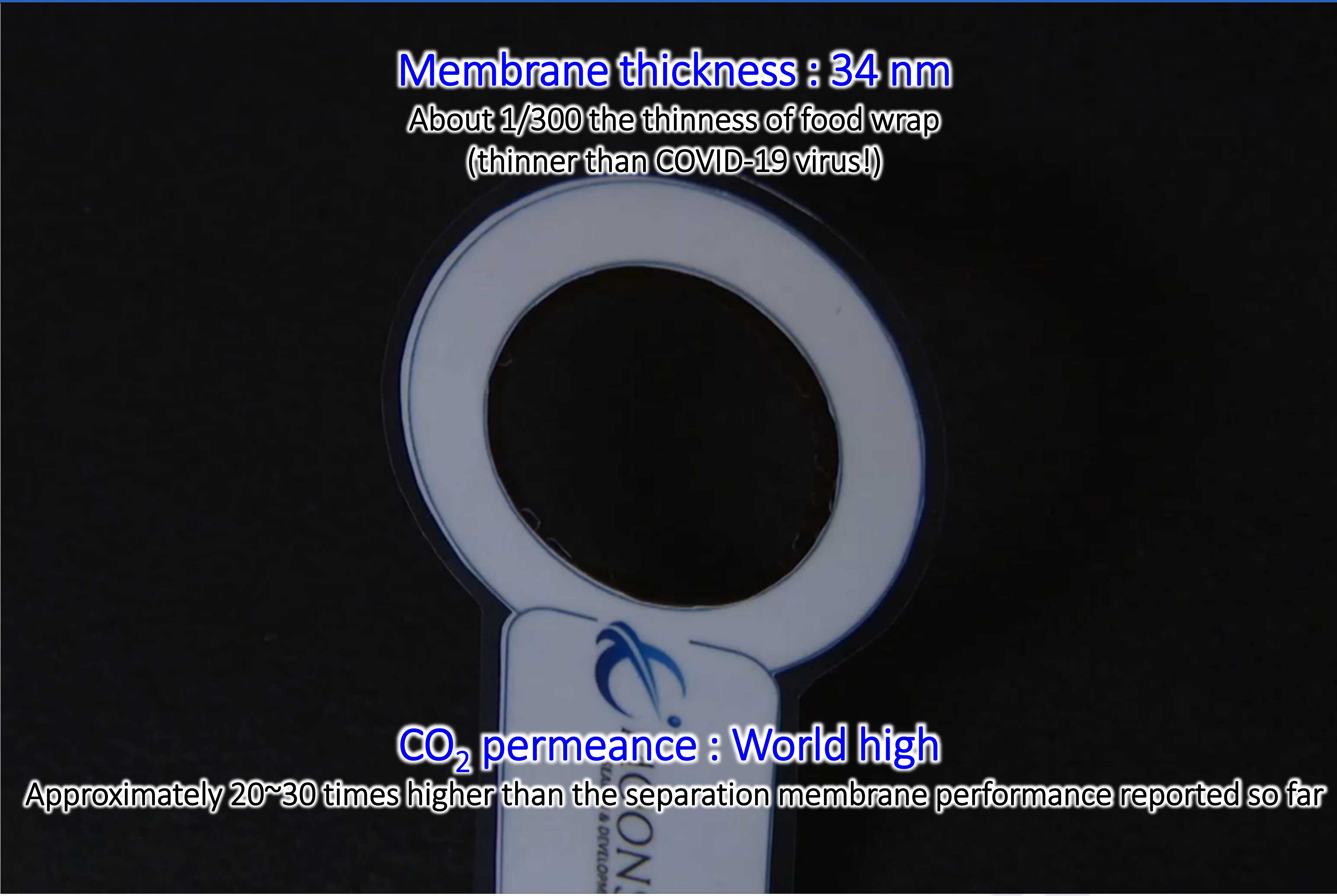
Development of Global CO_2 Recycling Technology towards "Beyond-Zero" Emission

PM : Shigenori FUJIKAWA
International Institute for Carbon-Neutral Energy Research, Kyushu University, Professor

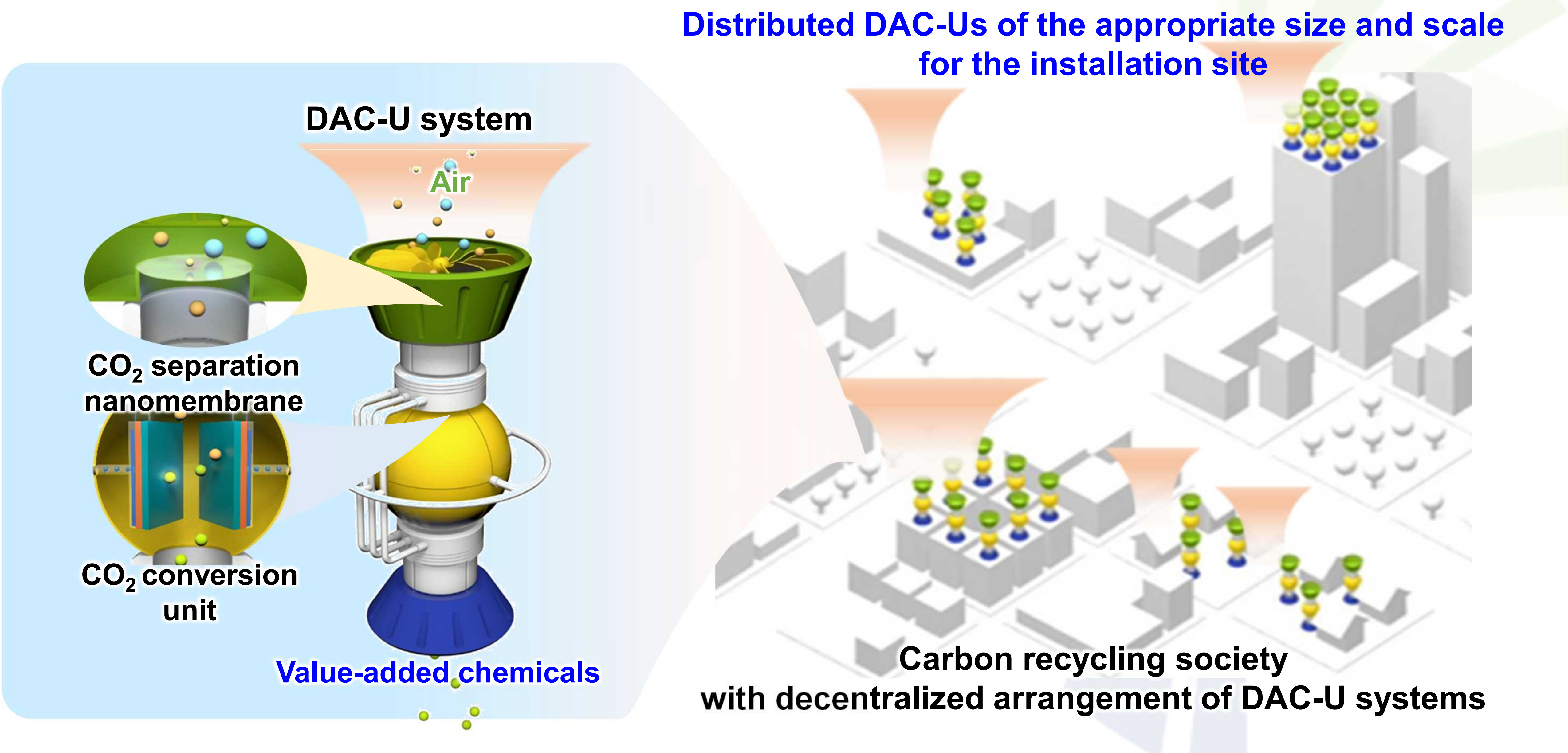
PJ partner institutes :
Kumamoto Univ., Hokkaido Univ., Univ. Tokyo, Kagoshima Univ., Osaka Inst. Tech., Shinshu Univ., Univ. Illinois at Urbana Champaign, Nanomembrane Tech. Inc.

CO_2 separation nanomembrane with the world's highest CO_2 permeability from Kyushu University



New carbon recycling society: resources from the air

The innovative separation nanomembranes with overwhelmingly high CO_2 permeability realizes CO2 capture directly from the atmosphere, which has been thought to be impossible until now. This membrane separation unit is integrated with an electrochemical or thermochemical CO_2 conversion unit to create the Direct Air Capture and Utilization (DAC-U) system, a continuous process from atmospheric CO_2 capture to carbon fuel production. The size-scalable DAC-U system will be distributed and deployed to contribute to the construction of a carbon-circulating society based on local production for local consumption.



"Moonshot for beyond Zero Emission Society"

MOZES

Moonshot for beyond Zero-Emission Society

CO_2

CO_2 capture nanomembrane

highly CO_2 permeable nanomembrane

1000 times concentration

MOONSHOT RESEARCH & DEVELOPMENT PROGRAM

CO_2 conversion

Permeate gas : concentrated CO_2 + N_2, O_2, Ar

CO_2 conversion under oxygen

MOONSHOT RESEARCH & DEVELOPMENT PROGRAM

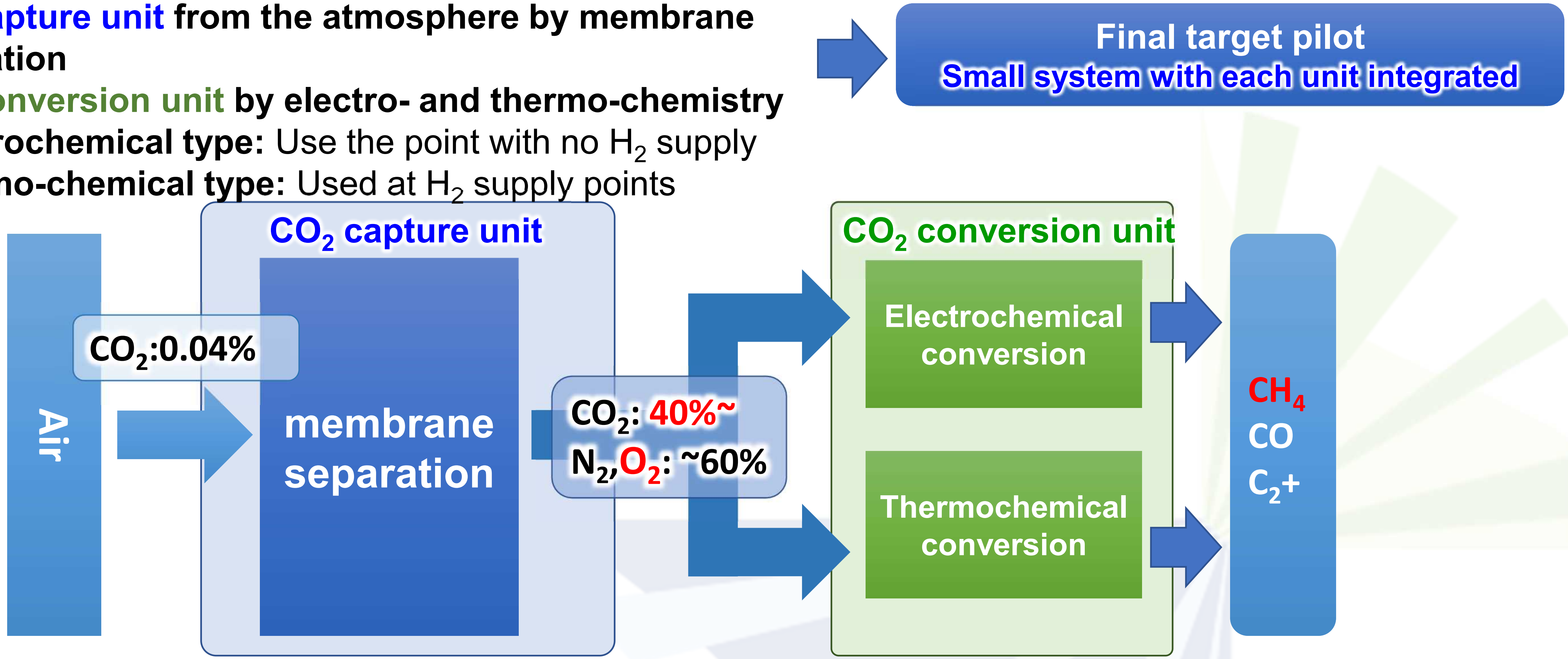
Green fuel production from CO_2 mixture gas

CO, CH_4, Ethanol, C_xH_y

Target pilots for this project

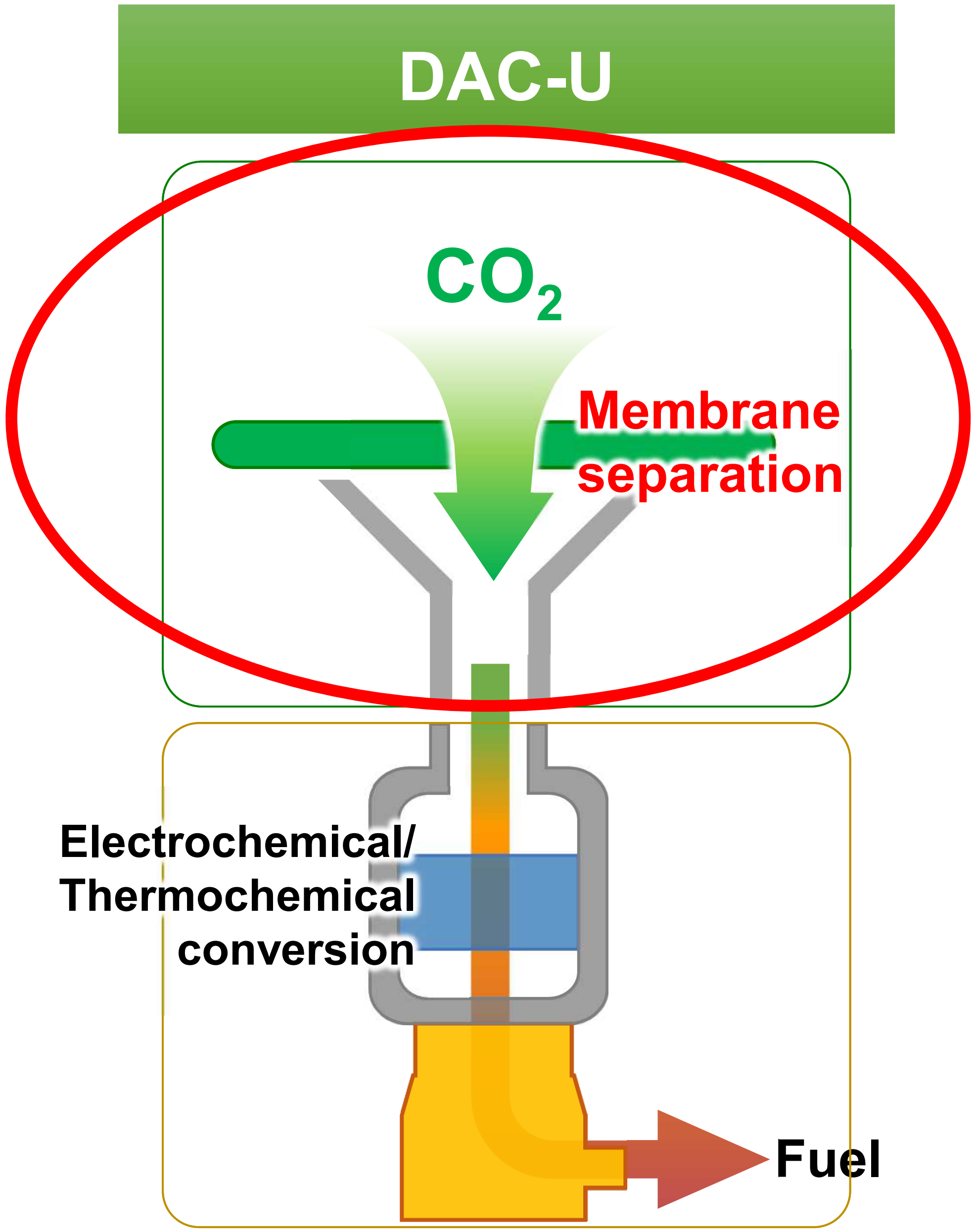
Target developments

- **CO_2 capture unit from the atmosphere by membrane separation**
- **CO_2 conversion unit by electro- and thermo-chemistry**
  Electrochemical type: Use the point with no H_2 supply
  Thermo-chemical type: Used at H_2 supply points



