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Novel Electrified Thermal Hydrogen Production Approaches

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Abstract. In this study, we investigated four advanced electrification-based hydrogen (H_2) production approaches, including direct Joule heating-assisted steam methane (CH_4) reforming (DJH-SMR), microwave (MW) heating-assisted chemical looping dry reforming of CH_4 (MWH-CLDRM), microwave heating-assisted CH_4 thermal pyrolysis (MWH-MTP), and microwave heating-assisted low-density polyethylene (MWH-LDPE) pyrolysis. In the DJH-SMR, we employed an electrically conductive catalyst in a fluidized bed and achieved 96% CH_4 conversion with H_2/CO ratio of over 3 (CO is the carbon monoxide) at bulk temperature T_b of 900°C and steam-to-carbon molar ratio of 1. In the MWH-CLDRM, we leveraged MW irradiations to selectively heat magnetite (Fe_3O_4), achieved 97% CH_4 conversion at T_b of 800°C in the fuel (reducer) reactor with an H_2/CO ratio of 2, while suppressed coke formation. With the MWH-MTP, we demonstrated 23% CH_4 conversion at T_b of 1065°C with 98% H_2 selectivity, while capturing 90% of produced solid carbon. The MWH-LDPE pyrolysis over an iron-nickel-alumina catalyst at T_b yielded 72% H_2 and 52% of carbon nanotubes. Obtained results highlighted the DJH-SMR's potential for H_2-rich streams, MWH-CLDRM's suitability for Fischer-Tropsch applications, MWH-MTP's promise for CO-free H_2 production when powered by renewable electricity, and MWH-LDPE pyrolysis's dual benefit of waste valorization and H_2 generation. Each approach exhibits high performance and scalability.

Key words. Electrified thermal hydrogen production, direct Joule heating, microwave heating, methane pyrolysis, chemical looping dry reforming of methane, plastic waste pyrolysis.

1. Introduction

The transition toward sustainable energy systems hinges on large-scale adoption of low-carbon technologies, while hydrogen (H_2) plays a vital role in this transformation. As a versatile energy carrier, H_2 enables deep decarbonization across hard-to-electrify sectors, such as chemical manufacturing, steel production, and heavy transport. However, current industrial-scale H_2 production approaches, in particular, steam methane (CH_4) reforming (SMR), remain carbon intensive. Industrial SMR plants typically emit about 9-11 kg of carbon dioxide (CO_2) per kg of H_2 produced [1]. The CO_2 emissions from the SMR plants mainly arise from the combustion of fossil fuels to provide the necessary heat, accounting for around 60% of the total emissions. The remaining emissions are produced from the chemical reactions involved, such as the water-gas shift reaction to increase the H_2 production rate and the incomplete reforming of natural gas, which undergoes secondary reactions or combustion, leading to CO_2 emission. CO_2 contributes to climate change by trapping heat in the atmosphere, leading to global warming, extreme weather, and ecosystem disruptions. These challenges highlight the urgent need for more sustainable H_2 production technologies.

Process electrification, especially through advanced heating methods, such as Joule and microwave (MW) heating, has emerged as a promising approach to decrease CO_2 emissions and process intensification [1,2]. Direct Joule heating (DJH) has a rapid, selective, and volumetric heating nature when exposed to electrically conductive materials, e.g., catalysts. By passing an electric current through a bed of electrically conductive catalysts, we can directly and uniformly heat the catalyst bed. In the study by Wismann et al. [3], the authors demonstrated DJH's effectiveness in SMR adopting a nickel (Ni)-zirconia (ZrO_2) catalyst coated on a metallic felt substrate (iron−chromium−aluminium tube), achieving a promising CH_4 conversion and H_2 yields. However, development of electrically conductive catalysts is crucial for a successful integration of DJH into catalytic processes.

