

## **Hydrogen Production**

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## QUESTIONS – H<sub>2</sub> PRODUCTION

 Why is there so much discussion about hydrogen?
 Hydrogen production – why should we and how can we produce green and blue hydrogen?
 How mature are the most sustainable routes to hydrogen and how their sustainability can be described?

# HYDROGEN PRODUCTION

## FUTURE PERSPECTIVE OF HYDROGEN

- Implementation of hydrogen as an alternative energy carrier
- Hydrogen as a sustainable feedstock for the industry
- Challenges and needs in hydrogen production for the future development remain – need for basic and applied research exist

## WHY $H_2$ ?

- Clean energy carrier, fostering green transition
- Important molecule in various industrial fields
- Emission-free fuel for vehicles and energy production → Less harmful compounds
- Production from water and renewable raw materials, bio-based wastes and side-streams
- One of the driving forces for global economy in the future



### GOVERNMENT'S CLIMATE POLICY: CLIMATE-NEUTRAL FINLAND BY 2035

https://ym.fi/en/climate-neutral-finland-2035

**The Chemical Industry Federation of Finland, 2021:** Cap the Carbon! Aiming for a Climate Neutral Finland

 10 of the most interesting technologies for achieving carbon neutrality in industry: ....New catalysts...



https://kemianteollisuus.studio.crasman.fi/file/dl/i/ii4Qug/M40Mi5b-uqOweSbta2zDOA/ Cap\_the\_Carbon\_.pdf

- Climate neutral Europe and the Green deal
  - The 2050 carbon neutrality target
  - The 2030 target of 55% emission reductions legally binding
- Finland: Carbon-neutrality by 2035
  - The world's first fossil-free welfare society
- The means to achieve the target
  - New decisions on climate policy and nearly emissions-free electricity and heat production by the end of 2030s
  - Reducing the carbon footprint of building
  - Promoting circular economy and climate-friendly food policy
  - More taxes on environmentally harmful activities
- The decline in biodiversity
  - Reforming the nature conservation legislation
  - Increasing the funding for the protection of biodiversity
  - Promoting sustainable use of natural resources
- Emissions reductions will be implemented in a way that is fair and just, both socially and regionally

#### **CAN FINLAND INCREASE CLEAN HYDROGEN PRODUCTION AND USE?**

- **Good wind resources** (offshore/onshore)
- A fairly strong electricity grid
- A full and working value chain for hydrogen
- Experience in large-scale industrial use of hydrogen
- Natural gas pipeline to store/carry hydrogen
- **Cost-effective transport of hydrogen by trucks**
- Large refinery and biofuel industry
- A major steel manufacturing company reforming the steel-making process - ~30% increase in hydrogen production and use
- Opportunity to create wide range of new H<sub>2</sub>-based businesses throughout the hydrogen value chain



Juhani Laurikko, Jari Ihonen, Jari Kiviaho, Olli Himanen, R

Ville Saarinen, Janne Kärki, Markus Hurskainen

#### Six areas in clean hydrogen:

- Hydrogen transmission & distribution
- Industrial applications of hydrogen
- Hydrogen for mobility
- Hydrogen for energy
- Residential applications of hydrogen

## AGENDA 2030 AND THE 17 SUSTAINABLE DEVELOPMENT GOALS (SDG) & TODAY'S MEGATRENDS AND DRIVING FORCES



By Eeva Forman, SYKE, http://www.syke.fi/fi-FI/Asiantuntijat/Eeva\_Furman(1877)

- Circular economy, Bioeconomy, Circular bioeconomy Low carbon society, Mitigation of climate change and biomass loss
- Energy transition, Hydrogen economy
- Renewable resources and energy
- Sustainable use of natural resources
- Sustainable production and products
- Integrated biorefineries of the future
- New value-added biofuels, biomaterials, biochemicals
- Enabling technologies, e.g., catalysts

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### HYDROGEN SUPPLY AND DEMAND, CLEAN HYDROGEN PRODUCTION



Zhang B, Zhang A-X, Yao R, Wu Y-H & Qui J-S (2021) progress and prospects of hydrogen production: Opportunities and challenges. J. Electronic Science and technology 19, 100080





