catalysts

MDPI

*Communication*

# Degradation of Sulfamethoxazole in Aqueous Solution by Low-Energy X-ray Irradiation

**Jun Yao [1], Weidong Rao [2], Hua Kong [2], Wentao Sun [2], Dengzhu Guo [2], Zhiwei Li [2,*] and Xianlong Wei [2,*]**

[1] Hunan Institute of Advanced Sensing and Information Technology, Xiangtan University, Xiangtan 411105, China
[2] Key Laboratory for the Physics and Chemistry of Nanodevices, School of Electronics, Peking University, Beijing 100871, China
* Correspondence: lzw111@pku.edu.cn (Z.L.); weixl@pku.edu.cn (X.W.)

**Abstract:** Antibiotic resistance has been a serious health threat of widespread concern, as antibiotics are difficult to degrade effectively in the environment. In this study, sulfamethoxazole (SMZ), a common antibiotic in an aqueous solution, was irradiated by low-energy X-ray to investigate the effect of the absorption dose, initial concentration, initial pH, irradiation energy and other conditions on the degradation of SMZ, as well as the kinetic mechanism of SMZ degradation. The results showed that low-energy X-ray irradiation could effectively degrade SMZ in an aqueous solution at different initial concentrations and acid-base degrees, and the degradation effect of irradiation in the range of 60–80 keV is independent of the energy of X-rays. The degradation rate of the SMZ solution, with an initial concentration of 10.70 mg/L, was 94.6% at an absorbed dose of 890 Gray, with a pH of 3.5. Similar to other works on the degradation of antibiotics by ionizing irradiation, the degradation of SMZ solutions conformed to a pseudo-first-order kinetic mechanism.

**Keywords:** antibiotic degradation; wastewater treatment; ionizing irradiation; low-energy X-ray; sulfamethoxazole



**Citation:** Yao, J.; Rao, W.; Kong, H.; Sun, W.; Guo, D.; Li, Z.; Wei, X. Degradation of Sulfamethoxazole in Aqueous Solution by Low-Energy X-ray Irradiation. *Catalysts* **2023**, *13*, 714. https://doi.org/10.3390/catal13040714

Academic Editor: Jorge Bedia

Received: 6 March 2023
Revised: 4 April 2023
Accepted: 8 April 2023
Published: 9 April 2023



## 1. Introduction

Antibiotics, which can effectively kill bacteria and resolve infection problems, play an important role in modern medicine. However, the problem of antimicrobial resistance due to the abuse of antibiotics has also gradually become a serious problem. The World Health Organization lists antimicrobial resistance as one of the top ten global health threats. The World Bank estimates that by 2050, 3.3% of the global GDP will be lost due to antimicrobial resistance [1,2]. Resistance to antibiotics results not only from the misuse of antibiotics but, more importantly, from the difficulty of effectively degrading antibiotics in the environment. Studies have shown that the metabolic pathways of antibiotics are relatively unique and will not completely degrade when they enter the body in humans or animals, most of which are in their original form or their intermediates when excreted into the natural environment [3]. Antibiotics are enriched in water bodies and will eventually be absorbed or consumed by aquatic organisms, inducing the development of drug resistance in microorganisms, thereby causing difficulty in the prevention and treatment of diseases in humans and animals [4]. Eliminating the antibiotic residues in various types of waste materials is essential for minimizing environmental pollution and ecological health risks from antibiotic contamination [5].

Due to their antimicrobial and persistent characteristics, antibiotics are often not effectively removed by conventional methods, which include physicochemical methods such as coagulation, flocculation, sedimentation and filtration [6–8], and biological methods, such as activated sludge and anaerobic digestion [9]. In contrast, newly developed physicochemical techniques, including oxidation and advanced oxidation, such as chlorination, ozone oxidation, Fenton oxidation, photolysis and photocatalysis [10], can overcome the

drawbacks of biological methods and effectively degrade antibiotics. Although these techniques effectively degrade antibiotics, their disadvantages are also obvious, such as slow degradation, high maintenance costs and unstable performance.

Irradiation degradation mainly refers to the removal of large amounts of contaminants by the fact that under ionizing irradiation, water generates strongly oxidizing (·OH) and strongly reducing species ($e^-_{aq}$, ·H) [11,12]. The common irradiation sources for ionizing irradiation include gamma rays, high-energy X-rays and electron beams. Currently, many studies have used gamma rays or electron beam irradiation as advanced oxidation processes for antibiotic removal, which includes norfloxacin, amoxicillin, ofloxacin, chloramphenicol [13–16], etc. Gamma rays have the strongest penetration compared to other irradiation sources and are relatively friendly to thermal-sensitive objects, with no secondary contamination after irradiation; however, radioactive cobalt source waste disposal is complex. The electron beam generated by linear accelerators is high in energy and power but has relatively weak penetration compared with gamma rays as well as high-energy X-rays, which are generally used for killing bacteria on thinner objects or object surfaces and require higher doses when used to degrade antibiotics [17,18]. Nevertheless, the problem common to all three classes of irradiation sources is the high energy and need for comprehensive protective measures; thus, all require decontamination treatments in a centralized irradiation venue, which greatly limits the range of applications and efficiency of irradiation degradation. Relatively, the low-energy X-rays (usually no more than 100 keV), which are safer due to their lower requirements for irradiation protection, have wider application scenarios. However, to the best of our knowledge, studies on the degradation of antibiotics by irradiation with low-energy X-ray sources are not currently available.

