Supporting Information for: Graphene Nanopores for Protein Sequencing

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Table S1: Summary of the MD simulations of the three-layer graphene membrane systems. The equilibration simulations of the first four peptide systems (listed above the horizontal line) began with the peptide chains threaded through the nanopore in an extended conformation, Figure 1a. The remaining systems (listed below the horizontal line) were built using already equilibrated peptide conformations. Simulations of water flow-driven transport are summarized in Table S3.
† Equilibration time in addition to 116 ns equilibration of $(FGFG)_{12}$.
∗ Equilibration time in addition to 125 ns equilibration of $(FKFG)_{12}$.
‡ Equilibration time in addition to 129 ns equilibration of $(FDFG)_{12}$.

Peptide	**Charge**	**Equilibration time**	**Voltage bias**	**Duration of transport simulations**		
				Run 1	Run 2	Run 3
AFKVQLQL PDNEVAQI SDYYPRNS IDTKEYMS TLTYGFNG NVTGDDTG	−4e	137 ns	1 V	905ns		
$(FDFG)_{12}$	−12e	129 ns	1 V	127 ns	60 ns	240 ns
$(FKFG)_{12}$	12e	125 ns	1 V	241 ns		
$(FGFG)_{12}$	0e	116 ns	1 V	147 ns		
$(FKDG)_{12}$	0e	43† ns	1 V	161 ns		
$(FKFQ)_{12}$	12e	7* ns	1 V	655 ns		
$(FKFK)_{12}$	24e	30† ns	1 V	72 ns	40 ns	30 ns
$(FRFG)_{12}$	12e	38† ns	1 V	137 ns	120 ns	210 ns
$(FDFQ)_{12}$	−12e	17‡ ns	1 V	662 ns		
$(FDFD)_{12}$	−24e	5† ns	500 mV -500 mV 1 V -1 V	70 ns 210 ns 27 ns 7 ns	120 ns 210 ns 60 ns 10 ns	120 ns 240 ns
$(FEFG)_{12}$	−12e	5† ns	1 V	167 ns	120 ns	210 ns
$(FGDG)_{12}$	−12e	5† ns	500 mV 1 V	180 ns 65 ns	210 ns 60 ns	120 ns 30 ns

Table S2: Summary of the simulations performed to determine the effect of the graphene membrane thickness and the peptide charge density on the translocation rate, Figure 2 of the main text. The initial configuration of the $(DGDG)_{12}$ system was obtained using the equilibrated conformation of the $(FDFD)_{12}$ peptide by replacing the amino acid side chains. The initial configurations of all other peptides were derived from the final equilibrated state of the $(DGDG)_{12}$ peptide. All peptide transport simulations were performed under a 500 mV transmembrane bias.

Peptide	**Charge**	**Equilibration time**	**Graphene layers**	**Duration of transport simulations**		
				Run 1	Run 2	Run3
$(DGDG)_{12}$	$-24e$	22 ns	1	12 ns	14 ns	14 ns
$(DGG)_{16}$	$-16e$	33 ns	1	13 ns	13 ns	13 ns
$(DGGG)_{12}$	$-12e$	34 ns	1	57 ns	47 ns	70 ns
$(DGGGG)_9DGG$	$-10e$	20 ns	1	50 ns	47 ns	85 ns
$(DGGGGG)_8$	$-8e$	20 ns	1	35 ns	35 ns	23 ns
$(DGGGGGGGG)_5DGG$	$-6e$	16 ns	1	58 ns	51 ns	84 ns
$(DGG)_{16}$	$-16e$	30 ns	2	15 ns	15 ns	15 ns
$(DGG)_{16}$	$-12e$	29 ns	3	45 ns	34 ns	72 ns
$(DGG)_{16}$	$-16e$	40 ns	5	54 ns	80 ns	42 ns

Table S3: Summary of the water flow-driven peptide translocation simulations, Figure 5 of the main text. The initial configuration of the $(FGFG)_{12}$ system was derived from the equilibrated conformation of the $(FQFQ)_{12}$ system.
† Equilibration time in addition to 116 ns equilibration of $(FGFG)_{12}$.

