University Research Priority Program Light to Chemical Energy Conversion

URPP LightChEC

UNIVERSITY OF CAMBRIDGE

Solar-driven Photoelectrochemical Systems and 'Artificial Leaves' for Efficient Waste-to-Fuel/Chemical Production

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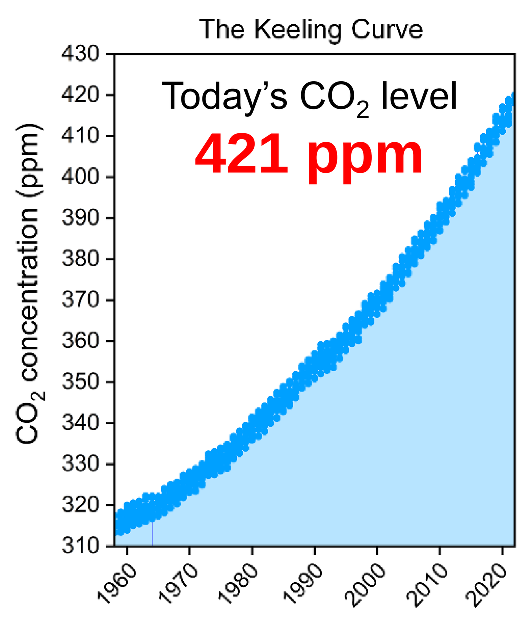
1. Broader Context and Introduction

Two Major Global Challenges[1]

1. Energy Crisis and Climate Change
Fossil fuels – Non-renewable – **Global Warming**
Constant rise in demand for energy
Need for cleaner sustainable sources of energy

2. Waste Generation and Pollution
Plastics, Biomass, etc. - Most end up in landfills
Existing recycling strategies **energy-intensive** and **polluting**
Sustainable utilisation of abundant waste resources



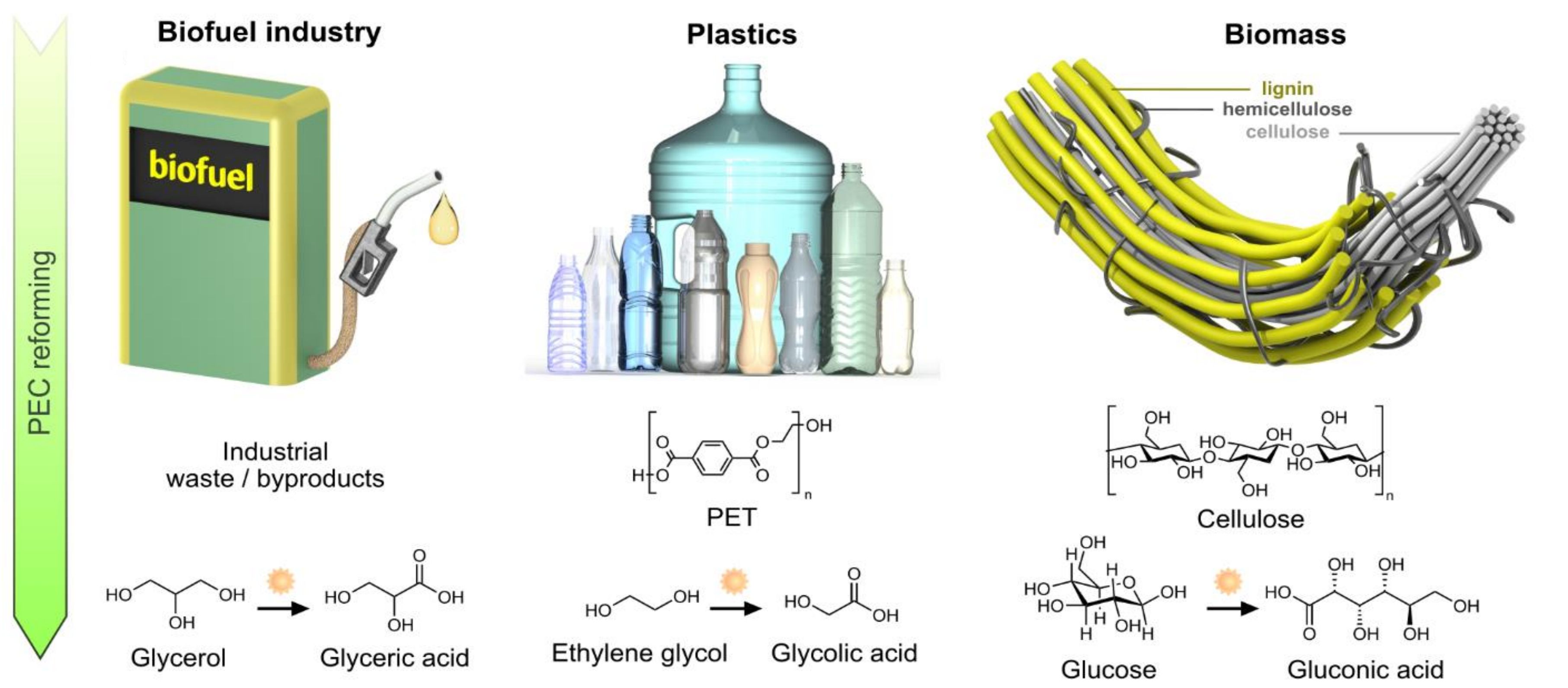
2. A Unifying Solution?

