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作品名稱　Greenhouse Gases Reduction: Conversion of Methane and Carbon Dioxide into Clean Energy

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Project Detail Form

Greenhouse Gases Reduction: Conversion of Methane and Carbon Dioxide into Clean Energy

SECTION 1: INTRODUCTION

In the upcoming years, both population and energy consumption are expected to increase dramatically [1]. Industrialization has led to a dramatic shift in the energy environment [2], with predictions of a 57% increase in demand for energy between 2002 and 2025 [3]. In addition to organic materials like trees and solid waste, fossil fuels like coal, natural gas, and oil provide more than 90% of the world's energy needs. Their overuse has resulted in the release of climate-altering greenhouse gases like carbon dioxide (CO_2) and methane (CH_4) into the atmosphere [4]. Scientists and other stakeholders are putting more emphasis on finding solutions to global warming, increasing energy production in order to meet increasing demands, and decreasing emissions of greenhouse gases. Using greenhouse gasses to make useful chemicals or fuels is one solution to both problems [5]. This motivated researchers to investigate the potential of CO_2 and CH_4 as clean energy sources. The process of dry reforming of methane (DRM) has been identified as a potentially successful strategy for transforming CO_2 into marketable syngas with a balanced H_2/CO composition [6], [7], [8], [9]. The economic viability of DRM, the reactor type, the availability of raw materials, and the intended use of the produced syngas are all-important considerations. Though DRM is gaining popularity, maintaining its long-term stability is difficult due to carbon accumulation from CO disproportionation and methane degradation [10], [11]. The catalyst used, as well as other parameters like as pressure, temperature, feed concentration, and reactor size, are critical to the process's effectiveness. In this scenario, a nickel catalyst on a La_2O_3/SiO_2 substrate with microspheres and a core-shell structure will be developed to improve

the conversion of greenhouse gases into profitable syngas. This catalyst is projected to improve the efficiency and performance of the DRM process significantly.

SECTION 2: Literature Review

The conversion of CH_4 and CO_2 into marketable syngas consisting of hydrogen and carbon monoxide makes DRM an extremely important catalytic process. For the DRM reaction, Ni-based catalysts have proven to be the most economical, highly active, and easily accessible option during the past three decades [12], [13]. The rapid deactivation of Ni-based catalysts, caused by carbon accumulation and/or sintering, is the fundamental obstacle to commercializing this approach [14], [15]. To improve metal dispersion and adjust the size of Ni particles, which in turn affects catalytic performance, mesoporous supports are used to confine Ni nanoparticles within porous channels. The methods of synthesis and the presence of promoters are also important factors in establishing the efficiency of the catalyst [16], [17], [18]. Because of their large surface areas, thermal resilience, and easy accessibility, fibrous silica (FS) supported catalysts have attracted more interest for their expanded use as catalysts [19]. Catalyst deactivation due to coke accumulation and active metal sintering can be prevented by adding suitable metals as promoters. In DRM, lanthanum is frequently employed as a promoter or support for Ni-based catalysts. This combination improves the catalyst's resilience to high temperatures and reduces the amount of coke that develops on it. Because of its exceptional chemical features such a high number of oxygen vacancies, reducibility, oxygen storage capacity, basic properties, and efficiency in removing coke, lanthanide-based oxides are also being actively explored by researchers. Because of their extraordinary durability in the circumstances of the DRM process, Lanthana-supported Ni

catalysts are currently the focus of intensive investigation. Incorporating rare earth oxides, particularly lanthanide oxides, is thought to be a promising strategy for improving the carbon resistance of nickel-based catalysts in the DRM process. Li et al. [20] developed nickel-based catalysts containing lanthanide oxides and used them for facilitating the DRM process. The size of the nickel nanoparticles generated throughout the process was regulated by the presence of lanthanide oxide.