MW heating (MWH) has gained significant attention due to its ability to rapidly, selectively, and volumetrically heat catalysts and/or solidus/liquidous reactants with appropriate electromagnetic properties [4]. These help enhance energy efficiency and minimize heat losses compared to conventional heating methods. Several studies have explored application of MWH in catalytic processes, including CH_4 reforming [4–6]. In the study by Fidalgo et al. [6], the authors reported that MWH-

assisted dry reforming of methane (MWH-DRM) with a Ni catalyst supported on activated carbon could improve CH_4 conversion and decrease solid carbon deposition. Likewise, in the study by Zhang et al. [5], the authors observed over 90% CH_4 and 95% CO_2 conversion at 800 in MWH-DRM adopting a platinum (Pt)/alumina (Al_2O_3) catalyst. These findings show the potential of MWH for process intensification in CH_4 reforming. Recent advances in MWH-assisted CH_4 thermal pyrolysis (MWH-MTP) have also demonstrated a significant potential for CO_2-free H_2 production [7]. This process directly decomposes CH_4 into H_2 and solid carbon, eliminating CO_2 emissions when powered by renewable electricity. The selective heating capability of MWs creates unique temperature gradients between solid and gas phases, enhancing both conversion efficiency and solid carbon capture compared to conventional heating.

Plastic gasification and pyrolysis are alternative sustainable approaches to produce circular H_2. Plastic pyrolysis offers advantages over gasification for H_2 production, including lower operating temperatures, minimized CO_2 emissions, and production of valuable byproducts, such as liquid hydrocarbons and solid carbon. Plastic pyrolysis, coupled with heterogeneous catalytic cracking, offers a more selective and controlled conversion approach compared to conventional thermal pyrolysis. Although plastic catalytic pyrolysis effectively produces H_2, its endothermic nature necessitates substantial energy input, often resulting in massive CO_2 emissions [8]. The catalytic pyrolysis of plastic waste, in particular, polyethylene, has been explored to produce valuable chemicals and fuels [8,9]. The incorporation of MW heating in pyrolysis has demonstrated potential for process intensification, enabling rapid and selective heating of feedstocks [8]. Catalysts play a key role in optimizing product yields and selectivity, in particular, for H_2 and aromatics, in plastic waste pyrolysis [9].

Advancing process electrification strategies, novel catalytic materials, and process intensification for sustainable H_2 production is critical in the transition to cleaner energy and climate change mitigation. By application of Joule and MW heating with catalytic CH_4 reforming or thermal CH_4 pyrolysis and plastic waste pyrolysis, in this study, we aim at contributing to the development of more efficient and environmentally friendly H_2 production approaches. Findings of this study will offer valuable insights into the potential of electrified CH_4 reforming and pyrolysis and plastic waste valorization, paving the path for further innovations in this field.

2. Experimental

We conducted DJH-assisted SMR (DJH-SMR) (Figure 1(a)) and MWH-MTP (Figure 1(b)) experiments in fluidized bed reactors (FBRs) to enhance heat and mass transfer efficiencies. In the DJH-SMR, we developed and experimentally evaluated a direct Joule heated FBR (DJH-FBR) for SMR with a novel electrically conductive Ni/molybdenum-carbide (Mo_2C)/Al_2O_3 catalyst. We synthesized the catalyst to combine high reforming activity of Ni, excellent electrical conductivity and coke resistance of Mo_2C, and mechanical stability and high surface area of Al_2O_3 support. For the reactor design, we employed axially aligned electrodes, with electrical current flowing through the fluidized catalyst bed to generate heat due to the bed electrical resistance as per the ohmic law. We investigated the reactors' performance across a range of bulk temperature T_b (700–900°C), steam-to-carbon (S/C) ratios (1–3), and superficial gas velocities U_g (5-20 cm/s), with a focus on evaluating the reactor performance, CH_4 conversion, H_2 yield, and operational stability of DJH-SMR.

In MWH-MTP, we employed MW absorber silicon carbide (SiC) particles in a FBR under MW irradiations at 2.45 GHz, leveraging their dielectric properties to achieve T_b as high as 1065°C, while capturing solid carbon deposits. We systematically studied the effects of T_b (900-1065°C), residence time (0.5-1.5 s), and inlet CH_4 concentration (20-50 mol.%) at U_g (5-15 cm/s) on CH_4 conversion and H_2 selectivity. We measured temperature distributions in the FBR employing pyrometers and thermocouples and assessed carbon deposition via Raman spectroscopy and mass balance [7].

