

ELECTROCHEMISTRY

Solid-state hydrogen storage goes electric

Electrochemistry enables reversible storage and release of hydrogen gas in a metal hydride

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Broad adoption of hydrogen as a versatile energy carrier is primarily hampered by a lack of safe and compact hydrogen storage. Hydrogen is often stored as compressed gas or cryogenic liquid, which requires high pressures or extremely low temperatures. Metal hydrides—compounds in which hydrogen is chemically bonded to solid metallic or intermetallic hosts—can store hydrogen with a high volumetric density (1). The inherent thermodynamic stability of these solid-state storage compounds provides safety but also requires impractically high temperatures for hydrogen release. On page 1252 of this issue, Hirose *et al.* (2) report an approach that electrochemically “pumps” hydrogen in the form of hydride ions (H^-) through a solid electrolyte into or out of a metal hydride. This elegantly circumvents the requirement of high temperature to free hydrogen from the metal hydride, offering a pathway to harness high-capacity hydrogen storage under practical release conditions.

Using electricity rather than temperature or pressure to drive hydrogen exchange between electrodes was first practically demonstrated in 1985 to investigate metal hydride thermodynamics (3). The Nernst equation—a central formula for calculating electrochemical cell potential under a given condition—also illustrates how a small voltage equates to many orders of magnitude change in hydrogen gas pressure, generating a large driving force for hydrogen exchange. For example, a small cell potential of 0.3 V is equivalent to a pressure of 1.4×10^{10} atm. However, using this thermodynamic driving force requires an ion-conducting electrolyte that is electronically insulating. Decades of work on hydride-ion conductors have underscored the difficulty in achieving this goal (4).

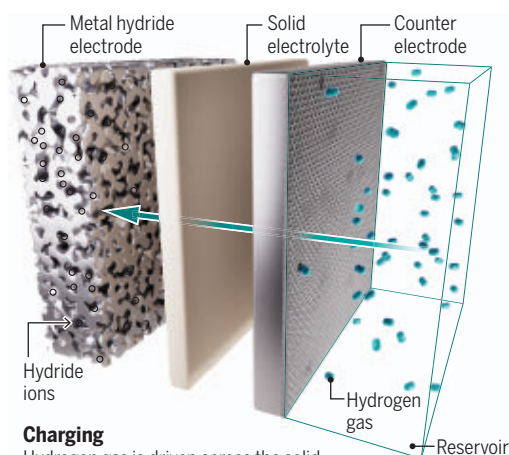
Hirose *et al.* developed a solid electrolyte by compositional tuning of a pseudo-ternary system in which barium, calcium, and sodium atoms form a compound with hydrogen atoms. This electrolyte exhibited good hydride ion conductivity ($\sim 2 \times 10^{-5}$ S/cm) at room temperature, low electronic conductivity (1.3×10^{-9} S/cm), and stability over a suitably wide voltage range. The hydride-

conducting electrolyte was sandwiched between a hydrogen-storage electrode and a counter electrode that delivered hydrogen gas to a reservoir (see the figure). This solid-state electrolyte could reversibly insert and extract hydride ions into and out of a diverse array of metal hydrides (such as TiH_2 , MgH_2 , NaAlH_4 , LiAlH_4 , NaBH_4 , and NaH) at moderate temperatures (60° to 100°C). Notably, magnesium hydride and sodium aluminum hydride electrodes achieved reversible hydrogen adsorption capacities that are close to their theoretical values, maximizing the full storage potential offered by these materials. At 90°C , the electrochemical cell with a magnesium hydride electrode achieved reversible hydrogen storage over 10 cycles (charge and discharge) at >99% of theoretical capacity (~2020 mA·hour/g), although at rather low charge and discharge rates. This achievement demonstrates the feasibility of the electrochemical approach to metal hydride hydrogen storage.

The study of Hirose *et al.* opens new directions to overcome the impractically high temperature requirements imposed by high-capacity metal hydride-based hydrogen-storage systems. Magnesium hydride has a theoretical gravimetric hydrogen-storage capacity (the amount of hydrogen that can be held per total mass) of 7.6 wt %, and much of this capacity was successfully accessed. However, the overall electrochemical cell configuration achieved a capacity of only 0.11 wt %. This is due, in part, to the thick, heavy solid-electrolyte layer (~ 450 μm) and a low active material ratio (20 wt % metal hydride) in the electrode. Furthermore, Hirose *et al.* acknowledged that voids in the cold-pressed solid electrolyte permit hydrogen-gas crossover. This leakage leads to a mixed storage mechanism in which the desired electrochemical hydride-ion insertion occurs simultaneously with an uncontrolled thermochemical reaction between the permeated hydrogen gas and the magnesium hydride electrode. The energy efficiency of the electrochemical system is also presently lower than that of thermal methods. The device of Hirose *et al.* required an energy input of ~ 81 kJ/mol, which is higher than that for optimized thermal desorption of hydro-

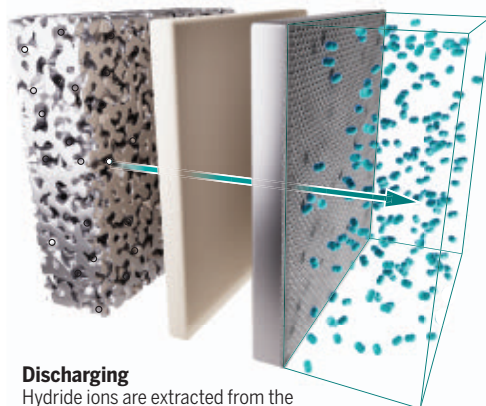
Electrochemical delivery of hydrogen

Metal hydrides can store hydrogen safely with a large volumetric density, but their inherent stability requires impractically high temperatures to release the stored hydrogen for use. A solid-state electrolyte can reversibly insert or extract hydrogen from the hydride at moderate temperatures (60° to 100°C) under an electric potential.



