

## Purification of the organic impurities contained in the used cyclotron target water using catalytic reaction of dissolved oxygen under UV irradiation

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### 1. Introduction

It is well known that the O-18 enriched target water contains various organic impurities such as acetone, ethanol, methanol, and acetonitrile, etc., after it is irradiated in a cyclotron. These organics may increase the target vapor pressure and decrease the target lifetime as a result [1]. More importantly, they inhibit the separation of generated F ions in the target medium after proton irradiation. For this purpose, UV photo-dissociation process dependent on the catalysts and wavelength has been widely investigated for removing organic materials in an aqueous solution [2,3]. In this investigation, we explored decomposition of the organics under the irradiation of UV wavelengths both 185 nm and 254 nm produced from the low pressure Hg lamp (L-lamp), by observing the concentration changes using the various spectroscopic means such as UV-VIS and GC. Effects of the dissolved oxygen (DO) for dissociation of organics were also evaluated.

### 2. Experimental

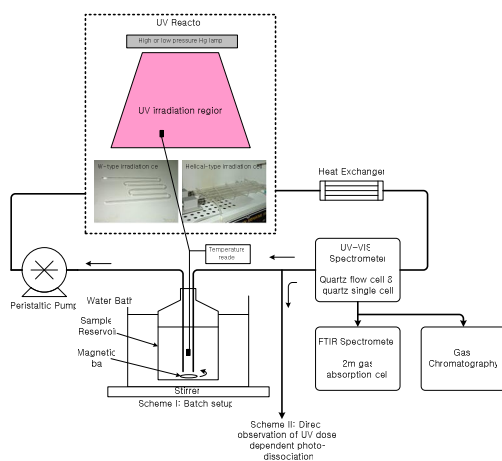


Figure 1. Configuration of experimental system of UV irradiation system and quartz irradiation cells.

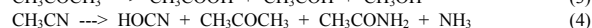
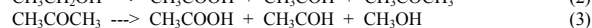
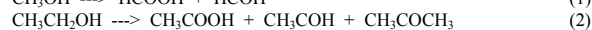
UV irradiation of the organics was performed by the batch setup using a peristaltic pump for circulating the sample at the fixed flow rate as shown in Figure 1. After the sample was irradiated in the UV reactor, it flowed through a quartz flow UV cell installed in the UV-VIS spectrometer (Thermo Spectronic Genesys 2) and was scanned at the certain wavelength ranges. By measuring the UV absorbance of the irradiated sample, it was

possible to estimate the efficiency of the organic removal, and to observe the stable dissociated intermediates produced from UV irradiation. At the same time, pH variance of the UV irradiated sample was measured during the experiment.

### 3. Results and Discussion

#### 3.1 UV Photo-Dissociation of the Organics

Three pathways of dissociation of the organics were assumed as follow; 1) dissociation by reaction with hydroxyl radicals produced from the UV water photolysis under irradiation of 185 nm, 2) direct C-C or C-H bond breakage of the organics by absorbing excess energy from the UV photons with 254 nm, and followed by reaction with hydroxyl radicals produced from the UV water photolysis as the same with 1), and 3) dissociation of the organics or organic fragments by catalytic reaction of DO under irradiation of 185 nm. Estimated reaction fragments under UV irradiation for four organic compounds contained in the water based on the above pathways are as follow;



When the organic compounds are dissociated under UV irradiation, it is assumed that acids and other stable intermediates are generated (Eq. 1~4). If DO exists in the medium, reactive oxygen or ozone might be produced under UV irradiation (Eq. 5~7). It is so reactive as to dissociate the organic substances easily, even more stable acids. Catalytic reaction of DO under UV irradiation was mainly used to remove the organics including those mentioned above and also acetic acid in the next section.

#### 3.2 Removal of the Organics

As shown in Figure 2 (a-i) and (b-i), the acetone concentration was decreased faster with aid of catalytic reaction of DO under L-lamp irradiation (mainly from irradiation of 185 nm) than the irradiation without DO according to UV absorbance changes at 264 nm as the

irradiation time increased. Also the concentration of acetic acid produced as a result of photo-dissociation of acetone was decreased (peak at  $< 220$  nm) faster about 3-fold with DO than that of without DO (Figure 2 (a-ii) and (b-ii)). In fact, acetic acid was not totally removed and acetone was even remained until the irradiation continued to 450 min under the UV irradiation without DO.

