

Optical Characterization of a Microwave Plasma Torch for Hydrogen Production

Babajide O. Ogungbesan, Rajneesh Kumar, and Mohamed Sassi

Abstract—Hydrogen sulfide (H_2S) is a very toxic gas that is produced in very large quantities in the oil and gas industry. It cannot be flared to the atmosphere and Claus process based gas plants are used to recover the sulfur and convert the hydrogen to water.

In this paper, we present optical characterization of an atmospheric pressure microwave plasma torch for H_2S dissociation into hydrogen and sulfur. The torch is operated at 2.45 GHz with power up to 2 kW. Three different gases can simultaneously be injected in the plasma torch. Visual imaging and optical emission spectroscopy are used to characterize the plasma for varying gas flow rates and microwave power. The plasma length, emission spectra and temperature are presented. The obtained experimental results validate our earlier published simulation results of plasma torch.

Keywords—Atmospheric pressure microwave plasma, gas dissociation, optical emission spectroscopy.

I. INTRODUCTION

ATMOSPHERIC pressure plasmas present considerable interest for wide range of environmental, bio-medical and industrial applications [1], [2] such as air pollution control [3], [4], waste water cleaning [5], bio-decontamination [6] and sterilization [7], material and surface treatment [8], [9], electromagnetic wave shielding [10], carbon beneficiation and nano-tube growth [11], [12], element analysis [13], etc. Due to the great applications of thermal plasma, plenty of work has been done using DC, RF and microwave plasma sources. Atmospheric microwave induced plasma (MIP) sources have been studied for decades due to their many advantages. Main advantages of high-power MIPs are electrode less operation, high-throughput atmospheric processing, efficient microwave to plasma coupling and availability of inexpensive source at 0.915 GHz and 2.45 GHz [14]. Because of these advantages, many types of atmospheric MIP sources have been developed. In particular, the Microwave Continuous Flow Reactor (MCFR) [15], [16], Surface Wave Sustained Plasma (SWSP) [17], Torch with Axial Gas Injection (TIA) [18], [19], Microwave Plasma Torch (MPT) [20] and Microwave Cavity Plasma [21], [22] are well-known types of plasma sources. The plasma gas used in plasma torch typically consists of argon, helium, nitrogen or air [23]. However, argon is often used for gasification and

waste treatment in thermal plasmas because it is relatively inert and can be obtained in ultra-pure form at a reasonable cost. It has a relatively low breakdown voltage and tends to minimize electrode wear [24]. To improve the applications of plasma torch, a better understanding of basic plasma parameters is desirable. One of the basic plasma parameters of interest is the gas kinetic or bulk gas temperature. The bulk gas temperature is related to the energy distribution among the heavier particles in the plasma. It can differ between different elements as well as it may be different than the plasma electron temperature. Generally, gas temperatures are referred by the rotational temperatures [26], [27]. However, the bulk gas temperature directly affects the kinetics of the chemical reactions in the plasma torch [27]. Therefore, study of gas temperature in plasma torch is important for hydrogen production, gas dissociation, waste treatment and other applications. Although several methods can be used to determine the gas temperature, optical emission spectroscopy (OES) is a well-used technique because it is simple, can be used *in situ* and is noninvasive [28], [29]. It also provides information on excited atomic and molecular states which enable the determination of the vibrational and electronic excitation temperatures of the plasma. In most of plasma torch applications, optical emission spectroscopy is used to find the molecular or rotational lines for estimation of rotational / gas temperature of nitrogen or air plasma. The rotational lines of the first negative $B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$ system of N_2^+ , particularly those of the (0-0) band have often been used to determine rotational and vibrational temperatures in the plasma torches. The accuracy of the technique is directly related to the number of rotational lines that can be used in global fitting or Boltzmann-plot techniques [30]-[35].

