



UK Government

# 1rsion of Captured CO<sub>2</sub> to Industrial Chemicals - CoCaCO<sub>2</sub>la

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## Acknowledgements

The CoCaCo2la project has received £518,777.19 in funding from BEIS under the UK ACT ERA-NET EC GA 691712. This funding has been made available from the Government's £1 Billion Net Zero Innovation Portfolio to provide key innovation to develop technologies needed to tackle climate change.



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# Project report

The report summarises the objectives and key findings of the project Conversion of Captured CO<sub>2</sub> to Industrial Chemicals - CoCaCO<sub>2</sub>la.

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## Executive summary

The CoCaCO<sub>2</sub>la project focused on developing innovative technologies to convert captured CO<sub>2</sub> into valuable products, primarily ethylene. A comprehensive technology gap analysis identified four main gaps: limited efforts in producing ethylene from CO<sub>2</sub> using amine solutions, the need for optimized hydrodynamic conditions for CO<sub>2</sub> reduction, the development of scalable electrocatalysts, and the absence of ethylene purification technologies. Six key performance indicators (KPIs) were established to measure CO<sub>2</sub> capture efficiency, utilization, energy efficiency, scalability, and product purity. The project selected and tested a solvent that showed optimal trade-offs in thermodynamics, reactivity, and sustainability, and enhanced CO<sub>2</sub> loading and absorption when mixed with known CO<sub>2</sub> capture solvents. The electrochemical cell model was validated, confirming higher product current density and lower undesirable hydrogen current density. Strategies for catalyst development were defined, and a copper salt was identified as a stable precursor for copper catalysts. Optimized production parameters led to well-adhered coatings, with durability tests showing up to 80% conversion of CO<sub>2</sub> to carbon-containing products. A silver facilitated transport membrane (Ag-FTM) demonstrated good selectivity and long-term stability for ethylene separation from gas mixtures. A flow-through electrochemical system successfully converted captured CO<sub>2</sub> to CO and ethylene with high efficiencies. Life Cycle Assessment (LCA) and Life Cycle Costing (LCC) frameworks were defined to evaluate environmental impacts, showing a significant reduction in climate change impact compared to conventional processes. A techno-economic assessment tool was created to estimate production costs. A bespoke testing system was developed to evaluate material performance in carbon capture and storage environments. The project has resulted in significant progress in CO<sub>2</sub> conversion technologies, addressing key gaps, optimizing processes, and evaluating environmental and economic impacts. The collaborative efforts and innovative solutions have the potential to advance carbon capture and utilization, contributing to more sustainable and efficient industrial processes.

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## Role and contributions of each project partners

The CoCaCO<sub>2</sub>la consortium consisted of a strategic alliance of partners, each playing a crucial role in the project based on their expertise. TWI (UK) is the project coordinator, leveraging its extensive experience in collaborative research projects and advanced thermal spray technology to oversee project performance and manage materials preparation for copper catalysts. Idaho National Laboratory (INL) utilised its capabilities in electrochemical processes and membrane-based gas separation to design, construct, and test the CO<sub>2</sub> electrolyser. The University of Leicester (ULEIC) contributed its expertise in surface engineering and electrochemistry to define and generate catalyst materials, collaborating with TWI on optimizing catalytic performance through advanced processing techniques. CERTH (Greece) developed and tested a new CO<sub>2</sub> capture solvent system, transferring its technology to work with copper catalyst electrodes, supported by its pilot-scale CO<sub>2</sub> capture units and process optimization expertise. TVS (UK) performed Life Cycle Analysis (LCA), Life Cycle Cost Analysis (LCCA), and Social Impact Analysis (SIA), developed a business plan for future technology exploitation based on these assessments. PTML (UK), part of the NSG Group, provided access to its glass manufacturing process and data on furnace waste gases, ensuring the technology development aligning with commercial needs. Collectively, these partners produced a wealth of knowledge and resources to advance the field of carbon capture and utilization, contributing to more sustainable industrial processes.

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## Short description of activities and results

A circular carbon economy aims to produce commodity chemicals from CO<sub>2</sub> emissions, turning them into value-added commodities such as polymers. Ethylene, a widely produced commodity chemical primarily used for polyethylene production, is a significant target due to its high CO<sub>2</sub> emissions. Traditional ethylene production processes are highly carbon-intensive, emitting 1 to 2 tons of CO<sub>2</sub> per ton of ethylene produced. By developing technologies to produce ethylene from CO<sub>2</sub>, significant carbon emission reductions can be achieved. **The CoCaCO<sub>2</sub>1a project focused on developing a low-temperature, intensified carbon capture and utilization (CCU) process to produce ethylene.**

### **CO<sub>2</sub> Capture and Delivery to the Electrochemical System**

The CO<sub>2</sub> capture process traditionally involves two main unit operations: absorption of CO<sub>2</sub> by amine-based solutions and stripping to release CO<sub>2</sub> by heating. This process is energy-intensive, particularly the stripping operation. **The CoCaCO<sub>2</sub>1a project proposed an isothermal capture and conversion process, where CO<sub>2</sub> release, conversion, and capture media regeneration occur within an electrolyser, forming an isothermal loop for carbon capture.**

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## WP 1: Technology Gap Analysis and KPI Definition -INL

The project explored various capture chemistries, including aqueous tertiary amine solutions, ionic liquids, covalent organic frameworks, and phase change solvents (PCS). Each approach has unique benefits and challenges. For example, alkaline solutions show promise for CO<sub>2</sub> capture and co-electrolysis to syngas but can suffer from carbonate salt deposition on the electrocatalyst. The use of PCSs, which capture CO<sub>2</sub> as bicarbonate, is emerging but underexplored, especially for ethylene production.

*Technology Gap 1:* Ethylene production from CO<sub>2</sub> captured in PCSs has not been reported.

### *Electrochemical Cell Configuration*

High purity CO<sub>2</sub> gas can be delivered to an electrochemical cell for conversion to desired products. Gas-fed electrolyzers (GFE) have made significant progress in terms of activity and selectivity towards ethylene. However, GFEs typically achieve low CO<sub>2</sub> conversions. The CoCaCO<sub>2</sub>la project explored liquid feed inputs where CO<sub>2</sub> is captured in an aqueous solution, aiming to optimize the electrochemical cell configuration for CO<sub>2</sub> reduction.

Two configurations were proposed to be considered: buffer layer and zero gap. The buffer layer configuration promotes effective CO<sub>2</sub> release but may result in higher cell voltages. In contrast, the zero-gap configuration offers lower cell voltages but may favour hydrogen evolution over CO<sub>2</sub> reduction.

*Technology Gap 2:* Need to define the optimum hydrodynamic conditions for captured CO<sub>2</sub> electrochemical reduction.

### *Electrocatalysts (Cathode)*

The cathode plays a crucial role in the electrochemical reduction of CO<sub>2</sub>. Copper (Cu) is the only metal known to produce C<sub>2</sub> products, including ethylene. The CoCaCO<sub>2</sub>la project aimed to develop electrocatalysts capable of operating under flooded conditions with low faradaic efficiency (FE) for hydrogen evolution. Current research highlights the potential of sputtered Cu on semipermeable PTFE membranes and anion exchange ionomers to enhance CO<sub>2</sub> conversion efficiencies.

*Technology Gap 3:* Developing an electrocatalyst capable of operating under flooded conditions with low FE for hydrogen evolution.

### *Membranes*

Membranes are essential for separating the anode and cathode compartments, enabling different chemistries or pH environments. Three types of membranes were considered:

- Cation Exchange Membranes (CEM): Transport H<sup>+</sup> ions from anode to cathode, facilitating CO<sub>2</sub> release but prone to water crossover, limiting local low pH development and CO<sub>2</sub> reduction efficiency.

