

Beyond Ionic Conductivity

Coordination-Transport-Interface Design Rules
for Durable Sodium Batteries

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The durability question

**A battery must
keep working,
cycle after cycle.**

Three papers connect three scales

Electrolytes

J. Mater. Chem. A · Review

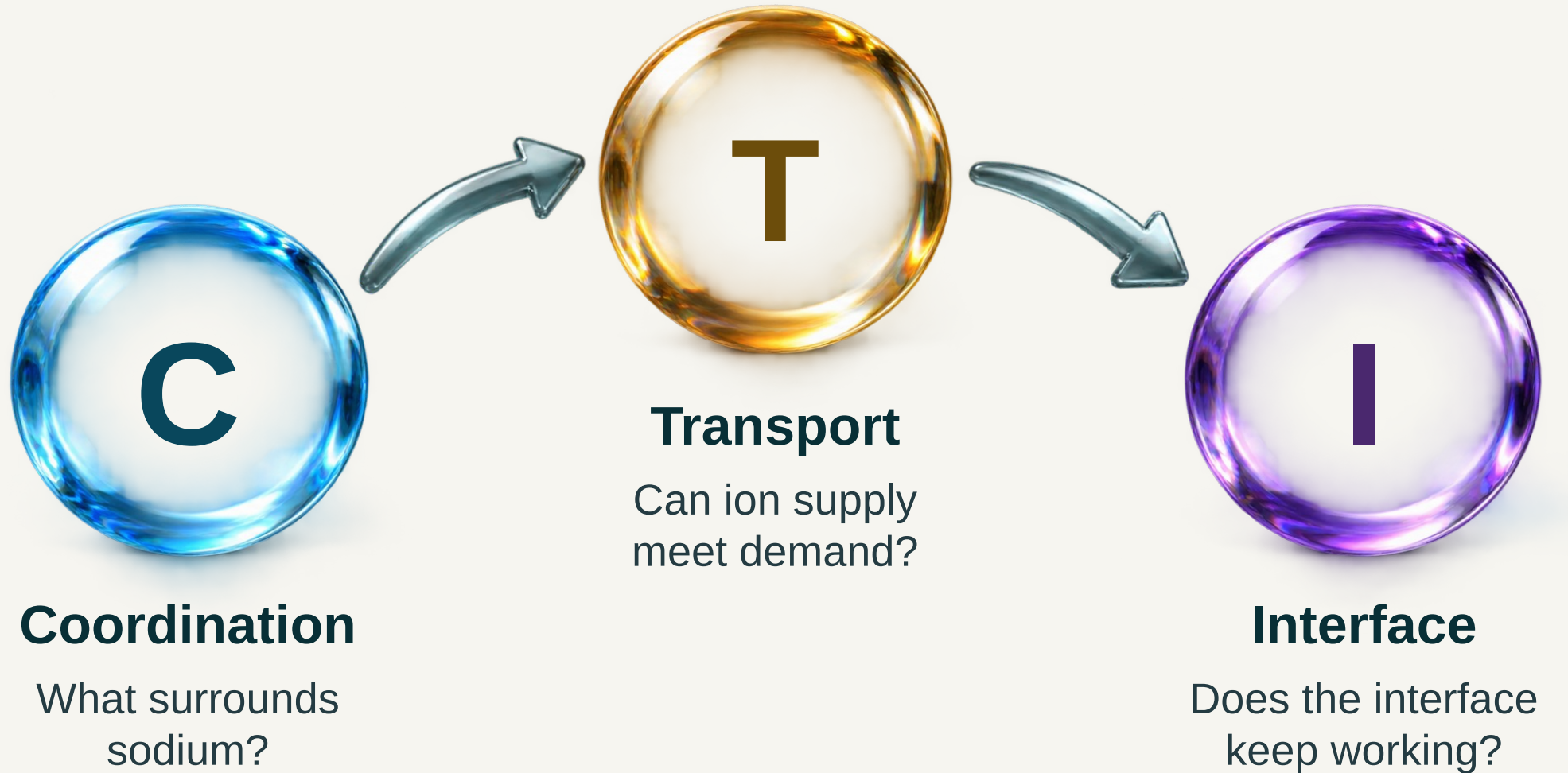
Carbon pores

Materials Horizons · Opinion

Full cells

Nanoscale Horizons · Focus

The CTI framework



The limiting process can change as the cell ages.