This paper presents the characterization of atmospheric microwave plasma of different gas combinations. A high-power, atmospheric microwave plasma torch is set-up for the waste treatment and hydrogen production from gases especially H_2S , CH_4 , etc. H_2S is a toxic chemical emitted in large quantities from natural sources and industrial processes [37], which cannot be flared to the atmosphere. Therefore simulation study on H_2S decomposition into hydrogen and sulfur has already been reported [36]-[39]. Experiments are carried out to characterize the plasma for getting the suitability for gas treatment (H_2S decomposition). Therefore air-nitrogen, air-air and air-argon gas plasmas are studied. Optical emission spectroscopy is conducted to characterize the plasma while first negative system of N_2^+ is used to estimate the gas temperature of different gas mixtures. Physical and spectroscopic characterization suggests that air-nitrogen

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waveguide, which is called afterglow plasma. By increasing the axial flow rate and microwave power, afterglow plasma can be formed in full length of quartz tube as shown in Fig. 2b. In this experiment, compressed air is used as a swirl flow gas while nitrogen, argon and air is used as the axial flow gas.

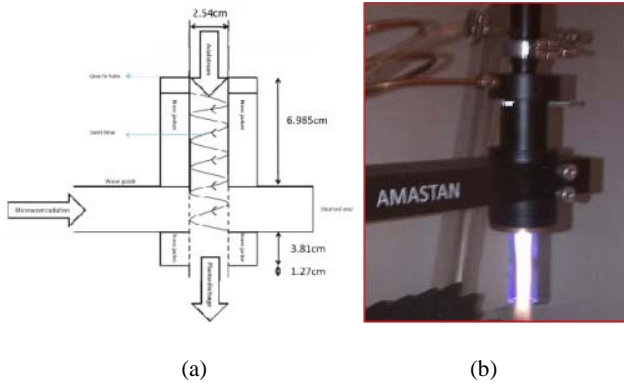


Fig. 2 Plasma formation mechanism

Therefore air-nitrogen, air-air and air-argon plasma are formed. The air-nitrogen plasma was ignited at 1.2 kW microwave power while air-argon plasma was ignited at 800 W at constant axial and swirl flow rates inside the waveguide. Afterglow plasma in the quartz tube can be visualized. It was found that the width of air-argon plasma was more than the air-nitrogen plasma in the afterglow region at constant operating parameters i.e. microwave power, axial flow rate and swirl flow rate. In other words, the swirl gas can efficiently quench the afterglow nitrogen plasma hence it forms in a cone shape unlike with the afterglow argon plasma, which forms in a rectangular shape. The picture of air-nitrogen and air-argon plasma in the waveguide region and afterglow region is shown in Fig. 3 (a) and (b).

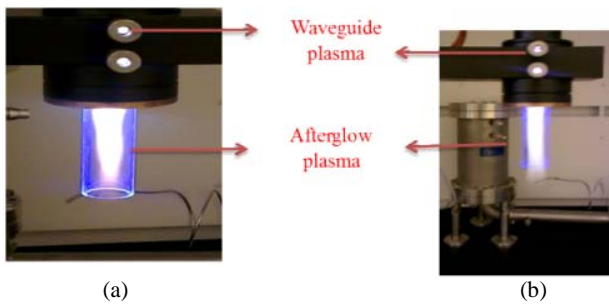


Fig. 3 Pictures of air-nitrogen plasma in (a) and air-argon plasma in (b)

The length of plasma in the quartz tube changes by varying the operating parameters such as the gasflow rates and microwave power. Hence, experiments are conducted with different gases to study the plasma formation with axial flow rate and microwave power. At constant microwave power and swirl flow rate, axial flow rate is varied from 40 l/min to 60 l/min and the length of plasma is measured. The length is measured visually by placing a scale behind the plasma. Measurement are taken from the top of waveguide where the plasma originates to the end of afterglow plasma. Experiments

are repeated at different constant microwave power. Results are shown in Fig. 4. It is suggested that plasma becomes longer when the axial flow rates increases. This is because, it is first created and sustained inside the waveguide, and as the axial flow rate increases, it is blown out farther. The axial flow rate can not be increased indefinitely otherwise the plasma is extinguished. This experiment is conducted with air-nitrogen, air-air and air-argon plasma and similar results are obtained.