The formation of Ni-$La_2O_2CO_3$ interfaces on Ni particles improves the stability of the catalyst by facilitating CO_2 adsorption and activation. The oxycarbonate can react with the coke in these interfaces [21]. Researchers were motivated to investigate alternative catalyst precursors due to low La_2O_3 surface area, which prevents an extensive nickel dispersion and consequently poor catalyst performance [22], [23]. The requirement for catalysts with a high surface area and inert support led scientists to investigate various silica-based compounds, such as SBA-15 and KIT-6. As a result, they used the PEG-assisted sol-gel method to produce a mesoporous Ni-La_2O_3/SiO_2 catalyst [24]. The Ni-La_2O_3 catalyst was synthesized on a fibrous silica substrate, specifically KCC-1, which comprises of high-surface-area silica nanospheres [25]. The basic sites provided by lanthana, La_2O_3, increase the reducibility of Ni^{2+} ions, leading to better nickel dispersion and higher selectivity in product formation. La_2O_3 and KCC-1 have been shown to increase reducibility, but the exact mechanism by which they do so is not yet known. Although using silica-based supports with a high surface area can improve the composite material's performance as a whole, there are still unresolved issues about how smaller Ni and lanthana nanoparticles (NPs) are distributed within the channels of the support structure, how these species are distributed on both the inner and outer surfaces of the support, and how they are functionalized. In order to clarify their functions, it is crucial to learn more about the interactions between Ni,

lanthana, and silica at the interfaces. The efficiency of these catalysts must be evaluated in terms of their mass of active component [26]. For this reason, we decided to synthesize a Ni-based catalyst supported by lanthana and fibrous silica (Ni/FSL) to evaluate its performance in the DRM process. This was done due to the fact that it has the potential to greatly improve efficiency, durability, conversion rates and selectivity of the desired products.

SECTION 3: Objectives

Research questions:

- What is the main aim of this research project?
- What is the novelty in this approach?
- What is the expected outcome of the research?
- What are the key findings and methodologies used in this study?
- How can the DRM process be optimized for large-scale industrial applications?

Purpose:

Our primary goal is to minimize greenhouse gas levels in the environment by converting them into a valuable product that can then be used to manufacture a range of chemicals to promote sustainability and meet energy demands. To achieve this, we attempted to create an affordable, highly effective, and anti-coking catalyst, such as Ni-FSL, to generate syngas via dry reforming of methane. Furthermore, we looked into the effect of adding La_2O_3 to fibrous silica. As in the previous study [27], this porous structure will have a good influence on the dry reforming of

methane by maintaining the active particles on the surface and enabling reagent access to the catalytic active sites. The additional oxides will have different basicity and ways of participating in the DRM process.

Novelty:

While Ni-based catalysts have showed considerable promise in the DRM conversion of CO_2 and CH_4 into syngas, there are a few limitations. We want to overcome these constraints by examining the effects of incorporating La_2O_3 into SiO_2 support. The uniqueness of this research is the mixing of fibrous silica with basic oxides in order to achieve high activity DRM catalysts, despite the fact that greater temperatures are necessary for excellent conversion in the case of Ni catalysts compared to other noble metals.

Hypothesis:

Our concept is that the presence of La_2O_3 and fibrous SiO_2 supports together will yield superior outcomes than previous studies because of the synergistic effects developed through their interaction.

SECTION 4: METHODOLOGY

Material and apparatus:

- Nickel precursor solution (e.g., nickel nitrate or nickel chloride).
- FSL support material (e.g., silica, alumina, or any other suitable support).
- Solvent (typically deionized water or a suitable organic solvent).
- Impregnation vessel (e.g., beaker or flask).
- Magnetic stirrer or mechanical stirrer.
- Heating source (e.g., hot plate or oven).
- Vacuum filtration setup (if necessary).
- Drying oven

Procedure:

The catalyst (Ni-FSL) was produced using the wet impregnation method, as described in the previous study [28]. In order to accomplish this, we dissolve the nickel precursor (nickel nitrate) in a suitable solvent to produce a homogenous solution with a Ni loading of 10% by weight. Fill the vessel with the weighed FSL support material. While stirring, gradually add the prepared nickel precursor solution to the support material. Wet the support material uniformly with the solution. Continued shaking for an adequate time facilitates the adsorption of Ni species onto the support. The surplus solvent will be evaporated using low heat (on a hot plate) to remove it. The impregnated support is dried in an oven overnight at 100 °C, calcined at 450 °C in Ar flow for 3 hours, and reduced under H_2 flow at 600 °C for 4 hours once the impregnation process is

completed. The catalyst must be cooled to room temperature in an inert environment after calcination.

