DESIGN AND FABRICATION OF ARC THERMAL PLASMA REACTOR FOR PETROLEUM SLUDGE TREATMENT

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DEDICATION

To Almighty Allah (SWT), then, my beloved parents, my family and friends

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ABSTRACT

Over 130, 000 metric tonnes of toxic petroleum sludge are generated yearly in Malaysia. The traditional methods of disposing of petroleum sludge are short of providing the much-needed benign treatment. A more robust treatment technique is therefore desirable. The thermal plasma treatment technique is employed to bridge the gap. In this research, a 4.7 kW thermal plasma reactor was designed and fabricated. The output current and the plasma temperature range were 5 – 200 A and 356 – 1694 ^oC respectively. After the treatment, the morphology of the sludge transformed from jelly-like to crystalline solid. A mass reduction of 36.87 – 91.40% and a total organic compound reduction of 21.47 – 93.76% were achieved in a treatment period of 2 – 5 minutes. The leaching test indicates that the heavy metals were stabilized in the solid, and hence, the solid is safe for secure landfill. The product gas is a mixture of carbon monoxide (CO), carbon dioxide (CO_2), hydrogen (H_2), water (H_2O), methane (CH_4), acetylene (C_2H_2) and ethylene (C_2H_4). The concentrations of the greenhouse gases, CH_4 and CO_2, were small. The lower heating value and the cold gas efficiency of the gas were 7.40 – 7.86 MJ/Nm^3 and 25.22 – 51.90% respectively. The efficiency is within the range of the efficiency of gasification of petroleum sludge in an updraft gasifier. Based on the operating cost estimation, a profit margin of RM 3.11/kg of sludge was achieved. Two quadratic models, one for cold gas efficiency and the other for CO/CO_2 ratio were developed. The developed models, using response surface methodology, showed a good fit with correlation coefficients of 99.32% and 99.66% for cold gas efficiency and CO/CO_2 ratio respectively. The optimum operating conditions for the treatment were arc current = 188.15 A, plasma gas flow-rate = 31.54 L/min and treatment time = 1.89 min. The optimum responses obtained from the optimization of the reaction system were 52.59% and 1.80 for cold gas efficiency and CO/CO_2 ratio respectively with the desirability of 1. Thermal plasma technique is, therefore, an alternative method for treating petroleum sludge.

ABSTRAK

Lebih 130,000 tan metrik enapcemar petroleum bertoksik yang dihasilkan setiap tahun di Malaysia. Kaedah pelupusan tradisional enapcemar petroleum kurang menyediakan rawatan tidak merbahaya sangat diperlukan. Oleh itu, teknik rawatan yang lebih mantap adalah diperlukan. Teknik rawatan plasma haba digunakan untuk merapatkan jurang ini. Dalam kajian ini, reaktor plasma haba 4.7 kW direka bentuk. Arus elektrik keluaran dan suhu plasma masing-masing adalah dalam julat 5 - 200 A dan 356 - 1694 ^{o}C. Morfologi enapcemar berubah dari seperti jeli kepada pepejal kristal selepas rawatan. Pengurangan jisim sebanyak 36.87 - 91.40% dan pengurangan jumlah sebatian organik sebanyak 21.47 - 93.76% telah dicapai dalam tempoh rawatan 2 - 5 minit. Ujian pengurasan menunjukkan bahawa logam berat telah distabilkan didalam pepejal dan oleh itu pepejal adalah selamat untuk ke tapak pelupusan. Gas produk adalah terdiri dan campuran karbon monoksida (CO), karbon dioksida (CO_2), hidrogen (H_2), air (H_2O), metana (CH_4), acetilena (C_2H_2) dan etilena (C_2H_4). Kepekatan gas rumah hijau, CH_4 dan CO_2, adalah sangat kecil. Nilai haba rendah dan kecekapan gas sejuk masing-masing adalah 7.40 - 7.86 MJ / Nm^3 dan 25.22 - 51.90%. Kecekapan tersebut adalah dalam julat kecekapan pengegasan enapcemar petroleum dalam pengegas naik. Berdasarkan anggaran kos operasi, margin keuntungan sebanyak RM 3.11/kg enapcemar telah dicapai. Dua model kuadratik, satu untuk kecekapan gas sejuk dan satu lagi untuk nisbah CO/CO_2 telah dibangunkan. Model yang dibangunkan menggunakan kaedah permukaan sambutan menunjukkan kesesuaian dengan pekali korelasi 99.32% dan 99.66% masing-masing untuk efisiensi gas dingin dan nisbah CO/CO_2. Keadaan operasi optimum untuk rawatan ialah arus arka = 188.15 A, kadar aliran gas plasma = 31.54 L/minit dan masa rawatan = 1.89 minit. Tindak balas optimum yang diperoleh daripada pengoptimuman sistem tindak balas adalah 52.59% dan 1.80 masing-masing untuk kecekapan gas sejuk dan nisbah CO/CO_2 dengan kebaikan 1. Teknik plasma haba adalah kaedah alternatif untuk merawat enapcemar petroleum.

