Research Article

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The degradation, biodegradability and toxicity evaluation of sulfamethazine antibiotics by gamma radiation

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Abstract: This study evaluated gamma radiation-enhanced sulfamethazine (SMT) degradation, which belongs to the heterocyclic sulfonamides, in different conditions in aqueous solution. The results showed that gamma irradiation could increase the SMT removal in aqueous solution, and the SMT degradation kinetic follow the modified pseudo-first-order kinetic. The degradation rate was nearly 95% at the absorbed dose of 4 kGy. The removal of total organic carbon (TOC) and total nitrogen (TN) could be also induced. When the radiation dose was 5 kGy, the removal rates were only about 6.8% and 10.5% for TOC and TN, respectively. Some ions including HCOO^- , CH_3COO^- and SO_4^{2-} released during SMT degradation were studied. The biodegradability and toxicity of intermediate products of the SMT degradation were also proposed in this experiment. The biodegradability of the SMT can be improved by the intermediate products of SMT degradation at the radiation dose of 1, 2 and 5 kGy. Gamma radiation could be used as a pretreatment technology before the biodegradation process of pharmaceutical waste water.

Keywords: sulfamethazine (SMT), gamma radiation, degradation, biodegradability, toxicity

1 Introduction

Antibiotics are one of the greatest inventions in the field of medicine in the world. They are widely used for curing human and animal diseases. They are also used in aquaculture and animal husbandry as an additive [1–3]. With the development of human society, drug abuse has become a serious threat to the safety of human beings and other organisms [4,5]. 70% of drugs produced are antibiotics every year in China [6]. Overproduced and used antibiotics enter natural water bodies through biological excretion, sewage discharge, surface runoff and soil infiltration, posing a serious threat to drinking water and endangering the environment, including human health and life safety [7–11].

Sulfonamides are mainly derived from sewage treatment systems, human production activities (agriculture, animal husbandry and aquaculture), hospitals and pharmaceutical plants [12–14]. Due to the special hazard of sulfonamides, related treatment technologies have gradually attracted attentions all over the world in recent years. Sulfamethazine (SMT) is an important veterinary drug. Its chemical structure is shown in Figure 1. It has been extensively used in fisheries and animal husbandry and has seriously threatened the health of water bodies. SMT is a kind of synthetic broad spectrum antibiotics, which can inhibit both Gram-positive and Gram-negative bacteria, and they are used especially for inhibition of Gram-positive bacteria. As the structure of SMT is similar to that of the *p*-aminobenzoic acid (the essential material for the synthesis of dihydrofolate), SMT can compete with *p*-aminobenzoic acid present in bacteria to block the synthesis of dihydrofolate, thereby inhibiting the synthesis of nucleic acid and the growth of bacteria.

The main treatment methods for SMT are physical methods, chemical methods and biological methods. The biological method is the most conventional method for waste water treatment. Antibiotic-resistant bacteria and

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Figure 1: The chemical structure of SMT.

antibiotic-resistant genes in microorganisms during biological treatment process lead to the formation of super bacteria, resulting in gene pollution [15–19]. The physical method is usually used for the primary treatment of waste water to achieve the preliminary removal of pollutants, but the pollutants will not be completely degraded. The chemical method is a simple and effective treatment method, which is used in the treatment of antibiotics frequently. Advanced oxidation process, a key technology of chemical oxidation, has obtained more attention of researchers [20]. As an emerging advanced oxidation technology, ionization radiation not only has the advantages of the advanced oxidation technology but also makes up for its disadvantages. Gamma irradiation appears to be more powerful, effective and safe [21,22]. In water radiolysis, three active species, hydroxyl radicals ($\text{OH}\cdot$), hydrated electrons (e_{aq}^-) and hydrogen atoms ($\text{H}\cdot$), play an important role in the degradation process. In the process of water treatment, ionizing radiation may trigger a variety of changes including chemical effects (e.g., decomposition and polymerization of contaminants), physical and chemical effects (e.g., colloidal denaturation) and biological effects (e.g., disinfection and sterilization). Irradiation technology has been used to treat a series of environmental problems such as dechlorination and dye degradation [23,24], sludge treatment [25], organic pollutant oxidation [26,27], insecticide removal [28] and drug degradation [29]. Irradiation technology is widely used in the purification of drinking water, surface water, groundwater, municipal sewage and industrial waste water [30].

