

Alkali metal-sulfur batteries

Pairing sodium-potassium anodes with solid electrolytes and semi-solid polysulfide cathodes to enable compact, safe and sustainable energy storage

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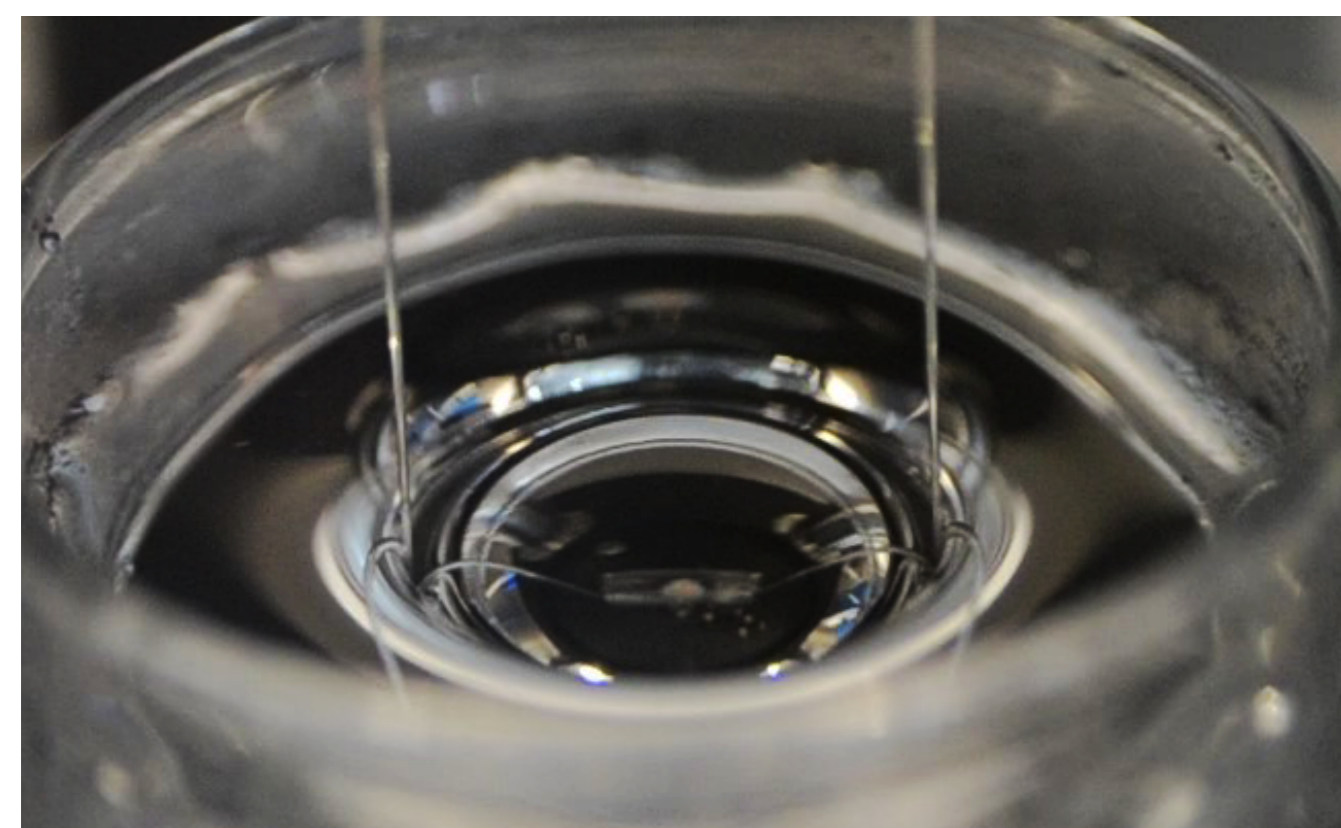


