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Decomposition of Organic Compounds by UV/TiO_2 System†

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The photodecomposition characteristics of EDTA, oxalic acid and ascorbic acid were investigated. Adsorption tests of EDTA, oxalic acid and ascorbic acid on a TiO_2 surface were also performed. For a one component system, the order of decomposition rate chosen from a COD measurement was oxalic acid > EDTA > ascorbic acid. For a two component system, the decomposition rate of a mixed EDTA + oxalic acid solution was fastest among the test solutions. Decomposition of an ascorbic acid + oxalic acid solution was more difficult than the decomposition of an EDTA + ascorbic acid solution. This can be explained by the adsorption characteristics of organic compounds onto the TiO_2 surface. For a three component system, which was a mixed solution of EDTA, oxalic acid and ascorbic acid, more than 95 % of the total organic carbon was decomposed during a 16 h period. The reaction time was reduced due to the successive application of Fenton process and photodecomposition process.

Key Words: Decomposition, Organic compounds, UV/TiO_2.

INTRODUCTION

Decontamination in the nuclear industry means the removal of radionuclides from a contaminated material. Electrochemical and chemical decontamination are known to be effective for the decontamination of the circuit equipment of a nuclear power plant. However, considerable volumes of liquid waste containing undesirable organic waste are generated in these processes[1]. The presence of organic complexing agents such as oxalic acid (OA), ascorbic acid (AA) and ethylenediaminetetraacetic acid (EDTA) complicates any procedure for the separation and concentration of radionuclides. In particular, they can increase the leachability of radionuclides into the environment from final solid wastes and can influence the safety of waste storage and disposal.

Many kinds of decomposition processes have been developed to treat organic compounds that are difficult to decompose. Those processes include a Fenton reaction, ozone treatment, ultrasonic method, radiolysis[2], biological decomposition and photolysis[3]. Hazardous organic solutions are changed into stable and safe inorganic radioactive liquid waste by the application of a decomposition process.

The objective of this study is to investigate the photodecomposition characteristics of surrogate liquid waste generated from a high concentration chemical decontamination process.

EXPERIMENTAL

Ascorbic acid, EDTA and oxalic acid were used as received (Junsei Chemical Co. Ltd.). A BLB fluorescent lamp was used as a light source (General Electric Co. F15T8/BLB, λ_{max} = 365 nm, 15 watt). A TiO_2 photocatalyst was used as received (Nippon Aerosil Co. Ltd. Degussa P25, 75 % anatase, 25 % rutile). COD was measured using a COD reactor (HACH, Model 45600) and DR 4000 UV spectrometer. The test conditions are listed in Table-1.

TABLE-1
EXPERIMENTAL CONDITIONS

	Concentration	TiO_2	Others
EDTA	6.5, 13, 19.5 mM	2 g/L	O_2 bubble
Oxalic aid	12.0, 24.0, 36.0 mM	1, 2, 3, 4 g/L	O_2 bubble
Ascorbic acid	2.9, 5.8, 8.7 mM	2 g/L	O_2 bubble
Mixed solution	[AA]: 3.3, 5.7 mM [EDTA]: 2.0, 13.4 mM [OA]: 10.0. 23.8 mM	2 g/L	O_2 bubble total TOC; 720 ppm

AA: Ascorbic acid, OA: Oxalic acid; TOC = Total organic carbon.

A photodecomposition test reactor is shown in Fig. 1. The inner diameter of the reactor column is 43 mm, while the outer diameter is 47 mm. The length of the cylindrical type photo-

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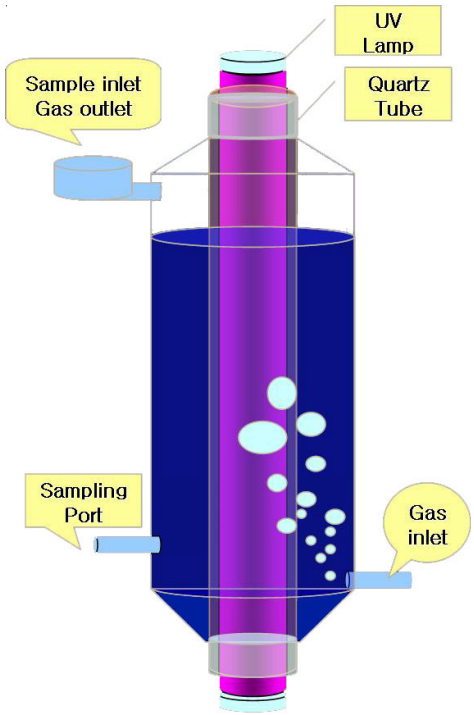

