Probing Electrostatic Potentials in Solution with Carbon Nanotube Transistors

CNF Project # 900-00
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Abstract:

We have used single-walled carbon nanotube transistors to measure changes in the chemical potential of a solution due to redox-active transition metal complexes. The interaction of the molecules with a gold electrolyte-gate wire changes the electrostatic potential sensed by the nanotube, which in turn shifts the gate-voltage dependence of the nanotube conductance.

Summary:

Carbon nanotubes (NTs) are promising candidates for nanoscale electrochemistry experiments. Semiconducting NTs can be used as field-effect transistors (FETs) in an electrolyte environment [1], and their high sensitivity makes them excellent sensors of molecules in solution (e.g., [2]). In previous sensing experiments, the analyte caused a shift in gate-voltage dependence of the nanotube conductance, which has been attributed to charge transfer from adsorbed molecules to the nanotube. We have explored the response of single-walled NT FETs to redox-active transition metal complexes, and have found that the signal stems from the changing chemical potential of the solution and does not depend on the proximity of the molecules to the NT [3].

We fabricated CVD-grown NTs in a FET geometry at the CNF, with 10 µm between the gold source and drain electrodes. A 60 by 25 µm microfluidic PDMS channel was sealed over the NT, and a gold wire in a reservoir on the end of the channel was used to set the electrochemical potential of the solution [1]. We introduced different redox-active molecules into the reservoir and studied the electrical response of the NT FET.

We found that oxidizing molecules generally caused a positive shift in the NT threshold voltage, whereas reducing molecules caused a negative threshold voltage shift. We explored this more carefully by making simultaneous measurements with a traditional electrochemical cyclic voltammetry setup, and we found that the threshold voltage shift varied with the change in chemical potential of the solution, as determined by the Nernst equation. By controlling the flow in the channel, we also determined that the response depended only on whether the molecules were near the electrolyte-gate wire, and not on their proximity to the NT.

To understand these data, we recall that the voltage applied to the electrolyte-gate wire sets the electrochemical potential of the solution, which is the sum of the chemical potential (set by the molecules in solution) and the electrostatic potential. If the nanotube senses only the electrostatic potential, then the shift in threshold voltage will simply be the change in the chemical potential, which agrees with our data. The source of the signal is the electrochemical interaction between molecules and electrolyte-gate wire, and the proximity of the molecules to the NT is irrelevant, as we observed.

To confirm this model, we measured the nanotube conductance versus both the voltage on the electrolyte-gate wire, which sets the electrochemical potential, and the potential of a Ag/AgCl reference electrode, which measures only the electrostatic potential. As expected, when we added a redox-active molecule, the conductance shifted versus the gate voltage, but it remained unchanged versus the reference electrode potential. We therefore conclude that NT FETs can be used as nanoscale reference electrodes to measure the electrostatic potential of a solution, which could lead to new kinds of nanoscale electrochemistry experiments.

References:


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Figure 1, top left: Electrolyte-gated NT FET. The gate wire establishes the electrochemical potential of the solution, which is the sum of the chemical potential set by the molecules and the electrostatic potential.

Figure 2, bottom left: Threshold voltage shift varies with change in chemical potential (oxidizing molecules cause a positive shift, reducing cause a negative shift).

Figure 3, above: The NT measures only the changing electrostatic potential, acting as a nanoscale reference electrode.