



Development of a Laboratory-Scale Thermal-Arc-Plasma Reactor and its Application in the Pyrolysis of Petroleum Oily Sludge

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Abstract: Waste treatment using thermal arc plasma is well established and laboratory/pilot scale plasma reactors were developed and their performances for the destruction of different hazardous wastes, other than petroleum oily sludge, were studied. This work aims to extend the plasma technology to the pyrolysis of hazardous petroleum oily sludge. A 4.7 kW thermal arc plasma reactor was developed using a standard TIG arc welding torch. The transferred arc plasma reactor was used to treat 20 g/batch of petroleum oily sludge. The prevailing temperature inside the reactor ranges between 356 – 1694 °C. The plasma arc temperature increased with increasing plasma arc current and also with increasing plasma gas flow-rate. A vitreous slag and a flue gas were generated as products. A mass reduction of between 36.87 – 91.40% and a TOC reduction of 21.47 – 93.76% were achieved in the treatment time of 2 – 5 min. The mass reduction was observed to increase with treatment time. However, the increase was more rapid between the 3rd and the 4th min of the treatment. The flue gas produced contains H₂ (43.79 – 50.97 mol%), H₂O (26.60 – 30.22 mol%), CO (8.45 – 11.18 mol%), CO₂ (5.12 – 10.35 mol%), CH₄ (2.17 – 3.38 mol%), C₂H₂ (0.86 – 2.69 mol%) and C₂H₄ (0.76 – 2.17 mol%). Thus, the thermal plasma reactor provides a suitable method of treating petroleum oily sludge.

Keywords: Thermal plasma, petroleum oily sludge; plasma arc temperature; mass reduction; carbon conversion.

1.0 Introduction

Thermal arc plasma technology has become a prominent waste treatment technique for a wide variety of waste because of the shortcomings of traditional waste disposal methods (A.M. Ali, Abu Hassan, & Abdulkarim, 2016). The arc plasma treatment technology has been identified as a potentially effective tool for producing less harmful by-products which can be used in building and road construction (Kourti et al., 2011; Tu et al., 2008). The innovative plasma technique involves subjecting waste material to high-temperature arc plasma such that the organics and the volatile species are gasified while the inorganics and non-volatiles are chemically bonded in a vitreous matrix, thereby making them resistant to leaching of heavy metals (Agon, 2015). Thermal arc plasma provides a suitable treatment technique for special waste disposal requirements. Advantages of thermal arc plasma treatment technique over conventional incineration include high-temperature regime, high waste volume reduction, low gas throughput, process flexibility in either oxidizing or a reducing environment, and can effectively treat a wide variety of waste types

There is an increase in the documented research, in the last two decades, concerning the destruction of hazardous wastes using thermal arc plasma technique. The growing interest of academic research in such an area cannot be unrelated to the ability of the technique to reduce waste volume by over 80% and produce benign byproducts. (A.M. Ali et al., 2016; Heberlein & Murphy, 2008). The plasma gasification of the organic portion of sludge has attracted interest as a source of energy and spawned process developments for the treatment of sludge from different sources (Bień, Celary, Morzyk, Sobik-Szołtysek, & Wystsalska, 2013; Celary & Sobik-Szołtysek, 2014; Cubas et al., 2014; Kim & Park, 2004; Leal-Quirós & Villafañe, 2007; C. Li, Lee, Huang, Fu, & Lai, 2007; O.L. Li, Guo, Chang, Urashima, & Saito, 2012; O. L. Li, Guo, Chang, Urashima, & Saito, 2015; O.L. Li, Guo, J.S., & Saito, 2015; Mohai & Szépvölgyi, 2005; Mountouris, Voutsas, & Tassios, 2008; Ramachandran & Kikukawa, 2002; Shie, Liau, Lin, & Chang, 2014; Sobiecka & Szymanski, 2014). Factors like treatment efficiency, plasma gas flow-rate, the treatment time of a batch operation, feed flowrate of continuous operation and inter-electrode separation were investigated.