We performed MWH-assisted chemical looping dry reforming of methane (MWH-CLDRM) in a fixed bed, employing magnetite (Fe_3O_4) as an oxygen carrier, creating selective thermal gradients that promoted reduction and oxidation (redox) reactions (see Figure 1(c)). We investigated the system's performance at T_b of 650-800°C, varying CH_4 concentrations (1.5-10 vol.%), and different CH_4-to-solid ratios (2-27 cm^3 $CH_4/g_{Fe3O4}\cdot min$), focusing on CH_4 conversion, H_2/CO ratio (CO is the carbon monoxide), carbon deposition prevention, and redox cycle stability [10]. Likewise, we investigated MW heating-assisted low-density polyethylene (MWH-LDPE) pyrolysis in a two-stage fixed bed system (see Figure 1(d)). In the first stage we carried out thermal decomposition at 500°C, followed by a catalytic chemical vapor deposition (CVD) at 800°C. SiC particles served as MW absorbers in the first stage, while an Al_2O_3-supported iron-nickel (FeNi) catalyst facilitated carbon nanotube (CNT) growth in the CVD stage. The investigation focuses on LDPE, which constitutes a major fraction (59 wt.%) of municipal solid waste. MW interaction with the deposited carbon promotes selective heating due to its high dielectric properties, accelerating the growth of CNTs. We tested various transition metal catalysts and performed 10-cycle tests on spent catalyst beds to evaluate catalyst stability.

We characterized the catalysts before and after reaction employing X-ray diffraction (XRD), scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM/EDX), Raman spectroscopy, and X-ray photoelectron spectroscopy (XPS) analysis. We evaluated performances of DJH-SMR, MWH-CLDRM, MWH-MTP, and MWH-LDPE pyrolysis based on CH_4 and plastic conversions, product yields, H_2 selectivity, and solid carbon formation, and compared to conventional heating to highlight the advantages of process electrification. We also monitored product gases by a Varian CP-4900 Micro Gas Chromatograph (GC).