## HOW TO PRODUCE CLEAN AND GREEN HYDROGEN?



Osman et al. 2021; <u>https://doi.org/10.1007/s10311-021-01322-8</u> IEA (2021) Global Hydrogen Review 2021, page 108; <u>https://iea.blob.core.windows.net/assets/5bd46d7b-906a-4429-abda-</u> e9c507a62341/GlobalHydrogenReview2021.pdf

- In 2020, sources of hydrogen production were:
  - Natural gas 59%, By-products 21%, Coal 19%, Oil 0.6%, Fossil fuels 0.7% (totally 90 Mt H<sub>2</sub>)
  - Totally fossil-based CO<sub>2</sub> almost 900 Mt/year



## **HYDROGEN PRODUCTION – CONVENTIONAL PROCESSES**



Bhandariet al. 2012; Life Cycle Assessment of Hydrogen Porduction Methods – A Review; IEK-STE

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### Reforming, Steam Methane Reforming (SMR)

- Methane and water vapour react over e.g., Ni/Al<sub>2</sub>O<sub>3</sub> or Ni/Al<sub>2</sub>O<sub>3</sub>-ZrO<sub>2</sub> catalyst (strongly endothermic reaction; ∆H<sub>SR</sub> = 206 kJ/mol) to hydrogen and carbon monoxide
  CH<sub>4</sub> + H<sub>2</sub>O + heat → CO + 3 H<sub>2</sub>
- Process conditions: 700 850 °C and 3 25 bar
  - Additional H<sub>2</sub> production by the reaction of CO and H<sub>2</sub>O via mildly exothermic ( $\Delta H_{WGSR}$ 
    - = -41 kJ/mol) water-gas-shift reaction
      - $\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2 + \text{heat}$





### **Goal gasification**

- Endothermic conversion of carbon to CO and H<sub>2</sub>
  - $C(s) + H_2O + heat \rightarrow CO + H_2 + CO_2 + CH_4$



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## **HYDROGEN PRODUCTION – CONVENTIONAL PROCESSES**



Bhandariet al. 2012; Life Cycle Assessment of Hydrogen Porduction Methods –A Review; IEK-STE

#### Autothermal reforming, ATR

- Methane and oxygen react (mildly exothermic reaction; ΔH<sub>ATR</sub> = -24.5 kJ/mol) to hydrogen and carbon monoxide
- Methane is partially oxidized

 $\begin{array}{r} \mathsf{CH}_4 + \mathsf{0.5O}_2 \rightarrow \mathsf{CO} + \mathsf{2H}_2 + \mathsf{heat} \\ 2\mathsf{CH}_4 + \mathsf{O}_2 + \mathsf{CO}_2 \rightarrow \mathsf{3H}_2 + \mathsf{3CO} + \mathsf{H}_2\mathsf{O} \\ 4\mathsf{CH}_4 + \mathsf{O}_2 + \mathsf{2H}_2\mathsf{O} \rightarrow \mathsf{10H}_2 + \mathsf{4CO} \end{array}$ 

• Process conditions: 950 – 1050 °C and 30-50 bar

#### Dry reforming, DR

- A strongly endothermic reaction between methane and carbon dioxide to CO and hydrogen
  - $\mathrm{CH_4} + \mathrm{CO_2} \rightarrow \mathrm{2CO} + \mathrm{2H_2}$

 $\Delta H_{DR}$  = 259 kJ/mol

• Ni-, Rh-, Ru-catalysts

#### Partial oxidation, POX

- Mildly exothermic reaction of methane and oxygen to CO and H<sub>2</sub>
  - $\mathbf{CH}_{4} + \frac{1}{2}\mathbf{O}_{2} \rightarrow \mathbf{CO} + \mathbf{2H}_{2} \qquad \qquad \Delta \mathbf{H}_{\mathsf{POX}} = -36 \text{ kJ/mol}$

#### Water-gas-shift reaction, WGSR

- Mildly exothermic ( $\Delta H_{WGSR}$  = -41 kJ/mol) reaction of CO and H<sub>2</sub>O to CO<sub>2</sub> and H<sub>2</sub>
  - $CO + H_2O \rightarrow H_2 + CO_2 + heat$
  - $Fe_2O_3$ -Cr<sub>2</sub>O<sub>3</sub> and CuO/ZnO catalysts at 300 450 °C and at 200 250 °C, respectively

