Supporting Information

Reverse translocation of nucleotides through a carbon nanotube

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Radial distribution function (RDF)

In Figure S1, we show the calculated ion-water RDFs inside and outside the CNT. Figure S1a shows the ion (K^+ or Cl^-)-oxygen-water RDFs in a 1 M KCl solution. Figure S2 displays the radial distribution functions (RDFs) of ion-oxygen-water when the ion (K^+ or Cl^- is inside the CNT of diameter 1.36 nm) and the surrounding oxygen on a water molecule can be either inside or outside the tube.

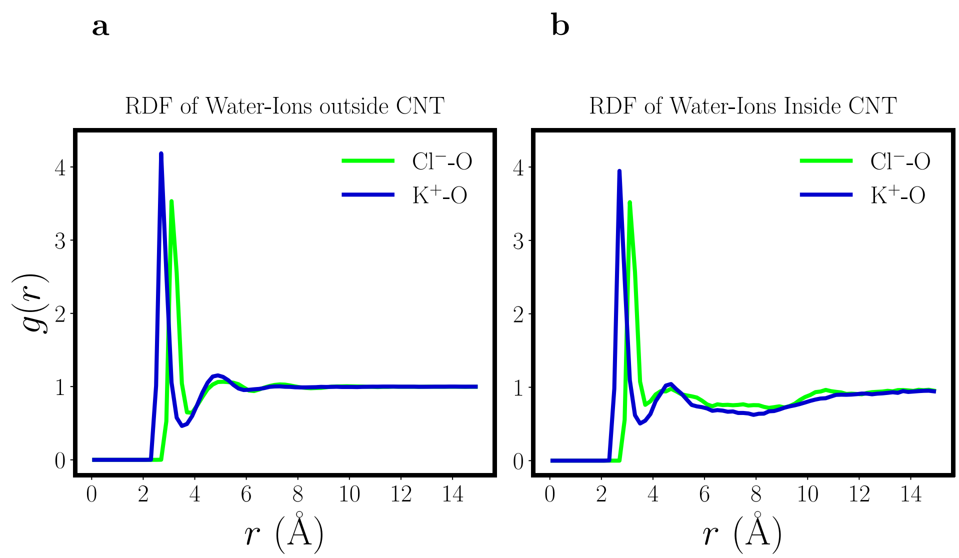

Figure S1 Radial distribution function (RDF) of water-ions. a. RDF of water (Oxygen) molecules with Cl^- and K^+ ions inside the CNT b. RDF of water (Oxygen) molecules with Cl^- and K^+ ions outside the CNT

These distribution functions are similar to the corresponding distribution functions in bulk water displayed in Figure S1a. However, in Figure S1b the number of water molecules around ions decreases on average in region between 6-10 Å due to presence of CNT. The results show a smaller solvation radius for K^+ ions compared with Cl^- ions.

Figure S2 shows the RDF of the adenine (center-of-mass)-ion (K^+ or Cl^-) with a significantly higher number of K^+ ions in the first shell compared with the number of Cl^- ions around the nucleotide, due to the negative charge on the adenine nucleotide.

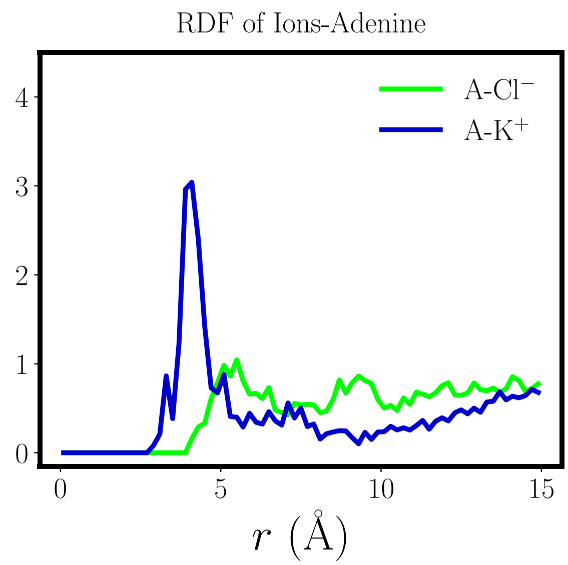

Figure S2. Radial distribution function (RDF) of water molecules with Cl^- and K^+ inside the CNT.