This study focuses on the SMT degradation effect of gamma irradiation in aqueous solution. The biodegradability and microbial toxicity by intermediate products during the degradation were also studied. The main objectives are as follows: (1) to investigate the degradation effect and kinetics of SMT at different absorbed doses; (2) to identify some organic acids and ions in the process; (3) to evaluate the biodegradability and microbial toxicity of radiolytic degradation of SMT in aqueous solutions.

2 Materials and methods

2.1 Reagents and preparation

SMT (A.R. grade, >99%), KOH (20%, w/v), NaCl (0.9%, w/v), K_2HPO_4 , $\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$, NaOH (20%, w/v), $\text{K}_2\text{S}_2\text{O}_8$ (4%, w/v) and KNO_3 were obtained from Beijing Chemical Factory. Phosphate buffer used for specific oxygen uptake rate (SOUR) experiments was prepared by adding 3.4 g K_2HPO_4 and 8.95 g $\text{Na}_2\text{HPO}_4 \cdot 12\text{H}_2\text{O}$ (3.55 g Na_2HPO_4) to 1,000 mL deionized water. Initial SMT solution concentration was 20 mg/L. The initial pH of the solutions was around 6.5 without adjustment. All the samples were filtered with 0.22 μm syringe microfilters (nylon 6) before being analyzed. The experiments in this study were performed at the ambient temperature of about 25°C.

2.2 Gamma irradiation

^{60}Co radiation facility used in the experiments was built in the Institute of Nuclear and New Energy Technology, Tsinghua University. The dose rate used in the gamma radiolytic experiments was 103 Gy/min. The radiation dosimetry was carried out by using a standard Fricke dosimeter. Radiation doses used in the experiment were 1, 2, 3, 4 and 5 kGy. The sample without radiation was used as a negative control. All the experiments in this study were performed at the ambient temperature under open air.

2.3 Chemical analysis

The concentration of SMT solutions was determined by the HPLC with diode array detector. The ions were detected by the Ion chromatograph (Dionex model ICS 2100) in this study, which was coupled with a Dionex IonPac AS19 analytical column (4×250 mm) and an IonPac AG19 guard column (4×50 mm). The column was maintained at 30°C. The injection volume was 10 μL . A mobile phase of methanol/water (35:65, v/v) was used. The flow rate of the mobile phase was 1.0 mL/min. The detection wavelength of SMT was 275 nm, and the retention time of 1.97 min.

The TOC was determined by a Shimadzu TOC-VCPH TOC analyzer (Shimadzu, Japan). The persulfate method was used to measure the total nitrogen (TN) [31].

2.4 Biodegradability and toxicity assessment

The activated sludge was obtained from Zijiang Apartment Reclaimed Water Plant, Tsinghua University. The activated sludge was aerated for 24 h to use all the organics in the system, and this process is called endogenous respiration (ER). Then, the sludge was washed with normal saline and centrifuged for purification for 10 min at 3,000 rpm. Then, the sludge was added into the phosphate buffer with the pH of 7.0. The final mixed liquid suspended solids (MLSS) was 20 g/L.

The biodegradability of the radiolytic solution was detected by the microrespirometer (skw-3, Shanghai University), which was used to determine SOUR. There were two flasks used as contrasts, and another two were set to test the ER. The other flasks were used to test the biodegradability of the radiolytic samples. All the experiments were conducted in conical flasks placed in the water at the ambient temperature.

Ethical approval: This research is not related to either human or animal use.

3 Results and discussion

3.1 Degradation and mineralization of SMT

Some free radicals and ions produced during the radiolysis of water were presented as follows [32]:

G values of the particles are presented in the brackets, indicating the number of one certain specie set per 100 eV. There are three reactive species with higher G value, including hydroxyl radicals ($\cdot\text{OH}$), hydrated electrons (e_{aq}^-) and hydrogen atoms ($\cdot\text{H}$). They play an important role in the radiation process for pharmaceuticals degradation in water.

