

Review

Advancing low-carbon methane production via integrated carbon capture and in situ methanation (ICCM) by dual function materials (DFMs): Progress and prospects

Yi Heng Chan^{a,*}, Sylvester Yew Wang Chai^b, Bing Shen How^c, Chung Loong Yiin^{d,e,*}, Bridgid Lai Fui Chin^{f,g}, Serene Sow Mun Lock^{h,i}, Mee Kee Wong^a, Muhammad Khairumuzdaniel Md Khairulanwar^a, Shin Ying Foong^j, Su Shiung Lam^{j,k,*}

^a PETRONAS Research Sdn. Bhd. (PRSB), Lot 3288 & 3289, off Jalan Ayer Itam, Kawasan Institusi Bangi, 43000 Kajang, Selangor, Malaysia

^b School of Mechanics and Construction Engineering, Jinan University, Guangzhou 510632, Guangdong, China

^c Biomass Waste-to-Wealth Special Interest Group, Research Centre for Sustainable Technologies, Faculty of Engineering, Computing and Science, Swinburne University of Technology, Jalan Simpan Tiga, 93350 Kuching, Sarawak, Malaysia

^d Department of Chemical Engineering and Energy Sustainability, Faculty of Engineering, Universiti Malaysia Sarawak (UNIMAS), 94300 Kota Samarahan, Sarawak, Malaysia

^e Institute of Sustainable and Renewable Energy (ISuRE), Universiti Malaysia Sarawak (UNIMAS), 94300 Kota Samarahan, Sarawak, Malaysia

^f Department of Chemical and Energy Engineering, Faculty of Engineering and Science, Curtin University Malaysia, CDT 250, 98009 Miri, Sarawak, Malaysia

^g Centre of New and Sustainable Energy Research and Ventures (CoNSERV), Curtin University Malaysia, CDT 250, 98009 Miri, Sarawak, Malaysia

^h Centre for Sustainable Energy, Environment and Social Innovation (SEnSI), Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak, Malaysia

ⁱ Department of Chemical Engineering, Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak, Malaysia

^j Higher Institution Centre of Excellence (HiCoE), Institute of Tropical Aquaculture and Fisheries (AKUATROP), Universiti Malaysia Terengganu, 21030 Kuala Nerus, Terengganu, Malaysia

^k Department of Chemical Engineering and Materials Science, Yuan Ze University, Taoyuan, Taiwan

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ABSTRACT

Integrated carbon capture and methanation (ICCM) couples CO₂ capture with in-situ conversion by employing dual-function materials (DFMs) that integrate a CO₂ sorbent and a methanation catalyst in a single bed. This compact carbon-capture-and-utilisation concept offers smaller, more heat-efficient, and potentially more cost-effective CO₂ capture and conversion process compared to conventional approach. This review consolidates recent progress in ICCM with a critical emphasis on DFM design and synthesis (e.g., impregnation, co-precipitation, sol-gel, mechanochemical processing), structure–function relationships, and pros/cons of each DFM synthesis technique. We benchmark how composition, sorbent–catalyst proximity, and process conditions (namely temperature, pressure, gas hourly space velocity and gas impurities) influence CO₂ uptake, conversion, CH₄ selectivity, and long-term durability under realistic conditions. Techno-economic and life-cycle perspectives are summarised to contextualise the roles of H₂ cost, reactor integration, and DFM lifetime. Finally, we outline key scale-up challenges and research priorities, and propose three facets for technology commercialization outlook, including (i) DFM engineering, (ii) reactor design/configuration and (iii) technology integration.

1. Introduction

Rising atmospheric CO₂ and the need to decarbonise hard-to-abate sectors have intensified interest in scalable carbon capture and

utilisation/storage solutions (Yusuf and Ibrahim, 2023). Although post-combustion capture is technically mature, its overall energy penalty and the need for downstream CO₂ conditioning, transport, and storage infrastructure can increase cost and complexity. Converting

List of abbreviations: CCM, Carbon capture and (ex situ) methanation; CCU, Carbon capture and utilisation; DAC, Direct air capture; DFM, Dual function material; GHSV, Gas hourly space velocity; ICCM, Integrated carbon capture and (in situ) methanation; LCA, Life cycle assessment; LCOM, Levelized cost of methane; PtG, Power-to-Gas; SNG, Synthetic natural gas; TEA, Techno-economic analysis; TRL, Technology readiness level.

* Corresponding authors.

E-mail addresses: chan.yiheng@petronas.com.my (Y.H. Chan), cliyin@unimas.my (C.L. Yiin), lam@umt.edu.my (S.S. Lam).

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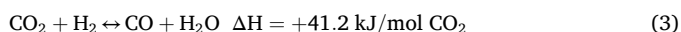
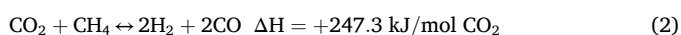
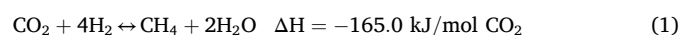
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captured CO₂ into storable fuels such as methane offers a pathway to valorise CO₂ while leveraging existing gas infrastructure (Smith et al., 2021). However, conventional ‘capture-then-convert’ schemes remain multi-unit processes. Process-intensified routes that integrate capture and conversion in fewer steps are therefore attracting increasing attention.

Integrated carbon capture and methanation (ICCM) is a process-intensified pathway in which CO₂ is first captured on a solid sorbent and subsequently hydrogenated to CH₄ (Eq. (1)) in the same reactor (typically under cyclic operation), minimising intermediate CO₂ release, compression, and handling. In a typical ICCM cycle, CO₂ is adsorbed/carbonated during a capture step and then transferred to adjacent catalytic sites during a reaction step where methanation proceeds. By combining capture and conversion, ICCM can reduce process footprint and potentially improve heat integration (Zhao et al., 2024a). Nevertheless, these benefits are realised only if the sorbent–catalyst system maintains high cyclic capacity, conversion, and stability over extended cycles under realistic operating conditions. Several studies have reported similar integrated capture-and-convert technique for production of various value-added products/chemicals, such as syngas (Eq. (2)) (Al-Mamoori et al., 2018), methane/synthetic natural gas (Wang et al., 2023), carbon monoxide (Eq. (3)) (Wang et al., 2022a), methanol (Jeong-Potter et al., 2024), etc.



DFMs enable ICCM by integrating (i) a CO₂ sorbent (often alkaline/alkaline-earth oxides or promoted supports) with (ii) a methanation catalyst (commonly Ni or noble metals) in close proximity (Omodolor et al., 2020). The exothermic nature of CO₂ methanation reaction provides heat for the endothermic CO₂ desorption, hence facilitating the mass transfer and CO₂ diffusion to the catalytic active sites for reaction with H₂ (Catarina Faria et al., 2023). High-performance DFMs require fast CO₂ uptake and release kinetics, accessible sorption sites, well-dispersed catalytic sites, and efficient interfacial transport of CO₂ from sorbent to catalyst during the reaction step. Design must also balance thermodynamics (carbonate stability), kinetics (surface reaction rates), and transport/thermal effects (diffusion limitations and hot-spots). Key durability challenges include metal sintering, pore blocking by carbonates, carbon deposition, and poisoning by sulfur/chlorine species present in practical gas streams.

Several reviews have discussed CO₂ methanation catalysts and broader carbon capture and utilization concepts, and a smaller number have addressed integrated capture-conversion at a conceptual level. For example, Lu et al. (2024) critically reviewed and discussed the design requirements and synthesis methods of DFMs, as well as the optimum operating conditions catered for various H₂ sources and conversion pathways. In addition, the performance of Ni-, Ru-, Rh-, and other metal-based DFMs, and the associated reaction mechanisms in integrated carbon capture and conversion processes were elucidated by Chen et al. (2022). Meanwhile, Shao et al. (2022) conducted a comprehensive review focusing on comparing reactor configurations and operating modes (i.e., dual-fixed-bed reactors with switching feed gas versus dual-fluidized-bed reactors with circulating DFM particles), providing critical insights towards realistic industrial demonstration of the process. Lastly, Sun et al. (2021a) analyzed in detail the influence of process parameters on the performance and stability of DFMs for three types of integrated capture and conversion processes, i.e., methanation, reverse-water gas shift, and dry methane reforming. However, a design-oriented synthesis for ICCM that systematically connects DFM synthesis routes and microstructure with cyclic performance metrics, realistic feed compositions, and scale-up constraints remains limited. In this review, we (1) map the ICCM/DFM literature and research trends

through bibliometrics, (2) compare DFM synthesis strategies and their structure–performance implications, (3) benchmark key operating parameters and cyclic performance metrics, (4) evaluate emerging and advanced ICCM techniques/technologies, (5) summarise techno-economic analysis (TEA) and life cycle assessment (LCA) insights, and (6) highlight practical challenges and propose way forward to guide translation beyond lab scale.

2. Bibliometric analysis of current literature on ICCM using DFMs

A bibliometric analysis was conducted to map research trends in ICCM and DFM-enabled capture–methanation. Publications were retrieved from Scopus using the query: “methane” OR “methane production” OR “CH₄” AND “ICCM” OR “in-situ methanation” OR “integrated carbon capture and methanation” OR “CO₂ capture” OR “carbon capture” OR “CO₂ adsorption” AND “dual-function” OR “dual function material”. A total of 94 documents were obtained. Documents published in languages other than English have been removed, and the remaining 93 documents comprising 85 articles and 8 reviews were used for further analysis. The documents were manually checked to ensure they are relevant to the subject matter. The final dataset of 93 documents was analysed using VOSviewer v1.6.20 by applying a minimum keyword occurrence threshold of 10 to quantify publication growth, dominant subject areas, leading journals/institutions, and keyword co-occurrence networks. The bibliometric results are discussed alongside technical developments to avoid purely descriptive trend reporting.

Fig. 1 shows the network visualization of keyword co-occurrence classification according to clusters. These clusters are: (1) catalyst or sorbent materials, (2) methanation performance with nickel catalysts, (3) integrated CO₂ capture, and (4) methanation reaction and process implementation. Cluster 1 presents keywords such as “alumina”, “ruthenium”, “sodium compounds”, “adsorption”, and “aluminum oxide”. This indicates a focus on dual-function catalyst-sorbent materials for ICCM. This cluster represents the development of integrated catalysts or support formulations that combine carbon capture and methanation functions within a single solid material, a key area of ICCM research. Cluster 2 contains terms like “nickel”, “nickel compounds”, “CH₄”, “carbon dioxide”, and “catalyst activity”. This cluster focuses on Ni-based methanation catalysts and their efficiency in converting CO₂ to methane, representing efforts to optimize methanation parameters (e.g., CH₄ yield, catalytic activity, and stability) for cost-effective Ni-based dual-function materials through material modification. These studies integrate fundamental catalysis with practical performance, ensuring efficient CO₂-to-CH₄ conversion in integrated processes. Cluster 3 consists of terms such as “CO₂ capture”, “CO₂ capture and utilization”, “direct air capture”, “performance”, and “Kyoto protocol”. This cluster investigates ICCM from a systems-level and environmental perspective, focusing on combining CO₂ capture technologies with methanation for climate mitigation and process efficiency. Lastly, Cluster 4 includes technical terms such as “methanation”, “hydrogenation”, “dual-function material”, and “flue gases”. This cluster corresponds to the reaction mechanism, kinetics, and the performance of DFMs for ICCM.

Fig. 2 illustrates the network visualization of keywords co-occurrence from 2022 to 2024. The evolution of ICCM research across time reveals a clear progression from fundamental catalyst development to system-level applications. Prior to 2022, research focused on validating the concept of ICCM, primarily using Ru- or Ni-based catalysts supported on adsorbent materials. From 2022 onward, the research focus shifted toward process enhancement and broadening applications, as shown by terms such as “direct air capture”, “performance”, and “Kyoto protocol”. These terms indicate a growing interest in enhancing the practical efficiency, durability, and scalability of ICCM systems. This transition reflects two key developments: (1) the extension of ICCM from flue gas treatment to ultra-dilute atmospheric CO₂ processing, and (2) increasing emphasis on techno-economic analysis and policy alignment.

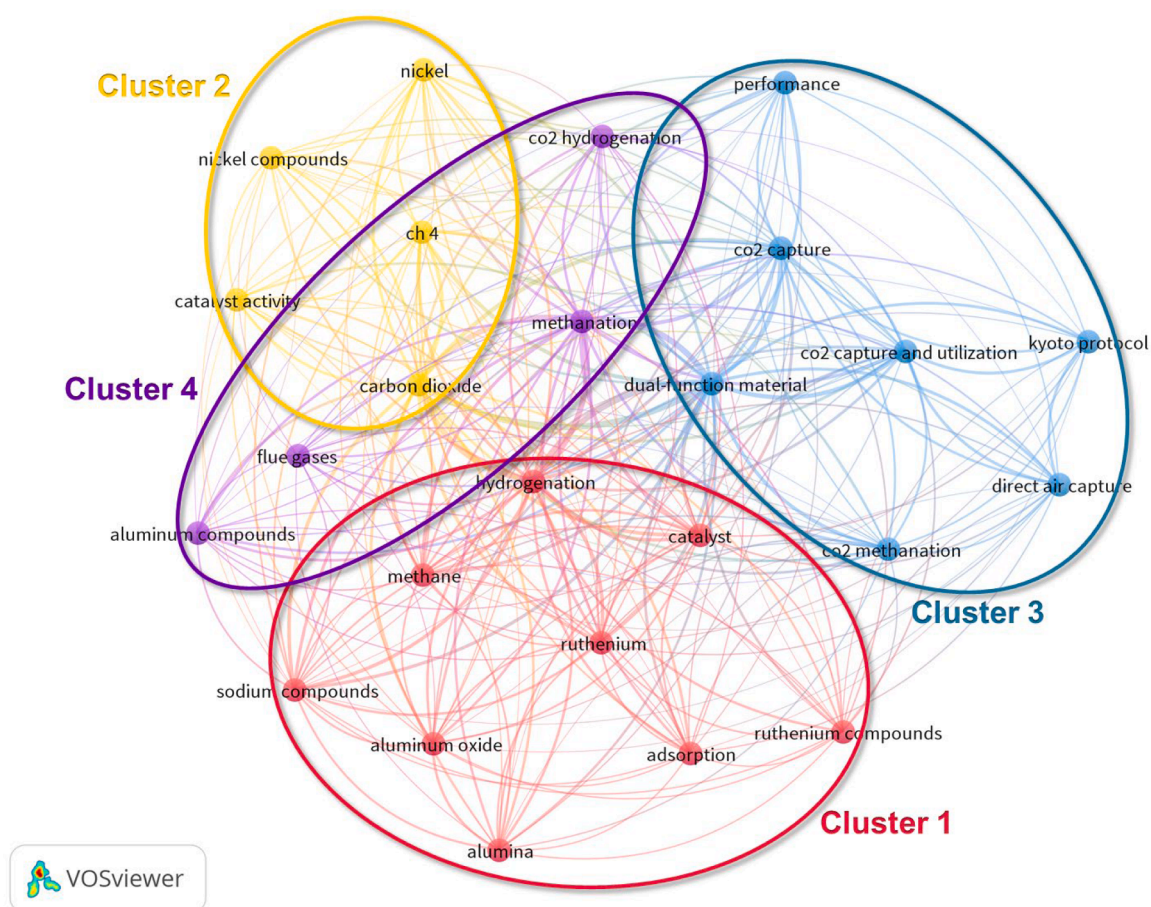


Fig. 1. Network visualization of keyword co-occurrence for integrated carbon capture and in-situ methanation using dual-functional materials according to clusters.

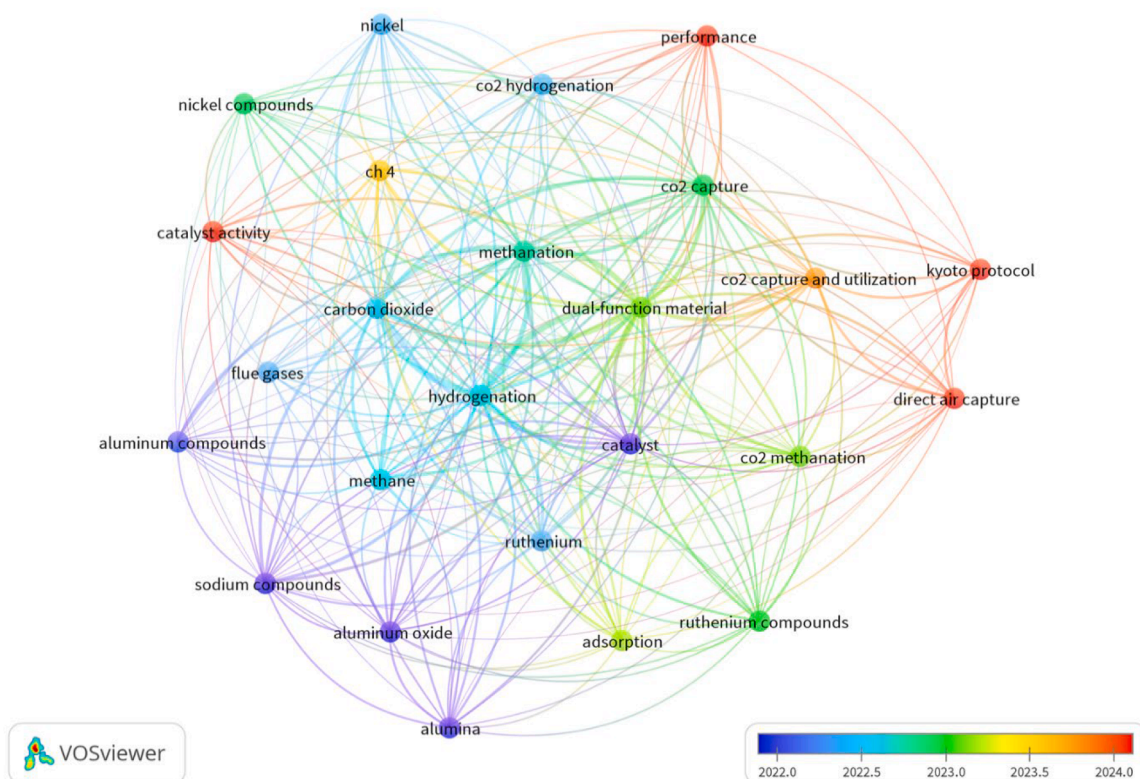


Fig. 2. Network visualization of keyword co-occurrence for ICCM using DFMs from 2022 to 2024.

