GAS FLARING REDUCTION TO SYNGAS USING LAYERED DOUBLE HYDROXIDE BASED COMPOSITE THROUGH PHOTOCATALYTIC DRY REFORMING OF METHANE

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ABSTRACT

Gas flaring reduction by utilizing methane for syngas production through dry gas reforming of methane is a favorable method, as compared to other syngas producing methods, as it utilizes both greenhouse gases (CO_2 and CH_4). Though, the dry reforming process is well studied, there are areas that are still being explored in optimizing the process. Currently, the focused area of research is improving the stability and activity of the catalysts used in the dry reforming of methane process. Activity of catalyst mainly depends upon support type, particle size, and dispersion on support, and synthesis method. Whereas catalyst deactivation is primarily due to coke deposition and sintering of metal precursor. In this work efficient well designed 2D/2D CoAl-LDH/g-C_3N_4 heterojunction for photocatalytic dry reforming of methane (DRM) for syngas production has been designed and fabricated. CoAl-LDH with different concentration coupled with g-C_3N_4 first tested for optimization of photocatalytic syngas production (CO, H_2), as prepared 15 wt.% CoAl-LDH/g-C_3N_4 exhibited efficient syngas production with proficient selectivity for CO and H_2. Productivity of H_2 of 15% wt. CoAl-LDH/g-C_3N_4 is about 4.8 fold that of pure CoAl-LDH and for CO is about 3.8 fold than that of pure CoAl-LDH. The improved photocatalytic activity could be attributed to unique structure and abundant active sties on surface. As compared to other heterojunction, 2D/2D CoAl-LDH/g-C_3N_4 heterojunction exhibit batter coupling interfaces and strong interfacial interaction, which can easily suppress the photo induced charge carrier's recombination and decreases the distance of transmission of charges. The good recyclability and efficient sorption process with different feed ratio (CH_4/CO_2) confirmed its stability and batter activity. Comparison with BRM process, gave opportunity to further extend the study for future improvement in shortcomings related to structure of heterojunction for better performance in BRM. Coupling CoAl-LDH with g-C_3N_4 in sheet-on-sheet heterostructure is an effective strategy towards syngas production through DRM process.

ABSTRAK

Reduksi gas flaring melalui kaedah pembentukan semula gas dengan mengutilasi gas metana bagi pengeluaran sintesis gas merupakan satu prospek yang terbaik berbanding kaedah lain. Teknik ini mengutilasi kedua-dua gas rumah hijau (CO2 dan CH4). Walaupun kajian melalui teknik pembentukan semula gas kering telah meluas, namun dalam mengoptimasikan proses ini, kajian perlu diperluaskan. Fokus kajian kini hanyalah terhadap mengimprovisasi stabiliti dan aktiviti pemangkin dengan menggunakan kaedah pembentukan semula gas kering melalui gas metana. Aktiviti pemangkin bergantung kepada jenis sokongan, saiz zarah, dispersi keatas sokongan dan juga kaedah sintesis. Faktor penting yang menyebabkan deaktivasi pemangkin adalah disebabkan oleh pemendapan kok dan pesinteran logam prekursor. Dalam kajian ini, 2D/2D CoAl-LDH/g-C_3N_4 heterojungsi direka dengan efisien untuk pembentukan semula gas kering fotokatalisis menggunakan gas metana (drm) untuk pengeluaran gas sintesis telah di fabrikasi. CoAl-LDH menggunakan konsentrasi berbeza di pasangkan dengan g-C_3N_4 diuji untuk optimisasi pengeluaran fotokatalisis gas sintesis ((CO,H_2), seperti yang disediakan 15 wt.% CoAl-LDH/g-C_3N_4 memiliki pengeluaran gas sintesis yang efisien dengan selektiviti yang profisien untuk CO and H_2. Produktiviti untuk H_2 of 15% wt CoAl-LDH/g-C_3N_4 adalah sebanyak 4.8 fold dan CO adalah sebanyak 3.8 fold daripada CoAl-LDH asli. Peningkatan aktiviti fotokatalisis disebabkan struktur unik dan tapak aktif yang banyak di atas permukaan. Berbanding dengan heterojungsi lain, 2D/2D CoAl-LDH/g-C_3N_4 heterojungsi memiliki bater interfasa gandingan dan interaksi interfasa yang kuat dimana memudahkan dalam menghalang rekombinasi foto-induksi karier cas dan mengurangkan jarak penularan cas. Kadar penggunaan semula yang baik dan penjerapan proses yang efisien dengan nisbah kemasukan yang berbeza (CH_4/CO_2) menentukan kestabilan dan aktiviti bater. Perbandingan proses BRM memberi peluang memperluaskan kajian struktur heterojungsi untuk prestasi yang lebih baik bagi penambahbaikan masa hadapan. Gandingan CoAl-LDH dan g-C_3N_4 diatas lapisan heterostruktur adalah merupakan satu strategi yang efektif terhadap pengeluaran sintesis gas melalui kaedah DRM.