Figure S2 indicates that there are more K^+ ions around the nucleotide rather than Cl^- ions, which is justified by the opposite charges of the phosphate group in the nucleotide and K^+ ions that generate attractive forces. The RDFs confirm the high affinity of the K^+ ion for the nucleotide in comparison to the Cl^- ion.

A video of reverse translocation of single adenine nucleotide through a CNT with a diameter of 1.36 nm (mp4) is included as supporting information.

Water Flow

Figure S3-5 show the cumulative flow of water inside and outside the narrow CNT of diameter 1.36 nm (inner diameter 1.11 nm) under different voltages of ~0.17V/nm, ~0.35V/nm, and 1V/nm. The water in Figure S3-5 flows in direction of K^+ ions through narrow tube and the negatively charged nucleotide translocates in same direction (reverse translocation).

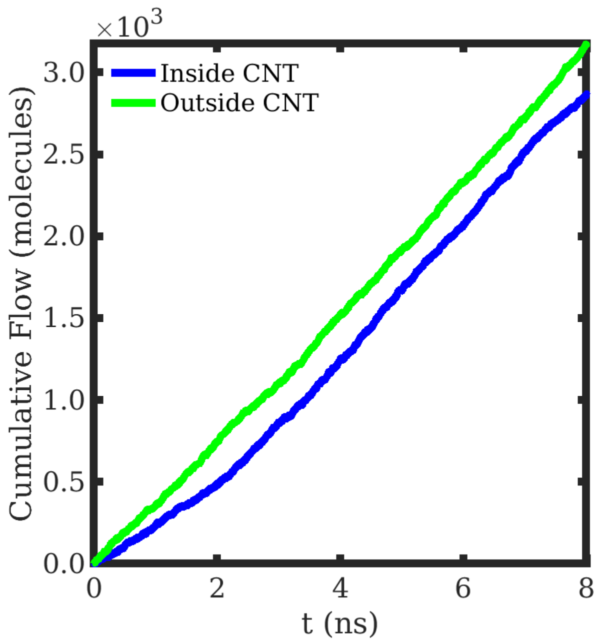

Figure S3. Cumulative water flow through the CNT of length 50 nm and diameter 1.36 nm under ~0.17 V/nm applied electric field with adenine inside and outside the CNT.

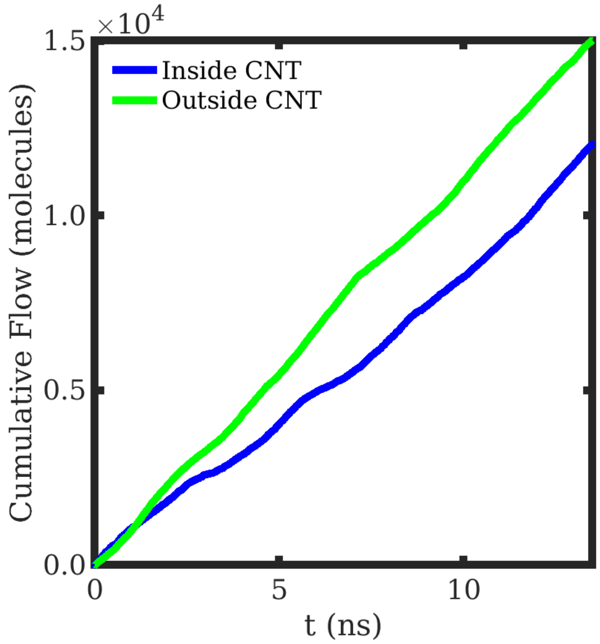

Figure S4. Cumulative water flow through the CNT with a length 50 nm and diameter 1.36 nm under ~0.35 V/nm applied electric field.

