

EERA AMPEA Workshop

Carbon Capture Storage & Utilization

March 10 - 11, 2021

Zoom Platform

Agenda

Day 1

Time	Topic	Presenter	Organization
09:00	Welcome and Steering Committee Meeting (separate agenda)	JPC S. Nakamae JPSC members	
Session JP CCS (Chair: M.L. Fontaine)			
09:30	Invited JP CCS - I CCUS – Recent Topics with Regard to Transport and Storage	R. Span J. Pearce	RUB – DE BGS - UK
10:00	Invited JP CCS - II From Clean Combustion to CCU: intriguing topics for process engineering and materials scientists	O. Senneca	CNR -IT
10:30	Q&A session		
10:45	Coffee break		
Session JP AMPEA I : Carbon Capture and Utilisation (Chair: R. Kiebach)			
10:55	Invited JP AMPEA Intensified catalytic conversion of CO ₂ into added-value hydrocarbons	J. Serra	ITQ CSIC -ES
11:15	Advanced materials for CCUS (at the University of Bologna): from nano-membranes to electrodes for CO ₂ reduction	M. Giacinti Baschetti G. Valenti	UniBo - IT
11:30	Efficient Combined CO ₂ Capture and Methanation	S. Cimino	CNR- IT
11:45	Thermochemical cycles for CO ₂ capture/utilization	G. Landi	CNR - IT
12:00	What would it take for renewable based electrosynthesis products to substitute those obtained from petrochemical processes?	J. Morante	IREC- ES
12:15	Q&A session		
12:30	End of first day		

Day2

09:00	Welcome	JPC S. Nakamae	CEA - FR
Session JP AMPEA II: Materials for Carbon Capture (Chair: A. Bieberle)			
09:10	Invited JP AMPEA Inorganic membranes for gas separation: opportunities and challenges	M.-L. Fontaine	SINTEF -NO
09:30	Materials and Processes for Solar Energy Based Capturing and Transforming of CO ₂ and Water into Hydrocarbon Feedstock	M. Roeb	DLR - DE
09:45	Ceramic gas separation membranes for the use in CCUS	W.A. Meulenbergh	FZ Jülich - DE
10:00	Current perspectives on advanced solid sorbents for CCS: the case of hybrid metal organic framework	M. Alfé	CNR-IT
10:15	Materials selection for the CHEERS project on Chemical Looping Combustion	A. Lambert	IPFEN - FR
10:30	Q&A session		
10:45	Coffee break		
Session JP AMPEA III: Carbon Conversion and Utilisation (Chair: J. Morante)			
10:55 Session 3	Invited JP AMPEA Carbon Capture Utilisation: some CEA on-going works and achievements	F. Ducros	CEA - FR
11:15	Invited JP AMPEA KEROGREEN - A plasma-based approach for CO ₂ neutral fuel production	S. Welzel	DIFFER - NL
11:35	Engineering photosynthetic microorganisms for direct solar chemical and fuel production from carbon dioxide	P. Lindblad	Uppsala - SE
11:50	Methanation of captured carbon dioxide in a solid oxide membrane reactor	E. Ruiz	CIEMAT- ES
12:05	Rational exploration of innovative materials by quantum simulation	P. Raybaud	IPFEN - FR
12:20	Electrocatalytic CO ₂ Reduction with Copper /Carbon Catalysts to C ₂ and C ₂₊ Value-added Products supported over Gas Diffusion Layers	S. Murcia	IREC - ES
12:35	Q&A session		
12:50	Conclusion of the workshop	JPC S. Nakamae	CEA - F

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CCUS – Recent Topics with Regard to Transport and Storage

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Abstract

Transport and storage are two essential steps in the CCUS, and in particular in the CCS process chain. For a long time, the focus of research has been mostly on the capture process itself, because the capture process has by far the highest energy demand in the process chain and because it dominates the additional investment and operating cost, if carbon capture from large power plants is considered. However, transport and storage determine requirements on processing of the captured CO₂, on allowable fluctuations in production rates, and finally on the potential of CCS technologies. With the increasing complexity of CCUS scenarios, it becomes obvious that there are still a number of questions that need to be answered for transport and storage.