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#### HOW TO PRODUCE $H_2$ ? – REFORMING & የማ (PHOTO)CATALYSIS AND SUSTAINABILITY IN HYDROGEN AND BIOGAS PRODUCTION AT ECE/UOULU

- Use of industrial side streams, organic load in wastewaters and biomass in H<sub>2</sub> production
- Use of microreactors, H<sub>2</sub>-selective membranes and CNT catalysts



cells

### HYDROGEN PRODUCTION – PHOTOCATALYTIC PRODUCTION OF H<sub>2</sub>







Adv. Funct. Mater. **2011**, 21, 126; JPCC, **2011**, 115, 2805; ACS Nano, **2008**, 2, 1492; Nano Res. **2011**, 4, 360; J. Photochem. Photobiol. A, **2010**, 216, 115., Catal. Comm. **2009**, 10, 538; Science, **2001**, 293, 269; ACS Appl. Mater. Interfaces, **2011**, 3, 229; Inorg. Chem., **2010**, 49, 2017.

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- Hydrogen has been reported to be produced photocatalytically in the early 1970's, UV + H₂O + semiconductive material → H₂
- Waste streams containing organic substances, e.g. alcohols and organic acids may also be used for photocatalytic H<sub>2</sub> production
  - $\rightarrow$  Combined water purification and energy production
  - $\rightarrow$  Energy by low costs
- Processes at low temperatures
- Immobilized and suspended catalyst systems
- Catalyst materials can be taylored for a particular reaction due to a large variety of phases, doping and decoration
- Large variety of synthesis methods to tailor size, phase, stoiciometry (PVD/CVD, solid-state, wet chemical reactions)
- Reaction with visible light pose green and renewable processes
- <u>Photocatalytic materials find use also in conventional catalysis,</u> <u>chemical sensors, nanoelectronics</u>

## **METHANE PYROLYSIS**



https://www.efzn.de/fileadmin/documents/Niedersaechsische\_Energietage/Vortr%C3%A4ge/2019/ NET2019\_FF1\_04\_Bode\_Rev1.pdf

- H<sub>2</sub> production through thermal decomposition, i.e., by pyrolysis of methane (mildly endothermic reaction)  $CH_4(g) \longrightarrow C(s) + 2H_2(g)$   $\Delta H_{DR} = 37 \text{ kJ/mol}$
- Low energy demand
- Solid carbon as the 2<sup>nd</sup> product: carbon black, carbon fibres, carbon nanotubes
- Pyrolysis occurs on the surface of the carbon particles
- Several possible reactor designs (moving-bed, fluidizedbed and liquid bubble column)
- Image on the right depicts a moving-bed reactor:
- Counter current flow
- Electrically heated
- Process temperature between 800-1400 °C
- Present reactor concept are not mature for industrial use (TRL 4-6)

### WATER ELECTROLYSIS (WE) AND ITS HISTORICAL BACKGROUND

- WE is a process which uses electricity to split water into hydrogen and oxygen
- The process produces clean energy with oxygen as the only by-product
- The generated H<sub>2</sub> can be injected into grid, used in a fuel cell or in chemicals and fuels production
- The energy demand for WE process can be supplied by electrical or thermal energy
- WE is an old technology known for over 200 years
- In 1789, Dutchmen Adriaan Paets van Troostwijk (1752–1837) and Jan Rudolph Deiman (1743–1808) observed the decomposition of water into a mixture of "combustible air" and "life-giving air" caused by electric discharges between two gold electrodes
- The phenomena of water decomposition initiated by direct current was observed by two Englishmen William Nicholson (1753–1815) and Anthony Carlisle (1768–1840) in 1800
- The basic physical law of electrolysis was discovered in 1834 by the English scientist Michael Faraday (1791– 1867)

### WATER ELECTROLYSIS (WE)AND ITS HISTORIC BACKGROUND

- By 1902 there were more than 400 industrial electrolyzers already in operation worldwide
- In 1940, the first large electrolysis plant with a capacity of more than 10,000 m<sup>3</sup> H<sub>2</sub>/h, built by the Norwegian company Norsk Hydro Electrolyzers, went into operation
- In 1948 the first pressurized electrolyzer was manufactured
- In 1966, the first solid polymer electrolyte membrane electrolyzer was built
- In 1972, the development of solid oxide electrolyte electrolyzers was started
- Today several companies are active in the manufacture and development of electrolysis technologies: Proton, Hydrogenics, Giner, and ITM Power being leaders in the field
- Water electrolysis, WE