The local environment around sodium

SSIP

Solvent-separated pair



CIP

Contact ion pair



AGG

Associated ion cluster



Na⁺: teal Anion: gold Solvent: pale blue

Local neighbors can change the first reaction

Ion supply under load

Can ion supply meet the electrode's demand?

Association

Useful chemistry can come with higher viscosity.

Temperature

Mobility can change strongly in polymers.

Current density Areal capacity Temperature

Verify local structure in LHCEs. Follow polarization under the stated conditions.

Resistance through the life of a cell

Follow the same cell through time.

01

Formation

Establish the
initial state

02

Early cycling

Locate emerging
changes

03

Later cycling

Test whether
function persists

Separate resistive contributions. Match state of charge and temperature.

Different electrolytes, different limitations

Electrolyte family	Where to look first
Ionic liquids / HCE / LHCE	Association and usable ion supply
Polymers	Temperature, mobility, and contact
Oxides	Grain boundaries, wetting, and contact
Sulfides	Reactivity and mechanical behavior

Measurements establish what limits the actual cell.

Halides and hydroborates

Halides

Sodium-metal side

Transport and compatibility can limit operation.

Oxidative stability can support cathode-side use.

Hybrid designs can give different regions different jobs.

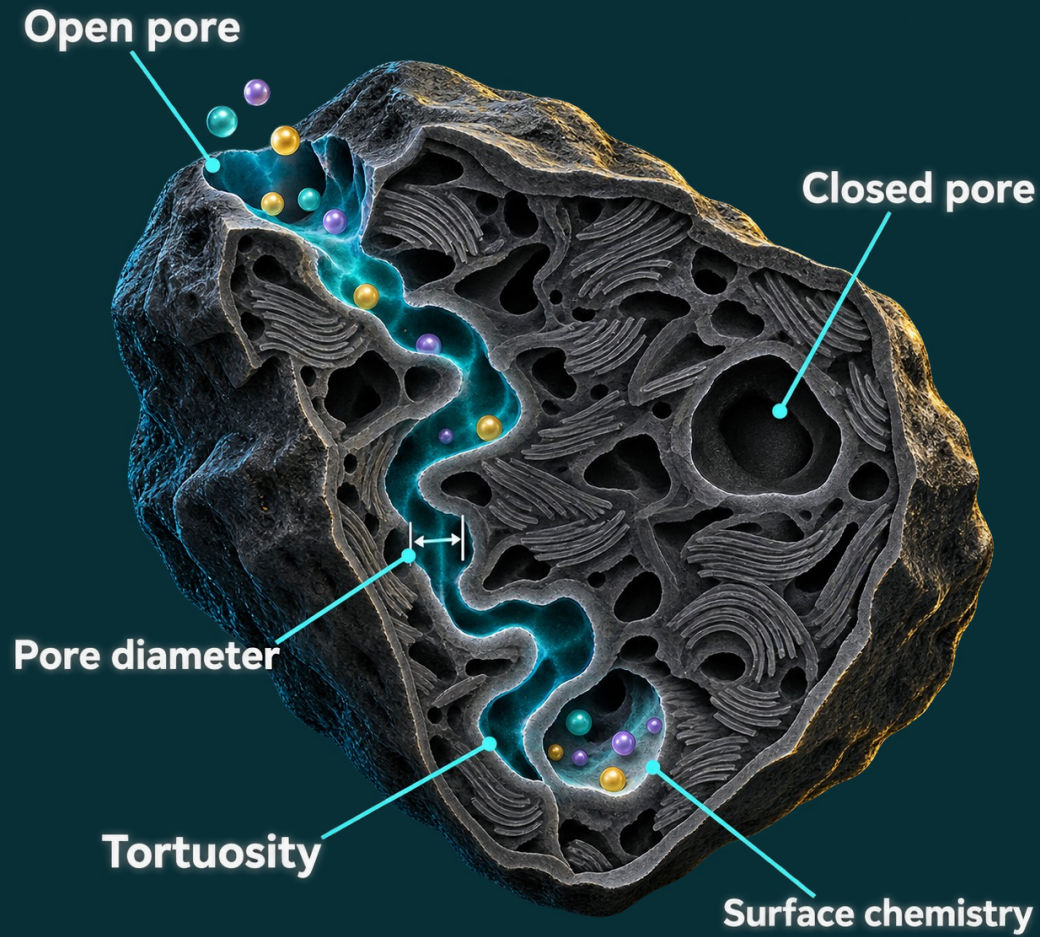
Hydroborates

Sodium-metal side

Favorable passivation in some systems.