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- Anion Exchange Membranes (AEM): Transport  $\text{OH}^-$ ,  $\text{HCO}_3^-$ ,  $\text{CO}_3^{2-}$  ions, maintaining alkaline environments at the anode but causing significant  $\text{CO}_2$  losses at the anode and limited  $\text{CO}_2$  release at the cathode.
  - Bipolar Membranes (BPM): Combine CEM and AEM properties, reducing element crossover and promoting high  $\text{CO}_2$  reduction efficiencies but requiring high operating voltages.

*Technology Gap 4:* Identify and optimize membrane types for efficient  $\text{CO}_2$  reduction in the proposed isothermal CCU system.

#### *Isothermal CCU Configuration*

The CoCaCO<sub>2</sub>la project envisioned an isothermal CCU system where  $\text{CO}_2$  capture and conversion occur within a pressurized electrolyzer. This configuration aimed to achieve high  $\text{CO}_2$  conversion rates while maintaining self-pressurization through gas evolution. Liquids and gases leaving the electrolyzer will be separated in a gas-liquid separation column, with downstream gas purification tailored to the specific gas mixture produced.

#### *Gas and Vapour Separations for Ethylene Recovery*

Ethylene recovery from the mixed-gas feed stream will be a complex process requiring multiple separation steps. Membrane separations are preferred over cryogenic distillation for their lower energy consumption. Facilitated transport membranes (FTMs) using  $\text{Ag}^+$  ions are promising for selectively binding ethylene over ethane, despite challenges related to  $\text{Ag}^+$  instability and membrane degradation. The project will focus on flat sheet technologies for initial separations, potentially scaling up to hollow fibre or spiral-wound modules for larger operations.

#### *Membrane Separation Technologies*

##### Glassy and Rubbery Polymers

Membrane separations for gas mixtures, particularly for ethylene/ethane separation, leverage the properties of glassy and rubbery polymers.

- Glassy Polymers: These polymers separate gases based on molecular size, using the solution diffusion model where permeability (P) is a product of diffusion (D) and solubility (S). Examples include polyetherimides (PEI), polyamide-imides (PAI), and polyimides (PI). These materials are effective in separating smaller kinetic diameter gases, such as ethylene, from larger molecules.
- Rubbery Polymers: These polymers separate gases based on solubility differences. Polydimethylsiloxane (PDMS) is a common example, partitioning gases with stronger solubility interactions at higher permeation rates. This property is useful for gases like ethane, which have higher solubility in rubbery polymers

#### *Challenges and Considerations*

The primary challenge in ethylene recovery from a gas mixture is the presence of hydrogen ( $\text{H}_2$ ), which constitutes a significant portion ( $\geq 70$  vol%) of the feed stream. Hydrogen's small

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kinetic diameter (2.89Å) and high permeation rate necessitate its removal before further gas separations. This stage is critical to avoid pressure drops and ensure efficient operation of subsequent separation processes.

*Hydrogen Separation:* Conventional polymeric membranes require multiple stages to effectively remove H<sub>2</sub>, potentially leading to significant pressure drops and increased capital and energy costs. Sintered noble metal membranes, such as palladium or platinum, offer excellent H<sub>2</sub> separation but demand high operational temperatures (>400°C), making them unsuitable for this application. Emerging electrochemical (EC) separation techniques present a potential solution but require further research to validate their effectiveness at high H<sub>2</sub> concentrations and scalability.

### *Ethylene Separation Strategy*

After addressing hydrogen separation, the focus shifted to the remaining gases—methane, carbon monoxide (CO), and ethylene. The separation process aimed to enrich ethylene concentrations from an initial 10-15 vol% to potentially 50 vol%, following the removal of other gases.

- Silver Facilitated Transport Membranes (FTMs): Research at INL has demonstrated that silver FTMs can achieve high selectivity and permeability for ethylene. Initial experiments will analyze a 1:1 mixture of ethylene/methane-ethane, considering the influence of CO if present in significant concentrations. The performance of silver FTMs in these conditions will be critically evaluated to optimize the separation process.

### **Key Performance Indicators (KPIs)**

The success of the gas separation technology was proposed to be measured using the following KPIs:

- Permeability and Selectivity: Achieving high permeability and selectivity for ethylene over other gases.
- Energy Efficiency: Minimizing energy consumption during the separation process.
- Operational Costs: Reducing capital and operational costs through optimized separation stages.
- Scalability: Ensuring the technology can be scaled from laboratory to industrial applications.
- Purity of Recovered Gases: Attaining high purity levels of recovered ethylene and hydrogen.

The technology gap analysis, KPIs were proposed as a part of WP1 and is reported in Deliverable D1.1-1.2. System definition was reported in Deliverable D1.3

A summary of the achievements and outcomes along with conclusions, future direction is presented below.

### **Achievements:**

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- Establishment of technology gap document
  - System's definition document
  - Key performance indicators for the project.

### **Outcomes:**

A living document containing a state-of-the art analysis was generated within the first quarter of initiating the project living document identifying four main technology gaps:

- Ethylene production from captured CO<sub>2</sub> is scarce with no efforts identified using amine capture solutions.
- Need to optimise the optimum hydrodynamic conditions for the electrochemical reduction of captured CO<sub>2</sub>
- It is necessary to develop a scalable electrocatalyst that can operate under flooded conditions with low faradaic efficiencies for hydrogen evolution.
- Ethylene purification technologies have focused on alkane/alkene separations but there is not reported technologies that aim to complex gas mixtures as those that can be generated through the electrochemical reduction of CO<sub>2</sub>.

Six key performance indicators were established targeting CO<sub>2</sub> capture efficiency, utilization, energy efficiency, scalability, and product purity.

A system definition document was prepared that highlighted:

- CO<sub>2</sub> capture and delivery system
- Integrated electrolyser cell system
- Gas purification/separation system

### **Conclusion**

This work package aimed to bridge significant technology gaps in the production of ethylene from CO<sub>2</sub>. By addressing challenges in CO<sub>2</sub> capture and delivery, electrochemical cell configuration, electrocatalyst development, membrane selection, and gas separation techniques, this work package supported the project to establish a viable, low-temperature CCU process for ethylene production, contributing to a circular carbon economy.

### **Future Directions**

Given the complexities and potential costs associated with hydrogen separation, a critical evaluation of H<sub>2</sub> recovery is essential. Integrating H<sub>2</sub> and CO<sub>2</sub> recovery processes could reduce overall system costs and enhance efficiency, though this integration lies beyond the current project's scope.

Research Needs: Further funding and evaluations are necessary to refine the separation process, particularly in integrating H<sub>2</sub> and CO<sub>2</sub> recovery and exploring advanced separation techniques.

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## WP2: CO<sub>2</sub> Capture and Delivery System

### Task 2.1: Assessment and Selection of CO<sub>2</sub> Capture Phase-Change Solvent

During the initial three months of the project, CERTH undertook Task 2.1, focusing on the assessment and selection of a CO<sub>2</sub> capture phase-change solvent. This task involved a systematic methodology to identify an appropriate solvent for the current application, including the creation of a solvent database, defining performance criteria, employing a multi-criteria assessment approach, and ultimately selecting a suitable solvent.