 $H_2O(I) \rightarrow H_2(g) + 0.5O_2(g)$  △H = 286 kJ/mol

- High energy demand, water as a raw material
- Endothermic ( $\Delta H > 0$ ) and non-spontaneous ( $\Delta G > 0$ ) chemical reaction

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# DIFFERENT APPROACHES FOR WATER ELECTROLYSIS IN INDUSTRIAL SCALE

- Alkaline Electrolyzers (AEL)
- Proton exchange membrane electrolyzers (PEM)
- Solid oxide electrolyzer cell (SOEC)



**PEM, AEL, AND SOEC ELECTROLYSIS** 



Ausfelder et al.; 978-389746-237-3

University of Oulu

### ALKALINE ELECTRO-LYZERS, AEL IN WATER ELECTROLYSIS



- The most mature technology
- It has been applied for large-scale hydrogen production in the industries since the beginning of the 20th century
- Conventional AEL are usually operated at current densities in the range of 300–500 mA/cm<sup>2</sup> and at corresponding cell voltages in the range of 1.9–2.4 V
- **Operating temperature:** 70-90 °C, and in either atmospheric or elevated pressure
- Higher efficiency and product purity (99.5+% H<sub>2</sub> purity)
- **System efficiency:** 60-80% depending on system size, purity and pressure levels
- **Pressurized AEL (up to 15 bar)** produced compressed hydrogen that is ready for grid injection with minimal energy input
- But they are more expensive (20-30%) than the atmospheric systems
- AEL cell is made of two electrodes that are immersed completely in 30 wt.% aqueous KOH electrolyte
- The electrodes are made of nickel or nickel plates ity of Outlu



### **Pros:**

• Durability, maturity, availability and comparatively low specific costs

#### Cons:

- Low current densities: significantly influence specific system size and H<sub>2</sub> production costs.
- Low operating pressures

- A microporous diaphragm separates the anode and cathode.
- Direct current is applied to the AEL, H<sub>2</sub> and OH ions evolves at the cathode according to the following reaction:

 $2\mathrm{H}_2\mathrm{O} + 2\mathrm{e}^{\scriptscriptstyle -} \to \mathrm{H}_2 + 2\mathrm{O}\mathrm{H}^{\scriptscriptstyle -}$ 

• The OH ions are migrating through the microporous separator and are oxidized at the anode according to the following reaction:

 $2\text{OH}^{-} \rightarrow 1/2\text{O}_2 + \text{H}_2\text{O} + 2\text{e}^{-}$ 

• Water is consumed but not KOH and therefore water has to be supplied continuously (neglecting physical electrolyte losses)

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## PROTON EXCHANGE MEMBRANE ELECTROLYZERS, PEM

- PEM development started in 1966 by General Electric
- First commercialization was in 1978
- System efficiency 60-70%
- Operation: 60-80 °C, 30-60 bar, voltage range: 1.6-2 V
- H<sub>2</sub> purity: 99.99+%



- Solid polymeric membrane is used as electrolyte
- PEM systems are usually operated at current densities of 1–2 A/cm<sup>2</sup>, which is about a factor of 4 higher compared to AEL
- At the anode, pure water is oxidized to O<sub>2</sub> and release protons that flow through the membrane and are reduced at the cathode to form H<sub>2</sub> according to the following respective reactions:
  - $H_2O \rightarrow 1/2O_2 + 2H^+ + 2e^-$
  - $2H^+ + 2e^- \rightarrow H_2$

<u>10.1016/B978-0-444-56352-1.00002-7</u>

## کہ PEM: Pros and cons



<u>10.1016/B978-0-444-56352-1.00002-7</u>

### Pros:

- Faster cold start, higher flexibility, and better coupling with dynamic and intermittent systems compared to AEL
- Can produced highly compressed hydrogen

### Cons:

• High cost of the catalyst support materials and the membrane

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### SOLID OXIDE ELECTROLYZER CELL, SOEC