Aim: Compact, safe and sustainable energy storage

- pairing abundant sodium-potassium anodes with sulfur cathodes
- address key aspects in four interconnected work packages spanning from alkali-metal fundamentals, via interfacial engineering and solid electrolyte synthesis to sulfur cathode development
- focus here on results related to alkali-metal fundamentals and interfacial engineering

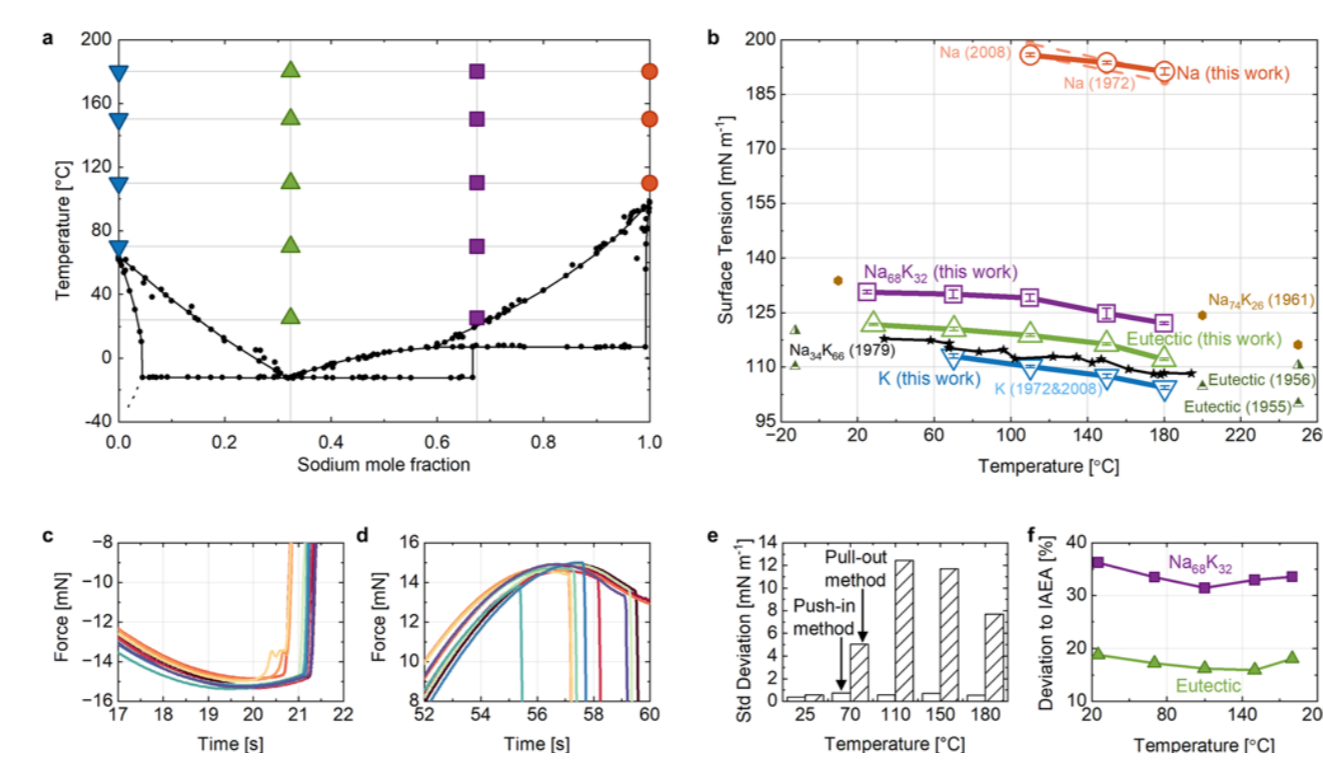
Fundamentals of alkali metals: Surface tension of sodium-potassium alloy

- precise knowledge of surface tension is a key requirement for interfacial engineering