#### *Methodology and Criteria*

A database of 30 solvents from existing literature was compiled, encompassing both phase-change and non-phase-change solvents. The selection criteria included:

- **Thermodynamic Properties:** Solubility parameter, vapor pressure, density, viscosity, and water solubility, expressed as the relative energy difference (RED).
- **Reactivity:** Represented by solvent basicity (pKa), indicating fast CO<sub>2</sub> reaction kinetics.
- **Environmental, Health, and Safety Properties:** Including cumulative energy demand, global warming potential, and Ecoindicator-99 for LCA, along with flash point, boiling point, lethal dose (LD50), permissible exposure limit, and other environmental impact indicators.

A Pareto analysis was conducted where the solvent density and basicity were maximized, while other properties were minimized. The analysis led to the identification of MCA as a promising solvent, with high aggregate index J values indicating its suitability over alternatives like CHP, which exhibited undesirable precipitation behaviour.

### Task 2.2: Experimental Studies on Selected Solvent MCA

CERTH initiated vapour-liquid-liquid equilibrium experiments with MCA, revealing high CO<sub>2</sub> loadings and good electrical conductivity. Experiments with MCA-KOH mixtures confirmed enhanced capture capacity and conductivity, even with minimal KOH addition. Preliminary tests by INL showed low viscosity and avoidance of foaming. Further experiments involved varying KOH concentrations in MCA solutions to measure phase equilibrium behaviour and electrical conductivity. For instance:

- 33 wt% MCA at 293-294 K: Electrical conductivity of 21.5-22.5 mS cm<sup>-1</sup> for CO<sub>2</sub> loadings of 0.883-1.004.
- 31 wt% MCA + 3 wt% KOH at 40°C: CO<sub>2</sub> loadings of 2.59-3.18 mol CO<sub>2</sub> /kg solvent.
- 27.5 wt% MCA + 10 wt% KOH: Biphasic behavior at 10 kPa CO<sub>2</sub> pressure with respective loadings of 2.06 and 1.88 mol CO<sub>2</sub> /kg for aqueous and organic phases.

These experiments indicated that adding KOH enhances CO<sub>2</sub> solubility and electrical conductivity without significantly altering solubility.

### Task 2.3: Electrolyser Model Development

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A preliminary model for the electrolyser was developed and validated against experimental data. The model includes species mass balances, charge and current balances, proton diffusion, and electrode reactions. Validation showed good agreement between simulated and experimental results, especially in terms of current density versus applied potential and electrolyte flowrate effects.

Further model extensions included evaluating electrolytes like MEA and MCA. Simulations revealed higher partial current densities for MEA, indicating increased reaction rates and less H<sub>2</sub> formation compared to KHCO<sub>3</sub>. A parametric sensitivity analysis identified key performance parameters such as catholyte flowrate, electrode-membrane distance, and applied potential.

### *Additional Experiments and Modelling*

Subsequent tasks involved measuring CO<sub>2</sub> loading in MCA+KOH and MCA+Piperazine mixtures, revealing that both mixtures, initially biphasic, transitioned to a single phase upon CO<sub>2</sub> addition. Experiments with MCA+AMP showed similar behaviour with enhanced CO<sub>2</sub> solubility but slightly decreased loading per mole of amine.

Two mathematical models were developed for electrochemical CO<sub>2</sub> reduction:

- High-Resolution Model: A detailed 2D model considering mass and current balances, showing concentration variations of CO<sub>2</sub> and CO.
- Voltametric Model: A lower resolution model used for techno-economic assessment of ethylene production, identifying electricity price and Faradaic efficiency as significant cost factors.

### *Techno-Economic Analysis*

Sensitivity analysis with the voltametric model estimated the base case ethylene production cost at 1,903 €/t, with electricity price and Faradaic efficiency being major influencers. The analysis highlighted the non-linear impact of CO<sub>2</sub> price and current density on production cost.

A brief summary of the achievements and outcomes along with conclusions is presented below.

### **Achievements:**

- Selected the desired solvent
- Performed equilibrium measurements for this solvent
- Simulated the CO<sub>2</sub> absorption for the desired solvent using an existing absorber model and developed an electrochemical cell model

### **Outcomes:**

- Selected solvent exhibits optimum trade-offs in several properties pertaining to thermodynamics, reactivity and sustainability

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- Selected solvents were tested experimentally in mixtures with various known CO<sub>2</sub> capture solvents including KOH, Piperazine and AMP.
  - The equilibrium measurements indicated that small additions of these components enhance the solvent CO<sub>2</sub> loading, hence benefiting both the absorption and the delivery of larger CO<sub>2</sub> amounts in the electrochemical cell
  - The model of the electrochemical cell was validated successfully using experimental data from literature, indicating very good match.
  - It was shown that MCA and MEA present higher product current density (equivalent to the product production rate) and lower H<sub>2</sub> current density (undesirable product) compared to base case (KHCO<sub>3</sub> as electrolyte).

## **Conclusion**

WP2 successfully identified and tested MCA as a viable CO<sub>2</sub> capture solvent, with significant progress in modelling and experimental validation. Further work focused on optimizing electrolyser performance and conducting comprehensive techno-economic assessments to support the overall project goals.

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## WP3: Catalyst Material Optimisation

### **Task 3.1: Catalyst Materials Selection**

The University of Leicester (ULEIC) led Task 3.1, focusing on the selection of catalyst materials. The primary approach involved a comprehensive review of technical literature. ULEIC identified, collated, and summarised relevant scientific articles and reviews. This information was then discussed with project partners to inform the selection process. A detailed Excel file summarising prior work on copper electrodeposition, different electrolytes, and gas-separation techniques was created and regularly updated. Laboratory accessories required for copper electrodeposition experiments have been ordered, and preparatory work, including risk assessments for experimental procedures, were prepared.

#### *Key Activities:*

- Literature review and collation of information.
- Preparation of an Excel summary file on electrodeposition and related techniques.
- Ordering of lab accessories and equipment.
- Drafting and circulation of the initial report to project consortium members.

### **Task 3.2: Development and Characterisation of Precursors for Catalysts**

Based on findings from Task 3.1, electrodeposition trials of copper have been performed under various conditions. These include different modes of deposition (constant current and voltage), varying concentrations, current densities, and substrates (mild steel, mild steel mesh). The physical characterisation of the deposited copper to understand its phase composition and crystal orientation was completed.

#### *Key Activities:*

- Conducting electrodeposition trials under varying conditions.
- Physical characterisation of the copper deposits using X-ray diffraction (XRD) and optical microscopy.
- Sharing results with consortium members.

#### *Challenges:*

The need for detailed investigation into the influence of different deposition parameters on the nature of copper deposits.

### **Task 3.3: Down selection of Precursors for Catalysts**

In Task 3.3, the suitability of different precursor candidates for copper electrodeposition was evaluated. Experiments were conducted using two different copper salts. It was found that electrodeposition from certain Cu-containing electrolytes yielded superior deposits compared to others. Deposition on various substrates, including titanium mesh, galvanised steel, and stainless steel, has been tested and characterised.

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*Key Activities:*

- Electrodeposition trials using different precursor candidates and substrates.
- XRD characterisation to identify the dominant crystal facets of deposited copper.
- Optimisation of electrodeposition parameters to achieve desired crystal orientations.

*Challenges:*

Poor deposition quality from chloride-based electrolytes requiring further optimisation.

### **Task 3.4: Optimisation and Production of Electrodes**

Task 3.4 focused on optimising the electrode production process based on insights from previous tasks. Experimental studies involved combining electrodeposited copper with sprayed copper on mesh substrates. Initial performance tests conducted at INL, USA, revealed that thicker deposits might be necessary to improve the conversion of CO<sub>2</sub>. Further experiments are in progress to refine the electrodeposition parameters and improve the performance of the copper catalysts.