Charging

Hydrogen gas is driven across the solid electrolyte as hydride ions, which are inserted into the metal hydride electrode for storage.



Discharging

Hydride ions are extracted from the metal hydride electrode and then combine to form hydrogen gas at the counter electrode.

gen (~72 kJ/mol). The authors note that further engineering of the constituent materials and the electrochemical device (such as by decreasing the electrolyte thickness) could substantially reduce the voltage needed for charging, potentially decreasing the energy requirement down to ~45 kJ/mol, which is considerably lower than that for thermal-based metal hydride storage.

Practical deployment of hydrogen storage systems will require durability for >1000 cycles. Electrodes change their volumes as hydrogen atoms move in and out. At present, maintaining the chemical and mechanical stabilities of electrodes and their interfaces with electrolytes under the practical number of cycles remains unproven and is a crucial area for future research. In addition, thermal management, which is also problematic for conventional temperature- and pressure-based metal hydride storage, is an essential aspect that needs to be considered for a large-scale electrochemical system (5). Hydrogen absorption is strongly exothermic (heat releasing), whereas desorption is endothermic (heat absorbing). For fast fueling of hydrogen gas, heat must be ejected at tens of kilowatts per kilogram of hydrogen gas during charge and supplied at a similar rate during discharge. Complex device-level heat management strategies such as fin and foam structures integrated into the metal hydride electrode, embedded heat-exchange tubes, and heat-pipe and phase-change augmentation have been reported (6). However, these additions increase the weight of the device and reduce gravimetric and volumetric hydrogen-storage densities.

Beyond the electrochemical approach of Hirose *et al.*, other promising directions to improve metal hydride-based hydrogen storage are under active exploration. These include nanostructuring the metal hydride particles to enhance the kinetics of hydrogen adsorption and desorption (7) and engineering the composition of the metal hydride to address the trade-off between capacity and stability (8). Although many hurdles need to be overcome, ongoing studies offer a credible and exciting proposition: By taming hydrogen in a solid state, a critical component to a future sustainable energy source could be unlocked. □

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GENOMICS

Genomic clues into the spread of deadly mosquitoes

Ancient and more recent human interactions shape mosquito vector evolution

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Vector-borne diseases threaten 80% of the world's population (1). At the top of the list is malaria, of which there are roughly 260 million cases each year (2), and dengue, which is present in 132 countries (3). As the world becomes more connected, populations of mosquitoes that were once geographically confined are now being transported long distances and adapting to new environments. Genomic data from present populations can be used to reconstruct the evolutionary and demographic history of lineages of mosquito vectors. These reconstructions can facilitate prediction of the movement and adaptation of modern mosquito populations and the development of interventions to prevent the spread of disease. On pages 1209 and 1208 of this issue, Boddé *et al.* (4) and Crawford *et al.* (5), respectively, report findings on the evolution of two mosquito vectors. Their results highlight the population health consequences of ancient and contemporary interactions between humans and deadly mosquitoes.

The *Anopheles funestus* species of mosquito is a major malaria vector across sub-Saharan Africa. Most studies on the evolution of malaria-transmitting *Anopheles* species have centered on the *Anopheles gambiae* species group, and important gaps in knowledge remain for *An. funestus*, a dominant vector in parts of east and southern Africa. Previous investigations have sought out the molecular basis of adaptations to environmental factors and vector control measures such as insecticides using targeted sequencing of limited regions of the genome, karyotyping (which detects chromosomal abnormalities), or whole-genome sequencing of limited geographic scope. High frequencies of insecticide-resistance mutations have been reported in this species (6), and structural variants such as chromosomal inversions have been linked to variable ecological conditions and adaptation (7), as observed in other malaria vectors.

To examine the molecular basis for adaptation in *An. funestus* at high genomic, spatial, and temporal resolutions, Boddé *et al.* analyzed mosquito genomes from 13 African countries to determine the level of connectivity across *An. funestus* populations and identify genomic loci affected by selective pressure. They found that the genetic diversity of many populations was structured by geography, whereas others appeared to be genetically connected across wide geographic distances. Notably, populations in the North Ghana and South Benin cohorts were more genetically distinct from neighboring populations than expected. Boddé *et al.* argue that these findings suggest the existence of distinct ecotypes (i.e., highly divergent lineages) within *An. funestus*. Such complex patterns of diversity mean that a universal strategy for vector control of this species would likely be ineffective and that such efforts should include locally tailored surveillance and intervention.

Boddé *et al.* also sequenced samples from museum specimens of *An. funestus* collected in 1927, before heavy insecticide use. Using these older specimens as a baseline, the authors found that insecticide resistance had emerged through the independent evolution of resistance mutations in some populations and importation in others.