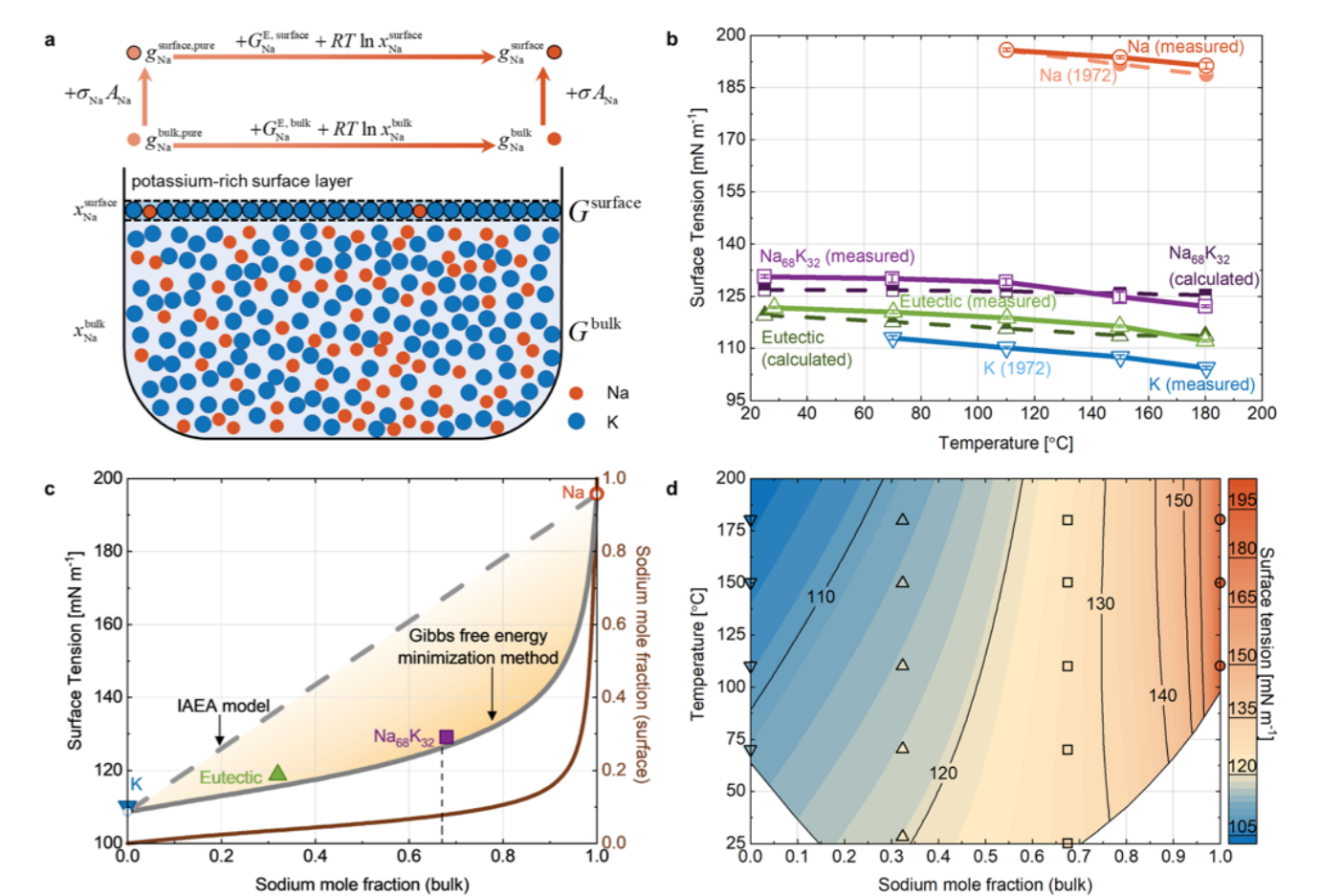
Du Noüy ring method. Measuring the surface tension of liquid alkali metals as a function of temperature and composition. Ranging from room-temperature up to 180 °C. Using a tensiometer with an attached Du Noüy ring. Compositions range from pure Na to pure K. Performed in Argon-filled glove box.

