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Development of plasma-based water treatment technologies based on the analyses of reaction fields by numerical simulations

N. Takeuchi
Department of Electrical and Electronic Engineering, Tokyo Institute of Technology
e-mail (speaker): takeuchi@ee.e.titech.ac.jp

Various types of plasmas in contact with liquid have been applied to water purification. The author and his group has developed plasma-based water treatment technologies for efficient and rapid decomposition of persistent organic compounds in water [1]. Understanding of the reaction fields in gas and liquid phases, with respect to mass transfer through their interface, is necessary to achieve efficient treatments, and a numerical simulation is a strong tool to understand such reaction fields. Numerical analyses of two types of plasmas in contact with liquid, namely, plasma generated within gas bubbles and plasma generated over a solution surface, are introduced in this abstract.

Plasma generated within gas bubbles has been used for ROS generation [2] and water purification using OH radicals [3]. There was a small hole in a ceramic plate and bubbles were produced from the hole by gas supply. Plasma was generated within the bubbles by applying a high voltage to the electrode under the hole with grounding the solution. When plasma was generated in O_2 bubbles, reactive oxygen species (ROS) such as O_3, $\cdot OH$, H_2O_2, and $HO_2\cdot$ were generated. It was found that the amount of produced H_2O_2 increased with increasing the discharge power while that of O_3 decreased. To understand the key reactions which determine the ROS generation, a zero-dimensional (0D) simulation model (global model) considering mass transfer between the gas and liquid phases was constructed [2]. By electron-impact dissociation of O_2 molecules, O radicals were generated. The O radicals were consumed to generate $\cdot OH$ or O_3 in bubbles via a reaction with H_2O or O_2, respectively, as follows: $\cdot O + H_2O \rightarrow 2\cdot OH$ or $\cdot O + O_2 + O_2 \rightarrow O_3 + O_2$. When the water vapor concentration was increased, the calculated H_2O_2 concentration increased, whereas the O_3 concentration drastically decreased. The calculated amounts of H_2O_2 and O_3 exhibited good agreement with experimentally obtained values. Therefore, it was concluded that when the discharge power increases, the vaporization of water is enhanced based on an increased heat flux from the plasma to the solution. Higher reaction rate of $\cdot O$ with H_2O results in higher concentrations of $\cdot OH$ and H_2O_2. In contrast, less vaporization with a lower power results in more reactions between $\cdot O$ and O_2, which generates O_3. When the plasma is driven by AC voltage, the discharge power can be controlled by using ballast capacitors. Lower power input with smaller ballast capacitances achieved a higher generation rate of O_3 [3].

Axisymmetric two-dimensional (2D) model was constructed for plasma generated over a solution surface, which was applied to decomposition of acetic acid [4]. A needle electrode was placed 1 mm above the solution surface with the gas composition of 97% Ar and 3% water vapor. Plasma was generated between the tip of the needle and the solution surface by applying a pulsed voltage. The solution contained acetic acid at a total organic carbon (TOC) concentration of 10 mg/L. The mass transfer of ROS, namely, $\cdot OH$, H_2O_2, and $HO_2\cdot$, through the gas–liquid interface was considered by assuming that a gas–liquid equilibrium is established in accordance with Henry's law and that the fluxes of these species are continuous. $\cdot OH$ in the gas phase was mainly generated via the dissociation of H_2O by metastable Ar atoms. Only a limited portion of the $\cdot OH$ generated in the vicinity of the plasma–solution interface could diffuse into the solution, while the rest was converted into H_2O_2. Figure 1 presents the liquid-phase concentration of $\cdot OH$ along the axisymmetric axis. The concentration near the plasma–solution interface peaked approximately 1 μs after the voltage increase and then decreased to 2% of its maximum value at 10 μs because of the loss reactions with ROS and acetic acid. In the simulated TOC concentration, the reaction rate of $\cdot OH$ with acetic acid was one to two orders of magnitude smaller than those with ROS. Even at a depth of 0.2 μm, the concentration of $\cdot OH$ was less than 1% of the maximum value near the interface. Therefore, the $\cdot OH$ penetration depth is considered to be less than 0.2 μm and the decomposition reactions of acetic acid are like interfacial reactions at the plasma–solution interface.

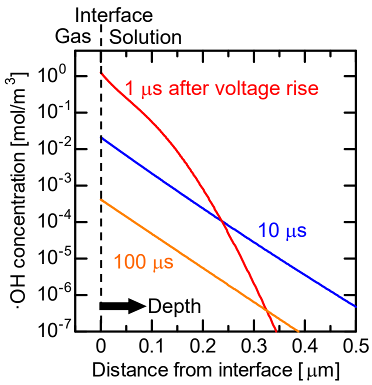

Figure 1. Distribution of the concentration of liquid-phase $\cdot OH$ along the axisymmetric axis [1].

References
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