

Melbourne Energy Institute

## Artificial Leaf for Hydrogen Production

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- 1. Solar Water Splitting
- 2. Cocatalyst Foils
- 3. GaAs Artificial Leaf
- 4. Triple-Junction Device
- 5. Conclusion and Outlook
- 6. Acknowledgements



## **Solar Water Splitting**





#### **Benefits and challenges of renewables:**

- Fossil fuels, CO<sub>2</sub> emissions, climate change.
- Renewables, readily available, cost-competitive.
- Intermittency a challenge, storage methods needed.



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Nat. Clim. Chang. 2017, 7, 243; IEEE Trans. Smart Grid 2012, 3, 850.



#### Molecular hydrogen H<sub>2</sub>:

- Very energy-dense chemical fuel (2.7× gasoline).
- Grey hydrogen, 3% of global CO<sub>2</sub> emissions.
- Green hydrogen economy has been proposed.





#### Water electrolysis:

- OER at the anode.  $2H_2O \rightarrow O_2 + 4H^+ + 4e^-$
- HER at the cathode.  $2H^+ + 2e^- \rightarrow H_2$
- Cell potential 1.23 V.
- +100 mV for the HER.
- +300 mV for the OER.



Potential



#### **Challenges:**

- Efficiency, stability, cost.
- Concentrated illumination can reduce material costs and improve energetics.
- Thermal management critical.
- Immersed devices, electrolyte can double as coolant.
- Transport heat from photoabsorbers to electrodes.
- All components maintain suitable temperatures.



### **Cocatalyst Foils**





#### **Earth-abundant cocatalysts:**

- Transition metals and compounds.
- Often nanostructured and therefore opaque.
- Deposited via solution-based methods.
- Difficult to combine with semiconductors.



Chem. Soc. Rev. 2014, 43, 7787; Nat. Rev. Chem. 2017, 1, 0003; Adv. Mater. 2020, 32, 1806326.



#### **Fully decoupled photoelectrodes:**

- Light absorption and catalysis are spatially decoupled during device operation.
- Cocatalyst deposition is decoupled from device fabrication



Adv. Energy Mater. 2022, 12, 2102752.



#### **Earth-abundant cocatalyst foils:**

- NiMo<sub>x</sub>/Ni foil HEC, deposited hydrothermally.
- NiFe(OH)<sub>x</sub>/Ni foil OEC, electrodeposited.





#### **Earth-abundant cocatalyst foils:**

- NiMo<sub>x</sub> flakes, NiFe(OH)<sub>x</sub> needles.
- Benchmark noble metal cocatalysts.







#### J-V characteristics in 1 M NaOH:

- NiMo<sub>x</sub>/Ni foil nearly as good as Pt/Ni foil.
- NiFe(OH)<sub>x</sub>/Ni foil better than  $IrO_x$ /Au/Ni foil.
- Collectively, earth-abundant cocatalyst foils as good as noble metals.





#### Stability in 1 M NaOH:

- Very stable over 3 days.
- Faradaic efficiency very close to 100%.





### **GaAs Artificial Leaf**





#### GaAs cells:

- Two cells in series required for solar water splitting.
- Direct band gap, can have thin film solar cells, concentrated illumination.





### **Construction of artificial leaf:**

- Two GaAs cells in series.
- Fully decoupled photoanode.
- Wired photocathode.
- Ag bars, Ag paint, glass, epoxy.







#### J-V characteristics in 1 M NaOH:

$$\eta_{\text{STH}} = \frac{1.23 \text{ (V)} \times J \text{ (mA/cm2)} \times \eta_{\text{F}}}{P_{\text{in}} \text{ (mW/cm2)}}$$

- 11.04 mA/cm<sup>2</sup> at 0 V.
- STH efficiency 13.6%.





#### Stability in 1 M NaOH:

- T1, T2, electrolyte replenished, lamp intensity reset.
- Epoxy delaminated at T2, GaAs cells corroded at T3.
- STH efficiency of over 10% for longer than 9 days.





# Comparison with previously reported devices:

- Systems measured under 1 sun.
- Most efficient with >7 days stability.
- Most efficient with earthabundant cocatalysts.





### **Triple-Junction Device**





#### **Triple-junction cells:**

- More subcells, higher PV efficiency.
- Double-junction cells "best" for solar water splitting.
- Triple-junction cells, excess photovoltage.
- Can adjust the ratio of cells to electrolysers.



Nat. Commun. 2016, **7**, 13237; Appl. Phys. Express 2015, **8**, 107101.



#### **Triple-junction cells:**

- Supplied by MicroLink Devices.
- InGaP/InGaAs/Ge.





#### **Construction of triple-junction device:**

- Fully decoupled photoanodes with  $NiFe(OH)_x/Ni$  foil.
- NiMo<sub>x</sub>/Ni foam cathodes (not pictured).
- Three photoanodes, four electrochemical cells.





#### J-V characteristics in 1 M NaOH:

- Each photoanode provides over 2 V.
- Each electrochemical cell requires 1.55 V.
- Components in series add linearly.
- 3 photoanodes, 4 electrochemical cells, STH **20.7%**.





### Stability in 1 M NaOH:

- >20% STH maintained for 40 hours, but epoxy not suitable for long-term stability.
- Single photoanode with better epoxy maintains very stable photocurrent for nearly 9 days.





#### **Comparison with previously reported devices:**

- Compares very favourably with other systems, both immersed and PV-EC.
- STH exceeded only by systems operating under concentrated illumination.





## **Conclusion and Outlook**





#### **Fully decoupled photoelectrodes:**

- Constructed using cocatalyst foils.
- Efficient, stable, earth-abundant.
- Paves the way for immersed devices operating under concentrated illumination.



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