Issues on Transport (R. Span)

Both pipeline and ship transport of CO₂ are in principal proven technology. Even at full commercial scale, there is little doubt that point to point pipeline transport of CO₂ can be realized in a safe and technically mature way. There are still a number of practical issues, like accurate flow metering according to fiscal terms and the availability of test and calibration facilities for full-scale flow meters, or the development of classifications / standards for large scale CO₂ tankers, but these issues may be considered engineering problems that can be solved based on existing knowledge.

However, the goal to develop industrial CO₂ transport networks capable to collect CO₂ from multiple industrial sources rather than from a single powerplant introduces a number of new challenges, whereby some of them involve open scientific questions. Monitoring and control of CO₂ transport networks with multiple users feeding CO₂ into the network, and potentially also extracting CO₂ from the network for CCU, requires detailed knowledge on all processes in the network, including online information about phase distributions and tracking of impurities. Mass-flow fluctuations in such networks also result in challenges to injection and storage.

Issues on Storage

The deep injection of CO₂ for permanent geological storage has been implemented in a number of projects worldwide, and builds on over three decades of experience of CO₂ injection for enhanced oil production, mainly within the USA. As Europe and some countries within it explore options to achieve net zero emissions, the storage of CO₂ is likely to be required on a significantly larger scale than previously expected. This CO₂ will be captured from industrial processes to support other decarbonisation options. Some gas-based electricity generation may also still be required to support wider renewable electricity generation. Where H₂ replaces natural gas in heating (either residential or industrial) then further CO₂ may be captured where the H₂ is produced through steam reforming of natural gas. In the future, so-called carbon-negative technologies may also be required including bioenergy with carbon capture and possibly direct air capture. All of these captured CO₂ streams will require a strategic transport and storage network, that must be able to accept the captured CO₂ at the rates supplied and with guaranteed storage available. The injection of these significant and potentially variable volumes requires understanding of both the availability of storage capacity and of the potential interactions and management of pressure responses at individual and between multiple storage sites. Furthermore, as CO₂ transport and storage moves towards more commercial operations, research is also needed to increase efficiencies and reduce costs, whilst providing robust evidence for the 'safety case' for each storage site. Research priorities include developing and improving our capabilities to remotely monitor the evolution of the CO₂ plume and demonstrating conformance between this observed site operation and predictions of future behaviour. In addition, all stakeholders require assurance that the risks, impacts and remediation options for a site failure that may lead to subsequent leakage are well understood.

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From Clean Combustion to CCU: intriguing topics for process engineering and materials scientists

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Abstract

Over the last decades a considerable research effort has been devoted worldwide to enable emission control and CO₂ capture from combustion of fossil fuels in power plants. Initially the main target was developing capture ready combustion technologies and combining them with economically sustainable systems of CO₂ capture and storage.

Nowadays researchers are confronting the more ambitious goal of integrating renewable energy, power generation from fossil fuels, CO₂ capture and utilization (CCU), energy storage, production of chemicals and materials. Within this renewed scenario, the concept of CO₂ as final product to be disposed of is being replaced by the vision that CO₂ can be used as carbon or energy vector within articulated and integrated process schemes.

In order to respond to this challenge it is necessary to combine expertise in many different sectors: chemical engineering, mechanical engineering, chemistry, biotechnology, IT, geology, social science, economics and last, but not least, materials science.

The present work examines a selection of present and future key technologies in the sector of clean combustion, CO₂ capture and utilization. Besides illustrating the state of the art of these technologies and how they contribute to the overall puzzle, the work will highlight the issues which might be of greatest interest for materials scientists, with the aim of identifying the possible synergies between the communities of CCU and materials science.

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Intensified catalytic conversion of CO₂ into added-value hydrocarbons

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Abstract

GHG emissions reduction policies to mitigate the alarming climate change can impact carbon-intensive industrial sectors, leading to loss of employment and competitiveness. Current multistage CCU technologies using renewable electricity to yield fuels suffer from low energy efficiency and require large CAPEX. Novel intensification process combines smart molecular catalysis and process intensification to bring out a novel efficient, flexible and scalable CCU technology, specifically, CO₂ conversion processes using renewable electricity and water steam to directly produce added-value hydrocarbons and synthetic fuels with balanced hydrocarbon distribution to meet the stringent specifications in transport, e.g. in aviation. The CO₂ reactors consists of a tailor-made multifunctional catalyst integrated with adsorbent, selective membranes and also electrochemical cells that enable to jointly realise electrolysis and/or water removal from hydrocarbon synthesis reaction. Intensified processes can potentially lead to breakthrough product yield and efficiency for chemical energy storage from electricity, specifically high CO₂ per-pass conversion and energy efficiency. In addition, these processes are generally compact, modular –quickly scalable- and flexible, thus, process operation and economics can be adjusted to renewable energy fluctuations. As a result, these technologies will enable to store more energy per processed CO₂ molecule and therefore to reduce GHG emissions per jet fuel tone produced from electricity at a substantial higher level.