*Key Activities:*

- Conducting experimental studies to combine different copper deposition methods.
- Testing electrode performance at INL, USA.
- Investigating the influence of electrode position and orientation on deposit quality.

*Challenges:*

Initial poor performance in CO<sub>2</sub> conversion, necessitating thicker copper deposits.

Significant progress has been made across all tasks within WP3. Key activities have included extensive literature reviews, electrodeposition trials, and physical characterisation of copper deposits. Initial experimental results have informed optimisation efforts, and the consortium has actively shared findings to guide the development of effective catalyst materials. Despite some delays due to administrative issues, the team has managed to adhere to revised timelines and deliverables.

A brief summary of the achievements and outcomes along with conclusions is presented below.

### **Achievements:**

- A detailed review has been carried to identify the key gaps in selected materials and suitable processes for development of catalyst
- Defined the strategy to down select the precursor candidates, for the spraying process, electrodeposition and the selection of experiments
- Defined characterization strategy and optimization of recipe specification for electrode production

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**Outcomes:**

- A detailed review summarizing the key findings and gaps from the existing literature, key publications and patents has been prepared and published as a review article.
- Precursors for spraying (copper wires, powders), electrodeposition were identified, selected and characterized by reviewing their availability from the market, suitability for each process (spray, electrodeposition)
- 2 different types of powders (spherical, dendritic) were chosen as precursor candidates for obtaining cold sprayed copper. Wire based precursors were identified as the suitable candidates for obtaining thermal spray copper
- Selection of electrodeposition experiments were defined by conducting preliminary trials involving several precursor chemistries
- Visual observation and physical characterization data suggested that copper salts are suitable precursor candidates for the development of stable copper catalysts

**Conclusion:**

WP3 has successfully advanced the understanding and development of copper electrodeposition techniques, providing a solid foundation for the production of efficient catalyst materials. Continued efforts will focus on refining these techniques to meet project goals and improve overall performance.

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## WP4: Electrode Development – TWI

WP4 focused on the development of electrodes, particularly through identifying methods for electrode production, sourcing suitable materials, and optimizing deposition techniques. TWI has made significant progress in these areas, involving the production and testing of electrode samples using various spray and electrodeposition methods. The tasks within WP4 are aimed at refining the production processes and improving the performance and durability of the electrodes.

### **T4.1: Electrode Production Process**

The report D4.1 was completed and shared with the consortium, detailing the materials specification, substrate preparation, spray methods, post-treatment procedures, and characterization plans. This report served as a foundational document guiding the subsequent experimental and process development activities in WP4.

### **T4.2: Spray Parameters for Electrodes**

#### *Material Sourcing and Initial Trials*

TWI received three copper powders with purity >99.5% for cold spray studies: dendritic copper powder (<53 $\mu$ m), fine spherical powder (15-38 $\mu$ m), and coarse spherical powder (15-53 $\mu$ m). Additionally, copper wire (1.6mm diameter, 99.9% purity) was sourced for twin wire arc spray.

Initial trials for arc spraying, flame spraying, low-pressure cold spraying, and high-pressure cold spraying were conducted. Parameters such as spray distance, voltage, and air pressure were varied, and samples underwent XRD, metallography examination, and oxygen level analysis.

#### *Observations and Adjustments*

Arc sprayed samples showed promising results on both copper and steel substrates in the first round.

Low-pressure cold spray samples experienced overheating and delamination, prompting parameter adjustments for subsequent trials.

High-pressure cold spray showed good results on copper substrates but significant delamination on steel, leading to substrate surface preparation optimization.

### **T4.3: Optimization of Electrodeposition Parameters**

Electrodeposition experiments were carried out using constant voltage and current with varying concentrations of copper salts at different time intervals. XRD characterization revealed electrodeposited copper.

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Two sets of experiments, designed using DOE with cathode position as an additional factor, were performed to optimize the process conditions for producing copper with high [1 0 0] peak intensity.

### *Results and Further Testing*

Two optimized conditions for high (1 0 0) facets were identified and detailed in deliverable D4.3.

Trials on sprayed copper foam substrates and bare copper foams were conducted, with samples sent to INL for further testing.

Durability tests in four different amine-based phase-changing solvents are in progress, with initial results showing promising electrochemical and corrosion properties.

#### **T4.4: Durability and Down-Selection of Electrodes**

Optimized conditions from T4.3 were used to deposit copper on copper foam substrates for CO<sub>2</sub> conversion efficiency testing at INL.

Durability tests in various amine-based solvents were performed with comparisons between Cu (1 0 0) and Cu (1 1 1) to understand the influence of crystal orientation on catalyst durability.

Initial results and further tests, including long exposure under reducing conditions, are detailed in deliverable D4.4.

#### **T4.5: Catalyst Production and Performance**

Early electrolysis testing indicated bulk substrates are unsuitable for catalytic performance, leading to trials with copper foam substrates.

Multi-coating deposition trials on copper foam substrates using various spray processes showed significant improvement in electrode selectivity, achieving up to 80% CO<sub>2</sub> conversion in the electrochemical cell.

Further optimization of coating methods and materials was carried out, with frequent discussions between TWI, ULEIC, and INL to review testing results and plan improvements.

### *Key Findings*

- **Spray Parameters:** Adjustments based on initial trials have improved coating quality and adhesion. Further optimization is needed for high-pressure cold spray on steel substrates.
- **Electrodeposition:** Optimized conditions for producing high (1 0 0) facet copper have been identified. Durability testing was performed to ensure long-term stability in various solvents.

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- Catalytic Performance: Copper foam substrates show high CO<sub>2</sub> conversion efficiency, and ongoing efforts aim to enhance performance through refined coating techniques and materials.

A brief summary of the achievements and outcomes along with conclusions is presented below.

### **Achievements:**

- Selected materials and suitable processes for development of catalyst
- Defined characterization strategy and production parameter optimization plan for electrode production
- DoE trials were carried out and well-adhered coatings were obtained using both thermal and spray processes.
- Physical characterization revealed a dominant [1 0 0] facet with electrodeposited copper on mild steel substrate.

### **Outcomes:**

- Copper wires and powders were selected by reviewing their availability from the market and suitability of being used in each spray process.
- Characterisation strategy was defined to efficiently evaluate microstructure and property of developed catalyst in multi-rounds of DoE trials.
- Both arc and flame sprayed copper coatings adhered well onto both steel and copper substrates and different levels of oxidization were observed.
- Low pressure and high pressure sprayed copper coatings experienced different level of delamination when depositing onto steel substrate but they adhere well with copper substrate. This has been successfully improved in round 2 by optimizing steel surface preparation.
- Electrodeposition of copper was performed based on the downselection of precursor candidates and experimental study with different operating conditions (mode of deposition, concentration, current density/voltage, substrates (mild steel, mild steel mesh), metal precursors.
- Electrodeposition was also performed in the selected current density range with varying anode to cathode distance.
- X-ray diffraction data revealed a dominant [1 0 0] facet for the electrodeposited copper on mild steel substrates and require a further study to understand the nature of the deposited copper in relation to its performance

### **Conclusion**

WP4 has made significant progress in developing effective electrode materials and deposition techniques. Through systematic experimentation and optimization, TWI has identified promising methods and materials for producing durable and efficient electrodes. Tests and

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refinements were carried out to enhance the performance of these electrodes, contributing to the overall success of the project.

