Atmospheric Pressure Plasma Generation in Contact with Water Column Jet Flow

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Abstract

Aiming for exploring unknown beneficial reaction pathways fixing nitrogen at the plasma - liquid water interface, this work specifically focuses on characterizing the reaction products resulted from the nitrogen plasma generation at the liquid water interface with a pulsed 60 kHz high voltage power source. The developed power source allows localized and intense energy input into the plasma advantageous for nitrogen dissociation rate but can limit the absolute energy input by an intermittent operation. For direct observation on the liquid phase reaction after the plasma exposure, liquid water jet flow is used to make a contact with the intermittent plasma-liquid interaction generated by the 60kHz power source. The plasma exposed liquid, supposed to be concentrated on the liquid, is selectively collected and quickly removed from the unexposed water by a newly developed gas blow liquid sampling. This can potentiate the liquid phase reaction studies. Further discussion on the plasma-liquid interface reaction will be presented.

1. Introduction

In recent years, plasma nitrogen fixation has been proposed as a sustainable nitrogen fixation method [1], which can fix nitrogen exclusively from air. The nitrogen fixation by plasmas powered by electricity has an advantage in accessibility to utilize renewable energy sources. However, in plasma nitrogen fixation, the energy cost to convert nitrogen molecule into storable forms of nitrogen, such as nitrate solution, is high relative to the existing methods [1,2]. In this study, we focus on exploring the nitrogen fixing reaction pathway in an amospheric pressure plasma at the liquid water interface, where full of complexities arise from molecular fluxes and large spatial gradients of the species densities and temperature, dominating resultant reaction processes. This interface reactions potentially involve a lead on the unresolved reaction pathways which may improve its energy cost, but the molecular flux modulation in the atmospheric pressure plasma at the liquid interface can be hardly modulated. This study focuses on development of a discharge plasma source at the gas-liquid interface with controllability in the atomic and molecular fluxes, resulting in modulation in the reaction mechanism. In addition, a liquid sampling system after after the plasma exposure has been develpoed to isolate the plasma exposed liquid from the unexposed liquid within 0.1 sec. These development can accelerate the nitrogen reaction pathway study in the plasma - liquid interaction.

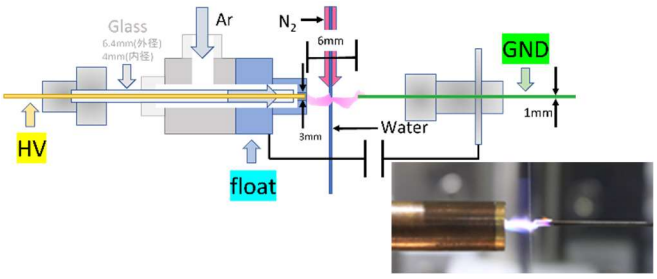

Fig. 1. Schematic of the high-frequency (~60 kHz) atmospheric pressure plasma discharge system and the photo of the discharge response

2. Experiment setup

Discharge frequency at approximately 60 kHz allows specific energy input advantageous for nitrogen dissociation,

thus may improve the atomic nitrogen flux into the liquid surface. The stability of the plasma-liquid interface influenced by surrounding gas flow may also be improved by an ionization mode transition from gamma to alpha mode, depending on the ion drift and electric field, respectively[3,4]. This is especially critical when the gas or liquid flow is given to the plasma liquid interface. Figure 1 shows a schematic of the high-frequency (~60 kHz) atmospheric pressure plasma discharge system and the photo of the discharge. A third capacitively coupled electrode and multiple carrier gas mixtures N_2/Ar are introduced to expand the discharge gap and enable adjustment of the power density. Unlike gliding arcs, which buoyancy force can play important role, the discharge is not arced but directed along the electric field, when the gas and liquid crossed flows are passed through the electrode gap. Figure 2 shows a schematic of the liquid sampling method by a crossed air blow, synchronized with the discharge plasma generation. This developed liquid sampling allows selective measurement of the plasma exposed liquid, locally generated by the intermittent plasma on the liquid. The distance from the plasma discharge to the air blow jet is set 20 cm as in Fig. 2, calculated advection delay is 58.8 ms. The concentration of nitrite NO_2^- is measured after the sampling.

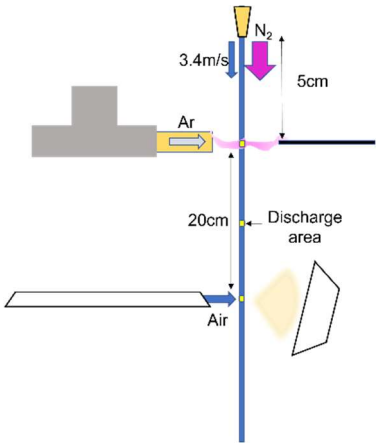

Fig. 2. Schematic of the liquid sampling method by a crossed air blow, synchronized with the discharge plasma generation

3. Results and discussion

The air blow pulse width is set to 20 ms. The measured NO_2^- concentration is shown in Fig. 3 as a function of the delay time of the air blow from the plasma discharge. Figure 3 shows that the peak concentration for the air blow starts at 40 ms and ends at 60 ms, which is roughly in a good agreement with the advection delay time. This demonstrates the localization of the active species in the liquid within the liquid injection flow. Details of the discharge control and the active species in the liquid phase will be presented.

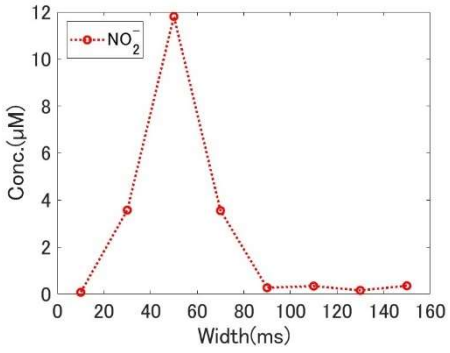

Fig. 3. Delay time of the air blow from the plasma discharge as a function of measured NO2- concentration

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