Oxidative stability can limit cathode-side use.

Inside a hard-carbon pore



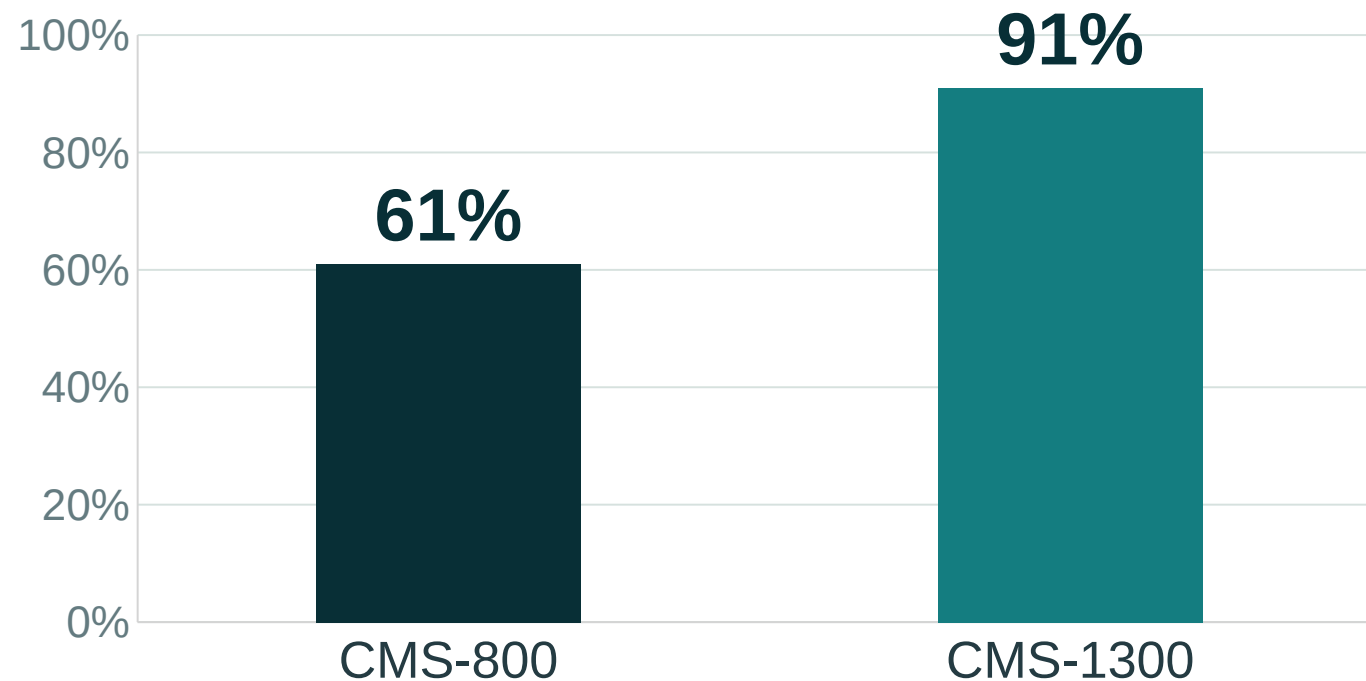
**The electrolyte
we mix is only
the beginning.**

Pore entrances and surfaces can
reshape the local environment.