Available reports indicate that the degree of antibiotic contamination in groundwater varies in different countries and regions, among which sulfonamide antibiotics are considered the most common antibiotic contamination in groundwater. In the analysis of antibiotic residues in European and Chinese water samples, sulfonamide antibiotics were found in wastewater at concentrations ranging from several tens to hundreds of ng/L [19,20], which are one of the highest levels among various antibiotics. At present, SMZ removals from aqueous solution are mainly via oxidation techniques. Garoma et al. [21] used $O_3$ to oxidize SMZ. Li et al. [22] adopted $H_2O_2$/UV to treat SMZ solutions. Trovó et al. [23] investigated the effect of the Fenton reagent on SMZ degradation. Abellán et al. [24] found that $TiO_2$ could facilitate SMZ degradation under UV irradiation, and SMZ removal was mainly controlled by the ·OH radical. Oyekunle et al. [25] summarized the development of several heterogeneous metals and non-metal catalysts used for persulfate activation for SMZ degradation. There have been studies on SMZ degradation using gamma ray [26–29] and high-energy electron beam [30] irradiation in ionizing radiation, but there has been no study on the degradation of SMZ by low-energy X-ray irradiation. In this work, we use low-energy X-ray irradiation to degrade sulfamethoxazole (SMZ) and investigate the effect of the absorption dose, initial concentration, initial pH, irradiation energy and other conditions on the degradation of SMZ. The kinetics of SMZ irradiation degradation are also studied. This study validates the feasibility of degrading antibiotics with low-energy X-rays and provides a prospective guide to this safe and easy handling method.

## 2. Experimental Design

### *2.1. Irradiation*

The irradiation experiments were carried out in our homemade irradiation chamber (Figure 1a). The irradiation chamber mainly comprises an X-ray source, a sample table, an ionizing irradiation dose-rate detector, a semiconductor cooling system, as well as a metal enclosure. The model of the X-ray source was the X80k1m-B from Beijing Kaiwei Technology Co., Ltd. (Beijing, China) with a filament current of 0.3 mA~1 mA and an anode energy range of 60–80 keV. The detector was a multi-purpose Magicmax-Universal X-ray detector with a German IBA probe XR. The cooling system was a semiconductor refrigeration module with a refrigeration power of 120 W. The metal enclosure,

800 mm × 600 mm × 600 mm in size, was made of lead and stainless steel, in which the lead plate thickness was 3 mm. The irradiation chamber contained a door with a self-locking device to guarantee no irradiation leakage during the experiment. Sulfamethoxazole (SMZ, purity > 99%) for irradiation was purchased from Meilun Yun (Beijing) Cell Biotech Co., Ltd. (Beijing, China), and both the analytical pure acetic acid and sodium hydroxide were purchased from Anhui Zesheng Technology Co., Ltd. (Hefei, China) The experimental water was all deionized water with a resistivity of 18.2 MΩ·cm (the water maker model was Merck Milli-Q, Germany). Before irradiation, a solution of a specific concentration of SMZ was prepared first and then poured into a card slot. The material of the slot was polypropylene with a height of 100 mm, a width of 80 mm and a thickness of 7 mm, in which the outer shell was 2 mm thick, and the solution was 3 mm thick. The card slot was then placed onto the sample table inside the irradiation chamber, and the location of the card slot was about 30 mm away from the X-ray window. The X-ray source was finally turned on to irradiate the SMZ solution.

