Optical emission analysis of hybrid air-water discharges

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Abstract : In this paper, hybrid air-water discharges were used to develop an optimal condition for providing a high level of water decomposition for hydrogen yield. Electrical and optical phenomena accompanying the discharges were investigated along with feeding gases, flow rates, and point-to-plane electrode gap distance. The primary focus of this experiment was put on the optical emission of the near UV range, with the energy threshold sufficient for water dissociation and excitation. The $OH(A^{2+},'=0\ X^2,''=0)$ band's optical emission intensity indicated the presence of plasma chemical reactions involving hydrogen formation. In the gaseous atmosphere saturated with water vapor the OH(A-X) band intensity was relatively high compared to the liquid and transient phases although the optical emission strongly depended on the flow rate and type of feeding gas. In the gaseous phase discharge phenomenon for Ar carrier gas transformed into a gliding arc via the flow rate growth. OH(A-X) band's intensity increased according to the flow rate or residence time of He feeding gas. Reciprocal tendency was acquired for N_2 and Ar carrier gases. The peak value of OH(A-X) intensity was observed in the proximity of the water surface, however in the cases of Ar and N_2 with 0.5 SLM flow rate peaks shifted to the region below the water surface. Rotational temperature (T_{rot}) was estimated to be in the range of 900-3600 K, according to the carrier gas and flow rate, which corresponds to the arc-like-streamer discharge

Key Words : optical emission, air-water discharges, plasma chemical, hydrogen production

1. Introduction

Recently,much attention has been devoted to hybrid air-water discharges in order to create efficient methods for sterilizing water [1], forming of UV radiation and shock waves [2], [3]. Another promising application is theproduction of hydrogen from water, instead of reforming of hydrocarbons [4].

2. Experimental setup

The experiments were carried out in a point-to-plane electrode system (see Fig. 1) with a needle type high voltage electrode and plane water surface. Plasma reactor chamber had a volume of 640 cm^3. The input electric power was adjusted with a regulating transformer and measured with a single-phase wattmeter (Model 7013, Hwa Shin Instru., South Korea). The earth electrode was connected to the ground via electrical circuit for measuring the discharge power (R=13 Ω, C=0.22 uF).

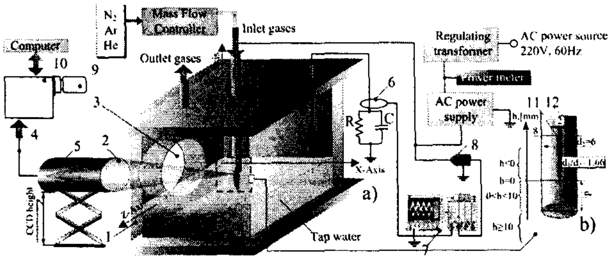

Figure 1. Experimental set up (a) and its detailed schematic drawing (b)

3. Experimental results and discussion

Two characteristic regions were observed in the total optical emission spectrum near UV radiation in the interval 290< λ <380 nm and visible range λ >380 nm, which are shown in Fig. 2. Several plasma chemical reactions including water decomposition in the humid gaseous state were under consideration, [5]:

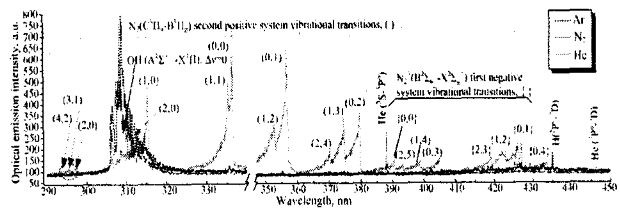

Figure 2. Spectrum for h=13mm and 0,5 SLM: soft UV and visible range. The OES was obtained for Ar, He and N_2 with following plug-in-power values-31, 35 and 42 W

Considered reactions clarify that OH radical monitors relative rate of water decomposition and hydrogen formation (R2, R4, R8-R11). In case of N_2 carrier gas OH(A-X) intensity was low compared to Ar and He. It was assumed that N_2 substitutes following humid air gas phase chemical reactions [6], [7].