Proposed conceptual framework

One electrolyte, two carbon structures

Initial Coulombic efficiency



Same electrolyte

1 M NaPF₆
EC:DEC (1:1)

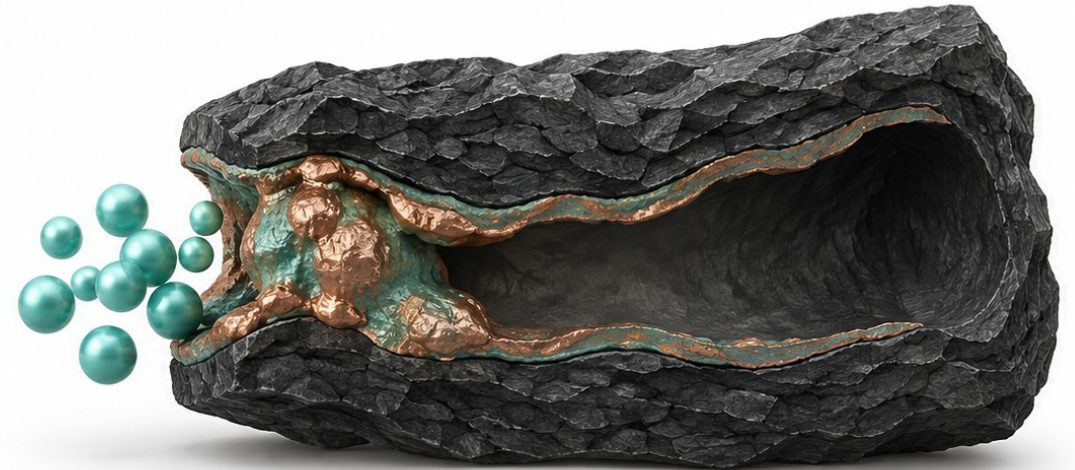
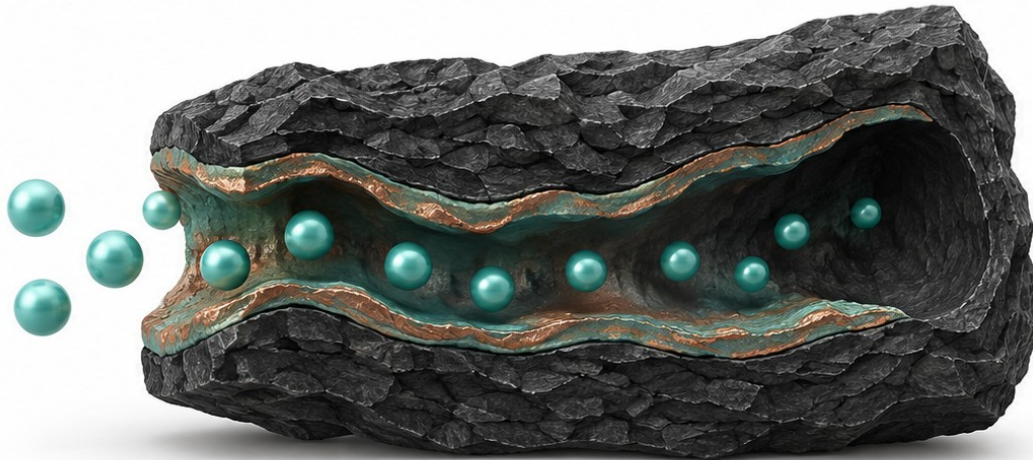
Na-metal half cells
25 °C, 50 mA g⁻¹
~1 mg cm⁻², 40 μL
0.005–2.5 V

30 percentage points in first-cycle efficiency

Interphase growth and pore access

Access preserved

Access restricted



The interphase must preserve pore access

Possible outcomes. Neither follows universally from SSIP, CIP, or AGG.