Peptide	**Charge**	**Equilibration time**	**Pore water velocity**	**Duration of transport simulations**
$(FGFG)_{12}$	0e	116 ns	0.29 nm/ns	736 ns
$(FGFG)_{12}$	0e	116 ns	2.85 nm/ns	767 ns
$(FGFG)_{12}$	0e	116 ns	7.17 nm/ns	112 ns
$(FQFQ)_{12}$	0e	10 ns†	0.23 nm/ns	697 ns
$(FQFQ)_{12}$	0e	10 ns†	2.11 nm/ns	665 ns
$(FQFQ)_{12}$	0e	10 ns†	5.66 nm/ns	640 ns

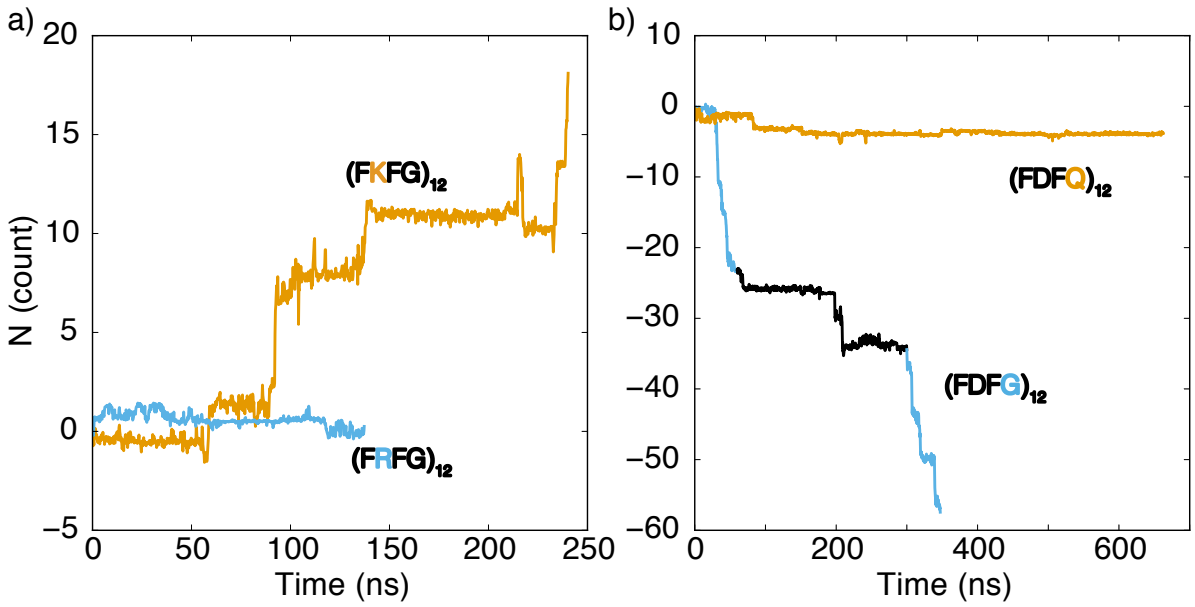


Figure S1: The effect of amino acid substitutions on electrophoretically driven translocation of charged peptides. All simulations featured in this figure were performed using a 2.2 nm diameter nanopore in a three-layer graphene membrane. The translocation progress is characterized by counting the number of amino acids transported through the mid-plane of the membrane in the direction opposite to that of the applied electric field. (a) Permeation traces of the $(FRFG)_{12}$ (blue) and $(FKFG)_{12}$ (orange) peptides under a transmembrane bias of 1 V. (b) Permeation traces of the $(FDFQ)_{12}$ (orange) and $(FDFG)_{12}$ (blue and black) peptides under a transmembrane bias of 1 V. The alternate coloring of the $(FDFG)_{12}$ trace indicates data from independent simulations that were combined to produce the total permeation trace.