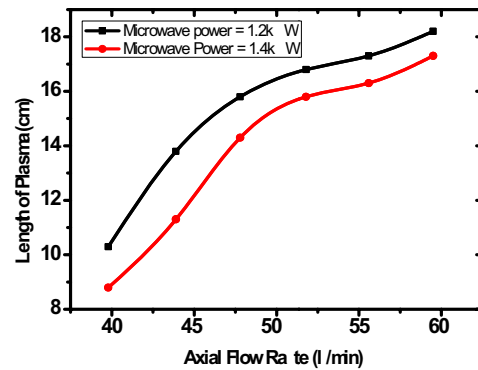


Fig. 4 Length of plasma with axial flow rate at different microwave power in the air-nitrogen plasma

Microwave power has an important effect on plasma formation. Thus, effect of microwave power on the plasma length is studied at constant operating parameters for the axial and swirl gas flow rates. Microwave power is varied from 1.2 kW to 1.6 kW and plasma length is visually measured by using a scale placed behind the plasma. Results are shown in Fig. 5. It is indicated that plasma length can be varied from 13 cm to 17 cm by varying the microwave power from 1.2 kW to 1.6 kW. This experiment is conducted with different constant axial flow rates and different gases like (air-air and air-argon) and similar results are obtained.

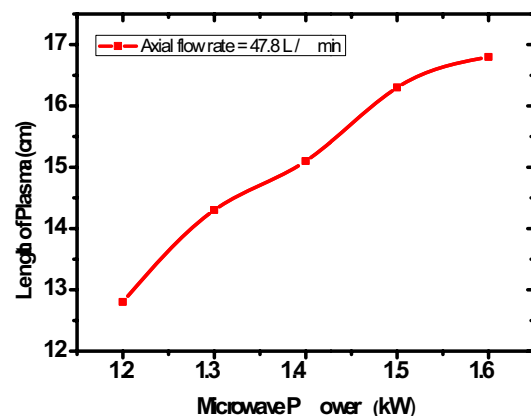


Fig. 5 Length of plasma with microwave power at constant axial flow rate of 47.8 l/min

This study helps to optimize the operating parameters to control the physical properties of different gas plasmas which play an important role during H₂S dissociation in the plasma

reactor.

Spectroscopic diagnostic is needed to understand the chemical kinetics of plasma species in the plasma torch.

B. Optical Emission Spectroscopy of Plasma

Optical emission spectroscopy (OES) based on plasma sources is a well-accepted technique for plasma characterization [29], [34-35]. Therefore, OES is used to characterize air-nitrogen, air-air and air-argon plasma. With the help of the OES results obtained, the gas temperature of different gas combination are estimated and intensity variation with operating parameters are studied, which are described in the sub-sections. A typical spectrum of air-nitrogen plasma in the range of 200 nm to 750 nm is shown in Fig. 6.

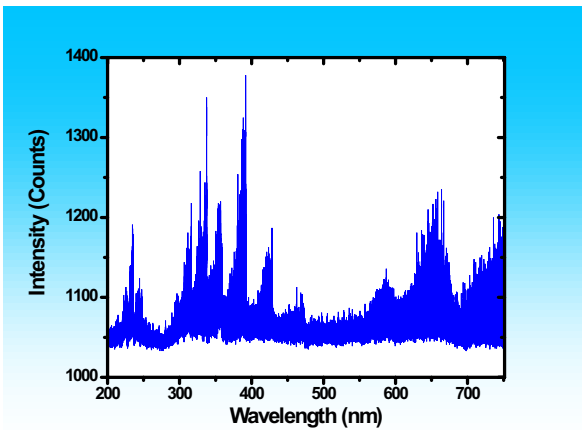


Fig. 6 A typical optical emission spectrum of air-nitrogen plasma in microwave plasma torch

1. Gas Temperature Measurement

Rotational temperature is assumed as gas temperature of air-nitrogen, air-air and air-argon plasma in waveguide region which are estimated by taking the spectrum from 200 nm to 750 nm. For the calculation of rotational temperature of the above three gas combinations, first negative band of $N_2^+(B^2\Sigma_u^+ - X^2\Sigma_g^+)$ have been considered. Strong bands from the (0-0) 391.4nm band head are found to contain an interference-free P-branch which is shown in Fig. 6.