KPI for 2024

- Development of CO_2 selective membranes with the CO_2/N_2, CO_2/O_2 selectivities of more than 30 and 10, respectively.
- CO_2 conversion from CO_2 mixed gas
  - Electrochemical conversion: Continuous production of CH4, CO, C_2H_4
  - Thermochemical conversion: continuous production of CO, CH_4



Realization of highly scalable and distributable CO_2 capture technology

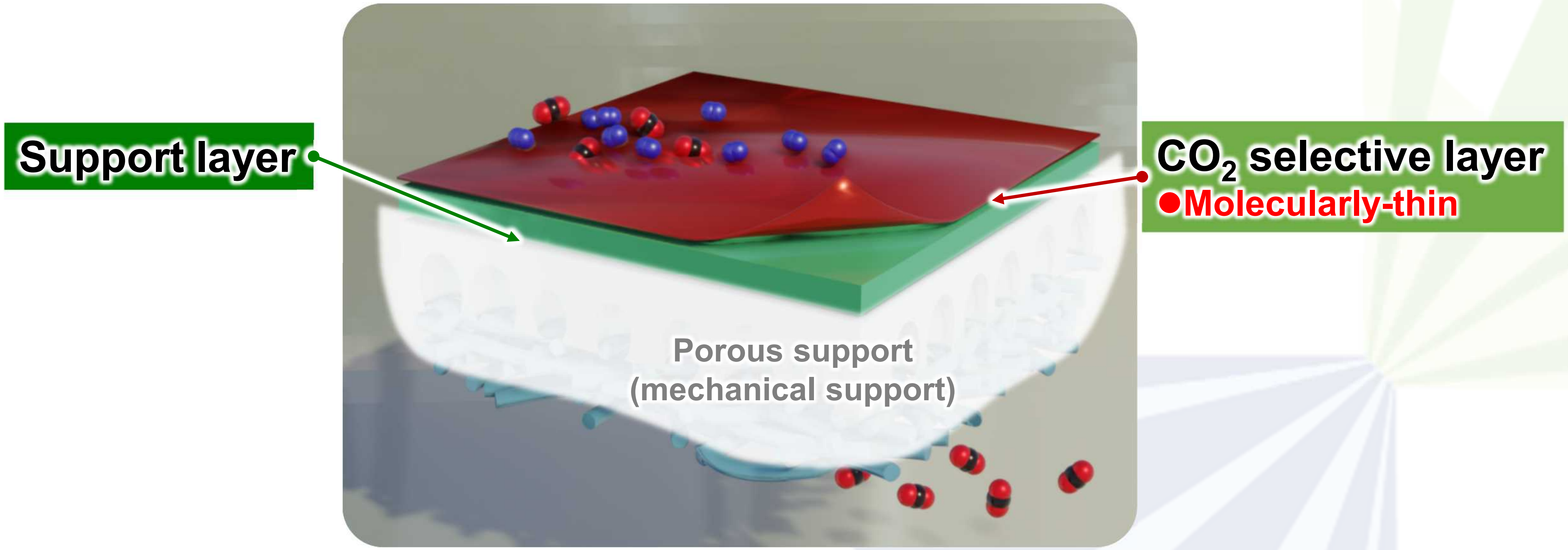
Shigenori FUJIKAWA (Kyushu Univ.)
Masashi Kunitake (Kumamoto Univ., Unit leader)
Tomoyasu HIRAI (Osaka Inst. Tech.)
Yoshiro KANEKO (Kagoshima Univ.)
Shin-Ichiro NORO (Hokkaido Univ.)
Toyoki KUNITAKE (NanoMembrane Tech. Inc.)

CO_2 capture research unit

Approach of membrane preparation

Membrane structure：Thin film composite of CO_2 selective and support layers

- **Support layer: Free-standing and highly gas permeable**
- **Selective layer：variety of CO_2 selective materials**



Development step

①Support layer

- Development of silicone material with high CO_2 permeabilities

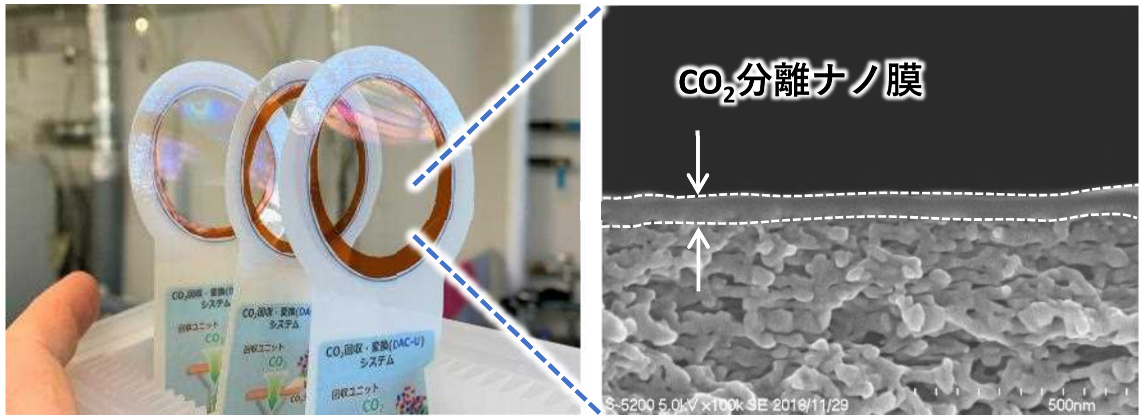
②Selective layer

- Systematic exploring CO_2-philic materials
→molecularly thin layer on a support layer

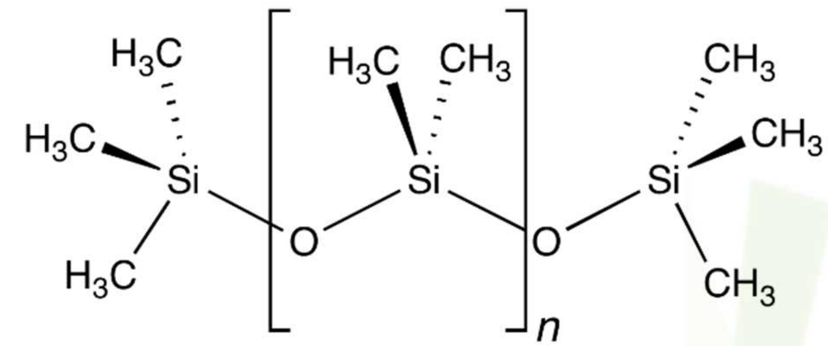
③ Controlled bonding of the selective layer to the supporting layer

④Large area production of separation nanomembranes

Development of silicone supported nanomembrane with high CO_2 permeability

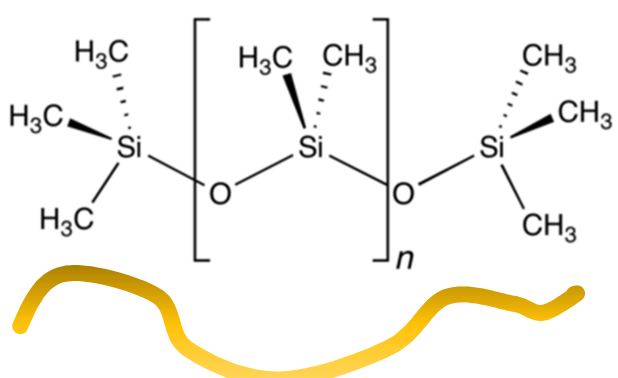


Fujikawa, et.al., Chem. Lett. **2019**, *48, 11, 1351-1354*



Polydimethylsiloxane (PDMS) (silicone)

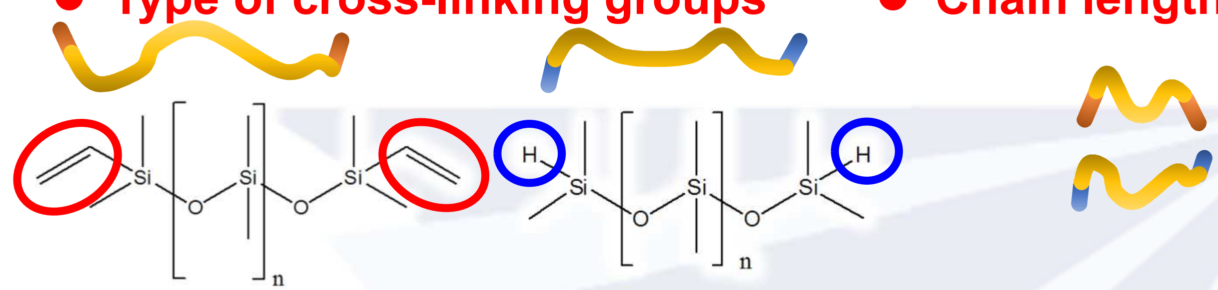
Basic molecular structure



Cross-liking

Terminally bifunctional PDMS

- Type of cross-linking groups
- Chain length

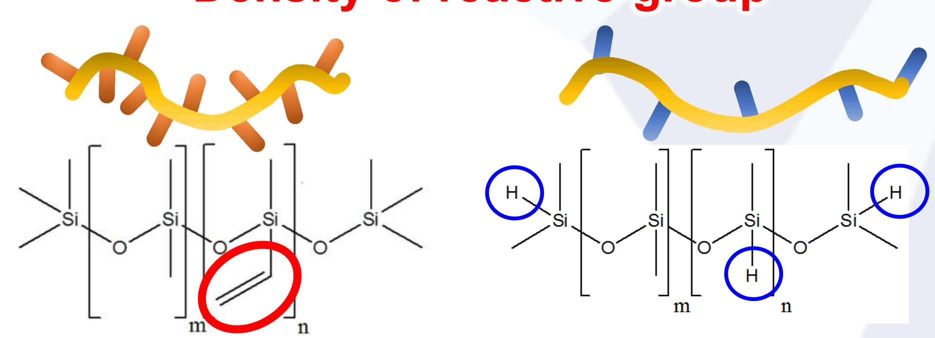


More than 10 types of synthesis completed

×

Side-chain reactive PDMS

- Density of reactive group



More than 20 types of synthesis completed

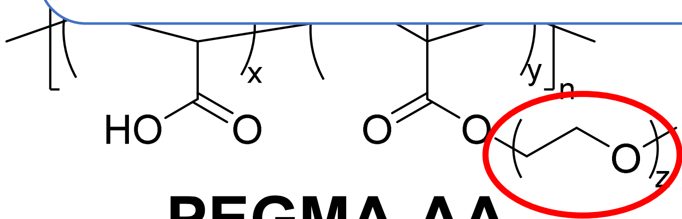
Candidate Structures

- Low cross-linking
- High molecular weight

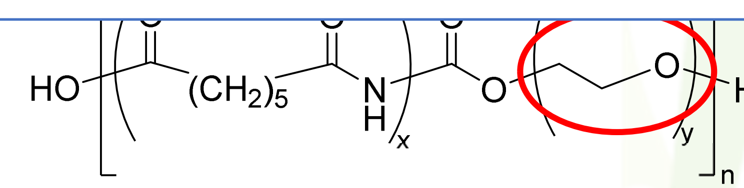
Development of highly CO_2 selective materials for membranes

2024 KPI

- Development of separation membranes with CO_2/N_2 and CO_2/O_2 selectivity of 30 and 10, respectively.



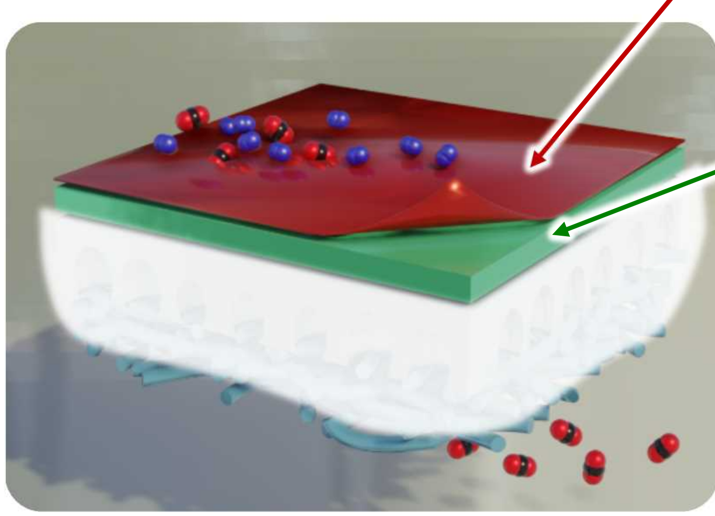
PEGMA-AA



Polyamide-PEG

	PDMS	PEGMA-AA	Polyamide-PEG
CO_2 / N_2	10.2	51.9	23~72
CO_2 / O_2	4.5	18.3	⇐ World top