Characterization techniques:

Several characterization approaches will be employed to investigate the suggested catalyst's physicochemical properties. Examples include N_2 physisorption (surface area and pore size distribution), XRD (crystallinity), FTIR (functional group), and advanced analytical instruments like XPS (oxidation state of metal atoms and oxygen vacancies), NMR (chemical environment of metal atoms), FESEM and TEM (morphology), CO_2–TPD (basicity).

Evaluation method:

The following formulae will be utilized to investigate the catalytic performance of the Ni/FSL catalyst.

$$\mathrm{CH_4\ conversion\ (\%)} = \frac{F_{CH_{4in}} - F_{CH_{4out}}}{F_{CH_{4in}}} \times 100 \quad (1)$$

$$\mathrm{CO_2\ conversion\ (\%)} = \frac{F_{CO_{2in}} - F_{CO_{2out}}}{F_{CO_{2in}}} \times 100 \quad (2)$$

$$\mathrm{Syngas\ ratio}\ \frac{H_2}{CO} = \frac{\text{mole of } H_2 \text{ produced}}{\text{mole of CO produced}} \quad (3)$$

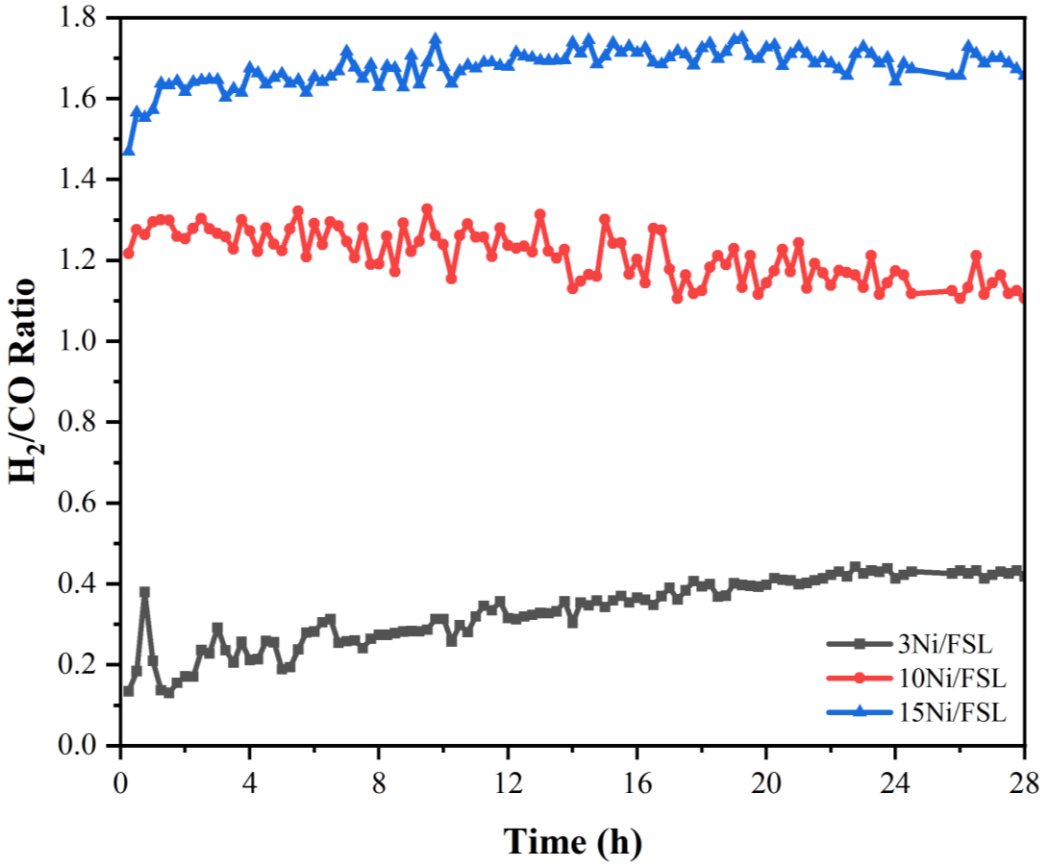
$$\mathrm{H_2\ yield} = \frac{F_{H_{2out}}}{2 \times F_{CH_{4in}}} \times 100 \quad (4)$$