Figures 2 and 3 show gamma radiation-induced SMT degradation in aqueous solutions with the increasing absorbed dose. The initial concentration of SMT was 20 mg/L. The radiation doses used in the experiment were 1, 2, 3, 4 and 5 kGy. One sample without radiation was used as a negative control.

Figure 2: Gamma radiation-induced degradation of SMT in aqueous solution (C : SMT concentration; C_0 : SMT initial concentration).

The results showed that the concentration of SMT was decreased rapidly with the increasing radiation dose. According to the results of pseudo-first-order kinetic parameters, R^2 values were more than 0.99. The SMT degradation kinetic follows the modified pseudo-first-order kinetic. The degradation rate of SMT was about 95% at the radiation dose of 5 kGy, which was the highest dose in this study. But the removal of TOC and TN was not that obvious. When the radiation dose was 5 kGy, the removal rates were about 6.8% and 10.5%, respectively. These indicated that nearly all the SMT molecules were degraded to some organic intermediates in the solutions without being mineralized in the SMT degradation process. In this stage, the reactive species produced by gamma radiation were mainly used to react with only SMT molecules and byproducts, and few of them were converted to carbon dioxide. These intermediate products were in competition with SMT molecules and reacted with reactive species produced by gamma radiation. The reaction probabilities between SMT molecules and reactive species decreased, and the rate of removal of SMT decreased thereafter. But the degradation of SMT molecules was the main reaction during the process. At high radiation dose, SMT molecules could be degraded almost completely to byproducts, resulting in a decrease in the removal rates of TOC and TN, and hence, the mineralization of SMT cannot be achieved.

3.2 Analysis of the radiolytic products

There were three compounds identified in the radiolytic solutions, including SO_4^{2-} , HCOOH and CH_3COOH .

Figure 3: TOC and TN removal at different absorbed doses (SMT initial concentration: 20 mg/L; dose rate: 103 Gy/min). (a) TOC and (b) TN.

Inorganic ions containing nitrogen, such as NO_2^- , NO_3^- and NH_4^+ , were not detected under any conditions in this study. Figure 4 shows the change in pH value and the formation of SO_4^{2-} , HCOOH and CH_3COOH with the increasing radiation dose. The results showed that the pH value decreased in the reaction process with the increasing radiation dose, and the solution was changed to acidic from partial neutral. The initial pH value of the SMT solution before reaction was about 6.4, and the final pH value was about 5.75 after irradiation. The change in pH value can be confirmed by the formation of SO_4^{2-} and two acids.

With the increase in the radiation dose, there was an increase in the SO_4^{2-} concentration, and the a maximum concentration was about 3.2 mg/L at 5 kGy, which was the highest radiation dose in the experiments. At higher radiation dose, more $\cdot\text{OH}$ radicals were produced in radiolytic solution. There were more SMT molecules oxidized by $\cdot\text{OH}$ radicals in the degradation process, resulting in more SO_4^{2-} ions released. But the quantity of the SO_4^{2-} could be limited by the initial SMT concentrations. Not all the SO_4^{2-} was released in this study at a radiation dose of 5 kGy. The formation trend of CH_3COOH was similar to that of SO_4^{2-} . At the radiation dose of 0–5 kGy, the CH_3COOH was produced rapidly and the concentration started to increase. The final concentration of CH_3COOH was about 2.45 mg/L at 5 kGy.

At the radiation dose of 0–4 kGy, the concentration of HCOOH started to increase, and the maximum concentration was about 0.95 mg/L. Then, the concentration decreased with the increase in radiation dose.

The corresponding inflexion points of the HCOOH concentration curves appeared at about 4 kGy. As more reactive species are produced at higher radiation dose, more HCOOH molecules are released from the SMT degradation process. The HCOOH molecules could be degraded at the same duration. When the radiation dose was high, the concentration of HCOOH decreased due to the mineralization by the reactive species.

Besides the aforementioned three compounds, several other intermediate products such as 4,6-dimethylpyrimidin-2-amine, 4-aminobenzenesulfonic acid, 4-nitrophenol 4-nitrobenzenesulfonic acid, 2-amino-6-methylpyrimidine-4-carboxylic acid and 4-amino-*N*-carbamimidoyl-benzenesulfonamide have been detected in our past research [33].