Final target CO_2/X: >30

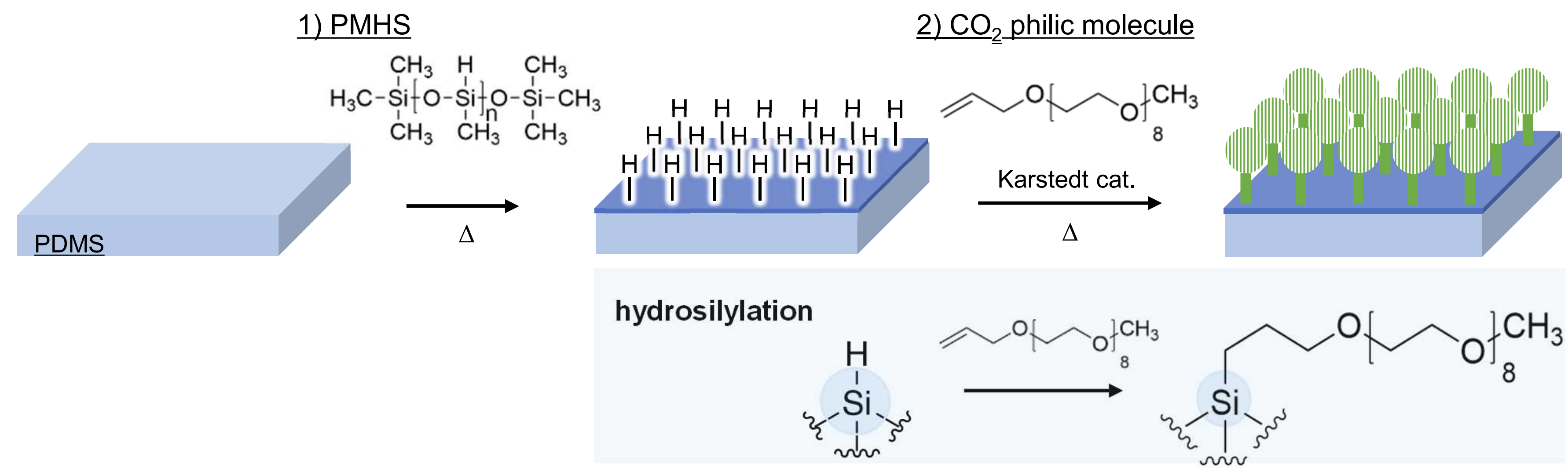


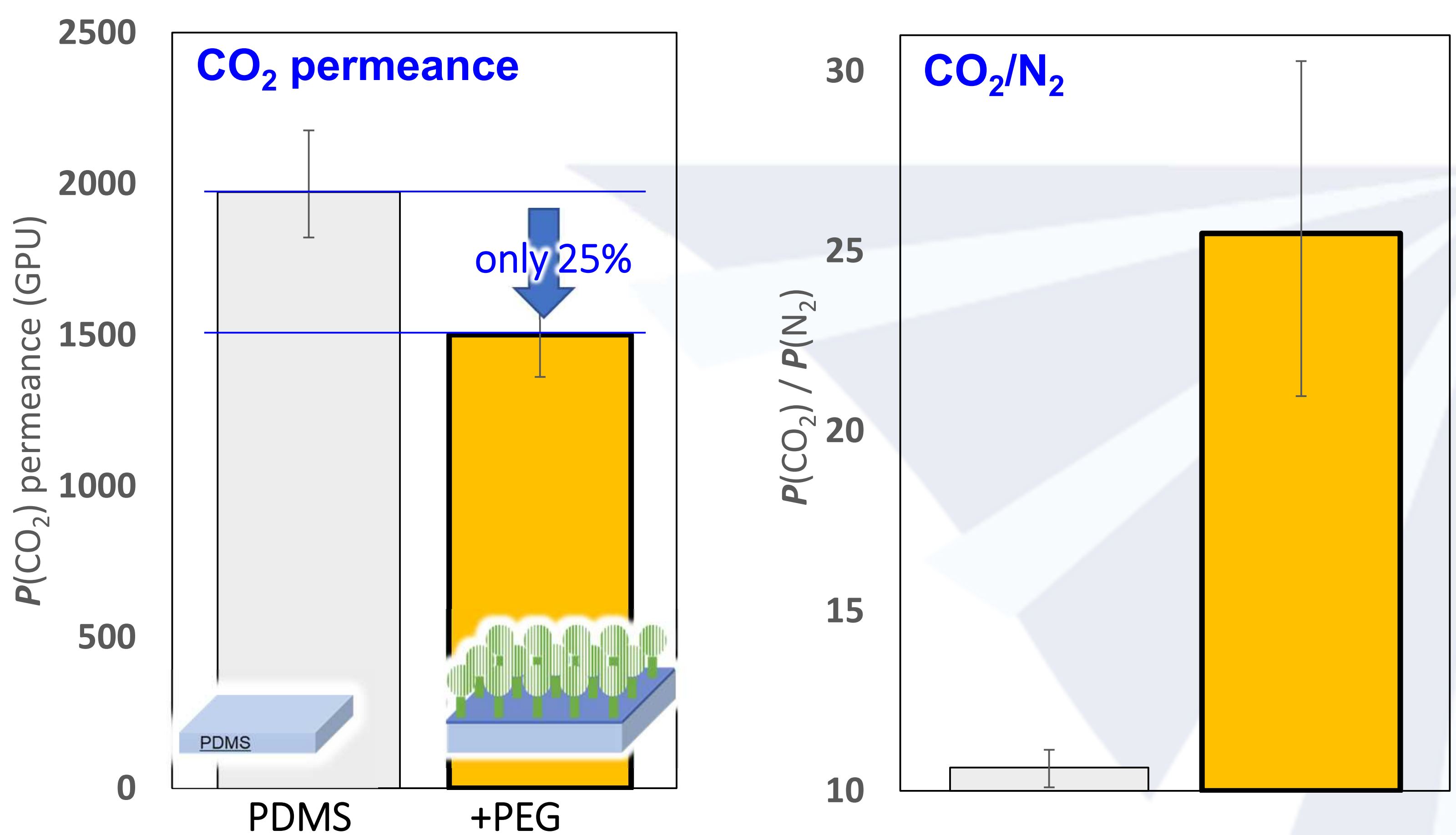
CO_2 selective layer (polar materials)

Support nanolayer (non-polar surface)

Challenge: Improving the compatibility of these layers

Selectivity enhancement by chemical modification of a membrane surfaces

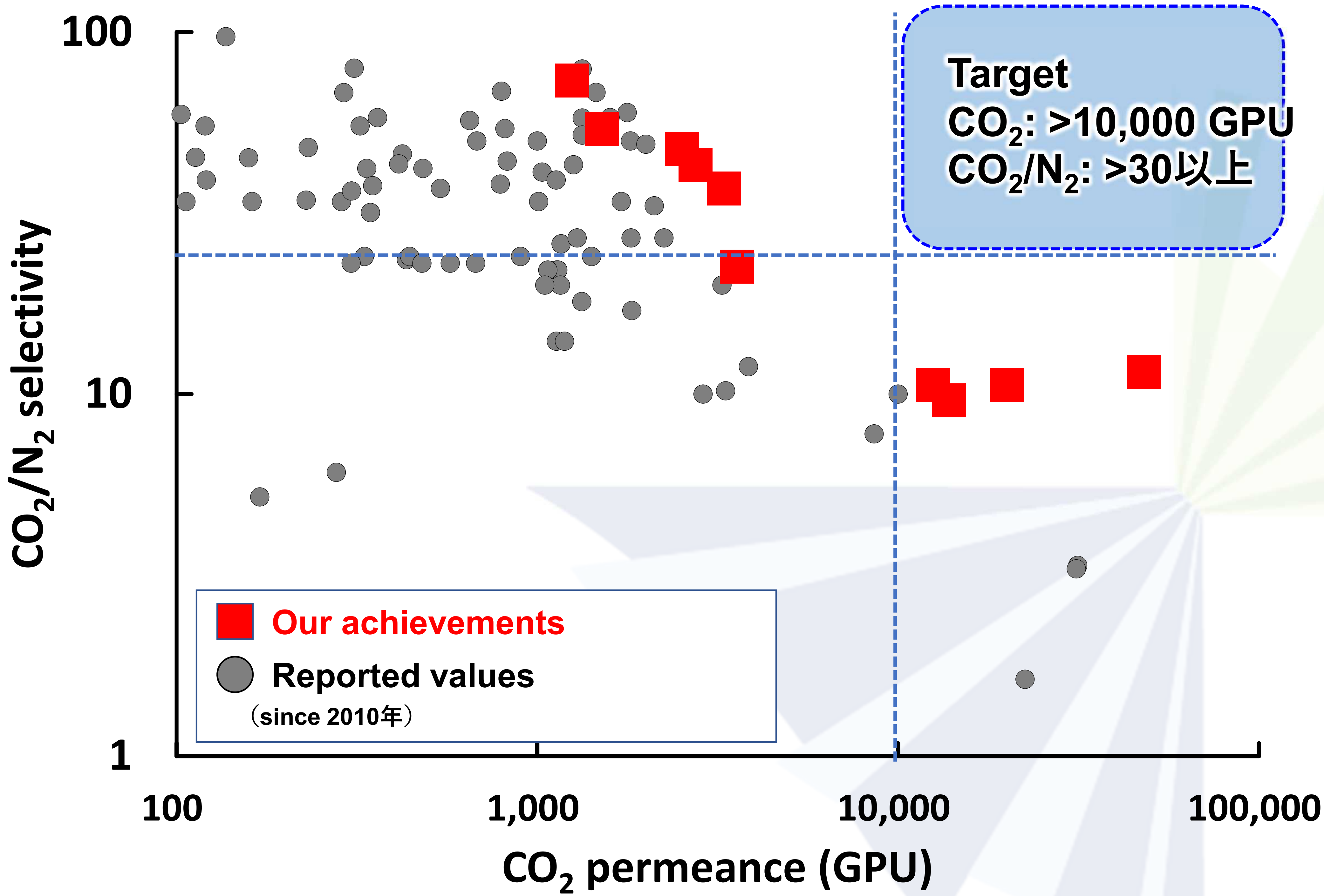




Improved selectivity without rapid CO_2 permeability loss

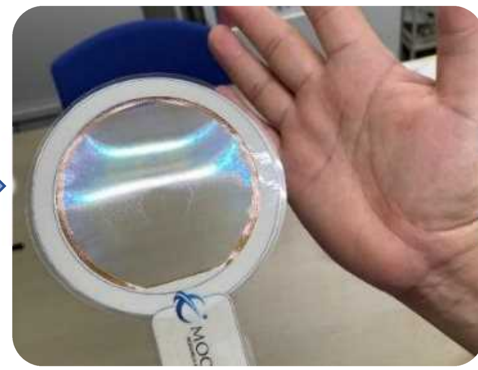
Performance of developed membranes (international comparison)

[11]

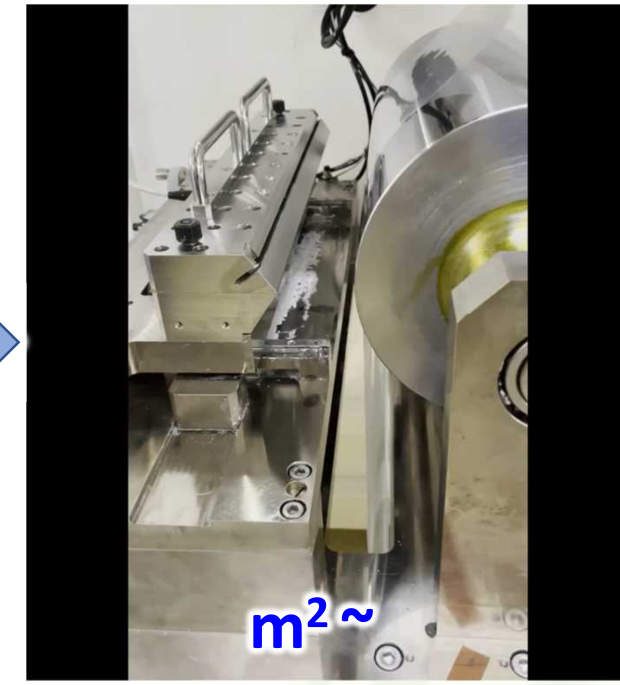


World top level!

