Research demonstrates surface diffusion enhanced ion transport through two-dimensional channels

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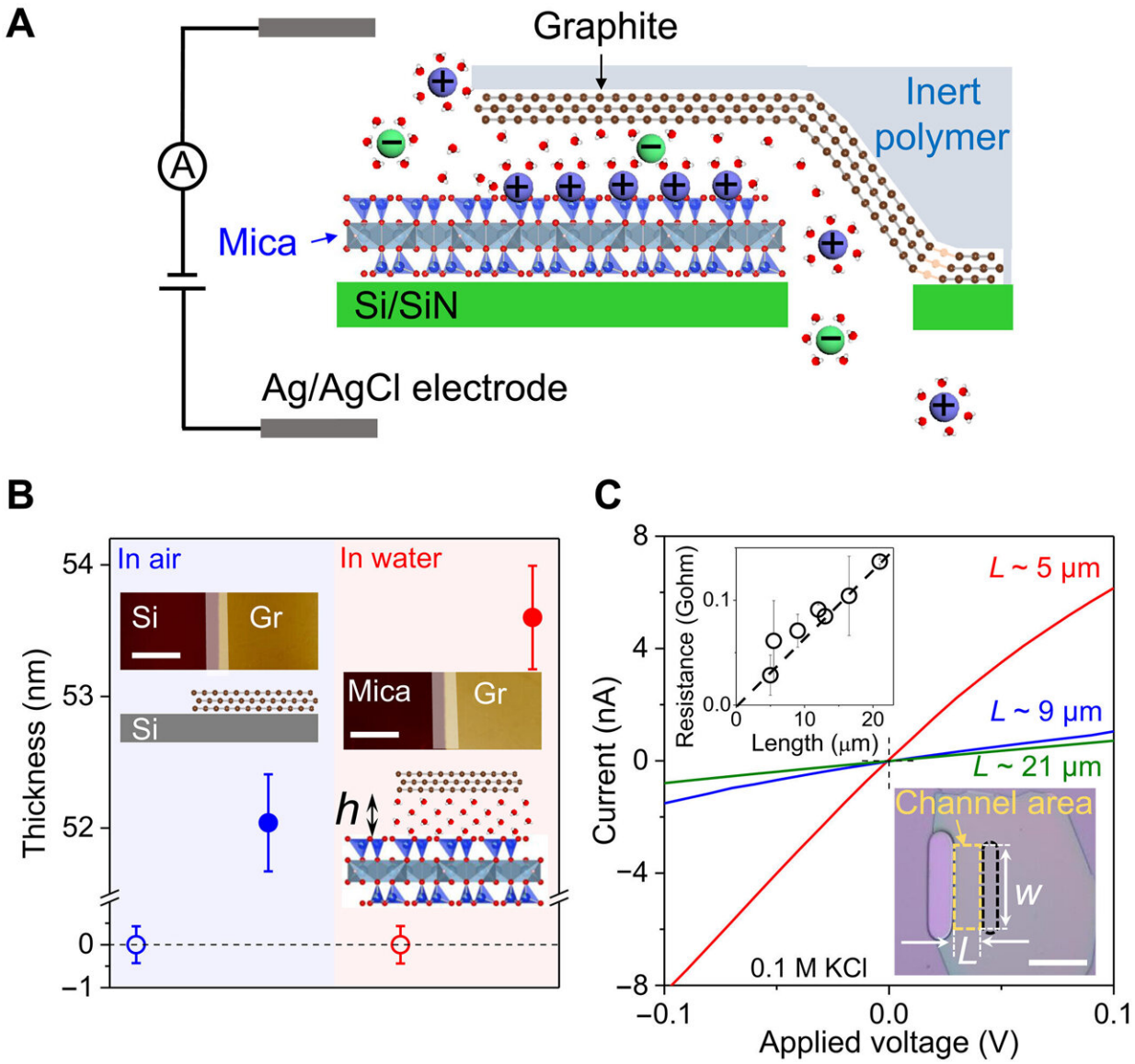


Nanofluidic channels made by graphite and mica heterostructures. (A) Schematic of our G-Mica channels and measurement setup. (B) AFM measurements of the top graphite (Gr) thickness when placed on the silicon (Si) substrate in air (before assembly) and on the mica substrate in aqueous solutions, respectively. The mean height of silicon (blue open symbol), mica (red open symbol), and graphite (filled symbols) is obtained statistically using all the data points in the AFM images (left and right insets, respectively), except for those in the step regions indicated by the white shadows. For comparison, the height of silicon and mica surface is deliberately set as zero. Left schematic shows pristine graphite on silicon. Right schematic shows water intercalation between graphite and mica in aqueous solutions, leading to an interlayer channel with height h. Scale bar, 0.5 μm. Error bars represent SD. (C) I-V characteristics of G-Mica channels with different length L. Top inset: Ionic resistance R for different L. Error bars represent SD. Bottom inset: Optical image of a representative G-Mica channel device. The black dashed area represents the aperture on silicon substrate, which is covered by top graphite. The yellow dashed box corresponds to the channel area, and the pink area is the opening on the inert polymer layer. w = 25 μm is the width for all channels. Scale bar, 20 μm. Credit: *Science Advances* (2023). DOI: 10.1126/sciadv.adi8493

Materials scientists have extensively studied fast ion permeation in nanofluidic channels in the past decades due to their potential within filtration technologies and osmotic energy harvesting. While the mechanisms underlying ion transport have yet to be understood, the process can be achieved in nanochannels developed in a carefully regulated manner.