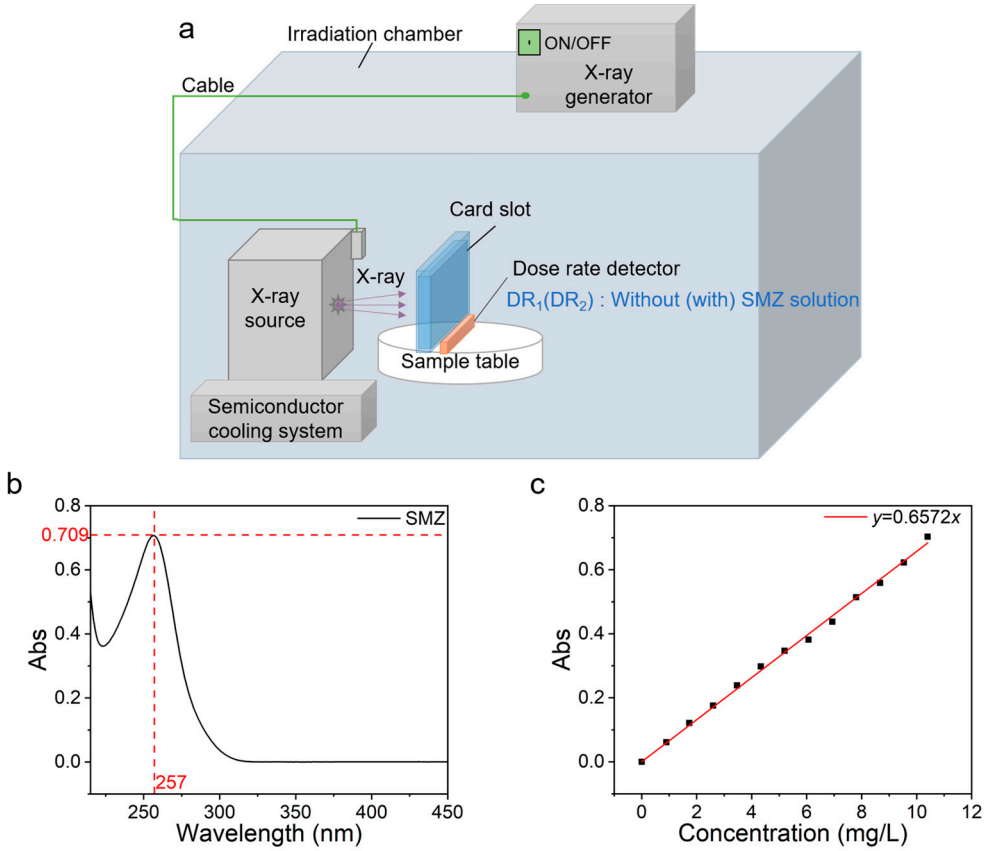


**Figure 1.** Irradiation and concentration calibration of SMZ solutions. (**a**) Schematic drawing of a low-energy X-ray irradiation device; (**b**) ultraviolet spectrogram of SMZ; (**c**) relationship between Abs and SMZ concentration.

As shown in Figure 1a, the dose rates $DR_1$ and $DR_2$ of X-ray irradiation are measured at the back of the slots with and without the SMZ solution, respectively, and the absorbed dose rate of the solution is $ADR = DR_1 - DR_2$. The total absorbed dose $AD = ADR \times t$, where $t$ is the duration of continuous irradiation.

### *2.2. Concentration Calibration of SMZ Solution*

After irradiation, the solution was poured into a quartz colorimetric dish and analyzed by an ultraviolet-visible near-infrared spectrophotometer UV (Agilent Cary5000, USA). The SMZ solution was scanned by light with a wavelength from 200 nm to 800 nm, and the absorbance spectra of the solution were obtained. The concentration of the solution

can be quantitatively analyzed by the wavelength of the maximum absorption peak on the spectrum. Figure 1b is a spectrum obtained by scanning an irradiated SMZ solution (baseline: NaOH, pH = 12). The effective wavelength ranges of 215 nm to 450 nm were selected. It can be seen that under alkaline conditions, the maximum absorption peak has a wavelength of 257 nm and an absorbance of 0.709. According to Bill's law [31], the absorbance is proportional to the concentration of the substance. Therefore, we compounded a total of 13 solutions of SMZ with a pH of 12 and concentrations from 0 to 10.4 mg/L. The absorbance of the solution at the wavelength of the maximum absorption peak was measured, and the curve in Figure 1c was obtained. Linear fitting was performed to obtain the relationship between the absorbance (Abs) and concentration © as Abs = 0.6571*c*, with the correlation $R^2$ as 0.999. In the subsequent experiments, the concentration of the solution can be calculated from the absorbance of the SMZ solution using this standard. For the validation experiment of norfloxacin (NOR) degradation, a similar concentration calibration process was used.

The exposure time of the X-ray irradiation can be controlled precisely by an automatic script, which gives our data excellent repeatability. In order to prove this, an experiment was conducted. Under the conditions of 80 keV, 1 mA, pH 12, and absorbed dose 156 Gray, we used low-energy X-rays to degrade SMZ independently three times. The calculated degradation rates of the three experiments are 56.46%, 56.45% and 56.37%, respectively, with a mean value of $\mu$ = 56.43%, a standard deviation of $\sigma$ = 0.051% and a standard error of $\sigma/\mu$ = 0.0009. These results show that the collected data in our manuscript have excellent repeatability.

## 3. Results and Discussion

### *3.1. Effect of Absorbed Dose on SMZ Degradation*

In order to study the kinetic mechanism of SMZ degradation by irradiation, we first studied the effect of the absorbed dose on the degradation rate of SMZ. As shown in Figure 2a, the concentration of SMZ, with an initial concentration of 10.70 mg/L, decreases with the increase of the absorbed dose at an energy of 80 keV, a beam of 1 mA and a pH of 3.5. At the absorbed doses of 260 Gray, 290 Gray, 430 Gray and 890 Gray, the degradation rates were 68.4%, 75.7%, 84.9% and 94.6%, respectively. The results of this study are similar to those of the degradation of sulfonamides by gamma or electron beam irradiation [27,32,33].

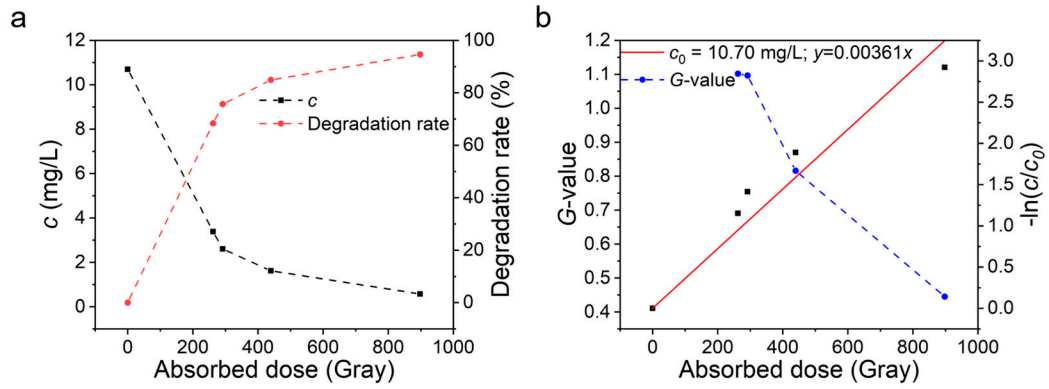


**Figure 2.** Effect of absorbed dose on SMZ degradation (energy = 80 keV; beam current = 1 mA; initial SMZ = 10.70 mg/L; pH = 3.5): (**a**) Concentration change and degradation rate diagram of SMZ; (**b**) G-value variation and kinetic fitting.