Fig. 1. Photodecomposition test reactor

decomposition test reactor is 300 mm. The UV lamp is surrounded by a quartz tube (inner diameter; 27 mm, outer diameter; 30 mm, length; 400 mm). The reactor volume is 200 mL. For easy dispersion of the photocatalyst and an adequate supply of oxygen, oxygen gas is passed through a gas inlet.

RESULTS AND DISCUSSION

Photodecomposition of organic compounds: Fig. 2 shows the photodecomposition of EDTA against the reaction time. The decomposition rate is increased with a decrease in initial EDTA concentration. After 8 h, the remaining portion of EDTA was 10, 20 and 35 % at 6.5, 13.0 and 19.5 mM EDTA, respectively.

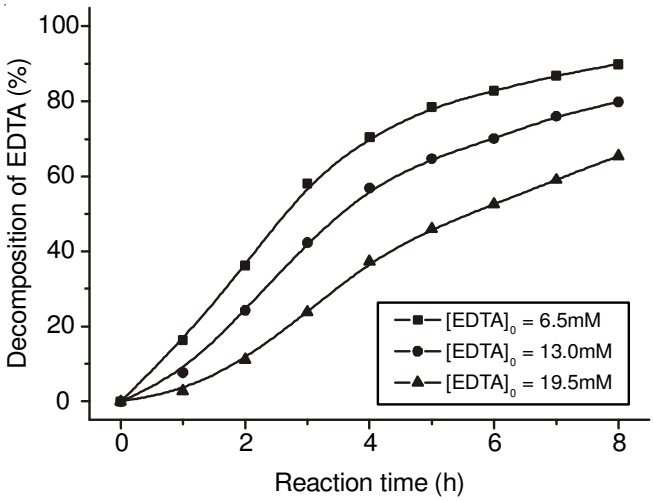

Fig. 2. Photodecomposition of EDTA *versus* reaction time

Fig. 3 shows the photodecomposition of oxalic acid (OA) against the reaction time. Contrary to the EDTA system, OA is decomposed steeply during the initial 2 h. For all the test

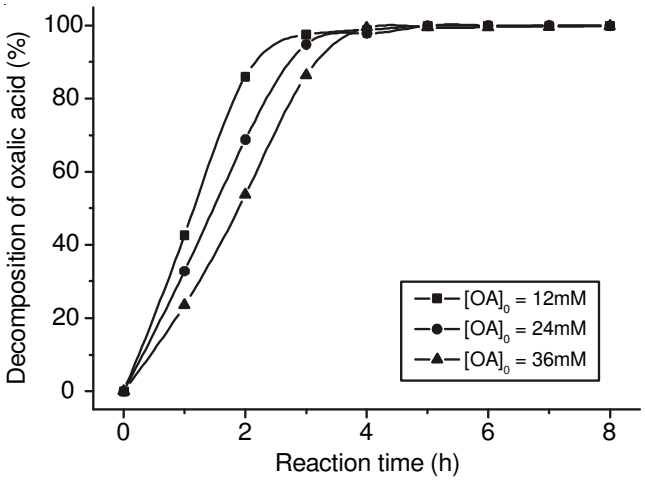

Fig. 3. Photodecomposition of oxalic acid *versus* reaction time

solutions, more than 99 % of the OA was decomposed within 4 h. Considering that the concentration of OA in a decontamination solution is 24.0 mM, it was found that OA itself is easily decomposed within 4 h.

Fig. 4 shows the photodecomposition of ascorbic acid against the reaction time. At 5.8 and 8.7 mM oxalic acid concentrations, the ascorbic acid was barely decomposed. However, the decomposition of ascorbic acid was initiated after 2 h of reaction time at 2.9 mM. More than 99 % of the ascorbic acid was decomposed within 8 h. The adsorption of ascorbic acid onto a TiO_2 surface is a control factor in the photodecomposition of ascorbic acid.