References

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Advanced materials for CCUS: from nano-membranes to electrodes for CO₂ reduction

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Abstract

This talk will present research activities ongoing at the University of Bologna on materials and processes for CO₂ capture, storage, and (photo)-electrochemical conversion.

Gas separation membranes do represent a viable alternative for carbon capture and several solutions have reached promising results, even if further improvements are needed to make this approach fully competitive. The University of Bologna within the NANOMEMC2 project [1] focused on the development of innovative facilitated transport membranes able to overcome the limitation of usual polymeric membranes and to reach higher separation performances thanks to the presence of active CO₂ carriers within the membranes.

Geological CO₂ storage involves injection operations, which usually entail sudden changes in CO₂ pressure and temperature, and possible phase transitions that have severe effects on the integrity of wells. Our geothermal research group is involved in the improvements of the T2Well-ECO2M (LBNL, Berkeley, USA), a numerical simulator for non-isothermal, multiphase, multicomponent flows in integrated wellbore-reservoir systems that can handle CO₂ injection in depleted hydrocarbon reservoirs.

The CO₂ reduction reaction is a two-way strategy for both environmental remediation and carbon-neutral circular economy. However, from a circular economy perspective, the conversion of CO₂ to useful compounds for use in industrial processes or as fuels, is an important goal that may be pursued by developing (photo)-electrochemical reduction processes powered by green or renewable energy sources, for example, wind, tidal, solar, or the so-called “blue energy” systems. The feasibility of the reaction strongly depends on the performance of the catalysts used. Besides activity, especially at low overpotential, and stability, the catalyst's ability to produce a single compound with high efficiency and selectivity is a key issue. The University of Bologna conducts research on the development of innovative nanomaterials and catalysts to for electrochemical, photochemical [2], and photo-electrochemical [3] CO₂ conversion. The combination of different nanostructures is a unique strategy to achieve a good selectivity and performance in the CO₂ conversion process.

References

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- [2] G. Valenti et al, ACS Appl. Energy Mater. 3, 8509 (2020)
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Efficient Combined CO₂ Capture and Methanation

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Abstract

Current multistage CCU technologies using renewable electricity to yield fuels suffer from low energy efficiency and require large capital expenditures. To increase the efficiency of the whole process and reduce its cost, a possible innovative solution is represented by the combined capture and hydrogenation of CO₂. The idea is to trap CO₂ from a CO₂-containing stream (e.g. combustion flue gases) using a dual functioning material (DFM) composed of a highly dispersed supported adsorbent which provides at the same time the CO₂ storage function and methanation active element to produce CH₄ upon reaction with renewable H₂ [1-2]. This process is operated cyclically by alternating phases where the DFM is exposed to the flue gas and CO₂ is selectively adsorbed, and phases where H₂ is fed and the stored CO₂ is hydrogenated to methane [2]. Notably, by eliminating a thermal swing process, the conversion of CO₂ to SNG using DFMs constrains the energy input to only renewable sources (in the form of H₂), thus allowing the CO₂ capture and utilization processes to approach carbon neutrality while integrating more renewable energy into the grid [2].

In a highly innovative scheme aimed at the process intensification, the DFM represents a CO₂ carrier that is continuously circulated from a sorption reactor to a methanation reactor and vice versa [3]. Dual interconnected fluidized bed technology appears perfectly suited to perform the chemical looping CO₂ Capture and Methanation process since it allows the recirculation of DFM particles between two reactors and ensures their efficient and independent temperature control. Therefore, we set out to experimentally investigate the key features capable to boost the overall performance of DFMs based on Ru and alkali metal (oxides) supported on high surface area aluminas with suitable mechanical and attrition resistance to be used in interconnected fluidized bed reactors. The general scope is to produce synthetic methane via an innovative, renewable energy driven, catalytic looping process, demonstrating effectiveness and reduction of greenhouse gas emissions while also addressing economic, regulatory, environmental and (critical) raw material constraints, as well as socio-economic impact related to the proposed technological pathway.