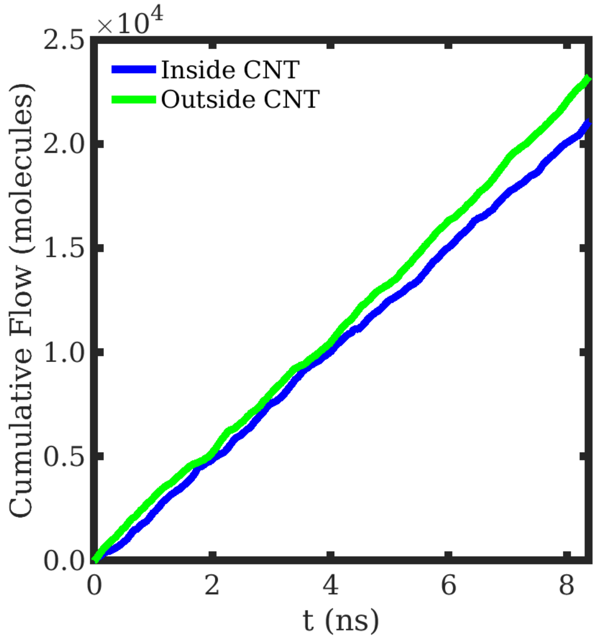

Figure S5. Cumulative water flow through the CNT with a length of 50 nm and a diameter of 1.36 nm under 1 V/nm applied electric field.

Figure S6 shows the cumulative flow of water inside and outside the wide tube. The water in Figure S6 flow in direction of Cl^- through wide tube where nucleotide translocates through same direction (normal translocation).

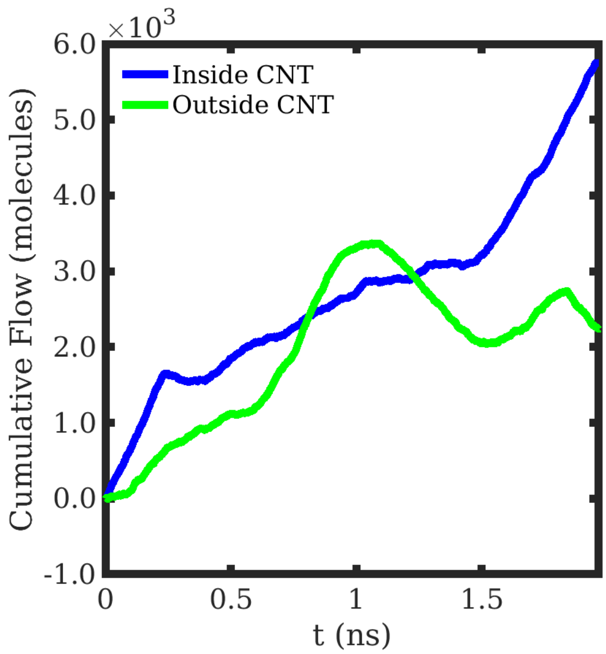

Figure S6. Cumulative water flow through the CNT with a length 20 nm and a diameter of 2.71 nm under 1 V/nm applied electric field.

Incorporation of a membrane in MD simulations

To investigate this, the translocation behavior of the nucleotide through CNT with a graphene nanosheet as a membrane, we developed a model presented in Figure S7a-c.

