Decomposition of Pharmaceuticals and Personal Care Products by Water Plasma

金, 淳浩

（様式２）

氏　　名　：金 淳浩

論 文 名　：Decomposition of Pharmaceuticals and Personal Care Products by Water Plasma
（水プラズマによる医薬品とパーソナルケア製品の分解）

区　　分　：甲

論 文 内 容 の 要 旨

Water plasma has received much attention in a view of the treatment of organic wastes. The water molecule offers higher plasma enthalpy and thermal conductivity than those of commonly used plasma gases such as N_2, O_2, and Ar. This is thus followed by higher heat transfer to the waste material for decomposition. Moreover, a large number of reactive radicals, such as ·H, ·O, and ·OH, as well as high temperatures were provided, by which the radicals accelerate the chemical reactions, produce a large amount of syngas like H_2 and CO, and suppress by-products formation.

Arc characteristics of the water plasma have not, however, been clarified despite the importance of the arc characteristics in thermal plasma torches. Therefore, a study on the fundamental arc fluctuation characteristics including temperature measurement of the water plasma is inevitable prior to applying to waste treatment.

Pharmaceuticals and personal care products (PPCPs) such as medicines, cosmetics, and detergents have been widely used for the purpose of therapeutic and beauty for both humans and veterinary. They, on the other hand, have raised significant concerns about their potential threats to the water system due to continuous discharge through bathing, excretion, and sewage treatment plants. Particularly, N, N-diethyl-m-toluamide (DEET) and Caffeine (CAF), commonly adapted as an insect repellent and psychoactive legal drug worldwide, respectively, have been broadly detected in aquatic environments due to their extensive usage. Moreover, they are hardly biodegraded for biorefractory, persistence, and bioaccumulation. Thus, an improved water treatment technique for the decomposition of PPCPs is very essential. The aims of this dissertation were: (i) to investigate arc fluctuation and temperature of water plasma with mist generation for application to waste treatment; (ii) to decompose DEET and CAF; (iii) to propose the decomposition mechanism of DEET and CAF.

In chapter 1, the introduction of thermal plasmas and the objective of this dissertation were presented.

In chapter 2, arc behavior and temperature distribution in water plasma with mist generation were successfully investigated. The effect of mist feeding rate on the arc was observed by speed cameras synchronized with arc voltage measurement. As the mist feeding rate increased, the arc voltage and length increased due to the higher drag force at constant arc current. The arc fluctuation rapidly increased due to a restrike phenomenon. Moreover, the high temperature area was also increased due to the increase of the arc voltage as well as heat convection. Therefore, it was feasible to control the arc fluctuation and area with a high temperature through a flow parameter. Based on this, the water plasma with a mist generation system is

expected to play a crucial role in the practical application of waste treatment.

In chapter 3, the effect of arc current (6.0, 7.5, and 9.5 A) on the decomposition of 2000 ppm DEET was investigated. The highest decomposition rate of 94.8% was obtained at an arc current of 9.5 A with an energy yield of 0.3 g/kWh within 10 min treatment. The contents of H_2 and CO_2 in the effluent gas increased at a higher arc current due to an enhanced oxidative environment. Moreover, total organic carbon (TOC) reduction rate increased from 69.7 to 91.4% with the increase of arc current. This results from the promoted formation of CO and CO_2 gases through the reactions with O radicals. The hydroxylated DEET was clearly observed in all experimental conditions. Based on seventeen main intermediates, the decomposition mechanism was proposed in detail, where electronic dissociation attacking weak C–N bond and hydroxylation to the ring structure were predominant factors at the early stage of decomposition.

In chapter 4, the decomposition of a high concentration of 20 g/L CAF was investigated according to different arc currents. At torch power of 0.8–1.1 kW, the removal efficiency of TOC and CAF increased with an increase of arc current, reaching 84.5% and 99.8% at 9.5 A, respectively. H_2, CO, CO_2, and N_2 were major effluent gaseous, of which the H_2 generation was more than 40% for all conditions. The nitrogenous compounds were measured as evidence for the reaction pathway of nitrogen and the CAF decomposition mechanism. In particular, the concentration of nitrate (NO_3^-) in the effluent liquids was the highest at 9.5 A, which results from a higher oxidation environment at a higher arc current. The detailed decomposition mechanism was proposed based on eleven intermediate products, in which it was found that electronic dissociation and hydroxylation brought about preliminary decomposition for the ring opening.

In chapter 5, the obtained results and conclusions were summarized and presented. Based on the obtained results through the study of arc characteristics and decomposition mechanism of PPCPs, water plasma is expected to be an alternative green technology for waste treatment. Furthermore, the decomposition of non-soluble organic compounds mixed with surfactant could be suggested for future work.