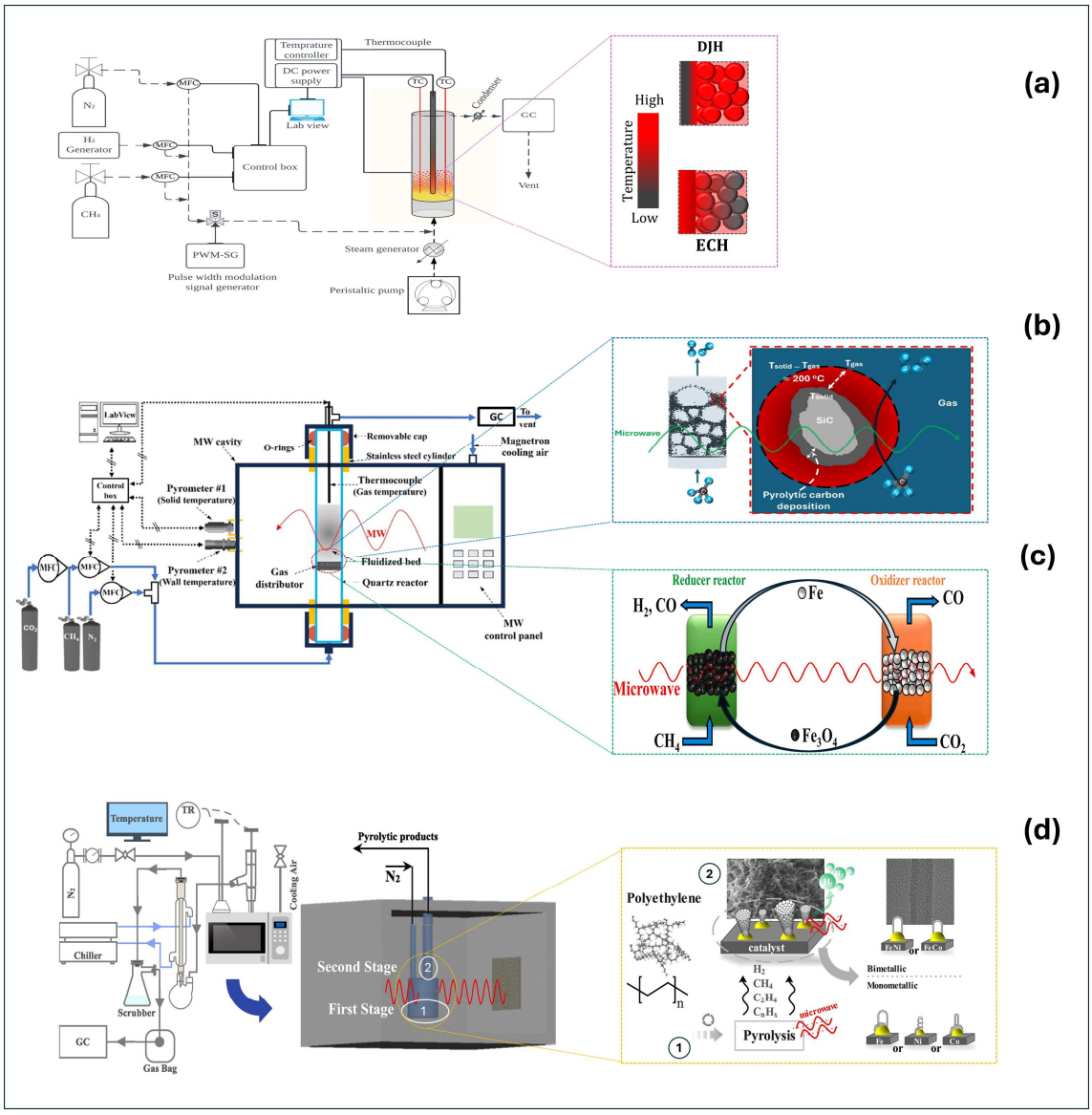


Fig.1. A schematic of (a) DJH-SMR, (b) MWH-MTP, (c) MWH-CLDRM, and (d) MWH-LDPE pyrolysis.

3. Results and Discussion

We presented details of H_2 production through DJH-SMR, MW-CLDRM, MW-MTP, and MW-LDPE pyrolysis and their features in the following subsections.

3.1. DJH-SMR

DJH-FBR configuration allowed localized, uniform heating of the catalyst particles overcoming the limitations of external conventional heating (ECH) methods that suffer from temperature gradients in the reactor, inefficient heat transfer, and delayed response times. Our results prove that DJH-SMR consistently outperformed external conventional heated FBR (ECH-FBR) of SMR by over 10% CH_4 conversion across all tested T_b. For instance, at T_b=700°C, the measured CH_4 conversion in the DJH-SMR matched the performance of ECH-SMR operated at T_b of 800°C, confirming that higher catalyst solid temperatures T_s created by DJH, enhanced reaction rates in DJH-SMR. The $Ni/Mo_2C/Al_2O_3$ catalyst demonstrated its highest activity at S/C molar ratio of 1, achieving up to 96% CH_4 conversion and a stable H_2/CO ratio of ~3. The activity of the produced catalyst was comparable to the commercial Ni-based SMR catalysts at the same operating conditions. Operating DJH-SMR at S/C=1 significantly decreases water and energy consumption, required for steam generation. Specifically, decreasing the S/C ratio from 3 to 1 decreases water usage by two-thirds and lowers the energy required for steam generation by approximately 150 kJ/mole of CH_4. This yields an overall energy savings of 30–35%. In addition, the incorporation of Mo_2C improved coking resistance and allowed stable activity at low S/C ratios, conditions under which commercial SMR Ni-based catalysts often deactivate rapidly due to carbon deposition.

We characterized fresh and spent $Ni/Mo_2C/Al_2O_3$ catalysts to assess structural integrity, phase composition, and changes induced during SMR operation. XRD of the fresh catalyst confirmed the successful formation of Ni and Mo_2C phases, while the spent catalyst showed the emergence of MoO_2 peaks, showing partial oxidation of Mo_2C during reforming (see Figure 2). XPS results

revealed the transformation of surface Mo species from carbide to oxide states, supporting the oxidation of the catalyst at higher S/C ratio. Despite these changes, we observed no sintering or structural degradation via SEM for the operating conditions explored here (see Section 2), and the catalyst maintained its porous structure. These findings show the stability and regenerative potential of the $Ni/Mo_2C/Al_2O_3$ catalyst, at low S/C operation, where Mo_2C facilitates a balanced in-situ carburization-oxidation cycle and resists deactivation.

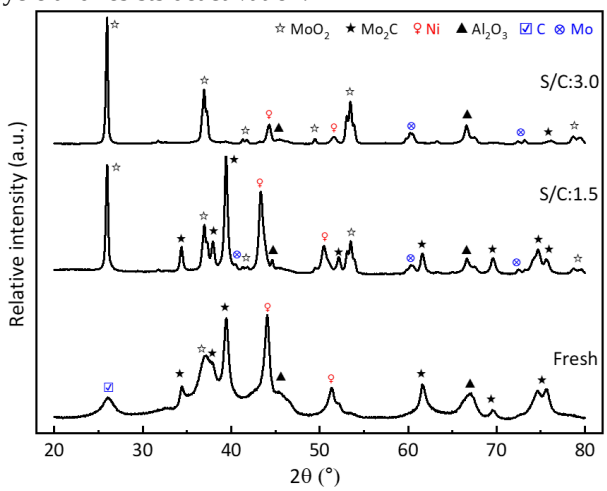


Fig. 2. XRD patterns of fresh and spent $Ni/Mo_2C/Al_2O_3$ catalyst employed in DJH-SMR.