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LIST OF ABBREVIATIONS

AC	-	Alternating Current
ASTM	-	American Standard Test Method/ American Society for Testing Materials
ANOVA	-	Analysis of Variance
APC	-	Air Pollution Control
CCD	-	Central Composite Design
CV	-	Coefficient of Variance
DC	-	Direct Current
DRE	-	Destruction and Removal Efficiency
DSS	-	Dutch Standard for Soil
DTG	-	Derivative Thermo – Gravimetric
EDS	-	Energy Dispersion Spectroscopy, Effective Disposal of Sludge
EQA	-	Environmental Quality Act
EQR	-	Environmental Quality Regulations
FAAS	-	Flame Atomic Absorption Spectrophotometer
FE-SEM	-	Field Emission Scanning Electron Microscopy
FT-IR	-	Fourier Transform Infrared Spectroscopy
GCMA	-	Gas Chromatography Mass Spectrometry
HHV	-	Higher Heating Value
ICP-OES	-	Inductively Coupled Plasma - Optical Emission Spectrometry
LHV	-	Lower Heating Value
MSW	-	Municipal Solid Waste
OFAT	-	One Factor At a Time
ORS	-	Oil Recovery from Sludge
OVAT	-	One Variable At a Time
PAED	-	Pulsed Arc Electrohydraulic Discharge

PAHs	-	Polynuclear Aromatic Hydrocarbons
PCBs	-	Polychlorinated Biphenyls
PCDD	-	Polychlorinated Dibenzo Dioxins
PCDF	-	Polychlorinated Dibenzo Furans
PGM	-	Plasma Gas Welding
Petronas	-	Petroliam Nasional Berhad
PHCs	-	Petroleum Hydrocarbons
RDF	-	Refuse Derived Fuel
RF	-	Radio Frequency
RGTS	-	Russian General Toxicological Standard
RSM	-	Response Surface Methodology
SEM	-	Scanning Electron Microscopy
SEOR	-	Surfactant Enhanced Oil Recovery
SW	-	Scheduled Waste
TC	-	Total Carbon
TCLP	-	Toxicity Characteristic Leaching Procedure
TGA	-	Thermogravimetric Analysis
TIC	-	Total Inorganic Carbon
TIG	-	Tungsten Inert Gas
TOC	-	Total Organic Carbon
TPR	-	Thermal Plasma Reactor
TPT	-	Thermal Plasma Treatment
US EPA	-	United State Environmental Protection Agency
VOCs	-	Volatile Organic Compounds
XRD	-	X-Ray Diffractometry

LIST OF SYMBOLS

A	-	Ampere
Cg	-	Syngas heating value (kJ/m^3)
Cp	-	TOC of product
Cs	-	TOC of untreated petroleum sludge
Ct	-	Sludge heat value (kJ/kg)
$\bar{E}$	-	Electric field
e^-	-	Electron
hv	-	Ultraviolet radiation
I	-	Input current (A)
kJ	-	kilo-Joule
kW	-	kilo-Watt
kWA	-	kilo-Watt-hour
k_B	-	Boltzmann constant
M_L	-	Mass of solid product
n_e	-	Electron density
n_i	-	Number density on ions
n_n	-	Number density on neutral atoms
P	-	Plasma torch power (kW), energy input (kWh)
T_e	-	Electron temperature
T_g	-	Gas temperature
T_i	-	Ion temperature
T_p	-	Plasma temperature
V	-	Applied voltage (V)
V_g	-	Volume of syngas (Nm^3)
$\langle Z \rangle$	-	Average charge state of ion

$+, A^+$	-	Cat-ion
O, A	-	Neutral atom
α	-	Degree of ionization
ϕ	-	Electric potential
η_g	-	Conversion efficiency
η	-	Cold gas efficiency
η_c	-	Carbon conversion efficiency