Conceptual illustration based on Santoso et al., Fig. 3. DOI: 10.1039/D6MH00519E

Matched tests of carbon and electrolyte

Same carbon

Same electrolyte

Different electrolyte

Controlled carbons

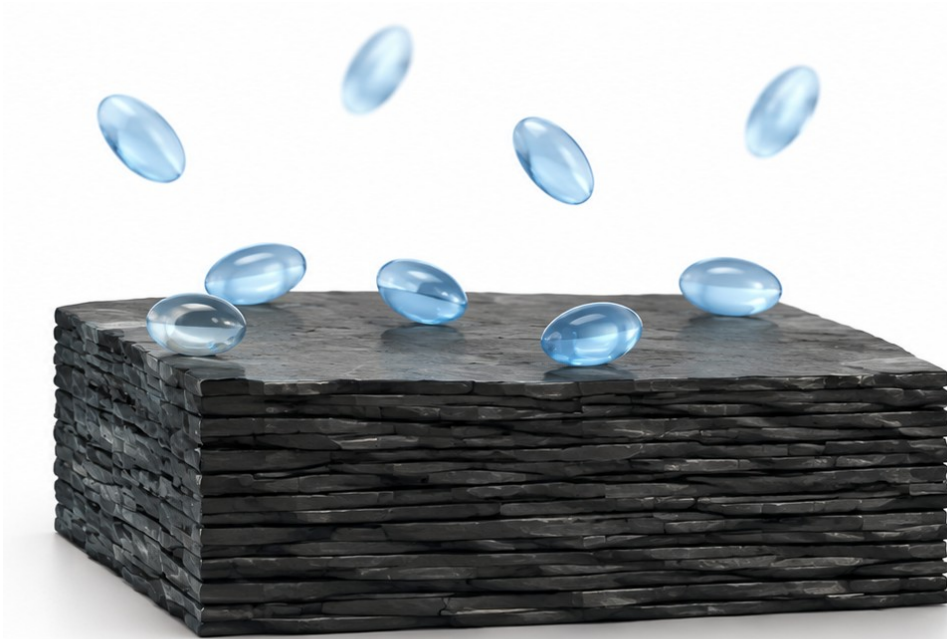
Connect local chemistry with retained pore access.

Complements insertion, adsorption, and pore filling.