References

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Thermochemical cycles for CO₂ capture/utilization

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Abstract

A thermochemical cycle (TC) is a process consisting of linked steps at varying reaction conditions; one or more materials are used, their composition is modified during the different steps, returning to their initial state at the end of the cycle. Accordingly, several processes fall in this definition such as, chemical looping combustion/gasification/reforming, water/carbon dioxide splitting, CO₂ capture and methanation, including those enabling capture storage and/or utilization. Moreover, due to their flexibility, TCs can be coupled with different energy sources, as clean energies such as solar, nuclear, and biomass resources [1]. TC reaction conditions significantly depend on the process and, thus, on the reactions occurring in each step [2, 3]. So, the successful development of processes based on TCs significantly depends on i) the development of suitable materials and ii) the development of highly performing reactors. From the “material” side, the main features generally required are high oxygen storage capacity, fast reduction/oxidation kinetics, low working temperatures, good thermal stability, good cyclability. From the “reactor” side, good heat and mass transfers are of paramount importance and, in processes using solar energy, good interaction with sun radiation must be guaranteed. From this point of view, fluidized bed reactors appear to best fit the requirements of thermochemical cycles.

We report our experience in the development of both materials and fluidized bed reactors for different chemical cycles. The general scope of activities related to materials is the design of catalysts/carriers based on different oxides (as perovskites, ceria, oxysulfates), whose features can be tuned with respect to the selected process. In addition to the features reported above, catalytic materials should also show good selectivity in presence of competitive reactions. The general scope of activities related to reactors is the development of interconnected fluidized bed configurations able to optimize multi-step thermochemical cycles. A novel two-stage fuel reactor for chemical looping combustion with oxygen uncoupling of solid fuels has been optimized to exploit at its best the reactive properties of the bed granular materials. Finally, a directly-irradiated fluidized bed autothermal reactor (DIFBAR) for concentrated solar thermal applications has been proposed and modelled. The very core of the reactor is a fluidized bed solar receiver/reactor coupled with a double pipe heat exchanger. The features of the DIFBAR reactor make it a valuable tool for solar driven thermochemical cycles.

References

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What would it take for renewable based electrosynthesis products to substitute those obtained from petrochemical processes?

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Abstract

Carbon dioxide reduction by electrochemical (dark and photo) procedures has become one of the most relevant challenges for the energy transition of our society, being the energy efficiency of this process its clue for assuring a clear industrial feasibility. Nevertheless, our society is raising an essential question: What would it take for renewably based electrosynthesis products to substitute those obtained from petrochemical processes?

In this contribution, the role of catalysts and reactor design will be reviewed with special attention paid in overpotential values and selectivity and productivity towards determined sub product such as CO, syngas, formic acid, methanol, or even methane or ethanol or double bonded carbon molecules such as ethylene. So, aside of the system design constraints, parameters as the overpotential values or charge transfer resistances, that are determining the final energy balance as well as the overall productivity, will be presented and discussed considering the conditions for diminishing as much as possible the overall cell voltage, looking for high range energy efficiencies in the solar energy conversion to chemical one in comparison with the efficiency achieved by solar energy conversion to electrical energy storage.

According to the potential performance systems and the capacities in design advanced catalysts and controlling characteristics at the nano scale level, it will be demonstrated the industrial feasibility and viability of the electrochemical conversion of CO₂, as alternative for the future energy models considering solar and dark refineries as energy paradigms delivering high current densities at low voltage cells improving energy balance with high stability, avoiding CO₂ emissions and with competitive energy cost of the produced product according to the cost of the CO₂ ton capture, the cost of the consumed renewal electricity and the required electrochemical system investment assuming different hypothesis about its lifetime.

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Materials and Processes for Solar Energy Based Capturing and Transforming of CO₂ and Water into Hydrocarbon Feedstock

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Abstract

Utilization of CO₂ as a feedstock is not only an interesting option for the production of valuable products, but also an important measure to counteract the effect of anthropogenic greenhouse gas emissions. Promising approaches to utilize CO₂ are electrochemical and thermochemical processes based on renewable energy supply such as heat and electricity from solar energy. The German Aerospace Center (DLR) investigates such processes which typically generate a mixture of CO and H₂ (= synthesis gas), which is then converted to liquid fuels like methanol, gasoline or kerosene or to more long-lasting products like chemicals, polymers or fibers allowing for negative emission scenarios.