In the P-branch, transitions around $J' = 40$ show a distortion due to perturbed energy levels. Therefore, only transitions up to $J' = 36$ were used for the rotational temperature calculation. The general expression relating the intensities of rotational lines (J', J'') with the rotational temperatures assuming rotational thermodynamic equilibrium is given by [40],

$$-\ln\left(\frac{I_{J'J''}}{S_J v_{J'J''}^4}\right) = a + B'J'(J'+1)\frac{1}{kT_R} \quad (1)$$

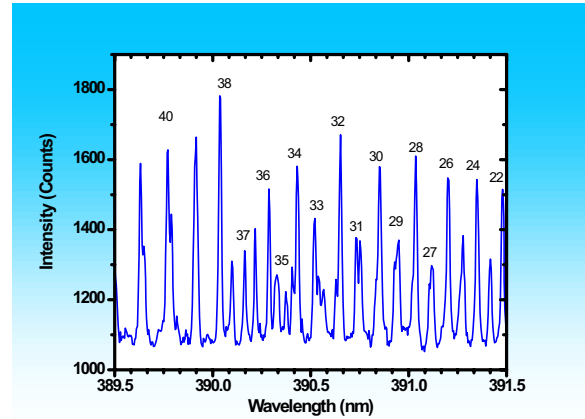


Fig. 7 Spectrum of the N_2^+ , 391.4 nm band head. The P- branch lines used for temperature calculation are labeled

In which $k(JK^{-1})$ is the Boltzmann constant, $T_R(K)$ is the rotational temperature, $B'(J)$ is the rotational constant for the rigid diatomic molecule, $\nu_{J'J''}$ is the frequency of the rotational line, S_J is the line strength which depends on the rotational quantum numbers of the transition levels and $I_{J'J''}$ is the intensity of the transition ($J' - J''$). The line strength S_J for the P branch can be given by [41],

$$S_J = \frac{(J'^2 - 0.25)}{J'} \quad (2)$$

The rotational constant for the upper state is $B'/k = 2.997^0K$ [42]. According to Eq. (1), a linear relationship is expected between the logarithmic term containing the intensity and the rotational energy term $B'J'(J'+1)$. It was also demonstrated that the population of the rotational levels between $J' = 26$ and $J' = 36$ can be described by a Boltzmann function to get a linear relation and rotational temperature can be calculated. It has also been noticed that the linearity of such a plot indicates to what extent a Boltzmann type energy distribution exists for the relevant molecule. Therefore the Boltzmann plots for the N_2^+ molecule (first negative system) in nitrogen-air, air-air and air-argon plasma is plotted. Fig. 8 and Fig. 9 show the Boltzmann plots for air-nitrogen and air-argon plasma respectively.

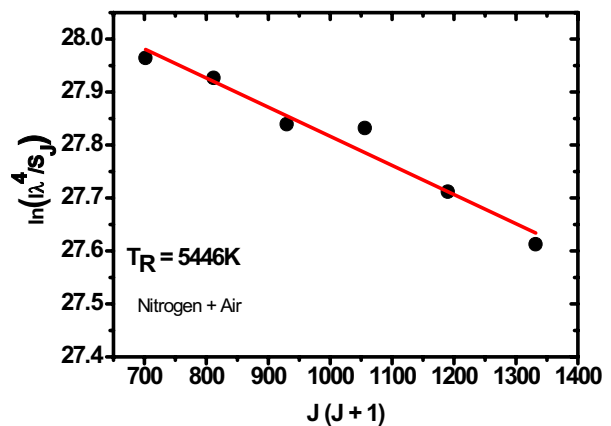


Fig. 8 Boltzmann plot for air-nitrogen plasma

With the help of Eq. (1) and Figs. (8 and 9), the rotational temperature is calculated. Rotational temperature of air-nitrogen, air-argon and air-air plasma in the waveguide region is 5446 °K, 5913 °K and 6100 °K respectively. Rotational temperature is related to the gas temperature hence above mentioned temperatures can be treated as the gas temperatures of different combination of gases. This gas temperature is enough to use this plasma torch for waste treatment and hydrogen production by dissociation of H₂S.

