Electronic Supplementary Material (ESI) for Chemical Science

Supporting Information

Transducing Methyltransferase Activity into Electrical Signals in a Carbon Nanotube-DNA Device

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SWCNT FET Fabrication:

The detailed procedure has been previously described (see reference 1). As before, we grew the individual SWNTs by chemical vapor deposition on degenerately doped silicon wafers with 300 nm SiO_2 on their surface. The SWNTs grown in this way are □1 to 2 nm in diameter. Metallic electrodes with a 20 μm separation (5 nm of Cr overlaid by 50 nm of Au) were deposited through a shadow mask onto the SWNTs. The nanotube devices were electrically tested using the metal pads as source and drain contacts and the silicon substrate as a global back gate.

SWCNT Cutting Procedure:

The procedure has been previously described (see reference 1). A window that is less than 10 nm in size was opened in a spin-cast layer of polymethylmethacrylate (PMMA) by using ultrahigh-resolution electron-beam lithography. The nanotubes were then locally cut through the open window via oxygen plasma ion etching (the operation conditions were 250 mTorr, 50 W RF power, 10 s exposure).

DNA Synthesis:

The procedure was adapted from reference 2. Amine-modified and unmodified oligonucleotides were prepared using standard phosphoramidite chemistry on an Applied Biosystems 3400 DNA synthesizer. Oligonucleotides modified with amines on both the 3’ and 5’ termini were prepared via solid phase synthesis using

reagents purchased from Glen Research, Inc. The solid phase synthesis was performed on 3'-PT-Amino-Modifier C3 CPG with the 5'-Amino-Modifier C3-TFA phosphoramidite added in the final step of the solid phase synthesis to leave protected amines at both the 3' and 5' ends. In all instances, the oligonucleotides were cleaved from the resin with concentrated ammonium hydroxide before being stringently purified with a C18 column. The purified oligonucleotides were quantified via UV-Visible spectroscopy and characterized via MALDI mass spectrometry. Complementary single strand DNA was hybridized by heating equimolar amounts of each strand in buffer containing 5 mM phosphate, pH = 7.1, 50 mM NaCl to 90 °C, followed by cooling to ambient temperature.

DNA Reconnection:

The procedure for DNA reconnection has been described in reference 2. Oxidatively-cut devices prepared above were incubated overnight in BupH^{TM} MES buffered Saline solution (pH 4.7, Pierce Biotech.) containing 5 mM EDCI and 10 mM Sulfo-NHS. The devices were then removed from the solution, washed with fresh buffer solution, and dried with a stream of nitrogen gas for device characterization. The as-formed devices were next incubated in the BupH^{TM} Phosphate Buffered Saline solution (pH 7.2, Pierce Biotech.) containing 10 μM amine-modified duplex DNA to form amide bonds between the DNA and carbon nanotube. The bridged devices were removed from the solution, washed with fresh buffer, and dried with a stream of nitrogen gas for device characterization.

DNA Methylation and Protein Detection:

The procedure used is the recommended one from the vendor of M.*Sss*I, New England Biolabs. It is at the following url:

http://www.neb.com/nebecomm/products/productM0226.asp

The DNA-bridged devices were incubated in NEBuffer 2 solution (pH 7.9, *New England Biolabs Inc.*) containing 6 nM of M.*Sss*I (*New England Biolabs Inc.*) and 300μM SAM (*New England Biolabs Inc.*) at 37 °C for 1 hour. Subsequently, the devices were removed from the solution, and dried with a stream of nitrogen gas for electrical characterization. The experiments without the SAM cofactor were performed under the same conditions, with the 300 μM SAM excluded.