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## WP5: Selection of gas separation systems – INL

The goal of WP5 was to develop effective membrane technologies for the separation of ethylene from a complex gas mixture resulting from the electrochemical conversion of CO<sub>2</sub> (EC- CO<sub>2</sub>). This work package included literature searches, technology analysis, experimental testing of membrane materials, and the development of facilitated transport membranes (FTMs) for ethylene separation. The work is divided into two main sub-sections: analysis of technologies for gas separation (Task 5.1) and the development of an electrolyzer product extraction and separation system (Task 5.2-5.4).

### **Task 5.1: Analysis of Technologies for Gas Separation**

#### *Literature Search and Initial Testing*

A comprehensive literature search was conducted to identify suitable membrane materials for the separation of ethylene from by-products such as water, hydrogen, unreacted CO<sub>2</sub>, and other heavy gases. Additionally, research on facilitated transport membrane (FTM) materials was performed. Initial gas permeation analyses were performed using various polymer membranes on gas mixtures containing hydrogen, carbon dioxide, methane, and ethylene. The setup for gas permeation instrumentation is complete, and these analyses aim to provide data on ethylene recovery.

#### *Development and Testing of PDMS Silver FTMs*

Preliminary tests were conducted on a novel polydimethylsiloxane (PDMS) silver facilitated transport membrane (FTM). Mixed-gas permeability tests were carried out on a simulated ethylene stream containing 2 vol% of nitrogen (N<sub>2</sub>), carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>), ethylene (C<sub>2</sub>H<sub>4</sub>), and ethane (C<sub>2</sub>H<sub>6</sub>) in a helium (He) balance. Notably, these mixed-gas experiments with an Ag-FTM have not been extensively studied in literature.

Results showed that the PDMS/Ag FTM displayed a selectivity ratio of 11.4 for ethylene over ethane over a 24-hour period in the mixed-gas feed stream, maintaining its performance without significant degradation. This is particularly promising given that polymer membranes generally do not exhibit high selectivity for ethylene over ethane, with the exception of TPX 80, which has lower gas permeabilities.

#### *Evaluation of Membrane Performance*

Further testing included the exposure of the PDMS/Ag FTM to different gas mixtures containing higher concentrations of CO<sub>2</sub> and CH<sub>4</sub>, along with ethylene. The Ag-FTM maintained a high selectivity ratio (~12 for ethylene over ethane and methane) over 30 days, even under exposure to problematic gases like CO<sub>2</sub>, CO, and CH<sub>4</sub>. This indicates the membrane's robust performance and suitability for ethylene recovery.

In addition to evaluating the effect of hydrogen exposure on the Ag-FTM, preliminary results suggest minimal changes in ethylene production after 24 hours of hydrogen exposure. This indicates potential for direct application in the electrolyzer feed stream.

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## **Tasks 5.2-5.4: Electrolyzer Product selection, extraction and separation system analysis, integration**

### *Experimental Setup and Initial Findings*

The tasks focused on analysing the product feed stream from the electrolyser to determine the best separation configuration for ethylene recovery. The remaining gases after initial separations, such as methane, CO, and ethylene, are targeted for further enrichment. Initial experiments used a 1:1 ethylene/methane-ethane composition to evaluate the FTM's performance.

### *Development of New Membranes*

Novel FTMs were fabricated and analysed for their suitability in separating the electrolyser gas feed stream. These membranes were characterized using mechanical, permeability, and optical techniques to ensure durability and efficiency. The goal was to achieve high olefin permeability and ease of blending in silver solutions.

### *Long-term Testing and Stability*

Long-term exposure tests were conducted with the novel PDMS/Ag FTM to simulate the gas stream from the electrolyser, particularly focusing on hydrogen, a reductive gas that can affect the active Ag<sup>+</sup> facilitator. Preliminary results showed that the Ag FTM maintained high selectivity ratios for ethylene over methane and hydrogen after exposure to a high concentration hydrogen atmosphere for seven days.

The Ag FTM demonstrated excellent gas permeabilities (>100 GPU) for ethylene and maintained selectivity even after prolonged exposure to problematic gases. This suggests that the FTM can effectively separate ethylene for extended periods, meeting the milestone of achieving ethylene selectivities close to 90% over ethane.

A brief summary of the achievements and outcomes along with conclusions is presented below

### **Achievements:**

- Fabricated a silver facilitated transport membrane (Ag-FTM) that is selective for ethylene
- Gas Permeability testing of the Ag FTM with various ratios of gas-mixtures containing ethylene and carbon dioxide, which shows long term stability and good ethylene production.
- Completed the report on the gas separation technologies for ethylene recovery

### **Outcomes:**

- The Ag FTM can enrich ethylene from a mixed gas feed stream that contains CO, CO<sub>2</sub> and methane.

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- The Ag FTM shows good selectivity for ethylene, even at low ethylene production (~2 vol%).

## **Conclusion**

WP5 has successfully developed and tested a novel PDMS silver facilitated transport membrane (FTM) capable of selectively separating ethylene from a complex gas mixture. The membrane demonstrated high selectivity and permeability for ethylene over extended periods, even under exposure to problematic gases. These promising results pave the way for further optimization and potential direct application in the electrolyzer feed stream, contributing significantly to the project's objectives in ethylene recovery and separation.

## **Future Work**

Future work will focus on testing the actual ethylene product feed stream from the electrolyzer, including further investigations into the effects of hydrogen and other chemical compounds on the FTM's longevity and performance. The goal is to stabilize the FTM under these conditions for short durations, allowing direct exposure from the electrolyzer product feed stream without additional separations.

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## **WP6: Electrolyser Integration and Optimisation (INL)**

The primary objective of WP6 is to develop and optimize an intensified electrochemical system for the conversion of captured CO<sub>2</sub> into value-added products such as ethylene. The activities conducted within this work package encompass the setup and validation of the electrochemical system, testing and optimization of various electrode materials and capture solutions, and the evaluation of system performance under different operational conditions.

### **Task 6.1- 6.3: Initial Setup, testing and validation**

In the first quarter, the electrochemical system setup was completed, and initial hydrodynamic and electrochemical tests were performed using capture formulations advised by CERTH partners. The chosen capture media demonstrated suitable viscosity and density characteristics, facilitating smooth flow through the system.

#### *Hydrodynamic Tests*

Preliminary hydrodynamic tests utilized a copper foam cathode with a current density of 100 mA/cm<sup>2</sup>. The electrochemical system setup, where cathode pressures of up to 80 psig were tested were reported in the deliverables (D6.1 & D6.2). Gas samples confirmed the presence of CO<sub>2</sub> and hydrogen, with qualitative and quantitative analyses.

#### *Electrochemical System Analysis*

The electrochemical system was tested with an amine capture solution proposed by WP2. Initial issues with gas analysis were resolved using a validated method, although efforts to enhance the gas analysis turnover rate continue. A 10 cm<sup>2</sup> single-pass electrochemical cell was integrated into the system, with preliminary tests conducted using a Cu powder-coated RVC electrode.

#### *Ethylene Production Tests*

Experiments for ethylene production employed a 2M KHCO<sub>3</sub> solution as the catholyte, fed at 1.4 mL/min. The electrochemical cell featured a two-compartment flow-through design with a bipolar membrane separating the anode and cathode. The cathode was a Cu powder-coated RVC electrode, while the anode used a Ni foam electrode with 1 M KOH anolyte. Figure 3 presents the conversion and ethylene selectivity results at 100 and 200 mA/cm<sup>2</sup>, indicating that higher current densities enhance ethylene selectivity at the cost of CO<sub>2</sub> conversion efficiency.

#### *Electric Conductivity Measurements*

CERTH measured the electric conductivity of various solvent mixtures, finding that higher KOH concentrations in MCA solutions increased conductivity. CO<sub>2</sub> partial pressure did not significantly affect conductivity, a beneficial attribute for fluctuating CO<sub>2</sub> feed applications. Figure 3 details these findings.