Results indicate that DJH-SMR employing $Ni/Mo_2C/Al_2O_3$ offers a scalable, energy-efficient alternative to conventional SMR, with lower CO_2 emissions, water, and energy consumption. Its rapid response and compact design make it ideal for distributed H_2 production and integration with renewables, supporting industrial decarbonization and a clean H_2 economy.

3.2. MWH-CLDRM

In MWH-CLDRM, we examined and compared the heating behavior of Fe_3O_4 and SiC. Observed T_b and T_s when employing Fe_3O_4 exceeded those of SiC under equivalent MW power, validating the superior MW absorption behavior of Fe_3O_4 due to its simultaneous interaction with electromagnetic components. In addition, dielectric measurements and thermal imaging confirmed uniform heating of the bed inventory. We also carried out twenty continuous redox cycles on the lab-scale MWH-CLDRM reactor. In the initial four cycles, we detected a decrease in the extent of Fe_3O_4 reduction by CH_4, likely due to sintering and surface agglomeration. However, we recorded a consistent Fe_3O_4-CH_4 reactivity after stabilization (upon completion of the first four redox cycles). At T_b of 800°C, for the stabilized Fe_3O_4, we achieved CH_4 conversion of up to 97% with an H_2/CO ratio close to 2. Applying the same operating conditions and adapting ECH, we achieved CH_4 conversion of up to 40% with an H_2/CO ratio close to 2. The MWH-CLDRM demonstrated the highest CH_4 conversion of 97% at T_b of 800°C, surpassing ECH-CLDRM. At T_b of 800°C, we did not detect coke deposition (<0.01 wt.%) applying the MWH-CLDRM, whereas the ECH-CLDRM showed signs of carbon accumulation, likely due to CH_4 gas-phase cracking. Consistent with the reduction step, the MWH reactor outperformed the ECH reactor during Fe oxidation with CO_2. Consequently, direct heating of Fe_3O_4 particles via the irradiated MWs accelerated CH_4-Fe_3O_4 redox reactions, while minimizing side (gas-phase) reactions.

3.3. MWH-MTP

In MWH-MTP, performed in a MW-heated FBR (MWH-FBR) employing chemically inert SiC particles, our key objective was to determine the process efficacy under MWH and comparing its performance with that of an ECH-FBR under identical operating conditions. We aimed at understanding the influence of MW-specific thermal effects, including solid–gas temperature gradients and formation of localized hotspots, on CH_4 conversion, H_2 selectivity, and carbon capture.

Under the MWH, the SiC particles were consistently ~200°C hotter than the gas phase, a temperature profile different from ECH-FBR, where thermal equilibrium between the phases was present. We achieved a maximum CH_4 conversion of 23% and H_2 selectivity of 98% in MWH-FBR. At comparable conditions, we achieved ~150% higher CH_4 conversion in the MWH-FBR compared to ECH-FBR. H_2 selectivity positively correlated with conversion, exceeding 90% when CH_4 conversion surpassed 10%, with both heating methods showing similar selectivity at comparable conversion levels.

We assessed the solid carbon capture efficiency on the fluidized particles by comparing the rate of carbon deposition on the fluidized particles to the total solid carbon produced. In MWH-FBR, we achieved a solid carbon capture ranging from 61% to 89%, averaging 70%, which was higher than the maximum 55% carbon capture efficiency we achieved in ECH-FBR under identical conditions. This enhancement was primarily due to the non-isothermal conditions in the MWH-FBR that localized carbon deposition on hot particle surfaces, limiting carbon formation in cooler zones, such as the bubble phase, freeboard or reactor wall.