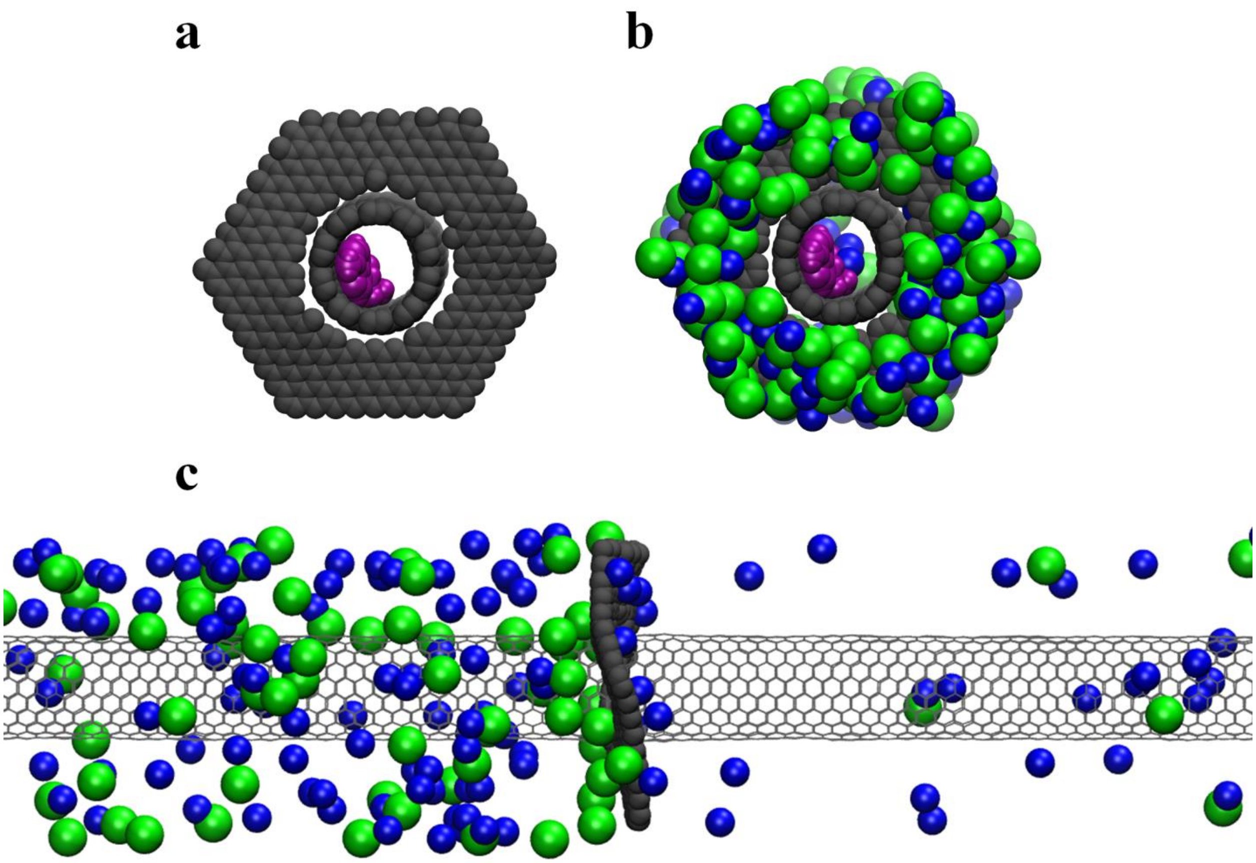

Figure S7. The CNT system with a graphene nanosheet as membrane in the middle of CNT. (a) A Representation of the graphene (grey sheet), CNT (grey cylinder), and a single nucleotide (purple) from front perspective. (b) A representation of Cl^- (green spheres) and K^+ (blue sphere) ions in the system.

The reverse translocation of nucleotide inside the CNT in the presence of graphene membrane was observed until the nucleotide reached the region in middle of the CNT next to the graphene membrane. In this region, we observed a resistance in the reverse translocation of nucleotide which is tentatively caused by accumulation of Cl^- and K^+ ions in either side of the graphene nanosheet outside of the CNT. Figure S7c shows there are numerous Cl^- ions accumulated on one side of the graphene nanosheet because of the force applied by electric field on Cl^-. Similarly, in the other side of the nanosheet, several K^+ ions are accumulated due to the electric force applied on K^+ ions in the direction they accumulate. Changing the position and composition of the membrane can change the behavior of ions and as a result affect translocation of the nucleotide.