Strategy: Use of sunlight-driven technologies to utilise abundant waste resources as feedstocks for the generation of **sustainable fuels** (green H_2, syngas, etc.) and **value-added products**

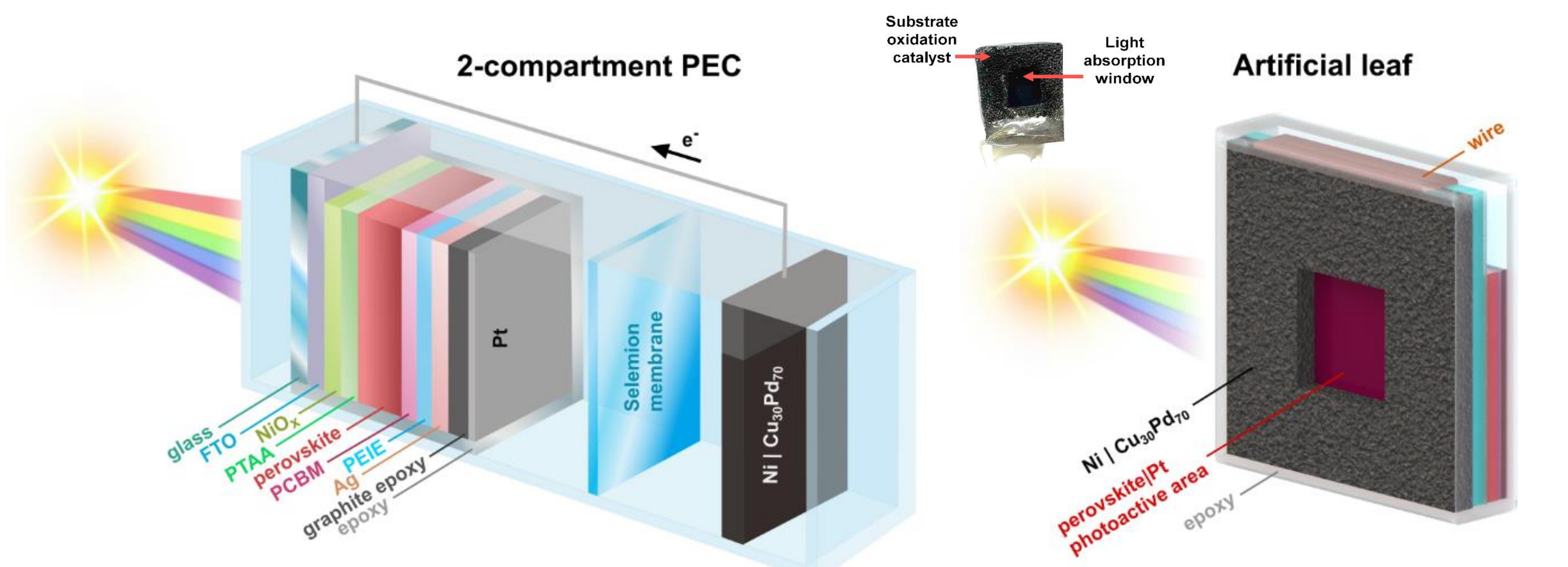
Limitations of existing photocatalytic (PC) reforming - **low product yields** (poor performance), **poor selectivites** (mixture of non-utilisable oxidation products), emission of undesirable CO_2 and **lack of versatility**[1,2]