Large scale preparation of nanomembranes

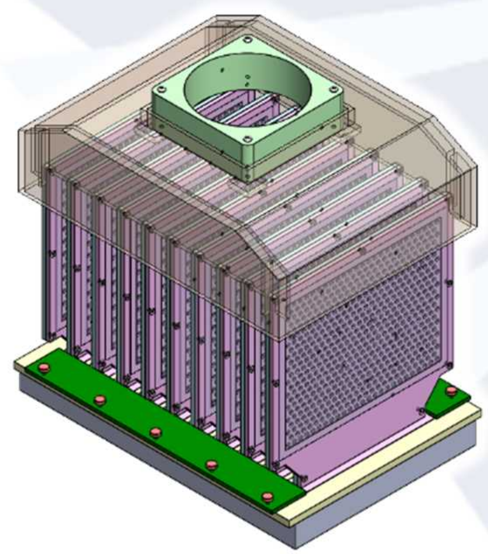


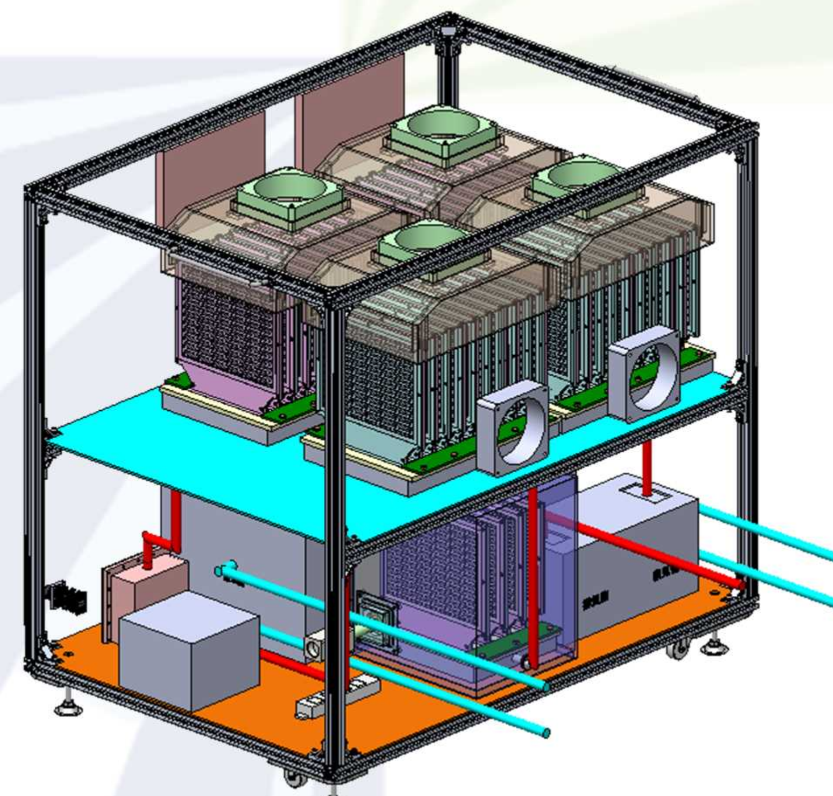
~ 10 cm^2



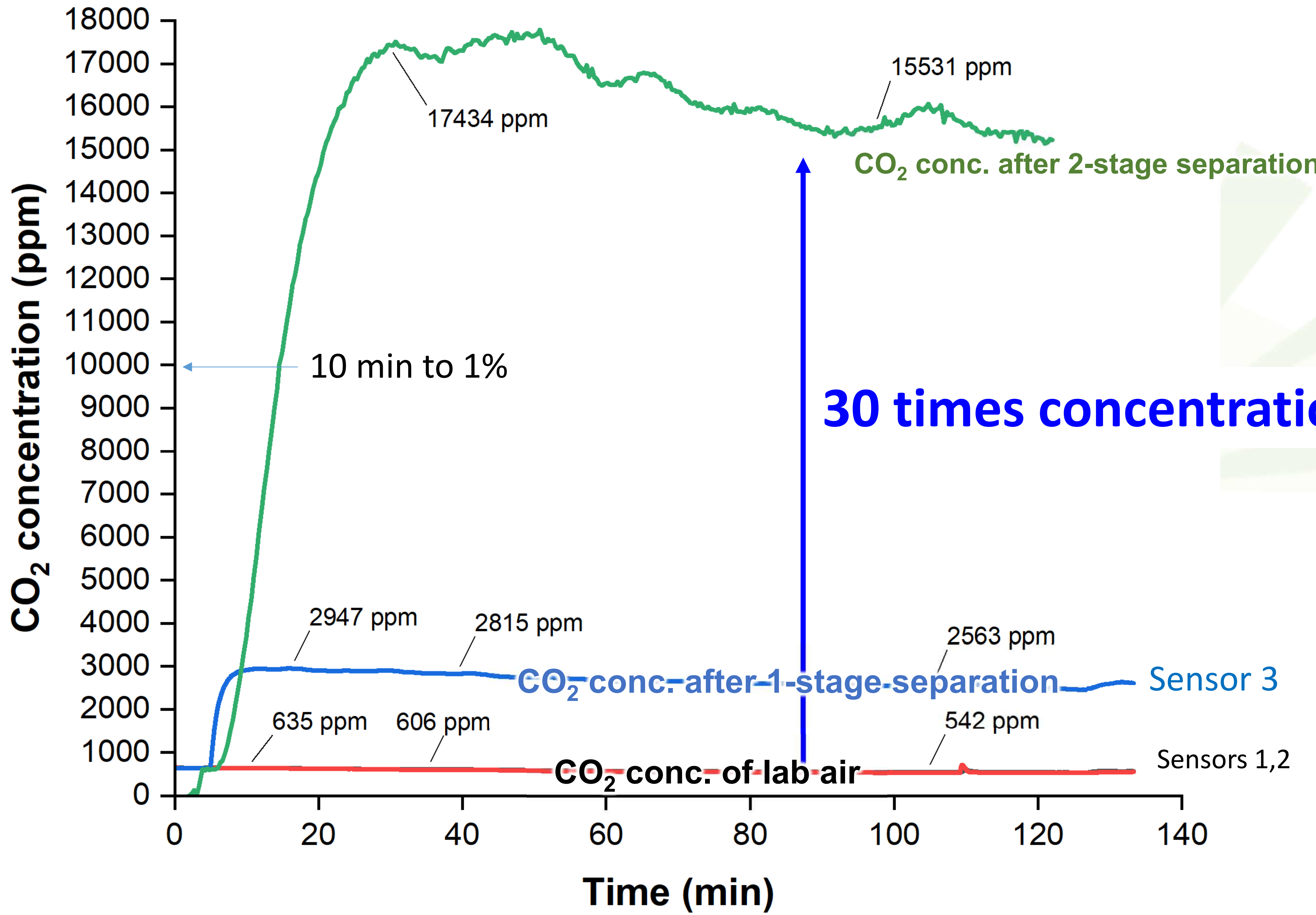
Length: >10m

Roll-to-roll process for membrane fabrication





Separation performance of 2-stage membrane separation module

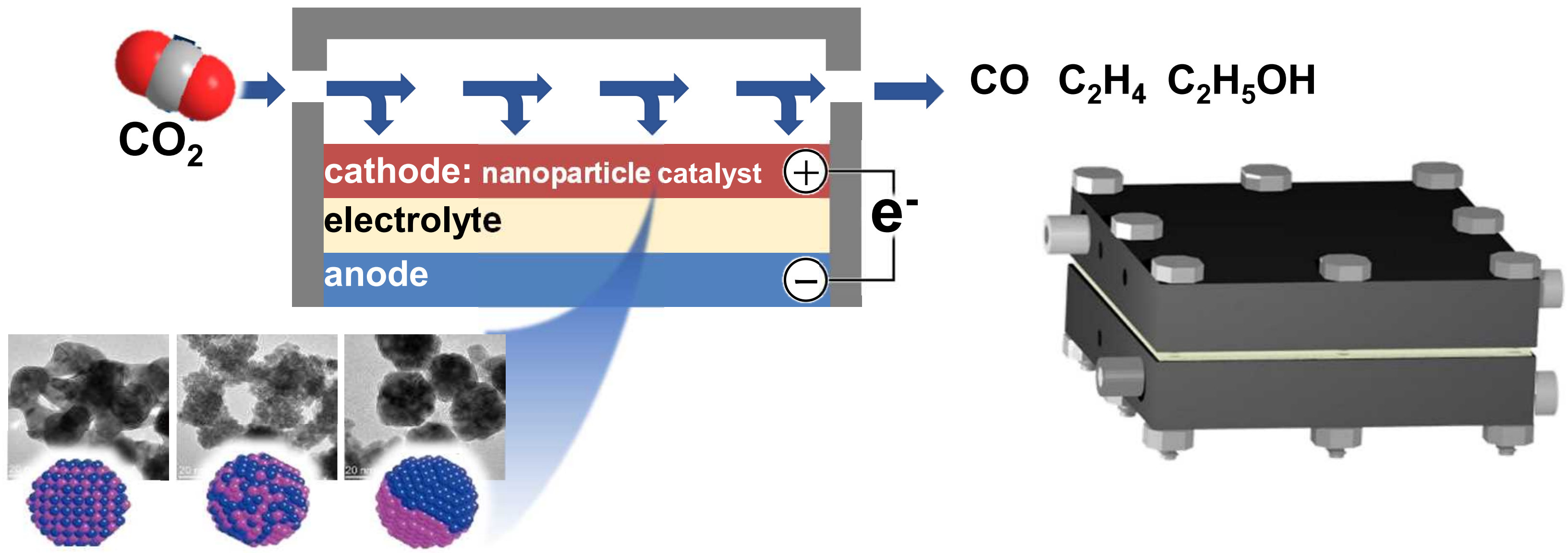


Demonstration of CO_2 enrichment at module level

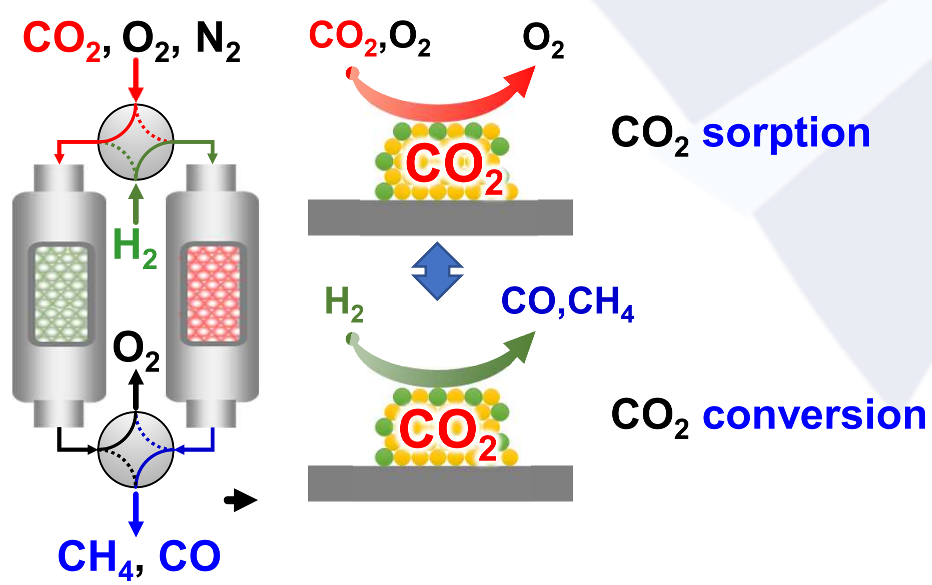
Development of CO_2 conversion unit using electrochemical and thermochemical reactions

Production of carbon resources from CO_2 mixed gas separated by separation nanomembranes

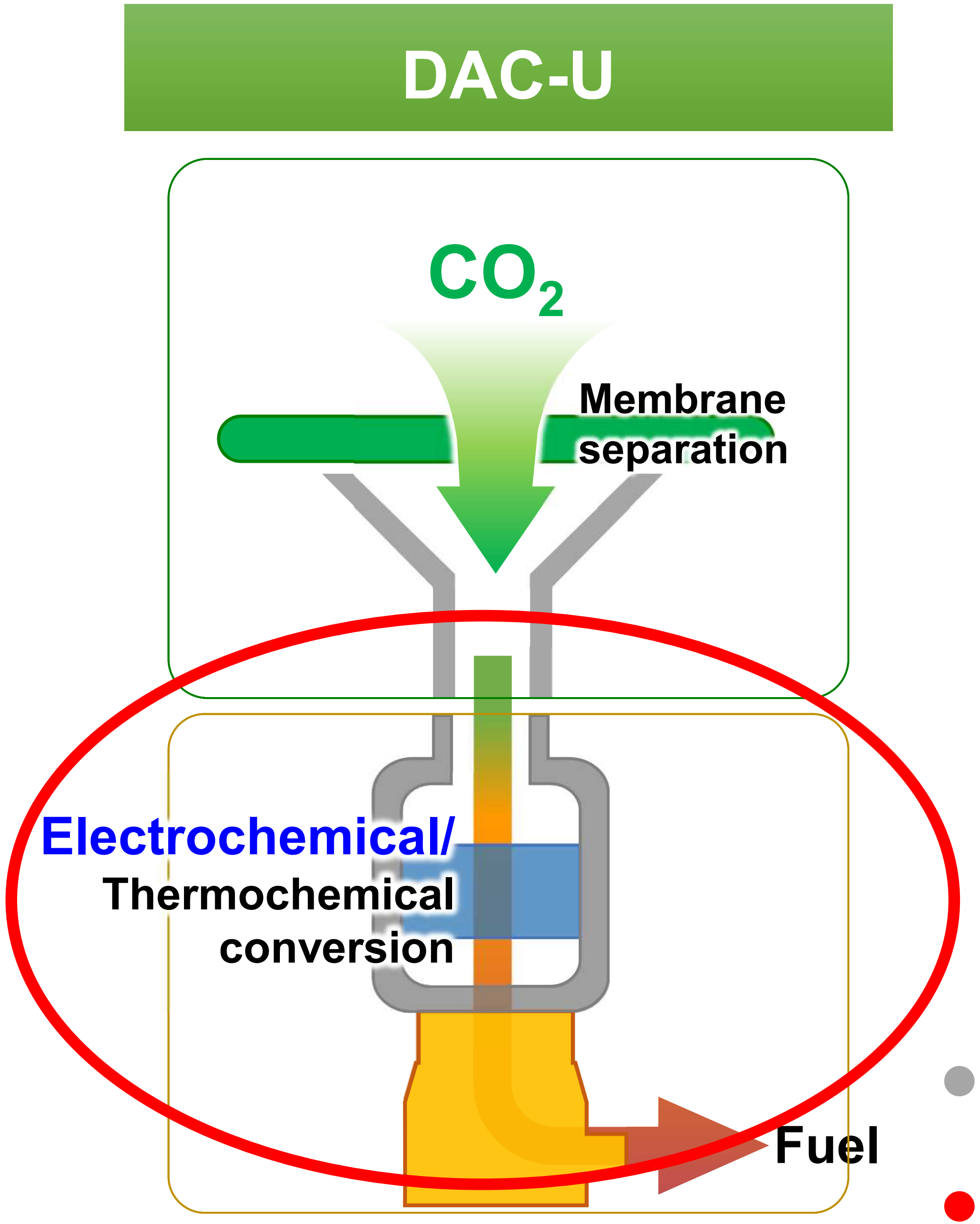
1. Production of basic chemicals and fuels by electrochemical conversion

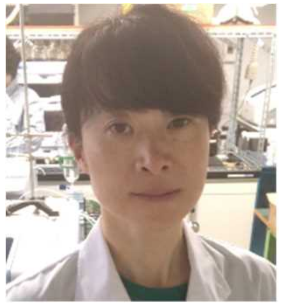


2. Production of C1 compounds by thermochemical conversion



Development of an electrochemical unit to produce carbon compounds from CO_2 mixed gas



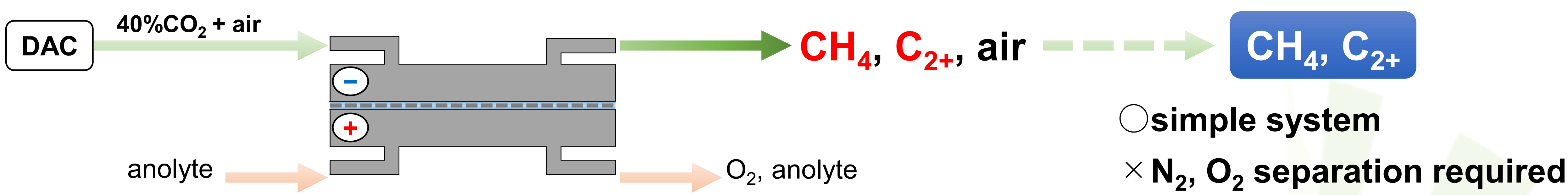


Miho YAMAUCHI （Kyushu Univ/）
Paul KENIS （Univ. Illinois at Urbana Champaign）

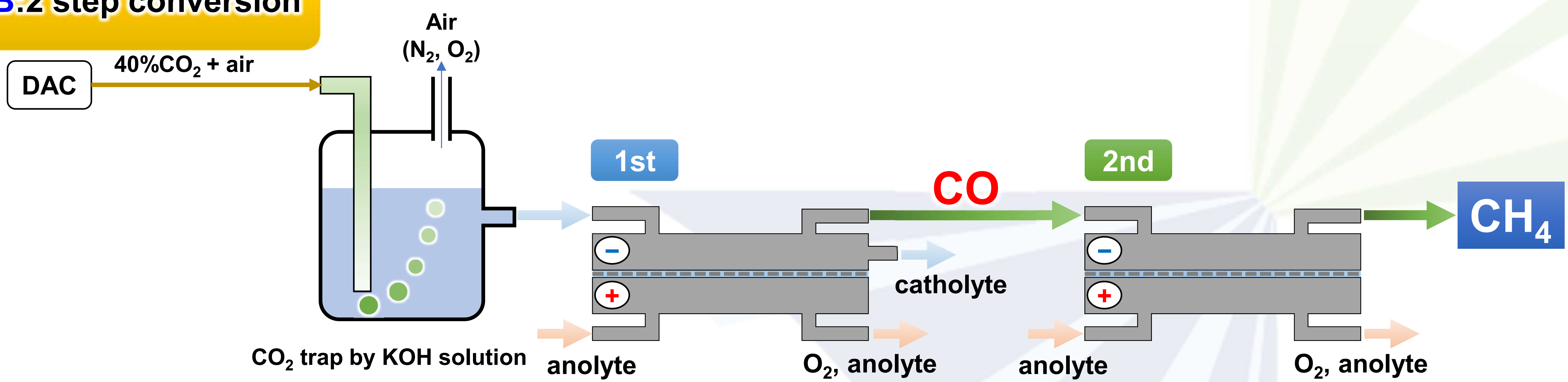
- Is it possible to fabricate large-area nanomembranes for CO_2 separation?
- What kind of chemical products can be produced?
- Can CO_2 be converted from O_2 mixed gas as feed gas?

Electrochemical CO_2 conversion combined with DAC [16]