Feasibility studies involving design and fabrication of thermal arc plasma reactors for hazardous waste destruction are also documented in the literature. In the USA a laboratory-scale thermal arc plasma reactor consisting of a highly instrumented furnace equipped with a 75 kW transferred arc plasma torch, was developed and used to study the physical and chemical behaviour of metal-spiked waste (nickel and chromium) in a high-temperature arc plasma regime (Cortez et al., 1996). In Thailand, a 20kW laboratory-scale, atmospheric-air DC plasma reactor was designed and fabricated using a non-transferred plasma torch and its performance was evaluated using electronic waste (Tippayawong & Khongkrapan, 2009). A research team in Brazil developed a small-scale, continuous-flow plasma reactor consisting of a torch with graphite electrodes and an integrated nebulization furnace. The reactor was used to eliminate carbon-

tetrachloride from liquid waste (Cubas, Carasek, Debacher, & De-Souza, 2005). In the Durgapur city of West Bengal, a 20 kg/hr plasma reactor for the treatment of waste plastic was developed, and its performance on the pyrolysis of waste plastic and energy generation was studied (Punčochář, Ruj, & Chatterj, 2012). Other similar studies involving design and evaluation of thermal plasma reactor for hazardous waste destruction were reported (Barcza, 1986; Khongkrapan, Thanompongchart, Tippayawong, & Kiatsiriroat, 2013; Szałatkiewicz, Szewczyk, Budny, Missala, & Winiarski, 2012, 2013; Tang, Huang, Zhao, Wu, & Chen, 2003; Townsend & Oehmig, 2014; Zhao, Huang, Wu, Li, & Chen, 2001).

It is obvious from the above discussion that waste treatment using thermal plasma technology has gain ground, and laboratory/pilot scale plasma reactors were developed and their performances for the destruction of hazardous waste were studied. However, it is not available to the knowledge of the authors, any attempt to develop a thermal arc plasma reactor that treats petroleum oily sludge. Thus, the present investigation was geared towards bridging this gap. In this study, a 20g batch-laboratory scale thermal arc plasma reactor was developed and used to treat petroleum oily sludge. Design parameters and the result of the test run of the reactor using petroleum oily sludge is, thus, presented.

2.0 Experimental set up and methodology

2.1 The Thermal Plasma Reactor

An exploded view of the plasma reactor is shown in Figure 1. The reactor consists of three major parts, the plasma torch, the anode and the furnace. The plasma torch is a standard TIG arc-welding torch. It is made up of a 2.4 mm diameter and 150 mm long lanthanated tungsten electrode (98% purity) inserted into a nozzle ejector. The nozzle ejector has an orifice opening through which argon gas (plasma forming gas) flows. The argon gas also cools the cathode electrode. The anode is a 10 mm diameter pure tungsten rod (98% purity). It is position vertically inside a jacketed brass that doubled as a holder and cooling jacket. The furnace is a hollow vertical cylinder of cast iron with horizontal extensions.

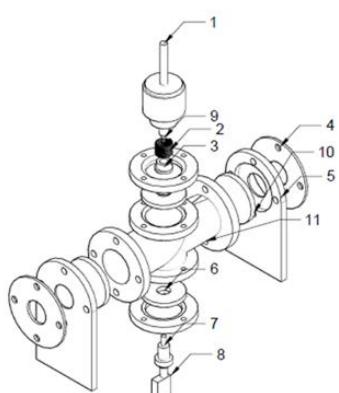


Figure 1. An exploded view of the thermal plasma reactor. (1) Plasma torch. (2) released spring, (3) cathode stopper, (4) back plate, (5) supporting leg, (6) marble plate, (7) anode, (8) cooling jacket, (9) cathode, (10) inspection window, (11) reactor chamber

Two Bakelite bars attached to both ends of the extensions supported the reactor in a vertical position. The inside top and the bottom of the furnace were protected with marble plates of thickness 10 mm each. A commercially available TIG generator (model: Master Weld TP - 2000) supplies the direct current needed for the generation of the arc. The specifications of the TIG master weld generator and the main components of the reactor are shown in Tables 1 and 2 respectively.