Our technology: efficient single-light absorber 'photoelectroreforming' (PEC reforming) devices which can reform a **diverse** range of waste feedstocks (see below) to green **hydrogen** and value-added chemicals with **high product formation rates** and **high selectivity**[3]

PC reforming

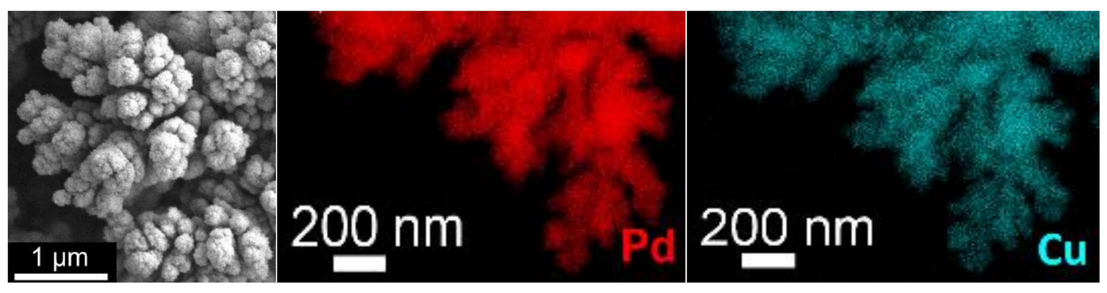


3. Design and Architecture of PEC systems

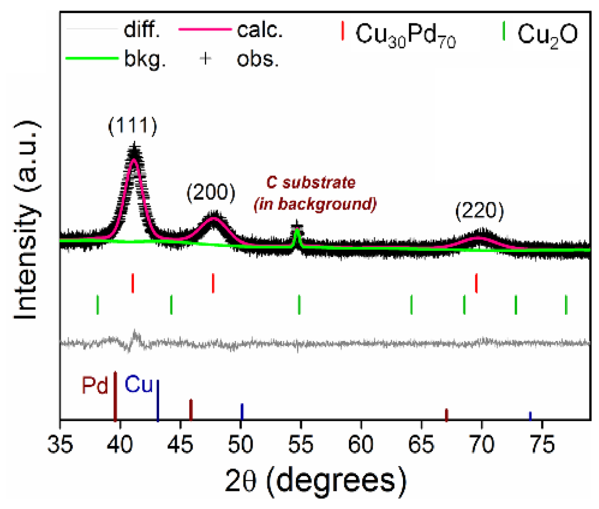


For designing the PEC devices, a **$Cu_{30}Pd_{70}$ oxidation catalyst** is integrated with **halide perovskite photocathodes**[4] employing Pt as H_2 evolution catalyst. These PEC devices are investigated in both **two-compartment** and **'artificial leaf'** configurations (see image above)[3]

Bimetallic $Cu_{30}Pd_{70}$ alloy micro-flowers (MFs), were developed and characterised as a selective waste oxidation catalyst for our PEC systems.