→ Powerful technique to accurately measure surface tension



Surface tension measurements. **a**, Na-K phase diagram introducing the experimental plan. **b**, Surface tension of pure K, pure Na, and Na-K alloy. **c,d**, Comparison of the minimum and maximum force in raw data. **e**, Average of standard deviations in push-in and pull-out methods. **f**, Deviation between measurements and simplified IAEA model. Evaluation.

- First systematic and precise measurements of NaK surface tension
- Force evaluation during moment of push-in enhances accuracy

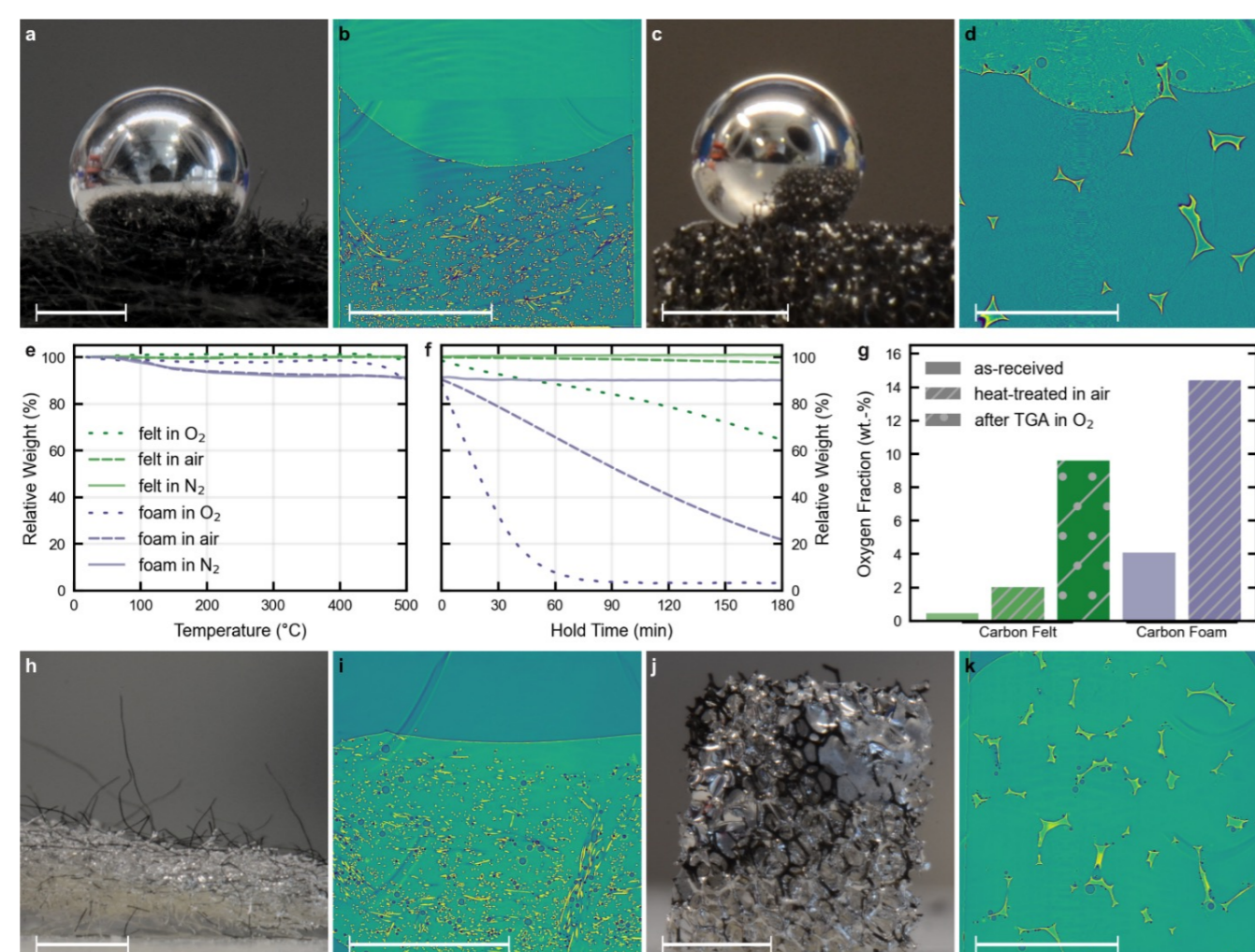


Modeling surface tension via Gibbs free energy minimization. **a**, Schematic. **b**, Model (dashed) vs. measurement (solid lines). **c**, Computed surface tension and Na mole fraction in surface phase versus Na mole fraction in bulk phase at 110 °C. **d**, Contour plot.

→ Gibbs free energy minimization modeling provides complete picture

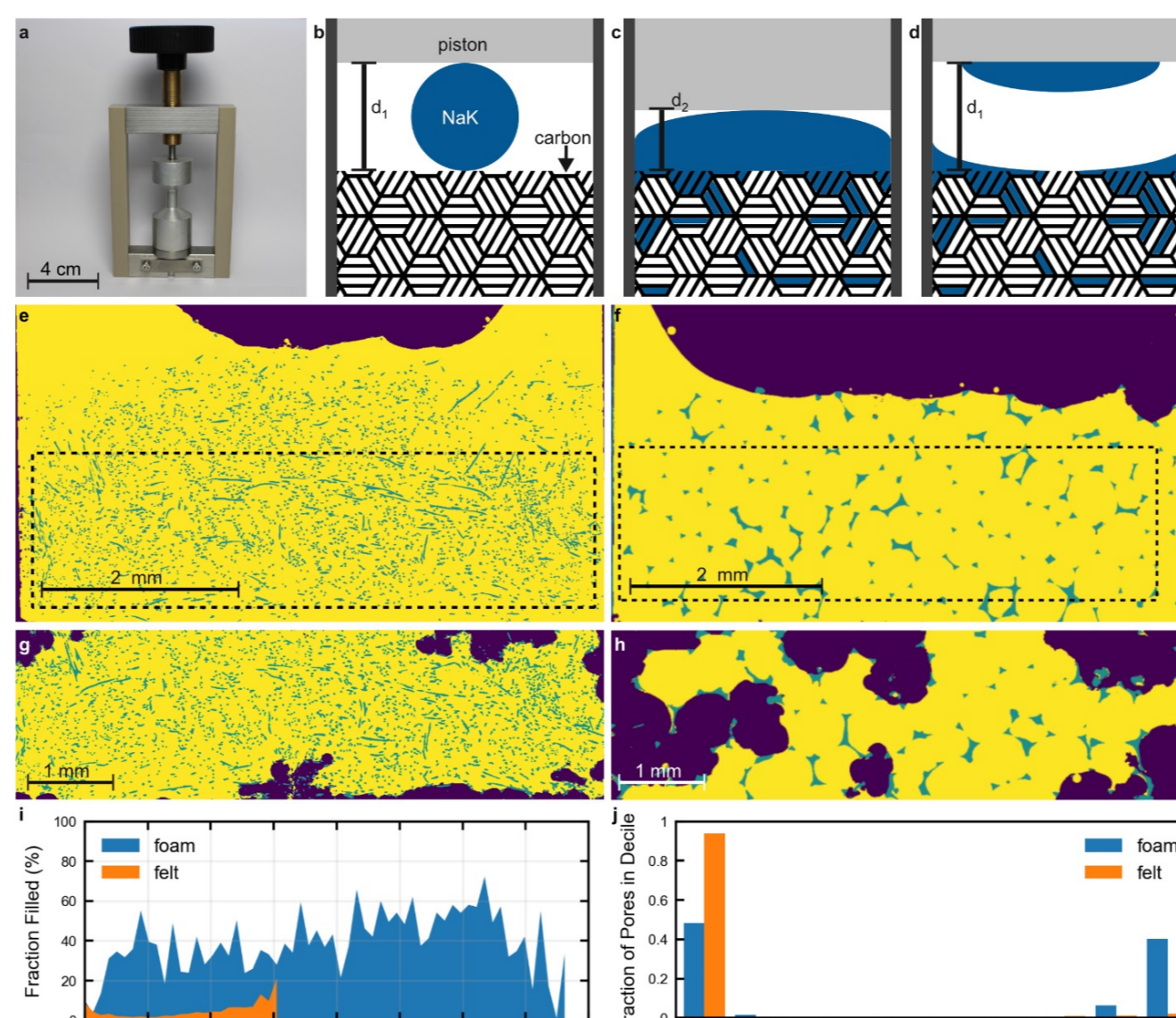
Interfacial engineering: Interactions between alkali-metal alloys and porous carbons