Table 1. TIG master weld specifications

Specification	Input rating	Output rating
Model	TP - 2000	
Efficiency (%)	80	
Power (kW)	4.7	
Voltage (V)	220/230/240	63
Current (A)	31	5 - 200

Table 2. Main components of the thermal plasma reactor

Component	Dimension				
	ID	OD	Thickness	Length/height	Material
Reactor furnace	85 mm	110 mm	-	116 mm	Cast iron
Cathode	-	2.4 mm	-	150 mm	Tungsten
Anode	-	10 mm	-	35 mm	Tungsten
Crucible	40 mm	-	4mm	47 mm	Mild steel

2.2 Experimental Method

The plasma arc temperature calibration was done prior to the pyrolysis experiment. All effort to ignite the arc when the gap between the electrodes was above 15 mm proved abortive. Likewise, when the gap was made too small, less than 5 mm, the two electrodes bridge together. Thus, a gap of 10 mm was maintained for both the reactor temperature calibration and the pyrolysis experiment. For the temperature calibration, a current of 100 A and argon flow-rate of 20 l/min were supplied to ignite and generate the arc plasma. The supplied current was increased gradually using a control nob while the plasma arc temperature was measured at intervals using an infrared thermometer (temperature range from 200 - 2200 °C). The procedure was repeated

with argon gas flow-rate of 25 l/min and 30 l/min respectively. The result of the plasma arc temperature measurement is presented in Figure 2.

The pyrolysis experiment was performed using petroleum oily sludge obtained from Petronas Penapisan Melaka Sdn. Bhd. The characteristics of the oily sludge were discussed elsewhere (Abubakar M. Ali et al., 2019). It was a black semi-solid cake characterized by high moisture, low volatiles and low fix carbon. It has an apparent density of 1.08 g/ml and a lower heating value (LHV) of 23.60 MJ/kg. The petroleum oily sludge was treated in the plasma reactor for 2, 3, 4 and 5 min respectively. In each run, 20 g of the wet oily sludge was placed in the plasma reactor and treated with arc plasma generated using an arc current of 160 A and argon gas flow-rate of 30 l/min. The set of the experimental run was repeated using thermal plasma generated with arc currents of 175 and 190 A respectively and argon gas flow-rate of 30 l/min. The flue gas generated in each run was passed through a cooling coil and a particle dust filter before collecting in a Teflon gas bag and analyzed in an offline FT-IR (Perkin Elmer, Frontier). The reactor was allowed to cool to room temperature and the solid remnant collected, weighed and analyzed using TOC analyzer (Model SSM-5000A) and ICP-OES machine (model: Agilent 710).

3.0 Results and Discussion

3.1 Temperature Profile Inside the Reactor

The plasma arc temperature profile (temperature vs arc current) is shown in Figure 2. The prevailing temperature inside the plasma reactor ranges between 356 - 1694 °C. At lower arc current, 100 - 140 A, the plasma arc temperature increases gradually from 360 - 600 °C. When the arc current is above 150 A, the increase in the plasma arc temperature is more rapid, from 600 - 1700 °C. This phenomenon could be explained by the increase in plasma density when arc current increases. At higher arc current, the plasma power is high which causes an increase in plasma density. Similar observations were reported by Tang and Huang (2005a) and (2005b). In the two separate studies, a direct increase in plasma arc temperature was observed when plasma power was increased. The effect of plasma gas flowrate on the prevailing plasma arc temperature inside the reactor is also depicted in Figure 2.0. At a current of 120 A and above the prevailing plasma arc temperature increases with plasma gas flowrate as manifested in the curves of 20, 25 and 30 l/min.