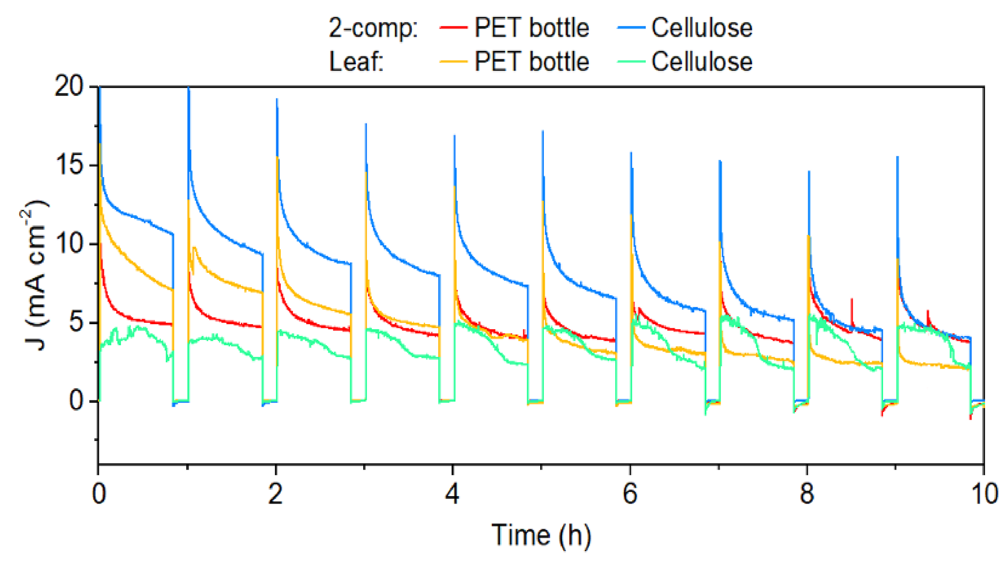
FESEM image and STEM-HAADF mapping of $Cu_{30}Pd_{70}$ alloy MFs

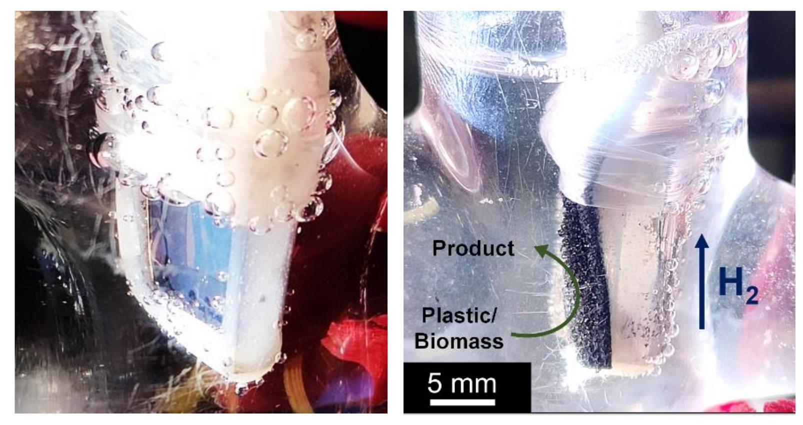


Rietveld refined PXRD pattern of the $Cu_{30}Pd_{70}$ alloy MFs

4. Bias-Free PEC Reforming of Polymeric Waste

The bias-free PEC experiments were carried out both in **two-compartment configuration (2-comp)** and an integrated **'artificial leaf'** configuration.[3]