Research performed at DLR covers the whole process chain from materials development over solar energy-based CO₂ capture to its conversion to liquid fuels, while the present contribution focusses on the capture aspects. Technologies to capture the carbon dioxide (CO₂) which is produced in several industrial processes such as in the cement industry are needed to reach the climate change goals. Among the different existent routes, the calcium oxide looping cycle (CaO-looping cycle) has gained more attention not only because it can be easily integrated in existent industries, using materials that are abundant, cheap, and not environmentally harmful, but also because the energy intensive calcination step of the process can be carried out using a renewable energy source as Concentrated Solar Power (CSP). However, some material issues connected to the continuous cycling processing such as reduction in the sorption capacity of particles as well as particles abrasion problems are the bottleneck of the CaO-looping cycle. As a result of it, the spent sorbent and the broken particles are taken out from the process to keep the CO₂ capture efficiency high and to avoid problems in the reactors respectively. Additionally, to capture the CO₂ pure from the cycle and to avoid further separation steps, the calcination reaction has to be done under a 100% CO₂ atmosphere, but there is few available information about the CaO-looping cycle under the mentioned conditions. We did a material study using Thermogravimetric Analysis (TGA) to investigate the material behavior under real CO₂ capture conditions (calcination in 100% CO₂ atmosphere, and carbonation in 30% CO₂ volume concentration which corresponds to the CO₂ concentration of a cement plant flue gas). We also defined process parameters and reactors concepts for the implementation of a solar CaO-looping cycle in a cement plant, where the spent sorbent and the abraded material could be used in the cement production process.

References

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Ceramic gas separation membranes for the use in CCUS

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Abstract

Carbon Capture Utilization and Storage (CCUS) is an important strategy in order to mitigate greenhouse gas emissions enabling a circular economy. Since CO₂ emissions typically occur at high temperature processes, ceramic gas separation membranes can provide the necessary separation and purification steps, which are key aspects in CCUS.

The presentation introduces different types of ceramic membranes able to separate CO₂, O₂, H₂ or other relevant gases from gas mixtures such as flue gases or synthesis gas. In particular membrane reactors are a promising option because of its energy efficiency enabling the combination of chemical reactions and gas separation (process intensification) [1]. The working principles are ionic transport (CO₃²⁻, O²⁻, H⁺) or molecular sieving in dense or porous membranes, respectively. State-of-the-art processing of membrane components as well as potential applications towards CCUS are described. To reach a high performance of the membrane systems thin film membranes, active surface layers and thermochemical and -mechanical stable supports with designed porosity are required. The production and characterization of membrane structures is explained using the example of sequentially tape cast and laminated supported membranes.

References

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Current perspectives on advanced solid sorbents for CCS: the case of hybrid metal organic framework

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Abstract

In the post-combustion CCS technology scenario, CO₂ adsorption on solid adsorbents is currently considered as a top-notch option [1]. Metal organic frameworks (MOFs), a wide family of crystalline, highly porous coordination polymers, are catalyzing a fast-growing interest in the CCS community [2] driven by their peculiar crystalline structure, high specific surface areas (up to 3000 m²/g) and tunable porosity and composition. These characteristics make them particularly suitable for selective CO₂ sorption applications also under realistic technological options. In this context, the empowering of MOF characteristics by hybridization with graphene related materials (GRM) is an attractive option that has already shown promising in the fields of gas storage and separation and for hydrocarbons adsorption [3]. The presence of GRMs embedded in the MOF crystal very often modifies the morphology of the parent MOF inducing the formation of a new porosity (micro and/or mesoporosity, depending on the MOF and on the GRM type) and thus the developing of higher total pore volume and surface area, particularly suitable for CCS applications. In this talk, after an overview on the synthesis, the characteristics and the applications of literature available MOF and MOF/GRM hybrids for CO₂ and CH₄ adsorption, the CO₂ adsorption capacities and selectivity toward CO₂ over CH₄ probed at high pressure of new-concept MOF/GRMs hybrids will be presented.