Meanwhile, performance and techno-economic analyses are receiving increasing attention from researchers, focusing on conversion efficiency, process integration, energy consumption, and life-cycle assessments. The growing presence of policy-related terms also suggests a more substantial alignment between ICCM research and global decarbonization targets.

Current studies have largely focused on the synthesis of DFMs and their performance under ideal conditions. However, limited work has addressed long-term stability under realistic environments that include moisture, oxygen, and impurities. The combination of CO₂ capture and its conversion to methane is complex, as the important operating conditions are often examined separately rather than as part of a complete system. Reaction kinetics under cyclic operation remain unclear, and advanced ICCM strategies, such as direct air capture (DAC) and solar-assisted systems, are still in the early stages, with limited data on energy efficiency and regeneration. Techno-economic and environmental assessments are also quite limited in this field. These issues represent important research challenges and will be further discussed in this review with dedicated sections covering material synthesis, process optimization, reaction kinetics, emerging technologies, economic assessments, and sustainability perspectives.

3. Synthesis of DFMs for ICCM

DFMs have been synthesized via multiple routes, including impregnation, sol-gel, washcoating, mechanochemical, co-precipitation, and physical mixing methods, each with distinct synthesis mechanisms as illustrated in Fig. 3. These mechanisms influence dispersion quality and the resulting DFMs structural features, which in turn strongly affect CO₂ uptake and CH₄ yield of the produced DFMs. For example, impregnation generally provides excellent metal dispersion but is prone to pore clogging at high loadings, whereas co-precipitation can promote intimate mixing but limits precise particle size control. Mechanochemical synthesis offers a solvent-free pathway capable of generating DFMs with unique morphologies. However, it provides limited flexibility for structural tailoring and is energy intensive. In contrast, physical mixing

is more energy efficient but produces discrete phase DFMs with weak synergistic interactions. To aid readers, Table 1 links each synthesis route to their resulting DFMs structural features, reported cyclic metrics (i.e., CO₂ uptake and CH₄ yield) and key advantages and limitations. The following sections provide a detailed breakdown of each synthesis pathway, highlighting their key characteristics and impact on DFMs' performance.

3.1. Impregnation

Impregnation is one of the earliest and commonly adopted synthesis methods for DFM in ICCM applications. Its preference stems from the maturity, simplicity, and versatility of the process, which allows flexible integration of a wide range of precursors and supports. The impregnation process involves dispersing catalytically active components from a precursor containing solution onto a high surface area porous support (Hagemeyer and Volpe, 2005; Omodolor et al., 2020; Zhdanov and Kasemo, 2000). There are several impregnation routes, including incipient wetness impregnation, wet impregnation, co-impregnation, and direct impregnation. Among these, incipient wetness impregnation is the most widely adopted pathway due to its efficient precursor utilization and minimal agglomeration risk (Mehrabadi et al., 2017; Cimino et al., 2022; Jo et al., 2023).

One of the earliest works on impregnation synthesis of DFM for ICCM applications can be dated back to 2014, conducted by Janke et al. (2014) featuring the synthesis of 10 wt.% Ru/ γ -Al₂O₃ DFM via the incipient wetness impregnation route. The DFM was calcined at a relatively low temperature of 250 °C, highlighting an important design consideration for Ru-based DFMs, as high temperature calcination can induce the formation of volatile RuO₄, which could lead to partial loss of Ru loading. Notably, this early DFM design lacked a dedicated CO₂ adsorbent such as CaO and Na₂O, which are commonly incorporated into DFM today, and the CO₂ adsorption reaction relies solely on the Ru catalyst. This resulted in over a 50% reduction in CO₂ uptake after the first cycle due to the irreversible loss of active CO₂ adsorption sites and lack of reversible adsorption functionality that an adsorbent would

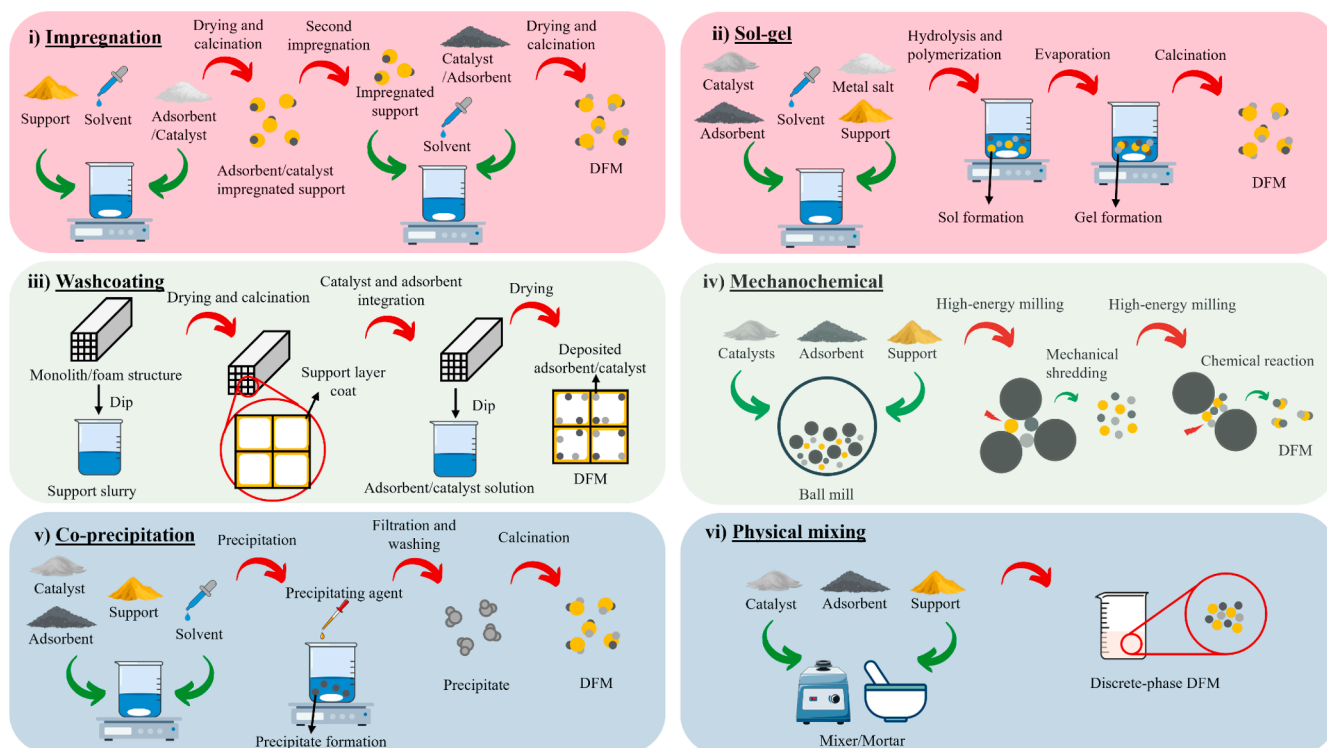


Fig. 3. Overview of dual-functional materials synthesis pathways.

Table 1

Summary of dispersion quality, structural features, advantages, and limitations of each DFM synthesis pathway.

Method		Dispersion Quality	Test Conditions	CO ₂ uptake, mmol CO ₂ /g _{DFM} ^a	CH ₄ yield, mmol CH ₄ /g _{DFM} ^a	DFM Structural Features	Advantages	Limitations	References
Impregnation	Incipient wetness impregnation	Low to moderate, with impregnation of support at an equal precursor solution volume to its pore capacity. The impregnation sequence may impact the dispersion quality.	<u>CO₂ capture</u> Temperature: 300 – 650 °C Pressure: 1 atm Gas: 5 – 10% CO ₂ /0 – 4.5% O ₂ /0 – 15% H ₂ O/0 – 100 ppmv SO ₂ /N ₂ <u>Methanation</u> Temperature: 300 – 650 °C Pressure: 1 atm Gas: 4 – 90% H ₂ /N ₂	0.13 – 0.68	0.21 – 1.05	Active phases are dispersed along the porous network.	• Efficient precursor utilization. • Minimal agglomeration risk during DFM synthesis. • Flexible precursors integration. • Close spatial proximity between adsorbent and catalyst.	• Powdered DFM tends to cause agglomeration, which leads to high operational pressure drop. • Often exhibit poor and uneven dispersion of precursors.	Mehrabadi et al. (2017); Arellano-Treviño et al. (2019a); Cimino et al. (2022); Jo et al. (2023); Duyar et al. (2015); Duyar et al. (2016)
	Wet impregnation	Moderate to high, with impregnation of support in excess solution that may enable deeper penetration and better dispersion. The impregnation sequence may impact the dispersion quality.	<u>CO₂ capture</u> Temperature: 250 – 650 °C Pressure: 1 atm Gas: 5 – 15% CO ₂ /0 – 10% O ₂ /0 – 10% H ₂ O/ N ₂ or Ar <u>Methanation</u> Temperature: 250 – 520 °C Pressure: 1 atm Gas: 5 – 100% H ₂ /N ₂ or Ar	0.07 – 14.10	0.06 – 8		• Flexible precursors integration. • Not dependent on capillary forces.	• Prone to clogging the support's pores due to impregnation under excess solution conditions. • Produce a low BET surface area DFM. • Powdered DFM tends to cause agglomeration, which leads to high operational pressure drop.	Arellano-Treviño et al. (2019a); Jo et al. (2023); Xiang et al. (2025); Sun et al. (2021b); Li et al. (2024a)
	Co-impregnation	Moderate to high, with impregnation of multiple precursors onto supports. Impregnation sequence and different combinations of precursors may impact the dispersion quality.	<u>CO₂ capture</u> Temperature = 280 – 650 °C Pressure: 1 atm Gas: 10% CO ₂ /Ar & CO ₂ /Ar 1/5 (vol%) <u>Methanation</u> Temperature = 280 – 520 °C Pressure: 1 atm Gas: 10% H ₂ /Ar	0.14 – 0.52	0.09 – 0.28		• Capable of integrating and promoting synergistic interaction between multiple precursors • Close spatial proximity between adsorbent and catalyst.	• Unable to control the dispersion of each precursor individually. • Powdered DFM tends to cause agglomeration, which leads to high operational pressure drop.	Arellano-Treviño et al. (2019a); Bermejo-López et al. (2023); Merkouri et al. (2022)
	One-pot impregnation	Moderate to high, with impregnation of multiple precursors onto supports in a single step. However, difficult to control the individual dispersion of each precursor.	<u>CO₂ capture</u> Temperature = 320 – 450 °C Pressure: 1 atm Gas: 7.5 – 10% CO ₂ /4.5% O ₂ /15% H ₂ O/N ₂ or Ar <u>Methanation</u> Temperature = 320 – 450 °C Pressure: 1 atm	0.59	0.58		• Simplicity, DFM can be synthesized within a single step.	• High batch-to-batch variance due to uncontrolled dispersion order. • Prone to encapsulation of active catalytic sites. • Powdered DFM tends to cause agglomeration, which leads to high	Arellano-Treviño et al. (2019a); Li et al. (2024a)

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Table 1 (continued)

Method		Dispersion Quality	Test Conditions	CO ₂ uptake, mmol CO ₂ /g _{DFM} *	CH ₄ yield, mmol CH ₄ /g _{DFM} *	DFM Structural Features	Advantages	Limitations	References
	Co-extrusion impregnation	High near the surface and low to moderate within the interior of the extrudate, with impregnation of precursors initiating at the surface and then migrating towards the interior of the extrudate.	Gas: 15 -30% H ₂ /N ₂ <u>CO₂ capture</u> Temperature: 180 – 380 °C Pressure: 1 atm Gas: 10 – 15% CO ₂ /N ₂ <u>Methanation</u> Temperature: 180 – 380 °C Pressure: 1 atm Gas: 5 – 100% H ₂ /N ₂	0.10 – 0.78	0.08 – 0.46	An enriched active layer on the surface of the extrudate.	- Produced DFM imposes a low operational pressure drop and easy handling. - High surface metal dispersion enhances CO₂ adsorption capacity. - High DFM stability. - Close spatial proximity between adsorbent and catalyst.	- operational pressure drop. - High dispersion variance. - Non-uniform dispersion can adversely impact CH₄ yield.	Liu et al. (2025); Catarina Faria et al. (2023)
Sol-gel	Standard sol-gel	High, with hydrolysis and polycondensation of a homogeneous precursor solution containing metal salts as a colloidal suspension, which ensures stable dispersion and uniform particle size in the sol.	<u>CO₂ capture</u> Temperature: 25 – 700 °C Pressure: 1 atm Gas: 400 ppm CO ₂ /21% O ₂ /N ₂ & 10% CO ₂ /10% H ₂ O/N ₂ <u>Methanation</u> Temperature: 400 – 700 °C Pressure: 1 atm Gas: 40% H ₂ /N ₂	7.40 – 16.22	4.70 – 14.94	Formation of a homogeneous polymeric network.	- Exhibit excellent operational stability. - Excellent phase integration.	- Long synthesis time. - May experience a large volume shrinkage of the resulting DFM.	Modan and Plăiașu (2020); Owens et al. (2016); Carter and Norton (2007); Jo et al. (2020); Feng et al. (2025)
	One-pot sol-gel		<u>CO₂ capture</u> Temperature: 550 °C Pressure: 1 atm Gas: 15% CO ₂ /N ₂ <u>Methanation</u> Temperature: 550 °C Pressure: 1 atm Gas: 100% H ₂	8.10 – 9.16	2 – 2.50		- Simplicity, DFM can be synthesized within a single step. - Close spatial proximity between adsorbent and catalyst.	- Synthesized DFM exhibits poor CH₄ yield and selectivity. - High risk of catalyst encapsulation after the CO₂ capture step.	Sun et al. (2021b); Yang et al. (2023); Wang et al. (2019)
	Pechini sol-gel	High, with the incorporation of chelation agent and glycols in the sol that promote greater porosity in support for enhanced dispersion of active sites.	<u>CO₂ capture</u> Temperature: 250 – 400 °C Pressure: 1 atm Gas: 10% CO ₂ / 10% O ₂ / 15% H ₂ O/Ar <u>Methanation</u> Temperature: 250 – 400 °C Pressure: 1 atm Gas: 10% H ₂ /Ar	0.58 – 0.88	0.41 – 0.47		- Produces DFM with higher porosity, enhancing dispersion of active sites. - Excellent phase integration.	- Long synthesis time. - Uses toxic reagents (i.e., glycol and chelation agent). - May experience a large volume shrinkage of the resulting DFM.	Modan and Plăiașu (2020); Carter and Norton (2007); Tsiotsias et al. (2024)
Washcoating	Foam washcoating	Low to moderate, with structured supports dipped into a	<u>CO₂ capture</u> ± <u>Methanation</u> Temperature:	N/A	CO ₂ conversion = 1.60 – 83.70%	Active phases anchored on the structure wall.	Exhibit excellent operational stability.	Synthesized DFM possesses low CO₂ conversion	Giani et al. (2006); Frey et al. (2016); Wang

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Table 1 (continued)

Method	Dispersion Quality	Test Conditions	CO ₂ uptake, mmol CO ₂ /g _{DFM} *	CH ₄ yield, mmol CH ₄ /g _{DFM} *	DFM Structural Features	Advantages	Limitations	References
Monolith washcoating	homogeneous suspension/slurry. Dispersion quality is dependent on slurry rheology and the type of structured support.	220 – 470 °C Pressure: 1 atm Gas: H ₂ /CO ₂ /N ₂ 36:9:10 (Molar ratio) & CO ₂ /H ₂ /N ₂ 1/4/1 (vol %)	0.13 – 1.06	0.03 – 0.56		• Provide excellent heat transfer capability. • Easier handling compared to powdered DFM.	efficiency and CH ₄ yield. • Foam possesses a limited surface area, which restricts active site availability. • Viscous washcoating solution may be prone to clogging the structure's pores.	et al. (2022b) ; Ma et al. (2019)
		<u>CO₂ capture</u> Temperature: 25 – 40 °C Pressure: 1 atm Gas: 400 ppm CO ₂ /air <u>Methanation</u> Temperature: 280 – 300 °C Pressure: 1 atm Gas: 15% H ₂ /N ₂				• High resistance against deactivation. • Exhibit excellent operational stability. • Produced DFM imposes a low operational pressure drop. • Provide excellent heat transfer capability. • Easier handling compared to powdered DFM.	• Synthesized DFM possesses low CO₂ conversion efficiency and CH₄ yield. • Viscous washcoating solution may be prone to the structure's pores.	Wang et al. (2022b) ; Lin et al. (2024) ; Abdallah et al. (2023)
Mechanochemical	High with precursors intensively mixed via ball milling, which generates high mechanical energy.	<u>CO₂ capture</u> Temperature: 200 – 700 °C Pressure: 1 atm Gas: 10% CO ₂ /Ar ₂ & 5 – 10% CO ₂ /15% O ₂ /N ₂ <u>Methanation</u> Temperature: 200 – 700 °C Pressure: 1 atm Gas: 5% H ₂ /Ar & 10 – 100% H ₂ /N ₂	0.10 – 7	0.03 – 0.73	Homogeneous nanostructured composite.	• Impart unique morphologies and configurations based on precursors combination that can significantly enhance ICCM performance. • Solvent-free technique. • Close spatial proximity between adsorbent and catalyst.	• Produces DFM with a relatively low BET surface. • Ball milling process is energy intensive. • Low flexibility to tailor DFM structure for potential catalytic enhancement.	Braga et al. (2025) ; Guo et al. (2024) ; Guan et al. (2025) ; Danielis et al. (2024) ; Sun et al. (2025)
Co-precipitation	High with co-nucleation of multiple precursors in an aqueous solution, where precipitating species adsorb and incorporate other precursors, resulting in homogeneous dispersion.	<u>CO₂ capture</u> Temperature: 180 – 440 °C Pressure: 1 atm Gas: 10 – 15% CO ₂ /N ₂ & 1:4:3 (CO ₂ : H ₂ : Ar) <u>Methanation</u> Temperature: 180 – 440 °C Pressure: 1 atm Gas: 1:4:3 (CO ₂ :H ₂ : Ar)	0.29 – 0.50	CO ₂ conversion = 35.21 – 86.02%	Homogeneous composite precipitate.	• Excellent phase integration. • DFM exhibits excellent CO₂ conversion efficiency and CH₄ selectivity.	• Prone to agglomeration, making it hard to achieve a controlled size.	Zhou et al. (2020) ; Li et al. (2023a) ; Fu and Lian (2024)

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Table 1 (continued)

Method	Dispersion Quality	Test Conditions	CO ₂ uptake, mmol CO ₂ /g _{DFM} *	CH ₄ yield, mmol CH ₄ /g _{DFM} *	DFM Structural Features	Advantages	Limitations	References
Physical mixing	Low with precursors only undergoing physical mixing without any chemical interactions or generating mechanical energy.	CO ₂ capture Temperature: 450 – 550 °C Pressure: 1 atm Gas: 10 – 15% CO ₂ /N ₂ Methanation Temperature: 450 – 550 °C Pressure: 1 atm Gas: 30 – 100% H ₂ /N ₂	0.10 – 15.30	0.10 – 8	Discrete and separated phases.	• Simplicity. • Does not rely on chemical interaction and bonding. • Less prone to encapsulation of active sites.	• Long distance between catalysts and adsorbents. • Poor phase integration. • Synthesized DFM often exhibits inconsistent operation performance.	Sun et al. (2021b); Li et al. (2024a); Heo et al. (2025); Sun et al. (2020)

* CO₂ adsorption and CH₄ production capacity are highly dependent on operating conditions, precursor types, and their loading during DFM synthesis.

provide.