In general, the rate of antibiotic degradation increases with the dose of absorption. The kinetic models of antibiotic degradation can be divided into zero order, first order and second order [34]. The relationship between the concentration and absorbed dose is $c - c_0 = -k_1 D$, $\ln(c/c_0) = -k_2 D$, and $(1/c - 1/c_0) = k_3 D$, respectively. Among them, c is

the concentration of the SMZ solution after irradiation, $c_0$ is its initial concentration, $k_1$, $k_2$, and $k_3$ are the dose constants, and D is the absorbed dose. As shown in Figure 2b, the relationship between $-\ln(c/c_0)$ and the absorbed dose is linear [35], and the correlation coefficient $R^2$ is higher than 0.97, indicating that the radiolysis of SMX conforms to pseudo-first-order reaction kinetics.

The G-value is an index of the utilization efficiency of active substances in the degradation of target pollutants. It can be defined as the number of particles that react (or are produced) when the irradiated medium absorbs 100 eV of irradiation energy [36]. The calculation method is shown in Equation (1), where the unit of the G-value is mol/J, $c_0$ is the initial concentration, and c is the concentration after irradiation (mol/L). D is the absorbed dose in Gray, $6.02 \times 10^{23}$ is the Avogadro number, and $6.24 \times 10^{16}$ is the conversion factor from Gray to 100 eV/L.

$$G = \frac{\text{molecularity}}{100\ \text{eV}} = \frac{(c_0 - c) \times 6.02 \times 10^{23}}{D \times 6.24 \times 10^{16}} = \frac{c_0 - c}{D} \times 1.0 \times 10^7 \quad (1)$$

As shown in Figure 2b, the G-value of SMZ under low-energy X-ray irradiation decreases from 1.10 mol/J to 0.44 mol/J with the increased absorbed dose, which can be explained from the basic principle of degradation. The first step of irradiation degradation of antibiotics is the reaction between rays and water. Under X-ray irradiation, the aqueous solution will produce several reactive substances [37], as shown in Equation (2) (the number in the brackets represents the G-value). Among them, ·OH is considered to play a leading role in the degradation of antibiotics [15]. However, with the increase of the absorbed dose and a decrease of the SMZ concentration, the reaction rate of free radicals and by-products increases, resulting in the reduction of effective oxidation free radical concentration (·OH) in the reaction with antibiotics, and then the reduction of the G-value. This is similar to the degradation of antibiotics previously studied [38,39].

$$H_2O \rightarrow e^-_{aq}(2.6) + \cdot H(0.6) + \cdot OH(2.7) + H_2(0.45) + H_2O_2(0.7) + H_3O^+(2.7) \quad (2)$$

A free radical test was conducted to validate the conclusion. Isopropanol (IPA), an effective radical scavenger of ·OH, was added to the aqueous solution of sulfamethoxazole. Figure 3a shows the degradation results (initial SMZ = 9.72 mg/L, energy = 80 keV, beam current = 1 mA, and pH = 12). At an absorbed dose of 76 Gray, the SMZ solution added with IPA almost did not degrade (with a degradation rate of 0.45%), while the degradation rate of the control group without IPA was 49.13%. Such a discrepancy is solid evidence for confirming the dominant role of ·OH in the degradation of SMZ.

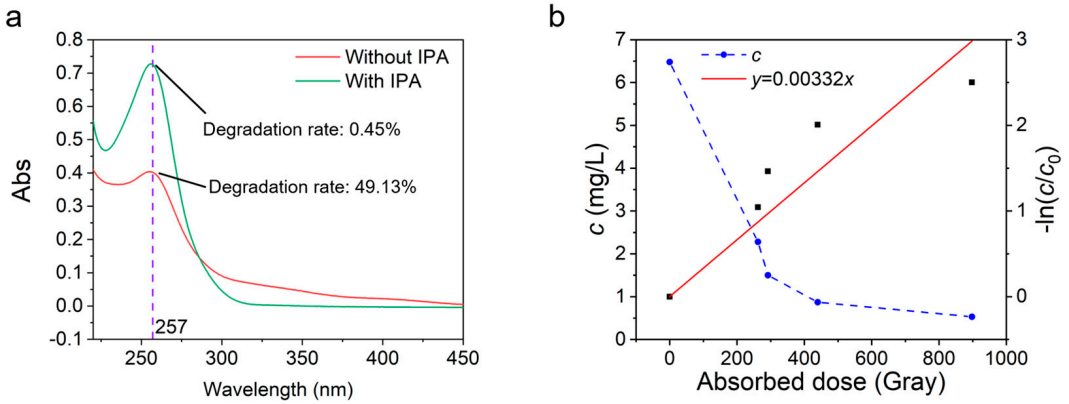


**Figure 3.** Validation tests of degradation kinetics model. (**a**) UV-visible spectrogram with or without IPA irradiation (initial SMZ = 9.72 mg/L, energy = 80 keV, beam current = 1 mA and pH = 12, absorbed dose = 76 Gray). (**b**) X-ray degradation of norfloxacin (initial NOR = 6.48 mg/L, energy = 80 keV, beam current = 1 mA, pH = 3.5).