3.3 Biodegradability and microbial toxicity of the products

The biodegradability and microbial toxicity of SMT radiolytic products were measured by specific oxygen uptake rate (SOUR) of the activated sludge, which was shown in Figure 5 [34]. The sludge was aerated for 24 h to reach ER stage before experiments. The oxygen uptake rates were measured by the gas pressure difference in the period of 6 h at different conditions, which were recorded in the curve. When the SOUR of the activated sludge was higher than the ER, the biodegradation was considered to be enhanced under this condition.

Figure 4: The variation in pH values and the formation of HCOOH, CH₃COOH, SO₄²⁻ with the increasing radiation dose (SMT initial concentration: 20 mg/L; dose rate: 103 Gy/min). (a) pH value, (b) HCOOH, (c) CH₃COOH and (d) SO₄²⁻.

The SOUR curve of the activated sludge treated by the SMT with no radiation was a little lower than the curve of ER, which meant that the SMT was toxic to microorganisms and could inhibit its respiration. The metabolic activity of the microorganism can be improved by the intermediate products of SMT degradation at the radiation dose of 1, 2 and 5 kGy. After 1 h reaction, the SOUR was increased about 14.3%, 7.1% and 14.3% at the radiation dose of 1, 2 and 5 kGy, respectively. Finally, the biodegradability of SMT was improved about 14.5%, 10.9% and 14.6%, respectively (after 6 h).

When the radiation dose was 4 kGy, which was a little higher in the experiments, the SOUR curve was much lower than the curve of ER. The inhibition ratio was about 16.7% after 1 h. At the end of the experiment, about 20% of the biodegradability was inhibited. The respiration of the activated sludge was inhibited greatly, and the metabolic activity of the microorganism was quite reduced. All these showed that some radiolytic

products caused inhibitory effects on the microorganism at the radiation dose of 4 kGy, which were not produced

Figure 5: SOUR of the activated sludge using SMT radiolytic products at different irradiation doses (SMT initial concentration: 20 mg/L; dose rate: 103 Gy/min; ER).

at lower radiation dose. With the increase in radiation dose, those products that showed the inhibition to the metabolic activity of the microorganism were degraded, and the biodegradability was recovered.

4 Conclusions

Gamma radiation had a great effect on the SMT degradation in aqueous solution. The SMT degradation kinetic follows the modified pseudo-first-order kinetic. The removal rate of SMT was about 95% at the absorbed dose of 5 kGy. However, the removal of TOC and TN was not as effective as that of SMT. When the radiation dose was 5 kGy, the TOC and TN removal were about 6.8% and 10.5%, respectively. Some ions including HCOO^- , CH_3COO^- and SO_4^{2-} released during SMT degradation were studied as well.

The SMT was inhibitive to the respiration of the microorganism, and the inhibition could be reduced or eliminated after irradiation under a certain absorbed dose, which meant that the biodegradability of the SMT could be improved by the gamma radiation. The biodegradability of the SMT can be improved by the intermediate products of SMT degradation at the radiation doses of 1, 2 and 5 kGy. The best promotion effect was at 1 kGy (14.5%) and 5 kGy (14.6%). Gamma radiation could be used as a pretreatment technology before the biodegradation process for pharmaceutical waste water.

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Conflict of interest: The authors declare no conflict of interest.