Based on this finding, Duyar et al. (2015) extended this work by incorporating CaO adsorbent into the system. The resulting 5 wt.% Ru, 10 wt.% CaO/ γ -Al₂O₃ DFM showed significantly improved and more stable CO₂ uptake capabilities, with some cycles exhibiting consistent capacity ranging from 0.30 to 0.60 mmol CO₂/g_{DFM}. Interestingly, despite using only half the Ru loading of the 10 wt.% Ru/ γ -Al₂O₃, the DFM demonstrated markedly improved methanation performance of up to five-fold increase in CH₄ yield (0.50 mmol CH₄/g_{DFM}) relative to the baseline (0.10 mmol CH₄/g_{DFM}), albeit the absolute CH₄ yield was still rather low. This highlights the key advantage of DFM over conventional single functional material in ICCM operation, where synergistic interaction between catalyst and adsorbent couples exothermic methanation with CO₂ desorption, promoting sequential capture and conversion. This study also compares DFM synthesis via the impregnation process with the physical mixing method, and it is shown that the former exhibits a higher CH₄ yield per Ru site. This is because impregnation can impart closer spatial proximity between the Ru catalyst and CaO adsorbent on the shared Al₂O₃ support. It was also reported that the impregnation sequence significantly influences DFM performance. When Ru was impregnated onto CaO/ γ -Al₂O₃, the CH₄ yield improved by 53% compared to the reverse order. This is attributed to the enhanced dispersion of the Ru catalyst over CaO-containing support during the impregnation process. On the flip side, impregnating CaO over Ru/ γ -Al₂O₃ would mask active Ru sites, decreasing methanation efficiency. Despite this, some studies still adopt the latter route (Cimino et al., 2022; Cimino et al., 2025), but the importance of proper sequencing is now widely adopted in various works (Duyar et al., 2016; Jeong-Potter et al., 2022a; Sapru et al., 2025).

Arellano-Treviño et al. (2019a) found that the incorporation of different precursors in impregnation plays a significant role in the final properties of DFM. Building on these insights, several studies have explored the co-impregnation of multiple catalysts, adsorbents, or even supports into a single DFM system (Jo et al., 2023; Xiang et al., 2025; Cimino et al., 2025). For example, Merkouri et al. (2022) impregnated both Ni and Ru catalysts as well as CeO₂ and Al₂O₃ supports into a single DFM. It is shown that the Ni-Ru, Na₂O/CeO₂-Al₂O₃ DFM exhibited a 0.09 and 0.12 mmol CO₂/g_{DFM} higher CO₂ uptake than the Ni, Na₂O/Al₂O₃ and Ru, Na₂O/CeO₂ system, respectively. This is due to the synergistic interaction of the integrated materials, where Ru helps in Ni dispersion, providing a closer proximity between adsorbents and catalysts while CeO₂ provide excellent redox properties and acts as a CO₂ promoter to the stable but rather inactive Al₂O₃ support (Bermejo-López et al., 2023; Merkouri et al., 2022).

Despite the superiority of incipient wetness impregnation (or dry impregnation route) over wet impregnation for DFM synthesis, some

studies still adopt the latter process (Mehrabadi et al., 2017). This is primarily due to the immersion of the support in excess precursor solution, which can facilitate better materials diffusion and dispersion during the impregnation stage (Jiao and Regalbuto, 2008). Unlike dry impregnation, wet impregnation is not dependent on capillary action, making it more suitable for supports that possess limited capillarity or strong adsorption properties (Ravanchi et al., 2018). However, the excess liquid also promotes precursor migration during drying, increasing the likelihood of pore blockage and particle agglomeration, which often results in lower BET surface area and inferior catalytic performance compared with incipient wetness impregnation (Cimino et al., 2022; Jo et al., 2023; Xiang et al., 2025). Despite the lower Ni and CaO loading in the 10 wt.% Ni, 10 wt.% CaO/Al₂O₃ DFM prepared by incipient wetness impregnation (Chai et al., 2020), it still exhibited 87 to 328% higher CO₂ uptake and 100% higher CH₄ yield compared to the 15 wt.% Ni, 15 wt.% CaO/Al₂O₃ (Bermejo-López et al., 2019) and 10 wt.% Ni, 15 wt.% CaO/Al₂O₃ (Kosaka et al., 2021) DFMs prepared by wet impregnation with higher adsorbent and catalyst loadings. However, there is still a lack of systematic comparison between these two methods in terms of their impact on the DFM morphology and performance at various ICCM operating conditions, highlighting a key direction for future research.

More recently, impregnation has been integrated with co-extrusion to address high-pressure drop issues and for easier handling of DFM. While traditional impregnation produces powder DFMs that are prone to agglomeration and harder to handle, co-extrusion produces DFMs in specific shapes that overcome these issues (Catarina Faria et al., 2023). Additionally, impregnation of co-extruded materials also enables unique dispersion behaviour, which can be beneficial for certain applications. In 2023, Catarina Faria et al. compared extrudate impregnation against conventional powder impregnation and found that impregnated extrudates showed higher surface concentration but higher catalyst dispersion variance. This resulted in superior CO₂ uptake (0.78 mmol CO₂/g_{DFM}) as compared to that of powder impregnation (0.69 mmol CO₂/g_{DFM}) due to more basic sites on the surface with comparable CH₄ yield (0.46 to 0.49 mmol CH₄/g_{DFM}) but CH₄ selectivity suffered due to non-uniform dispersion, with higher concentration externally compared to internally.

In a more recent study, Liu et al. (2025) compared co-extruded impregnated DFMs (1 wt.% Ru, 20 wt.% Na₂CO₃/ γ -Al₂O₃ (RuNaAl) and 2 wt.% Ni, 20 wt.% Na₂CO₃/ γ -Al₂O₃ (NiNaAl)) with their physically mixed and conventionally impregnated counterparts. The co-extruded DFMs showed superior CH₄ yield, outperforming physically mixed DFMs by 107% to 137% and conventionally impregnated ones by 9% to 15%. Notably, the co-extruded RuNaAl and NiNaAl samples achieved CH₄ yields of 0.31 and 0.09 mmol CH₄/g_{DFM}, respectively, at a low

temperature of 220 °C, while physically mixed and conventional impregnated DFM produced negligible CH₄ at the same temperature. This allows the co-extruded samples to exhibit exceptional stability in this temperature range, as metallic sintering is less prone to occur. The superior performance of the co-extruded samples is attributed to their enhanced spatial proximity (<10 μm) between the adsorbents and catalysts within the DFM, which is significantly shorter than their respective counterparts (> 200 μm).

In contrast to all the multi-step impregnation methods discussed so far, Li et al. (2024a) have examined a one-pot approach, which integrates all materials within a single step. Although this method offers a simplified synthesis process, the resulting DFM exhibited inferior performance compared to its multi-step impregnated counterpart. This is primarily due to the proneness of the one-pot method to cause encapsulation of active catalytic sites. This is evident by the significant drop in CO₂ uptake and CH₄ yield from 1.79 to 0.59 mmol CO₂/g_{DFM} and 1.78 to 0.58 mmol CH₄/g_{DFM}, respectively, when compared to DFM synthesized via the sequential impregnation route.

3.2. Sol-gel method

The sol-gel method is a type of wet chemistry technique that has been widely employed for advanced materials production, owing to its versatile nature, which allows chemical modification of the precursor at the molecular level (Dave and Lockwood, 2012). In other words, one can produce tailored nanoparticles with the desired properties, size, and shape (Han et al., 2024); and with fine dispersion through its polymer matrix (Branda et al., 2023). In general, it involves four steps which begin with “sol” formation (i.e., forming an aqueous precursor solution that contains metal salts, acting as a colloidal suspension); followed by hydrolysis and polycondensation, which transform precursors into a polymer; and finally ends with calcination to remove the solvent (Mushaq and Sathyakam, 2025). To date, this method has been applied to generate various DFM, including Al, Zr, and Ce modified Fe, CaO (Gao et al., 2025), Ni-promoted Fe, CaO (Sun et al., 2022a), and FeNiMn, CaO (Zhao et al., 2024b) for CO formation; CaO-based sorbent for ethylene production (Zhang et al., 2023); cobalt oxide nanoparticles for water purification (Dakave et al., 2024); while this method has also been used in the ICCM applications.

Back in 2020, a group of Korean institutions proposed a novel ICCM process using 9.3 wt.% Ni, CaO DFM derived from sol-gel method (Jo et al., 2020). The resultant DFM exhibited a decent BET surface area of about 14.0 m²/g, capable of achieving excellent CO₂ uptake, CH₄ yield and CH₄ selectivity of 15.49 mmol CO₂/g_{DFM}, 14.94 mmol CH₄/g_{DFM} and 96%, respectively, at 600 °C. More interestingly, the study also revealed that the sol-gel derived DFM can exhibit long-term stability of the yielded DFM, in which the resultant CO₂ uptake remains stable (ranging between 60.2% to 70.5%). A year later, Sun et al. (2021b) proposed a one-pot sol-gel approach to synthesize a similar Ni-based DFM. Despite having a decent CO₂ uptake of 9.16 mmol CO₂/g_{DFM}, the respective CH₄ yield and selectivity were merely 2.0 to 2.5 mmol CH₄/g_{DFM} and 54% to 69%, respectively, depending on the Ni loading.

More recently, Feng et al. (2025) proposed the use of Ca(OH)₂ as an alternative sorbent to replace CaO so that the methanation can perform better at a lower temperature region (i.e., 400 °C), resulting in a greater CO₂ uptake of 8.96 mmol CO₂/g_{DFM} and CH₄ yield of 8.60 mmol CH₄/g_{DFM}. To enhance its stability, metal doping of ZrO₂ can be considered (drop in CO₂ conversion after 5 cycles reduced from 6% to 3%). Apart from Ni-based DFM, Tsiotsias et al. (2024) explored the potential of Ru-based DFM (1.5 wt.% Ru, 12 wt.% Na₂O/Al₂O₃) for ICCM application. In that work, the impact of two sol-gel method was investigated. Generally, the Pechini sol-gel method that includes glycols on top of the chelating agent (i.e., citric acid) can offer greater porosity in the support, causing a better dispersion of active sites. This results in an overall greater CH₄ yield, i.e., 2% and 13% higher compared to that

of DFM generated using the normal sol-gel method and precipitation approach.

3.3. Washcoating

Washcoating method is often being misinterpreted as impregnation due to their similar preparation nature (Jeong-Potter et al., 2022b). Unlike impregnation, the former coats a pre-formed slurry or viscous suspension of active material onto a structured support, such as monoliths and foams (Lin et al., 2024). Similar to impregnation, it also offers high flexibility in the DFM synthesis, capable of integrating diverse precursors and support materials.

The use of washcoating in material synthesis can be traced back to 1995 (Xiaoding et al., 1995) but its adoption for ICCM application only gained traction in 2016. Frey et al. (2016) integrated dual Ni-Ru catalyst onto ceria-zirconia (CZ) washcoated silicon carbide (SiC), aluminium (Al), and alumina (Al₂O₃) open cell foams for CO₂ methanation. Among the three washcoated DFM, Ni-Ru/CZ-SiC exhibits the highest CO₂ conversion value of 7.8% followed by Al₂O₃ and Al at 3.7% and 2.3%, respectively. These low conversion values are tied to the inherently low surface area of the foam supports, which limits the overall BET surface area of washcoated DFM (<33.8 m²/g) (Giani et al., 2006). While foam structures offer good stability, their limited surface area restricts active site availability and weaker catalytic activity. Moreover, the more viscous nature of washcoating solutions can also block the foam's pores and lead to poor dispersion or agglomeration of the catalyst, further reducing the effective surface area (Wang et al., 2022b). Additionally, the DFM lacks reversible adsorption functionality due to the absence of an actual adsorbent. Despite the relatively low conversion value, these DFM exhibit excellent CH₄ selectivity of up to 91.3%, 89.9% and 83%, respectively.

To enhance the CO₂ conversion efficiency, Ma et al. (2019) integrated graphene oxide onto Ni-SiO₂/Ni-foam. This study shows that graphene oxide contributes positively to the metal-support interaction by serving as a connective interface between the Ni-SiO₂ catalyst and the Ni foam. The formation of Ni-SiO₂ on graphene layer facilitated a high dispersion of Ni sites, leading to an enhanced CO₂ conversion of up to 83.70%, compared to 66% for the counterpart without graphene.

Aside from foam washcoating, Abdallah et al. (2023) washcoated 1 wt.% Ru, 10 wt.% Na₂O/γ-Al₂O₃ onto ceramic monoliths. This washcoated ceramic monoliths shows a CO₂ uptake of 1.06 mmol CO₂/g_{DFM} with a CO₂ conversion efficiency of 58%, comparable to that of washcoated foam DFM. A critical finding from this study is the excellent stability of the washcoated monolith DFM, which exhibits stable operation of up to 100 cycles (450 hours) under varying conditions, such as dry or humid conditions. This is tied to the structured monolithic DFM design, which mitigates pressure drop, a common issue in conventional impregnated DFM operation.

All the abovementioned washcoated DFM exhibit excellent CH₄ selectivity due to Ru content. However, since Ru is an expensive noble metal, thriftiness its loading to low levels is essential for the commercialization of these DFM. To this end, Lin et al. (2024) extended the previous work by reducing the Ru content from 1% to 0.25%. Despite this 75% reduction in Ru loading, the washcoated DFM is able to maintain its high stability, proven by the absence of major deactivation after aging performance tests. It is also shown that after even 250 hours of operation, the Ru crystalline size only shows a minimal increase from 2 nm to approximately 25 nm. However, reducing Ru loading may compromise ICCM performance; thus, optimal reduction is necessary to maintain efficiency of DFM. Overall, this study indicates that washcoated monolith DFM design can minimize sintering or agglomeration, allowing the system to maintain low pressure drop and favourable heat transfer.

Further supporting the low-pressure drop advantage, García-Bordejé et al. (2026) demonstrated that Na-Ru washcoated cordierite monoliths exhibit comparable operational efficiency to their raw powder

counterparts but achieve a significantly lower operational pressure drop. This finding reinforces the benefit of the washcoated monolith structure; however, the relatively low CO₂ conversion and CH₄ yield remains a key challenge for these DFMs.

3.4. Mechanochemical

Mechanochemical synthesis has recently emerged as a sustainable method for preparing DFMs, with key studies appearing from 2024 onwards, offering a solvent-free alternative to conventional solvent-based routes such as impregnation and sol-gel methods (Braga et al., 2025; Guan et al., 2025). This approach involves the use of ball milling to integrate catalyst, adsorbent, and support within a single reactor, significantly reducing synthesis time and treatment requirements. Through ball milling, mechanochemical synthesis facilitates intimate microscale interaction between precursors, imparting DFMs with unique morphologies and configurations that can significantly enhance CO₂ uptake and catalytic performance. However, the ball milling process can be energy-intensive, which may negatively impact the overall energy efficiency of the system (Ravichandran et al., 2018).

A study conducted by Danielis et al. (2024) showed that ball milling Ni and Ru catalysts with CaO and ceria-alumina (CA) produced well-dispersed nanoparticles (~ 4 nm), in close proximity with Ca-based species and catalytic sites, creating a high density of adsorbent-catalyst interface. This allows the prepared DFM to exhibit excellent CO₂ uptake and CH₄ yield of up to 0.27 mmol CO₂/g_{DFM} and 0.18 mmol CH₄/g_{DFM}, respectively. Compared to similar samples prepared by conventional wet impregnation technique, DFMs prepared by the mechanochemical synthesis route exhibit an 11% to 30% increase in CO₂ uptake, despite having a lower surface area. This study also highlights the adverse impact of large Ni particles in increasing the risk of sintering and reducing the BET surface area. Reduced BET surface area of DFMs can adversely impact their performance by restricting gas diffusion and limit the accessibility of active sites; thus, suggesting that decreasing Ni loading may provide a higher activity of milled DFMs (Tan and Gunawan, 2023; Porta et al., 2021).

