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Chemical Kinetic Simulation in Aqueous Solution for Plasma-Induced Chemical Processing
プラズマ誘起液中化学プロセスにおける液中化学反応シミュレーション

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Chemical kinetic simulations for more than twenty chemical species inside aqueous solution in contact with plasma source were performed with simple diffusion process to examine anomalous penetration of bactericidal radicals observed in past experiments. Penetration lengths of the radicals are significantly limited to sub-mm or less due to their theoretical short lifetimes while a mm-order penetration was suggested by the experiments. Some effects by external electric field, such as electrophoresis or electro-osmosis flow, should be considered in the assessment of the anomalous penetration because source of ions and radicals is a plasma jet in the atmospheric air attached with the solution, which can supply a large potential fluctuation in the realistic situation.

1. Introduction

Low-temperature plasmas in an atmospheric air have a possibility of new medical technology with less physical stress for a livig body and have vigorously been investigated by many authors to develop an application for soft-materials[1]. In particular, a low-frequency (LF) plasma jet proposed by Kitano et al.[2] is expected to be widely used for plasma process in aqueous solutions from nano-particle synthesis[3] to medical application in a wet-condition because the plasma jet is formed in a quite low temperature and then does not heat nor break the irradiated materials while its chemical reactivity is high enough[4].

In their in-vitro experiments, *E. coli* in low pH-controlled solution was drastically inactivated by a irradiated plasma jet in a short duration less than one minute[5]. This reduced pH method has been applied to dental therapy[6]. After the careful assessment based on an acid dissociation constant (pKa = 4.7), they concluded that superoxide ($O_2^{\bullet -}$) and its conjugate acid, hydroperoxide radical ($HOO^{\bullet}$) should play an important role to inactivate[7] and penetrate the container of the solution whose depth is 10 mm in the exposure duration though their lifetimes are too short to survive in such deep solution.

In the present work, we have numerically modeled a system of chemical kinetics in aqueous solution with some other physical processes in order to estimate a penetration depth of the radicals and clarify important chemical species or exposure conditions for the anomalous penetration.

2. Numerical Modeling

As a first step of our computational work, we have developed a chemical kinetic code with 25 chemical species and selected 66 reactions. A linear diffusion process with species-depended but constant diffusion coefficients was installed in the code for a simplicity. The system of stiff differential equations is solved by the well-known matrix solver, LSODE which was developed and distributed by Lawrence Livermore National Laboratory[8]. Simulations were conducted with several boundary conditions of the input flux of the chemical species, which mimic the conditions of the interface between the air and the solution as the final reactants are reasonably created at the end of the simulations.

3. Result and Discussion

Unfortunately, the key radicals (e.g., $HOO^\bullet$) for inactivation hardly penetrate in the order of mm while the sub-mm penetration is found in a specific condition. Some effects by an external electric field, such as electrophoresis or electro-osmosis flow, should be installed in the assessment of the anomalous penetration because source of ions and radicals is the plasma jet in the atmospheric air attached with the solution, which can supply a large potential fluctuation in the realistic situation. So we are upgrading our code with electric mobility of the species in the solution to include the effects of the fluctuating external electric field.

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