Depletion of oxygen atoms and molecules, facilitating hydrogen recombination and electrons attachment, by N_2 environment results in N_xO_y formation. NO and NO_2, in the presence of water are converted to acidic products by reacting with OH radical. Thus, the pH level of water solution in our reactor decreased from 6.25 to 4.09 after discharges. Tap water analysis showed that after discharging concentration of NO_2^- and NO_3^- anions increased in 3 and 5.6 times correspondingly, in comparison with original water's composition.

Q-branch intensity, as the reference peak of OH(A-X) band was measured to investigate the effect of gap distance, flow rate and type of the carrier gas on hydroxyl formation. Flow rate and gap distance influence on OH(A-X)-band intensity are shown in Fig. 3. To investigate distribution of OH(A-X)-band head intensity spatially, CCD camera was mounted onto a vertical positioner, thus adjusting the height level of positioner the intensity of OH(A-X)-band was determined as a function of distance from the water surface. In Fig. 4, peak value of OH(A-X)-band optical emission intensity occurs mostly at the proximity of the water surface. It is partially proved by [8].

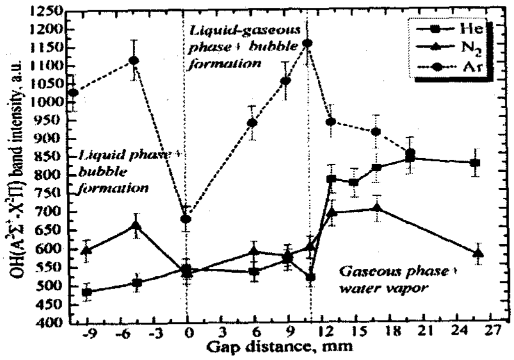

Figure 3. OH(A-X) second positive system band intensity as a function of gap distance (h) for 0.5 SLM flow rate and maximum applied voltage.

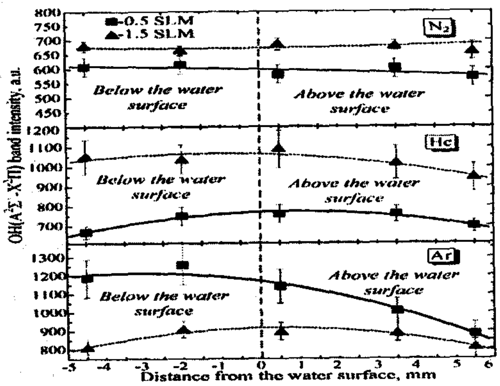

Figure 4. Full fitting curves representing OH(A-X)-band intensity distribution in the axial direction of 17 mm gap distance.

However, in the case of Ar and $N_2$with 0.5 SLM peaks are shifted to the region below the water surface. Apparently, that was caused by weak flow rate, since discharge is not formed as a series of scattered streamers, but as a single plasma channel, capable to penetrate into the water for different depth. To derive plasma gas temperature, by comparing experimental spectra with theoretical, $OH(A^2\Sigma^+ \rightarrow X^2\Pi)$ doublet transition was used. T_{rot} is plotted together with discharge power in accordance with gap distance. $T_{rot}$is changed within the range of 900-3600 K for different carrier gases and flow rate. This range corresponds to the streamer to an arc transition, [9]. T_{vib}/T_{rot}ratio was obtained as the 2~3 factor, which indicated that transient discharge is more inclined to thermal equilibrium, though plasma is still characterized as non-thermal.

4. Conclusions

It was demonstrated that ac hybrid discharge initiate intense UV radiation and chemically active species formation. This type of the discharge causes free radical reactions within and around the sheath of plasma channel. The $OH(A^2X, v'=0 \rightarrow X^2\Pi, v''=0)$ optical emission intensity was measured as a function of applied voltage, gap distance and flow rate of feeding gases. The mechanism for strong OH(A-X)(0,0) emission due to the decomposition of water accompanied with UV radiation was proved. The trend of OH(A-X) emission in respect with gap distance and flow rate was in good agreement with plug-in, discharge power and T_{rot}. For the weak flow rates less than 0.5SLM the highest intensity for OH(A-X) band was measured below the water surface, which indicates that UV radiation can penetrate into the water.

Acknowledgements

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