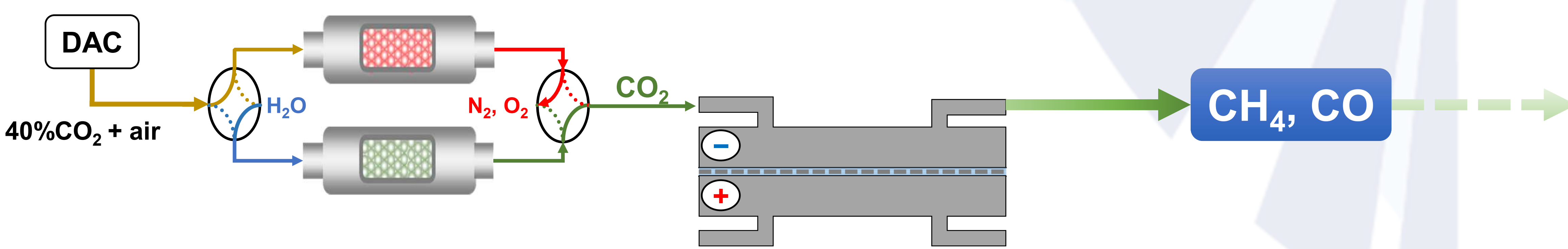
A: Direct conversion



B: 2 step conversion



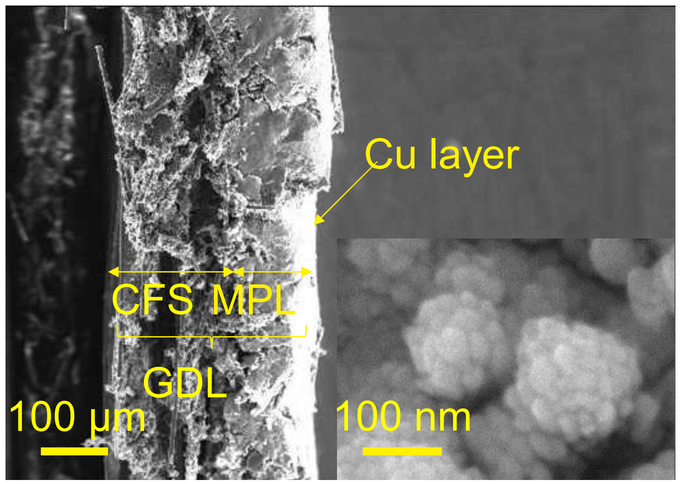
C: O_2 removal

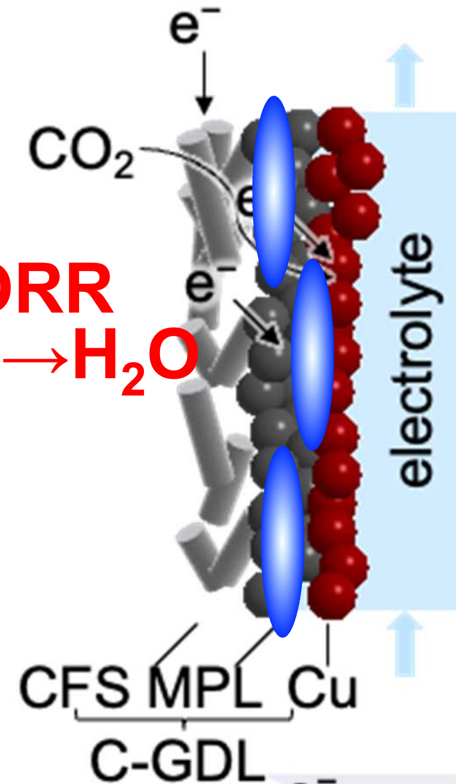


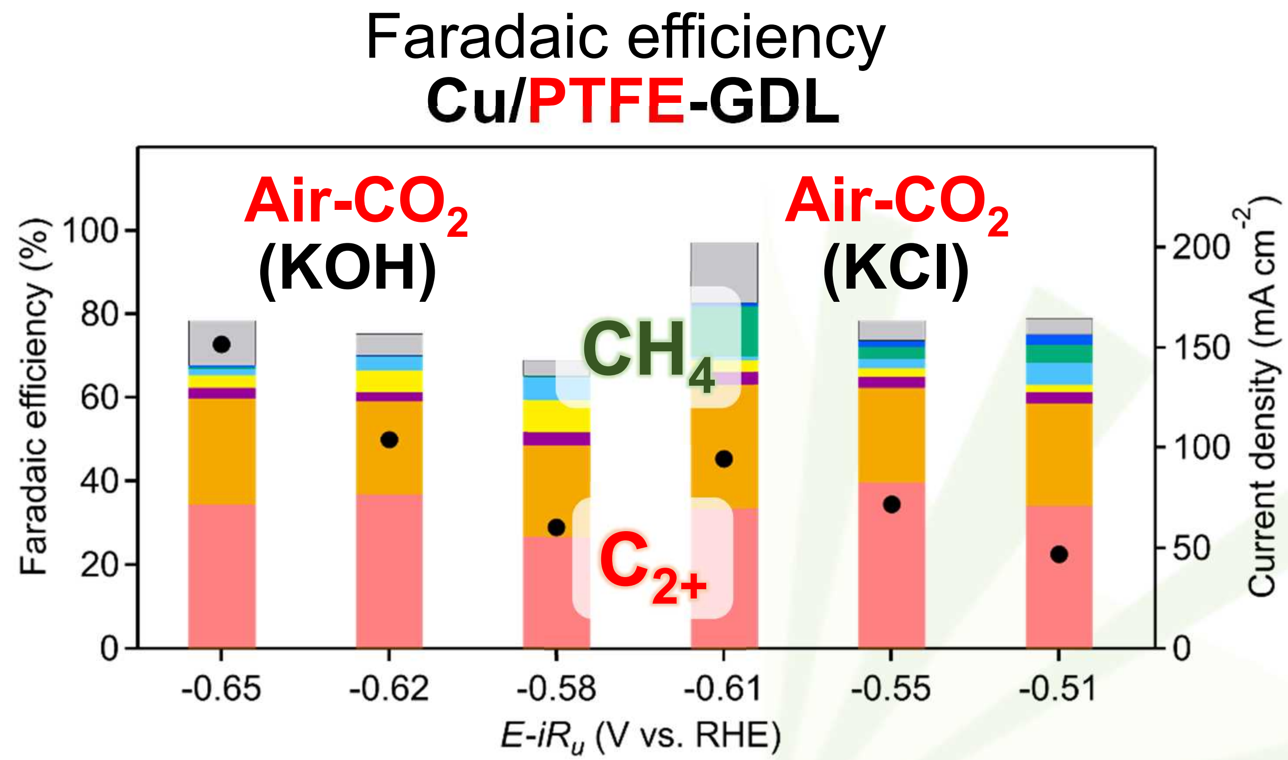
Type: A

Direct CO_2 conversion using 60% air 40% CO_2 mixed gas

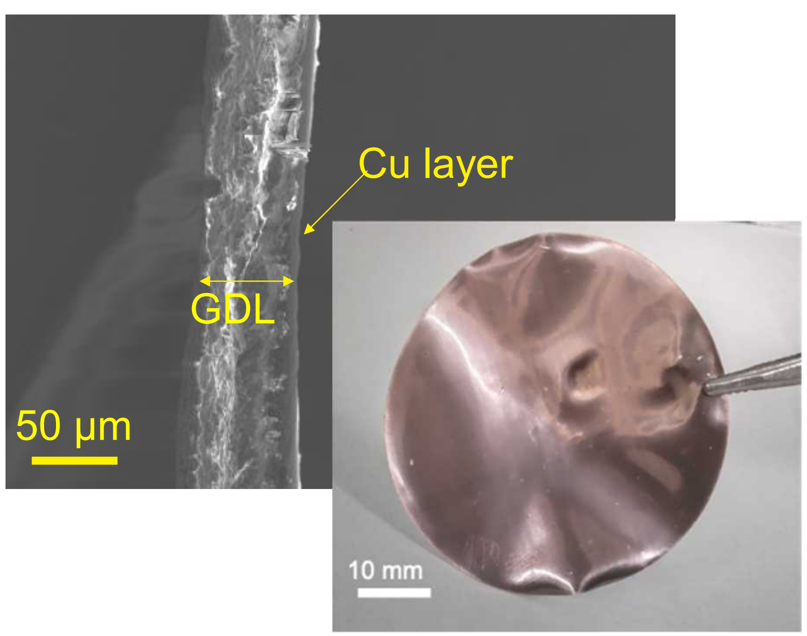
Cu/C-GDL

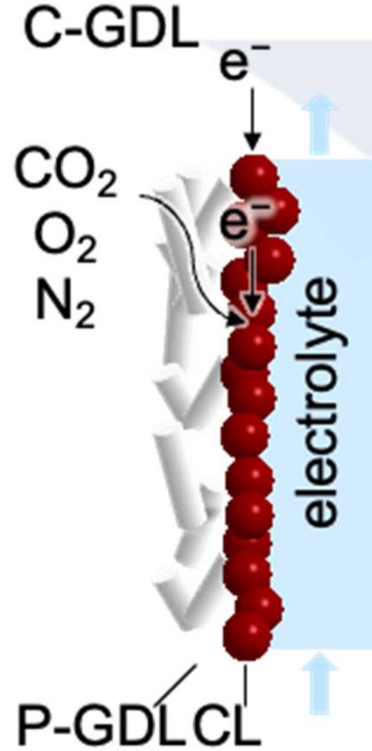


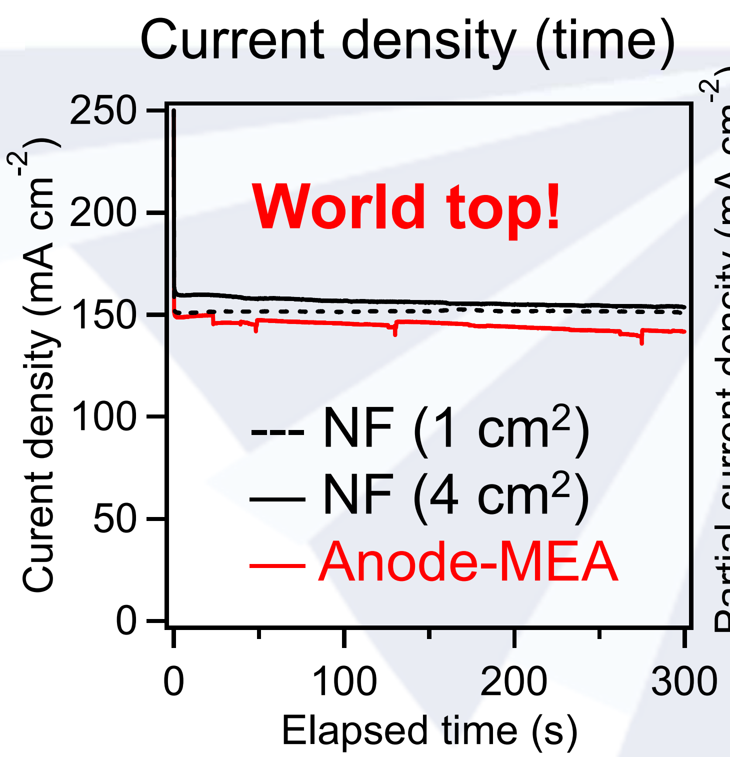


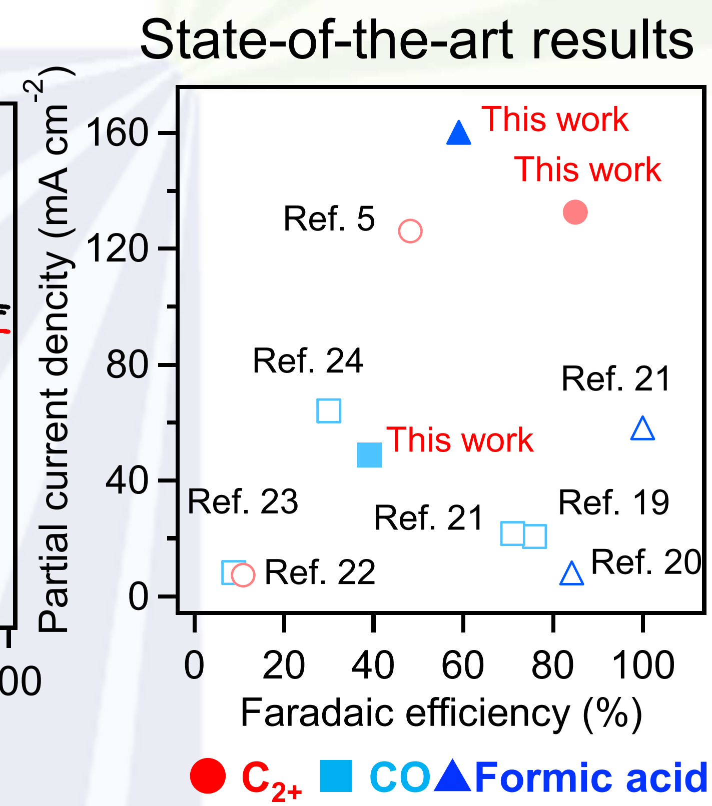


Cu/PTFE-GDL









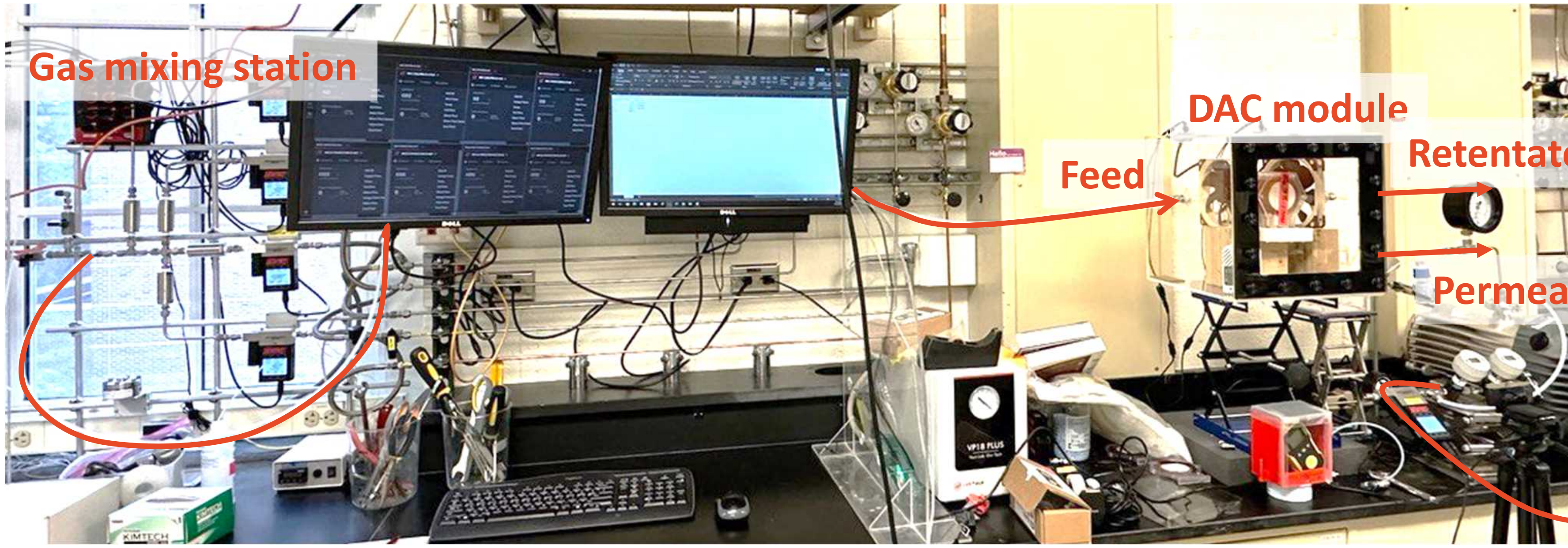
with Dr. Anzai

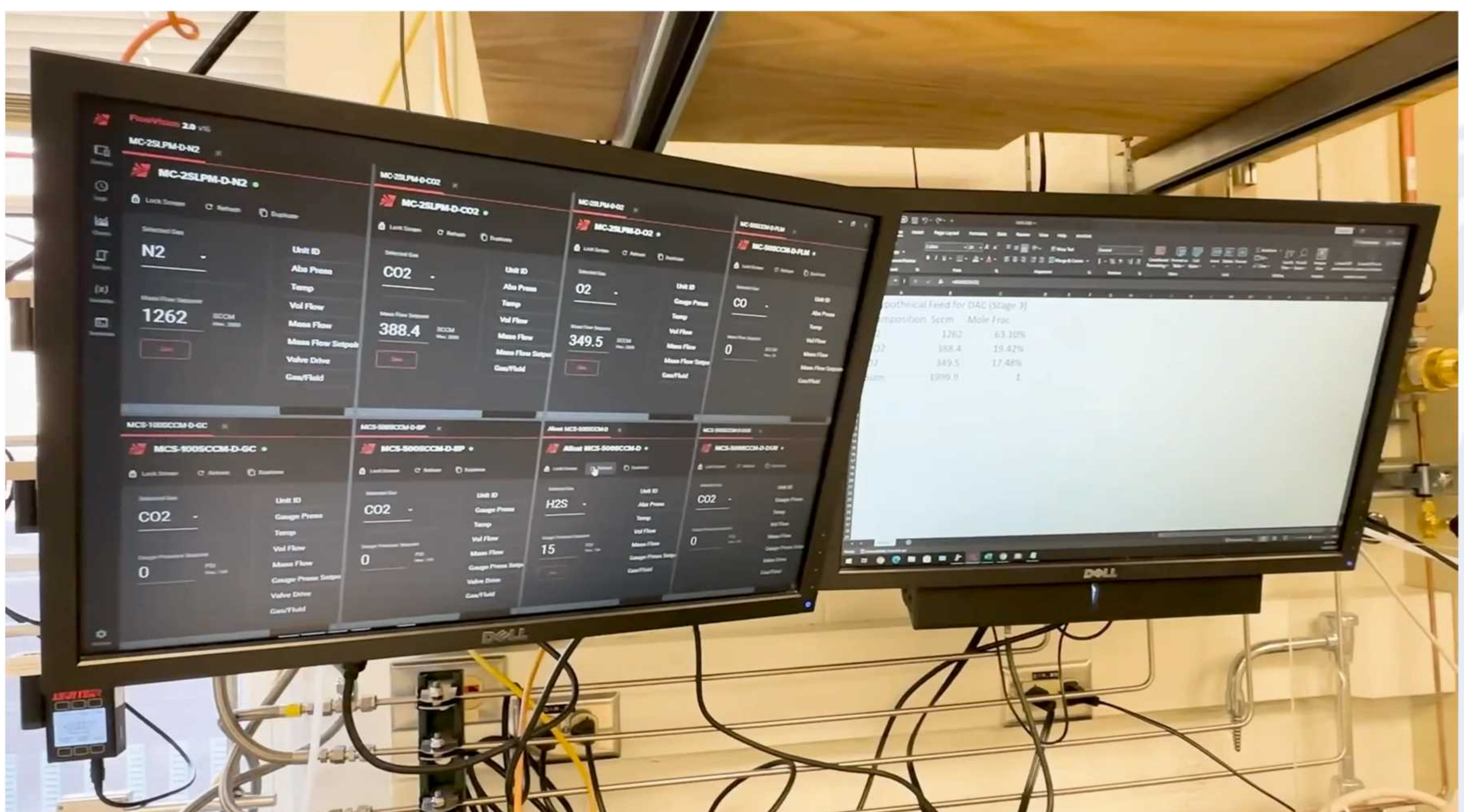
Chem. Comm., 59, 11188-11191 (2023), inside cover