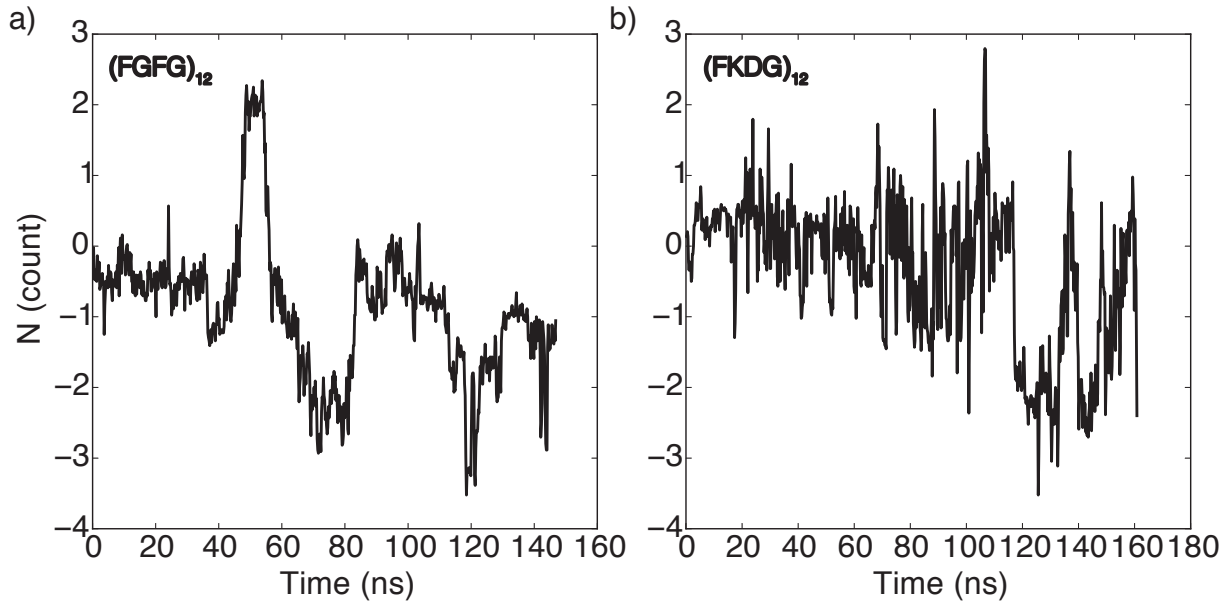
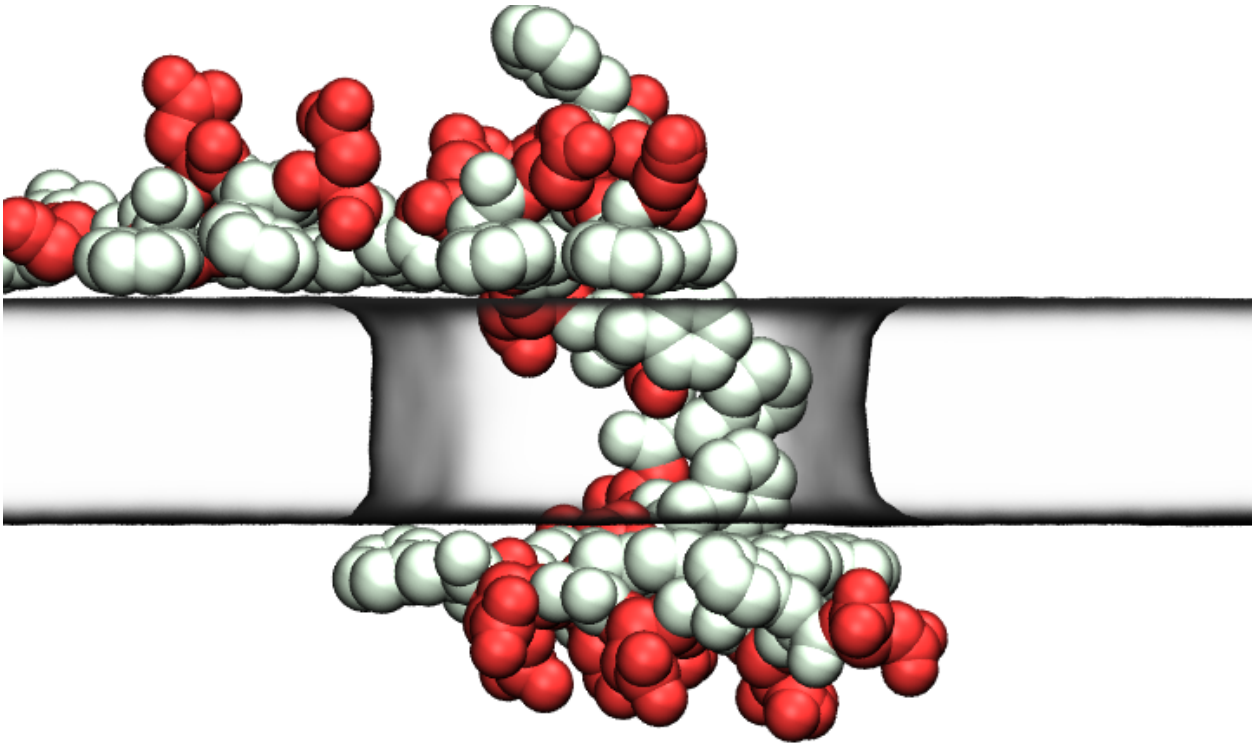