3.2 Mass and Volume Reduction

Two products, flue gas and a vitreous slag, were obtained from the pyrolysis of petroleum oily sludge in the thermal plasma reactor. The mass reduction was computed using Equation 1. A mass reduction of 36.87 - 91.40% was achieved in the treatment time of 2 - 5 min. The variation in mass reduction as a function of treatment time is shown in Figure 3. Between the 2nd and the 3rd minutes of treatment, there is a gradual increase in mass reduction with increased in the treatment time. This is the time when evaporation of the moisture in the sludge as well and

the decomposition of the oily sludge takes place. However, from the 3rd to the 4th minutes, the increase in mass reduction was very sharp, indicating that evaporation was completed at the 3rd minutes and that the entire thermal energy was used in the pyrolysis of the oily sludge. Beyond the 4th minutes, the mass reduction was insignificant. This signifies that the pyrolysis of the sludge was almost completed at the 4th minutes. The variation in arc current has a significant influence on the mass reduction of the oily sludge as shown in Figure 3. Within the three plasma arc currents considered, 160, 175 and 190 A, the highest mass reduction was obtained at an arc current of 190 A, while the lowest was at 160 A.

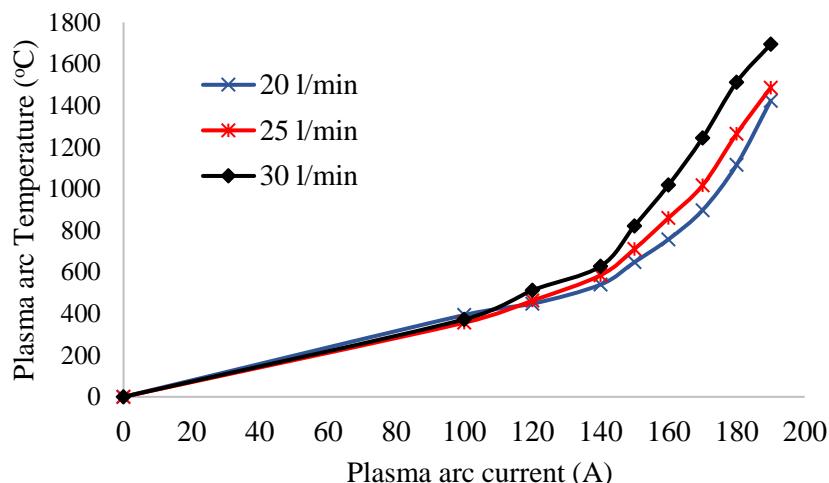


Figure 2. Variation of plasma arc temperature with plasma arc current

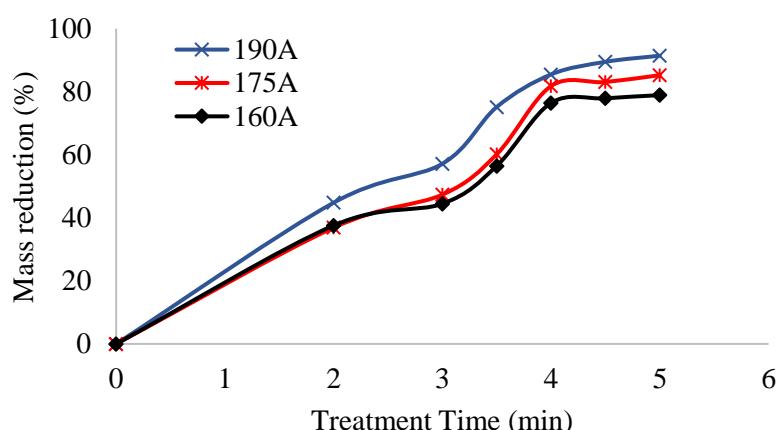


Figure 3. Variation of mass reduction with treatment time and arc current

$$\text{Mass reduction} = \left(\frac{\text{mass of sludge} - \text{mass of slag}}{\text{mass of sludge}} \right) \times 100 \quad (1)$$