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CHAPTER 1

INTRODUCTION

1.1 Problem Background

There is a growing concern on the sludge generated from petroleum processing and refining due to the presence of high concentration of petroleum hydrocarbons (PHCs) and heavy metals (Robertson *et al.*, 2007; Wang *et al.*, 2007). Improper or insufficient disposal can cause serious environmental consequences (Suganthi *et al.*, 2018) and health implications on human (Fytili and Zabaniotou, 2008; Hu *et al.*, 2013). Methods employed for petroleum sludge disposal include landfills, agricultural application (Nasreen and Kalsoom, 2018), solidification/stabilization (Benlamoudi *et al.*, 2018; Murshid *et al.*, 2018) and incineration (Xie *et al.*, 2014). However, these technologies present considerable drawbacks. Disposal of sludge into landfills causes greenhouse gas emissions (CH_4 and CO_2) and seepage of heavy metals into soil and groundwater (Battikhi, 2014; Celary and Sobik-Szołtysek, 2014; Okoro and Adoki, 2014). The disposal method is no longer a viable solution due to the rising cost of land and shutdowns of landfills (Sanin *et al.*, 2011). Farmland application of sludge is being hindered by the limited uptake capacity of soil and the potential pollution by heavy metals (Hu *et al.*, 2013; Varavipour *et al.*, 2009).

Incineration has been applied to the treatment of industrial sludge (Chen, 2004; Gong *et al.*, 2018; Liu *et al.*, 2009b; Scala and Chirone, 2004; Zhou *et al.*, 2009). It is one of the most promising and competitive technology. Incineration has the ability to destroy hazardous constituents such as organics and pathogens and to reduce the volume and mass of landfill material to barest minimum (Hu *et al.*, 2013; Pan *et al.*, 2013; Zhao *et al.*, 2010; Zheng and Koziński, 2000). However, the technology is challenged with the emission of great quantities of volatile organic compounds (VOCs) and fly ash that needs further treatment (Hu *et al.*, 2013; Xie *et al.*, 2009), and the cost of pollution-control of the flue gases is very high (Quina *et al.*, 2008).

Currently and in the recent past, thermal plasma technique is being applied to the treatment of waste (Ali *et al.*, 2018; Ducharme and Themelis, 2010; Leal-Quiros, 2004; Panepinto and Genon, 2014; van der Walt *et al.*, 2017). The technology has the ability to gasify the organic components of the waste and vitrify the inorganics (Mazzoni and Janajreh, 2017; Sanlisoy and Carpinlioglu, 2017). The advantages of thermal plasma technology over other treatment methods include high reaction temperatures, independence of additional fuel, shorter residence times, high energy densities, high heat transfer rate, lower volume of stack gases, lower ash volume, lower unburned components in the ash and reduced footprint (Kaldas *et al.*, 2007; Kezelis *et al.*, 2004; Tang *et al.*, 2013).

The thermal plasma technology has also been demonstrated as an effective and environmentally friendly technique for the treatment of tannery sewage sludge (Bień *et al.*, 2013; Celary and Sobik-Szołtysek, 2014), ship oil sludge (Kaldas *et al.*, 2007), wastewater sludge (Leal-Quirós and Villafañe, 2007; Mountouris *et al.*, 2008), storm-water sludge (Li *et al.*, 2015a; Li *et al.*, 2012; Li *et al.*, 2015b), wet paper sludge (Shie *et al.*, 2014), electroplating sludge (Li *et al.*, 2007; Ramachandran and Kikukawa, 2002), galvanic sludge (Abdulkarim *et al.*, 2018; Cubas *et al.*, 2014) and a mixture of sewage sludge and fly ash (Kim and Park, 2004; Sobiecka and Szymanski, 2014). This research considers the use of a thermal plasma reactor to treat petroleum sludge. The treatment method is expected to address the leaching of heavy metals and reduce the generation of greenhouse gases.

1.2 Problem Statement

About 133,260 MT of petroleum sludge is generated yearly from petroleum and petrochemical processes in Malaysia (D.O.E., 2012). The sludge is harmful due to the presence of high concentration of petroleum hydrocarbons (PHCs) and heavy metals. Improper or insufficient disposal can cause serious environmental consequences and health implications. Traditional methods for treating petroleum sludge include incineration, stabilization/solidification and biodegradation. These methods have considerable drawbacks. Incineration is challenged with the generation of toxic fly and bottom ash. There is the production of finely inhalable particles, laced with toxic metals, alongside fly ash. Whereas, the bottom ash contained leachable heavy metals (Solution, 2018).

Waste stabilization/solidification using cement as a binding agent is another waste disposal method applied to industrial sludge. The technology has the ability to retained heavy metals in a hardened solid matrix and prevents leaching of the heavy metals from the matrix into the environment. The technology which is best suited for inorganic waste sludge may not be suitable for petroleum oily sludge. The oily sludge is primarily organic. Organic compounds form protective a layer around cement grain, hindering hydration and retarding setting process (Omar *et al.*, 2008). The alternative biodegradation treatment is a slow process; it takes 6 – 24 months to complete. Heavy metals and some chlorinated compounds are not amenable to biodegradation. The slow pace of the process of biodegradation allows the migration of heavy metals into the surface and groundwater. Also, the interaction of the atmospheric air with the sludge causes emission of CO_2 and VOCs (Ubani *et al.*, 2013).