#### *Amine-Based Formulations Evaluation*

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INL evaluated six amine-based formulations against CO<sub>2</sub>-saturated 1 M KOH. Conductivity decreased significantly upon CO<sub>2</sub> saturation, highlighting the need for additional supporting electrolytes like KCl, which doubled the conductivity. Further tests with amine formulations involving piperazine and 2-Amino-2-methyl-1-propanol were performed. Evaluations of TWI-provided electrodes revealed carbon layer formation instead of ethylene production, likely due to insufficient active surface area. TWI produced new porous electrodes for further testing at INL. Continued testing of amine capture solutions against benchmark conversions using Ag-painted carbon electrodes yielded results. The performance with 0.5 M MCA was comparable to 0.5 M KHCO<sub>3</sub>, but higher MCA concentrations reduced CO<sub>2</sub> conversion efficiency.

#### **Task 6.4: Optimization of Operating Conditions**

Efforts to optimize operating conditions involved down-selecting electrode synthesis methods for maximum ethylene production. Eight electrodes combining flame and arc spraying with different Cu sources were tested, with electrode FF1 showing the highest performance for ethylene production. The FA2 electrode exhibited notable activity towards CO, suggesting potential for further research.

A brief summary of the collaborative efforts, achievements and outcomes along with conclusions is presented below

##### *Collaborative Efforts:*

Collaborations with WP4 facilitated the advancement of deliverable 4.4 and progress on task 6.3 and deliverable 6.4. High-throughput synthesis methods (electrodeposition, flame spray, and arc spray) yielded electrodes with varied catalytic activities. AS electrodes were 100% selective towards CO, while FS electrodes showed higher selectivity for ethylene, albeit with lower stability over extended operation periods.

##### **Achievements:**

- A flow through electrochemical system with an effective area of 10 cm<sup>2</sup> was assembled and hydrodynamically tested to be fed by CO<sub>2</sub> captured solutions, allow gas separation, and on-line analysis of the gas products.
- Electrochemical system has been benchmark using CO production to verify successful conversion of captured CO<sub>2</sub>. Current efficiencies as high as 40% were obtained with CO<sub>2</sub> conversions over 80%.
- Preliminary results have demonstrated formation of ethylene with carbon selectivity as high as 70% and CO<sub>2</sub> conversions ranging from 40-70% in a single pass

##### **Outcomes:**

- Measured the electrical conductivity of the selected solvent and its mixtures and indicated that the selected solvents already exhibit high ionic conductivity, so it is suitable for use in the electrochemical cell and that the addition of the other solvents further enhanced the electrical conductivity.

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- Production of ethylene from captured CO<sub>2</sub> has been verified. Challenges addressed on electrode architecture and electrocatalytic activity have been identified leading to collaborative discussions among the involved work packages.

### **Conclusion and Summary:**

WP6 successfully established an intensified electrochemical system, conducted preliminary hydrodynamic and electrochemical tests, and evaluated various electrode materials and capture solutions. Initial results demonstrated promising ethylene selectivity and CO<sub>2</sub> conversion, with optimization of operational parameters and electrode synthesis methods. Future efforts will focus on enhancing system performance and stability to achieve project targets.

### **Future direction:**

Future tasks include testing new porous electrodes from TWI, optimizing operational parameters (e.g., back pressure, flow rate, current density), and further evaluating the performance of amine capture solutions in the electrochemical cell. These efforts aim to enhance the efficiency and selectivity of CO<sub>2</sub> conversion to value-added products, contributing to the clean energy sector.

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## WP7: Carbon Footprint and Sustainability Assessments – TVS

### **Task 7.1: Life Cycle Assessment (LCA)**

During the period from 1st February to 30th April 2022, Task 7.1 commenced with defining the functional unit as 1 kg of ethylene production from a CO<sub>2</sub> -rich gas stream, adopting a cradle-to-gate system boundary. The required mass and energy data flows were derived from outcomes of WP2-WP6.

From 1st April to 30th June 2022, literature reviews on the environmental impacts of both thermochemical CO<sub>2</sub> utilization and conventional fossil-fuel-based ethylene production were conducted. Spreadsheets for data collection from WP2-WP6 were prepared, and the life cycle inventories for the CO<sub>2</sub> capture and delivery unit were developed using mass balance calculations and process unit designs based on quicklime plant flue gas.

Data for the carbon capture, electrochemical conversion, and gas separation units were inventoried. Using the LCA methodology IMPACT World+ Midpoint version 1.03 and SimaPro 9.5.0.0 software, the environmental footprints of 1 kg of ethylene production were evaluated across 18 midpoint impact categories, including climate change and human toxicity.

### **Task 7.2: Life Cycle Cost (LCC) Analysis**

An initial framework for Life Cycle Cost (LCC) analysis was developed, identifying cost parameters for the techno-economic assessment of CoCaCO<sub>2</sub>la processes. Costs for conventional ethylene production methods were reviewed, and a detailed cost analysis was initiated for the integrated process units.

The study estimated equipment costs and performed a cost-effective analysis for the European context. CERTH provided a detailed techno-economic assessment of CO<sub>2</sub> capture using MEA and MCA solvents. The capture costs were determined to be 31 €/tn for MEA and 23 €/tn for MCA, with significant contributions from blower costs for flue gas delivery.

### **Task 7.3 : Social Impact Analysis**

A questionnaire for social impact analysis was developed based on literature studies. The survey was made available on TVS's citizen engagement portal, "Citizen tone," since May 2023, and data collection was performed with 36 completed and 22 partial responses.

CERTH conducted a detailed techno-economic assessment of alternative CO<sub>2</sub> capture and utilization configurations using MEA and MCA solvents. The assessment evaluated direct and indirect configurations for CO<sub>2</sub> reduction in electrolysis cells. The results showed a 9.7% average reduction in ethylene production costs compared to the base case, with MCA being more cost-effective than MEA.

### **Overall Summary**

WP7 focused on comprehensive assessments of the carbon footprint, life cycle costs, and social impacts of ethylene production through CO<sub>2</sub> electroreduction. Initial LCA and LCC

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frameworks were established, and data collection from WP2-WP6 was initiated. Preliminary results highlighted potential cost savings and environmental benefits of the integrated CO<sub>2</sub> capture and utilization process. Work performed includes optimizing process parameters and finalizing social impact assessments to ensure the sustainability and economic feasibility of the CoCa CO<sub>2</sub> la project.

**Achievements:**

- LCA and LCC frameworks have been defined for CoCa CO<sub>2</sub> la project.
- Data inventories of three units are being carried out for evaluating environmental impacts.
- An online software tool has been developed for conducting social impact analysis.

**Outcomes:**

- The environmental footprints of the ethylene product derived from CO<sub>2</sub> -rich gas stream.
- Comparing the environmental impact results of the ethylene production for the CoCa CO<sub>2</sub> la project processes with the conventional processes.
- An excel template has been created as a techno-economic assessment tool.
- Capital investment and total product cost estimation are being prepared for the ethylene production from secondary literature in context of European region.
- A survey results of social impact analysis

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## WP8 : Dissemination, Engagement and Exploitation Planning - TWI

### **Task 8.1: Advisory Committee Formation and Coordination**

Progress has been made in forming the advisory committee, with ULEIC working alongside TWI to finalize the required paperwork for formal agreement. An advisory committee consisting of two advisors has been established. Regular monthly coordination and management calls with all consortium members are being conducted, facilitated by a sharefile set up by TWI for central project information storage.

