Mineralization of Oxalic Acid by Ni(II) With Gamma Radiation

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1. Introduction

Various organic solvents are used in many countries' nuclear industries to eliminate the metal scales produced in cooling pumps, piping, etc [1]. Organic acids such as oxalic acid, citric acid, and formic acid are commonly used, and various studies are being conducted to remove organic acids that advanced oxidation processes (AOPs) [2]. Highly reactive species are generated in water radiolysis using gamma radiation. Then, hydroxyl radical (•OH) and sulfate radical (SO₄*-) can formed by the Ni(II) and potassium persulfate (PDS). The objectives of this study is to formed hydroxyl radical and sulfate radical by combining Ni(II)/PDS with gamma radiation.

2. Materials and methods

2.1 Materials and sample preparation

Oxalic acid, NiSO₄·6H₂O, K₂S₂O₈ (PDS) were purchased from Sigma-Aldrich. Oxalic acid solution of 2 mM was irradiated for effect of Ni(II):PDS on oxalic acid degradation. Fixing the initial Ni(II) concentration at 1 mM and varying the PDS concentration to obtain molar ratio of 1:0, 1:1, 1:2 and 1:5.

2.2 irradiation sources

The samples were irradiated at the absorbed doses

from 5 to 50 kGy and dose rate of 10 kGy/hr. Gamma radiation was achieved using a ⁶⁰CO source (Nordion Inc., Canada) at the Korea Atomic Energy Research Institute (KAERI, Korea).

2.3 Analysis methods

Total organic carbon (TOC) for mineralization was determined using a TOC-VCSN TOC analyzer (Shimazu, Kyoto, Japan). A digital pH meter was used for pH measurement (Orion, Singapore).

3. Result and discussion

3.1 Effect of Ni(II) on the mineralization of oxalic acid

Fig. 1 shows that the effect of various Ni(II) on the degradation of oxalic acid by gamma radiation. The degradation efficiency of oxalic acid was increased with the addition of Ni(II), and was decreased when excessive concentration of Ni(II) was added. The decreased efficiency with the addition of Ni(II) can be explained by the following equation:

$$Ni(II) + \bullet OH \rightarrow Ni(III) + OH$$
 (1)

Excess concentration of Ni(II) occur due to the hydroxyl radical scavengers.

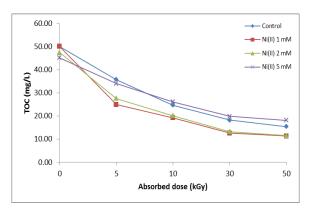


Fig. 1. Effect of different Ni(II) concentration on oxalic acid ($C_0 = 2$ mM) degradation using gamma radiation.

3.2 Effect of Ni(II) and PDS on the mineralization of oxalic acid

Fig. 2 shows the degradation of oxalic acid at different molar ratios of Ni(II) to PDS using gamma radiation. The degradation efficiency increased with an increase in PDS concentration. When 5 kGy of an absorbed dose, TOC of oxalic acid was removed about 88% at the Ni(II):PDS = 1:5 molar ratio. The initial TOC of oxalic acid was decreased before gamma radiation.

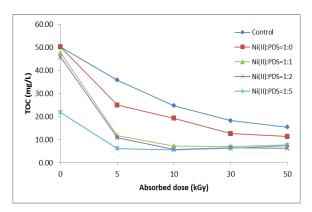


Fig. 2. Effect of 1 mM Ni(II) and various PDS concentration from 1 to 5 mM on oxalic acid ($C_0 = 2$ mM) degradation using gamma radiation.

The reason is considered to be an oxidation reaction by the Fenton-like reaction. The hydroxyl radical and sulfate radical were formed by a reaction of Ni(II) and PDS. Oxalic acid is removed by the effect of hydroxyl radical and sulfate radical. But, excess PDS addition resulted in a decrease of degradation efficiency.

4. Conclusion

The mineralization of oxalic acid was investigated combining Ni(II)/PDS with gamma radiation. The this study, the Ni(II)/PDS/gamma radiation process shows a high dependency on the concentration of Ni(II) and PDS. Ni(II)/PDS/gamma radiation process is more effective than the gamma radiation or Ni(II)/gamma radiation process.

REFERENCES

- [1] Enda, Masami, et al. "System and method for chemical decontamination of radioactive material." U.S. Patent No. 7,087,120. 8 Aug. 2006.
- [2] Thomas, Daniel A., et al. "Real-time studies of iron oxalate-mediated oxidation of glycolaldehyde as a model for photochemical of aqueous tropospheric Environmental science & technology 50.22 (2016): 12241-12249.