Supporting Online Material for

**Fuel-Powered Artificial Muscles**

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I. Actuator Materials
Cantilever-based nanotube actuators used in Fig. 1A, which comprise carbon nanotube sheets laminated with a layer containing Pt catalyst, were made by sequential vacuum filtration of (1) 100 ml of an aqueous solution comprising an ultrasonically dispersed mixture of 10 mg of SWNTs (HiPco nanotubes from Carbon Nanotechnologies Inc.) and 0.2 g of Triton X-100 surfactant (Aldrich) to make the first layer of the cantilever and (2) 6 ml of an aqueous solution containing an ultrasonically dispersed mixture of 60 mg Triton X-100 and 6 to 10 mg carbon-supported Pt catalyst (80 wt. % of 1:1 by weight Pt on carbon from Engelhard Corporation, mixed with 20 wt. % Nafion ionomer from DuPont) to form the opposing layer of the cantilever. After air drying and peeling from the 45 mm diameter, 5.0 $\mu$m pore size poly(tetrafluoroethylene) filter, the bilayer sheet was cut with a razor blade to form 3 mm x 30 mm x 26 $\mu$m cantilever strips. The SWNT sheet strips used for tensile actuator measurements (in the one-compartment cell of Fig. 1B) were made analogously, except that the dispersions in (1) and (2) were mixed and vacuum filtered in a one-step process. As evidenced by SEM micrographs, these latter sheets contain rather uniformly distributed Pt catalyst, while sheets made by successive filtration of nanotubes and Pt show separate nanotube and Pt-containing layers. The NiTi shape memory alloy used for the continuously shorted fuel cell muscles was obtained from Dynalloy, Inc. as a 152 $\mu$m diameter Flexinol® wire having a 70°C austenitic phase transition. These wires were used as received, without further thermomechanical treatment. The Pt black catalyst (HiSPEC 1000 with a nominal surface area of 27 m$^2$g$^{-1}$ from Alpha Aesar) was attached to the shape memory wire by immersing the wire in a Pt-hexane slurry, evaporating the hexane, and using compressed air to remove unbound catalyst. The resulting weight gain of the shape memory wire was 1.3-1.5%.

II. Actuation and Electrochemical Potential Measurements
Tensile strain measurements as a function of various constant mechanical loads were made for all tensile actuators using the Perkin Elmer Dynamic Mechanical Analyzer (DMA 7e). Electrode potentials during actuation were measured using either saturated calomel or Ag/AgCl reference electrodes, from which potentials versus NHE were derived. Electrical connection to one end of each actuating nanotube electrode was made using Ag-filled epoxy (covered with an insulating epoxy) that connected to platinum wire leads. This end and the opposite end of the actuator strip were clamped in the grips of the DMA. The counter-electrode was a high-surface-area carbon felt contacted by Pt wire. The nanotube electrode in the one-compartment cell was initially cycled in the electrolyte to maximize electrode wetting. This was accomplished by cycling the fuel and oxidant (separated by a N$_2$ sweep of the cell) or by sweeping potential in the 0 to 1 V range (vs. NHE). For the one compartment nanotube fuel cell muscle, strain and potential changes due to the oxidation and reduction of H$_2$ and O$_2$, respectively, were measured by purging the cell with oxygen and hydrogen for
~30-45 minutes for each gas with a few minutes of nitrogen purge in between gases to avoid direct contact of H\textsubscript{2} and O\textsubscript{2} in the presence of Pt catalyst. For comparison purposes, the same carbon nanotube composite electrode used in the nanotube fuel cell muscle was also used as the working electrode in the electrically powered muscle. In the case of the electrically powered muscle, strains at varying applied potentials were measured and compared to the gas-induced (H\textsubscript{2} and O\textsubscript{2}) strains.

Fig. S1. Charge (red) and discharge (blue) curves for a cantilever-based nanotube fuel cell muscle. The observed actuator stroke during chemically driven charge injection was a 2 mm deflection of a 3 cm long nanotube cantilever in ~5 seconds, as the nanotube electrode potential increased to ~0.8 V (versus NHE). The opposite actuator deflection, obtained when the hydrogen and oxygen electrodes were shorted, occurred in a second. Breaking the connection between electrodes caused recharging of the nanotube muscle and return to the deflection of the initially charged state. The rates for the charge and discharge parts of the actuation cycle differ, likely because the charging rate is controlled by gas diffusion to the opposite electrodes and oxidation and reduction at these electrodes to form electrochemical double layers, while the discharge rate is controlled by H\textsuperscript{+} diffusion and electron transport between electrodes.