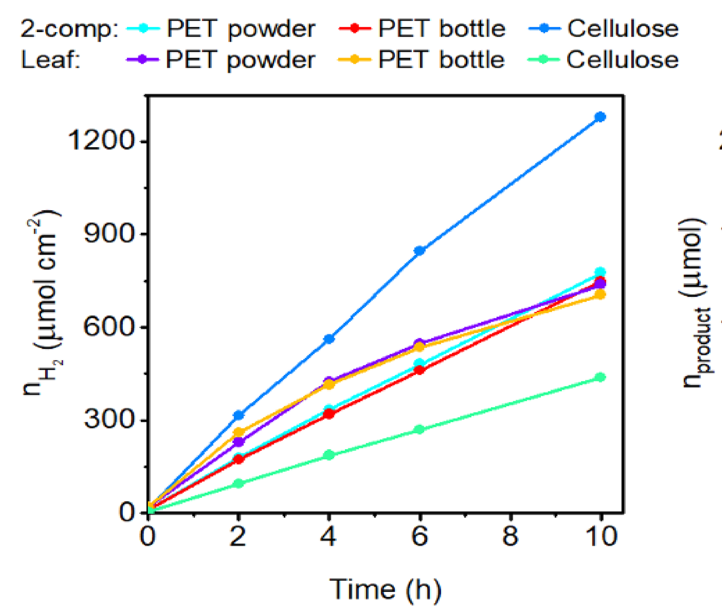
Perovskite|Pt photocathode

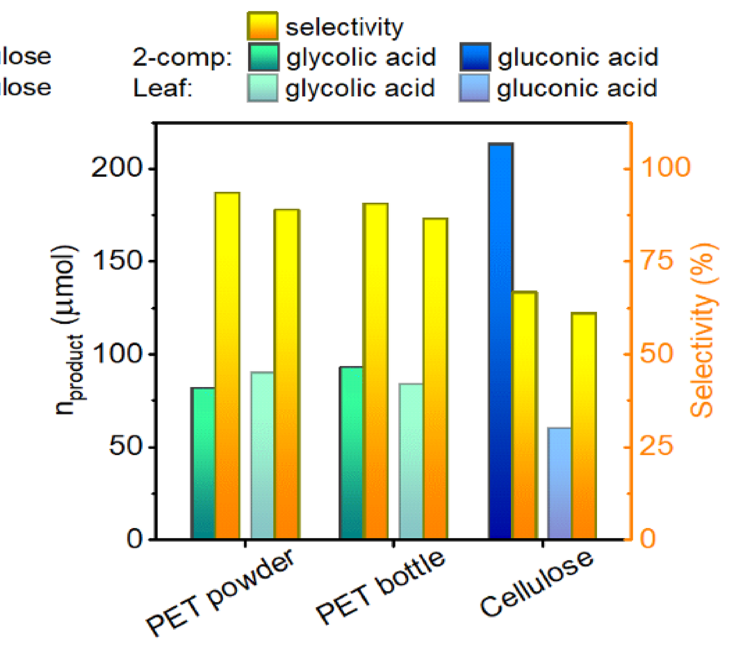
Integrated $Cu_{30}Pd_{70}$|perovskite|Pt 'artificial leaf'

H_2 production in 10 h (µmol H_2 cm^{-2})

