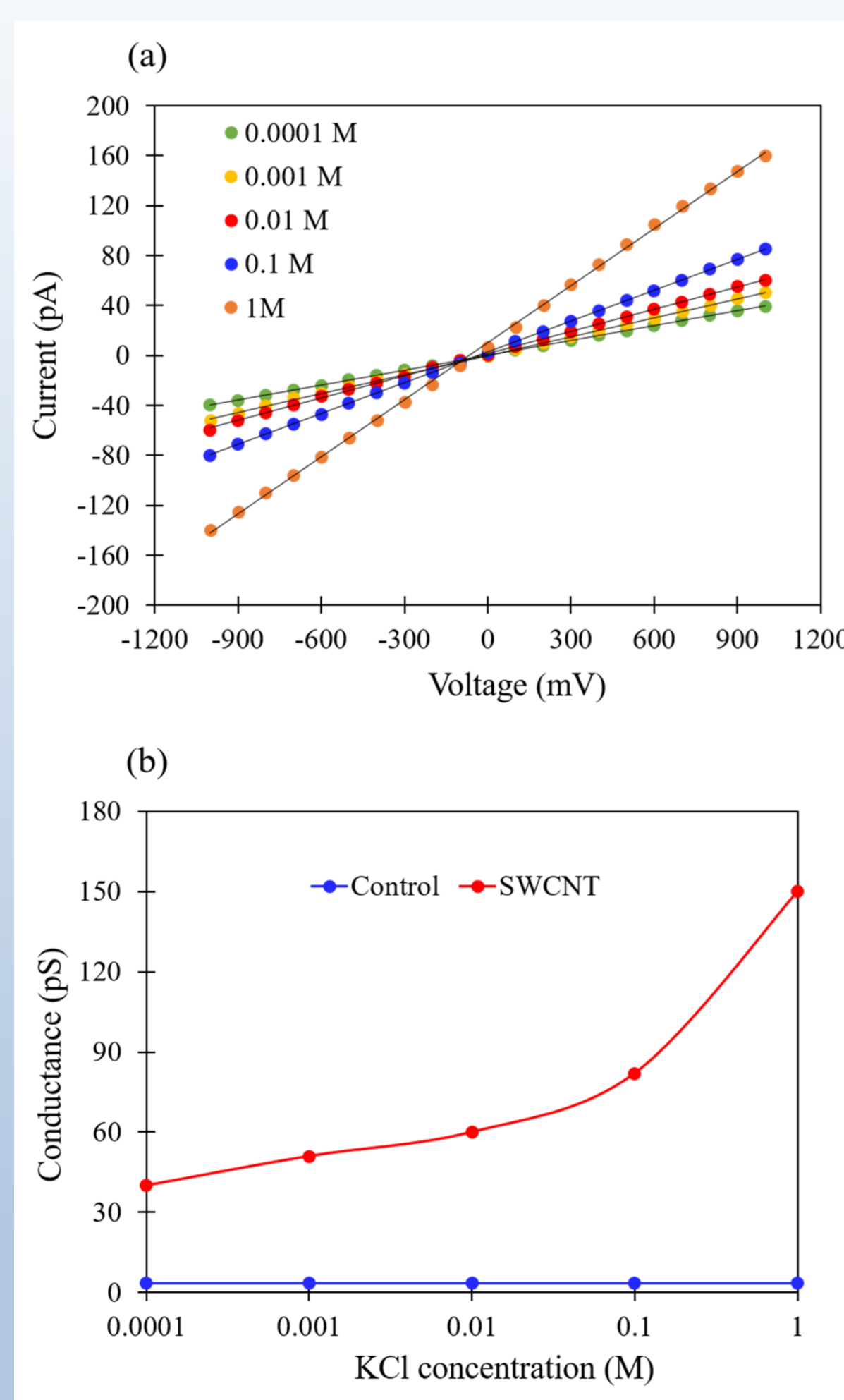


Fig. 4 Ionic transport. (a) Ionic current as a function of applied voltage, for different KCl concentrations (CNT side). (b) Ionic conductance as a function of KCl concentration: CNT (red points) and control (blue points).