This hypothesis aligns with a study conducted by Guo et al. (2024), who synthesized Ni, MgO DFMs via mechanochemical process at varying Ni loading. It was found that decreasing the Ni loading from 80 mg to 50 mg significantly increases the BET surface area and pore size of the DFMs from 3.68 m²/g to 18 m²/g and 0.053 cm³/g to 0.11 cm³/g, respectively. This allows the latter to achieved comparable CO₂ uptake of 0.31 mmol CO₂/g_{DFM} (vs 0.37 mmol CO₂/g_{DFM}) and CH₄ yield of 0.22 mmol CH₄/g_{DFM} (vs 0.27 mmol CH₄/g_{DFM}), despite having a low Ni loading. However, further decrease in Ni loading to 20 mg would adversely impact the BET surface area and pore size to 9.94 m²/g and 0.076 cm³/g, respectively. As compared to the typical physical mixing process, mechanochemical synthesized DFM exhibits better textural properties in terms of BET surface and pore size, contributing to enhanced CO₂ uptake and catalytic activity. It was also observed that the DFMs display uniquely irregular spherical structures that can potentially inhibit particle growth, contributing to long-term operational stability and improve CO₂ uptake. However, some studies consider these irregular structures as a disadvantage, which may adversely impact catalytic performance (Ravichandran et al., 2018; Liu and Corma, 2018). Furthermore, the mechanochemical process often lacks precise control over the shape of the final product. As a result, the irregular morphology limits the flexibility to tailor DFM structures, potentially restricting opportunities to optimize catalytic activity through shaping (Zhang et al., 2024a). Nevertheless, these results highlighted that the potential of mechanochemical process in optimizing DFM microstructure to maintain output performance while reducing metal content, offering both cost and sustainability advantages.

More recently, Sun et al. (2025) explored the utilization of natural materials such as eggshells and hematite as Ca and Fe substitutes for conventional sources to prepare DFMs via mechanochemical process.

The characterization results show that the mechanochemical process effectively enhances the spatial proximity between Fe and Ca species, where calcined eggshell exhibited an adhesive granular morphology with large pores, enabling nanoscale hematite to uniformly disperse within the cavities of the CaO skeleton, producing well-dispersed DFMs. The derived DFMs exhibited excellent CO₂ uptake of 7 mmol CO₂/g_{DFM} but a low CH₄ yield of approximately 0.1 mmol CH₄/g_{DFM} due to high selectivity of >99% towards CO formation. However, the stability of the DFMs is prone to degradation due to the reduction and re-oxidation of Ca₂Fe₂O₅, resulting in an approximate 30% reduction in CO₂ uptake. To remedy this, MgO was incorporated into the DFMs to help stabilize it, maintaining stable operation for up to 20 cycles and only experiencing a 15% decrease in CO₂ capacities thereafter.

Aside from focusing on precursors, Braga et al. (2025) examined the impact of milling parameters (i.e., milling intensity and type of precursors) on the performance of mechanochemical synthesized DFM. The finding indicates that milling intensity exerts the strongest impact on DFM as higher milling intensity produces a stronger chemical interaction between precursors with support, which enhances CO₂ uptake despite having lower surface areas. This highlights that interaction between precursors and support plays a more crucial role in CO₂ capture rather than its surface area.

3.5. Co-precipitation method

Co-precipitation is a synthesis method used to form solid composites in an aqueous solution that contains two or more metal ions. Generally, when one soluble compound is precipitated from the solution, other soluble metal ions may be adsorbed onto the nucleic formed, and subsequently can be removed from the solution as a homogeneous composite precipitate (Zueva, 2018). Given its nature to be operated under atmospheric conditions, this method often received merits in terms of cost-effectiveness and operation friendliness (Usuki et al., 2025; Guan et al., 2022).

One of its earliest applications for ICCM is reported online in 2019 (published in Zhou et al., 2020), where a bi-function 2D-layered Ni, MgO/Al₂O₃ nanosheets is synthesized for simultaneous CO₂ capture and methanation. Their results demonstrated excellent CO₂ capture efficiency of nearly 100% at 250 °C, with near-complete conversion of the captured CO₂ to CH₄ over Ni active sites. A similar promising outcome was also observed in a more recent work conducted by Fu and Lian (2024). In that work, Al-doped Ni/CeO₂ dual-function catalysts derived from co-precipitation were able to attain a CO₂ conversion of approximately 90% with CH₄ selectivity approaching 100% at 240 °C. However, the risk of agglomeration appeared to be a common conundrum faced by co-precipitation methods compared to other methods, making it harder to achieve size control (Li et al., 2023a). To overcome this issue, one would need to consider adding additional surfactants or polymers (Kaewsanee et al., 2023; Sangsuriyong et al., 2022).

3.6. Physical mixing

Physical mixing refers to the mechanical blending of individual components, which, under the context of DFM preparation refers to the catalysts and adsorbents. Generally, such a synthetic method does not rely on chemical interactions and bonding. As a result, the catalyst and adsorbent remain as discrete particles and have a larger distance between active sites. The increased distance weakens synergistic interaction and would typically reduce DFM performance. However, this increased separation can be beneficial in certain cases, particularly in oxide-based DFM (i.e., CaO), where excessively intimate catalyst and adsorbent contact leads to active-site blockage during carbonation.

For example, Sun et al. (2021b) investigated the performance of Ni/CaO-based DFM derived from physical mixing method and compared their performance with counterparts synthesized via the one-pot method. They demonstrated that an increment in the distance

between active sites (Ni) and sorbents (CaO) prevented newly formed CaCO_3 during carbonation from covering the Ni active sites. As a result, the physically mixed DFM exhibited 24% and 26% higher CO_2 conversion and CH_4 selectivity than the one-pot synthesized DFM, respectively. This observation suggests that excessively intimate proximity may not always be beneficial particularly in DFM systems employing adsorbents that undergo significant volumetric expansion during CO_2 capture step, which may lead to catalyst active-site blockage. Nevertheless, it is also highlighted the stability limitations of physically mixed DFMs, suggesting that insufficient catalyst-adsorbent interaction may negatively affect overall DFM performance.

In contrast, Li et al. (2024a) reported that Ni/AlCaO DFM synthesized via impregnation method exhibited superior CO_2 uptake and CH_4 yield (i.e., at least 2.03 and 2.06 times higher) compared with physically mixed DFMs. This enhanced performance was attributed to the incorporation of Al, which acted as a structural buffer to mitigate the volume expansion associated with CaO carbonation and reduced the coverage of Ni active sites by carbonated products. Therefore, the impregnated DFM was able to maintain the advantages of close catalyst-adsorbent proximity while preserving catalytic accessibility. These findings indicated that DFM synthesis pathway that can achieve strong catalyst-adsorbent interaction while preventing active-site blockage, are expected to offer improved ICCM performance compared to conventional physical mixing method. Overall, the performance of DFMs synthesized via physical mixing is often system-dependent and inconsistent, limiting the reliability of this synthesis approach (Heo et al., 2025).

4. Effect of process conditions on CO_2 adsorption and methanation by DFMs

4.1. Effect of temperature

CO_2 methanation is highly exothermic, hence temperature and heat removal strongly influence both kinetics and catalyst stability during the ICCM cycling. It typically proceeds efficiently at moderate temperatures

below 500°C (Sun et al., 2022b). Noble metal-based catalysts have been reported to exhibit superior catalytic performance due to their exceptional catalytic activity and high methane selectivity (Sun et al., 2022b). Examples include ruthenium (Ru) (Wang et al., 2016; Proaño et al., 2019), gold (Au) (Guo et al., 2018), and palladium (Pd) (Park and McFarland, 2009). Moreover, the interface between active metals and their supports plays a pivotal role in influencing catalytic performance (Guo et al., 2018; Parastaev et al., 2020). A reducible oxide-like catalyst, such as those supported on redox-active materials like cerium oxide (CeO_2) (Zhang et al., 2017; Du et al., 2012), titanium dioxide (TiO_2) (Zhan et al., 2016; Tang et al., 2017), plays a critical role in low-carbon methane production by combining CO_2 adsorption with in situ catalytic hydrogenation via strong metal-support interactions. Fig. 4 shows the three-regime temperature framework explaining the dominant physicochemical constraints governing the integrated CO_2 capture and in situ methanation over dual-function materials (DFMs). The classification of the three-regime temperature framework is linked mainly to the CO_2 sorbent chemistry and involved the activity of methanation catalysts reported in literature (Zhang et al., 2024b; Omodolor et al., 2020; Guo et al., 2023). At low temperatures ($<200^\circ\text{C}$), the CO_2 capture is thermodynamically favoured but methanation is kinetically limited. At this low temperature range, it is often linked with amine-functionalized and zeolitic sorbent (Zhang et al., 2024b). Intermediate temperatures ($\sim 300^\circ\text{C}$ – 380°C) provide an optimal balance between CO_2 release, methanation kinetics, and cyclic stability. At such intermediate temperatures, MgO -, hydrotalcite-, and Na_2CO_3 -based DFMs coupled with Ni catalysts are commonly employed (Zhang et al., 2024b). At high temperatures (>400 – 450°C), fast reaction kinetics are sometimes offset by reduced CO_2 uptake, increased hotspot formation, sintering of active metals, and enhanced reverse water–gas shift (RWGS) activity. At even higher temperature range (500 – 650°C), the dominant material systems are CaO-based dual-function materials incorporating Ni or Ru catalysts, which enable efficient calcium-looping-assisted CO_2 capture coupled with in-situ methanation through reversible carbonation–calcination cycles. (Zhang et al., 2024b). The schematic is synthesized from

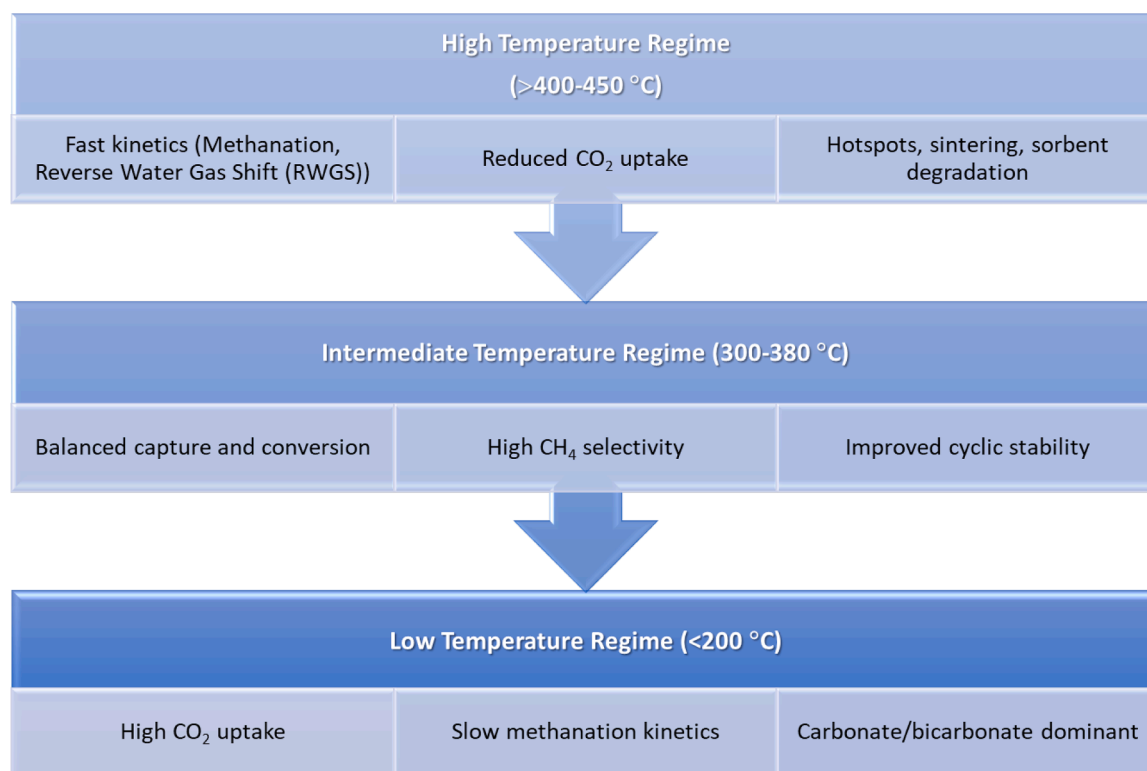


Fig. 4. Three-regime temperature framework for integrated CO_2 capture and in-situ methanation over DFMs.

experimental trends reported in the literature enlisted in Table 2, which compiles the performance of DFMs across a range of operating temperatures.

According to Shen et al. (2025), the influence of reaction temperature affects both the CO₂ capture and catalytic conversion creating a synergistic effect. Theoretically, a higher uptake capacity tends to occur at lower temperatures in the adsorbent (Shen et al., 2025). However, catalytic conversion typically requires higher temperatures, which promotes CO₂ release and activates the gas molecules. This leads to a mismatch in the optimal reaction temperatures for these two stages. This issue is less pronounced in exothermic methanation as the reaction performs well at moderate temperature and allows the CO₂ capture efficiency to improve with steady in situ methanation (Shen et al., 2025). Sun et al. (2023) studied on the dual functional hybrids of alkali metal salts (AMS)-promoted porous CaMgO adsorbents (AMS/CaMgO) with non-precious Ni_xCo_y bimetallic catalysts, facilitating for the ICCM technology under intermediate temperature of 350 °C. In this technology, there are three distinct steps involved, i.e., (i) adsorption-based CO₂ capture from simulated flue gas, (ii) intermediate N₂ purging, and (iii) catalytic CO₂ methanation with H₂ as the reducing agent. It was concluded that with the incorporation of these two processes of CO₂ capture and methanation conversion at the temperature of 350 °C shows a synergistic effect between the adsorption and conversion that increase the efficiency of the CO₂ methanation, with a conversion efficiency and CH₄ selectivity reaching 93.4% and ~88%, respectively. However, for the reverse water–gas shift (RWGS) and dry reforming of methane (DRM) at elevated temperatures, the experiments demonstrated a temperature compatibility between the two steps, with RWGS and DRM closely interlinked in the overall integrated performance. Furthermore, variations in the packing configuration of the sorbent and catalyst within the system were found to significantly influence product selectivity between CH₄ and CO.

In another study, Jo et al. (2023) investigated on the developed a Ru/K₂CO₃–MgO (Ru/KMg) catalyst-sorbent for ICCM at temperature of 60–320 °C for carbon capture and 320 °C for methanation. The regeneration behaviour of the catalyst-sorbent was examined using temperature-programmed desorption (TPD) in the atmosphere of N₂ and temperature-programmed surface reaction (TPSR) in H₂, subsequent to carbonation conducted over a temperature range of 60–320 °C. It was reported that at the low carbonation temperatures of 60 °C and 120 °C, the CO₂ was primarily captured through the formation of KHCO₃, meanwhile MgCO₃ and K₂Mg(CO₃)₂ phases were detected at all investigated temperature range from 60 °C to 320 °C. Additionally, the CO₂ capture capacity exhibited a clear decreasing trend with increasing carbonation temperature, highlighting the temperature-dependent nature of the process. The desorption of CO₂ from the KHCO₃ phase did not influence the H₂ composition at temperatures below 200 °C during the methanation process, despite an increase in the carbonation temperature from 60 °C to 120 °C. The decarbonation of the MgCO₃ and K₂Mg(CO₃)₂ phases proceeded more rapidly under H₂ compared to N₂, due to the immediate reaction of the desorbed CO₂ with H₂ to form CH₄ based on the Le Chatelier's principle. Jeong-Potter et al. (2022c) carried out an investigation into the synthesis of Ru–Ni catalytic combinations, intimately integrated with Na₂O supported on γ-Al₂O₃ DFMs. This study reported that Ni-only DFMs are prone to oxidation in the presence of O₂ and cannot be effectively reactivated below temperature of 550 °C which limits their suitability for flue gas applications. The addition of a small amount of Ru, however, facilitates NiO reduction at typical DFM operating temperature of ~300 °C that corresponds to flue gas conditions, and consequently increases the number of active sites available for CO₂ methanation.

Park et al. (2021) evaluated on the performance of the physical mixtures of NaNO₃/MgO (sorbent) and Ru/Al₂O₃ (catalyst), prepared through different methods by integrating the CO₂ capture and conversion cycles. In this study, the CO₂ sorption behaviour and catalytic activity were investigated using thermogravimetric analysis and

spectroscopic measurements and fixed-bed reaction experiments. Cyclic experiments were conducted to assess the influence of temperature on integrated CO₂ capture and methanation. While isothermal operation at 300 °C provided a baseline, a modified strategy applied CO₂ sorption at 300 °C and methanation at 320 °C in the first cycle, followed by subsequent cycles at 300 °C. In this study, the sorption and methanation capacities declined more rapidly in cycles following methanation at temperature of 320 °C compared to cycles conducted entirely isothermally at temperature of 300 °C. Specifically, the CO₂ sorption capacities decreased from 2.93 mmol CO₂/g to 2.07 mmol CO₂/g, while CH₄ production capacities declined from 1.90 to 1.40 mmol CH₄/g over subsequent cycles. These results suggest that performing methanation at the elevated temperature of 320 °C accelerates the deactivation of the MgO sorbent which is likely due to its tendency to sinter during thermal cycling. Although high initial sorption and methane production were achieved, the sorbent stability improvement under high-temperature methanation remains a significant challenge.

