

TEMPERATURE PROFILE OF PACKED-BED NON-THERMAL PLASMA
REACTOR AND ITS EFFECT ON TOLUENE DECOMPOSITION

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TEMPERATURE PROFILE OF PACKED-BED NON-THERMAL PLASMA
REACTOR AND ITS EFFECT ON TOLUENE DECOMPOSITION

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DEDICATION

To Allah (SWT),

To my beloved parents,

Rosdi Salleh and Che Salbiah bt Awang Mat,

To my husband,

Mohd Akram Mat Deris

To my son,

Muhammad Aqil Hadif

And to my siblings,

Mohd Zulhilmie, Nur Izzati, Mohammad Zahran, and Muhammad Zulhazreen

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ABSTRACT

This study aims to profile real plasma temperature inside the packed-bed (PB) non-thermal plasma (NTP) reactor using a fiber Bragg gratings (FBG) and its effect on toluene decomposition efficiency. PB reactor was designed and fabricated by packing some dielectric material of barium titanate (BaTiO_3) pellets between two stainless steel electrodes. The FBG was embedded inside the reactor to measure the plasma temperature within the plasma stream. Plasma temperatures for four carrier gases, helium (He), argon (Ar), nitrogen (N_2), and air were profiled at different applied voltages ranging between 4 and 16 kV based on their breakdown voltage to determine suitable gases for toluene decomposition process that has good temperature stability and no arc formation. For noble gases He and Ar, the plasma temperatures are in the range of 25-80°C and 60-170°C, respectively, while those of N_2 and air are in the range of 28-200°C. Air was selected as carrier gas for toluene decomposition process due to higher plasma temperature, no arc formation and higher free oxygen radicals in the plasma stream. The results show that the plasma temperature increases with the increase in applied voltage, and with the decrease in flow rate and toluene input concentration. The average plasma temperature for toluene decomposition in air is in the range of 100-260°C when measured under applied voltage of 14-19 kV, carrier gas flow rate of 1.0-2.0 L/min and toluene input concentration of 500-8400 ppm. Complete toluene decomposition efficiency has been achieved under plasma parameters of 18 kV, 2.0 L/min and 500 ppm. From this finding, plasma temperature profiling using FBG sensor can be used as plasma diagnostic tool to replace Fourier Transform Infrared spectroscopy (FTIR) instrument and as indicator when toluene decomposition process is complete.

ABSTRAK

Kajian ini bertujuan untuk memprofilkan suhu sebenar plasma di dalam *packed-bed* (PB) reaktor plasma bukan terma (NTP) menggunakan parutan gentian Bragg (FBG) dan kesannya terhadap kecekapan penguraian toluena. Reaktor PB direkabentuk dan difabrikasi dengan memadatkan sejumlah bahan dielektrik barium titanat (BaTiO_3) pelet di antara dua elektrod keluli kalis karat. FBG dimasukkan ke dalam reaktor untuk mengukur suhu dalam aliran plasma. Suhu plasma bagi empat gas pembawa, helium (He), argon (Ar), nitrogen (N_2), dan udara telah diprofilkan pada voltan berbeza antara 4 dan 16 kV berdasarkan voltan rosak mereka untuk mencari gas yang sesuai untuk proses penguraian toluena yang mempunyai kestabilan suhu yang baik dan tiada pembentukan arka. Untuk gas lengai He dan Ar, suhu plasma masing-masing berada dalam lingkungan 25-80°C dan 60-170°C, manakala untuk N_2 dan udara berada dalam lingkungan 28-200°C. Udara dipilih sebagai gas pembawa untuk proses penguraian toluena disebabkan faktor suhu plasma yang lebih tinggi, tiada pembentukan arka dan lebih banyak penghasilan radikal oksigen bebas dalam aliran plasma. Keputusan menunjukkan suhu plasma bertambah dengan peningkatan voltan, dan dengan pengurangan kadar aliran dan kepekatan input toluena. Suhu purata plasma untuk penguraian toluena dalam udara berada dalam lingkungan 100-260°C apabila diukur pada voltan antara 14-19 kV, kadar aliran gas pembawa 1.0-2.0 L/min dan kepekatan input toluena 500-8400 ppm. Kecekapan penuh penguraian toluena dicapai pada parameter plasma 18 kV, 2.0 L/min dan 500 ppm. Daripada dapatan ini, pemprofilan suhu plasma menggunakan penderia FBG boleh digunakan sebagai alat diagnostik plasma untuk menggantikan instrumen spektroskopi inframerah transformasi Fourier (FTIR) dan sebagai penanda apabila proses penguraian toluena lengkap.