References

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Materials selection for the CHEERS project on Chemical Looping Combustion

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Abstract

The CHEERS project is a collaborative project between Europe and China that aims at demonstrating carbon capture by Chemical Looping Combustion (CLC) of petcoke at the scale of 3 MW_{th}, bringing the technology to TRL7. As part of the project, Work Package 3 consists in selecting oxygen carriers with sufficient reactivity towards petcoke, and capable of sustaining the high sulfur and metals content of this fuel.

Natural ores (several ilmenite and manganese ores) and synthetic materials (CMTF perovskites developed by SINTEF [1]) have been tested using IFPEN's batch fluidized bed to measure petcoke combustion kinetics as well as their sensitivity to sulfur. CMTF perovskites' reactivity and mechanical resistance were promising, but passivation in the presence of sulfur could not be avoided.

Two natural ores, an ilmenite from Norway and a Mn ore from China, were selected based on their reactivity, and on their agglomeration and mechanical resistance properties. Further testing was performed in IFPEN's 10 kW_{th} circulating CLC unit, first activating the particles with methane, then using petcoke as fuel, and the aged particles were characterized.

This work was supported by the European Union's Horizon 2020 Research and Innovation Program (CHEERS project No. 764697)

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Carbon Capture Utilisation : some CEA on-going works and achievements

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Abstract

The future place of carbon molecules in energy and chemistry will be shortly discussed in the framework of French and international contexts relative to renewable energies (some of these trends will be illustrated through the main European demonstrator projects). CEA takes benefit of this situation and is now structured around many programs related to energy transition; one of them being dedicated to “Carbon Circular Economy”. The structure of this program will be briefly presented both in terms of fundamental and applied research topics, and illustrated in the case of electro-thermo-catalytic conversion from basic resources (CO₂, carbon loaded wastes, water, power) to molecules of interests. Three common opened issues will be highlighted: (i) various chemical pathways could be considered to produce methane, methanol, e-fuels (and other related molecules of interest) with specific features regarding energy and carbon budgets, and related costs, (ii) corresponding conversion units should be operated in a technico-economical optimal way, (iii) critical components will be identified.

Examples relative to ongoing works on these issues will be provided. The merits of chemical pathways such as Power-to-X, Biomass-to-X, and Power-and-Biomass-to-X will be discussed and illustrated through the Méthycentre project [1]. Optimal conversion unit control and management will be discussed in terms of both whole process numerical modelling and mathematical optimisation, and illustrated through the Jupiter 1000 project [2]. A specific emphasis will be dedicated to two main components of the conversion chain: the electrolyser and the catalytic conversion reactors. CEA high temperature electrolysis technology, main advantages and results will be presented. Some achievements regarding catalytic fixed bed reactors will be presented in the framework of methane and liquid fuel synthesis.

A brief focus will be provided on a specific CEA initiative concerning fundamental researches in the Carbon Circular Economy field. This focus concerns about 20 PhDs or Postdoctoral researchers who have started working on this field by the end of 2020.

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KEROGREEN - A plasma-based approach for CO₂ neutral fuel production

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Abstract

Aviation is a striking example of a sector that is difficult to decarbonise. The KEROGREEN project is a collaborative European project on producing green kerosene [1]. The new approach and process of KEROGREEN reduces overall CO₂ emission by creating a closed carbon fuel cycle and at the same time creates long-term large-scale energy storage capacity. The concept starts from water and air and is entirely powered by renewable electricity. The target product is aircraft grade kerosene synthesised through (i) the splitting of CO₂, (ii) separation and upgrading of the dissociation products, (iii) syngas formation by water-gas-shift, (iv) Fischer-Tropsch synthesis and (v) further refinery steps (hydrocracking). The KEROGREEN project aims at combining these different technologies in a container-sized plant and bringing them all to TRL level 3-4 [1,2].

This contribution discusses two aspects among the initial two steps in the process chain, i.e. (i) the CO₂ splitting by means of a gas discharge (CO₂ plasmolysis), and (ii) the separation of oxygen from the CO₂/CO/O₂ gas mixture by means of solid oxide electrolysis cells (SOCs). The focus will be thereby on the material aspects and future challenges.