Sun et al. (2021b) employed different Ni/CaO-based DFMs to evaluate the ICCM process at temperature of 550 °C by changing the distances between active sites and sorbent phases to create different volume spaces for CaCO₃ growth during the carbonation reaction. At the temperature of 550 °C, it was revealed that the distance between the active sites and the sorbent via physical mixing markedly enhanced the CO₂ conversion and CH₄ selectivity of the 1%Ni/CeO₂–CaO physical catalyst, achieving values of 62% and 84%, respectively, under atmospheric pressure with CO₂ captured from a 15% CO₂/N₂ stream. The improved performance at this elevated temperature is linked to the preservation of the well-dispersed Ni active sites on the CeO₂ nanorod surface which reduced their deposition by newly formed CaCO₃ during carbonation. Sun et al. (2022b) utilised Ru/CeO₂ as a catalyst and involved physical mixing with MgO as an adsorbent with three various CeO₂ morphologies that consists of rod, particle, and cube CeO₂ which was prepared using hydrothermal method. Interestingly, although the CO₂ desorption capacity of Ru/rod-CeO₂–MgO declines markedly from 58.5 to 5.3 μmol/g_{DFM} over nine cycles, the CH₄ yield exhibits only a modest decrease of approximately 20% (from 0.34 to 0.27 mmol/g_{DFM}) at temperature of 300 °C. Additionally, the Ru/rod-CeO₂–MgO catalyst demonstrates remarkable catalytic stability, achieving greater than 95% CO₂ conversion with 5% H₂/N₂ from the sixth cycle onwards. Duyar et al. (2015) reported a low CH₄ yield of 0.5 mmol/g from the synthesised Ru/CaO DFMs for the ICCM process. This was mainly linked to the limited sorbent loading. Nevertheless, the optimal temperature of Ru-based catalysts for CO₂ methanation is starting from temperature 200 °C to 300 °C, which is considerably below that required for carbon capture with the aid of CaO-based sorbents (Guo et al., 2018). The disadvantages of Ru-based catalysts include their high cost and limited resistance to poisoning, which restrict their industrial applicability. In contrast, Ni-based catalysts represent promising alternatives for the proposed ICCM process due to their economic feasibility and their compatibility in operating temperature between CO₂ methanation and CO₂ capture when coupled with CaO-based DFMs (Chen et al., 2017; Solis-Garcia et al., 2017). Therefore, establishing the optimal temperature alignment is essential for DFMs to deliver integrated and synergistic performance.

Most studies operate in moderate temperature windows where methanation rates are sufficiently high while limiting metal sintering and overly stable carbonate formation. In practice, DFMs which combines alkaline sorbent, for example CaO or Na₂CO₃-based, with Ni- or Ru-based methanation catalysts are stereotypically functioned within a wide temperature a range of ~250–650 °C, which mostly depends on the reaction step and material formulation (Tsiotsias et al., 2020; Merkouri et al., 2023; Li et al., 2023b). The CO₂ capture step over CaO based sorbents is normally conducted at temperature range of 450–650 °C, where carbonation to CaCO₃ is thermodynamically fast and rapid carbonation kinetics are attained (Criado et al., 2018; Sun et al., 2024a). Therefore, the cyclic ICCM operation usually targets a compromise

Table 2Summary of CO₂ adsorption and in situ methanation performance using various DFMs in ICCM systems.

DFMs	Operating conditions		Performance				Application/ Feed Category*	Reference
	CO ₂ adsorption	CO ₂ methanation	CO ₂ uptake capacity	CO ₂ conversion (%)	CH ₄ yield (mol/kg)	CH ₄ selectivity (%)		
Alkali Metal Salts/ CaMgO Ni _x Co _y	Temperature: 350 °C Pressure: - Gas: 45:55 (v/v) for CO ₂ /N ₂ Flowrate: 50 mL/min	Temperature: 300 – 400 °C Pressure: - Gas: • Reduction step: 10 vol% H ₂ /N ₂ • CO ₂ capture step: 45 vol% CO ₂ /N ₂ (simulated flue gas) • Regeneration step: 10 vol% H ₂ /N ₂ • Purge step: pure N ₂ Flowrate: 50 mL/min Gas hourly space velocity (GHSV): 7500 – 22500 mL/g.h	350 °C: 14.4 mmol/g, 375 °C: 14.6 mmol/g, 400 °C: 14.4 mmol/g	350 °C: 90%, 375 °C: 60.5%, 400 °C: 43.5%	350 °C: 9.97 mmol/g, 375 °C: 5.53 mmol/g, 400 °C: 2.70 mmol/g	350 °C: 19.0%, 375 °C: 31.8%, 400 °C: 53.3%	Fundamental/ ideal condition	Sun et al. (2023)
Ru/ K ₂ CO ₃ –MgO	Temperature: 60 – 320 °C	Temperature: 280 – 320 °C Gas: 90 vol% H ₂	280 °C: 1.06 mmol/g, 320 °C: 1.07 – 1.15 mmol/g	100%	-	97.4 – 100%	Fundamental/ ideal condition	Jo et al. (2023)
Ru–Ni, Na ₂ O/ Al ₂ O ₃	Gas: • No O ₂ and steam (7.5% CO ₂ /N ₂) • Simulated flue gas (7.5% CO ₂ , 4.5% O ₂ , 15% H ₂ O, balance N ₂) Flowrate: 200 mL/min GHSV: 7700/h	Temperature: 320 °C Gas: 90 vol% H ₂ Flowrate: 200 mL/min GHSV: 7700/h	-	-	1.07 – 1.119 mmol/g	-	Realistic flue gas	Jeong-Potter et al. (2022c)
5% NaNO ₃ / 1%Ru/ Al ₂ O ₃	Temperature: 300 °C Gas: 10% CO ₂ / balance N ₂ with 11% absolute humidity Flowrate: 20 mL/min	Temperature: 280 – 320 °C Gas: 10% CO ₂ / 40% H ₂ /balance N ₂ Flowrate: 20 mL/min	-	70 – 96%	-	-	Flue-gas-relevant simplified feed	Park et al. (2021)
1%Ni/CeO ₂ – CaO	Temperature: 550 °C Pressure: 1 atm Gas: 15% CO ₂ /N ₂ Flowrate: 50 mL/min	Temperature: 550 °C Pressure: 1 atm Gas: Pure H ₂ Flowrate: 50 mL/min	15.3 mmol/g	62%	8 mmol/g	84%	Flue-gas-relevant simplified feed	Sun et al. (2021b)
Ru/CeO ₂ –MgO	Temperature: 300 °C Gas: 35% CO ₂ /N ₂ Flowrate: 100 mL/min	Temperature: 300 °C Gas: 5% H ₂ /N ₂ Flowrate: 50 mL/min	-	55.7 – 59.8%	0.29 mmol/g	-	Fundamental/ ideal condition	Sun et al. (2022b)
CaO/γ-Al ₂ O ₃ , Ru/γ-Al ₂ O ₃	Temperature: 320 °C Gas: 10% CO ₂ /N ₂ Flowrate: 30 mL/min	Temperature: 320 °C Gas: 4% H ₂ /N ₂ Flowrate: 26 mL/min	0.3 – 0.6 mmol/g	49.6 – 88.5%	-	-	Flue-gas-relevant simplified feed	Duyar et al. (2015)
Ru–K/Al ₂ O ₃	Temperature: 300 °C Pressure: 1 atm Gas: CO ₂ /N ₂ (dry)	Temperature: 350 °C H ₂ partial pressure: 0.15 – 0.60 atm Gas: H ₂ /N ₂	-	0.15 atm: 40%, 0.30 atm: 68%, 0.60 atm: 95%	0.15 atm: ~0.4, 0.60 atm: ~0.95	-	Fundamental/ ideal pressure study	Porta et al. (2023)
Ru–Ba/Al ₂ O ₃	Temperature: 300 °C Pressure: 1 atm Gas: CO ₂ /N ₂ + H ₂ O	Temperature: 350 °C H ₂ partial pressure: 0.15 –	-	Similar trend (exact % not reported)	Similar trend (lower than Ru–K)	-	Flue-gas-relevant simplified feed	Porta et al. (2023)

(continued on next page)

Table 2 (continued)

DFMs	Operating conditions		Performance				Application/ Feed Category*	Reference
	CO ₂ adsorption	CO ₂ methanation	CO ₂ uptake capacity	CO ₂ conversion (%)	CH ₄ yield (mol/kg)	CH ₄ selectivity (%)		
K/Cu–Zn–Al (CZA) [†]	Temperature: 300 °C Pressure: 1 bar Gas: CO ₂ /N ₂	0.60 atm Gas: H ₂ /N ₂ Temperature: 300 °C Pressure: 1 – 3 bar Gas: H ₂ -rich	-	1 bar: ~50%, 3 bar: 94.4%	-	-	Other CO ₂ hydrogenation system	Jeong-Potter et al. (2024)
Na–Ru/Al ₂ O ₃ (Na-DFM)	Temperature: 320 °C Pressure: 0.1 – 5.0 MPa	Temperature: 320 °C Pressure: 0.1 – 5.0 MPa	-	Energy conversion efficiency decreases from 66.1% to 60.3% as pressure increases	-	CH ₄ concentration >90 vol% at ≥0.7 MPa	Fundamental/ideal pressure simulation	Jeong-Potter et al. (2022b); Jeong-Potter et al. (2022d); Chen et al. (2024a)
(Li–Na–K)NO ₃ –MgO–Ni/CeO ₂ -phy (Mg-DFM)	Temperature: 300 °C Pressure: 0.1 – 5.0 MPa	Temperature: 300 °C Pressure: 0.1 – 5.0 MPa	-	Lowest Energy-to-steam efficiency in 0.2 – 5.0 MPa range	-	CH ₄ concentration >90 vol% at ≥0.7 MPa	Fundamental/ideal pressure simulation	Huang et al. (2021); Huang et al. (2023); Chen et al. (2024a)
Ni ₅ –CeLi (Li-DFM)	Temperature: 320 °C Pressure: 0.1 – 5.0 MPa	Temperature: 320 °C Pressure: 0.1 – 5.0 MPa	-	Energy conversion efficiency less sensitive to pressure	-	CH ₄ concentration ≤88.4 vol% at 5 MPa	Fundamental/ideal pressure simulation	Ly et al. (2023); Chen et al. (2024a)
Ni–Na ₂ CO ₃ /γ-Al ₂ O ₃ (Na-DFM)	Temperature: 340 °C Pressure: 0.1 – 0.4 MPa Gas: 5 vol% CO ₂ in N ₂	Temperature: 340 °C Pressure: 0.1 – 0.4 MPa	-	From 40% to 95% (from 0.1 to 0.4 MPa)	From 1.5 to 3.3 mmol/g (increasing with pressure)	0.1 MPa: ~90% 0.2 MPa: ~96% 0.4 MPa: ~99%	Flue-gas-relevant simplified pressure study	Fan et al. (2025)

(-) data are not available.

[†] Methanol-producing system, included to demonstrate pressure effect on CO₂ hydrogenation.

* Application/feed category was assigned based on the reported feed composition. “Realistic flue gas” refers to feeds containing CO₂ together with O₂ and H₂O, while “flue-gas-relevant simplified feed” refers to CO₂/N₂ or humid CO₂/N₂ mixtures with CO₂ concentrations relevant to flue gas but without the full impurity matrix. “Fundamental/ideal condition” refers to simplified model feeds or simulation-based studies.

temperature window of 300–400 °C which ensures adequately fast methanation kinetics while allowing earlier captured CO₂ can be free and hydrogenated in situ (Jeong-Potter and Farrauto, 2021; Merkouri et al., 2023).

From a thermodynamic standpoint, CO₂ methanation via the Sabatier reaction is strongly exothermic as shown in Eq. (1). In accordance with Le Chatelier’s principle, the exothermic character of the reaction leads to enhanced methane formation at lower temperatures due to a shift of the equilibrium toward the products (Acierno et al., 2025). As a result, equilibrium-controlled operation at lower temperatures enhances CO₂ conversion and CH₄ selectivity; however, this thermodynamic advantage is accompanied by kinetic limitations (Acierno et al., 2025). The apparent activation energy for CO₂ hydrogenation is on the order of ~100 kJ mol^{−1}; consequently, intrinsic reaction rates decrease sharply as the temperature is reduced from 300 °C to around 250 °C, often by approximately one order of magnitude—rendering equilibrium conversions unattainable at practical space times (Sterk et al., 2022; Mohd Ridzuan et al., 2022). Additionally, high pressures enhanced the production of methane (Rönsch et al., 2016).

Beyond thermodynamic considerations, kinetic limitations further constrain the operating window. Methanation over Ni-based catalysts become rate-limited below ~250–280 °C despite favourable equilibrium constraints, whereas reaction rates increase sharply with temperature and peak in the 300–400 °C range under practical space velocities (Rönsch et al., 2016; Alrafi et al., 2020). At higher temperatures greater than temperature of 450 °C, RWGS kinetics become increasingly competitive, reducing CH₄ selectivity (Merkouri et al., 2023). Although elevated temperature accelerates carbonate decomposition and improves CO₂ availability near metal sites, excessively rapid CO₂ release

can induce local stoichiometry imbalances (Sun et al., 2021b; Arellano-Treviño et al., 2019b). Consequently, for temperature ranging from 300 °C–400 °C represents the most favourable kinetic compromise for most DFM-based ICCM systems (Merkouri et al., 2023).

Beyond reaction thermodynamics and kinetics, heat and mass transfer effects strongly influence ICCM performance because CO₂ methanation is highly exothermic. Fixed-bed DFM systems operating at ≥350–400 °C commonly experience local temperature rises of 30–100 °C, which accelerate metal sintering, support phase transformations, and sorbent degradation, leading to reduced CO₂ uptake and cyclic stability (Park et al., 2021; Sun et al., 2023; Jeong-Potter et al., 2022a). Additional mass-transfer limitations arise from mismatches between CO₂ release and consumption rates or from diffusion barriers imposed by dense carbonate layers (Jo et al., 2023; Shen et al., 2025).

Overall, most studies operate DFMs within moderate temperature windows that balance methanation activity with sorbent and catalyst stability. However, the optimal temperature range is inherently system-specific, depending on sorbent chemistry, catalyst composition, spatial integration of capture and catalytic sites, and cycle design. Reported ICCM performance is also highly sensitive to pressure, space velocity, H₂/CO₂ ratio, and cycle time, as these parameters govern equilibrium constraints, intraparticle diffusion, and hotspot severity. To improve comparability across studies, future work should consistently report a standardized set of operating conditions such as temperature, pressure, GHSV, H₂/CO₂ ratio, and cycle protocol, together with cyclic performance metrics (CO₂ uptake per cycle, conversion, CH₄ selectivity, stability over N cycles) and a complete carbon balance.

4.2. Effect of pressure

The growing emphasis on carbon-neutral technologies has propelled interest in ICCM, a process designed to convert CO₂ emissions into synthetic natural gas (SNG) through the use of renewable hydrogen. DFM, which serve simultaneously as CO₂ adsorbents and methanation catalysts, are fundamental to the ICCM process. Among the process parameters influencing ICCM performance, operating pressure plays a uniquely complex role, directly impacting both the adsorption of CO₂ and its subsequent catalytic methanation (Sun et al., 2024b). This section examines recent findings on how variations in pressure affect ICCM performance using DFMs, drawing from validated experimental and simulation-based studies. A summary of the data discussed is provided in Table 2. To improve the practical interpretation of pressure effects, the studies in Table 2 are further categorized according to their feed/application category, namely realistic flue gas, direct air capture (DAC), flue-gas-relevant simplified feed, fundamental/ideal condition, or other CO₂ hydrogenation system. This classification is important because pressure effects obtained from dry CO₂/N₂ model feeds or simulations cannot be directly extrapolated to practical CH₄ combustion flue gas, which typically contains 4–10% CO₂, 2–5% excess air, 8–20% steam and balance N₂. Similarly, DAC studies should be treated separately because the CO₂ concentration is only around 400–425 ppm in air and pressurizing such a dilute stream is generally less practical than pressurizing a concentrated or partially enriched CO₂ stream.

In general, increasing pressure improves CO₂ adsorption by enhancing the thermodynamic favourability of gas–solid equilibrium. This effect is particularly beneficial in ICCM systems that rely on low-concentration CO₂ sources, such as flue gas from power plants (Soo et al., 2024). One clear demonstration of this effect comes from the work of Fan et al. (2025), who evaluated a Ni–Na₂CO₃/γ-Al₂O₃ DFM under total pressures ranging from 0.1 to 0.4 MPa. Their findings showed that CO₂ conversion, reaction kinetics and CH₄ selectivity rose over this range. The enhancement in performance under higher pressure conditions was attributed to both increased CO₂ capture and more efficient hydrogenation in the methanation step (Zhang et al., 2025). However, the pressure study by Fan et al. (2025) should be interpreted as a flue-gas-relevant simplified study rather than a fully realistic flue-gas study. Although the CO₂ concentration used in the study falls within the lower range relevant to combustion flue gas, the feed did not include excess O₂ and steam. Therefore, the reported improvement in CO₂ conversion and CH₄ yield with pressure provides useful insight into adsorption–methanation coupling, but further validation is still required under realistic CH₄ combustion flue gas containing CO₂, O₂, H₂O, and N₂.

Pressure also alters the kinetics and product distribution during in situ methanation. A thermodynamic sensitivity study by Chen et al. (2024a) modelled the performance of ICCM systems using three different DFMs namely Na–Ru/Al₂O₃, Mg–DFM, and Li–DFM over a broad range of pressures, from 0.1 to 5.0 MPa. For the Na–DFM, CH₄ concentration in the product gas rose from 71.8% at atmospheric pressure to 87.5% at 5.0 MPa. This enhancement in methane content aligns with the requirements for natural gas pipeline compatibility, especially in terms of heating value and hydrogen content. However, this gain came with a measurable cost: energy-to-SNG efficiency declined from 66.1% to 60.3% as pressure increased, highlighting a trade-off between gas quality and energy efficiency. In the same study, Mg-based DFMs maintained high SNG quality and relatively stable energy performance across the pressure range, suggesting that material composition significantly influences pressure sensitivity. Conversely, Li-DFMs showed comparatively low methane content, even under elevated pressures, indicating that not all materials benefit equally from pressurization. These variations underscore the importance of selecting pressure-compatible DFMs based on their thermodynamic and kinetic behaviour (Su and Shen, 2024).

Experimental findings further support these trends. In a study by

Porta et al. (2023), Ru-based catalysts supported on K- and Ba-promoted alumina were tested under varying hydrogen pressures. CO₂ conversion increased from 40% to 95% as H₂ pressure rose from 0.15 to 0.60 atm, while CH₄ selectivity improved in tandem. Although this study did not operate under a full ICCM configuration, the results nonetheless confirm that elevated pressure, especially hydrogen partial pressure, strongly enhances methanation performance. Despite the clear advantages of higher pressure for reaction kinetics and thermodynamics, practical considerations impose limitations. Elevated pressures introduce additional equipment and energy costs, especially for gas compression and system sealing (Osman et al., 2024). For this reason, moderate pressure regimes, typically around 0.3 to 0.7 MPa, are often recommended as a compromise between performance and feasibility. In this context, Fan et al. (2025) suggested 0.4 MPa as an optimal point, where CH₄ yield and CO₂ conversion are near their maxima without excessive energy penalties.