References

- [1] Singer RS, Finch R, Wegener HC, Bywater R, Walters J, Lipsitch M. Antibiotic resistance – the interplay between antibiotic use in animals and human beings. *Lancet Infect Dis*. 2003;3:47–51. doi: 10.1016/S1473-3099(03)00490-0.
- [2] Cabello FC. Heavy use of prophylactic antibiotics in aquaculture: a growing problem for human and animal health and for the environment. *Environ Microbiol*. 2006;8:1137–44. doi: 10.1111/j.1462-2920.2006.01054.x.
- [3] Kümmerer K. Antibiotics in the aquatic environment – a review – Part II. *Chemosphere*. 2009;75:435–41. doi: 10.1016/j.chemosphere.2008.12.006.
- [4] Wright GD. The antibiotic resistome: the nexus of chemical and genetic diversity. *Nat Rev Microbiol*. 2007;5:175–86. doi: 10.1038/nrmicro1614.
- [5] Aminov RI. The role of antibiotics and antibiotic resistance in nature. *Environ Microbiol*. 2009;11:2970–88. doi: 10.1111/j.1462-2920.2009.01972.x.
- [6] Ledjeri A, Yahiaoui I, Kadji H, Aissani-Benissad F, Amrane A, Fourcade F. Combination of the Electro/ Fe^{3+} /peroxydisulfate (PDS) process with activated sludge culture for the degradation of sulfamethazine. *Environ Toxicol Phar*. 2017;53:34–39. doi: 10.1016/j.etap.2017.04.022.
- [7] Yuan F, Hu C, Hu X, Qu JH, Yang M. Degradation of selected pharmaceuticals in aqueous solution with UV and UV/ H_2O_2 . *Water Res*. 2009;43:1766–74. doi: 10.1016/j.watres.2009.01.008.
- [8] Song C, Gao NY, Gao HW. Transmembrane distribution of kanamycin and chloramphenicol: insights into the cytotoxicity of antibacterial drugs. *Mol Biosyst*. 2010;6:1901–10. doi: 10.1039/b921810f.
- [9] Chen J, Zhou X, Zhang Y, Gao HP. Potential toxicity of sulfanilamide antibiotic: binding of sulfamethazine to human serum albumin. *Sci Total Environ*. 2012;432:269–74. doi: 10.1016/j.scitotenv.2012.06.003.
- [10] Sirés I, Brillas E. Remediation of water pollution caused by pharmaceutical residues based on electrochemical separation and degradation technologies: a review. *Environ Int*. 2012;40:2012–229. doi: 10.1016/j.envint.2011.07.012.
- [11] Zhang R, Zhang G, Tang J, Xu WH, Li J, Liu X, et al. Levels, spatial distribution and sources of selected antibiotics in the East River (Dong jiang), South China. *Aquat Ecosys Health Manage*. 2012;15:210–8. doi: 10.1080/14634988.2012.689576.
- [12] Tamtam F, Mercier F, Le BB, Eurin J, Dinh QT, Clement M, et al. Occurrence and fate of antibiotics in the Seine River in various hydrological conditions. *Sci Total Environ*. 2008;393:84–95. doi: 10.1016/j.scitotenv.2007.12.009.
- [13] Luo Y, Xu L, Rysz M, Wang YQ, Zhang H, Alvarez PJJ. Occurrence and transport of tetracycline, sulfonamide, quinolone, and macrolide antibiotics in the Haihe River Basin, China. *Environ Sci Technol*. 2011;45:1827–33. doi: 10.1021/es104009s.
- [14] Hruska K, Franek M. Sulfonamides in the environment: a review and case report. *Vet Med*. 2012;57:1–35. doi: 10.17221/4969-VETMED.
- [15] Baran W, Adamek E, Ziemińska J, Sobczak A. Effects of the presence of sulfonamides in the environment and their influence on human health. *J Hazard Mater*. 2011;196:1–15. doi: 10.1016/j.jhazmat.2011.08.082.
- [16] Proia L, Lupini G, Osorio V, Perez S, Barcelo D, Schwartz T, et al. Response of biofilm bacterial communities to antibiotic pollutants in a Mediterranean river. *Chemosphere*. 2013;92:1126–35. doi: 10.1016/j.chemosphere.2013.01.063.
- [17] Fan Y, Ji Y, Kong D, Lu JH, Zhou QS. Kinetic and mechanistic investigations of the degradation of sulfamethazine in heat-activated persulfate oxidation process. *J Hazard Mater*. 2015;300:39–47. doi: 10.1016/j.jhazmat.2015.06.058.

