Selective ion transport and crystal growth in 2-dimensions inside graphene oxide membranes

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Highly anisotropic membranes can be obtained by stacking together nanosheets of 2-dimensional materials like graphene.
In such membranes, diffusion of molecules is strongly favoured along the main plane (the xy plane), and strongly hindered along the z axis perpendicular to it. This yields new exotic properties, such as unimpeded permeation of water[1] or selective sieving of small molecules and gases,[2] allowing to obtain tunable permeability *and* selectivity, beyond the traditional limits of conventional membranes.[3]
Thanks to their excellent performance, high chemical versatility and easy production, these membranes are promising for water purification, water desalinization, energy storage and biochemical sensing.

In this work, we describe the forced transport, on millimeters scale, of ions moving along the 2-dimensional inner channels of a graphene oxide membrane (GOM). Ion transport is achieved by applying a low negative voltage (0.5 V) between two water reservoirs located at the opposite edges of a graphene oxide membrane (GOM). Thanks to applied voltage, ions are driven along the GOM, travelling on macroscopic distances (1-5 mm), then being measured by an electrochemical pulsed technique.
In this way, we could easily compare the transport of most common cations (Li^+, Na^+, K^+, Mg^{2+}, Ca^{2+}) along the membrane, showing that ionic transport is inversely proportional to the charge density of the ions.
We also studied the presence and chemical state of the ions directly inside the membrane; noteworthy, the electrically-driven salts and hydroxides different from the ones present in the source solution form in the GOM, due to preferential intercalation of ions. As example, sodium carbonate can form in the membrane by intercalation of ions coming from sodium chloride solutions, or lithium carbonate from lithium chloride.
By using a positive voltage we could also observe preferential transport of anions (F^-, Cl^-) in the GOM, overcoming electrostatic repulsions between the functional groups of GO and the anions.
The tunable transport and assembly of crystals of anions and cations in GOM is a useful tool for both fundamental studies, to study crystallography in constrained spaces, and for concrete applications related to water treatment and purification.

References

[1] A. Geim et al., Science, (2012) **335**, 442.
[2] R. Nair et al., Nature Materials, (2017) **16**, 1198; Carbon, (2017) **120**, 145.
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Figures

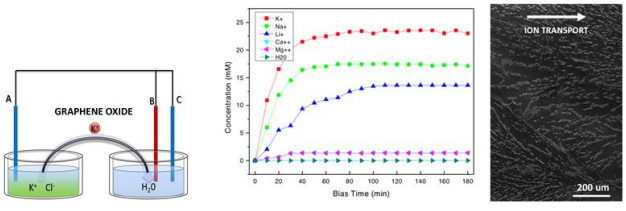

Figure 1: *Left: scheme of the electrochemical setup used. Center: different ion transport observed along the GO membrane for different cations. Right: SEM image of crystals grown into the GOM aligned along the ion flux.*