Microwave plasma-driven conversion of CO₂, aimed at producing CO feedstock for synthetic fuels has recently re-gained attention [3]. Reported conversion rates vary between 10% and 80% and are strongly dependent on the process conditions [4]. These plasmas are typically characterised by gas temperatures of several 1000 K. Recently, the fundamental processes in CO₂ microwave plasmas – in a more homogeneous discharge regime as well as in a constricted mode with strong gradients in e.g. gas temperature -- have been disentangled [5]. These results in turn can be translated into reactor design and future research directions.

A main challenge of such a plasma-based CO₂ splitting process is fact that CO remains mixed with O₂ and residual CO₂. Therefore, efficient gas separation and recuperation are essential for obtaining pure CO, which can be used further downstream in the water gas shift and Fischer-Tropsch reactions. The combination of a plasmolysis with solid oxide electrolyte cells (SOCs) has the potential to tackle this gas separation challenge.

High-temperature SOCs are advanced electrochemical energy storage and conversion devices with high conversion/energy efficiencies. They offer attractive high-temperature electrolysis routes which can convert CO₂ or CO₂/H₂O into valuable chemical feedstock, enabling large-scale energy storage/conversion and facilitate the integration of renewable energies into the electric grid. SOCs typically consists of an ion conducting electrolyte, an anode and a cathode where electrolysis reactions take place. The high operating temperature and the carbon–oxygen double-bond of CO₂, which is difficult to activate, put forward strict requirements for SOC cathode. Suitable cathode materials should be of high catalytic activity and excellent long-term stability.

For the KEROGREEN process the synergistic combination of SOCs and plasma processes is examined where CO₂ activation is used to relax the SOCs material requirements. A significant part of this contribution will be spent on elaborating on different concepts, first results and remaining challenges [6].

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Engineering photosynthetic microorganisms for direct solar chemical and fuel production from carbon dioxide

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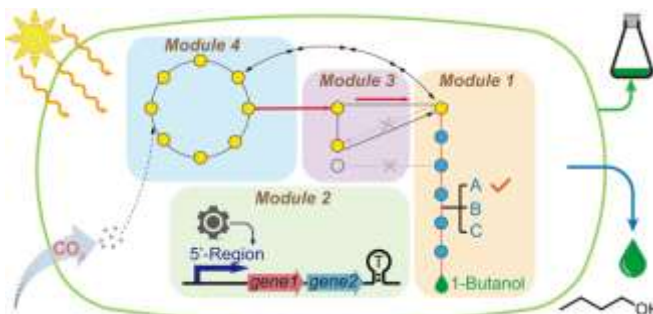
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Abstract

Cyanobacteria, photosynthetic microorganisms with the same type of photosynthesis as plants and algae, can be engineered to produce solar chemicals and solar fuels in direct processes from carbon dioxide [1]. This presentation will outline our strategies to engineer cyanobacteria to produce alcohols using butanol and ethanol as examples from solar energy and carbon dioxide. Introduction of a single gene encoding KivD resulted in isobutanol producing strains of *Synechocystis* PCC 6803 [2]. Knowledge based modelling of the identified bottleneck KivD resulted in strains with significantly increased isobutanol production [3]. Using our best isobutanol strain in long-term experiments a cumulative titer of 911 mg per L was observed with a maximal rate of 43.6 mg per L and day [4]. A similar approach to systematically engineer cyanobacteria to produce 1-butanol resulted in cells with a cumulative titer of 4.8 g per L and a maximal rate of 302 mg per L and day [5], quickly doubled to 600 mg 1-butanol per L and day [6]. Moreover, cyanobacteria engineered for increased growth/CO₂-fixation showed increased product formation, exemplified with the alcohol ethanol [7, 8].

Figure. Systematic modular engineering of a photosynthetic microorganism resulted in cells with significant levels and rates of 1-butanol production directly from CO₂ [5].



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Methanation of captured carbon dioxide in a solid oxide membrane reactor

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Abstract

Conversion of captured carbon dioxide to grid compatible synthetic natural gas may contribute to diminish its emissions and the reliance on fossil reserves, enabling its recycling as a renewable, safe and established fuel, as well as the storage of the renewable energy required for the production of the electrolytic hydrogen and to power the chemical conversion process [1].