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LIST OF SYMBOLS/ABBREVIATION

A	-	Absorbance
Ar	-	Argon
A_e	-	Area under experimental toluene absorption band
A_s	-	Area under standard database toluene absorption band
AC	-	Alternating Current
ATR	-	Attenuated Total Reflectance
Ar	-	Argon
APPJ	-	Atmospheric Pressure Plasma Jet
BaTiO ₃	-	Barium titanate
BTEX	-	Benzene, Toluene, Ethyl benzene and Xylene
DBD	-	Dielectric Barrier Discharge
C	-	Carbon
C.C	-	Continuous Current
CD	-	Corona Discharge
CFCs	-	Chlorofluorocarbons
CH ₄	-	Methane
CO	-	Carbon monoxide
CO ₂	-	Carbon dioxide
C ₇ H ₈	-	Toluene
c	-	Molar concentration
DC	-	Direct Current
ESP	-	Electrostatic precipitators

EPA	-	Environmental Protection Agency
ϵ	-	Molar absorptivity
\bar{e}	-	Electron
FBGs	-	Fiber Bragg Gratings
FTIR	-	Fourier Transform Infrared Spectroscopy
GAD	-	Gliding Arc Discharge
Ge	-	Germanium
GPIB	-	General Purpose Interface Bus
HD	-	High – intensity arc Discharge
HNO ₃	-	Nitric acid
He	-	Helium
Hg	-	Mercury
HV	-	High Voltage
H ₂ O	-	Water
H ₂ O ₂	-	Hydrogen peroxide
I	-	Intensity transmitted light
I ₀	-	Intensity of incident light
KBr	-	Potassium bromide
L	-	Grating length
L_m	-	Pathlength experimental
L_s	-	Pathlength standard gas
LTE	-	Localized Thermodynamic Equilibrium
l	-	thickness
MnO ₂	-	Manganese dioxide
NIST	-	National Institute of Standards and Technology
NTP	-	Non-Thermal Plasma
Ne	-	Neon
NO	-	Nitrogen oxide
NO _x	-	Nitrogen oxides

NO ₂	-	Nitrogen dioxide
N ₂	-	Nitrogen
N ₂ O	-	Nitrous oxide
N_s	-	Concentration standard database (100 ppm)
n_e	-	Electron density
n_{eff}	-	effective refractive index
OH	-	Hydroxide
OES	-	Optical Emission Spectrometer
OSA	-	Optical Spectrum Analyser
O ₂	-	Oxygen
O ₃	-	Ozone
PB	-	Packed- Bed
PD	-	Pulsed Discharged
PE	-	Polyethylene
P	-	Power
PM	-	Particulate matter
PMT	-	Photomultiplier tube
PM ₁₀	-	Coarse particle
PM _{2.5}	-	Fine particle
ROS	-	Reactive Oxygen Species
RSM	-	Response Surface Methodology
SD	-	Surface Discharge
SVE	-	Soil Vapor Extraction
SO ₂	-	Sulfur oxide
TP	-	Thermal Plasma
T	-	Temperature
T_{avg}	-	Average temperature
T_e	-	electron temperature
T_g	-	gas temperature

T_i	-	ion temperature
T_o	-	Heavy particles temperature
T_{rot}	-	Rotational temperature
T_v	-	Vibrational temperature
t	-	Time
TiO_2	-	Titanium dioxide
UV	-	Ultraviolet
VD	-	Volume Discharge
VOCs	-	Volatile Organic Compounds
V	-	Voltage
α	-	Thermal expansion coefficient
ζ	-	Thermo – optic coefficient
λ	-	Wavelength
λ_B	-	Bragg wavelength
λ_{Bo}	-	Initial Bragg wavelength
$\Delta\lambda_B$	-	Bragg wavelength shift
ΔT	-	Temperature difference
Λ	-	Bragg grating period
Y- Al_2O_3	-	Gamma alumina