A representative from TWI attended the CCUS conference in Rotterdam to present the project, furthering dissemination efforts. A press release about the project was published on the TWI website, and a webinar on decarbonization and carbon management is planned for 1st December 2022.

### **Publications:**

- Penot et al. (2024) - Electrochemical Characterization of Electrodeposited Copper in Amine CO<sub>2</sub> Capture Media, Materials (MDPI)
- Kyriakides et al. (2023) "Modeling and Assessment of Electrochemical Reduction of CO<sub>2</sub> in Amine-based Capture Solvents" - 33rd European Symposium on Computer-aided Process Engineering.
- Kyriakides et al. (2023) "Optimal design of Electrochemical Reduction of CO<sub>2</sub> in Amine-based Capture Solvents" - 26th Conference Process Integration, Modelling and Optimisation for Energy Saving and Pollution Reduction.
- Tzirakis et al. (2023) "CO<sub>2</sub> Solubility in Aqueous Solutions used in CO<sub>2</sub> Capture Applications" - 26th Conference Process Integration, Modelling and Optimisation for Energy Saving and Pollution Reduction.
- K K Maniam et al. (2023)- "Progress in Electrodeposited Copper Catalysts for CO<sub>2</sub> Conversion to Valuable Products", Processes (MDPI)

### **Task 8.2: Business Planning**

Work on the business model canvas and value proposition canvas was performed, with a focus on components such as customer segments, value proposition, channels/marketing, customer relationships, revenue streams, key resources, key activities, key partnerships, and cost structure. The ethylene market size, stakeholder analysis, SWOT analysis, and value proposition are being developed. The LCC report will further enhance the understanding of cost structure and potential revenue.

### **Task 8.4: Dissemination Activities**

The dissemination efforts include the presentation of the paper "Life Cycle Assessment studies of ethylene production through the electroreduction of captured CO<sub>2</sub> from a quicklime plant" at the 26th Conference of Process Integration, Modelling and Optimisation for Energy Saving and

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Pollution Reduction in Thessaloniki, Greece. This paper has been accepted for publication in the peer-reviewed journal Cleaner Engineering and Technology.

John Klaehn presented “Facilitated transport membranes for small scale ethylene production” at the ACS Spring 2023 National Meeting, highlighting work by INL researchers on novel facilitated transport membranes (FTMs) designed for electrocatalytic conversions to ethylene.

Corentin Penot presented “Performance evaluation of copper catalysts for CCUs” during EUROCORR’23 at Brussels, Belgium

### **Task 8.5: Impact of CO<sub>2</sub> CO<sub>2</sub> Gas Impurities on Electrolyser Materials**

TWI designed, built, and commissioned a bespoke testing system for performance and compatibility assessment of materials in carbon capture and storage (CCS) environments. This dynamic system allows for continuous injection of impurities like water and H<sub>2</sub>S into a CO<sub>2</sub> stream to test material durability and performance. These tests simulate the challenging conditions encountered in CCUS operations, focusing on the potential risks of corrosion and cracking in electrolysers due to impurities.

The testing system enables monitoring and control of impurity concentrations within the test cell, providing insights into the long-term performance of electrolyser materials. This approach supports the optimization of electrolyser design and material selection, enhancing the overall efficacy and sustainability of CCUS systems.

### **Conclusion and overall summary**

WP8 has made significant developments, activities in the formation of advisory committees, dissemination of project information, and business planning. Regular coordination among consortium members ensures effective project management. Dissemination activities include conference presentations, publications, and a planned webinar, while business planning efforts focus on developing a comprehensive business model canvas and competitive analysis. The establishment of a bespoke testing system for evaluating the impact of CO<sub>2</sub> impurities on electrolyser materials demonstrates TWI's commitment to optimizing CCUS technologies, ensuring their long-term viability and performance.

## Project impact

**Table 1 – Project impact based on WPs**

WP	Key expected result	Impact	Status
2	a) Select appropriate solvent, (b) derive experimental data and demonstrate its suitability for this application, (c) demonstrate scale-up potential using process models	Transform conventional carbon capture systems into ones that directly transform the captured CO <sub>2</sub> into value-added products, contribute to development of net-zero industrial systems	Expected results (a) and (b) have been attained. In the process of performing the simulations to attain result (c). The impacts in terms of the overall process will be quantified in the end of the project based on the design characteristics (e.g. equipment capacity, CO <sub>2</sub> captured, product yield etc.) that will be attained.
3	Enhance the electrochemical reduction of CO <sub>2</sub> into valuable C <sub>2</sub> -based chemicals like ethylene, thereby promoting the use of carbon capture, utilization, and storage (CCUS) technologies.	By developing advanced copper catalysts and optimizing electrodeposition techniques, the project contributes significantly to the emergence of CCUS. The project's findings will aid in designing more efficient and durable catalysts, crucial for scaling up CCUS technologies. This progress supports the reduction of industrial carbon emissions and aligns with global efforts to mitigate climate change.	The project has actively progressed through catalyst material selection, development, and optimization phases, ensuring robust groundwork for future CCUS applications focusing not only on ethylene but also syn gas
4	specification and characterization of materials and processes for electrode production, optimization of spray parameters for various spraying techniques, systematic	The development of optimized copper electrodes for CO <sub>2</sub> conversion contributes significantly to the advancement of Carbon Capture, Utilization, and Storage (CCUS) technologies. Efficient CO <sub>2</sub> conversion to	Optimisation techniques and characterization of catalysts were refined to adapt to the commercial readiness. Continued progress in this area

	electrodeposition of copper to identify optimal performance parameters, and durability assessment and enhancement of electrode materials for CO2 conversion	valuable chemicals like ethylene and ethane enhances the viability and attractiveness of CCUS in mitigating climate change.	will further increase the chances of successful commercialization.
5	Identify suitable membrane materials and conduct initial testing on mixed gas streams.	Completed literature search and preliminary gas permeation analyses. Developed a testing protocol and summary of the literature, paving the way for effective ethylene and syngas recovery technologies.	Testing was adapted and performed with catalysts coated by different methods, achieved ethylene and syngas separations effectively.
6	(a) Establish an electrochemical system and conduct preliminary tests using various electrode materials and capture solutions, (b) Optimize operating parameters and electrode synthesis methods to enhance ethylene production and CO2 conversion efficiency	Completed the setup and initial testing, achieving successful conversion of CO2 and demonstrating high selectivity for ethylene production.  Achieved significant progress with electrodes showing high ethylene selectivity and optimization efforts aimed at further improving system performance and stability.	Completed the tests with catalysts coated by different techniques
7	a) LCA analysis b) LCC techno-economic assessment c) social impact	To derive environmental, techno economic and social acceptance evaluation of the concept.	LCA and LCC frameworks have been defined for CoCaCO2Ia project.  An online software tool has been developed for conducting social impact analysis.

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## Implementation

Describe the implementation of the project results in relation to the SET plan Implementation Actions (no 9 on CCUS), Mission innovation research priorities and how you have engaged industry in your work.

The CoCaCO<sub>2</sub>la project has made significant progress in aligning its research with the SET plan Implementation Actions No. 9 on Carbon Capture, Utilization, and Storage (CCUS) and Mission Innovation research priorities as outlined below.