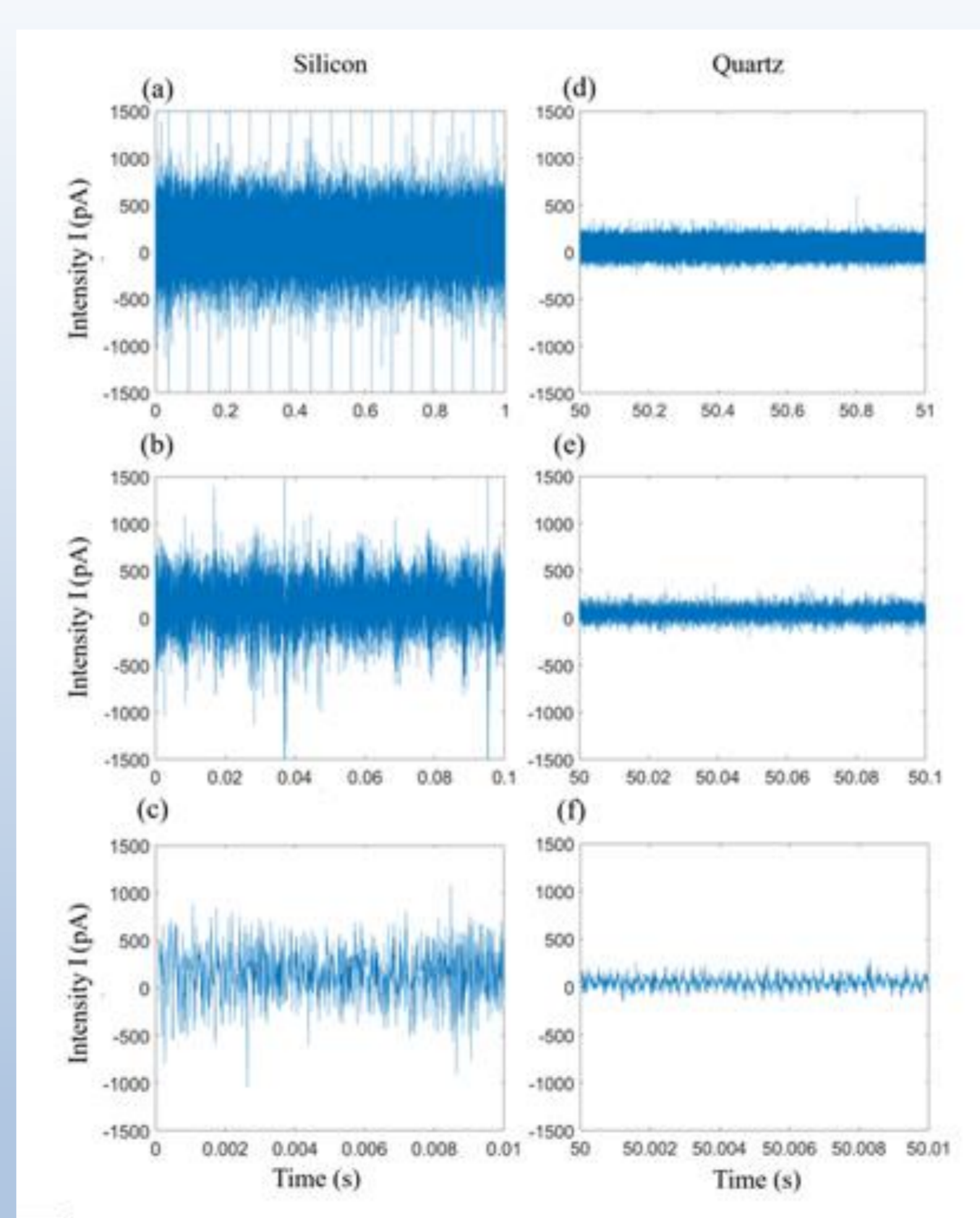


Fig. 5. Noise analysis. (a-f) Current traces for a CNT nanofluidic device on a SiO₂/Si substrate (a-c) and on a quartz substrate (d-f) at different time scales. The KCl concentration is 1 M and the sampling rate is 100 kHz in both cases. Devices: LB13 (Quartz) and LB18 (Silicon). (g) Comparison of the background noise spectra for a device on silicon and two devices on fused quartz substrates ([KCl] = 1 M in all cases). (h) LPSD for CNT and control at +1 V (device CR05). This CNT LPSD is best fitted with a power law $1/f^a + b$ with $a = -1.06$ (green curve). The CNT LPSD at 0 V is also shown as reference.