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Plasma, also referred to as ‘ionized gas’ is generated when thermal, electrical or light energy reacts with a gas. An electron is moved out from the gas atom outermost shell, and thus forms a collection of free-moving ions and electrons. Plasma is divided into two general categories, namely, thermal plasma (TP) and non-thermal plasma (NTP). In thermal plasma, all species of particles, including neutral atoms, neutral molecules, ions and electrons, are in thermal equilibrium. Meanwhile, in NTP, most of the discharged energy is transformed into the production of high energetic electrons, instead of heating the neutral and ions that remain at or near room temperature. Therefore, the electrons, ions and neutrals are not in thermal equilibrium. This NTP technology with a high production of energetic electrons leads to a higher production of active chemical species, and thus becomes a promising tool for numerous applications, including surface treatment, plasma actuators, biomedical domain, sterilization, plasma medicine, as well as environmental and industrial pollutant abatement [1–5]. This NTP technology is widely used to treat various environmental pollutants such as diesel exhaust cleaning [6], water treatment [1,7] , air pollutants [8–10] and Volatile Organic Compounds

(VOCs) [11–16]. VOCs are carbon-based chemicals that can easily evaporate to vapor or gas at room temperature, due to their low boiling point. Some examples of VOCs include benzene, toluene, xylene (BTEX), styrene, methane, ethane, methyl chloride and formaldehyde. VOCs are widely released by residential areas, pharmaceuticals industries, high-tech and commercial industries, petrochemical industries, as well as household products such as solvent and paint thinner, lubricants, gasoline, oil refineries, detergents and dry cleaning fluids [17,18].

The conventional techniques available to control VOCs emission include adsorption, absorption, condensation and membrane separation technology, which is only categorized as a recovery process [19–21]. These techniques do not destroy VOCs, but are able to be recovered and reused. Conversely, NTP technology is an abatement process which destructs VOCs. VOCs should be controlled and treated because short and long term exposure to VOCs has adverse health effects on humans, animals and the surrounding environment [22–24]. The long exposure might cause kidney failure, child birth defects and even death.

NTP process for VOCs pollutant removal could be generated by methods including an electron beam and electrical discharges such as pulse power discharge [25], corona discharge [26] and dielectric barrier discharge (DBD) [27–30]. In this study, DBD with a designed packed-bed (PB) reactor was constructed using a several basic elements including two electrodes, a high voltage power supply, a discharge gap between the electrodes, and dielectric materials and a carrier gas.

Previous studies in the related literature mostly focused on plasma properties such as energy density [31–34] and electron temperature [35–37], but there is a lack of related studies on real plasma temperature within plasma streams inside the

plasma reactor. Real plasma temperature measurement plays an important role in chemical reactions and in-situ measurement, mainly because it is temperature-dependent. This study focuses on monitoring the real plasma temperature using established optical tools, Fiber Bragg Grating (FBGs) which is a fast response technique to profile the plasma temperature during the entire VOCs decomposition process using an NTP system. FBGs are embedded inside the PB reactor and is monitored in real-time via the installed LabVIEW software application. Three plasma parameters, including applied voltage, carrier gas flow rates and VOCs (toluene) input concentrations were varied to examine the impact on the plasma temperature profile. The correlation regarding plasma temperature behavior, toluene decomposition rate and its by-product formation, were thoroughly investigated.

1.2 Problem Statement

The fundamental study of temperature is crucial in the NTP decomposition process because it plays a vital role for chemical reactions during gas phase analysis and plasma diagnostic, especially for in-situ measurements. The increment of temperature enhances the chemical reaction rate by breaking and dissociating more VOCs molecular bonding and enhances the VOCs decomposition. To date, information regarding real plasma temperature behavior inside plasma reactors remains undiscovered, since studies on temperature monitoring techniques within the plasma streams are lacking. The conventional technique commonly employed in NTP technology to measure the plasma temperature is using a thermocouple, but this is limited to the outer plasma reactor wall due to metal based component leads to electrical field interruption.