3.3 TOC and Carbon Conversion

The total organic carbon (TOC) of both the petroleum oily sludge and the vitreous slag is shown in Table 3. At relatively low arc current (160 A) and short treatment time (2 – 3.5 min) the TOC of the vitreous slag is relatively high (30.08 – 41.15%). Conversely, at higher arc current (175 – 190 A) and longer treatment time (4 – 5 min), the TOC of the vitreous slag is in the range of 3.40 – 14. 15%. Generally, at constant arc current, the TOC of the vitreous slag decreased with an increase in the treatment time. Similarly, at constant treatment time, the TOC decreased with an increased in the plasma arc current.

The TOC of the vitreous slag was compared with that of the untreated oily sludge in the carbon conversion expression shown Equation 2 (Sattar, Leeke, Hornung, & Wood, 2014). The variation of carbon conversion with treatment time is shown in Figure 4. At an arc current of 160 A and a treatment time of 2 – 3.5 min, the carbon conversion is 54.47 – 44.79%. This is an indication of low gasification of the hydrocarbons in the sludge. However, at higher arc current, 175 – 190 A, and longer treatment time, 4 – 5 min, the carbon conversion is in the range of 74 – 94%. This shows that effective gasification is achieved at higher arc current and longer treatment time. The observed behavior could be related to the combined effect of evaporation and gasification. At the early stage of the treatment, 2 – 3.5 min, evaporation of the moisture takes place alongside the gasification of the hydrocarbons, thereby reducing the degree of the gasification. However, when the moisture was removed, the entire thermal energy was available for gasification, thereby, getting higher conversion. In addition, higher arc current generates higher thermal energy thereby enhancing endothermic gasification reactions.

$$\text{Carbon Conversion (\%)} = \left[1 - \frac{\text{TOC of Product slag}}{\text{TOC of oily sludge}} \right] \times 100 \quad (2)$$

Table 3. TOC of the petroleum oily sludge and vitreous slag

Treatment time	TOC (%)		
	160 A	175 A	190 A
2	41.15	36.55	31.92
3.5	32.69	30.08	29.82
4	26.78	14.15	13.17
5	8.87	5.16	3.40
Untreated oily sludge	54.48		

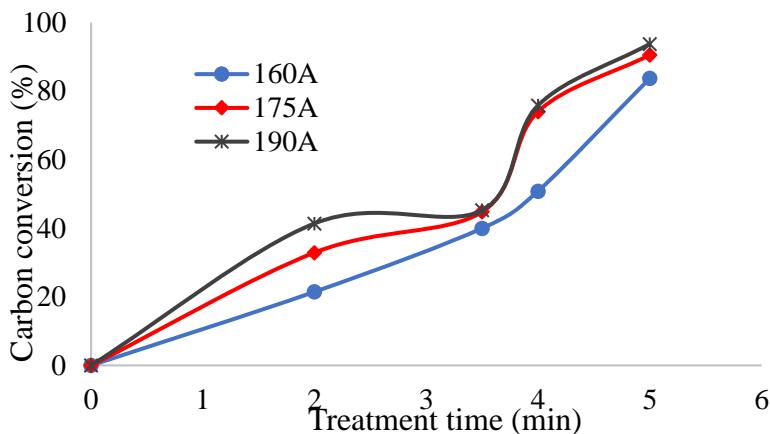


Figure 4. Effect of treatment time on carbon conversion

3.4 Flue Gas Yield

The flue gas yield from the plasma pyrolysis of petroleum oily sludge ranges between 0.78 – 1.61 Nm³/kg of sludge. The yield obtained is higher than the yield obtained from the catalytic pyrolysis of petroleum oily sludge for the production of hydrogen-enriched syngas (Huang et al., 2015). The higher yield obtained in this study could be related to the cracking of a more stable compound at high temperature. The variation of the flue gas yield with treatment time is shown in Figure 5. At the plasma arc current of 160 A, when the treatment time was increased from 2 to 4 min, the flue gas yield increased gradually from 0.78 – 1.1 Nm³/kg of sludge respectively and remained constant thereafter. However, at 175A and 190A, a rapid increase was observed from the 2nd to the 4th min. Increase in residence time provides sufficient time for the gasification reaction to reach completion thereby increasing conversion of the sludge to flue gas.