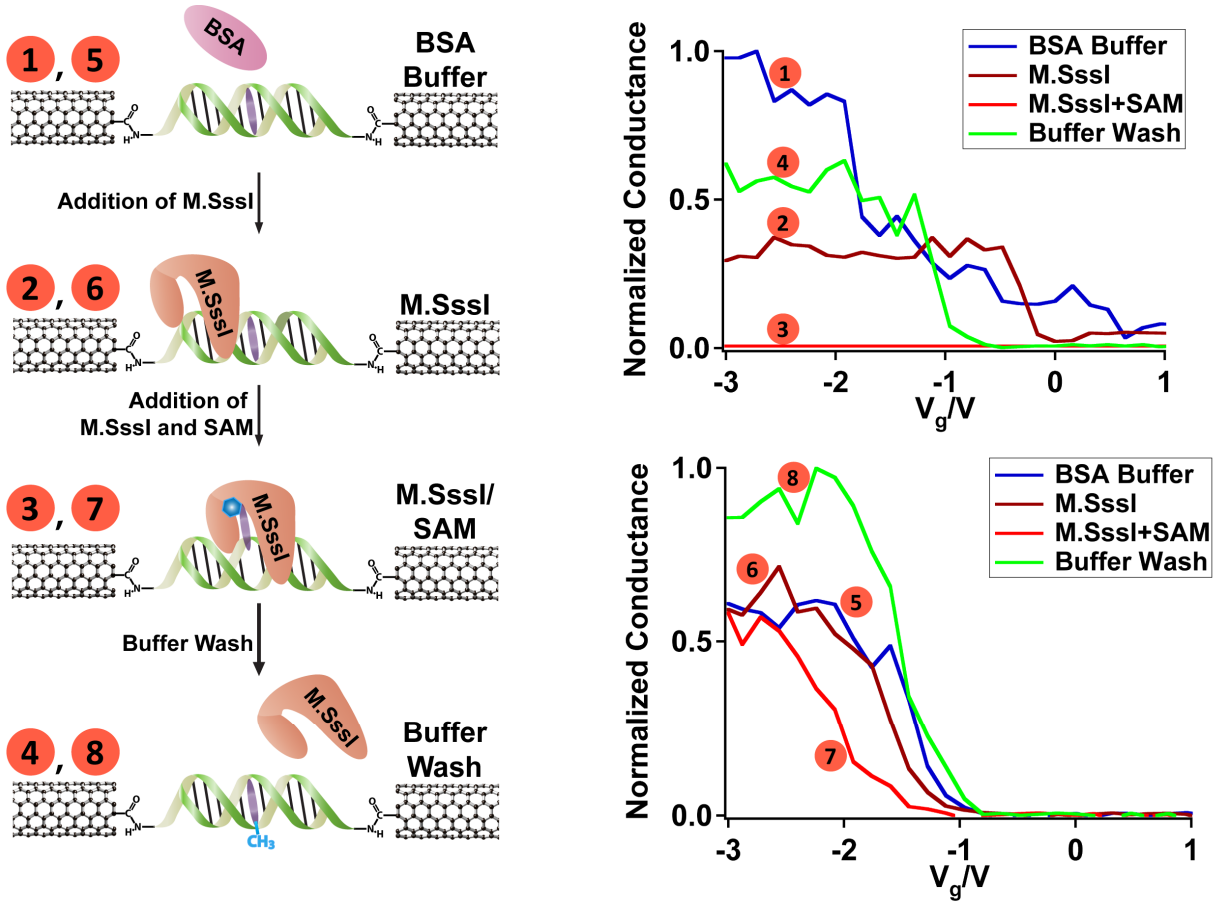

Fig. S1 Normalized conductance curves that correspond to Figure 4 in the text. Left: Protein detection at a DNA bridged device. Right: The normalized conductance curves of steps 1-4 are in the upper right panel and steps 5-8 are in the lower right penal. The buffer wash conditions are 50 mM NaCl, 10 mM Tris-HCl, 10 mM $MgCl_2$, 1 mM Dithiothreitol, pH 7.9. The sequence was NH_2-5'-GACAGTCGACATGTC -3'-NH_2.

References

1) X. Guo, J. P. Small, J. E. Klare, Y. Wang, M. S. Purewal, I. W. Tam, B. H. Hong, R. Caldwell, L. Huang, S. O'Brien, J. Yan, R. Breslow, S. J. Wind, J. Hone, P. Kim, C. Nuckolls, *Science.* **2006***, 311,* 356–359.

2) X. Guo, A. A. Gorodetsky, J. Hone, J. K. Barton, C. Nuckolls, *Nat. Nanotechnol.* **2008,** *3,* 163-167.