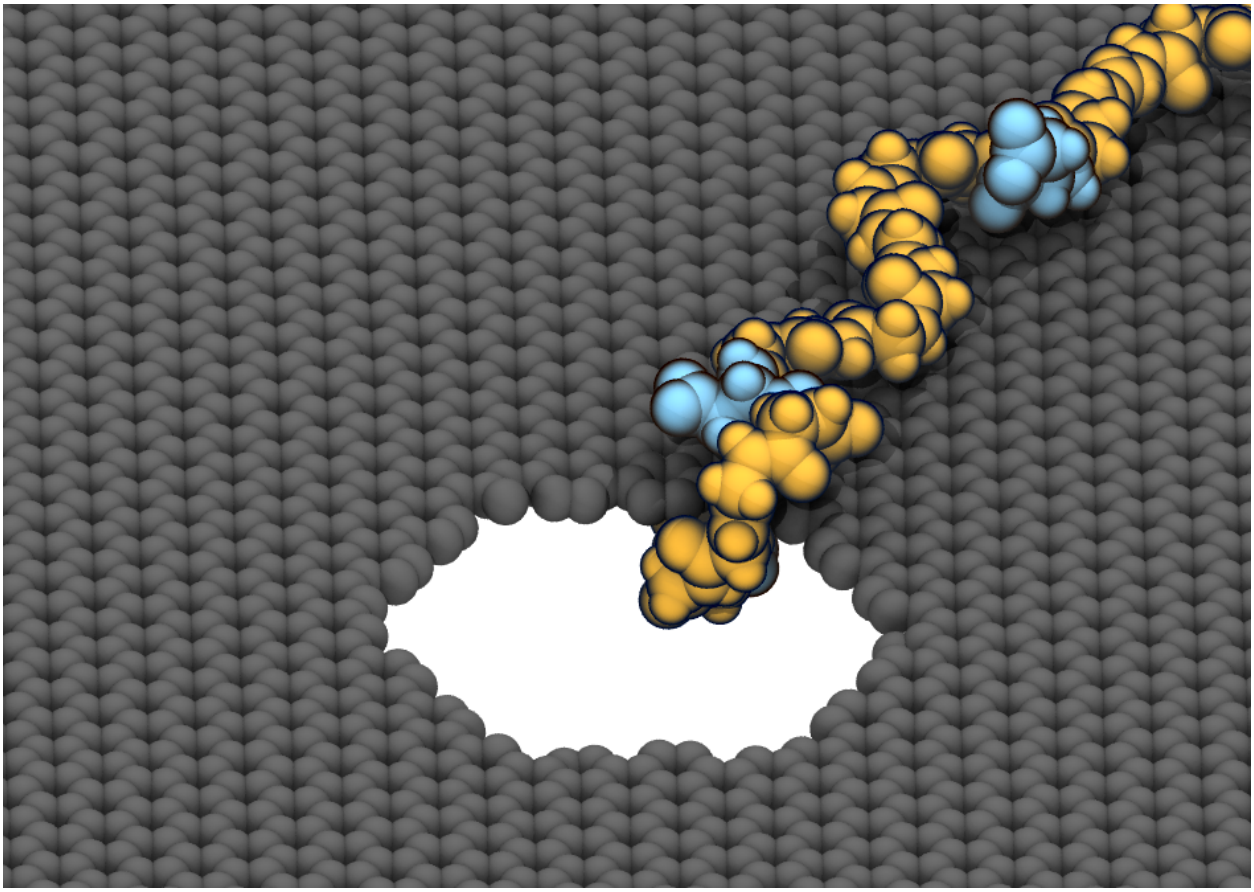