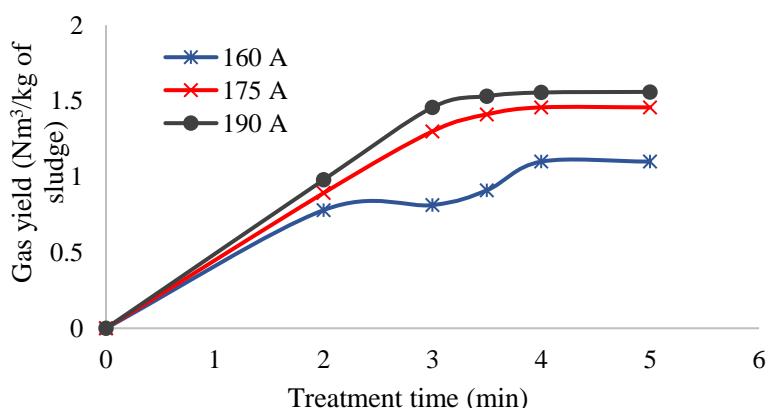


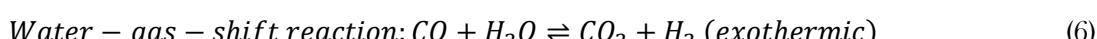
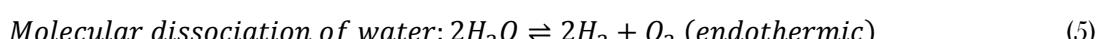
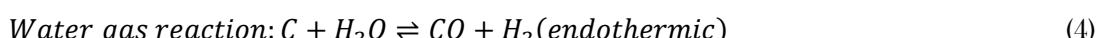
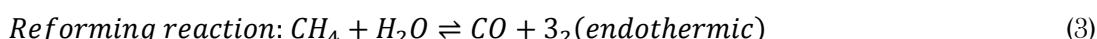
Figure 5. Effect of treatment time on flue gas yield

3.5 Composition of Flue Gas

The composition of the flue gas obtained from thermal plasma pyrolysis of petroleum oily sludge is as follows; H₂ (43.79 – 50.97 mol%), H₂O (26.60 – 30.22 mol%), CO (8.45 – 11.18 mol%), CO₂ (5.12 – 10.35 mol%), CH₄ (2.17 – 3.38 mol%), C₂H₂ (0.86 – 2.69 mol%) and C₂H₄ (0.76 – 2.17 mol%). H₂ and H₂O were the major components in the flue gas followed by the oxides, CO and CO₂. The hydrocarbons, CH₄, C₂H₂ and C₂H₄, were in small concentrations. The high-temperature plasma breaks the molecular bonds in the sludge and gasifies the hydrocarbons into simple molecules such as H₂ and CO. Evaporation at the periphery account for the large concentration of water-vapour in the flue gas. The portion of the oily sludge at the periphery receives less heat when compared with the portion at the centre where the oily sludge is in direct contact with the plasma flame. Thus, evaporation alongside gasification takes place at the periphery.

The variation of flue gas composition with plasma arc current is shown in Figure 6. The concentration of H₂ was observed to increase while that of CO₂ decreased when the plasma arc current increased from 160 – 190 A. This trend is based on the thermodynamics of the gasification reactions involved. Around the plasma flame, H₂ and CO were formed through endothermic gasification reactions such as steam reforming (Equation 3), water-gas reaction (Equation 4) and molecular dissociation of water (Equation 5). An increase in plasma arc current increased the plasma arc temperature thereby, shifting the equilibrium towards the products.

