Thermocatalytic Decomposition of Methane: A Review on Carbon-Based Catalysts

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Table S1. Summary of recent ordered-mesoporous carbons used for methane decomposition studies in the literature.

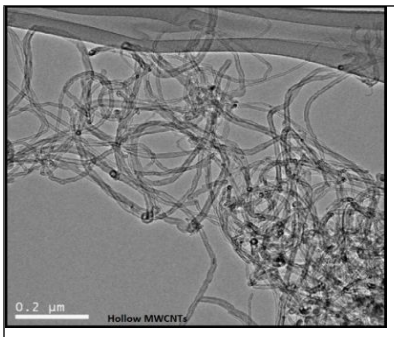
Catalyst	BET Surface area, $m^2\ g^{-1}$	Temperature, °C	Methane conversion	Hydrogen produced, mol. g cat^{-1}	Reference
CMK-5	1940	1000	NA	0.95	[1]
CMK-3	1323	1000	NA	0.45	[1]
OMC	2154	850	28% to 5%	0.275	[2]
CMK-3	1400	950	NA	1.52	[3]
DUT-19	2420	950	NA	1.27	[3]
Ni/3DOMC	884	850	58% to 50%	NA	[4]

CMK: carbon mesostructured by KAIST
OMC: ordered mesoporous carbon
DUT: carbide-derived carbon developed at Dresden University of Technology
3DOMC: three dimensionally ordered mesoporous carbon

Table S2. Initial CDM conversion rates of carbon obtained from various carbon sources, at 850°C [5].

S.No.	Carbon type	Source	Initial CDM rate per unit mass, K_m (T= 850°C) (mmol/min.g)
1.	Activated carbon	Coconut shell, KE	1.76
2.	Activated carbon	Hardwood	2.04
3.	Activated carbon	Lignite	1.77
4.	Activated carbon	Peat	1.63
5.	Activated carbon	Phenol, resin	1.66
6.	Activated carbon	Petroleum coke	1.43
7.	Glassy carbon	-	0.95
8.	Carbon black	Black Pearls 120	0.22
9.	Graphite	Natural	0.02
10.	Fullerene soot	-	1.9

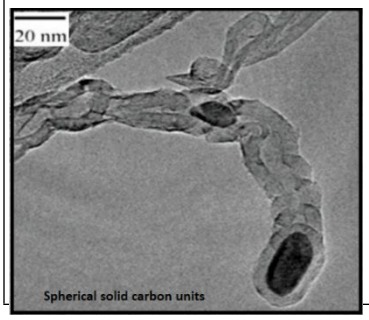
Table S3. Overview of filamentous carbons produced during the CDM reactions under different reaction conditions.



Reaction conditions: High temperature conditions

- Carbon nucleation rate > diffusion rate
- Nucleation/precipitation near metal/gas interface
- Examples: 3%Ru/Ac, activated biochar.

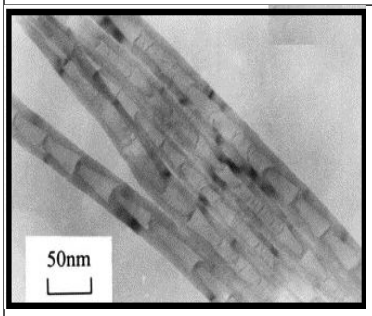
Hollow MWNTs, CNTs, and Carbon microtubes. Reprinted with copyright permission from [6].



Reaction conditions: Low temperature conditions

- Carbon nucleation rate < diffusion rate
- Nucleation/precipitation near metal/support interface
- Examples: 30%Fe/AC, K_2CO_3/CC (Coal char), 10%Fe/AC, 10%Ni/C (coal char)

Solid MWCNTs, CNFs. Reprinted with copyright permission from [7]



Reaction conditions: High temperature conditions

- Traces of hydrogen in the feed + promoter
- Examples: Ni-Cu/CNT.
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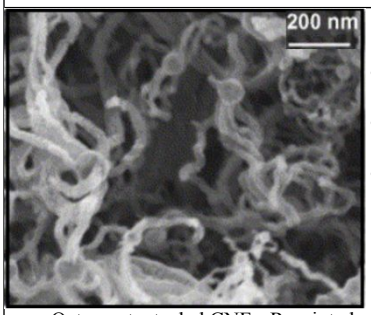
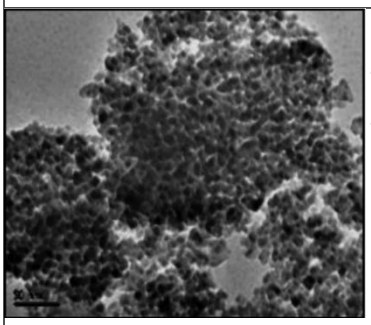
Bamboo- CNTs. Reprinted with copyright permission from [8].



Reaction conditions: Low temperature conditions

- Traces of hydrogen in feed
- Comparatively larger particle sized metal catalysts under testing
- Examples: 1%Ni/graphite

Octopus tentacled CNFs. Reprinted with copyright permission from [9].



Reaction condition: very high temperature conditions (>800°C)

- Non-supported catalysts
- Examples: Fe/graphite, Fe_2O_3/graphite

Nano-onions carbon deposits. Reprinted with copyright permission from [10]

Table S4. Summary of few studies concerning the catalyst-regeneration techniques and the conclusions.

Catalyst regeneration technique	Catalyst used	Conclusion	Reference

Pulsed or continuous introduction of oxidizing agents, such as air, CO_2, and CO_2 under cyclic decomposition of methane	Carbon black, activated carbon, carbon nanofibers	A portion of the original catalyst was also gasified besides deposited carbon, rendering the process insufficient to retain the original activity of catalyst.	[11,12]
Deep regeneration by chemical looping combustion (CLC) using H_2O and O_2 as gasifying agents	Activated carbon	More than 30% of spent catalyst could be recovered with heat generation that in turn would assist in methane decomposition.	[13]
Simultaneous dosing of CO_2 and CH_4 as feedstock	Activated carbon (pine wood biomass derived)	Efficient regeneration with increased H_2 yield due to the simultaneous processes of carbon deposition, oxidation of deposit, decomposition of its structure by CO_2, and partial oxidation of the original catalyst.	[14]
Gasification of spent catalyst with steam and CO_2 mixture	Activated carbon	Increase in catalyst porosity and methane decomposition rate.	[15]

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