Type: A

Concentration of Air-CO_2 and direct CO_2 conversion

ILLINOIS





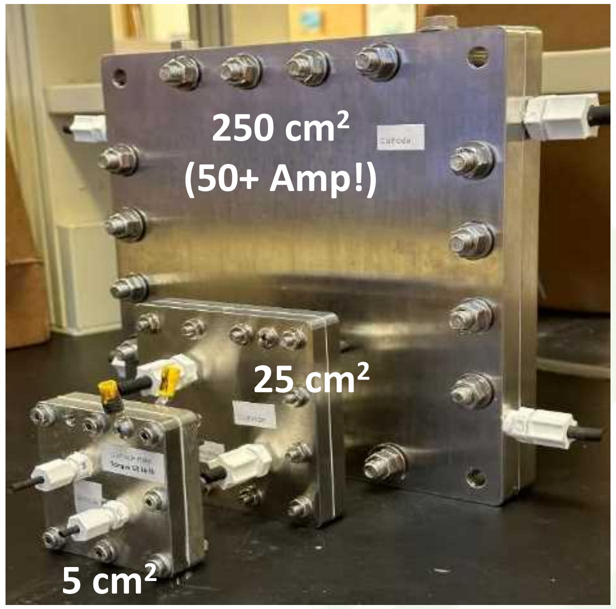
Mr. Vijay Shah
Mr. Benjamin Sit

Type: A

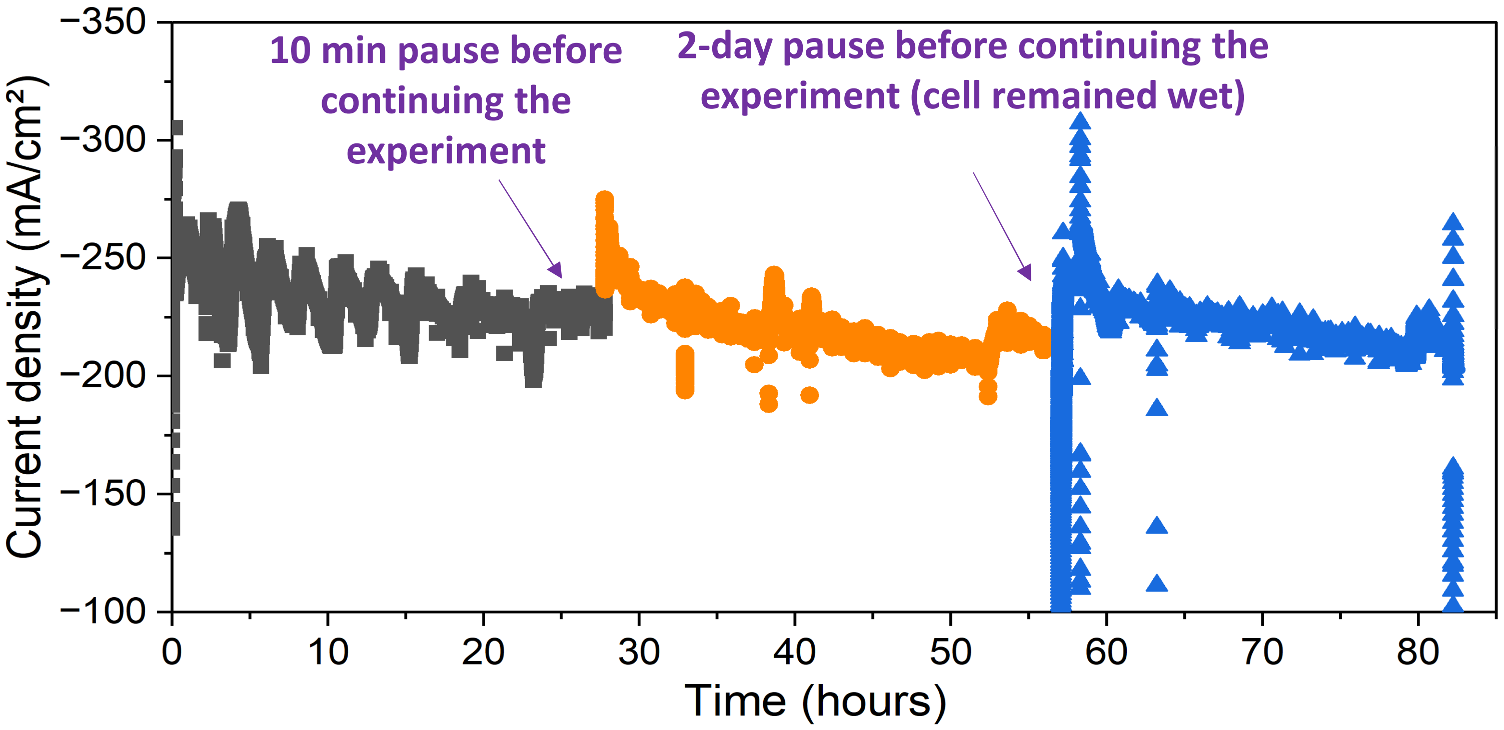
Large MEA Cell for CO_2 conversion and performance evaluation

ILLINOIS

- UIUC has tested CO_2 to CO using pure CO_2 with 25 and 100 cm^2 MEA cell for durability study
- Long-term tests of 25 cm^2 cell showed high conversion of CO_2 to CO and stable current density over more than 80 h.
- UIUC will perform CO_2 to CO conversion using larger cell with NEDO mixed gas.

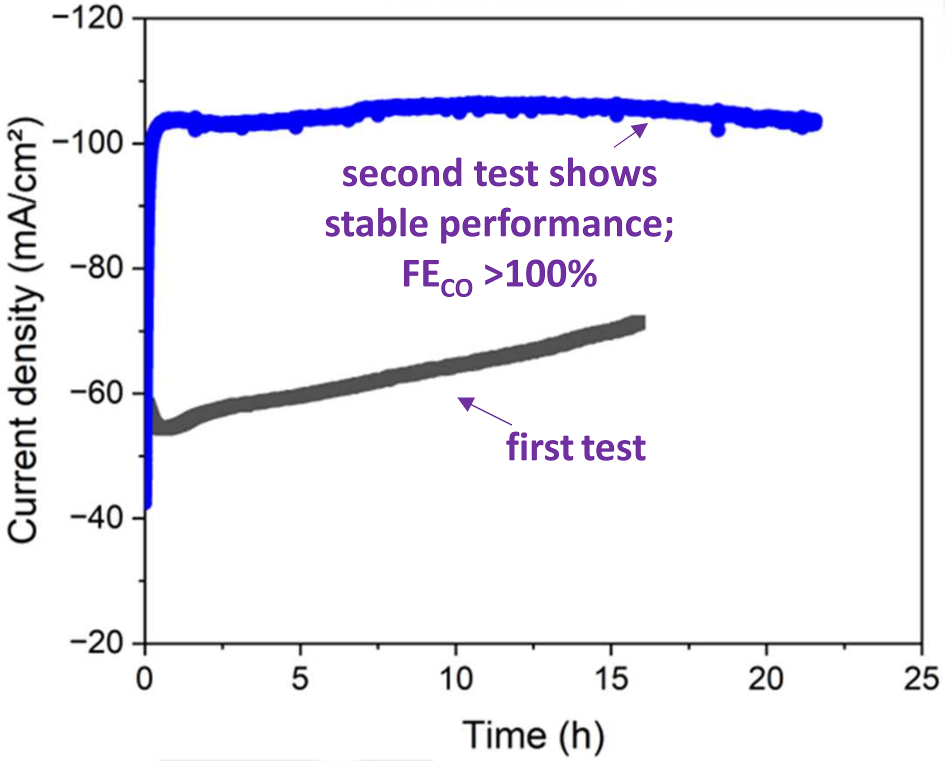


Current density change over time（25 cm^2 MEA cell）



MEAセル
大面積化

100 cm^2 MEA cell

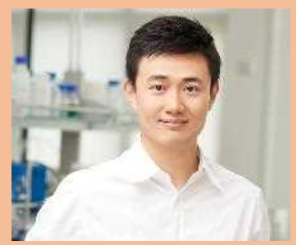


Global Trends in Direct Electrochemical Conversion of Air CO_2

CH_4 production from CO_2 gas mixed with O_2 is only a few examples in the world.

Moonshot challenge

competitor



Prof. Qi Lu
(Tsinghua Univ.)

Moonshot team

MOONSHOT
RESEARCH & DEVELOPMENT PROGRAM

	Benchmark	Our achievements
Source gas	**80%CO_2,20%O_2**	**40%CO_2,12%O_2**
cell	**Batch cell**	**Flow cell**
products	H_2, C_1, C_2	H_2, C_1, C_2
Current density	<6 $mAcm^{-2}$	CH_4 : **19.7 $mAcm^{-2}$** C_{2+}: **132 $mAcm^{-2}$**

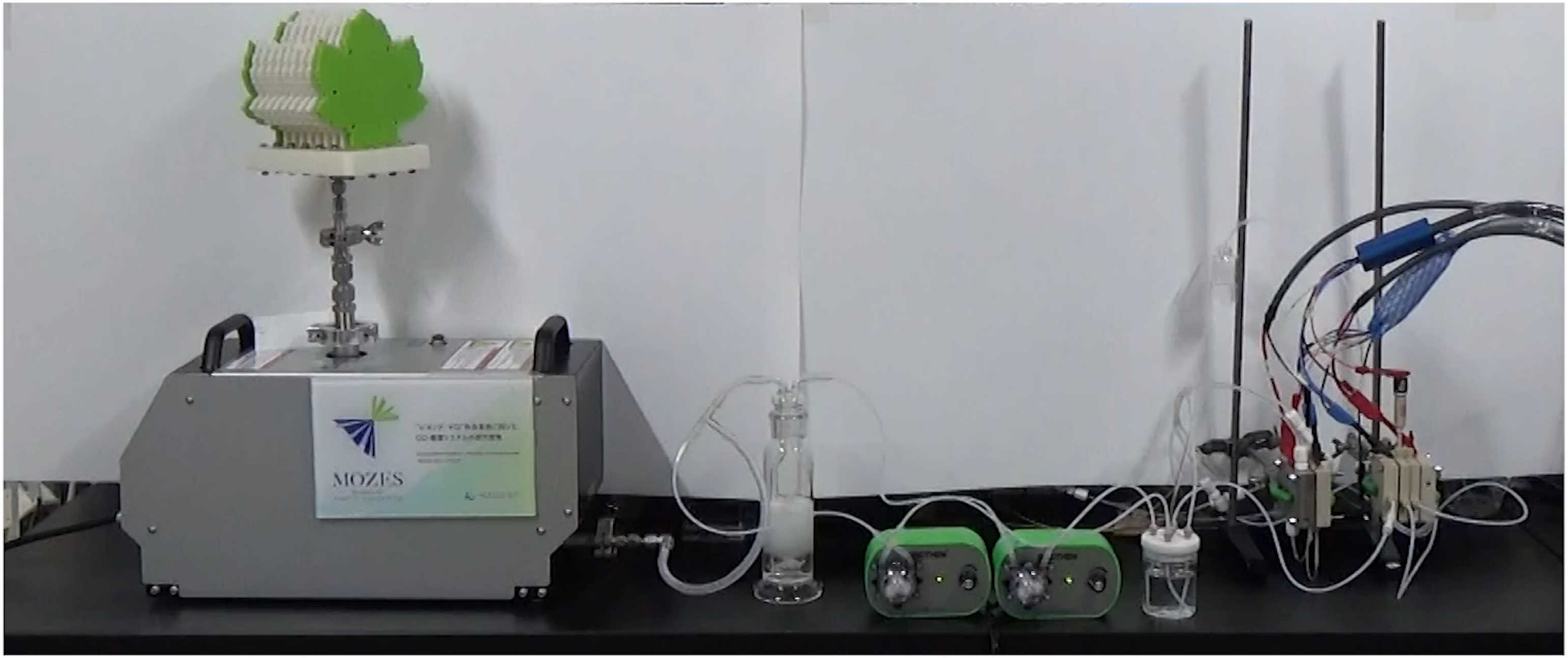
Nature Commun. **2022**, 11, 3844

C1 and C2 production at high current density even with low CO_2 feed gas concentration

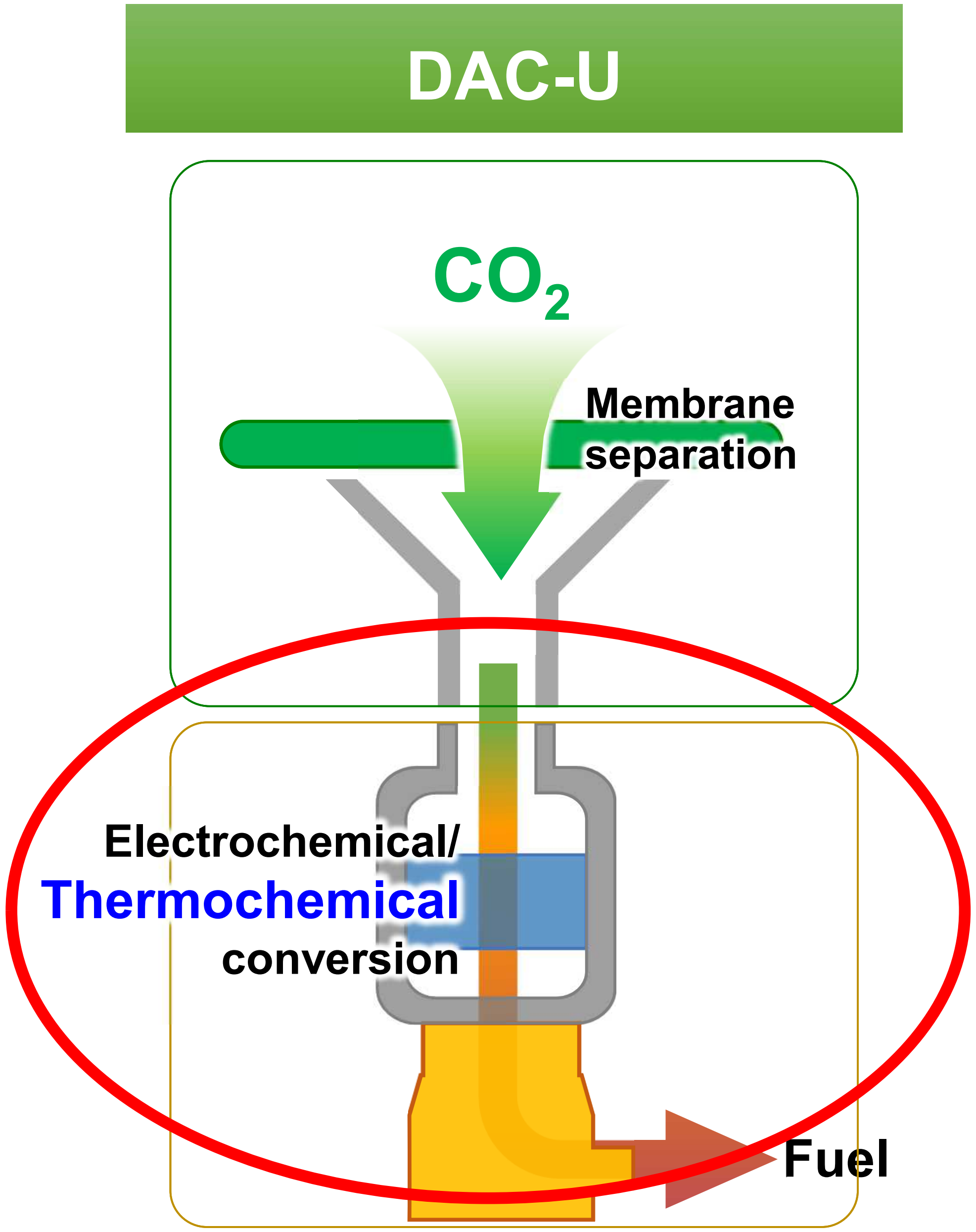
A

Continuous conversion by direct connection of DAC and electrochemical unit

Direct Air Capture and Utilization (DAC-U) system
~ m-DAC combined with electrochemical conversion system ~