As indicated in the Fig.6, the wavelength range between 200nm and 750nm emission spectrum is recorded. It is observed that spectral line intensity depends on the operating parameters i.e. microwave power, axial flow, radial and axial positions, etc. Therefore experiments are carried out to measure the effect of operating parameters on spectral lines of first negative system ($B^2\Sigma_u - X^2\Sigma_g$), second positive system ($C^3\Pi - B^3\Pi$) and first positive system ($B^3\Pi - A^3\Sigma$) of nitrogen molecule of different gases plasmas in the waveguide region.

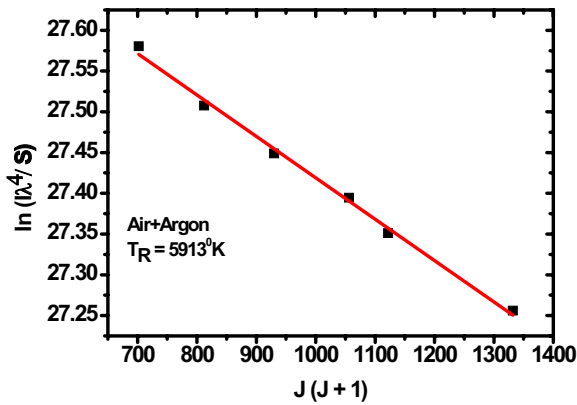


Fig. 9 Boltzmann plot for air-argon plasma

2. Variation with Axial Flow Rate

Intensity of different spectral lines is measured at different axial flow rates of gas at constant microwave power of 1.2 kW. Measurements are shown in Fig. 10. Results indicate that line intensity increases when axial flow rate is increased from 40 l/min to 60 l/min.

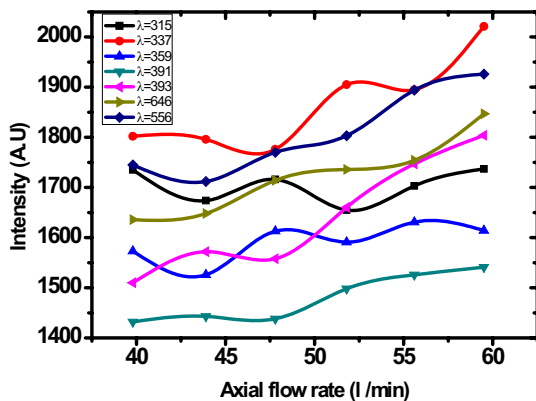


Fig. 10 variation in the intensity of spectral lines with the axial flow rate at constant operating parameters in the waveguide plasma

3. Variation with Microwave Power

Intensity of spectral lines is measured when microwave power is varied from 1.2 kW to 1.6 kW at constant operating parameters as shown in Fig. 11.

It can be concluded from Fig. 10 and Fig. 11 that spectral intensity of waveguide plasma is more enhanced by microwave power than with the axial flow rate. Experiments are also conducted with air-argon and air-air plasma. Obtained results are similar. Therefore plasma characteristics for dissociation of H₂S depend on the microwave power and axial flow rate of a gas.

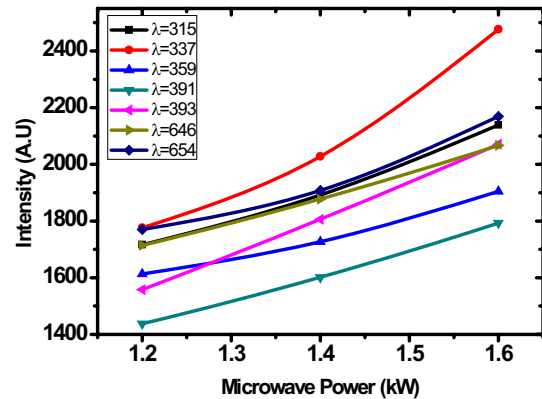


Fig. 11 Variation in the intensity of spectral lines with the microwave power at constant operating parameters