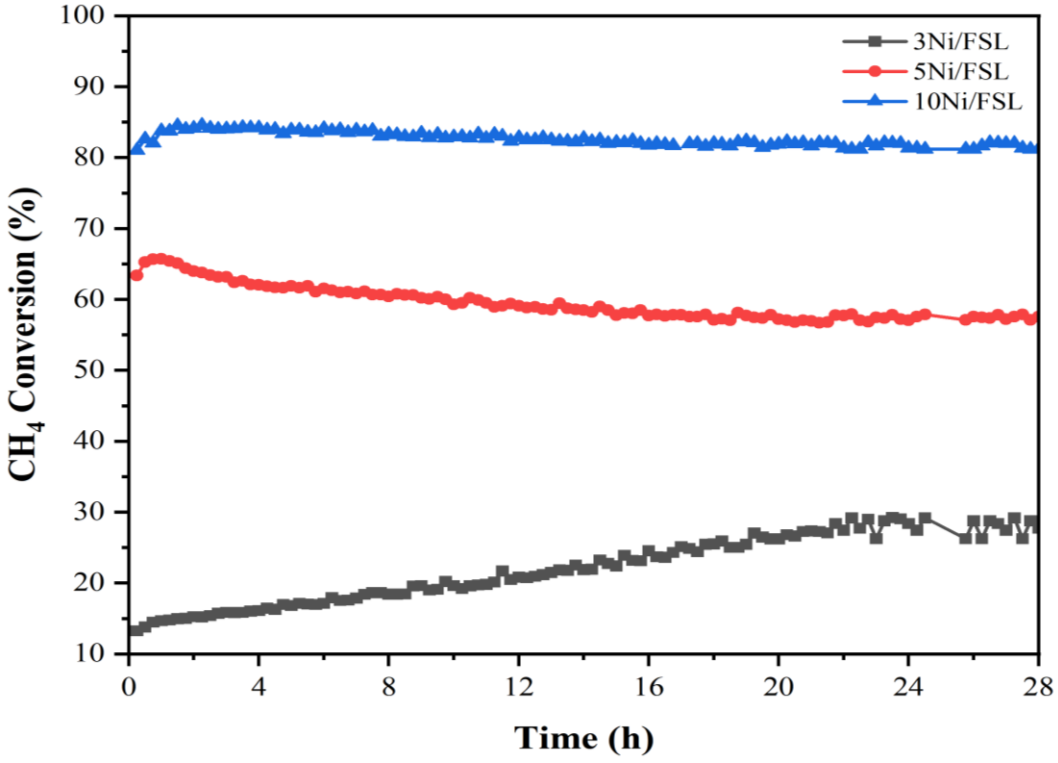
$$\mathrm{CO\ yield} = \frac{F_{CO_{out}}}{F_{CH_{4in}} + F_{CO_{2in}}} \times 100 \quad (5)$$

