

大気圧プラズマから液中へ供給された活性化学種の反応拡散シミュレーション Reaction-diffusion simulation of reactive species in liquid exposed to atmospheric-pressure plasmas

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Recently biomedical applications of low-temperature atmospheric-pressure plasmas (APPs) have attracted much interest in the plasma science community. Recent studies have shown that application of APPs directly to living cells and tissues has some therapeutic effects which are caused by reactive oxygen species (ROS) / reactive nitrogen species (RNS). These reactive species generated in the gas phase by APPs do not interact directly with tissues and cells of living bodies. When a living tissue is exposed to a low-temperature APP, chemically reactive species need to be transported through a liquid layer. During this transport process, some of the species may react with others and, therefore, the observation of gas-phase chemically reactive species does not directly convey information on abundant chemically reactive species in the vicinity of the tissue. In this study, both zero-dimensional (global model) chemical reaction simulation and one-dimensional reaction-diffusion simulation have been performed to investigate the concentration distributions of reactive species in water that is exposed to an APP.

The simulation model newly developed in this study is a one-dimensional system of a gas and a liquid that contact each other through a flat gas-liquid interface. The initial condition of the liquid is assumed to be pure water (pH=7) with dissolved oxygen and nitrogen in equilibrium with air at 1 atm. The gas phase species generated by plasma discharge are assumed to be in steady state and to enter water at their thermal velocities. The outward flux of chemical species from water to gas phase is determined from Henry's law. This simulation model incorporates 37 species and 111 chemical reactions in water at room temperature. The governing equations for chemical reactions in the liquid phase are reaction-diffusion equations. If the system consists of N species that are represented by X_i with $i = 1, 2, \dots, N$, the following equations hold;

$$\frac{\partial [X_i]}{\partial t} = D_i \frac{\partial^2 [X_i]}{\partial z^2} + \tilde{R}_i \quad (1)$$

where $[X_i]$ represents the concentration of X_i in water, D_i is the diffusion coefficient of X_i in water, and z is the coordinate representing the depth in water. The

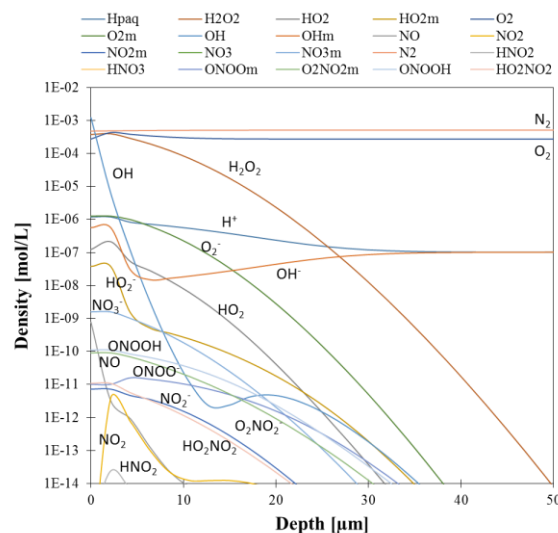


Fig. 1. Concentration distributions of reactive species at $t = 1 \times 10^{-2}$ sec (one-dimensional simulation). The supplied radicals are only OH ($2.5 \times 10^{-5} \text{ mol} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$) and NO ($1.9 \times 10^{-7} \text{ mol} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$).

source \tilde{R}_i represents the chemical reaction term determined from rate equations.

When reactive species such as H_2O_2 , HO_2 , OH , NO , NO_2 , NO_3 , and O_3 dissolve into water from a plasma, the results of the zero-dimensional simulation (the diffusion term of Eq. 1 is ignored) shows that variety of species such as nitrate ion (NO_3^-), peroxyxynitrous acid (ONOOH), and superoxide radical anion (O_2^-) are generated in water and one of the most dominant species is hydrogen peroxide (H_2O_2). It is also found that the solution becomes acidic due to the generation of nitric acid.

Figure 1 shows the concentration distribution of the reactive species obtained from the one-dimensional simulation of the reaction-diffusion equations. The dissolving species are only OH and NO. It is found that OH radicals supplied to water by the plasma are confined to a very thin layer near the gas-liquid boundary due to their high-rate reactions whereas highly stable species such as H_2O_2 diffuse into water.