The effect of pressure also extends to CO₂ partial pressure, which can be decoupled from total system pressure. Fan et al. (2025) examined the impact of varying CO₂ concentrations (2.5% to 20% by volume) at constant atmospheric pressure. While higher CO₂ content led to greater adsorption capacity, the study noted diminishing returns beyond a certain threshold, suggesting the presence of equilibrium limitations or pore saturation effects in the DFM. When total pressure was simultaneously increased with CO₂ concentration, both capture and methanation efficiency improved, reaffirming the synergistic benefits of total and partial pressure elevation (Quan et al., 2025). Moreover, data from long-term simulations reveal that energy penalties associated with pressure do not scale linearly. According to Chen et al. (2024a), at 5.0 MPa, electricity consumption across different DFMs converged to similar values, reducing the relative energy advantage of one material over another. This suggests that beyond a certain point, pressure-induced gains plateau, and further increases may offer limited returns unless the entire system is re-optimized for high-pressure operation.

DFMs operating at high pressure may suffer from mechanical degradation, especially if the support structure is not engineered for stress resistance (Thornton et al., 2016). Additionally, impurities in industrial flue gas, such as SO_x and NO_x, can accelerate DFM deactivation under pressure, further complicating long-term performance (Rezaei and Jones, 2013; Rezaei and Jones, 2014). From a systems integration standpoint, pressure also influences heat management strategies. Methanation is a highly exothermic reaction, and at elevated pressures, the rate of heat release can lead to localized hot spots, potentially damaging the catalyst. In ICCM configurations that lack effective heat dissipation, this could limit cycle duration or require operational compromises, such as temperature moderation or diluted reactant streams. Some modelling studies, including the one by Chen et al. (2024a), assume idealized heat removal systems to simulate performance. However, these assumptions may not hold in real-world applications, where heat transfer efficiency is constrained by reactor design and material limits.

From an application viewpoint, the role of pressure differs across the main feed clusters. For realistic flue gas, moderate pressurization may improve CO₂ uptake and methanation, but compression of N₂, O₂, and steam introduces additional energy and equipment penalties. For DAC, increasing total air pressure is usually unattractive because a large volume of N₂ and O₂ must be compressed to increase the very small CO₂ partial pressure. Therefore, DAC-oriented DFM studies should be discussed separately from flue-gas studies. For fundamental or ideal studies, pressure trends remain valuable for understanding thermodynamics and kinetics, but they should not be treated as direct evidence of practical performance unless realistic feed compositions are tested.

As summarized in Table 2 and further illustrated in Fig. 5, the influence of pressure on ICCM performance is highly system-dependent, varying with DFM composition, operating window, and reactor configuration. While increasing pressure generally improves CO₂ conversion and enhances CH₄ concentration, the magnitude of these gains and their

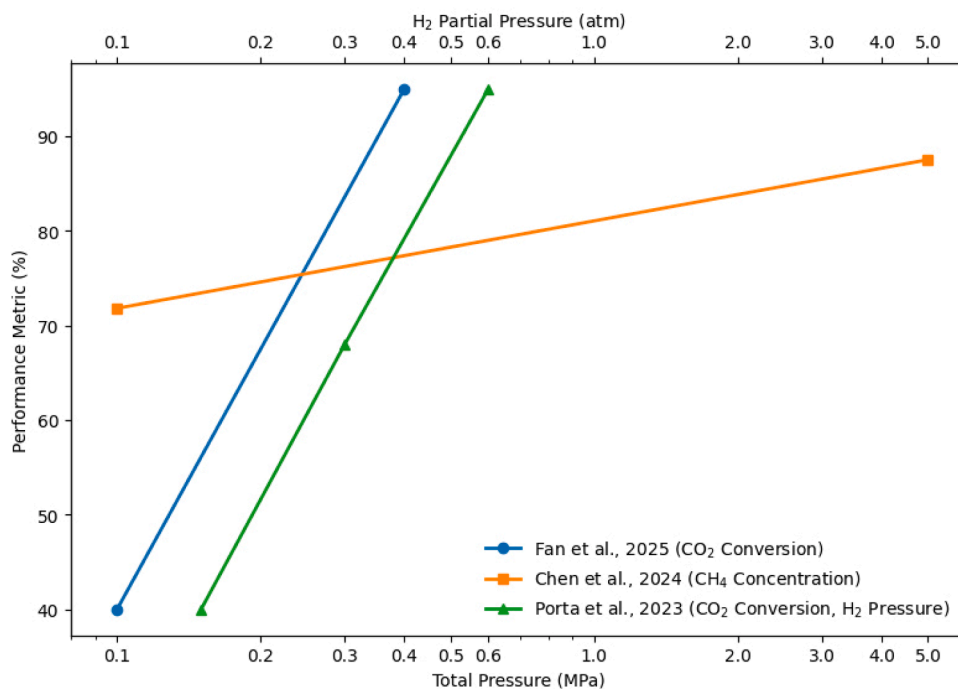


Fig. 5. Pressure-dependent performance trends in ICCM using DFMs. Reported data illustrate the effect of total pressure on CO₂ conversion (Fan et al., 2025) and CH₄ concentration (Chen et al., 2024a), as well as the influence of H₂ partial pressure on CO₂ conversion (Porta et al., 2023).

impact on overall energy efficiency differ across materials. This indicates that pressure selection should not be treated as a universal optimization parameter but rather evaluated in relation to the specific performance objective, whether maximizing methane purity, achieving high CO₂ conversion, or maintaining favourable energy efficiency. Future work should therefore emphasize long-term evaluation under practically relevant pressure conditions, supported by techno-economic analysis that realistically considers compression demand, operational cost, and infrastructure requirements.

4.3. Effect of gas hourly space velocity (GHSV)

GHSV influences the ICCM process using DFMs, as it determines the gas-solid contact time and subsequently governs both the CO₂ adsorption and successive catalytic methanation performance. It is defined as the volumetric flow rate of the feed gas divided by the total reactor volume under standard temperature and pressure (STP), with units of h⁻¹ (Nozaki et al., 2023). In most studies, GHSV is calculated based on the empty reactor volume (i.e., the superficial reactor volume), making it equivalent to the inverse of the apparent gas residence time (Nozaki et al., 2023). A lower GHSV prolongs the residence time of the reactant gases, thereby enhancing gas-solid interactions between the reactants and the active sites of the DFM (Omodolor et al., 2020). This promotes more effective CO₂ adsorption and facilitates the hydrogenation of adsorbed CO₂ intermediates into CH₄. Conversely, increasing the GHSV shortens the gas-solid contact time, which can lead to incomplete CO₂ capture, lower CO₂ conversion, and reduced CH₄ selectivity owing to insufficient hydrogenation of the adsorbed CO₂ species (Omodolor et al., 2020).

During the CO₂ capture stage, GHSV plays a critical role by determining the gas-solid contact time, which influences the effective utilisation of the sorbent's adsorption capacity (Danish et al., 2021). Adequate contact time facilitates the diffusion of CO₂ molecules into the porous structure and increases their interaction with the available adsorption sites, thereby enhancing CO₂ capture efficiency (Danish et al., 2021). Zhang et al. (2024b) had mentioned that solid sorbents such as CaO-, MgO-, and hydrotalcite-based materials able to maximise

the CO₂ uptake with adequate residence time involved. According to Cafiero et al. (2026) and Shen et al. (2025), the GHSV is essential during the subsequent methanation stage in achieving the effective contact between the adsorbed CO₂ species, hydrogen, and metallic active sites such as Ni and Ru by allowing the Sabatier reaction to proceed in forming CH₄ effectively. Ye et al. (2019) developed a Ni/SiO₂ catalyst using the ammonia evaporation method (AEM), which generated a lamellar nickel phyllosilicate structure and promoted the uniform dispersion of nickel species on the SiO₂ support. In this study, the reduction of nickel phyllosilicate resulted in the formation of highly dispersed and confined Ni nanoparticles (4.2 nm) within the porous SiO₂ support, which possessed a high surface area of 446.3 m²/g. Consequently, the Ni/SiO₂-AEM catalyst demonstrated superior methanation performance, delivering a high methane yield with remarkable stability over 100 h at a GHSV of 10,000 mL/(g_{cat}·h) and maintaining activity for an additional 60 h under an increased GHSV of 30,000 mL/(g_{cat}·h) at 370°C. Compared with the Ni/SiO₂-IM catalyst synthesized by the impregnation method (IM), the AEM-derived catalyst exhibited improved methane yield and enhanced resistance to deactivation under the same operating conditions. Wang et al. (2024) investigated hydroxylated TiO₂-induced high-density Ni clusters to overcome the activity-selectivity trade-off in CO₂ hydrogenation. In comparison, additional catalytic activity tests were conducted to evaluate the product selectivity of 10Ni/TiO₂-OH and 10Ni/TiO₂-Ref1, which possess distinctly different Ni structures. By varying the GHSV, the product selectivity of both catalysts was compared under similar CO₂ conversion levels. Notably, even when the GHSV was increased to 800,000 mL g_{cat}⁻¹ h⁻¹, 10Ni/TiO₂-OH still exhibited significantly higher CO₂ conversion than 10Ni/TiO₂-Ref1 operated at a much lower GHSV of 20,000 mL g_{cat}⁻¹ h⁻¹. Jia et al. (2019) developed a Ni/ZrO₂ catalyst through a plasma decomposition of a nickel precursor at atmospheric pressure (~150°C), followed by hydrogen reduction at 500°C. The plasma treatment produced highly dispersed Ni(111) nanoparticles with strong Ni-ZrO₂ interactions and abundant oxygen vacancies, which enhanced CO₂ adsorption, activation, H₂ dissociation, and hydrogen spillover, resulting in superior low-temperature methanation activity. Notably, the plasma-treated catalyst achieved a CO₂ conversion of 71.9%, a CH₄ yield

of 69.5%, and a turnover frequency (TOF) of 0.61 s^{-1} at 300°C under a high GHSV of $60,000\text{ h}^{-1}$, demonstrating excellent catalytic activity despite the short gas–solid contact time. In contrast, the thermally prepared catalyst exhibited significantly lower CO_2 conversion (32.9%), CH_4 yield (30.3%), and turnover frequency, TOF (0.39 s^{-1}) under the same reaction conditions.

Overall, the reported studies highlight the significant influence of GHSV on CO_2 methanation performance, as it governs the residence time of reactants and their accessibility to catalytic active sites. A general consensus among Ni-based catalysts is that structural optimisation, such as achieving high metal dispersion, strengthening metal–support interactions, and tailoring surface characteristics, is important for CO_2 activation enhancement and sustaining catalytic activity at elevated GHSV values. Nevertheless, the effectiveness of these strategies changes that relies on the support material, catalyst synthesis approach, and the resulting active site properties. While certain catalysts benefit from improved CO_2 adsorption and activation, others achieve enhanced durability by suppressing Ni particle agglomeration and preserving active sites during reaction. Consequently, the development of efficient CO_2 methanation catalysts requires the synergistic optimization of catalyst properties and operating parameters to overcome the limitations associated with shortened residence times under high-throughput conditions.

4.4. Effect of impurities

Although temperature and pressure are commonly discussed as the main operating parameters in ICCM, practical deployment of DFMs will also depend strongly on their resistance toward impurities in real CO_2 -containing streams. This issue deserves specific attention because many ICCM studies still rely on simplified feeds, typically CO_2/N_2 during capture and H_2/N_2 during methanation. Such conditions are useful for identifying intrinsic DFM behavior, but they do not fully represent post-combustion flue gas, direct-air-capture streams or biogas-derived CO_2 . Realistic gases may contain H_2O , O_2 , SO_x , NO_x , CO , CH_4 , H_2S , siloxanes and particulate matter, depending on the source. These species can affect ICCM through competitive adsorption, oxidation of catalytic metals, irreversible sulfate or nitrate formation, pore blockage, hydrothermal ageing and disruption of sorbent–catalyst interfaces. Therefore, impurity tolerance should be considered a core DFM design requirement rather than a secondary stability issue (Omodolor et al., 2020; Gharamaleki et al., 2025).

Water vapor is one of the most important impurities because it is present in many CO_2 sources and is also formed during CO_2 methanation. Its effect is complex and should not be interpreted only from CO_2 uptake data. In some alkaline sorbents, moisture can promote bicarbonate or hydroxylated carbonate formation, thereby improving low-temperature CO_2 capture. This behavior has been reported for Ru+ $\text{Na}_2\text{O}/\text{Al}_2\text{O}_3$ DFMs under ambient capture and in-situ methanation conditions (Jeong-Potter et al., 2022b). However, improved humid capture does not always lead to higher methane production. Liu and Chen (2024) showed that steam can strongly affect in-situ hydrogenation over Ru- $\text{Na}_2\text{CO}_3/\gamma\text{-Al}_2\text{O}_3$ DFM. At elevated steam content, moisture-driven carbonate desorption becomes more competitive, leading to lower adsorbent conversion and reduced CH_4 selectivity. This finding highlights an important benchmarking issue whereby high CO_2 uptake under humid conditions may overstate DFM performance if the captured species are released as CO_2 instead of being converted to CH_4 .

Oxygen is particularly problematic for Ni-based DFMs. Ni is attractive because it is inexpensive and active for methanation, but it can be oxidized during exposure to O_2 -containing flue gas. Arellano-Treviño et al. (2019a) identified this as a key limitation for Ni-containing DFMs under flue-gas conditions. Once metallic Ni is converted to NiO, the subsequent H_2 step must reduce NiO back to Ni within the available cycle time. If this redox recovery is incomplete, methane productivity decreases even when CO_2 uptake remains acceptable. This creates a

fundamental conflict in Ni-based ICCM operation because the capture step may expose the material to oxidizing gas, while the conversion step requires reduced metallic sites. Bimetallic and promoted formulations can partly address this oxygen sensitivity. Ru can promote H_2 dissociation and facilitate the reduction of oxidized Ni species at lower temperature. This effect has been linked to improved availability of active hydrogenation sites in Ru–Ni DFMs (Proaño et al., 2020). More recently, Peng et al. (2025) reported that Cu-promoted Ni–Ca DFMs improved cyclic behavior under O_2 -containing flue gas. Onrubia-Calvo et al. (2025) also showed that partial replacement of Ni with Ru in $\text{La}_{0.6}\text{Ca}_{0.4}\text{NiO}_3/\text{CeO}_2$ enhanced integrated CO_2 adsorption and methanation under NO -, H_2O - and O_2 -containing flue gas. Nevertheless, noble-metal promotion should not be viewed as a universal solution. Ru improves redox resilience, but it also increases material cost and may reduce the economic appeal of Ni-based DFMs. A more realistic design target is to identify the minimum Ru loading required to maintain cyclic redox reversibility under impurity-containing feeds.

NO_x species introduce another layer of complexity because they can affect both sorbent and catalyst functions. Unlike O_2 , which mainly changes the oxidation state of metallic sites, NO and NO_2 can form nitrate or nitrite species on alkaline sorbents and oxygen-vacancy-rich supports. These species may compete with carbonate formation, alter surface basicity and disturb the transfer of captured CO_2 species to nearby catalytic sites. Li et al. (2024b) investigated the influence of H_2O , O_2 and NO_2 on CeNiCa DFMs for integrated CO_2 capture and in-situ methanation. Their results showed that these impurities oxidized the catalyst and modified the surface chemistry, thereby affecting both capture and conversion. The NO_x effect remains less systematically understood than the effects of H_2O , O_2 and SO_2 . Future studies should distinguish the roles of NO and NO_2 , quantify nitrate accumulation during cycling, and determine whether short H_2 pulses can fully regenerate NO_x -exposed DFM surfaces.

SO_2 and H_2S can poison metallic methanation sites and react with alkaline sorbents to form stable sulfates or sulfites. These sulfur-containing species are generally more difficult to regenerate than carbonates. As a result, even low sulfur levels may cause cumulative loss of working capacity over repeated cycles. Cimino et al. (2022) studied Na–Ru/ Al_2O_3 DFM under realistic feeds containing SO_2 , H_2O and O_2 . The study reported sulfur tolerance and partial self-regeneration of Ru catalytic sites. However, this tolerance should not be interpreted as complete sulfur immunity. Cimino et al. (2024) later showed that Li–Ru/ Al_2O_3 could retain part of its methanation functionality under SO_2 -containing flue gas, but CO_2 capture capacity still declined gradually because of sulfur accumulation. This distinction is critical because a DFM may appear catalytically stable over short testing periods while its sorbent function is already deteriorating. The sulfur sensitivity of Ni-based systems appears more severe. Xie et al. (2025) provided mechanistic evidence that SO_2 deactivates Ni-based materials for integrated CO_2 capture and methanation. The decline in performance was attributed to sulfate or sulfite formation, loss of basic sites and deterioration of Ni catalytic activity. This finding is important for evaluating the real cost advantage of Ni-based DFMs. Although Ni reduces catalyst cost relative to Ru, the economic benefit may be weakened if deep upstream desulfurization becomes necessary. Therefore, sulfur tolerance should be assessed using coupled indicators such as cyclic CO_2 uptake, carbonate conversion, CH_4 yield, sulfur balance, sulfate formation and regeneration ability. CH_4 selectivity alone is insufficient because it does not reveal whether the sorbent function is being progressively lost.