There is, therefore, the need for a more robust treatment method that will drastically reduce the volume of sludge and at the same time prevent or minimize the production of greenhouse gases and prevent leaching of heavy metals into the groundwater. Thermal plasma treatment technology poses these qualities. The high-temperature regime in the thermal plasma has the ability to gasify organic compounds into a synthetic gas of economic value and solidify the inorganics in a vitreous matrix, preventing the leaching of heavy metals into the surrounding.

At present, there is no documented literature on the treatment of petroleum sludge in arc thermal plasma reactor. Similarly, there is no known research available to the researcher on the design and fabrication of thermal plasma reactor for petroleum sludge treatment. The propose arc thermal plasma reactor will treat petroleum sludge and reduced the volume to less than 10%. The reactor will also convert the petroleum sludge to syngas that can be used for electricity generation and vitreous slag that encapsulate heavy metals and prevent their leaching to the environment. Furthermore, the proposed mathematical model equation will provide an understanding of the effect of process variables (arc current, residence time and plasma gas flow-rate) on the arc thermal plasma reactor (TPR) performance (cold gas efficiency and CO/CO_2 ratio).

1.3 Objectives of the Study

The main objectives of this study are as follows:

1. To characterize petroleum oily sludge prior to thermal plasma treatment.
2. To design and fabricate a thermal plasma reactor (TPR)
3. To evaluate the performance of the TPR on the treatment of petroleum sludge.
4. To optimize the system using response surface methodology (RSM).

1.4 Scope of Study

The scope of this research includes the collection of the sludge from Petronas Penapisan Melaka Sdn. Bhd. The characterization of the sludge to determine the moisture, volatiles, ash content, fixed carbon, total organic carbon (TOC), elemental composition and morphological structure. The designing (using AutoCAD 2018) and fabrication of the arc plasma reactor. The analysis of the solid and gaseous products to determine the TOC, heavy metal composition and the composition of the gas. The conduct of the leaching test on the product solid to evaluate the effectiveness of the reactor system in encapsulating the heavy metals in the solid matrix. And finally, the optimization of the treatment system.

1.5 Significances and Original Contributions of This Study

This project is significant because the world cannot do without petroleum product and it is only available through exploration and refining, thus hazardous petroleum sludge is inevitable. The safe and environmentally friendly treatment method is a better solution to hazardous and harmful effects of petroleum sludge. Most of the traditional methods of treating petroleum sludge are short of providing complete destruction of harmful VOCs and fly ash. TPT will break the molecular bond and convert the VOCs into simple compounds like CO and H_2 that can be used for energy generation. Methane and carbon dioxide are major contributors to Global Warming, they must be removed or reduced to barest level from exhaust gas to meet environmental regulations. Companies require technology that will provide them with the tool to remove these toxic gases from their exhaust gas. The fabricated thermal arc plasma reactor is an approach for future undertaking and would be able to customize the technology for particular requirement. The research offers an alternative solution to National Oil and Gas company.

1.6 Organization of the Thesis

This thesis consists of five (5) chapters, the introduction, the literature review, the methodology, the result and the discussion of the result and the conclusion section. Each chapter provides detail information about the specific research area. A more elaborate description of chapters 2 – 5 is given below.

Chapter 2 is the literature review. It covers background information related to the research which includes general information on petroleum and petrochemical sludge and thermal plasma technology. It also covers existing information on the applications of thermal plasma technology in industrial sludge treatment and modelling and optimization using response surface methodology (RSM).

Chapter 3 is the methodology section. The chapter described in detail the process equipment used for data collection and data analysis. It detailed out process equipment and specification, the method for sample collection and characterization, procedure for sludge treatment in TPR, procedure for products characterization and analysis and finally, the procedure for performance optimization.

Chapter 4 is the section for result and discussion. The chapter focuses on the discussion of the findings from the characterization of petroleum sludge and evaluation of the products obtained from the plasma treatment of the petroleum sludge in a transferred arc TPR. Parameters like plasma temperature, total organic carbon (TOC), leachability of heavy metals, CO/CO_2 ratio and product gas efficiency were discussed. The chapter also presents a detailed discussion of the modelling and optimization of the thermal plasma reactor for the treatment of petroleum sludge.

Chapter 5 is the conclusion and recommendation section. In this chapter, the conclusions drawn from the research findings are presented. Also, recommendations for future study, in this research area, is given.

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