Under the same conditions (energy 80 keV, beam current 1 mA, pH 3.5), we then used low-energy X-ray to degrade norfloxacin, another common antibiotic. As is shown in Figure 3b, norfloxacin, at an initial concentration of 6.48 mg/L, has a degradation rate of 91.8% at an absorbed dose of 890 Gray, which shows a similar trend and degradation level with those of sulfamethoxazole, indicating that low-energy X-ray irradiation is a broad-spectrum degradation method for antibiotics. The degradation data of norfloxacin fitted well using pseudo-first-order reaction kinetics, which further proved the kinetic mechanism of degradation under X-ray irradiation.

### *3.2. Effect of Initial Concentration on Degradation of SMZ*

To study the effect of the initial concentration on SMZ degradation, we compounded the SMZ solutions with an initial concentration of 10.70 mg/L and 37.54 mg/L, respectively, and a 30 mL solution for each concentration was irradiated in the irradiation chamber with the energy of 80 keV, a beam of 1 mA and a pH of 12.

The corresponding SMZ degradation rates at different absorption doses are shown in Figure 4a. The initial concentration of SMZ significantly affects the degradation rate. At the absorbed dose of 890 Gray, when the initial concentration was 10.70 mg/L and 37.54 mg/L, the degradation rates of SMZ were 91.5% and 85.2%, respectively.

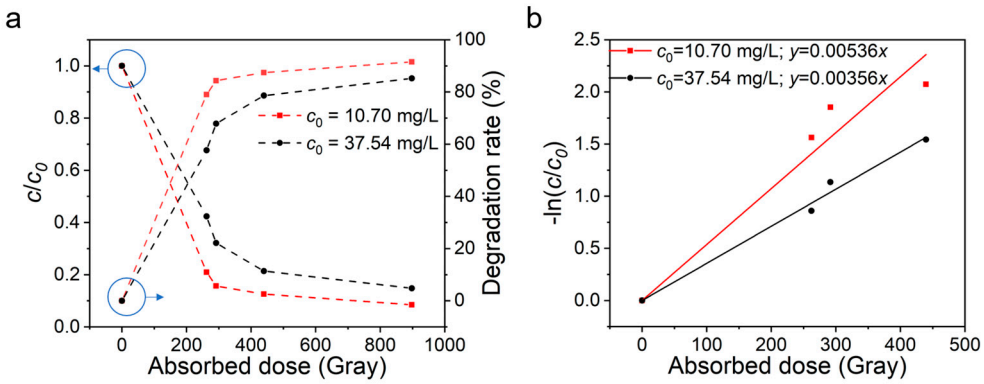


**Figure 4.** Effect of different initial concentrations of SMZ on degradation efficiency (energy = 80 keV; beam current = 1 mA; pH = 12): (**a**) Degradation of SMZ at different initial concentrations and absorbed doses; (**b**) pseudo-first-order dynamics of experimental data fitting.

According to the pseudo-first-order kinetic model, the relationship between the antibiotic concentration and absorbed dose can be formulated as $\ln(c/c_0) = -kD$. As shown in Figure 4b, for the SMZ solution with initial concentrations of 10.70 mg/L and 37.54 mg/L, the dose constants are $5.36 \times 10^{-3}$ Gray$^{-1}$ and $3.56 \times 10^{-3}$ Gray$^{-1}$, respectively. This shows that low-concentration SMZ shows a high degradation rate during radiolysis. On the one hand, at a high SMX concentration, a large number of intermediates were produced during radiolysis, consuming more free radicals, such as ·OH, $e_{aq}^-$ and ·H, thus reducing the reaction rate of free radicals and SMZ [27]. On the other hand, the SMZ solution with an initial concentration of 37.54 mg/L was first reduced to 10.70 mg/after some dose of irradiation with the left part to keep degrading. Therefore, under the same total absorbed dose, the final concentration of the solution with the initial concentration of 10.70 mg/L is certainly lower.

### *3.3. Effect of pH Value on Degradation of SMZ*

In order to study the effect of irradiation degradation under different pH environments, we selected NaOH and $CH_3COOH$ as the baselines, with pH values of 12 and 3.5, respectively. The UV-visible spectra of the SMZ solution of NaOH and $CH_3COOH$ are shown in Figure 5a. It can be found that the maximum peak positions of the spectra under different baselines are different (257 nm and 267 nm, respectively), indicating that the absorption effect of SMZ changes under different pH conditions.