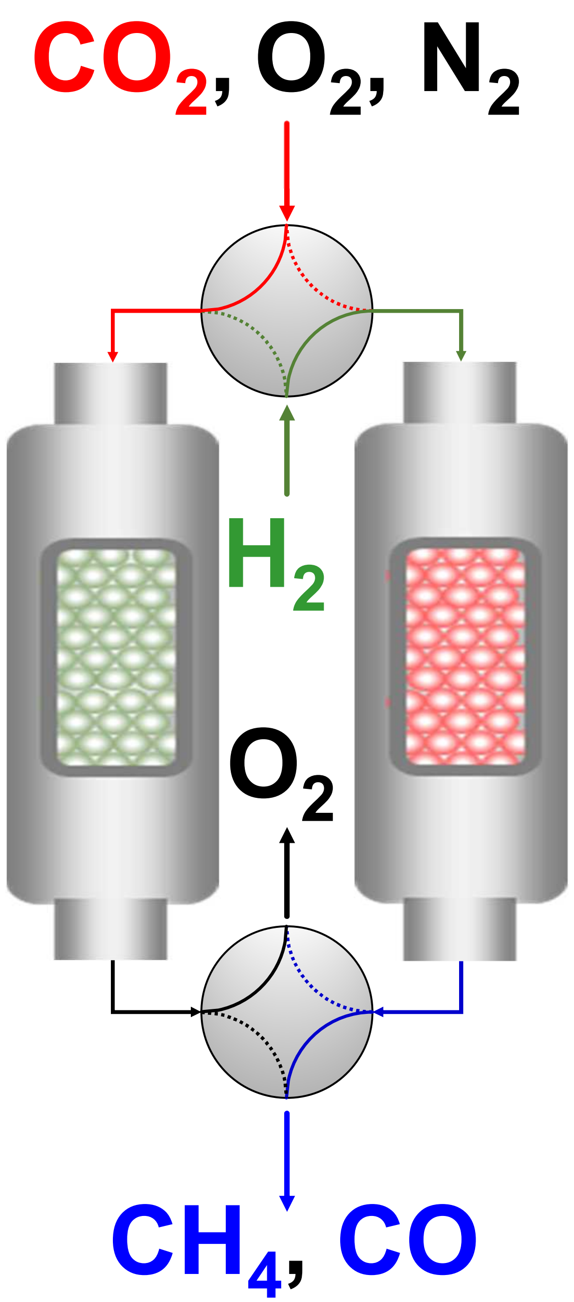
Development of a thermal conversion unit to produce carbon compounds from CO_2 mixture gas from DAC



Prof. Ken-ichi SHIMIZU (Hokkaido Univ.）

Conversion of CO_2 mixed gas from DAC to CH_4 and CO

Thermochemical production of CO, CH_4 from CO_2 mixed gas



High technological challenge

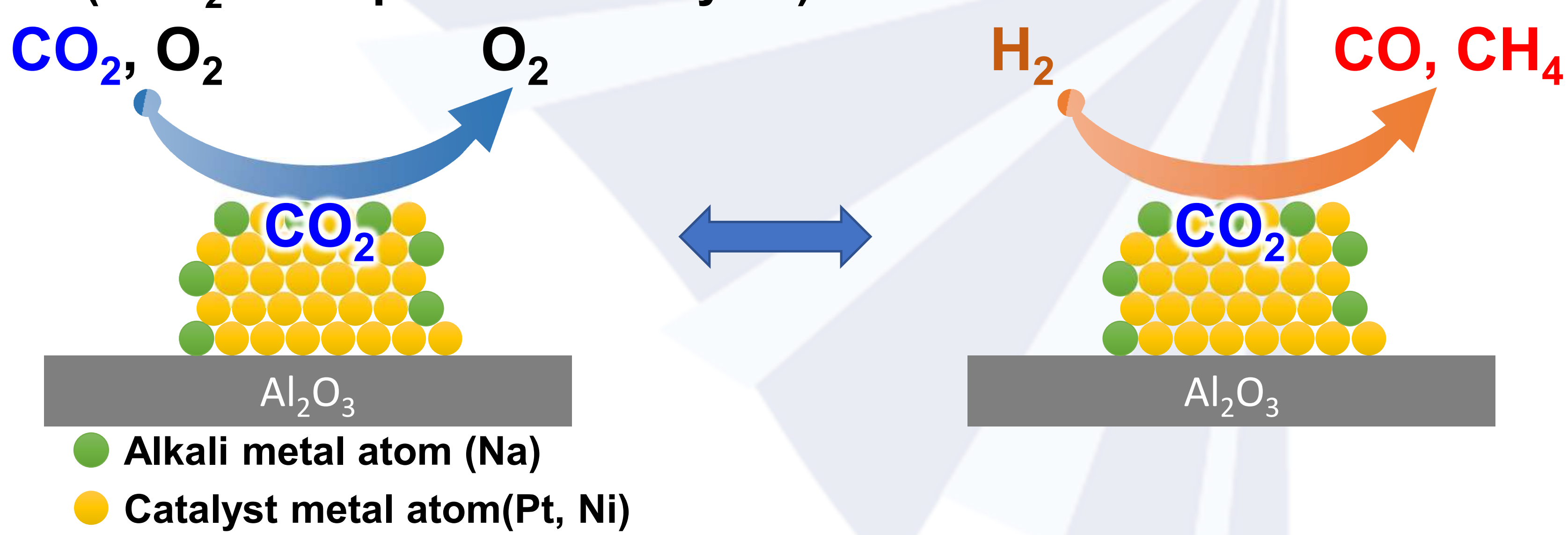
Producing CO,CH_4 from CO_2/O_2 mixed gas
→ there is almost no example in the world

H_2 does not react with O_2, but reacts with CO_2

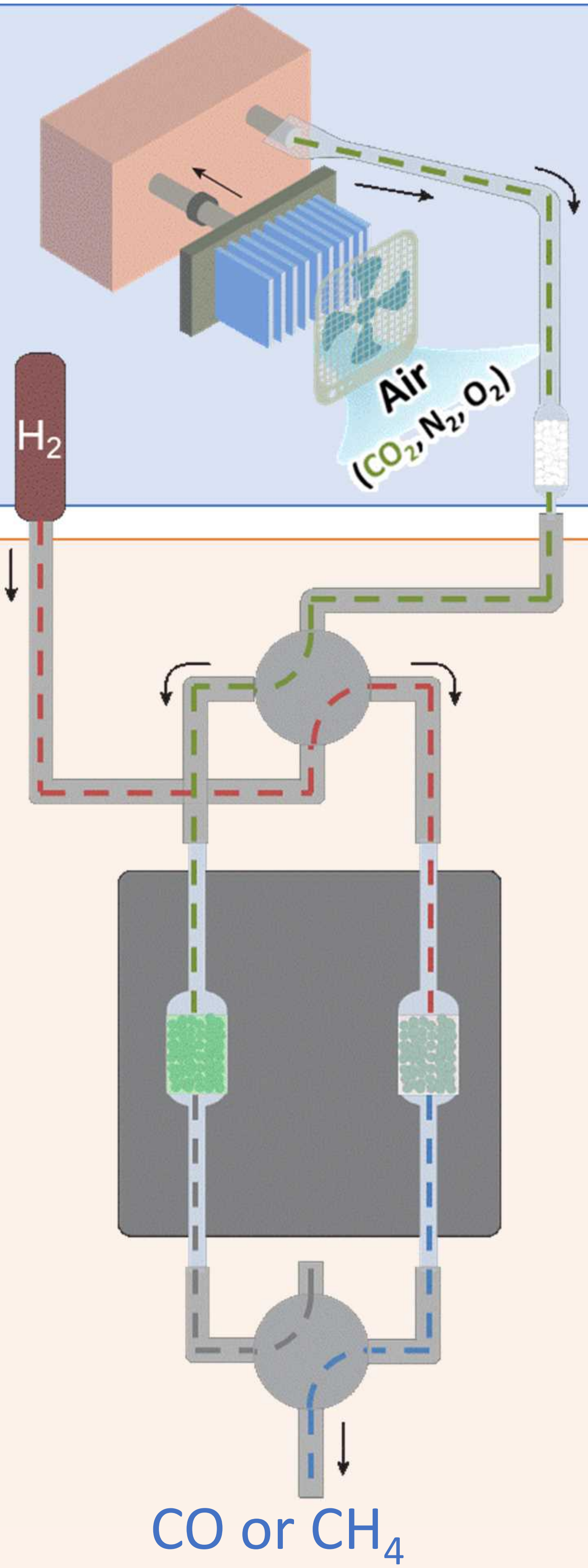
O_2 removal and CO_2 hydrogenation

■ O_2 removal
(+CO_2 absorption on catalysts)

■ hydrogenation



Construction of an integrated system by connecting DAC and thermo-chemical CO_2 conversion unit [24]



Direct Air Capture (DAC) by CO_2 capture nanomembranes

Enrichment of CO_2 from the air (400 ppm → 2000 ppm)

Continuous CO_2 adsorption & Reduction

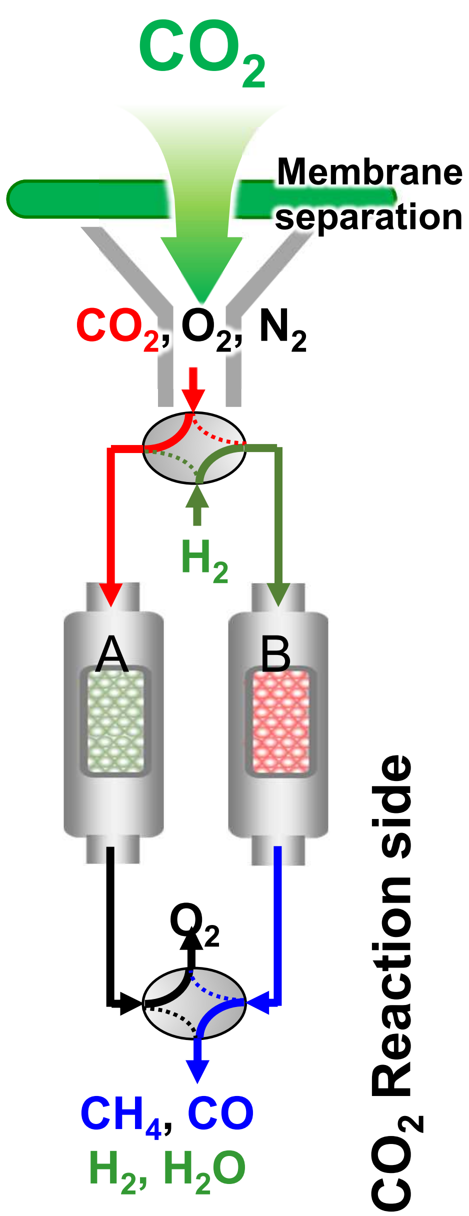
Advantages

✓Conversion of concentrated $CO_2/O_2/N_2$ mixtures to CH_4 and CO "directly" and in one step

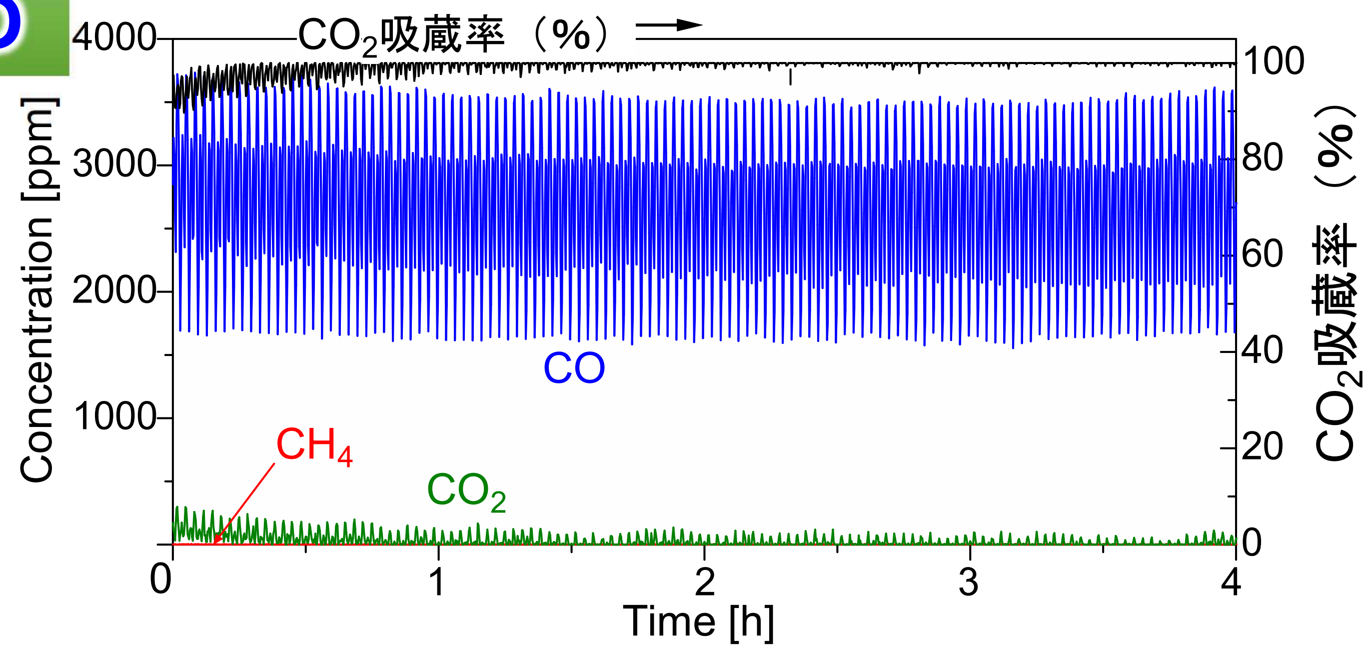
Challenges

✓Requires highly selective, low-temperature catalysts

Continuous production of CO, CH_4 by DAC-U system



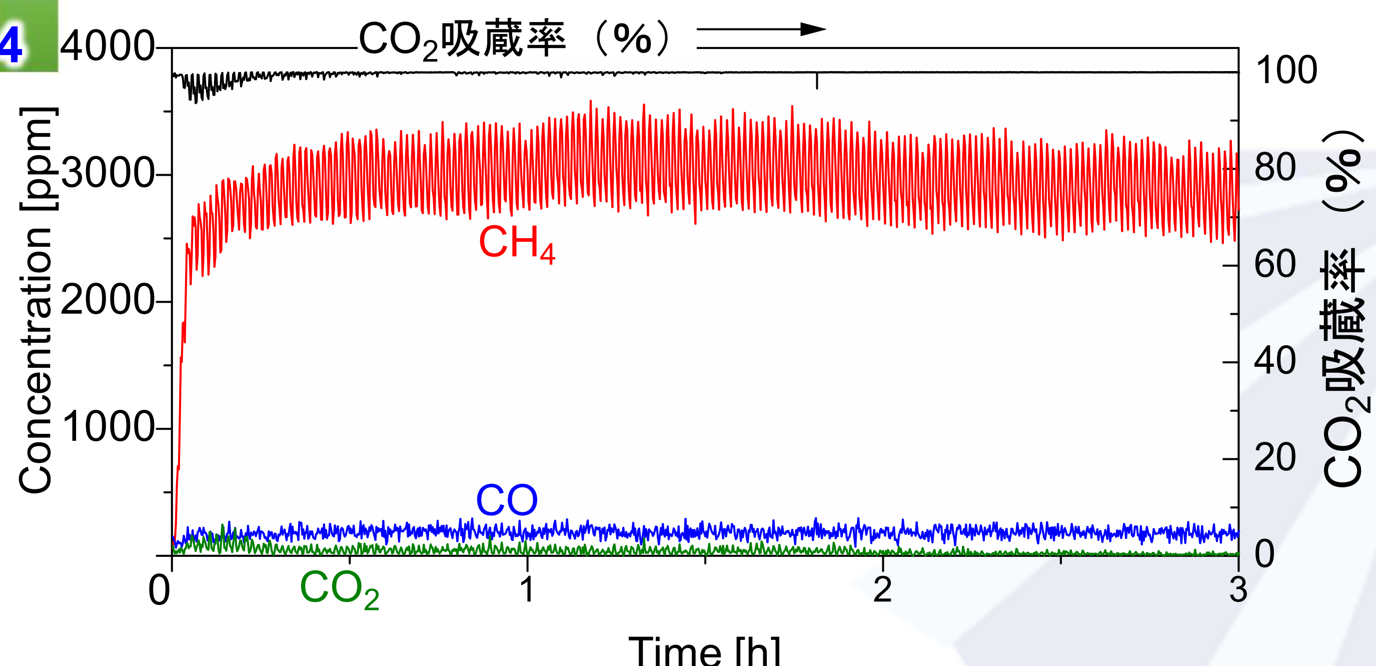
CO



selectivity>98%
Conversion rate >90%

Reaction condition
Catalyst amount：500 mg
Temp.：450°C
Source gas：100 mL/min
H_2 flux：100 mL/min
Valve change：every 30 sec