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LIST OF ABBREVIATIONS

SMR	-	Steam Reforming of Methane
DRM	-	Dry reforming of Methane
EOR	-	Enhanced Oil Recovery
Syngas	-	Synthesis gas
Syncrude	-	Synthesis crude
POM	-	Partial Oxidation of Methane
ATR	-	Autothermal Reforming
LNG	-	Liquified Natural Gas
LPG	-	Liquified Petroleum Gas
GTL	-	Gas To Liquid Technology
FT	-	Fischer-Tropsch
RWGS		Reverse -Water- gas -shift
GHG		Greenhouse gas
GGFR		Global Gas Flaring Reduction
UNFCCC		United Nations Framework Convention on Climate Change
BCM		Billion Cubic Meters
EIA		Energy Information Administration
IEA		International Energy Agency
OPEC		Organization of Oil Exporting Countries
CAPEX		Capital Expenditure
OCM		Oxidative Coupling of Methane
NOCM		Non-Oxidative Coupling of Methane

LIST OF SYMBOLS

δ	-	Minimal error
D, d	-	Diameter
F	-	Force
v	-	Velocity
p	-	Pressure
I	-	Moment of Inersia
r	-	Radius
Re	-	Reynold Number
CH_4		Methane
H_2		Hydrogen
Kb/d		Thousands barrels per day
Mscf/d		Million Standard Cubic Feet of Gas Per Day
CO		Carbon monoxide
λ		Wavelength
g		gram
µm		Micrometer
cm		Centimetre
m		metres
nm		Nanometre
n		No of moles
V		volume
Eg		Band Gap
A		Area
CB		Conduction Band
VB		Valance Band

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Regardless of efforts to tackle global warming and climate change, the burning of fossil fuels is still found to be the main contributing factor for the increase of greenhouse gases in the atmosphere [1]. It is well known that oil and gas production sites and refineries are main sources of greenhouse gas emission due to releasing flare and flue gases. Flue gas is the mixture of gases produced during combustion of fossil fuels and acts as pollutant, whereas, flare gas emission occurs when the surplus process gas is burnt in gas flares before releasing to the atmosphere [2].

Natural Gas flaring is the process in which associated gas from wells, refineries and hydrocarbon processing plants are burned either for disposal purposes or as a way to release pressure [3]. This practice of burning gas is now recognised as an important environmental problem. About 150 billion cubic meter of natural gas is flared worldwide, which contaminates the surrounding environment with almost 400 Mt carbon dioxide per year [3, 4]. The estimated losses of flared gas are the single largest loss in many industrial operations such as oil and gas production, chemical plants, refineries and coal plants. Wastes or losses occurred due to the flaring includes natural gas, fuel gas, nitrogen and process gases [5]