- [18] Yahiaoui I, Aissani-Benissad F, Fourcade F, Amrane A. Removal of a mixture tetracycline-tylosin from water based on anodic oxidation on a glassy carbon electrode coupled to activated sludge. *Environ Technol.* 2015;36:1837–46. doi: 10.1080/09593330.2015.1013571.
- [19] Adamek E, Baran W, Sobczak A. Assessment of the biodegradability of selected sulfa drugs in two polluted rivers in Poland: effects of seasonal variations, accidental contamination, turbidity and salinity. *J Hazard Mater.* 2016;313:147–58. doi: 10.1016/j.jhazmat.2016.03.064.
- [20] Zhang TQ, Dong FL, Luo F, Li C. Degradation of sulfonamides and formation of trihalomethanes by chlorination after pre-oxidation with Fe(vi). *J Environ Sci.* 2018;73:89–95. doi: 10.1016/j.jes.2018.01.016.
- [21] Getoff N. Factors influencing the efficiency of radiation-induced degradation of water pollutants. *Radiat Phys Chem.* 2002;65:437–46. doi: 10.1016/S0969-806X(02)00342-0.
- [22] Wang JL, Wang JZ. Application of radiation technology to sewage sludge processing: a review. *J Hazard Mater.* 2007;143:2–7. doi: 10.1016/j.jhazmat.2007.01.027.
- [23] Ma HJ, Wang M, Yang RY, Wang WF, Zhao J, Shen ZQ, et al. Radiation degradation of Congo Red in aqueous solution. *Chemosphere.* 2007;68:1098–104. doi: 10.1016/j.chemosphere.2007.01.067.
- [24] Chen WS, Liang JS. Decomposition of nitrotoluenes from trinitrotoluene manufacturing process by electro-Fenton oxidation. *Chemosphere.* 2008;72:601–7. doi: 10.1016/j.chemosphere.2008.03.005.
- [25] Park W, Hwang MH, Kim TH, Lee MJ, Kim IS. Enhancement in characteristics of sewage sludge and anaerobic treatability by electron beam pretreatment. *Radiat Phys Chem.* 2009;78:124–9. doi: 10.1016/j.radphyschem.2008.09.010.
- [26] Al-Sheikhly M, Poster DL, An JC, Neta P, Silverman J, Huie RE. Ionizing radiation-induced destruction of benzene and dienes in aqueous media. *Environ Sci Technol.* 2006;40:3082–8. doi: 10.1021/es052533j.
- [27] Hu J, Wang JL. Degradation of chlorophenols in aqueous solution by γ -radiation. *Radiat Phys Chem.* 2007;76:1489–92. doi: 10.1016/j.radphyschem.2007.02.058.
- [28] Basfar AA, Mohamed KA, Al-Abduly AJ, Al-Kuraiji TS, Al-Shahrani AA. Degradation of diazinon contaminated waters by ionizing radiation. *Radiat Phys Chem.* 2007;76:1474–9. doi: 10.1016/j.radphyschem.2007.02.055.
- [29] Sánchez-Polo M, López-Peñalver J, Prados-Joya G, Ferro-García MA, Rivera-Utrilla J. Gamma irradiation of pharmaceutical compounds, nitroimidazoles, as a new alternative for water treatment. *Water Res.* 2009;43:4028–36. doi: 10.1016/j.watres.2009.05.033.
- [30] Hevroy TH, Golz AL, Hansen EL, Xie L, Bradshaw C. Radiation effects and ecological processes in a freshwater microcosm. *J Environ Radioact.* 2019;203:71–83. doi: 10.1016/j.jenvrad.2019.03.002.
- [31] Lee B, Lee M. Decomposition of 2,4,6-trinitrotoluene (TNT) by gamma irradiation. *Environ Sci Technol.* 2005;39:9278–85. doi: 10.1021/es0489590.
- [32] Wasiewicz M, Chmielewski AG, Getoff N. Radiation-induced degradation of aqueous 2,3-dihydroxynaphthalene. *Radiat Phys Chem.* 2006;75:201–9. doi: 10.1016/j.radphyschem.2005.08.015.
- [33] Liu YK, Wang JL. Degradation of sulfamethazine by gamma irradiation in the presence of hydrogen peroxide. *J Hazard Mater.* 2013;250:99–105. doi: 10.1016/j.jhazmat.2013.01.050.
- [34] Sun WH, Chen LJ, Tian JP, Wang JL, He SJ. Degradation of a monoazo dye Alizarin Yellow GG in aqueous solutions by gamma irradiation: decolorization and biodegradability enhancement. *Radiat Phys Chem.* 2013;83:86–9. doi: 10.1016/j.radphyschem.2012.10.014.