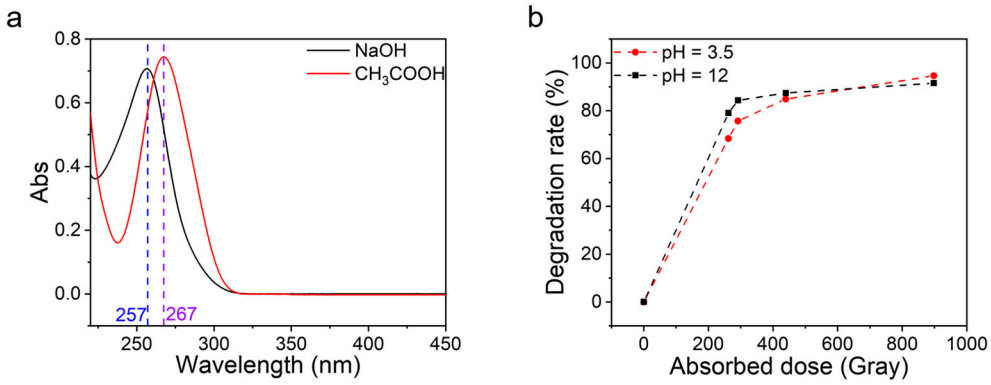


**Figure 5.** Effect of pH value of initial solution on degradation of SMZ: (**a**) Ultraviolet-visible spectrogram of NaOH and $CH_3COOH$; (**b**) degradation rate diagrams of SMZ (energy = 80 keV; beam current = 1 mA; initial SMZ = 10.70 mg/L).

As shown in Figure 5b, under acidic and alkaline conditions, the degradation rates of SMZ are slightly different because, in the process of low-energy X-ray irradiation degradation (the energy is 80 keV, the beam current is 1 mA, the initial concentration is 10.70 mg/L), the pH value of the initial solution will affect the number of active particles produced by the irradiated aqueous solution, thus affecting the degradation effect of SMZ. However, the degradation rates of solutions with different pH values are very close, and there is no significant difference. This shows that the degradation of SMZ by low-energy X-ray irradiation is effective in various pH solutions.

### *3.4. Effect of Irradiation Energy on SMZ*

In order to study the effect of low-energy X-ray energy on the irradiation degradation rate of SMZ, a fixed beam current of 1 mA, a pH value of 12, an initial concentration of 10.70 mg/L and X-ray energies of 60 keV, 70 keV and 80 keV were used to irradiate SMZ. The maximum SMZ irradiation absorbed dose was set at 890 Gray. Figure 6 shows that the degradation rates of SMZ at 890 Gray are 91.5%, 90.5% and 90.4%, respectively. The results show that when the absorbed dose of the SMZ solution is fixed, the irradiation energy has little effect on the degradation rate of SMZ in the range of 60–80 keV. The reason may be that at the same volume and concentration, the G-values of free radical particles, such as ·OH, $e_{aq}^-$ and ·H by low-energy X-ray irradiation of the SMZ solution are equal.

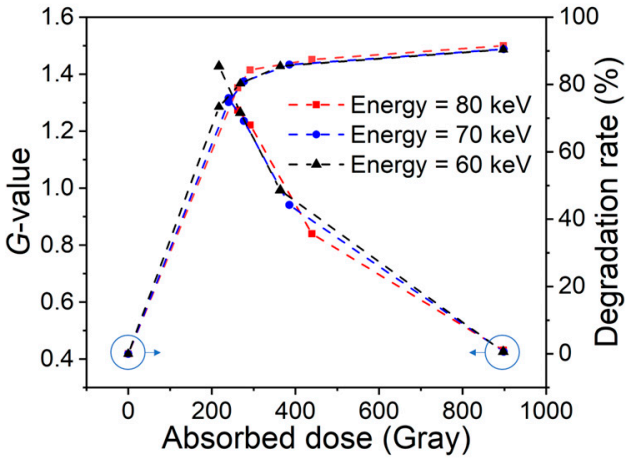


**Figure 6.** Effect of irradiation energy on degradation of SMZ (beam current = 1 mA; initial SMZ = 10.70 mg/L; pH = 12).

## 4. Conclusions

In this study, low-energy X-ray irradiation was used to irradiate the aqueous solution of SMZ, and it was found that low-energy X-ray irradiation could effectively degrade SMZ in the aqueous solution. The experimental results show that low-energy X-ray irradiation is an effective method for degrading antibiotics, with good degradation effects for solutions with different initial concentrations and pH values. Considering the low protection requirements and wide application range of low-energy X-ray sources, this technology is expected to be widely used in the future. This study provides valuable guidance for solving the problems of antibiotic wastewater pollution and the antibiotic resistance caused by it.

**Author Contributions:** Conceptualization, X.W., D.G. and J.Y.; methodology, X.W., J.Y. and W.R.; software, J.Y.; validation, J.Y., H.K. and W.S.; formal analysis, J.Y. and Z.L.; investigation, J.Y.; resources, X.W.; data curation, J.Y.; writing—original draft preparation, J.Y.; writing—review and editing, X.W. and Z.L.; visualization, J.Y.; supervision, X.W. and Z.L.; project administration, J.Y.; funding acquisition, X.W. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research was funded by NSF of China, grant number 62022007.