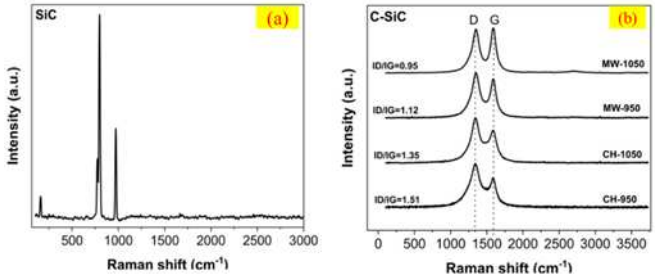


Fig. 3. Raman spectra of (a) SiC, (b) C-SiC produced in MWH-FBR and ECH-FBR at different temperatures [7]

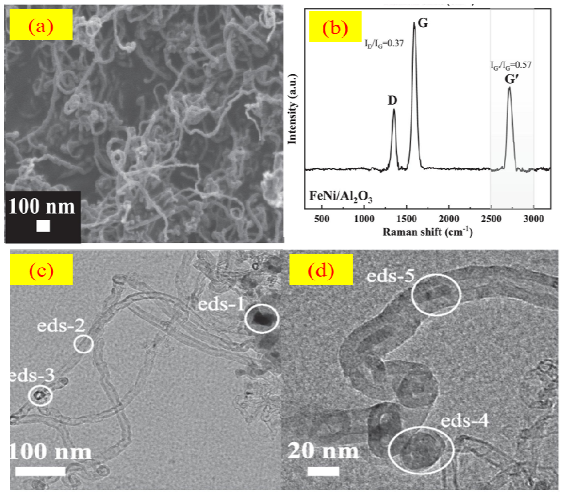
As shown in Figure 3, results of Raman spectroscopy indicated that MWH produced pyrolytic carbon with a higher degree of graphitization as compared to ECH. The band intensity ratio was lower in carbon from MWH-FBR at T_b=1050°C (0.96), compared to a higher ratio of 1.34 in ECH-FBR at the same T_b, suggesting larger in-

plane crystallite size and higher sp^2 content under MWH. One can attribute these results to the higher effective temperature at the particle surface and the presence of hotspots in MWH-FBR.

The transient behavior of the process over 18 hours of time-on-stream revealed a gradual increase in CH_4 conversion and T_s under MWH. This was driven by the accumulation of pyrolytic carbon, which enhanced the dielectric properties and improved the heating efficiency of the particles. As carbon loading increased from 0.45-4.2 wt.%, the T_s increased from 855°C to 980°C, leading to a stable CH_4 conversion plateau of around 10% at extended operation times. Unlike catalytic processes that typically suffer from catalyst deactivation and a decrease in CH_4 conversion, the adopted MWH-FBR for MWH-MTP showed self-enhancing behavior and a stable CH_4 conversion at extended time-on-stream.

3.4. MWH-LDPE pyrolysis

In this study, we evaluated Al_2O_3-supported Fe, Ni, and FeNi catalysts for carbon deposition and H_2 production in an MWH-assisted two-stage LDPE pyrolysis process. $FeNi/Al_2O_3$ indicated the best performance, achieving >97% LDPE conversion, 76 vol.% H_2, and 460 mg/g plastic CNT yield over 10 stable cycles. As shown in Figure. 4, Raman and SEM-EDX analyses confirmed high CNT purity and structural uniformity, with filamentous carbon making up ~70% of solids. Results presented in Figure 4(a) demonstrate that the CVD stage entangled CNT growth, confirming improved density and yield compared to monometallic counterparts.



(e)

Element(AT%)	C	O	Al	Fe	Ni
eds-1	88.7	5.5	2.9	2.4	0.5
eds-2	99.7	0.3	0.0	0.0	0.0
eds-3	99.5	0.5	0.0	0.0	0.0
eds-4	99.8	0.2	0.0	0.0	0.0
eds-5	96.3	2.3	0.0	1.1	0.3

Fig. 4. (a) SEM, (b) Raman, (c and d) TEM images and (e) EDX elemental spectra of the generated carbon nanotubes over $FeNi/Al_2O_3$ catalyst [11].