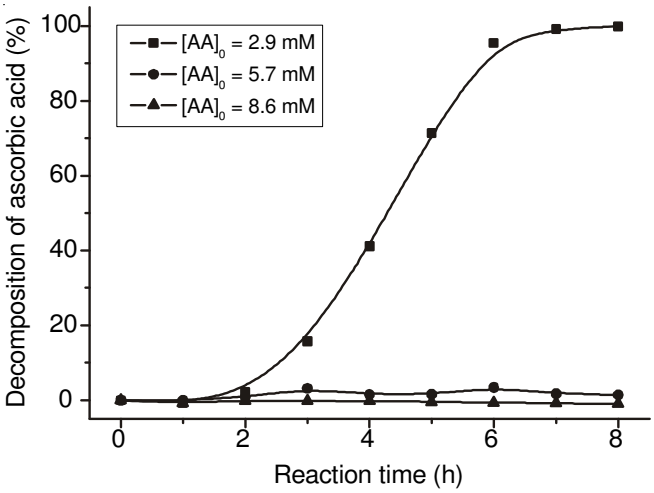

Fig. 4. Photodecomposition of ascorbic acid *versus* reaction time

Fig. 5 shows the photodecomposition of two component systems against the reaction time. The decomposition rate of a mixed EDTA + OA solution is fastest among the test systems. For the EDTA + AA system, decomposition of organic compounds is inhibited by the adsorption of AA at the initial stage. The decomposition of AA on the TiO_2 surface then accelerated the decomposition of EDTA and AA. However, only OA is decomposed in a mixed solution of AA + OA during the 12 h reaction time.

Fig. 6 shows the adsorption characteristics of organic compounds on the TiO_2 surface. The amount of adsorbed organic compounds is linearly increased with the increase of the

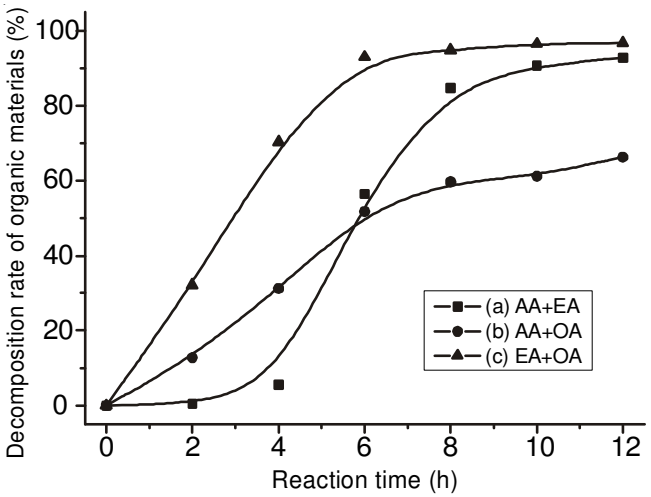

Fig. 5. Photodecomposition of two-component systems *versus* reaction time

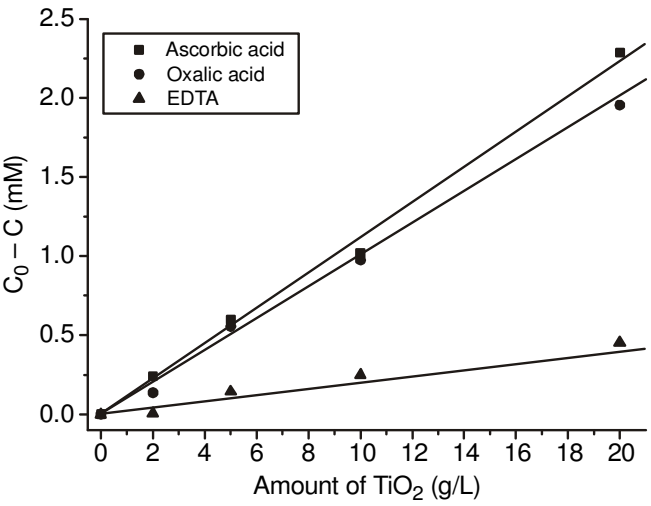

Fig. 6. Adsorption of ascorbic acid, oxalic acid and EDTA *versus* the amount of TiO_2

amount of TiO_2. Ascorbic acid and oxalic acid are easily adsorbed onto the TiO_2 surface. However, only a small amount of EDTA is adsorbed. Comparing Figs. 3 and 4, it was found that adsorbed OA is easily decomposed by UV light. Contrary to OA, adsorbed AA shielded the UV light and inhibited the photodecomposition reaction.