Methane, a prime component of hydrocarbon family, and considered as a cheapest energy source, compared to other fossil fuels. Yet often it is neglected as a major GHG contributor , with more severe potency of almost 30 times and lifetime of 100-year as compared to Carbon dioxide.[6] Oil and Gas industry is a major contributor to CH4 emission via gas flaring, the process in which methane gas is burned-off from oil and gas fields as a mean of safety measure for pressure relieve.[7] This practice of gas flaring cause not only environmental problems, but also contribute to wastage of gas, which otherwise would have been utilized for energy generation.[8] According to one estimate around 150 billion cubic meter of natural gas is flared as a routine practice in oil and gas fields around the world, which directly contribute to environmental contamination with almost 400 Mt CO2 per year.[1] Wastage of valuable gas from oil and gas industry is the single biggest loss in terms of volume of flared gas.

The situation of flaring may reduce, due to application of Dry reforming of methane (DRM), which utilizes both CO2 and CH4 for production of the industrially valuable synthesis gas (syngas), which is mixture of CO and H2.[9] Given by equation (1.1).

$$CH_4 + CO \leftrightarrow 2CO + 2H_2, \quad \Delta H_{298k} = 247 kJ/mol \quad (1.1)$$

$$CO_2 + H_2 \leftrightarrow CO + H_2O, \; \Delta H_{298k} = 41\, kJ/mol \quad (1.2)$$

Regardless of advantages of DRM process, the production of syngas from equation (1) requires energy intensive operating conditions, which is highly endothermic process (temperature of 800 ◦C.[10].This heat requirement is supplied through combustion of fossil fuels, which further increase the GHG emission associated with syngas production. Moreover, catalyst deactivation during DRM process has remained a serious obstacle towards its industrial application.[11] Solar energy driven photocatalytic process, is a promising technique, which displaces conventional thermal reforming with solar reforming, thus reducing the reaction temperature thereby, avoiding CO2 emission by adopting the green approach to DRM, also provide better resistance to coke formation[6].

However, few studies reported on solar energy driven reaction of CH4 and CO2. In a recent study , it was investigated that plasmonic metal based catalysts can be used for the acceleration of DRM process.[12] Various studies reported for photocatalytic DRM using transition metal oxide semiconductor catalysts.[13-15] In one study, SrTiO3 catalyst exhibited 3.8% methane conversion under 700 ◦C reaction condition. In another study, Rh/SrTiO3 catalyst reported, which exhibited yield of almost 50% and DRM conversion at reaction condition of under 150 ◦C.[16] Moreover, various metal oxides such as, tin oxide (SnO_2), titanium dioxide (TiO_2), and tungsten oxide (WO_3) were studied as a semiconductor photocatalysts for photocatalytic DRM process.[17] Furthermore, in various studies, magnesium oxide (MgO) was used at low temperature for reduction of CO_2 to CO in the gas phase for Photocatalytic DRM reaction.[15] Recently, combination of Pt/TiO_2 with SiO_2 light diffuse reflection surface for efficient DRM photocatalytic reaction.[11] In another study La- modified TiO_2 (La/TiO_2) was used under UV light for photocatalytic DRM reaction.[10] In all these studies mentioned, the main issues encountered with respect to catalyst activity was, lower catalyst stability due to catalyst deactivation, higher heat requirement due to endothermicity of reaction and photothermal mode of heat addition, and expensive catalyst used.