2-compartment
PET powder: **776**
PET bottle: **748**
Cellulose: **1280**

Artificial Leaf
PET powder: **737**
PET bottle: **705**
Cellulose: **438**

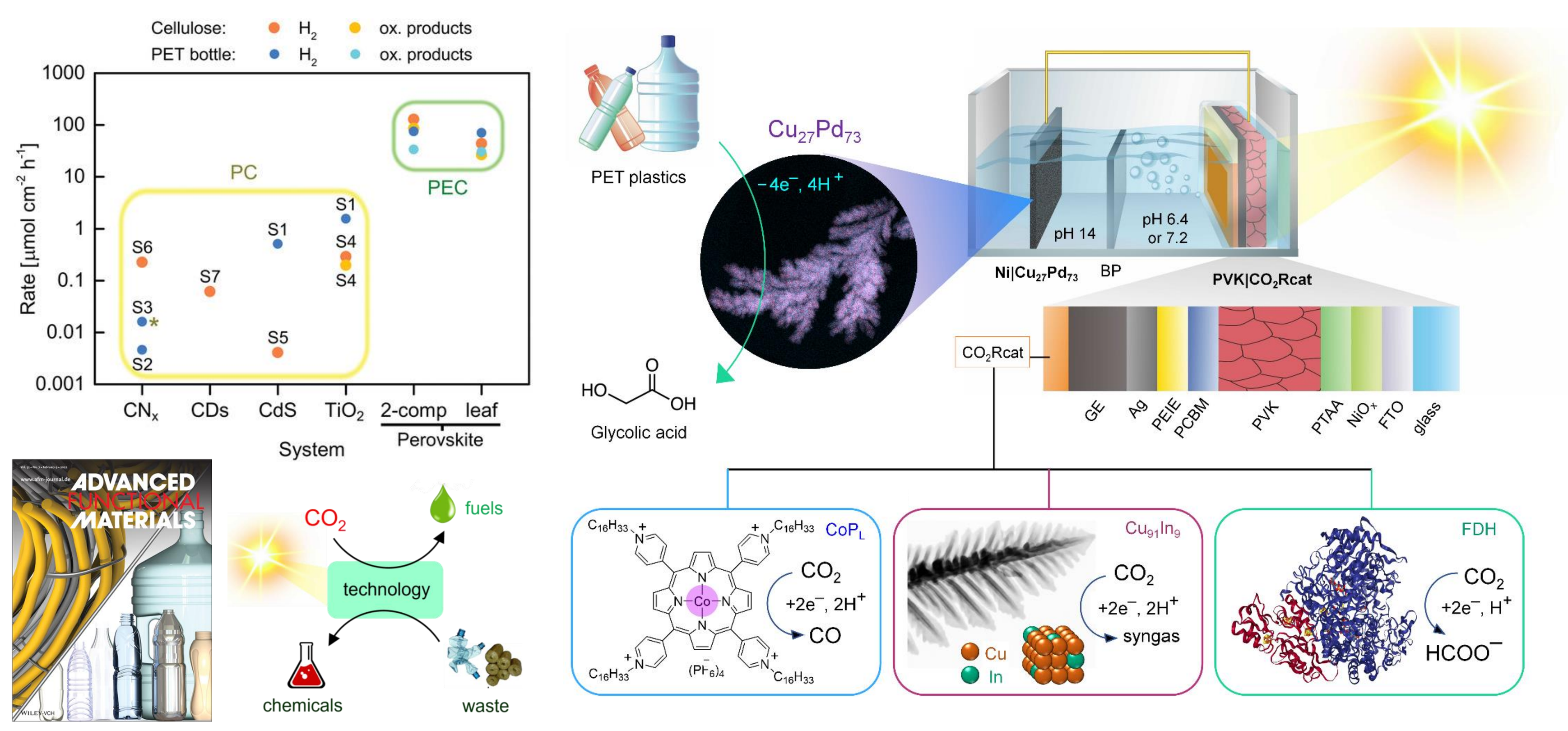




Oxidation products
PET powder – **Glycolic acid** [**2-comp** - **82 µmol** (Selectivity **94%**) **Leaf** - **90 µmol** (Selectivity **89%**)]
PET bottle – **Glycolic acid** [**2-comp** - **93 µmol** (Selectivity **91%**) **Leaf** - **84 µmol** (Selectivity **87%**)]
Cellulose – **Gluconic acid** [**2-comp** - **213 µmol** (Selectivity **67%**) **Leaf** - **60 µmol** (Selectivity **61%**)]

5. Conclusions and Beyond H_2 Production

- **First** demonstration of **bias-free**, **single light-absorber PEC systems** for **efficient** waste reforming from **diverse sources**
- PEC systems achieved product formation rates of up to ~130 µmol cm^{-2} h^{-1} - **10^2 – 10^4 times higher** than conventional PC reforming. High selectivity of oxidation products **(60-90%)** obtained
- **Configuration versatility**: Two-compartment beneficial for non-transparent waste streams and integrated 'artificial leaves' enable easy device retrieval and reuse
- The high performance single light-absorber PEC system can be envisioned to perform other fuel forming reactions such as **CO_2 reduction**, thereby increasing the product scope in future development



6. Acknowledgements

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7. References

[1] Uekert, T. *et al. Nat. Sustain.* **2021**, *4*, 383-391.
[2] Nguyen, V. *et. al. ACS Catal.* **2021**, *11*, 4955.
[3] Bhattacharjee, S. *et al. Adv. Funct. Mater.* **2022**, *32*, 2109313.
[4] Andrei, V. *et al. Nat. Mater.* **2020**, *19*, 189-194.
[5] Bhattacharjee, S. *et al. Nat. Synth.* **2022** (in review)

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