4. Spectral Intensity of Afterglow Plasma with Lateral Positions

In the visual observations, it has been shown in Fig. 3 that afterglow plasma in quartz tube spreads radially and axially in different manners for air-nitrogen and air-argon plasmas. We see an opportunity to support the visual observations by measuring the spectral line intensity at different lateral positions on the radial axis of the afterglow plasma in the quartz tube. Therefore variation in intensity of a spectral line at different lateral positions of afterglow plasma is studied at different operating parameters. This experiment is carried out by fixing the optic fiber receiver to a movable stand. Measurements are recorded from different lateral positions by moving the optic fiber from the center of afterglow plasma ($x=0$) to the inner surface of quartz tube ($x=\pm 12$ mm). A typical lateral profile of intensity is shown in Fig. 12. Maximum intensity is recorded from the central part of the afterglow plasma. Lateral profiles with operating parameters are also studied.

Lateral profiles of intensity are measured at different axial flow rates of gas while other operating parameters are kept constant. It is noticed that the ratio of intensity at center and edge increase as axial flow rates of gas is increased. However, the swirl flow gas also prevents the diffusion of afterglow plasma towards the quartz tube wall. In another set of experiment, intensity of spectral line is measured at different microwave power while the other operating parameters are kept constant. Ratio of intensity at the center and edge increases when microwave power is increased. It can be suggested with these experimental results that the central part (-5 mm $< x < +5$ mm) of the afterglow plasma torch emits the

maximum radiation which means that it contains the highest concentration of plasma species. It is also noticed that central part of afterglow plasma is affected by the axial flow rate, microwave power and gas composition. However, microwave power has a profound effect as compared to the rate of axial flow on the afterglow plasma formation in the quartz tube. This study can help to understand the controlling of afterglow plasma properties with the operating parameters at the time of H_2S dissociation in the plasma reactor.

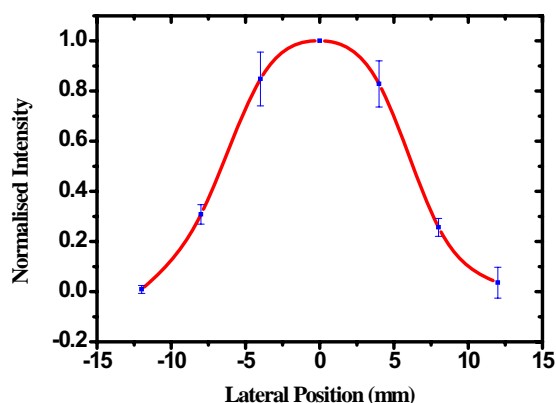


Fig. 12 Lateral profile of intensity across the diameter of afterglow plasma in the quartz tube

IV. CONCLUSION

Atmospheric pressure plasma of air-nitrogen, air-air and air-argon is formed by microwave power of 1.6 kW and 2.45 GHz frequency. The length of plasma depends on axial flow rate and microwave power. Full length of plasma up to 18 cm is achieved by axial flow rate of 60 l / min or by microwave power of 1.6 kW. Interestingly, air-argon afterglow plasma spreads radially more than air-nitrogen afterglow plasma at similar swirl flow rate, axial flow rate and microwave power. First negative system of N_2^+ from the optical emission spectrum of different plasmas is used for the Boltzmann plot. The gas temperature of air-nitrogen, air-air and air-argon plasma in the waveguide region is estimated as 5446 $^{\circ}K$, 6100 $^{\circ}K$ and 5913 $^{\circ}K$ respectively. Spectral intensities of full spectrum are increased by axial flow rate or microwave power. Species concentration is more affected by microwave power than axial flow rate while swirl flow rate is kept constant. Plasma properties make this plasma torch suitable to use in H_2S dissociation into hydrogen and sulfur. However Species density, temperature, velocity and dissociation are under investigation and will be reported in the near future.

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