Particulate and inorganic impurities are also relevant, especially for coal-fired flue gas. Chen et al. (2024b) investigated the effect of coal ash on Na–Ru/ Al_2O_3 DFM during integrated CO_2 capture and methanation. Coal ash contamination affected both the adsorbent and catalyst components. The adsorbent showed slower carbonate conversion and poorer desorption kinetics, while contamination and sintering of Ru sites contributed to the decline in methanation ability. This type of deactivation differs from molecular poisoning by SO_2 or NO_x because it

involves physical masking, pore blockage and inorganic deposition. Consequently, DFM testing with clean simulated gas may overestimate performance for coal-derived CO₂ streams. Hot-gas filtration, particulate removal and mechanically robust DFM shaping should therefore be considered part of ICCM system design. Impurities in biogas or fermentation-derived CO₂ streams differ from those in flue gas. These streams may contain high CH₄ content, moisture, trace H₂S and volatile organic or silicon-containing species. [Merkouri et al. \(2022\)](#) demonstrated integrated CO₂ capture and dynamic catalysis using brewery fermentation off-gas over a NiRuCa DFM. This type of real-feed study is valuable because it moves beyond synthetic binary or ternary gas mixtures. However, the number of real-feed ICCM studies remains limited, and DFM lifetime cannot yet be generalized across different CO₂ sources. Biogas-derived ICCM will require more attention to H₂S and siloxane tolerance because these species are known to deactivate conventional methanation catalysts and may also interact with alkaline sorbents.

A further challenge is that impurity effects are rarely additive. Steam may improve bicarbonate formation during capture but suppress methane selectivity during hydrogenation. O₂ may oxidize Ni, while Ru may partly restore reducibility. SO₂ may become more damaging in the presence of O₂ and H₂O because sulfate formation becomes more favorable. NO_x may interact with oxygen vacancies on reducible supports, changing both redox behavior and carbonate chemistry. Therefore, testing impurities one by one provides mechanistic clarity but does not fully predict long-term DFM behavior under mixed feeds. A DFM that survives individual H₂O, O₂ or SO₂ exposure may not necessarily remain stable when these species coexist.

Overall, impurities reveal a major gap between laboratory DFM performance and industrial ICCM viability. The most active material under pure CO₂/N₂ and H₂/N₂ cycling may not be the most practical material under flue gas, direct-air-capture or biogas conditions. Future impurity studies should report coupled capture–conversion indicators, including CO₂ uptake, CH₄ yield, CH₄ selectivity, carbon balance, metal oxidation state, sulfur or nitrogen accumulation, pore structure, mechanical integrity and regeneration behavior. Standardized testing should progress from dry model feed to humid feed, O₂-containing feed, sulfur- and NO_x-containing feed, and finally mixed-impurity cycling over extended periods. Promising mitigation strategies include Ru-thrifted Ni–Ru catalysts, redox-active CeO₂- or ZrO₂-containing supports, oxygen-tolerant perovskite-derived catalysts, sulfur-resistant sorbents, structured monoliths with lower pressure drop, and upstream guard beds for sulfur or particulate removal. Ultimately, impurity management must be integrated into DFM design, reactor configuration and techno-economic assessment because gas-cleaning cost, DFM replacement frequency and regeneration efficiency will determine whether ICCM can move from proof-of-concept studies to industrial deployment.

5. Emerging and advanced ICCM techniques

Electrification of chemical processes is emerging as a potential decarbonization solution of various industries, especially with the gradual availability of renewable energy ([Giannikopoulos et al., 2024](#)). In this context, microwave-powered ICCM lab-scale studies have been reported in the scientific literature. [Acher et al. \(2023\)](#) studied the performance of DFM under microwave irradiation for CO₂ capture and methanation. By physically mixing CO₂ adsorbent material (ZrO₂ and Fe₃O₄) and methanation catalyst (Ru/SiO₂), optimum performance of ~35–50 μmol CH₄/g_{DFM} was obtained, with 100% CH₄ selectivity. However, a low CO₂ conversion of only ~20% was reported and this warrants further optimisation or intensification strategies to further improve the performance and economic viability of MW-powered ICCM for commercial implementation.

In another study by [Acher et al. \(2024\)](#), Co-based supported catalyst was tested for its efficacy of CO₂ methanation under the presence of microwave, with the use of SiC and Fe₃O₄ as the microwave absorber

and CO₂ sorbent, respectively. A rather low CH₄ selectivity of 40% and CO₂ conversion of 70% were achieved at microwave power of 100 W. The authors ascribed this to the formation of CO as byproduct and recommended to vary the H₂ flowrate or pressure to boost the selectivity of CH₄. [Zhu et al. \(2024\)](#) optimized the DFM formulation, consisting of Ni-loaded CaO mixed with carbon nanotubes for ICCM operation driven by microwave ([Fig. 6](#)). At optimum Ni loading of 20 wt%, the DFM achieved CO₂ uptake of 3.31 mmol/g_{DFM}, and CO₂ conversion and CH₄ selectivity of 71.01 % and 98.17 %, respectively. As the Ni loading increased from 1 wt% to 20 wt%, its dispersion and the metallic surface area within the DFM were enhanced significantly, providing higher accessibility for in situ CO₂ hydrogenation to take place.

Apart from microwave-assisted ICCM, other innovative techniques such as induction heating, photothermal, electrocatalytic and plasma have also been appraised and are emerging as alternative routes ([Wang et al., 2025](#); [Hadi et al., 2025](#)). However, these studies only focused on the standalone CO₂ capture or CO₂ methanation process, rather than as an integrated in situ process. [Li et al. \(2021\)](#) employed non-thermal plasma in the capture and in situ conversion of CO₂ to CO, and similar emerging technology could be replicated for CH₄ production in the future. In this context, [Zhu et al. \(2025\)](#) proposed that the use of (i) photothermal CO₂ adsorbents and catalysts in a photothermal reactor, or (ii) DFMs in a reactor powered by concentrated light could be explored for ICCM concept.

In summary, the proof of microwave-assisted ICCM concept has been successfully demonstrated via various studies in the lab-scale using domestic microwave. While some studies showed promising preliminary results, extensive exploratory and fundamental studies are still required to further mature and derisk the technology. Firstly, development of DFMs that are compatible and tailored for microwave is critical. As microwave involves selective heating, the design or formulation of DFMs which incorporate the use of microwave absorber (such as SiC and carbonaceous materials) is critical for the success of microwave-driven ICCM technology. In addition, from the operation perspective, the robustness of DFMs towards the impurities in the CO₂ and H₂ gases needs to be established and understood, as most literature studies only tested binary or ternary gas mixtures which are not representative of the realistic conditions. The impact of these impurities (such as moisture, O₂, SO_x, NO_x and CH₄) needs to be evaluated thoroughly as they could impact the longevity and stability of the DFMs, which plays a crucial part of the operating expenditure (OPEX). Lastly, from techno-economic-and-environmental viewpoints, the competitiveness of microwave-assisted ICCM technology relies heavily on the availability of cheap renewable electricity and green H₂ to lower the cost of e-methane production ([Santamaría et al., 2025](#)).

6. Techno-economic analysis (TEA) of ICCM

Techno-economic analysis (TEA) is a critical tool for evaluating the viability of emerging technologies. It not only determines financial feasibility based on technical and economic data but also identifies key parameters that influence economic performance ([Chai et al., 2022](#)). This enables technical teams to assess potential risks and supports informed decision-making regarding research and development, investment, and market strategies. Given the benefits of ICCM, such as process intensification, reduced system footprint, and improved energy efficiency, the technology is gaining momentum in carbon management research. While there is increasing interest in developing DFM with higher CO₂ capture capacity, enhanced catalytic activity, and better process integration, a clear knowledge gap remains in understanding the cost-effectiveness of ICCM at an industrial scale. To the best of the authors' knowledge, only limited TEA studies have been published to date that quantify the economics of methane production via ICCM using DFM. This section assesses the techno-economic potential of ICCM comparing it with carbon capture and ex situ methanation pathway and discusses the opportunities and challenges toward driving the

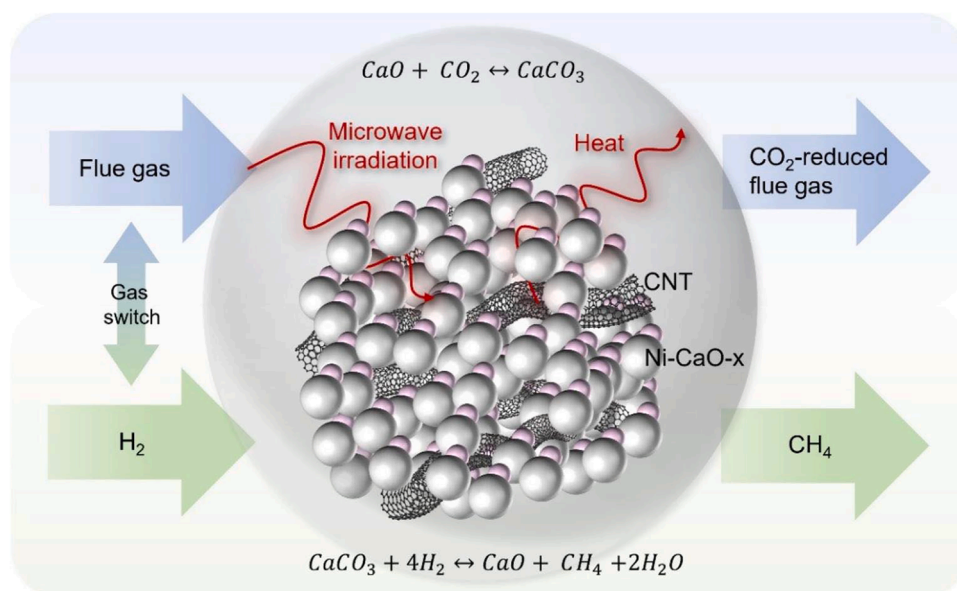


Fig. 6. Illustration on the working principle of microwave-driven ICCM using DFMs (Zhu et al., 2024).

commercial viability of ICCM. Table 3 summarizes these studies, comparing ICCM with other integrated carbon capture and methanation processes.

Lv et al. (2024) and Chen et al. (2024c) evaluated the mass and energy balances of ICCM systems that convert CO₂ from flue gas into methane. In principle, ICCM integrates CO₂ capture and methanation within a single system, employing DFM that circulates between a CO₂ capture reactor and a methanation reactor (Lv et al., 2024; Chen et al., 2024c). Taking into account the capital costs, material and utility expenses, revenues from methane sales and heat recovery, and applied a carbon tax (75.0 US\$/tCO₂e), the resulting CO₂ capture-and-conversion cost ranged from 0.98–2.0 \$/kg CO₂. Hydrogen production emerged as the primary cost driver. Among the three energy sources evaluated for hydrogen and electricity production, namely coal, wind, and solar, coal-based energy is economically superior. Wind and solar will only be cost competitive if the cost of these renewable sources become comparable to coal-based production. The estimated levelized cost of methane

(LCOM), based on CO₂-to-CH₄ molar conversion is rather high, ranged from 4.6–7.4 \$/kg, for a flue gas feed capacity of about 49 ton/hr (Chen et al., 2024c).

In the work by Lv et al. (2024), a significantly larger ICCM system was modeled, processing more than 2,100 ton/hr of flue gas from a 1000 MWe coal-fired power plant. Two fixed beds were used for CO₂ capture and in situ methanation. In this setup, CO₂ adsorbed as CaCO₃ in the first bed is decomposed in the second bed, where the released CO₂ is hydrogenated to produce methane. The heat released during methanation is directly used for CaCO₃ decomposition, enhancing the energy efficiency of the system. The overall energy consumption of ICCM was found to be three times lower than that of conventional CO₂ capture with ex situ methanation (CCM). With waste heat recovery, the projected LCOM was 0.61 \$/kg CH₄ and could be further reduced to 0.52 \$/kg CH₄ with the application of carbon tax incentives (Lv et al., 2024).

An earlier economic assessment conducted in 2018 examined the direct methanation of flue gas from a natural gas power plant as a

Table 3

Summary of TEA studies for ICCM and CCM processes using various CO₂ gas sources.

CO ₂ capture and conversion configuration	DFM/Catalyst/Archaea	CO ₂ source	LCOM (\$/kg CH ₄)	Carbon tax (\$/tCO ₂)	Reference
in-situ	unspecified	Flue gas	0.61 ^{a,b}	35.40	Iaquaniello et al. (2018)
	CaO and unspecified catalyst	Coal-fired power plant flue gas	0.52 ^a	35.40	Lv et al. (2024)
ex-situ	Na-Ru/Al ₂ O ₃ DFM	Coal-fired power plant flue gas	4.64 ^c	88.50	Chen et al. (2024c)
	unspecified	Biogas	2.56 ^{a,b}	-	Vo et al. (2018)
	CaO	Coal-fired power plant flue gas	1.17 ^a	0.00	Chirone et al. (2022)
	Cobalt/Molybdenum	Coal-to-liquid plant flue gas	2.09 ^d	11.00	Chiuta et al. (2016)
	Ni/MgAl ₂ O ₄	Cement plant flue gas	2.82 ^a	-	Chauvy et al. (2021)
	Ni-based	Automotive Shredder Residue flue gas	1.95	68.00	Wasnik et al. (2025)
	unspecified	Flue gas	1.77 ^{a,d}	77.88	De Paolis et al. (2025)
	Ni/MgAl ₂ O ₄	Direct air capture	2.30 ^{a,d}	-	Coppitters et al. (2023)
	Ni/MgAl ₂ O ₄	Direct air capture	4.58 ^{a,d}	35.00	Choe et al. (2023)
	Ni-based	Direct air capture (CaO)	5.78 ^a	0.00	Tregambi et al. (2023)
	Ni/MgAl ₂ O ₄	CO ₂	3.34 ^d	234.3	Haider et al. (2022)
	unspecified	Biogas	2.11 ^{a,d}	295.00	Elhaus et al. (2024)

(-) data are not available.

^a estimated based on 1 € = 1.18 \$

^b estimated based on volumetric cost of methane with conversion factor of 0.66 kg/Nm³ CH₄.

^c estimated based on levelized CO₂ conversion cost. Conversion rate of 0.365 kg CH₄/kg CO₂.

^d estimated based of levelized cost of energy with conversion factor of 53,940 kJ/kg CH₄.

strategy for surplus energy management. In this context, ICCM was proposed as a solution to mitigate issues related to renewable energy curtailment, which can otherwise result in economic losses, increased dependence on fossil fuels and elevated CO₂ emissions. As an alternative to conventional energy storage systems, such as pumped hydro and batteries, chemical energy storage via Power-to-Gas (PtG) pathways demonstrated a reasonably low LCOM, estimated at 0.61 \$/kg. This competitive cost was largely attributed to the assumption of cost-free renewable electricity and hydrogen production used as inputs in the process (Iaquaniello et al., 2018).

Carbon capture with ex situ methanation (CCM) systems use sequential units for CO₂ capture, typically using amine scrubbing or solid sorbent, followed by subsequent methanation in fixed-bed reactors. CO₂ feedstock for methanation originates from various sources with varying concentrations. The three main sources are industrial processes such as power plant and cement plant, direct air capture (DAC) and biogas from anaerobic digestion of animal waste or agricultural residues. When compared to CCM, ICCM requires lower capital investment by reducing the number of reactor and heat exchanger. Eliminating external heating requirement for sorbent regeneration contributes to operation expenses saving (Lu et al., 2024; Freyman et al., 2023). An overall energy efficiency of up to 89% has been reported for ICCM systems that utilize excess heat from the exothermic methanation reaction for in situ CO₂ desorption (Chen et al., 2024c).

Across reported TEA cases, LCOM spans a wide range, with variability dominated by different boundary conditions, which include the assumed price of H₂, electricity supply, source of CO₂, CO₂ credit and plant scale. The LCOM gathered from multiple CCM studies ranges from 1.17 to 2.9 \$/kg for flue gas capture. However, DAC incurs significantly higher costs due to the dilute nature of atmospheric CO₂ and the associated energy penalty for capture (Choe et al., 2023; Tregambi et al., 2023). Biogas presents a compelling alternative, as it contains a relatively high concentration of CO₂. In biogas-based CCM system, hydrogen can be injected into a methanation reactor containing CO₂-rich biogas, either direct from anaerobic reactor or post CO₂ capture. Unlike catalytic systems, these configurations employ hydrogenotrophic methanogenic microbes to convert CO₂ and H₂ into methane under milder temperature and pressure conditions. Another study found that in situ biological methanation offers a marginally lower methane selling price (approximately 2.56 \$/kg) than conventional biogas upgrading routes with prior CO₂ separation (2.68 \$/kg) (Vo et al., 2018). In terms of near-term commercial feasibility, CCM processes utilizing flue gas or biogas are currently more attractive than ICCM with DFMs, owing to their higher technology readiness levels (TRLs) and operational maturity (Long and Murphy, 2019; Pesuit et al., 2017; Younas et al., 2016).

For future implementation, ICCM process shows promising potential, particularly due to process simplification, internal heat integration, and lower capital investment. Nevertheless, the LCOM of synthetic methane remains above market prices for natural gas (~\$0.506/kg) in the absence of carbon incentives (Lv et al., 2024). The economic viability of ICCM systems is highly sensitive to the cost of green hydrogen and the carbon pricing mechanisms. Despite the early promise, current TEA studies for ICCM often rely on simplified models that lack detailed thermodynamic, kinetic, and mass transfer considerations. Many simulations assume ideal simultaneous CO₂ adsorption and catalytic conversion using DFM, without accounting for degradation effects. Contaminants such as O₂, SO₂, and H₂O in the feed gas have been shown to reduce adsorption capacity and catalytic performance, emphasizing the need for detailed studies on DFM deactivation and impurity accumulation (Zheng et al., 2016; Jeong-Potter et al., 2022b; Cimino et al., 2023).

Furthermore, catalyst durability, including mechanical stability under cyclic operation, is critical for real-world viability (Gharamaleki et al., 2025; Ströhle et al., 2020). Advancing ICCM technologies toward industrial readiness will require robust engineering design of dual-function reactors, particularly for effective heat management and

resistance to sintering or attrition. In conclusion, while ICCM presents an attractive pathway for potential cost-effective and compact methane production from CO₂, comprehensive techno-economic and durability studies are needed to fully realize its potential in the decarbonization landscape.

7. Life cycle assessment (LCA) of ICCM

ICCM using DFMs offers a novel pathway that combines CO₂ capture and conversion to synthetic methane in a single step. This integrated configuration offers potential advantages in energy efficiency, system simplification, and overall environmental performance across the process life cycle (Liu et al., 2023). However, despite the rapid expansion of experimental studies on DFMs at low TRLs, comprehensive life cycle assessment (LCA) studies dedicated to ICCM remain relatively limited compared to laboratory-scale performance evaluations (Sun et al., 2021a).