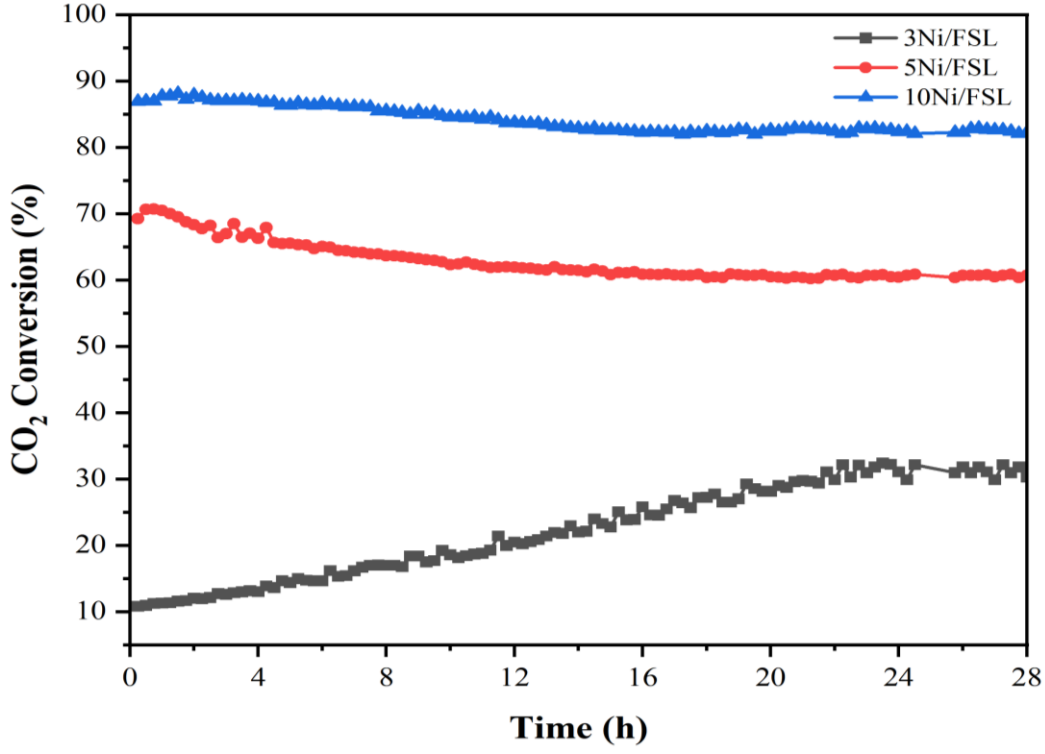
Where, F_{CO2in} = inlet molar flow of CO_2; F_{CO2out} = outlet molar flow of CO_2; F_{CH4in} = inlet molar flow of CH_4; F_{CH4out} = outlet molar flow of CH_4.

SECTION 5: RESULTS

- The prepared Ni-FSL catalysts were tested for their catalytic activity, and it was found that a Ni/FSL catalyst with a 3% Ni content had a conversion rate of 11% for CO_2 and 9% for CH_4.
- However, the conversion rates for CO_2 and CH_4 at 5% Ni/FSL are 69% and 64%, respectively.
- The 10% Ni/FSL catalyst showed the highest catalytic activity, converting 87% of CO_2 and 81% of CH_4.
- Moreover, a higher proportion of Ni in the catalyst results in greater H_2 and CO yields.


1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
H2/CO Ratio
0
4
8
12
16
20
24
28
Time (h)
3Ni/FSL
10Ni/FSL
15Ni/FSL