An interphase in motion

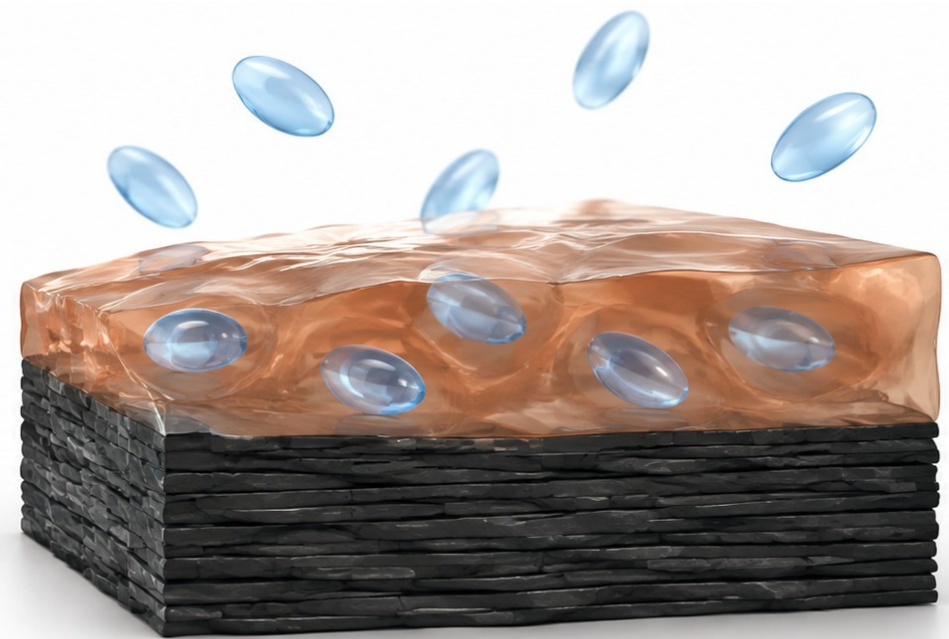
Adsorption

AT a surface



Absorption

INTO the interphase



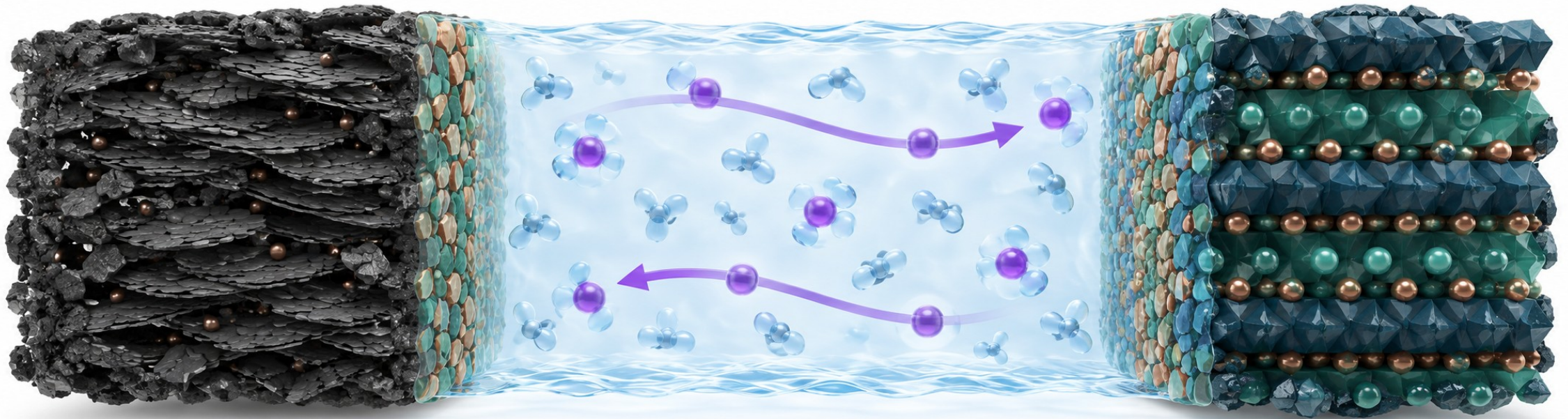
Solvent can enter and change the interphase

Two electrodes, one shared electrolyte

Hard-carbon anode

Shared electrolyte

Cathode



Each electrode can affect the other

Possible pathways require verification in sodium-ion cells.

Conceptual illustration based on Wisan et al., Fig. 1b–c. DOI: 10.1039/D6NH00245E

The sodium budget

A finite sodium budget

Initial losses

Formation consumes some sodium.

Later side reactions

Continued loss draws from the same budget.

Electrode balance and electrolyte amount define the test.

Formation and operating history

01

Formation

Control the
early history

02

Operating stress

Specify voltage,
current, and time

03

Diagnostics

Compare resistance
and losses

A test should expose the failure we aim to prevent.

Evidence for an interphase mechanism

- | | | |
|----------|--------------------|--|
| 1 | Performance | Does the cell perform better? |
| 2 | Composition | Which products are present, and where? |
| 3 | Evolution | How does the interface change with time? |
| 4 | Cause | Does a targeted change test the explanation? |

A proposed guide to matching claims with evidence.

Conditions that make results comparable

The operating cell	The measurement
Current density and areal capacity	Impedance state of charge and temperature
Temperature and sodium inventory	Formation and sample handling
Electrolyte amount	Matched diagnostic protocol
Solid cells: pressure and architecture	The same basis for comparison

Clear conditions make a comparison meaningful.

Design rules 1–3

01 Connect coordination to interface behavior

Check the chemistry and follow resistance during use.

02 Test transport at the required ion flux

State current density, areal capacity, and temperature.

03 Separate resistance contributions

Identify the process that limits the actual cell.

Design rules 4–6

04 Follow resistance over time

Measure the change through formation and cycling.

05 Match the intervention to the failure

Give each coating, additive, or interlayer a clear purpose.

06 Compare under shared conditions

Make inventory, loading, pressure, and history explicit.

When interface resistance grows

A worked example: contact loss or chemical change?

Test	Possible cause	Intervention to test
Pressure response and contact imaging	Contact loss	Compliant interface or better contact
Chemical change and resistance evolution	Continued decomposition	Chemical passivation

Both processes can occur. Test the predicted response.

Experiments that test the framework

Carbon pores

Does local chemistry preserve access?

Solid interfaces

Is the change physical or chemical?

Full cells

What happens under a limited inventory?

Proposed tests with controls for alternative explanations

Proposed experiments based on the three articles



Preserve a working interface, cycle after cycle.

Beyond Ionic Conductivity

Thank you

The people behind the science.



Our coauthors and collaborators

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