461c Gated Chemical Transport through Vertically Aligned Carbon Nanotube Membranes

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A promising architecture for ion-channel mimetics is a composite membrane structure containing vertically aligned carbon nanotubes, with inner core diameters of 7 nm, passing across a polystyrene matrix film. Plasma oxidation during the fabrication process introduces carboxylic acid groups on the CNT tips that are modified using carbodiimide mediated coupling between carboxylic acid on the CNTs and accessible amine groups of the functional molecule. The entrances to CNT's cores were thus functionalized with aliphatic amines of different lengths, charged dye molecule and an aliphatic amine elongated by spacers containing poly-peptides. The transport through the membrane of two differently sized but equally charged molecules, ruthenium bi-pyridine [Ru-(bipy)3+2] and methyl viologen [MV+2], was studied in a U-tube permeation cell with flux quantified by UV-vis Spectroscopy. Relative selectivity of the permeates was seen to vary from 1.9 to 3.6 as a function of tip-functionalization chemistry. Anionic charged functional groups are seen to sharply increase flux of cationic permeates. This effect is reduced at higher solution ionic strength consistent with shorter Debye screening length. Using a hindered diffusion to model observed selectivities was consistent only with a geometry of only CNT tip functionalization, not along the length of CNT core. Biologically active desthiobiotin was also shown to be reversibly coordinated to stretavidin, showing a reduction in ionic flux through CNT. Pressure driven flux of a variety of solvents (H2O, hexane, decane ethanol, methanol) are highly accelerated deviating by 5 orders of magnitude from classic Newtonian flow.