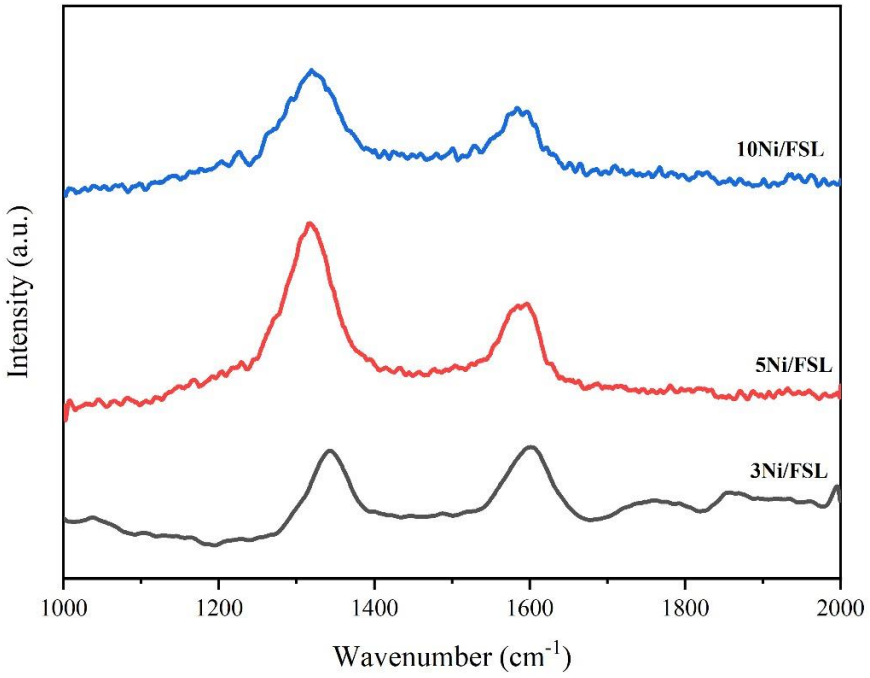

3Ni/FSL
5Ni/FSL
10Ni/FSL
CH_4 Conversion (%)
Time (h)
100
90
80
70
60
50
40
30
20
10
0
4
8
12
16
20
24
28

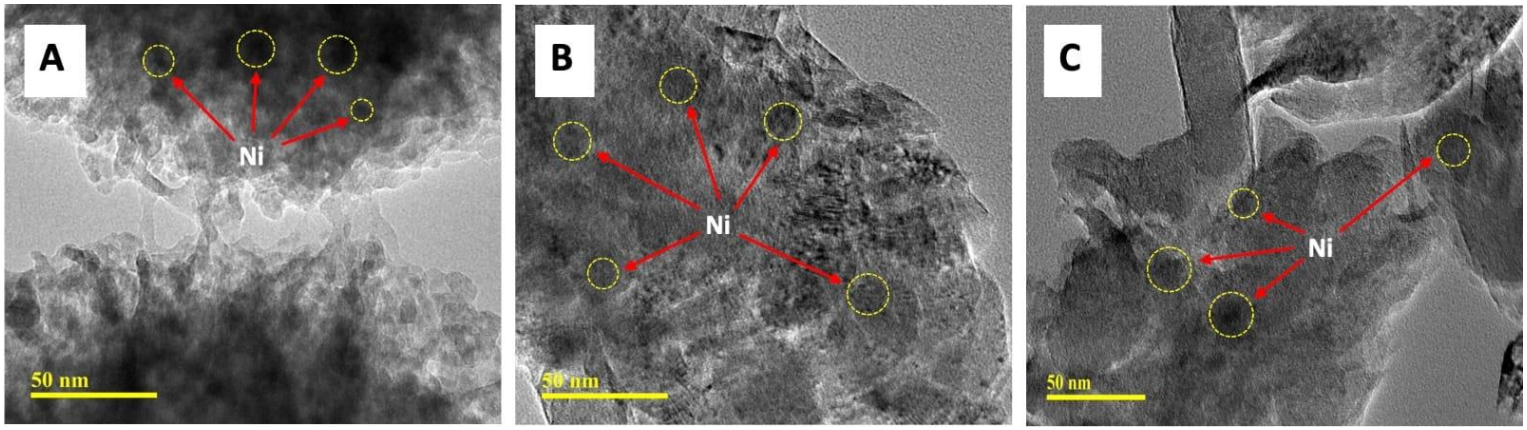

3Ni/FSL
5Ni/FSL
10Ni/FSL
CO_2 Conversion (%)
Time (h)
100
90
80
70
60
50
40
30
20
10
0
4
8
12
16
20
24
28

Spent catalyst characterization:

After 28 hours at 750oC, a thorough study of the materials was carried out using morphological, spectral, and physical analyses in order to learn more about the differences between various catalysts. Investigations into DRM have unequivocally demonstrated how coke production impairs the efficiency of Ni-based catalysts. Amorphous, graphitic, and filamentous carbons were among the species of carbon that were found during the reaction. We present TEM images of the catalyst samples after a 28-h operation time at 750°C. These images reveal that there is evidence of larger Ni particles, formed due to Ni agglomeration as a result of high-temperature sintering. Several of the Ni NPs were dislodged from the supporting material (FSL) due to the growth of filamentous carbon. Notably, a substantial accumulation of carbon was observed on all utilized catalysts of the filamentous type, except for 3Ni/FSL, which revealed the presence of amorphous carbon after the reaction. Interestingly, some Ni nanoparticles can be observed positioned at the ends of the filaments, serving as evidence of the positive impact of filamentous carbon on Ni dispersion. Although the average size of Ni particles has slightly increased compared to the fresh catalysts, this underscores the catalysts' resilience to sintering. Prior research has suggested that the formation of carbon nanotubes typically does not lead to catalyst deactivation. This can be credited to the scattering of Ni NPs at the extremities of the carbon filaments, which additionally functions to stabilize the catalyst for an extended duration. Conversely, the formation of amorphous carbon in the case of 3Ni/FSL fully envelops the Ni NPs, leading to unstable catalytic activity over the course of the reaction. The Raman spectra analysis further validated the existence of various carbon species. The presence of sp2-bonded carbon deposition was evident from the distinct peaks