Existing LCA-related assessments of ICCM exhibit methodological heterogeneity, particularly in terms of functional unit and system boundary definitions. Reported functional units include 1 kg CH₄ produced, 1 kg CO₂ captured and converted, and 1 MWe-h electricity generated, which complicates direct benchmarking against conventional carbon CCM systems or other CO₂ utilisation routes. Similarly, system boundaries vary from gate-to-gate simulations to cradle-to-gate analyses incorporating hydrogen production and material synthesis. Only limited assessments extend toward broader life-cycle considerations, including material replacement and end-of-life management. Such variability in methodological assumptions significantly influences reported GHG intensities and resource consumption metrics. This section reviews the few existing LCA studies available to date that have been performed for ICCM process using DFMs, in which the focus was found to be majorly devoted to evaluating either emissions, energy use or resource consumption within a confined boundary or section of the process, necessitating further studies in this area.

Across available studies, several environmental hotspots consistently emerge. H₂ production is identified as the dominant contributor to global warming potential. When H₂ is derived from fossil resources, life-cycle emissions increase substantially conversely, renewable H₂ from wind or solar electrolysis markedly improves environmental performance. Chen et al. (2024c) demonstrated that under renewable wind-powered hydrogen, ICCM could achieve optimal net CO₂ emissions of -1.57 kg CO₂ per kg CO₂ captured (considering CO₂ emissions offsets of the produced methane and heat recovery), with a comprehensive energy consumption of -20.68 MJ per kg CO₂. The study further reported that ICCM systems could reach 89% energy conversion efficiency, comparable to conventional systems, indicating enhanced heat recovery potential. However, the environmental and economic viability of ICCM remains highly sensitive to hydrogen cost. When renewable H₂ falls below \$1.4/kg, comparable to coal-based hydrogen, ICCM becomes both environmentally and economically competitive.

Electricity and heat supply represent another major hot spot, particularly under elevated reaction pressures. Chen et al. (2024a) investigated ICCM performance under varying pressures and reported that at 5.0 MPa, electricity consumption was nearly identical across different DFM-based configurations. Although CO₂ methanation is strongly exothermic ($\Delta H^\circ \approx -165 \text{ kJ mol}^{-1} \text{ CO}_2$), insufficient heat integration may reduce the anticipated energy advantages. Nevertheless, integration of capture and methanation units has demonstrated thermal efficiency improvements of approximately 5% compared to conventional Sabatier processes (Kothandaraman et al., 2021), highlighting the potential benefit of process coupling.

DFM manufacturing, stability, and circulation rate constitute an additional environmental and resource hotspot. The effective design of DFMs with well-integrated adsorption and catalytic sites plays a pivotal role in governing carbon emissions, energy utilisation, and material consumption (Sun et al., 2024b). Nickel (Ni) and ruthenium (Ru) are

widely recognized as highly active methanation catalysts (Tsotsias et al., 2021), and their incorporation into DFM has been extensively studied to enhance CH₄ selectivity (Sun et al., 2022b). For instance, Ce-modified CaO-supported Ni catalysts demonstrated efficient operation at 320°C without additional heat input (Sun et al., 2019), contributing to improved process sustainability relative to stepwise CCM systems. Wang et al. (2017) reported that Ru-Na₂CO₃ exhibited stable performance over at least 10 adsorption-methanation cycles under O₂-free CO₂ feed, indicating durability benefits that reduce material replacement frequency. However, Chen et al. (2024c) highlighted that Ru-based DFMs such as Ru-Na₂CO₃/Al₂O₃ suffer from high cost and limited resistance to flue gas contaminants. Alternative formulations, including NaNO₃-promoted Ni/MgO, showed higher CO₂ adsorption capacity but lower methanation reactivity and selectivity, illustrating the performance trade-offs that influence both material efficiency and life-cycle impact.

Comparative DFM scenario analysis further reveals resource-related trade-offs. Chen et al. (2024a) reported that Na-DFM achieved superior energy conversion efficiency (82.74%), comprehensive energy consumption (-14.61 MJ/kg CO₂), and net CO₂ emissions (-1.18 kg CO₂/kg CO₂) relative to Mg-DFM and Li-DFM systems. Additionally, Na-DFM enabled greater SNG production and enhanced waste heat and water recovery. However, this improved performance required a substantially higher circulation rate of 94.22 kg/kg CO₂, compared to 8.31 kg/kg CO₂ for Mg-DFM and 4.21 kg/kg CO₂ for Li-DFM, thereby increasing material demand and potentially influencing embodied emissions. These findings demonstrate that DFM composition, active component fraction, and capture-hydrogenation mechanisms significantly affect both process efficiency and life-cycle sustainability.

Process-level comparisons between ICCM and CCM systems further highlight integration benefits. Lv et al. (2024), using Aspen Plus simulation, reported that ICCM emitted 83.6 kg CO₂ per MWe-h compared to 148.93 kg CO₂ per MWe-h for CCM when waste heat recovery was incorporated. ICCM also reduced sorbent loss to 0.5%, compared to 5% in CCM, thereby lowering material replenishment demand. Moreover, the ICCM configuration required only one-third of the non-renewable coal consumed by CCM to complete carbon capture in a 1000 MWe coal-fired power plant scenario, demonstrating additional resource conservation advantages.

Overall, while dedicated cradle-to-grave LCAs specifically focused on ICCM with DFMs remain scarce, preliminary findings consistently indicate reduced life-cycle emissions and improved energy utilization relative to conventional CCM, particularly under renewable hydrogen integration and effective heat recovery. However, reported environmental benefits are highly sensitive to functional unit selection, system boundary definition, hydrogen source, electricity mix, DFM production assumptions, and catalyst lifetime. The methodological heterogeneity observed across studies underscores the importance of transparent reporting and harmonized LCA frameworks to enable robust benchmarking against alternative CO₂ utilization technologies. Without such consistency, differences in reported environmental performance may reflect variations in assumptions rather than intrinsic technological superiority. Continued systematic LCA evaluations are therefore essential to accurately quantify the environmental potential of ICCM within the broader carbon capture and utilization landscape.

Therefore, continued research and development are essential to optimize system performance, address existing limitations, and fully realize the environmental potential of this emerging technology. In addition, conducting a comprehensive LCA using established tools and reliable databases is critical for accurately evaluating the full spectrum of environmental impacts associated with ICCM. Given the emerging nature of ICCM technologies, a cradle-to-grave assessment, encompassing raw material extraction, operational phases, and end-of-life treatment, is vital to uncover potential environmental trade-offs and unintended consequences that may not be apparent through partial or simplified evaluations. Utilizing standardized and systematic LCA

methodologies, such as those aligned with ISO 14040/44 guidelines, ensures transparency, reproducibility, and scientific rigor in the assessment process. This methodological consistency not only enhances the credibility of individual studies but also facilitates robust benchmarking and cross-comparison among various technologies, system configurations, and process conditions reported in the literature. Without this level of methodological alignment, conclusions drawn from disparate studies may be misleading or incompatible, impeding evidence-based decision-making by policymakers, researchers, and industry stakeholders. Therefore, a methodical, tool-based LCA approach is indispensable for positioning ICCM as a verifiable and environmentally sound solution in the broader context of carbon capture and utilization (CCU) strategies. Additionally, a summary table has been added Table 4 to harmonize and compare assumptions across available ICCM LCA studies.

8. Challenges, outlook and future perspectives for industrial deployments

ICCM via DFMs is a critical enabler to the realization of potential efficient and cost-effective decarbonization by strategizing process intensification to lower the footprint of conventional CCM. Based on the review conducted, the concept has been proven quite successfully in laboratory settings and is currently at TRL 4. To further mature and derisk the technology towards industrial implementation, there are still some technical gaps to be critically addressed, which can be summarized into 3 facets (namely DFM engineering, reactor design/configuration and technology integration), as illustrated in Fig. 7.

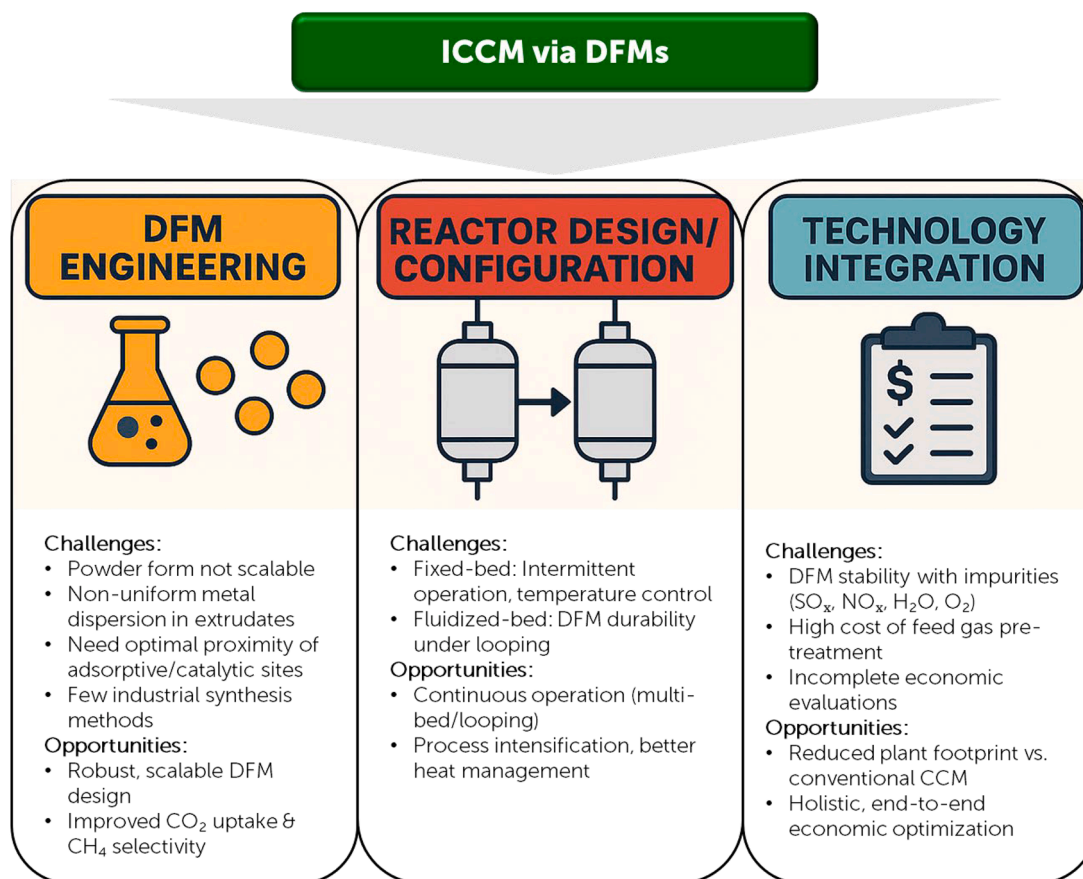
First and foremost, DFM engineering or design is the core aspect of the ICCM technology. Current state-of-the-art reveals the primary focus of most studies is still exploratory and fundamental in the context of DFM formulation and synthesis route optimization, both of which play direct role in dictating the physicochemical properties of DFMs and their efficiency of carbon capture and conversion to CH₄. In particular, DFMs in powder form currently being used for laboratory evaluation is inappropriate for commercial adoption in which pelletized, extrudate or monolithic forms are preferable for minimizing the pressure drop across the reactor bed. However, some studies reported the non-uniform dispersion of active metals in extruded DFMs. Besides, another challenge in DFM design is to engineer optimal spatial proximity between the adsorptive and catalytic species within the DFM matrix, balancing the diffusion of adsorbed CO₂ to nearby catalytic sites for methanation reaction via the spillover mechanism (Ahmadov et al., 2024), while preventing the catalytic sites to be covered by the adsorbed CO₂. Theoretically, the amount of CH₄ produced depends heavily on the amount of CO₂ captured. Furthermore, the selection and loading of active metals in DFMs will play a part in their resistance towards O₂. For example, the reducibility of nickel to its active metallic state is lower compared to that of ruthenium (Tsotsias et al., 2025). Hence, the challenges stated above should be addressed delicately by engineering robust DFMs towards optimizing CO₂ uptake, CO₂ conversion and CH₄ selectivity for realistic operating envelope of ICCM. Apart from that, the scalability of DFM synthesis method should also be taken into consideration during the early stages of research and development. Although various DFM synthesis techniques are attempted by scholars (Section 3), only few techniques are industrially established and commercially feasible for mass production of DFMs (e.g., co-precipitation and impregnation (Munnik et al., 2015)).

Another aspect to scrutinize is the reactor design and configuration for ICCM process via DFMs. There are a few reactor configurations proposed for industrial ICCM process; fixed-bed reactor (Faria et al., 2025) or interconnected fluidized-bed reactors (Chen et al., 2024a). These configurations will determine the operating mode of the ICCM process. For fixed bed reactor, intermittent switching between CO₂-containing gas and H₂ would be required for a single-unit operation, as the DFM in the reactor needs to be saturated with CO₂ before the

Table 4

Summary of LCA studies on ICCM systems.

Study	Functional unit	System boundary	H ₂ source	Key hotspot identified	Main findings
Sun et al. (2021a)	1 kg CH ₄	Cradle-to-gate	Grid/fossil H ₂	H ₂ production	Integrated system reduces compression/transport emissions
Lv et al. (2024)	1 MWe-h	Gate-to-gate (Aspen simulation)	Process-integrated	Electricity consumption	ICCM emitted 83.6 kg CO ₂ /MWe-h vs 148.93 kg CO ₂ /MWe-h (CCM)
Chen et al. (2024a)	1 kg CO ₂	Industrial-scale scenarios	Renewable H ₂	DFM circulation rate	Na-DFM showed improved energy efficiency but higher circulation demand
Chen et al. (2024c)	1 kg CO ₂ captured	Cradle-to-gate	Wind / Solar H ₂	H ₂ source sensitivity	Net emissions -1.57 kg CO ₂ /kg CO ₂ (wind)

**Fig. 7.** Challenges and outlook of ICCM technology via DFMs from three main perspectives.

methanation step. This would subsequently result in intermittent CO₂ capture and production of methane. In this context, two or more units of reactor would be needed for continuous CO₂ capture and methanation by switching CO₂-containing gas and H₂ feed streams between the reactors, similar to the concept of pressure swing adsorption. Besides, the control of reactor temperature, which could be challenging due to the endothermic and exothermic nature of adsorption and methanation, respectively, is critical as it could affect the overall process kinetics and subsequently the cyclic operation. As for interconnected fluidized-bed reactors, DFM is circulated in between two reactors (adsorption/carbonation and methanation) in a chemical looping manner. However, information on DFMs' durability under fluidized bed chemical looping conditions is still lacking in the literature and warrants further investigation (Massa et al., 2024).

Lastly, from the technology integration perspective, the economic feasibility of ICCM technology as a potential substitute to conventional established CCM technologies entails further evaluation. ICCM via DFMs could potentially reduce the footprint of conventional CCM

technologies. However, the robustness, tolerance and stability (lifespan) of DFMs when exposed to various impurities in the feed gas (e.g., SO_x, NO_x, H₂O, O₂, etc) could directly impact the overall economics. The pre-treatment or conditioning of feed gas prior to the carbon capture and/or methanation processes often constitutes a significant portion to the overall production cost due to stringent requirements (Scholes et al., 2016). These aspects are often being excluded from current economic evaluation in the literature (Kim and Léonard, 2025). Hence, it is imperative to optimize the robustness and stability of DFMs against these impurities and to conduct a holistic end-to-end economic evaluation for the entire plant's battery limit (i.e., not just limited to the ICCM technology package but inclusive of feed gas pre-treatment or conditioning, post-processing, DFMs replacement/change out, etc).

9. Conclusions

ICCM via DFMs offers a promising route to intensify CO₂ capture and methanation, potentially reducing unit operations and enabling

modular deployment. The reviewed literature demonstrates that DFM's composition, synthesis route, and cyclic operating strategy jointly control CO₂ uptake, conversion, CH₄ selectivity, and durability. Among various DFM synthesis methods, co-precipitation and impregnation are commercially mature. In terms of ICCM process, temperature and pressure play crucial influence in the performance of DFMs for CO₂ capture and methanation. Based on the review, operating temperature ranging from 300 – 550 °C is commonly reported for ICCM process, depending on the DFMs used, feedstock quality and system configuration. As DFMs reported are mostly intended for CO₂ capture from flue gas, the operation pressure would be below 5 bar, though some modelling studies simulated the performance up to 50 bar. TEA studies reveal that ICCM shows strong potential for future implementation due to process simplification, heat integration, and lower capital costs, but its economic viability remains highly dependent on green hydrogen prices and carbon incentives as current production cost of SNG is still higher than the market price of natural gas. Moving beyond lab scale requires (i) standardized reporting and benchmarking (complete carbon balance, cyclic metrics, and comparable operating conditions), (ii) long-term stability tests under realistic feeds (impurities such as H₂O, CO, SO_x, NO_x, sulfur/chlorine species), (iii) reactor and heat-management strategies to mitigate hotspots and maintain sorbent/catalyst integrity, and (iv) harmonised TEA/LCA with transparent assumptions. Addressing these priorities will clarify the realistic deployment niches of ICCM and accelerate translation to pilot-scale demonstrations. Lastly, enhancement and development from three perspectives, i.e., DFM engineering, reactor design/configuration and technology integration, are needed to achieve industrial readiness and successful deployment of the ICCM technology.

CRediT authorship contribution statement

Yi Herng Chan: Writing – review & editing, Writing – original draft, Visualization, Conceptualization. **Slyvester Yew Wang Chai:** Writing – original draft. **Bing Shen How:** Writing – original draft. **Chung Loong Yiin:** Writing – original draft. **Bridgid Lai Fui Chin:** Writing – original draft. **Serene Sow Mun Lock:** Writing – original draft. **Mee Kee Wong:** Writing – original draft. **Muhammad Khairumuzdanial Md Khair-ulanwar:** Writing – original draft. **Shin Ying Foong:** Writing – original draft. **Su Shiung Lam:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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