This work presents a study of CO₂ methanation in a solid oxide electrolyte membrane reactor, combining H₂ production and CO₂ hydrogenation in the same unit, under conditions representative of post-combustion carbon dioxide capture streams. A Cu electrocatalytic film (cathode), active for CO₂ methanation, was coated by electroless [2] on a commercial (CoorsTek Membrane Sciences) anode (Ni-BZCY)-supported solid electrolyte (BZCY) candle and afterwards characterized, both as prepared and after testing, by SEM-EDX, XRD and XPS. Characterization results revealed that electroless technique allowed the deposition of a very thin Cu film (<2 microns), rendering a low electrical resistance and enhanced proton transfer to the catalyst surface and reactant/product diffusion, and some signs of degradation associated with the mechanical or chemical stability of the anode-supported electrolyte candle under the high temperatures and concentrated CO₂ atmospheres utilized.

The effect of temperature (between 325-550 °C under open circuit) and applied potential (between -2 and 2 V at 450 °C) on CO₂ conversion and selectivity to CH₄ and CO were studied at bench scale, under atmospheric pressure and feeding the reacting gas mix (42 NI/h, H₂/CO₂=4) and a diluted H₂ stream (source of H⁺) to the cathode and anode chamber, respectively. CH₄ was preferentially formed over CO on Cu/BZCY. CH₄ selectivity increased with temperature up to 400 °C. At higher temperatures, where methanation (exothermic) and reverse water gas shift (endothermic) reaction is retarded and favored, respectively, CH₄ selectivity starts to decrease, while CO selectivity increases. A temperature of 450°C was selected for the potentiostatic tests to assure high values of co-ionic/electronic conductivity of BZCY and CH₄ yield. Cu/BZCY can be electrochemically enhanced for CO₂ methanation, for both positive and negative potentials, showing an “inverted volcano” behavior.

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Rational exploration of innovative materials by quantum simulation

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Abstract

To reduce the ecological footprint of current industrial applications, the transition from fossil fuels towards alternative cleaner fuels is strongly encouraged. Nevertheless, such a transition will require to design innovative processes and their related materials (many catalytic ones) by gaining atomic scale insights on concepts often located at the crossroads of chemistry and physics.

Through various examples, this lecture will highlight how quantum simulation combined with well defined experiments may help to identify relevant materials and rationalize their atomic scale properties in emerging applications. In particular, some of the following case studies addressed at IFPEN will be illustrated:

- the identification of metal hydrides for hydrogen storages based on their predicted enthalpy of formation [1]
- the decoding of reaction mechanisms involved in biomass conversion catalyzed by transition metal sulfides [2]
- the prediction of optimal catalysts and electro/photocatalysts based on volcano curve relationships [3]
- the tuning of opto-electronic properties of two-dimensional semiconducting heterojunctions for CO₂ reduction photocatalysts [4]

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Electrocatalytic CO₂ Reduction with Copper/Carbon Catalysts to C₂ and C₂₊ Value-added Products supported over Gas Diffusion Layers

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Abstract

Circular economy of CO₂ is nowadays a hot issue for our society. Nevertheless, its successful implementation still requires a lot of new knowledge in reliable systems. Electrochemical CO₂ reduction (ECO₂R) is one of several promising strategies to mitigate CO₂ emissions. Developing novel advanced functional nanostructured catalyst materials and systems for electro conversion of CO₂ to alternative fuels or value-added products in order to meet the global energy needs and so as a mean to curb the increasing amounts of CO₂ in the atmosphere is of utmost importance. Till date, copper (Cu) and copper based materials are the only heterogeneous catalyst systems that has shown a propensity to produce valuable hydrocarbons and alcohols such as ethylene and ethanol¹.

Copper nanoparticles were synthesized by several wet methods including chemical reduction and hydrothermal synthesis. The presence of Cu₂O revealed by XRD and the crystal orientation have shown to have influence on the distribution of products. The formation of Cu₂O/Vulcan in close contact leads to a superior faradaic efficiency to ethylene (FE_{C₂H₄} > 25%), while the low CH₄/C₂H₄ indicates this structure favours the dimerization steps, leading to C₂s products. The electrodes were evaluated in flow filter-press cell with Gas Diffusion electrodes under neutral and alkaline electrolyte conditions. A clear correlation between working voltage, structural electrocatalyst properties, pH and product distribution was observed, with higher selectivity towards ethylene generally obtained at intermediate potentials. Other aspects such as hindering the Hydrogen Evolution Reaction and evaluating the role of halide species were also considered. The results and both selectivity favouring and limitation factors were discussed and potential solutions were addressed here.

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