At the periphery, that is outside the plasma core region, CO₂ and H₂ were formed through water-gas-shift reaction (Equation 6), thereby reducing the concentration of CO in the flue gas. The observed behaviour was also reported in the literature (Huang et al., 2015; Motlagh, Klyuev, Surendar, Ibatova, & Maseleno, 2018). The former studied the characteristic behaviour of catalytic pyrolysis of petroleum sludge. They reported an increase in H₂ concentration and a decrease in CH₄ and CO concentrations, in the product, when the pyrolysis temperature was increased. The latter developed a kinetic model for steam gasification of oily sludge and observed that an increase in temperature caused an increase in H₂ concentration with a corresponding decrease in CO concentration.



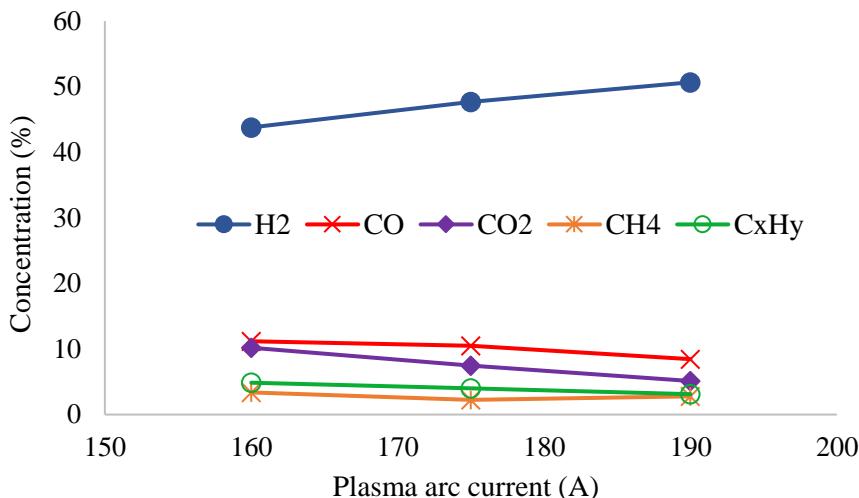


Figure 6. Effect of plasma arc current on flue gas composition

4.0 Conclusions

A 4.7 kW thermal arc plasma reactor was developed using a standard TIG arc welding torch. The transferred arc plasma reactor was used to treat 20 g/batch of petroleum oily sludge. The prevailing temperature inside the reactor ranges between 356 - 1694 °C. The plasma arc temperature increased with increasing plasma arc current and also with increasing plasma gas flow-rate. A vitreous slag and a flue gas were generated as products. A mass reduction of between 36.87 - 91.40% and a TOC reduction of 21.47 - 93.76% were achieved in a treatment time of 2 - 5 min. The mass reduction was observed to increase with increased treatment time. However, the increase was more rapid between the 3rd and the 4th min of the treatment. A flue gas yield of 0.78 - 1.61 Nm³/kg of sludge was obtained, the composition of the gas was H₂ (43.79 - 50.97 mol%), H₂O (26.60 - 30.22 mol%), CO (8.45 - 11.18 mol%), CO₂ (5.12 - 10.35 mol%), CH₄ (2.17 - 3.38 mol%), C₂H₂ (0.86 - 2.69 mol%) and C₂H₄ (0.76 - 2.17 mol%). Thermal plasma reactor is thus, an alternative method of treating petroleum oily sludge.

Recommendation

Further research should consider an upgrade of the reactor system to treat petroleum oily sludge on continuous operation. Since petroleum oily sludge is considered by EPA as a source of polychlorinated dibenzo-para(p)-dioxin and polychlorinated dibenzofurans, further research should consider evaluating these compounds to ascertain the effectiveness of the plasma pyrolysis in removing the compounds.

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