CH_4



selectivity>90%
Conversion rate>90%

Reaction condition
Catalyst amount：300 mg
Temp.：300°C
Source gas：500 mL/min
H_2 flux：100 mL/min
Valve change：every 1 min

2022 KPI

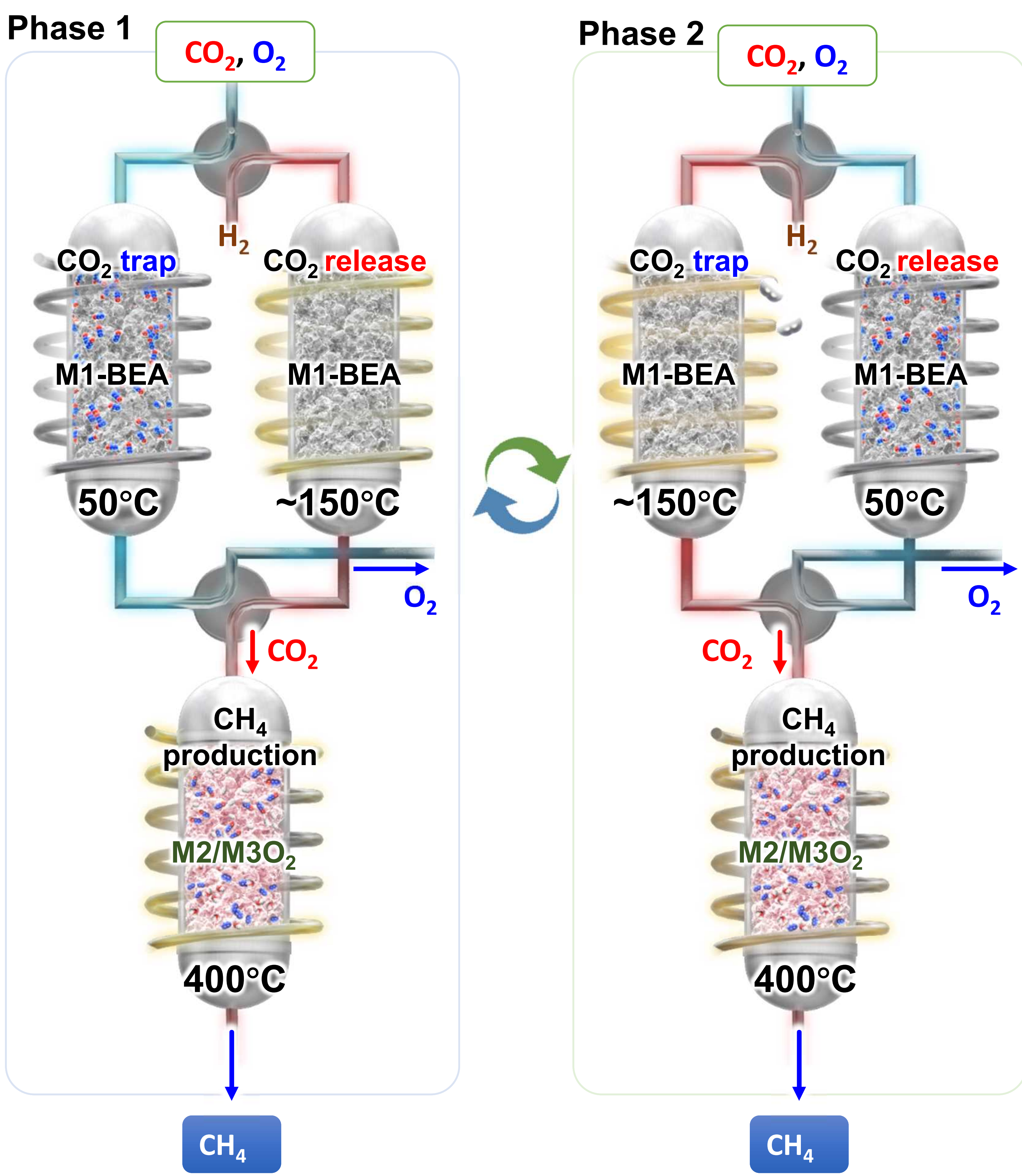
Achieved "Demonstration of CO and CH_4 conversion from CO_2 mixed gas

(Patent pending, paper in preparation)

Future task

- **Optimization of H_2 introduction conditions**
- **Recycling unreacted H_2**

Toward the CH_4 production with high concentration



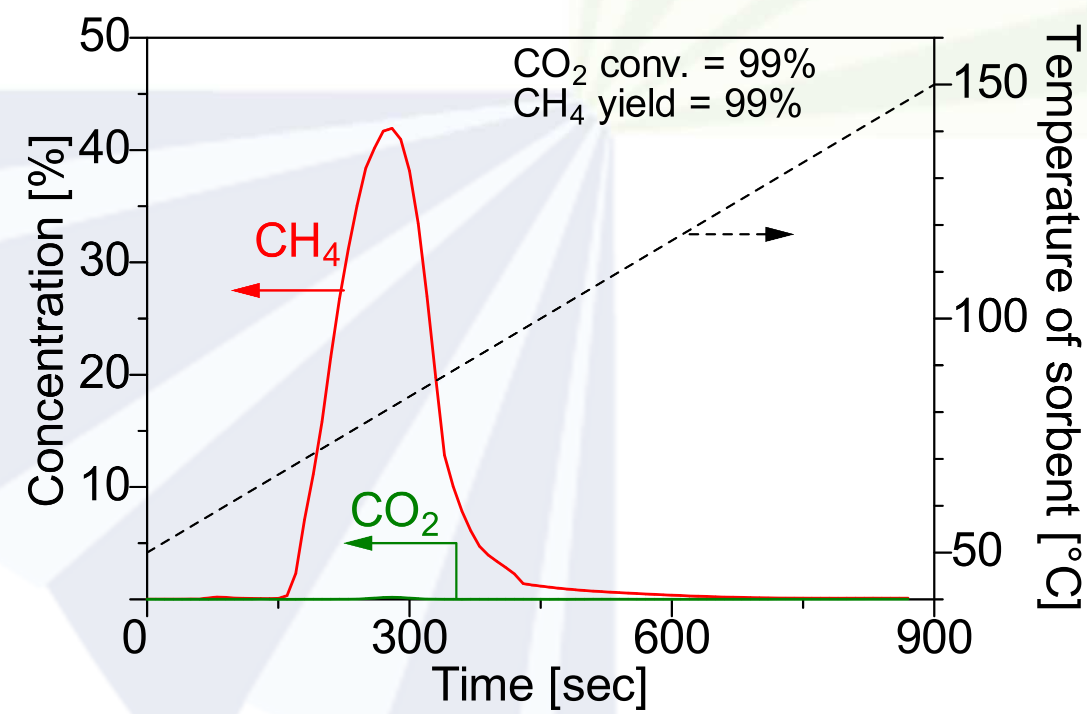
Reaction condition

1. O_2 removal

- CO_2 sorbent：M1-BEA Si/Al=5 (2.0 g)
- Gas
  - 10% CO_2/10% O_2/He (100 mL/min)
  - 100% H_2 (100 mL/min)
- Sorbent temp.：50 ºC～150 ºC
  heating rate : 10 ºC/min
- Switching time：15 min

2. CH_4 production

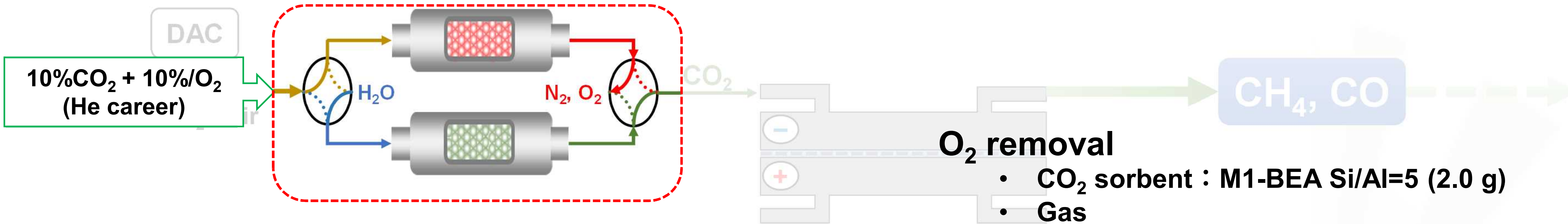
- catalyst：M2/M3O_2 3.0 g
- Reaction temp.： 400 ºC



High concentration CH_4 production from 10% CO_2/10% O_2

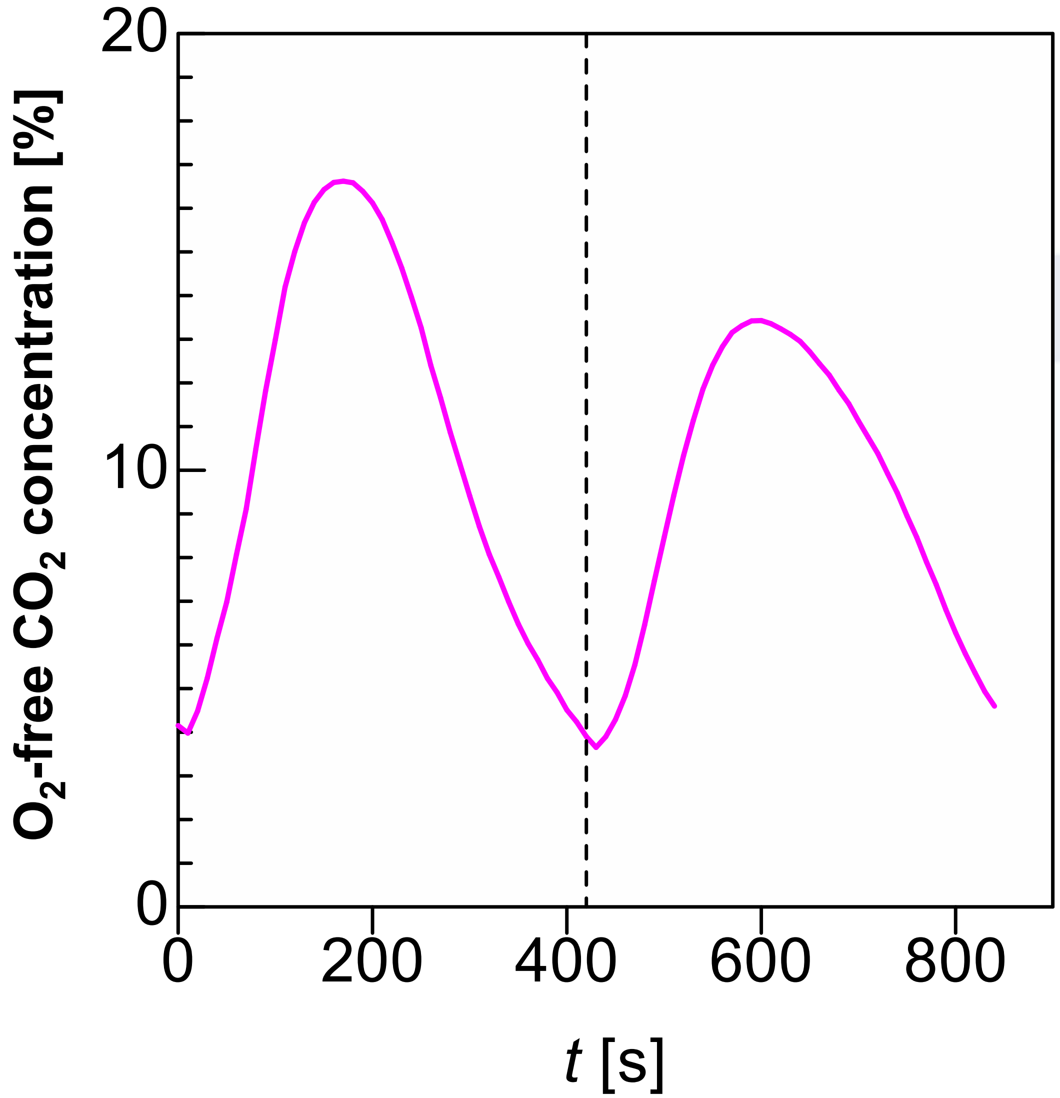
Lower temperatures for O_2 removal process

Type C:O_2 removal



O_2 removal

- CO_2 sorbent：M1-BEA Si/Al=5 (2.0 g)
- Gas
  - 10% CO_2/10% O_2/He (100 mL/min)
  - He (100 mL/min)
- Temp.：40 ºC～100 ºC
  Heating rate : 30 ºC/min)
- Switching：7 min



O_2-free CO_2 production from 10%CO_2/10%O_2

Coupled with electrochemical conversion

Candidate scenarios for social implementation of DAC-U

Features of DAC-U

- Compact and modular
- Decentralized distribution
- Size scalable

Utilization of small dispersibility: Methane →Use as fuel

System scale	application	CO_2 capture unit	product	conversion	
				with H_2 supply	no H_2 supply
Small to medium size	Residential houses small stores	1 ~ 2	CH_4	thermochemical	electrochemical
	Office building apartment building	multiple	CH_4	thermochemical	electrochemical

Utilize scalability: Assume CO, C_2H_2 →Use as industrial raw materials

System scale	application	CO_2 capture unit	product	conversion	
				with H_2 supply	no H_2 supply
Large	factory	massive	CO, C_2H_2	thermochemical	electrochemical

Activities for social implementation

June 23, 2023　Press release

sojitz
New way, New value

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双日、ナノ分離膜を用いたDAC技術の2020年代後半の実用化に向け新会社を設立

～九州大学発の革新的技術の社会実装を加速化～

PDF版[259.4 KB]

2023年6月12日
双日株式会社

双日株式会社（以下「双日」）は、2022年2月の九州大学との覚書締結を通じてDAC技術（membrane-based Direct Air Capture、以下「m-DAC™(*1)」）の2030年までの実用化に向け調査・研究を進めてきましたが、2020年代後半に社会実装を前倒しすべく新会社Carbon Xtract（カーボン エクストラクト）株式会社（以下「Carbon Xtract」）を設立しました。

地球温暖化対策として世界各国で2050年にCO_2排出ネットゼロを目指していますが、IEA(International Energy Agency/国際エネルギー機関)は「化石燃料などの消費抑制による排出削減で達成できるのは現時点の排出量の90%強で、2050年の排出ネットゼロには、2030年時点でDAC技術による7千万トン程度のCO_2の直接吸収が必要」と報告しています(*2)。この実現に向けて日本でも2023年5月12日にGX（グリーントランスフォーメーション）推進法が成立し、DACを始め脱炭素に関わる革新的技術の社会実装を後押しするための先行投資支援体制と市場整備が推し進められています。

Company	**Carbon Xtract Company**
President	Tetsuo Moriyama
Establishment	May 26, 2023
Business	Development and sales of equipment and products utilizing technology to selectively capture carbon dioxide from the atmosphere using separation nanomembranes

Nov. 24, 2023 press release

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Topics
トピックス

ナノ分離膜を用いた革新的CO_2回収技術を持つCarbon Xtract株式会社に九州大学初となる出資・事業参画

2023.11.24

トピックス

このたび九州大学は、九州大学発の実用開発中ナノ分離膜を用いた、大気からの直接的二酸化炭素（以下「CO_2」）回収技術（membrane-based Direct Air Capture、以下「m-DAC®(※1)」）と回収したCO_2の利活用技術の実用化に賛同・推進すべく、2023年5月に双日株式会社（以下「双日」）が主体となって設立した Carbon Xtract株式会社（以下「Carbon Xtract」）に、本学として初めての出資による事業参画を行います。

m-DAC®は、空気を膜でろ過するだけでCO_2を回収・濃縮するという世界で初めての革新的技術であり、これを装置化すれば様々な場所でのCO_2回収が可能になります。

分離膜によって、エアーフィルターのように大気からCO_2を回収・濃縮し、様々な有用物質に変える装置「Direct Air Capture and Utilization (DAC-U®)シ

First direct investment from Kyushu Univ.

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