- alkali-metal management (storage, release, transport) is essential for high-performance anodes



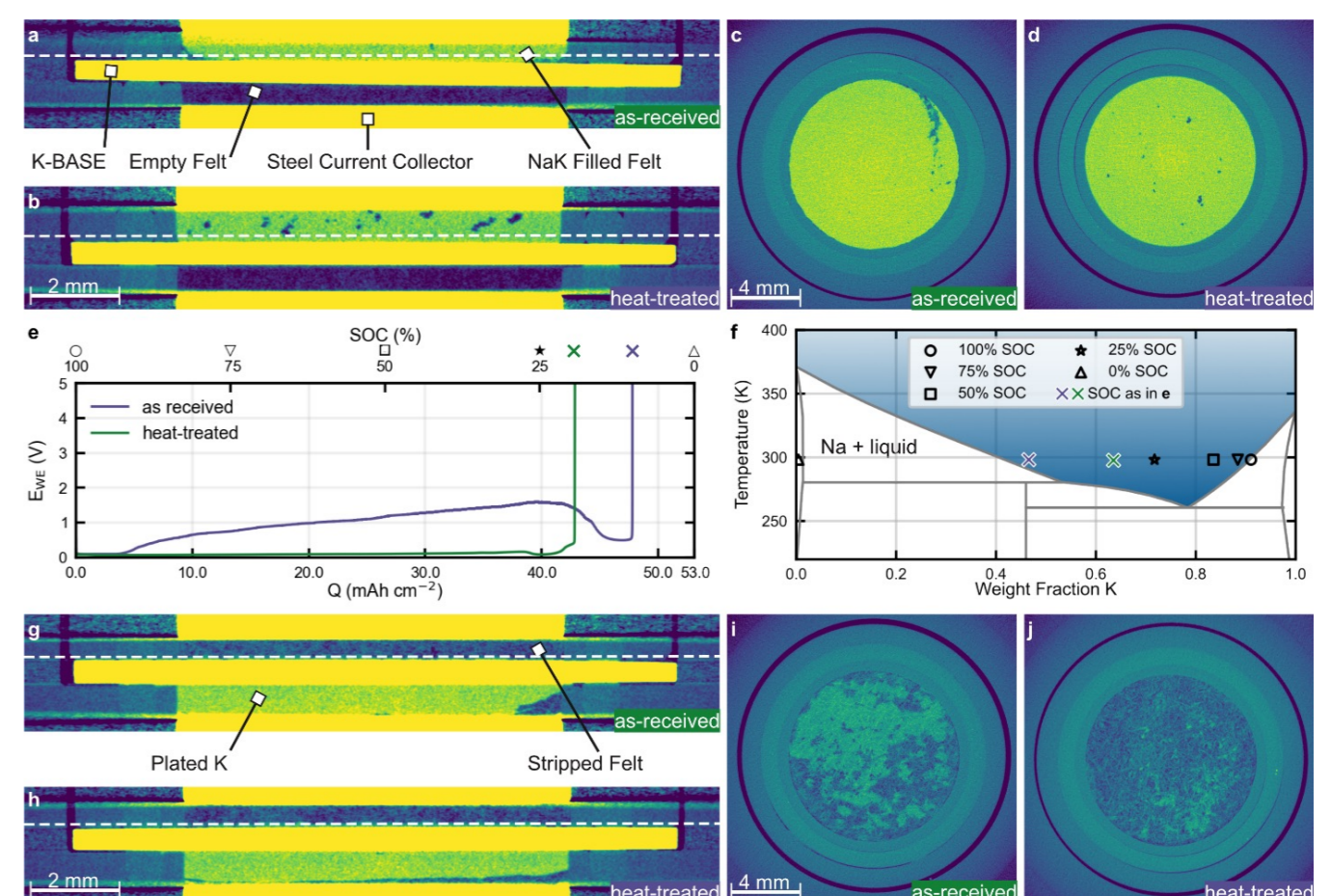
Wetting interactions between liquid alkali metals and porous carbon hosts as a function of texture and thermal pretreatment. **a-d**, Photographs and X-ray computed tomography (XCT) scans of NaK droplet resting on as-received carbon felt and foam in Argon atmosphere. **e**, Thermogravimetric analysis (TGA) of carbon felt and carbon foam with a temperature increase up to 500 °C and, **f**, a subsequent 3 h isothermal treatment. **g**, Elemental analysis obtained via energy-dispersive X-ray spectroscopy (EDX) showing that the measured atomic concentration of oxygen in the samples increases with the heat treatment. **h-i**, Photograph and XCT scan of heat-treated carbon felt and foam, which got spontaneously wetted by NaK. All scalebars are 2 mm.