**Institutional Review Board Statement:** Not applicable.

**Informed Consent Statement:** Not applicable.

**Data Availability Statement:** Not applicable.

**Conflicts of Interest:** The authors declare no conflict of interest.

## References

1. World Health Organization. *Global Antimicrobial Resistance Surveillance System (GLASS) Report: Early Implementation 2020*; World Health Organization: Geneva, Switzerland, 2020.
2. World Health Organization. *Global Antimicrobial Resistance and Use Surveillance System (GLASS) Report: 2022*; World Health Organization: Geneva, Switzerland, 2022.
3. Huang, C.-H.; Renew, J.E.; Smeby, K.L.; Pinkston, K.; Sedlak, D.L. Assessment of potential antibiotic contaminants in water and preliminary occurrence analysis. *J. Contemp. Water Res. Educ.* **2011**, *120*, 4.
4. Yang, W.; Fulidong, F.; Ma, J.; Cui, Y. Research progress of antibiotic pollution and treatment technologies in China. In Proceedings of the E3S Web of Conferences, Shanghai, China, 18–20 September 2020; p. 04004.
5. Sodhi, K.K.; Kumar, M.; Balan, B.; Dhaulaniya, A.S.; Shree, P.; Sharma, N.; Singh, D.K. Perspectives on the antibiotic contamination, resistance, metabolomics, and systemic remediation. *SN Appl. Sci.* **2021**, *3*, 269. [CrossRef]
6. Li, S.-Z.; Li, X.-Y.; Wang, D.-Z. Membrane (RO-UF) filtration for antibiotic wastewater treatment and recovery of antibiotics. *Sep. Purif. Technol.* **2004**, *34*, 109–114. [CrossRef]
7. Back, J.O.; Obholzer, T.; Winkler, K.; Jabornig, S.; Rupprich, M. Combining ultrafiltration and non-thermal plasma for low energy degradation of pharmaceuticals from conventionally treated wastewater. *J. Environ. Chem. Eng.* **2018**, *6*, 7377–7385. [CrossRef]
8. Back, J.O.; Stadlmayr, W.; Jabornig, S.; Winkler, F.; Winkler, K.; Rupprich, M. Removal of arsenic from water with non-thermal plasma (NTP), coagulation and membrane filtration. *Water* **2018**, *10*, 1385. [CrossRef]
9. Wallace, J.S.; Garner, E.; Pruden, A.; Aga, D.S. Occurrence and transformation of veterinary antibiotics and antibiotic resistance genes in dairy manure treated by advanced anaerobic digestion and conventional treatment methods. *Environ. Pollut.* **2018**, *236*, 764–772. [CrossRef] [PubMed]
10. Homem, V.; Santos, L. Degradation and removal methods of antibiotics from aqueous matrices–a review. *J. Environ. Manag.* **2011**, *92*, 2304–2347. [CrossRef] [PubMed]
11. Buxton, G.V.; Greenstock, C.L.; Helman, W.P.; Ross, A.B. Critical Review of rate constants for reactions of hydrated electrons, hydrogen atoms and hydroxyl radicals (· OH/· O− in Aqueous Solution. *J. Phys. Chem. Ref. Data* **1988**, *17*, 513–886. [CrossRef]
12. Bielski, B.H.; Cabelli, D.E.; Arudi, R.L.; Ross, A.B. Reactivity of $HO_2/O^-_2$ radicals in aqueous solution. *J. Phys. Chem. Ref. Data* **1985**, *14*, 1041–1100. [CrossRef]
13. Alsager, O.A.; Alnajrani, M.N.; Alhazzaa, O. Decomposition of antibiotics by gamma irradiation: Kinetics, antimicrobial activity, and real application in food matrices. *Chem. Eng. J.* **2018**, *338*, 548–556. [CrossRef]
14. Sayed, M.; Khan, J.A.; Shah, L.A.; Shah, N.S.; Khan, H.M.; Rehman, F.; Khan, A.R.; Khan, A.M. Degradation of quinolone antibiotic, norfloxacin, in aqueous solution using gamma-ray irradiation. *Environ. Sci. Pollut. Res.* **2016**, *23*, 13155–13168. [CrossRef]
15. Wang, M.; Zhang, L.; Zhang, G.; Pang, T.; Zhang, X.; Cai, D.; Wu, Z. In situ degradation of antibiotic residues in medical intravenous infusion bottles using high energy electron beam irradiation. *Sci. Rep.* **2017**, *7*, 39928. [CrossRef] [PubMed]