In a new report now published in *Science Advances*, Yu Jiang and a research team in physical chemistry of solid surfaces in China described the development of two-dimensional nanochannels with their top and bottom walls containing atomically flat graphite and mica crystals.

The distinct wall structures and properties allowed the investigation of

interactions between ions and interior surfaces. The team noted enhanced ion transport within the channels that are orders of magnitude faster than in bulk solutions, providing insights into surface effects on ion transport at the nanoscale.

Nanoscale ion transport

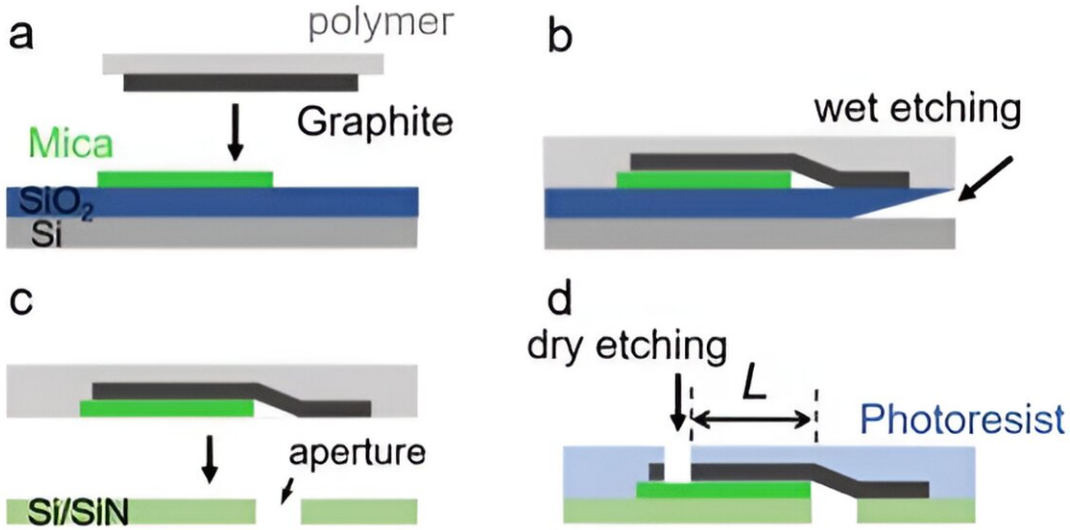
Mechanisms of nanoscale ion transport can outperform their macroscale counterparts due to their transport rates. Examples include fast ion flow through protein channels in cell membranes in a process that is critical for the essential functioning of life. These include ion permeation through nanoporous membranes for water purification, ion separation and osmotic power generation. To understand the mechanisms of fast ion transport at the nanoscale, researchers must create nanochannels with well-regulated geometry and interior structures.

Yu Jiang and team investigated the origin of fast ionic transport within nanochannels containing ion adsorption sites in the interiors. The simplified design minimized the chance of contaminating channel interiors with chemicals and polymers during fabrication to study adsorption effects on pristine surfaces.

During the experiments, Jiang and colleagues assembled mechanically exfoliated graphite and mica crystals and transferred them to an aperture on silicon substrates. They aligned the graphite/mica heterostructures with the aperture for the top graphite layer cover, while the bottom layer aligned with the aperture at their edges as determined by the transfer method.

The scientists used an atomic force microscope to measure the thickness of the top graphite on mica in aqueous solutions. They then measured the mean height of mica and graphite surfaces in the channel region. Since graphite and mica layers can delaminate at high salt concentrations of 2

M with relatively large ionic currents through the channels, they used solutions with salt concentrations equal to or smaller than 0.1 M for experimental accuracy.



Device fabrication and characterization. Fabrication flow of graphite-mica channels. (a) A graphite flake is transferred onto mica via dry transfer technique. (b) and (c) The graphite-mica stack is transferred onto the aperture using wet transfer technique. (d) Channel length is defined by dry etching methods. Credit: *Science Advances* (2023). DOI: 10.1126/sciadv.adi8493

Additional experiments

The scientists estimated the effective height of the channels seen by ions and confirmed the height characterized by atomic force microscopy. During the experiments, they filled the two reservoirs with various chloride solutions of 0.1 M and 0.01 M concentrations, respectively, to create a concentration gradient.

Jiang and colleagues studied the surface effects of the channel's interior upon ion transport and measured the ionic conductivity of potassium chloride as a function of its bulk concentration. The team investigated the ion transport process in the G-mica channels and narrowed the number of possible mechanisms by performing additional measurements.

Outlook

The high conductance and selective ion adsorption on mica surfaces indicated considerable surface diffusion. The scientists introduced a quantitative expression for ion transport in the graphite-mica channels to provide insights to related mechanisms.

They described the surface conductivity to be due to the migration of adsorbed cations while considering the effective surface salt number density, the surface mobility of adsorbed cations, and focused on the transport of monovalent cations. The relatively large adsorption energy of cations limited their desorption, before migration to highlight the importance of mica for ion transport.

In this way, Yu Jiang and colleagues highlighted surface diffusion as an additional ion transport path in nanofluidics to provide ionic conductivity that are orders of magnitude higher than in bulk solutions. The value is among the highest reported from single nanochannels. The capacity to create channels using mica group crystals that have preferences of adsorbing diverse cations can distinguish ions that depend on their adsorption energies for ion transport and sensing applications.

More information: Yu Jiang et al, Surface diffusion enhanced ion transport through two-dimensional nanochannels, *Science Advances* (2023). DOI: 10.1126/sciadv.adi8493