10.1016/j.mset.2019.03.002

- SOEC is at early stage of development
- A thin, dense solid oxide layer is used as electrolyte
- The core SOEC components are usually made up of ceramic materials
- SOEC uses waste heat instead of part of the electricity needed in the process
- SOEC operates under high temperature (800-1000 °C): higher efficiency (>90%) compared to AEL and PEM
- The solid electrolyte is typically made up of ZrO<sub>2</sub> doped with 8% Y<sub>2</sub>O<sub>3</sub>
- At the cathode, steam is reduced to produce H<sub>2</sub>:

 $H_2O+2e^- \rightarrow H_2+O^{2-}$ 

21 Doctoral student Ahmed Dahiru & Prof. Riitta Keiski, UOulu, Faculty of Technology, Environmental and Chemical Engineering Research unit

## SOEC: PROS AND CONS



<sup>10.1016/</sup>j.mset.2019.03.002

### **Pros:**

- Significant increase in ionic conductivity and the rates of electrochemical reactions due to high-temperature operation
- Reduction in total electricity demand that can be compensated by supply of thermal energy

### Cons:

- Material decomposition and short-term stability
- Difficult to operate with fluctuating and intermittent power

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## ELECTROLYSIS AND PHOTOCATALYSIS -COMPARISON





- **Hydrolysis** (electrolytic water splitting) is technology to produce hydrogen using energy from renewable source (solar light, wind, hydropower).
- The reaction has a standard potential of -1.23 V.
- Efficiency of modern hydrogen generators is measured by energy consumed per standard volume of hydrogen (MJ/m<sup>3</sup>).
- **Practical electrolysis** (using a rotating electrolyser at 15 bar pressure) consume 50 kilowatt-hours per kilogram (180 MJ/kg), 15 kilowatt-hours (54 MJ) more is needed if the hydrogen is compressed e.g. for use in hydrogen cars
- Hydrogen can be produced using photocatalysis, i.e., water splitting (under research) and from organic molecules (e.g., ethanol, organic load in wastewaters) in aqueous phace (under research) using UV or solar light and semiconductive photocatalysts

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## H<sub>2</sub> PURIFICATION

- Hydrogen has to be purified before it is used e.g., in chemical industry, transportation, in fuel cells.
- Hydrogen purification can be done mainly with three methods:
  - Chemical (e.g. ,adsorption, absorption)
  - Physical (e.g., metal hybrides, cryogenic)
  - Selective diffusion (vaious types of membranes)
- Typically, it is purified by using a **pressure swing adsorption** (PSA) purity higher than 99.99% is obtained.
- The adsorbents used most often are e.g., activated carbon, silica gel, alumina and zeolites.
- Another widely used purification method is membrane purification: 1) Inorganic membranes (metallic or ceramic membranes) and 2) organic membranes (polymeric membranes)
- The selection of H<sub>2</sub> purification technique is solely dependent on the operating conditions, specifications and the end use requirements
  - **Criteria:** low T and p, O<sub>2</sub> and Cl<sub>2</sub> resistance, easy operation, high selectivity and fluxes, durability.

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H<sub>2</sub> stream purification from a chlorate process: possible purificaion steps needed after the water scrubbing unit:



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## HYDROGEN PRODUCTION – TECHNOLOGY READINESS LEVEL, TRL

### TECHNOLOGY READINESS LEVEL (TRL)

TRL	Definition
9	Actual system "flight proven" through successful mission operations
8	Actual system completed and "flight qualified" through test and demonstration (ground or space)
7	System prototype demonstration in a space environment in a space environment
6	System/subsystem model or prototype demonstration in a relevant environment (ground or space)
5	Component and/or breadboard validation in relevant environment
4	Component and/or breadboard validation in laboratory environment
3	Analytical and experimental critical funsciotn and/or charateristic proof-of-concept
2	Technology concept and/or application formulated
1	Basic principles observed/reported

Rybicka, J.; Journal of Cleaner Production 112 (2016) 1001-1012; http://dx.doi.org/10.1016/j.jclepro.2015.08.104

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# TRL OF KEY HYDROGEN PRODUCTION TECHNOLOGIES



### HYDROGEN PRODUCTION – TECHNOLOGY READINESS LEVEL, TRL

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2	Technology concept and/or application formulated
1	Basic principles observed/reported



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Notes: Technologies included are at large prototype or at a more advanced stage. CCUS = carbon capture utilisation and storage. For more detailed information on individual technology designs for each of these technologies, and designs at small prototype stage or below, please visit: <a href="https://www.iea.org/articles/etp-clean-energy-technology-quide">www.iea.org/articles/etp-clean-energy-technology-quide</a>.