observed in the spectra. Specifically, at 1585 cm-1 the graphite carbon band (G-band) and at 1348 cm-1 the disorder-generated band (D-band) represented two separate peaks associated with sp2-bonded carbon deposition [29]. The intensity of the bands in the spectra is indicative of the amount of carbon present, while the D-to-G band ratio (ID/IG) is utilized to assess the level of graphitization in carbon formations. Consequently, a higher ID/IG ratio suggests a greater tendency for the rapid formation of graphitic carbon compared to amorphous carbon. This heightened graphitic carbon formation rate makes the catalyst more susceptible to deactivation and poses challenges for the rejuvenation process [29]. The I_D/I_G ratios of the 3Ni/FSL, 5Ni/FSL, 10Ni/FSL, 15Ni/FSL, and 20Ni/FSL samples were determined to be 0.89, 1.79, 1.6, 1.52, and 2.75, respectively. These ratios indicate the presence of both graphitic carbon and amorphous carbon in all the samples. This suggests that there is a minimal amount of carbon deposition on the 3Ni/FSL catalyst, with the majority of the carbon adopting a disordered amorphous structure. This type of carbon deposition carries a lower possibility of catalyst deactivation. The higher I_D/I_G ratio in the highest line suggests that the carbon structures that developed this spectrum had more essential flaws.


10Ni/FSL
5Ni/FSL
3Ni/FSL
Intensity (a.u.)
1000
1200
1400
1600
1800
2000
Wavenumber (cm-1)


A
Ni
50 nm
B
Ni
50 nm
C
Ni
50 nm

SECTION 6: INTERPRETATION & CONCLUSIONS

Conclusion:

- The 10% Ni/FSL nanocomposite had the highest CH_4 and CO_2 conversion rate and showed the most promising catalytic stability.
- In addition, 15% Ni/FSL produced greater amounts of H_2 and CO than 3% Ni/FSL and 10% Ni/FSL.
- The preceding data show that increasing the content of Ni in a catalyst increases its ability to transform reactants into products, resulting in better products yield.

Value add, impact and applications:

This innovative method has the dual advantage of lowering GHG concentrations in the environment while also promoting the production of syngas (CO/H_2), a valuable chemical molecule. By turning CO_2 and CH_4 into valuable substances such as syngas (CO/H_2), we not only address change but also stimulate economic growth and innovation, which coincides with our commitment to a more promising and prosperous future.

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【評語】030035

In the present work, a nickel catalyst on a La203/Si02 substrate with microspheres and a core-shell structure has been developed to improve the conversion of greenhouse gases into profitable syngas. The process of dry reforming of methane (DRM) has been identified as a potentially successful strategy for transforming C02 into marketable syngas with a balanced H2/C0 composition. A nickel catalyst on a La203/Si02 substrate with microspheres and a core-shell structure is developed to improve the efficiency and performance of the DRM process. It was assumed that the presence of La203 and fibrous Si02 supports together would yield superior outcomes than previous studies because of the synergistic effects developed through their interaction. To achieve this, they synthesized Ni-based catalyst supported by lanthana and fibrous silica (Ni/FSL) to generate syngas via dry reforming of methane. The data showed that 10% Ni/FSL nanocomposite had the highest CH4 and C02 conversion rate and 15% Ni/FSL produced greater amounts of H2 and C0 than 3% Ni/FSL and 10% Ni/FSL. Increasing the content of Ni in a catalyst increases its ability to transform reactants into products, resulting in better product yield. The authors have done a good literature search and addressed the goal, hypothesis, and novelty in their report. The presentation is concise and easy to understand.