Recently, layered double hydroxides, (LDHs) have received considerable attention due to its various applications in various fields, such as catalysts, use as absorbents for CO_2, catalyst precursors, as anion exchangers photoactive materials, degradation of pollutants and hydrogen production.[18] LDHs have normally higher specific surface area, which implies that more active sites for catalytic reaction.[19] Layered double hydroxides are type of hydrotalcite clay, comprise of positive layer and interlayer anion for exchange. Among various layered materials, LDHs are significant layerd photocatalyst material with structure comprise of brucite like layer with MO_6 ictahedra edge sharing structure.[20]Therefore, MO_6 octahedra edge sharing structure helps in formation of two dimensional sheets (2D) consists of metal cations coordinated with OH group in six folds. LDHs have certain advantages of being uniform distribution of metallic cations, higher stability, lower cost, and adjustable composition.[21] Recently, layered double hydroxides materials attracted researchers' attentions in photocatalysis.[22] As it has advantages of containing transition metal elements, tunable band gaps, environment friendly and higher photocatalytic activity under visible light. However, Pure LDHs exhibit lower catalytic activity due to high recombination rates of electron and hole and slow charge carriers mobility to the surface.[23] Among various layered double hydroxides materials CoAl-LDH found to be excellent photocatalytic agent due to its appropriate redox potential and higher visible light harvesting abilities.[24]

Therefore, its application particularly focused for various processes comprise of photoreduction reactions under visible-light irradiation., also for photocatalytic degradation reactions.[20]

Graphitic carbon nitrade (g-C3N4), a non-metallic semiconductor, due to its excellent abilities of exhibiting lower cost, suitable band gap position (2.7 eV), and higher stability is desirable for its application towards efficient energy production owing to presence of earth-abundant elements.[25] Therefore, g-C3N4 application towards DRM is promising for higher conversion without thermal system constrain. However, in spite of promising activity, g-C3N4 still faced with restriction of limited activity due to higher recombination rate.[10] Hence, requirement of further modification necessary to achieve the desired results of higher activity.

Various literature regarding restricting charge carriers through modification of its intrinsic structure have been reported.[10, 26] Two-dimensional (2D) morphology system for photocatalytic process have proved to be helpful in suppressing charge carrier recombination also reducing the distance of transmission for charge carriers.[27] Several studies confirmed the fabrication of g-C3N4 with material such as LDHs (NiAl-LDH), Znln2S4 and phosphorus (black) increased the charge carrier separation.[22, 28] Therefore, doubled layer hydroxides (LDHs) are preferred for its unique double layered structure and excellent photocatalytic performance can be utilized with g-C3N4 to lower the charge recombination and enhance the photocatalytic activity.[29] CoAl-LDH has been used previously in various studies for different applications, such as CO2 reduction, degradation of pollutants like RhB and Congo red, and recently for photocatalytic hydrogen production.[23] However, CoAl-LDH application for syngas production in dry reforming of methane, never reported before.

Over the last few years, performance of LDHs has been improved by compounding various materials.[30] In current study, CoAl-LDH combined with g-C3N4 for photocatalytic reforming of methane to syngas (CO,H2) by suppressing the charge carriers recombination and enhancing the syngas production. CoAl-LDH was synthesized through co-precipitation method and g-C3N4 was obtained through calcination, and later CoAl-LDH was coupled in different percentages with g-C3N4. The performance of synthesized layered 2D composite was tested through dry reforming of methane in fixed bed photoreactor under visible light irradiation. The 2D/2D CoAl-LDH/g-C3N4 composite exhibited higher photocatalytic activity for syngas production. In addition, the composite was tested with variation in feed ratio of CH4/CO2 to examine the effect on yield and selectivity of H2 and CO (syngas). Comparison between dry reforming and bi reforming was conducted to evaluate the catalyst performance for yield. Finally, catalyst stability was evaluated for catalyst life determination in terms of continuous production of CO and H2 under same operating condition in various cycles. The construction of CoAl-LDH composite with g-C3N4 will pave path for reducing GHG emissions and inhibit wastage of valuable energy resource through gas flaring processing and allow reforming of natural gas (methane) to efficient renewable fuel through DRM photocatalytic process.

