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Photocatalytic degradation of acesulfame K: Optimization using the Box–Behnken design (BBD)

Seong-Nam Nam^{a,c}, Hyekyung Cho^{b,d}, Jonghun Han^c, Namguk Her^c, Jaekyung Yoon^{b,*}

^a Department of Civil and Environmental Engineering, Chung-Ang University, 84 Heukseok-ro, Dongjak-gu, Seoul 06974, Republic of Korea

^b New and Renewable Energy Research Division, Hydrogen Laboratory, Korea Institute of Energy Research, 71-2 Jang-dong, Yuseong-gu, Daejeon 305-343, Republic of Korea

^c Department of Civil and Environmental Science, Korea Army Academy at Yeongcheon, 495 Hoguk-ro, Yeongcheon-si, Republic of Korea

^d Department of Chemical and Biomolecular Engineering, Yonsei University, 50 Yonsei-ro, Seodaenum-gu, Seoul 03722, Republic of Korea

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ABSTRACT

In this research, photocatalytic degradation of acesulfame K, one of the most popular artificial sweeteners, has been carried out under variations of the initial concentration, pH, concentration of persulfate, and amount of natural organic matter (NOM). The removal efficiencies for 30-min, 60-min and 180-min reaction time have been applied to response surface methodology using the experimental responses obtained by a four-factor-three-level Box–Behnken design (BBD). This provided 29 experimental data for the initial concentration of acesulfame K ranging from 300 to 900 $\mu\text{g/L}$, pH of solution ranging from 4 to 10, persulfate concentration ranging from 0 to 10 mg/L, and amount of natural organic matter (NOM) ranging from 0 to 5 mg/L, which were consecutively coded as A, B, C, and D at three levels (−1, 0, and 1). The analysis of variance (ANOVA) tests with 95% confidence limits determined the significance of independent variables and their interactions consisting of the polynomial regression equation. The optimum values of the selected variables were determined by numerical optimization, and the experimental conditions were found to reach complete mineralization for 30 min and thereafter, at initial concentration of 887.2 $\mu\text{g/L}$; pH of 4; persulfate concentration of 9 mg/L, and NOM concentration of 5 mg/L.

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1. Introduction

Artificial sweeteners, e.g., acesulfame, aspartame, cyclamate, saccharin, and sucralose, have been used as sugar substitutes in considerable amounts in beverages, food, health and dietary products, pharmaceuticals, and animal feed (Kroger et al., 2006). These sweeteners have no or low calories, and are a few hundreds to thousands times stronger than sugar; thus their uses have been substantially increased by commercial industries. Following ingestion, artificial sweeteners are excreted from human bodies mostly unchanged at rates of >90% of the ingested dose

(Mayer and Kemper, 1991), and are discharged into aquatic environments through wastewater treatment plants (WWTPs), because some of these compounds are not efficiently degraded through WWTPs. For example, removal efficiencies of acesulfame and sucralose were reported to be less than 20% (Scheurer et al., 2009; Buerge et al., 2009), consequently, these are detected to ng/L to mg/L levels in wastewater (Loos et al., 2013), surface waters (Muller et al., 2011; Perkola and Sainio, 2014), groundwater (van Stempvoort et al., 2011), and drinking water (Scheurer et al., 2010). Of artificial sweeteners, acesulfame is particularly resistant to degradation in WWTPs; thus, it was found

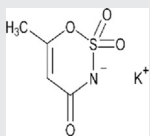
* Corresponding author.

E-mail addresses: gyoon@kier.re.kr, toxicanions@hotmail.com (J. Yoon).

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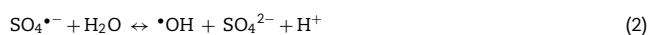
Table 1 – Chemical information of acesulfame K.

	Formula	Molecular weight (g/mol)	Solubility ^a (g/L)	pK _a ^b	Log K _{ow} ^c	Log K _{oc} ^d	Structure
Acesulfame K	C ₄ H ₄ KNO ₄ S	201.24	9.1 × 10 ²	3.02	−1.33	1.302	

^a Based on WSKOW v1.41.
^b Based on ChemAxon.
^c Based on KOWWIN v1.67 estimate.
^d Based on PCKOCWIN v1.66.

at similar to concentrations in a septic tank and a ~15-year old septic plume (Robertson et al., 2013). Due to its persistence in the environment, acesulfame has been used as a suitable wastewater tracer in groundwater (Buergel et al., 2009; Scheurer et al., 2010; Muller et al., 2011; van Stempvoort et al., 2011; Loos et al., 2013; Robertson et al., 2013; Perkola and Sainio, 2014), as well as surface waters (Spoelstra et al. 2013).