The project's Work Package 1 (WP1) focused on technology gap analysis and key performance indicator (KPI) definition, addressing innovative approaches to CO<sub>2</sub> capture and conversion. This included exploring capture chemistries such as aqueous tertiary amine solutions, ionic liquids, covalent organic frameworks, and phase change solvents (PCS). Major technology gaps were identified, including the unreported ethylene production from CO<sub>2</sub> captured in PCSs and the need for optimized hydrodynamic conditions for CO<sub>2</sub> electrochemical reduction. WP1 also explored electrochemical cell configurations, proposing buffer layer and zero-gap configurations to enhance CO<sub>2</sub> reduction efficiency and identifying the need for scalable electrocatalysts and optimized membrane types.

WP2 assessed and selected phase-change solvents for CO<sub>2</sub> capture, identifying MCA mixed with KOH as a promising solution due to its high CO<sub>2</sub> loading and conductivity. Techno-economic analyses highlighted the importance of electricity prices and Faradaic efficiency on ethylene production costs. WP3 optimized catalyst materials for CO<sub>2</sub> reduction, with the University of Leicester leading efforts in copper electrodeposition techniques. WP4, led by TWI, focused on electrode production processes, identifying spray parameters and developing high CO<sub>2</sub> conversion efficiency electrodes.

WP6 developed and optimized an electrochemical system for converting captured CO<sub>2</sub> into value-added products like ethylene. Initial setup, testing, and validation involved hydrodynamic tests using copper foam cathodes and optimizing various operational conditions. The system showed promising results with ethylene selectivity and CO<sub>2</sub> conversion efficiencies, emphasizing the need for ongoing optimization. Future tasks include testing new porous electrodes, optimizing parameters like back pressure and flow rate, and evaluating amine capture solutions.

WP7 focused on comprehensive assessments of the carbon footprint, life cycle costs, and social impacts of ethylene production through CO<sub>2</sub> electroreduction. An initial Life Cycle Assessment (LCA) framework was established, defining the functional unit as 1 kg of ethylene production from a CO<sub>2</sub>-rich gas stream and employing a cradle-to-gate system boundary. Data from WP2-WP6 informed mass and energy flows, and environmental impacts were assessed using the IMPACT World+ Midpoint methodology and SimaPro software. An initial framework for Life Cycle Cost (LCC) analysis was developed, identifying cost parameters for the techno-economic assessment of CoCaCO<sub>2</sub>la processes, and highlighting the cost-effectiveness of MCA over MEA solvents. Social Impact Analysis was initiated, developing a questionnaire based on literature studies and collecting responses via TVS's citizen engagement portal.

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WP8 focused on dissemination, engagement, and business planning. An advisory committee was formed, and dissemination activities included conference presentations, publications, and a planned webinar. Business planning efforts involved developing a comprehensive business model canvas and competitive analysis. A bespoke testing system was established to evaluate the impact of CO<sub>2</sub> impurities on electrolyser materials, optimizing CCUS technologies for long-term viability.

Throughout the project, industry engagement has been pivotal. Collaboration with industrial partners ensured the relevance and applicability of research findings (through dissemination activities), facilitating the translation of laboratory results into scalable industrial solutions. By addressing key technology gaps and optimizing CO<sub>2</sub> capture and conversion processes, the CoCaCO<sub>2</sub>la project significantly advances CCUS technologies, aligning with the SET plan and Mission Innovation priorities for a sustainable carbon economy.

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## Collaboration and coordination within the Consortium

Describe the collaboration and coordination within the Consortium. Comment on the effectiveness of management structures and governance procedures. Add a special focus on the added value of trans-national collaboration on CCUS.

The CoCaCO<sub>2</sub>1a project, focussed on converting captured CO<sub>2</sub> into valuable products like ethylene, exemplifies the value of trans-national collaboration in the field of carbon capture, utilization, and storage (CCUS). Through a strategic alliance of partners with specialized expertise, the consortium effectively addressed critical technology gaps, including the optimization of CO<sub>2</sub> reduction conditions, development of scalable electrocatalysts, and enhancement of purification technologies. Key performance indicators (KPIs) were established to measure progress in areas such as CO<sub>2</sub> capture efficiency, energy efficiency, scalability, and product purity. Significant advancements were made, including the validation of an electrochemical cell model, the development of durable copper catalysts, and the demonstration of an effective silver facilitated transport membrane (Ag FTM) for ethylene separation. The project's management structure, coordinated by TWI (UK), facilitated efficient collaboration and resource allocation, supported by monthly coordination calls and centralized information storage.

Partners such as Idaho National Laboratory (INL), the University of Leicester (ULEIC), and CERTH (Greece) brought their unique capabilities to the table, from designing CO<sub>2</sub> electrolyzers, catalysts to developing new CO<sub>2</sub> capture solvents. Life Cycle Assessment (LCA) and Life Cycle Costing (LCC) conducted by TVS (UK) highlighted the project's environmental and economic benefits, highlighting a significant reduction in climate change impact compared to traditional processes. Dissemination efforts included conference presentations, publications, and planned webinars, ensuring the project's findings reached a broad audience.

The advisory committee, comprising expert from INL played a crucial role in guiding the project's progress, supported by regular updates and strategic planning sessions. The development of a bespoke testing system by TWI for evaluating material performance in carbon capture environments further demonstrated the consortium's commitment to overcoming operational challenges in CCUS technologies.

Overall, the collaborative efforts within the CoCaCO<sub>2</sub>1a consortium have yielded innovative solutions and substantial progress in CO<sub>2</sub> conversion technologies, showcasing the added value of trans-national collaboration in advancing sustainable and efficient industrial processes.

## Dissemination activities (including list of publications)

### Publications

- Penot et al. (2024) - Electrochemical Characterization of Electrodeposited Copper in Amine CO<sub>2</sub> Capture Media, Materials (MDPI)

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- Kyriakides et al. (2023) “Modeling and Assessment of Electrochemical Reduction of CO<sub>2</sub> in Amine-based Capture Solvents” - 33rd European Symposium on Computer-aided Process Engineering.
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  - Tzirakis et al. (2023) “CO<sub>2</sub> Solubility in Aqueous Solutions used in CO<sub>2</sub> Capture Applications” - 26th Conference Process Integration, Modelling and Optimisation for Energy Saving and Pollution Reduction.
  - K K Maniam et al. (2023)- “Progress in Electrodeposited Copper Catalysts for CO<sub>2</sub> Conversion to Valuable Products”, Processes (MDPI)

### Conference Presentations

- John Klaehn presented “Facilitated transport membranes for small scale ethylene production” at the ACS Spring 2023 National Meeting, highlighting work by INL researchers on novel facilitated transport membranes (FTMs) designed for electrocatalytic conversions to ethylene.
- Corentin Penot presented “Performance evaluation of copper catalysts for CCUs” during EUROCORR’23 at Brussels, Belgium

### Webinar:

<https://www.youtube.com/watch?v=bIKW7C8T2T0>

### Press release on TWI website:

- <https://www.twi-global.com/media-and-events/press-releases/2024/cocaco2la-carbon-capture-and-conversion-for-net-zero-goals>
- <https://www.twi-global.com/media-and-events/press-releases/2023/twi-expert-presenting-at-act-workshop-in-paris>
- <https://www.twi-global.com/media-and-events/press-releases/2023/cocaco2la-project-details-shared-at-act-workshop>
- <https://www.twi-global.com/media-and-events/press-releases/2023/cocaco2la-project-paper-published>
- <https://www.twi-global.com/media-and-events/press-releases/2022/cocaco2la-webinar-to-discuss-carbon-management-for-net-zero>
- <https://www.twi-global.com/media-and-events/press-releases/2022/cocaco2la-project-represented-at-ccus-conference-rotterdam>
- <https://www.twi-global.com/media-and-events/press-releases/2022/cocaco2la-project-converting-co2-to-ethylene>

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