16. Cho, J.Y.; Chung, B.Y.; Hwang, S.A. Detoxification of the veterinary antibiotic chloramphenicol using electron beam irradiation. *Environ. Sci. Pollut. Res.* **2015**, *22*, 9637–9645. [CrossRef] [PubMed]
17. Silindir, M.; Özer, A.Y. Sterilization methods and the comparison of E-beam sterilization with gamma radiation sterilization. *Fabad J. Pharm. Sci.* **2009**, *34*, 43.
18. Fan, X.; Niemira, B.A. Gamma ray, electron beam, and X-ray irradiation. *Food Saf. Eng.* **2020**, *179*, 471–492.
19. Carvalho, I.T.; Santos, L. Antibiotics in the aquatic environments: A review of the European scenario. *Environ. Int.* **2016**, *94*, 736–757. [CrossRef]
20. Zeng, H.; Li, J.; Zhao, W.; Xu, J.; Xu, H.; Li, D.; Zhang, J. The Current Status and Prevention of Antibiotic Pollution in Groundwater in China. *Int. J. Environ. Res. Public Health* **2022**, *19*, 11256. [CrossRef]
21. Garoma, T.; Umamaheshwar, S.K.; Mumper, A. Removal of sulfadiazine, sulfamethizole, sulfamethoxazole, and sulfathiazole from aqueous solution by ozonation. *Chemosphere* **2010**, *79*, 814–820. [CrossRef]
22. Li, W.; Lan, M.; Peng, X. Removal of antibiotics from swine wastewater by UV/H2O2 combined oxidation. *Environ. Poll. Control* **2011**, *33*, 25–28.
23. Trovó, A.G.; Nogueira, R.F.; Agüera, A.; Fernandez-Alba, A.R.; Sirtori, C.; Malato, S. Degradation of sulfamethoxazole in water by solar photo-Fenton. Chemical and toxicological evaluation. *Water Res.* **2009**, *43*, 3922–3931. [PubMed]
24. Abellán, M.; Bayarri, B.; Giménez, J.; Costa, J. Photocatalytic degradation of sulfamethoxazole in aqueous suspension of $TiO_2$. *Appl. Catal. B Environ.* **2007**, *74*, 233–241. [CrossRef]
25. Oyekunle, D.T.; Gendy, E.A.; Ifthikar, J.; Chen, Z. Heterogeneous activation of persulfate by metal and non-metal catalyst for the degradation of sulfamethoxazole: A review. *Chem. Eng. J.* **2022**, *437*, 135277. [CrossRef]
26. Kim, H.Y.; Kim, T.-H.; Cha, S.M.; Yu, S. Degradation of sulfamethoxazole by ionizing radiation: Identification and characterization of radiolytic products. *Chem. Eng. J.* **2017**, *313*, 556–566. [CrossRef]
27. Wang, J.; Guo, Z.; Shen, X.; Guo, Q.; Zhao, Y.; Zhu, S.; Guo, Z. Gamma irradiation-induced decomposition of sulfamethoxazole in aqueous solution: The influence of additives, biological inhibitory, and degradation mechanisms. *Environ. Sci. Pollut. Res.* **2017**, *24*, 23658–23665. [CrossRef]
28. Wang, S.; Wang, J. Radiation-induced degradation of sulfamethoxazole in the presence of various inorganic anions. *Chem. Eng. J.* **2018**, *351*, 688–696. [CrossRef]
29. Zhuan, R.; Wang, J. Degradation of sulfamethoxazole by ionizing radiation: Kinetics and implications of additives. *Sci. Total Environ.* **2019**, *668*, 67–73. [CrossRef]
30. Kim, T.-H.; Kim, S.D.; Kim, H.Y.; Lim, S.J.; Lee, M.; Yu, S. Degradation and toxicity assessment of sulfamethoxazole and chlortetracycline using electron beam, ozone and UV. *J. Hazard. Mater.* **2012**, *227*, 237–242. [CrossRef] [PubMed]
31. Mäntele, W.; Deniz, E. UV–VIS absorption spectroscopy: Lambert-Beer reloaded. *Spectrochim. Acta Part A Mol. Biomol. Spectrosc.* **2017**, *173*, 965–968. [CrossRef] [PubMed]
32. Liu, Y.; Wang, J.; Zhou, Z.; Zheng, X.; Zhao, L.; Yu, A. The degradation, biodegradability and toxicity evaluation of sulfamethazine antibiotics by gamma radiation. *Open Chem.* **2020**, *18*, 1188–1194. [CrossRef]
33. Sági, G.; Bezsenyi, A.; Kovács, K.; Klátyik, S.; Darvas, B.; Székács, A.; Mohácsi-Farkas, C.; Takács, E.; Wojnárovits, L. Radiolysis of sulfonamide antibiotics in aqueous solution: Degradation efficiency and assessment of antibacterial activity, toxicity and biodegradability of products. *Sci. Total Environ.* **2018**, *622*, 1009–1015. [CrossRef] [PubMed]
34. Espenson, J.H. *Chemical Kinetics and Reaction Mechanisms*; Citeseer: Princeton, NJ, USA, 1995; Volume 102.
35. Khan, J.A.; Shah, N.S.; Khan, H.M. Decomposition of atrazine by ionizing radiation: Kinetics, degradation pathways and influence of radical scavengers. *Sep. Purif. Technol.* **2015**, *156*, 140–147. [CrossRef]
36. Basfar, A.; Mohamed, K.; Al-Abduly, A.; Al-Shahrani, A. Radiolytic degradation of atrazine aqueous solution containing humic substances. *Ecotoxicol. Environ. Saf.* **2009**, *72*, 948–953. [CrossRef] [PubMed]
37. Cooper, W.J.; Cadavid, E.; Nickelsen, M.G.; Lin, K.; Kurucz, C.N.; Waite, T.D. Removing THMs from drinking water using high-energy electron-beam irradiation. *J. -Am. Water Work. Assoc.* **1993**, *85*, 106–112. [CrossRef]
38. Sánchez-Polo, M.; López-Peñalver, J.; Prados-Joya, G.; Ferro-García, M.A.; Rivera-Utrilla, J. Gamma irradiation of pharmaceutical compounds, nitroimidazoles, as a new alternative for water treatment. *Water Res.* **2009**, *43*, 4028–4036. [CrossRef]
39. Shuibo, X.; Ting, C.; Zhiping, L.; Yuqi, W.; Yingjiu, L.; Hui, L. Amoxicillin degradation in aqueous solution by gamma-ray irradiation. *Chin. J. Environ. Eng.* **2015**, *9*, 73–78.