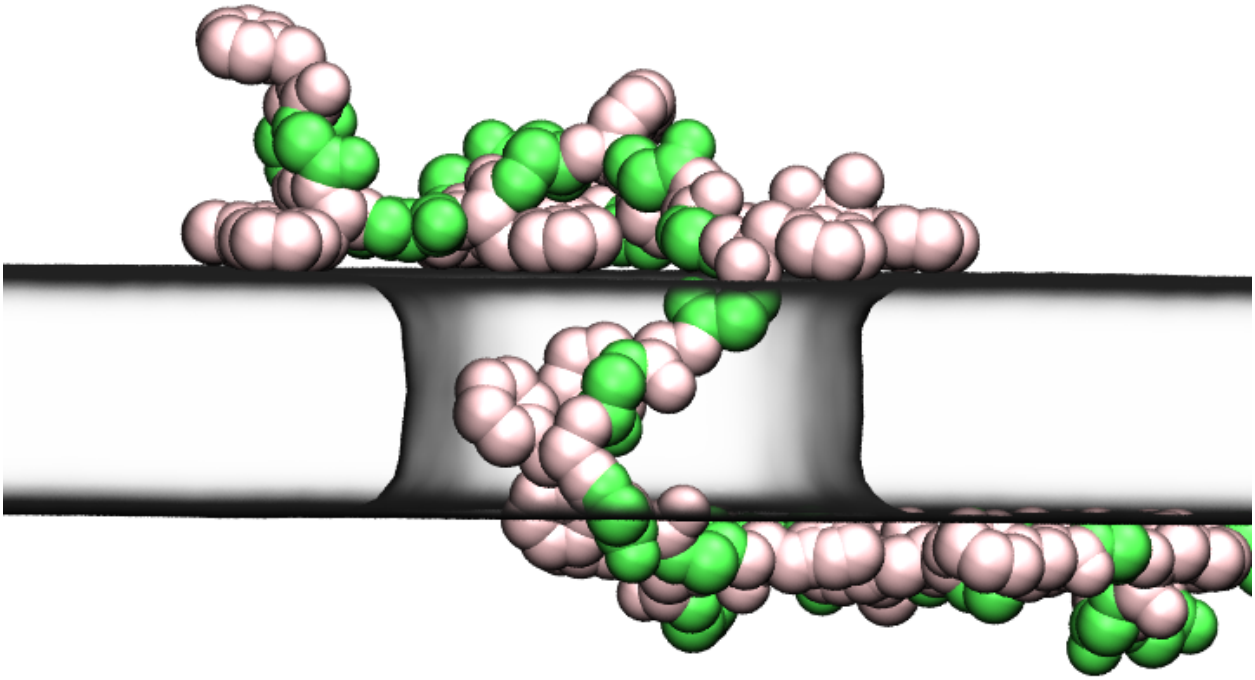
Figure S2: The translocation behavior of two uncharged peptides subject to a transmembrane bias of 1 V. (a,b) The number of amino acids transported through the mid-plane of the membrane in the direction opposite to that of the applied electric field is plotted against the simulation time for the $(FGFG)_{12}$ (panel a) and $(FKDG)_{12}$ (panel b) peptide systems.



Supplementary Movie 1: Electric field-driven transport of an $(FDFD)_{12}$ peptide through a nanopore in a 3-layer graphene membrane. Atoms of the phenylalanine and aspartic acid residues are shown using white and red van der Waals spheres, respectively. Graphene is shown as a translucent surface, with the details of individual atoms smoothed to increase clarity. Water and ions are not shown. Notice the stepwise character of the peptide transport.



Supplementary Movie 2: Electric field-driven transport of a $(DGGGGGGGG)_5DGG$ peptide through a nanopore in a single-layer graphene membrane. Atoms of the aspartic acid and glycine residues are shown using orange and light blue spheres, respectively. Carbon atoms of the graphene membrane are shown using gray van der Waals spheres. Water and ions are not shown.



Supplementary Movie 3: Water flow-driven transport of an $(FGFG)_{12}$ peptide through a nanopore in a 3-layer graphene membrane. A 2.85 nm/ns water flow through the pore was produced by applying a constant force to each of the water molecules present in the system. Atoms of the phenylalanine and glycine residues are shown using white and green van der Waals spheres, respectively. Graphene is shown as a translucent surface, with the details of individual atoms smoothed to increase clarity. Water and ions are not shown. Notice the the stepwise character of the translocation process.