Photocatalysis, which is one of the advanced oxidation processes (AOPs), has been extensively applied for degradation of a variety of pharmaceuticals and recalcitrant organic compounds that are known to be less effective to conventional water and wastewater treatment processes (Romero et al., 2011; Dimitrakopoulou et al., 2012; Kanakaraju et al., 2014). Many metal oxides and sulfides such as TiO₂, ZnO, WO₃, ZrO₂, Fe₂O₃, CdS and ZnS have been used for the photocatalytic degradation since those can respond to visible light (Cao et al., 2015; Chong et al., 2010; Baran et al., 2015; Villa et al., 2015). In TiO₂-based photocatalysis, several active oxygen species (e.g., hydroxyl radical, superoxide radical, and hydrogen peroxide) are formed by the reactions with generated electrons and holes. Of them, hydroxyl radical ([•]OH) is frequently considered as the major reactant responsible for the photocatalytic degradation of organic chemicals (Hoffmann et al., 1995). On the other hand, the addition of persulfate ion (S₂O₈²⁻) to the photocatalytic process is reported to significantly increase the removal efficiency (Wang and Hong, 1999; Subramonian and Wu, 2014; Ahmadi et al., 2015). S₂O₈²⁻ can generate the sulfate radical (SO₄^{•-}) in aqueous solution (Eq. (1)) (Kolthoff and Miller, 1951; Waldemer et al., 2007; Matzek and Carter, 2016). SO₄^{•-} reacts with H₂O to produce [•]OH (Eq. (2)) (Hayon et al., 1972; Ghanbari and Moradi, 2017).



Persulfate can also be used as an electron acceptor in the photocatalysis process, producing sulfate radical that has a higher redox potential (2.5–3.1 V) with a longer lifetime compared to [•]OH through the following reaction (Eq. (3)) (Anipsitakis and Dionysiou, 2003).



Therefore, the persulfate is supposed to accelerate the photocatalytic degradation. Assuming that the photocatalytic process is applied to wastewater containing artificial sweeteners, the presence of organic matter would be one of the important parameters to be considered because natural organic matter (NOM) is present in almost every type of water, and plays a role as a hydroxyl radical scavenger, supposing that they are highly reactive with each other ($k_{C,OH^{\bullet}} = 3.8 \pm 1.9 \times 10^8 \text{ M}_C^{-1} \text{ s}^{-1}$ (Arakaki et al., 2013), $k_{NOM,OH^{\bullet}} = 1.02 \pm 0.10 \times 10^8 \text{ M}_C^{-1} \text{ s}^{-1}$ (Donham et al., 2014)).

In assessing the effects of the multivariate results, response surface methodology (RSM) has been proven to be useful for developing the regression model. In photocatalytic process, using RSM, multiple parameters can be optimized by systematic variation of all variables in a well-designed experiment with a minimum, but the required number of experiments.

The present study presents an experimental approach in combination with RSM. Thus, the purpose of this study was to investigate the photocatalytic degradation of acesulfame using the immobilized nano-TiO₂/UV/PS system and to optimize experimental variables using a Box-Behnken design, one of the experimental designs for RSM. Many researches under the similar reaction system to this study have performed to investigate kinetics and mechanisms along with varying parameters (Moradi et al., 2016; Ahn et al., 2017). Thus, based on the understanding of known discoveries, this study does not repeatedly cover similar mechanistic studies, but the optimization of the degradation conditions using the lab-generated immobilized TiO₂ nanotube system.

2. Materials and methods

2.1. Reagents and chemicals

Acesulfame potassium and ammonium persulfate were purchased from Sigma-Aldrich, Korea. The Suwannee River natural organic matter (SRNOM) used as a representative NOM was obtained from the International Humic Substance Society (IHSS, Atlanta, Georgia, USA). Water (HPLC grade) and acetonitrile (HPLC grade) were purchased from J.T. Baker.

2.2. Photocatalysis experiments

Photocatalytic experiments were implemented using the self-rotating photocatalytic system, which is made up of the UV light source, tubular reactor vessel, and self-rotating body (~90 rpm of rotating speed) with four-blade impellers and TiO₂ nanotubes on Ti meshes (each mesh: 2 cm × 5 cm in W:H). The physicochemical properties (e.g., structural morphology, crystalline phase, size, etc.) of anodized TiO₂ nanotubes on Ti meshes as well as its preparation and fabrication processes can be found in our previous paper (Kim et al., 2013).

During the experiments, 1-L of synthesized solution was continuously circulated by 4-blade impeller incorporating with a peristaltic pump to keep the solution homogeneous. Temperature (20 ± 1 °C) and dissolved oxygen concentration (7.6 ± 0.9 mg L⁻¹) were maintained by recirculating water through a closed and double jacketed reactor vessel. The UV irradiation was provided by a 1000 W xenon lamp (Oriel, USA), in which the light emission was above the 300 nm and the irradiated light intensity was measured to be ca. 68 mW cm⁻². More details of the system are well described in the previous paper (Kim et al., 2013), and readers are encouraged to refer it for further information. During the irradiation experiments, 2 mL of samples for acesulfame K were withdrawn every at 30 min time interval for 180 min (Table 1).

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