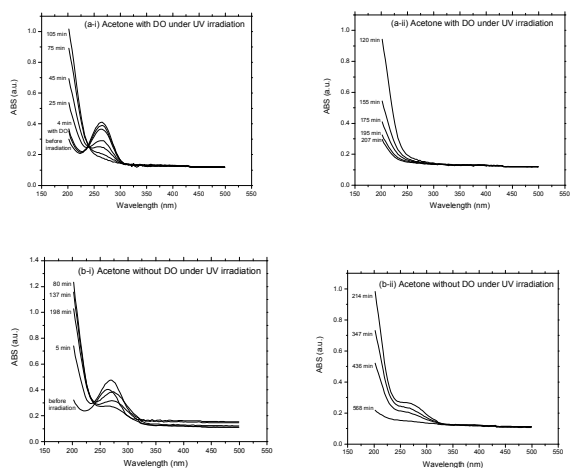


Figure 2. UV-VIS spectrum of acetone removal (a) with and (b) without DO under L-lamp UV irradiation.

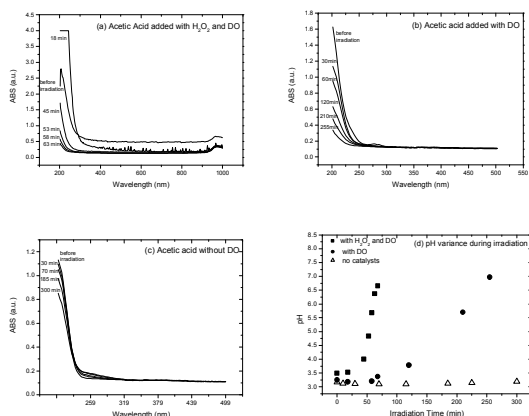


Figure 3. L-lamp UV irradiation of acetic acid ( $\text{CH}_3\text{COOH}$ ) in water with aid of the catalysts (a)  $\text{H}_2\text{O}_2$  and DO, (b) DO only, (c) without catalysts and (d) pH variance during irradiation.

Figure 3 shows the variation of UV spectrum taken during irradiation of acetic acid (a) with aid of both hydrogen peroxide and DO, (b) with aid of DO alone, and (c) without aid of catalysts. Figure 3(d) also shows the variance of pH during irradiation. Acetic acid was removed completely after 1 hr of irradiation according to the absorbance at  $< 250$  nm with hydrogen peroxide and DO. It took almost 4 hours to remove acetic acid when DO alone is added in the medium while acetic acid was not removed at all without aid of any catalytic reaction. These were matched well with the pH changes shown in Figure 3(d).

### 3.3 Overall Removal Efficiencies

Overall efficiency of the purification scheme was evaluated by batch setup L-lamp UV irradiation. Organic impurities of ethanol, methanol, acetone, and acetonitrile contained in the deionized water with a volumetric concentration of 0.1% were prepared. The experiments were performed with and without DO respectively until the organics were removed completely. As shown in Figure 4, organic impurities could be removed entirely within 2 hours for acetone, and within 3 hours for methanol and ethanol, and 5 hours for acetonitrile with aid of a catalytic reaction of DO under UV irradiation, while it took even more than 20 hours for a removal of acetonitrile when DO was not presented in the sample.

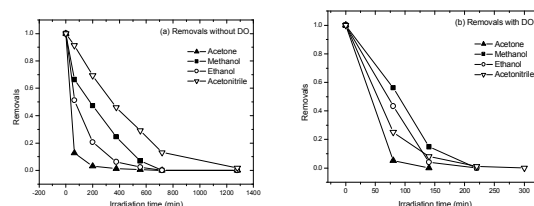


Figure 4. Removal of the organic impurities under L-lamp UV irradiation (a) with aid of DO and (b) without aid of DO.

## 4. Conclusion

Photo-dissociation with aid of the catalytic reaction of DO under the L-lamp UV irradiation was the most efficient, and expedite the removal of the organic impurities contained in the water faster about 4 times when compared to the case without aid of DO under the same conditions. Also, acids produced from UV irradiation of the organics were not removed easily without aid of catalytic reaction of DO or hydrogen peroxide. Consequently, DO presented in an aqueous solution plays an important role for the photo-dissociation process during the UV irradiation without producing any unwanted by-product.

## REFERENCES

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