Japanese Journal of Applied Physics **54**, 01AF03 (2015) **REGULAR PAPER**
http://dx.doi.org/10.7567/JJAP.54.01AF03

Computational study of temporal behavior of incident species impinging on a water surface in dielectric barrier discharge for the understanding of plasma–liquid interface

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Received March 6, 2014; accepted June 19, 2014; published online November 26, 2014

A lipid bilayer is a basic structure of the cell membrane and is stable in liquid solution. In this study, we analyzed dielectric barrier discharge (DBD) containing water on a quartz substrate using a one-dimensional fluid model. To simulate atmospheric pressure plasma for practical use, a tiny amount of N_2 gas (0.5 ppm) was added to He gas ambient as an impure gas. The calculated current–voltage (I–V) characteristics reproduced the measured ones qualitatively. We focused on the behavior of DBD at the plasma–liquid interface and analyzed the temporal behavior of the electric field strength and incident fluxes of charged, excited, and radical species on the water surface. By varying the gap length, it was shown that the maximum electric field strength in an AC cycle saturated at gap lengths ≥0.15 cm. The incident fluxes of N_2^+ and He_2^+ on the water surface are almost the same and show strong correlations with the electric field in the vicinity of the water surface.
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1. Introduction

Non-equilibrium atmospheric pressure plasma (NEAPP) has been researched for a wide area of applications since it does not require a high-vacuum pump and is therefore cost-effective. Plasma can serve as a powerful source of free radicals and/or chemically reactive species that arise from atoms and molecules of ambient gas. Of particular interest among reactive species generated by NEAPP are reactive oxygen species (ROS) and reactive nitrogen species (RNS).[1–3)]

Hashizume et al. examined the effect of atomic and excited molecular oxygen species on inactivating *Penicillium digitatum* spores. They used a non-equilibrium atmospheric-pressure radical source, which supplies only neutral radicals, and evaluated the contribution of atomic oxygen and excited molecular oxygen species to the inactivation of *P. digitatum* spores.[4)] Both in vitro and in vivo studies of NEAPP action on cancer were performed. It was shown that cold plasma application selectively eradicates cancer cells in vitro without damaging normal cells and significantly reduces tumor size in vivo.[5,6)] This is explained by the contribution of ROS.[7–9)] Kim et al. reported that a significant overproduction of ROS and a reduction in cell viability are induced by the plasma exposure of cancer cells. Normal cells were observed to be less affected by plasma-mediated ROS, and their viability shows smaller changes than that of cancer cells.[9)]

Dielectric barrier discharge (DBD) can be generated and is stable under atmospheric pressure. DBD is used for blood coagulation and the treatment of mammalian cells.[10)] There are other reports on the development of a DBD setup for biomedical application.[11,12)] To understand the effect of plasma on the cell surface in a buffer solution that forms an interface with plasma, we have studied the irradiation of DBD in Ar or He gas to a lipid bilayer, a basic structure of the cell membrane.[13–16)] We found that pores were formed on a lipid bilayer by the DBD irradiation.[16)]

Murakami et al. analyzed the neutral and ionic systems of a radio-frequency-driven atmospheric-pressure plasma in a helium-oxygen mixture (He–0.5% O_2) with air impurity levels from 0 to 500 ppm with relative humidities from 0 to 100% using a zero-dimensional, time-dependent global model. They reported that air impurity at a few hundred ppm crucially changes the plasma from a simple oxygen-dependent plasma to a complex oxygen–nitrogen–hydrogen plasma.[17–19)] Tani et al. investigated the formation of free radicals in an aqueous solution exposed to different types of non-contact atmospheric-pressure helium plasma using the spin-trapping technique. Both hydroxyl radical (OH•) and superoxide anion radical (O_2^-•) adducts were observed when neutral oxygen gas was additionally supplied to the plasma.[20)] Van Ganes and Bogaerts demonstrated that, in their zero-dimensional, semi-empirical model of an argon plasma jet flowing into humid air, reactive H, N, O, and OH radicals are formed in large quantities after the nozzle exit, and that H_2, $O_2(^1\Delta_g)$, O_3, H_2O_2, NO_2, N_2O, HNO_2, and HNO_3 are predominantly formed as "long-living" species.[21)] These results indicate that ROS and RNS are mostly generated in NEAPP when air and/or humid impurities are present in a plasma source gas. Since plasma for biomedical applications must come into contact with water and living tissues, the generation of ROS and RNS cannot be excluded.

Numerical modeling is a powerful tool for understanding the effects of these reactive species on water and living tissues. The target of such modeling is divided into three phases: (1) the generation of charged, excited, and radical species in plasma containing air impurities and their incidence at the plasma–liquid interface, (2) the chemical reactions of these species with water molecules and the behavior of these species in water, and (3) the interaction between the reactive species and living tissues at the liquid-living tissue interface.

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