1.2 Problem Statement and Research Hypothesis

The recent studies over carbon dioxide (CO_2) utilization technologies with respect to controlling greenhouse gas (GHG) emissions has often neglected methane (CH_4), which is another main GHG contributor with impacts 25 times than that of CO_2 also the lifetime span of a 100-year, leading to more severely environmental deterioration[6]. Oil and Gas industry is the main contributor towards methane emission and burning of natural gas, which is often called as gas flaring, a routine practice in oil and gas industry. Natural gas flaring practice wastes valuable energy resource and enhances global warming effects.

There are different natural gas utilization technologies, among them syngas production through reforming methods is a best option of natural gas utilization in Gas to liquid technology (GTL) process, because of GTL is being marked as a clean and environmentally friendly fuel source worldwide[7]. Syngas production is a first primary step in GTL process, as it is used further as a feed for conversion into synthetic crude, in a reaction based on Fischer-Tropsch (FT) process. Besides, Syngas can be produced through various reforming methods i.e. Steam reforming of methane (SRM), Partial oxidation of methane (POM).Dry reforming of methane (DRM).DRM can be considered more suitable for Fischer-Tropsch synthesis because of H_2/CO molar ratio of unity, whereas Steam reforming of Methane (SRM) process gives higher molar ratio of H_2/CO, which limits its usage in FT process[31].However, in all above reforming technologies, the energy requirement is very high, needs high temperatures, which result into further combustion of fossil fuel, thus contributing to CO_2 emission. Natural gas flaring reduction through technologies such as LPG, LNG and GTL are expensive because of more energy requirement and more greenhouse gas emissions.

Catalysts used in reforming process, have issues such as, catalyst deactivation and sintering, which causes lower productivity during reforming process. Deactivation of catalyst normally occurs due to endothermicity and high temperature of reforming reaction.

Based on the discussed problems and by considering the above-mentioned perspective, following hypothesis is formulated by keeping in view the possible solutions:

a) Gas flaring reduction can be mitigated, through its utilization towards production of energy efficient renewable fuel.

b) Gas flaring reduction using photocatalytic Dry reforming Technology is economical and environment friendly. Required low temperature and green energy solution to gas flaring problem.

c) CoAl(LDHs)/g-C_3N_4 composite photocatalyst can enhance the photocatalytic productivity of synthesis gas (CO,H2). Also have batter stability then thermally driven Dry reforming process.

1.3 Project Objectives

Syngas Production through photocatalytic Dry reforming of Methane (DRM) is a proficient strategy towards solution of Gas flaring reduction and further utilization to energy efficient renewable fuels. In this regard following are the objectives of the current study:

(a) Gas flaring reduction through utilization in reformation to Synthesis gas (H_2,CO) for energy efficient fuel.

(b) Synthesis of CoAl-LDH/g-C_3N_4 composite photocatalyst for higher productivity of syngas. Characterization of synthesized catalyst sample to study the morphology, structure and elemental characteristics and its influence on higher yield of syngas.

(c) Parameters Study on synthesized photocatalyst such as Feed ratio and Stability.

1.4 Scope of Study

The scope of the study is as under, aims at gas flaring reduction and utilization to efficient energy, improving the productivity of photocatalyst, towards improving the performance of overall photocatalytic dry reforming of methane.

i. Synthesis of CoAl-LDH/g-C_3N_4 composite photocatalyst using Co-precipitation method.

ii. Characterization of photocatalyst so to study the structural, morphological and surface characteristics using XRD,SEM,EDX, FTIR and PL.

iii. Natural gas (CH_4) along with Carbon dioxide (CO_2) is Tested (flared gas) as a feed in photoreactor for Dry reforming process to Produce synthesis gas.

iv. To test the stability of synthesized photocatalyst composite and evaluate the effect of parameters such as feed ratio on productivity of photocatalyst.

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LIST OF PUBLICATIONS

Title	Journal	Impact Factor
Recent Development in Natural Gas Flaring Reduction and Reformation to Energy Efficient Fuels: A review	Energy & Fuels Journal by American Chemical Society	(Q1,IF=3.1)