PHOTOCATALYTIC CONVERSION OF CARBON DIOXIDE TO METHANE USING RGO/Au-TNTs

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SUPERVISOR’S DECLARATION

I hereby declare that I have checked this thesis, and in my opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Master of Science.

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I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

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ABSTRAK

Kepelbagaian bahan nanostruktur dan penerokaan sifat-sifat fizikal, kimia dan optik bahan tersebut adalah sangat diteliti pada masa kini. Kajian ini memberi penekanan kepada pengurangan gas karbon dioksida (CO_2) yang besar di atmosfera kepada bahan api hidrokarbon yang berharga dengan penggunaan bahan novel semikonduktor fotokatalis. Titanium dioksida (TiO_2) adalah salah satu daripada semikonduktor fotokatalis yang paling meluas untuk diaplikasi. Walaubagaimanapun, ia mempunyai kelemahan utama dari segi jurang tenaga tinggi iaitu 3.2 eV dan kadar rekombinasi pembawa caj yang tinggi. Oleh kerana nilai jurang tenaga yang tinggi, penyerapan tenaga hanya berlaku di rantau ultraviolet (UV) spektrum elektromagnetik. Tambahan itu, spektrum suria hanya mengandungi 5% rantau UV sekaligus menyukarkan penyerapan tenaga oleh bahan fotokatalis di bawah spektrum suria. Oleh itu bahan TiO_2 telah diubahsuai bagi mengurangkan jurang tenaga dan kadar rekombinasi pembawa caj sekaligus membolehkan penyerapan cahaya melalui specktrum suria. Pengubahsuaian telah dilakukan melalui gabungan anodalis elektrokimia dan pemendapan elektrokimia. Selepas pengubahsuian, kecekapan penyerapan cahaya dapat dilihat melalui analisis UV-Vis melalui sifat LSPR nanopartikel Au. Di samping itu, jurang tenaga fotokatalis telah berkurang secara drastik yang seterusnya menunjukkan kadar rekombinasi e^-/h^+ yang lebih rendah dicapai melalui analisis PL. Prestasi bahan fotokatalis telah dikaji melalui penukaran hasil CO_2 ke CH_4. Penukaran hasil CO_2 ke CH_4 adalah mengikut turutan TNTs <RGO-TNTs <Au-TNTs <RGO/Au-TNTs dengan bacaan 4.1% <12.46% <22.32% <33.1%. Hasil yang signifikan diperoleh dengan menggunakan RGO/ Au-TNTs. Jumlah hasil CH_4 yang diperoleh selepas 2 jam melalui RGO/ Au-TNTs adalah 8.07 kali lebih tinggi daripada TNTS. Kesimpulannya, TiO_2 yang digabungkan dengan Au telah berjaya disintesis melalui kaedah pemendapan elektrokimia yang lebih mudah. Selain itu, kecekapan penyerapan cahaya yang kelihatan lebih lama meningkatkan kadar penggabungan semula e^-/h^+ dan meningkatkan kecekapan penukaran hasil CO_2 ke CH_4. Oleh itu, pendekatan ini membuka banyak laluan untuk bahan semikonduktor fotokatalis digunakan di bawah spektrum suria yang besar bagi menghasilkan bahan api hidrokarbon dari CO_2 yang berlebihan di atmosfera.

ABSTRACT

The diversity of nanostructured material synthesis and exploring the proficient physical, chemical, and optical properties in order to investigate its catalytic efficiency is one of the most researched areas nowadays. This present study emphasizes on the reduction of immense CO_2 gas in the atmosphere to valuable hydrocarbon fuel with the utilization of synthesized novel nanostructured photocatalyst. Titanium dioxide (TiO_2) is one of the most widespread semiconductor photocatalysts for photocatalytic applications. Despite its eminence, it has major drawbacks in terms of higher bandgap (3.2 eV) and high recombination of photogenerated charge carriers. Due to its wide bandgap, the photoexcitation occurred only in the ultraviolet (UV) region of the electromagnetic spectrum. Moreover, the UV region is only 5% in the solar spectrum whereas the visible region comprises a total of 53%. Thus, the higher charge carrier recombination, with less visible light utilization during photoexcitation of TiO_2 is one of the major challenges in photocatalytic domains. For this reason, in this study, a TiO_2 based nanocomposite photocatalyst with enhanced visible light efficiency was developed through the combined electrochemical anodization, electrochemical deposition, and immersed method. The visible light absorption efficiency of the photocatalysts was revealed through UV-Vis analysis due to the LSPR nature of Au nanoparticles. In addition, the bandgap energy of the photocatalyst was reduced drastically which further shows a lower e^-/h^+ recombination rate attained through PL analysis. The photocatalytic performance of the prepared photocatalysts for the conversion of CO_2 to CH_4 yield follows an ascending order of TNTs <RGO-TNTs < Au-TNTs <RGO/Au-TNTs which are 4.1% <12.46% <22.32% <33.1%. The significant result obtained by utilizing RGO/Au-TNTs photocatalyst, for the reduction of CO_2 to CH_4. The total CH_4 yield obtained after 2 h of photocatalytic performance for the RGO/Au-TNTs is 8.07 times higher than TNTs. To conclude, Titanium dioxide nanotube incorporated with Au was successfully synthesized through a facile electrochemical deposition method as well induced simple experimental set-up. The prolonged visible light absorption efficiency improved the TNTs e^-/h^+ recombination rate and enhanced the photocatalytic CO_2 conversion efficiency towards visible light by employing LSPR effective Au nanoparticles and highly active RGO. Therefore, this approach opens the numerous paths for the efficient visible light photocatalyst (VLP) for utilizing a huge solar spectrum to produce hydrocarbon fuels from the excessive CO_2 in the atmosphere.

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LIST OF SYMBOLS

°C	degree celsius
g	gram
cm^{-1}	per centimetre
h	hour
mL	mililitre
mol	mole
mol L-1	mol per litre
nm	nanometre
%	percentage
s	second
cm^2	square of centimetre
µm	micrometre
W/cm^2	watt per centimetre square
kPa	kilopascal

LIST OF ABBREVIATIONS

Ag	Silver
Au	Gold
CO_2	Carbon dioxide
CB	Conduction band
CH_4	Methane
e^-	Charge/electron
E_f	Fermi energy
E_g	Bandgap energy
GO	Graphene oxide
GHG	Greenhouse gases
Hg	Mercury
H^+	Proton
h^+	Carrier/hole
hυ	Photon energy
IPCC	Intergovernmental panel on climate change
LSPR	Localized surface plasmon resonance
-OH	Hydroxyl
•OH	Hydroxyl radicals
Pt	Platinum
RGO	Reduced graphene oxide
SPR	Surface plasmon resonance
TiO_2	Titanium dioxide
TNTs	Titanium dioxide nanotube
UV	Ultraviolet
VLP	Visible light active photocatalyst
VB	Valence band
Xe	Xenon

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