## HYDROGEN AS AN ENABLER OF ENERGY TRANSITION

Pulling forward early demand to enable clean hydrogen scaling will follow sequencing of demand sector "take off" over next 3 decades



SOURCE: SYSTEMIQ analysis for the Energy Transitions Commission (2021)



### H<sub>2</sub> VIA REFORMING & PHOTO-CATALYSIS FROM INDUSTR-IAL SIDE STREAMS AND BIOMASS AT ECE/UOULU



- Reforming in H<sub>2</sub> production over nanostructured base metal (e.g., over conventional catalysts, CNTs, nanofibres) catalysts.
- Photocatalytic H<sub>2</sub> production over non-PGM nanomaterials.
- Catalytic routes from biomass, industrial gases (including CO<sub>2</sub>), organic load in WWs, water to fuels (e.g., hydrogen, alcohols, hydrocarbons)
- Purification of gaseous streams using membrane technology and hybrid systems (e.g., photocatalytic membranes and membrane reactors).
- Catalyst deactivation studies in reactive conditions (poisoning, structural changes).
- Sustainability assessment analyses.
- Thermodynamic calculations and reactor design.

# THE BIO-CCU PROJECT



### ECE/UOULU:

- CO<sub>2</sub> purification Development of membrane gas purification for CO<sub>2</sub> containing effluent streams (e.g., lowgrade biogas)
  - Modification of waste materials to membranes
- Hydrogen sourcing Development of catalysts for biogas upgrading to hydrogen (link to on-going GREENE project)
- Development of new circular material-based catalysts and conversion routes for green fuel production (P2G, P2C)
  - Modification of waste materials to catalysts and membranes
  - Applications of hybrid technologies (catalytic membranes, adsorption/catalysis)

### Process modelling and simulation

- Development of mechanistic models for selected process concepts (mass and energy balances)
- Sensitivity analysis, flexibility/controllability of processes
- Support in techno-economic analysis and life cycle assessment
  - Integrating the process developments and process models to TEA/LCA

#### FOSSIL FREE BIOHYDROGEN AND CLEAN ENERGY FROM BIOGAS -**GREEN AT ECE/UOULU**



- The project supports the hydrogen economy, self-sufficiency and local energy production and contributes to the carbon neutrality 2035 target in Finland.
- The goal is to develop a new **thermochemical method** that utilizes biogas in the production of fossil-free biohydrogen.
- The developed method enables the utilization of biogas in hydrogen production in an atom- and energy-efficient way in order to promote low-emission, flexible and efficient decentralized energy production.
- The development includes reactor design including pretreatment, analytics and automation, the development and testing of carbon-resistant and sulphur-tolerant catalysts, Kestävää kasvua ja työtä -ohjelma and pilot-scale equipment construc



Doc. Esa Muurinen, Doc. Satu Pitkäaho, UOulu Environmental and Chemical Engineering Research Unit www.oulu.fi/environmentalengineering/greene



### HYDROGEN PRODUCTION QUESTIONS



- 1. Why is there so much discussion about hydrogen?
- 2. Hydrogen production why should we and how can we produce green and blue hydrogen?
- 3. How mature are the most sustainable routes to hydrogen and how their sustainability can be described?



## Many thanks!

### Environmental and Chemical Engineering Research unit, ECE

https://www.oulu.fi/environmentalengineering/



### Environmental and Chemical Engineering

Material Characterization development and measurements

Unit operation Process development modeling and control

Advanced data analysis and optimization

assessment

Sustainability

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Thanks to Dr.-Ing. Philip Biessey from Ruhr-Universität Bochum, Institut für Thermo- und Fluiddynamik, for his lectures in 2022 and the lecture materials that motivated and helped me during the preparation of these lectures