In addition, the temperature measured outside the plasma reactor does not indicate the actual plasma temperature because the nature of plasma is strongly

localized. A laser gun technique is also inappropriate for plasma temperature measurement inside the PB reactor due to the presence of dielectric beads within the plasma stream. Therefore, the real plasma temperature monitoring inside the plasma reactor is necessary to study the temperature within the plasma stream.

FBGs seem to offer an alternative method to measure the real plasma temperature within the plasma streams inside the plasma reactor, since they are highly temperature sensitive, small, flexible, able to operate at temperatures up to and beyond 1000°C [38], have a fast response technique, and are free from electrical fields and electromagnetic field interruption, since FBGs are insulator-based components. Besides, FBGs optical sensing is able to be placed in between tight packed dielectric beads without affecting the dielectric properties of the beads. This FBGs sensor is connected via the LabVIEW program in order to allow the actual real plasma temperature profiling.

1.3 Objectives

The objectives for this research are as follows:

- 1) To design and fabricate atmospheric pressure non-thermal packed-bed plasma reactor system for VOCs decomposition
- 2) To profile plasma temperature within plasma streams inside packed-bed reactor through an in-situ and real-time temperature measurement using FBGs
- 3) To determine the impact of applied voltage, carrier gas flow rate and toluene input concentration on plasma temperature profile
- 4) To investigate the behavior of plasma temperature profile with toluene decomposition efficiency and its by-products from the decomposition process

1.4 Scope of Study

In this study, the NTP system was developed with a self-designed and fabricated PB reactor. This system was tested for the PB reactor, which consists of barium titanate (BaTiO_3) dielectric pellets in between the discharged electrodes, and is suitable to be operated within the applied plasma parameters range. The FBGs is embedded inside the plasma reactor to allow plasma temperature to be monitored within the plasma stream via in-situ and real-time process. The plasma temperature profile is monitored by observing Bragg wavelength shift, $\Delta\lambda_B$ from Optical Spectrum Analyser (OSA), and is displayed using the LabVIEW program via GPIB connector. Emitted radiation from plasma generation is collected simultaneously with the recorded plasma temperature using special grade fibre, Optical Emission Spectrometer (OES) by monitoring the profiles of peaks intensity stability. When applied voltage is higher, the emission of intensity peaks is increased and since FBGs are made up of Ge-doped based fiber it could absorb UV radiation when reached maximum operated voltage and permanently altered the refractive index. Thereby, monitoring the profiles of peaks intensity stability using OES indicate and ensure harmless UV emission range, which is safe for FBGs temperature measurement without permanently altering the refractive index.

The plasma temperature profiles are investigated under the influence of plasma operating parameters including applied voltage (14 – 19 kV), carrier gas flow rates (1.0 – 2.0 L/min) and toluene initial concentration (500 – 8400 ppm). By varying the plasma parameters, the behavior of plasma temperature profile is thoroughly investigated and correlated with the decomposition efficiency of toluene and its by-products.

1.5 Significance of Study

The information of the real plasma temperature is significant in the NTP decomposition process, since the chemical reaction process and quantitative analysis used such as Fourier Transform Infrared (FTIR) spectroscopy and laser spectroscopy for in-situ measurement are very dependent on temperature. For quantitative measurement using FTIR, Beer-Lambert Law is strongly recommended and the absorbance is temperature dependent parameter. Since the analysis dependent on standardly-acquired temperature database, hence calculation referring at inaccurate temperature database influenced to the error in temperature measurement and thereby causes inaccuracy in the quantitative analysis of VOCs decomposition efficiency. The fast response technique of FBGs provides novel knowledge, since it is able to monitor and profile the real plasma temperature within the plasma stream inside the plasma reactor. Therefore, FBGs can be applied to investigate the impact of the plasma parameters on the plasma temperature behavior, and can be performed as a suitable technique to measure VOCs decomposition efficiency.

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