Fig. 7 shows the photodecomposition of three-component systems against the reaction time. The total organic carbon is 720 ppm in the condition described in (a), while (b) shows the real concentration of the decontamination solution. For the system in (a), 95 % of organic compounds was decomposed during the initial 6 h and 99 % was decomposed within 16 h. For the system in (b), 68 % of the organic compounds was decomposed during the initial 6 h and 95 % was decomposed within 16 h.

Fig. 8 shows the effect of the successive application of Fenton and photodecomposition processes on a three component system. Through application of the Fenton process, 80 % of organic compounds was decomposed during the initial 2 h, while 97 % was decomposed within 10 h using the UV/TiO_2 system. However, 85 % of the organic compounds had decomposed within only 10 h using the UV/TiO_2 system.

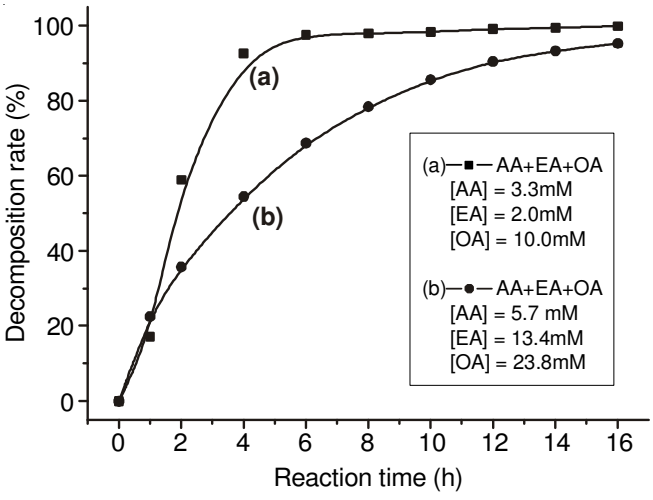

Fig. 7. Photodecomposition of three component systems *versus* reaction time

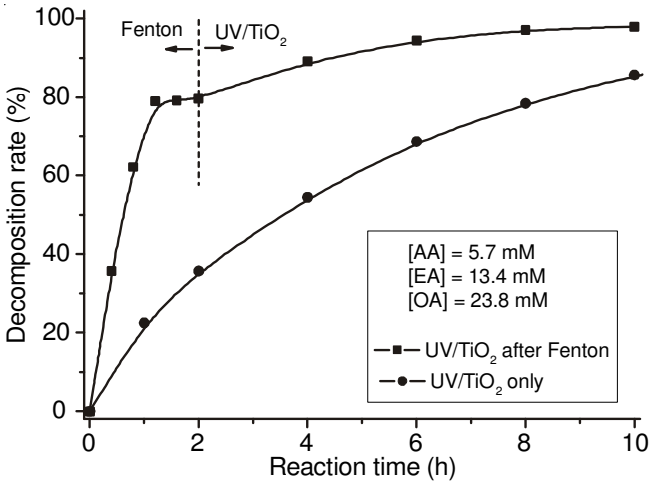

Fig. 8. Successive application of Fenton and photodecomposition processes

Conclusion

To decompose the organic compounds in liquid waste generated from the chemical decontamination of a nuclear facility, a test on a UV/TiO_2 photodecomposition process was performed. The organic compounds tested were easily decomposed using the UV/TiO_2 system. However, it was found that the adsorption of ascorbic acid onto the surface of TiO_2 particulates inhibited the decomposition of organic compounds during the initial stage. To facilitate the photodecomposition reaction, a Fenton process was applied prior to the UV/TiO_2 process. By the application of these two processes, the decomposition time could be reduced and the decomposition rate was significantly increased.

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