→ Simple heat treatment renders porous carbons philic to NaK



Forced wetting interactions between as-received, alkali-metal repellent carbon hosts and liquid NaK droplets. **a**, Photograph of the tomography cell with an adjustable piston. **b-d**, Schematic drawings of the interactions between the NaK droplet with the porous hosts showing, **b**, the initial state, **c**, the state when the piston is lowered, and, **d**, after the piston is retracted. **e-f**, Slice through a tomography measurement of the as-received carbon felt and foam. Dashed box indicates the analyzed control volume. **g-h**, Control volume shown in **e** and **f**, after piston has been lowered to 0.6 mm and 1 mm, respectively, above the felt. **i**, Fraction of filled pores depending on the pore radius for felt and foam. **j**, Distribution of pore filling.

→ Forced wetting into phobic porous carbons scales with pore size



Comparing electrochemical performance of as-received and heat-treated carbon felt. **a-b**, Vertical slice through an XCT scan of an electrochemical cell with as-received carbon felt and heat-treated carbon felt, respectively, on both sides and an initial volume of NaK in upper compartment. The compartments are separated by a potassium-β"-alumina solid electrolyte (K-BASE). **c-d**, Horizontal slice through the same cells pictured in **a** and **b** on the height of the white dashed line. **e**, Constant current discharge at 0.5 mA cm⁻² for electrochemical cells with as-received and heat-treated carbon felt samples. **f**, Na-K phase diagram with theoretical state of charge (SOC) values and their respective composition marked at room temperature. **g-h**, Vertical slice through an XCT scan of an electrochemical cell with as-received carbon felt and heat-treated carbon felt, respectively, after most of the initial potassium has been electrochemically transferred to the lower compartment. **i-j**, Horizontal slice through the same cell pictured in **g** and **h** on the height of the white dashed line.

→ Heat-treated sample shows ten times lower overpotential

References

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