3.5. Comparison of electrified thermal H_2 production approaches

In Table 1, we presented a comparative overview of four electrified approaches developed and studied in this work for producing H_2 or syngas. For syngas generation, DJH-SMR and MWH-CLDRM offer distinct advantages. The DJH-SMR delivers a high H_2/CO ratio of 3 and with downstream water-gas shift reaction, making it particularly well-suited for H_2-rich streams. In contrast, the MWH-CLDRM operates without steam, eliminating the need for energy-intensive steam generation and lowering energy consumption through heat integration between the endothermic reduction and exothermic oxidation stages. This simplifies process design, improves energy efficiency, and lowers operating costs. A further advantage of the MWH-CLDRM lies in application of CO_2 as an oxidant in this approach to regenerate the reduced Fe_3O_4 oxygen carrier. This not only utilizes CO_2, thus decrease greenhouse gas emissions, but also converts it into CO, a versatile chemical building block for value-added products, such as phosgene, acrylates, benzaldehyde, citric acid, and toluene. In contrast to reforming, the MWH-MTP offers a carbon-neutral approach for H_2 production by avoiding CO_2 emissions altogether. This approach produces high purity H_2 gas alongside solid carbon, enabling direct in-situ solid carbon capture. The MWH-LDPE pyrolysis demonstrates the potential for chemical recycling of plastic waste, producing high purity H_2 gas and high-quality CNTs. Compared to CH_4 pyrolysis, the plastic pyrolysis can also generate liquid hydrocarbons (C_6–C_{23}) and waxes. In our MWH-LDPE system, we minimized these byproducts to <10 wt.%, ensuring efficient conversion of LDPE to H_2 and CNTs.

H_2 production with H_2/CO ratio of 3 by DJH-SMR, make it ideal for applications that require pure H_2 streams, such as ammonia synthesis, fuel cell feedstock, and hydrotreating in petroleum refining. Its compact design and electrically heated catalyst bed also allow for modular deployment in remote or distributed energy systems powered by renewable electricity. MWH-CLDRM has a great potential for Fischer-Tropsch synthesis and syngas-to-methanol conversion due to its tunable H_2/CO ratio of 2. MWH-MTP provides a carbon-free pathway for H_2 generation, while capturing solid carbon in the process. This feature makes it a key candidate for industries, such as green steelmaking, semiconductor manufacturing, and H_2 turbines, where CO and CO_2 impurities can be problematic. MWH-LDPE pyrolysis is advantageous for synthesizing CNTs, which are applied in batteries, conductive composites, and electrochemical electrodes. In addition, the H_2 produced from the MWH-LDPE pyrolysis can be utilized in fuel cells for clean power generation, as a low-emission fuel for transportation, or as a reducing gas in various chemical processes.

Table1. Comparison of electrified thermal H_2 production approaches explored in this study.

Approach	Bed material type	T_b (°C)	CH_4 or LDPE conversion (%)	Product	Carbon amount
DJH-SMR	$Ni/Mo_2C/Al_2O_3$	700–800	96	Syngas (H_2/CO of 3)	NA
MWH-CLDRM	Fe_3O_4	800	97	Syngas (H_2/CO of 2)	Minimal coke (<0.01 wt.%)
MWH-MTP	SiC	900–1065	23	High purity H_2 (98 vol.%)	CNT and black carbon (128 mg/g SiC)
MWH-LDPE pyrolysis	$FeNi/Al_2O_3$	500 (pyrolysis) - 800 (CVD)	97 wt.%	High purity H_2 (76 vol.%)	CNT (460 mg/g plastic)

4. Conclusion

In this study, we highlighted DJH-SMR, MWH-CLDRM, MWH-MTP, and MWH-LDPE pyrolysis as promising electrified H_2 production approaches. By combining selective volumetric heating with advanced materials, these approaches improve process efficiency, decrease greenhouse gas emissions, and enable carbon capture and waste valorization. We achieved over 96% CH_4 conversion in the DJH-SMR employing fluidizable and electrically conductive catalysts. We employed Fe_3O_4 as an oxygen carrier in the MWH-CLDRM and leveraged MW-induced thermal gradients to enhance CH_4 conversion (97%) and syngas quality, while minimizing coke formation. MWH-MTP showed high H_2 selectivity (98%) and effective solid carbon capture. The MWH-LDPE pyrolysis converted LDPE plastic waste into H_2-rich gas and CNTs, offering a sustainable approach for plastic waste upcycling and H_2 generation.

Despite the benefits mentioned, challenges remain regarding material stability, scalability, and economic feasibility. Future work should focus on enhancing catalyst and MW absorber durability, optimizing reactor design, and performing detailed techno-economic and life cycle analyses for these approaches. By addressing these challenges and leveraging the synergistic benefits of these electrified approaches, they hold a great potential for decreasing the CO_2 footprint of H_2 production, mitigating environmental impacts, and advancing both a sustainable H_2 economy and a circular economy for plastic waste.

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