

Article

Reductive Degradation of N-Nitrosodimethylamine via UV/Sulfite Advanced Reduction Process: Efficiency, Influencing Factors and Mechanism

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Abstract: N-nitrosodimethylamine (NDMA), as an emerging nitrogenous disinfection byproduct, is strictly controlled by multiple countries due to its high toxicity in drinking water. Advanced reduction processes (ARPs) are a new type of water treatment technology that can generate highly reactive reducing radicals and make environmental contaminants degrade rapidly and innocuously. In this study, a systematic investigation on the kinetics of the NDMA degradation via the chosen UV/sulfite ARP and the impacts of some key parameters of reaction system was conducted. The results indicated that the UV/sulfite ARP was an efficient and energy saving method for the reductive degradation of NDMA. A total of 94.40% of NDMA was removed using the UV/sulfite ARP, while only 45.48% of NDMA was removed via direct UV photolysis under the same reaction conditions. The degradation of NDMA via the UV/sulfite ARP followed pseudo-first-order kinetics. Increasing both the UV light intensity and sulfite dosage led to a proportional increase in the NDMA removal efficiency. The alkaline condition favored the degradation of NDMA, with the removal efficiency increasing from 21.57% to 66.79% within the initial 5 min of the reaction when the pH increased from 3 to 11. The presence of dissolved oxygen substantially decreased the removal efficiency of NDMA due to the formation of oxidizing superoxide radicals, which competed with NDMA by capturing the reducing active radicals during the reaction. An analysis of the degradation products indicated that several refractory intermediates such as dimethylamine, methylamine and nitrite were completely decomposed via the UV/sulfite ARP, and the final degradation products were formate, ammonia and nitrogen.

Keywords: N-nitrosodimethylamine; UV/sulfite ARP; reductive degradation; influencing factors; mechanism



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1. Introduction

Disinfection is vital in providing safe drinking water and preventing waterborne disease. However, disinfectants could react with various organic and inorganic substances, which are widely present in raw water and generate a range of harmful substances as disinfection byproducts (DBPs). Since Rook first found chloroform in disinfected water and confirmed its carcinogenicity to rodents, DBPs have gradually attracted attention. In order to reduce the generation of conventional DBPs such as Trihalomethanes (THMs) and Haloacetic acids (HAAs), more and more drinking water plants have used chloramine disinfection instead of chlorine disinfection in recent years. However, chloramine can form increased levels of nitrogenous DBPs (N-DBPs) such as N-nitrosamines (NAs), haloacetamides (HAMs) and halonitromethane (HNMs) [1]. Compared with conventional carbonaceous disinfection by-products (C-DBPs), N-DBPs have higher genotoxicity and cytotoxicity [2]. Therefore, although the amount of N-DBPs in disinfected water is generally lower than that of C-DBPs, its threat to human health cannot be ignored.

As an emerging N-DBP, N-nitrosodimethylamine (NDMA) has received much attention from the drinking water treatment industry recently due to it being strongly carcinogenic and frequently detected [3,4]. According to a national survey in China, NDMA had the highest concentration in finished and tap water, with an average concentration of 11 and 13 ng L⁻¹, respectively [5]. In Canada, the maximum concentration of NDMA for tap water reached 67 ng L⁻¹ [6]. The USEPA classified NDMA as a probable human carcinogen and established a 10⁻⁶ lifetime cancer risk concentration of 0.7 ng L⁻¹ in drinking water. The maximum concentration of NDMA in drinking water specified by the World Health Organization shall not exceed 100 ng L⁻¹. Although the regulatory level of NDMA in drinking water varies around the world, reducing the concentration of NDMA in disinfected water is still an important task in the drinking water treatment process in order to ensure the safety and health of the public.

NDMA is difficult to remove using physical methods such as air stripping, absorption or reverse osmosis due to its low molecular weight and polar functional groups [7,8]. In addition, NDMA has shown to be resistant to oxidation by ozone or other oxidants due to low second-order rate constants ($5.3 \times 10^{-2} \text{ M}^{-1} \text{ s}^{-1}$). Direct UV photolysis is the most commonly used method for the degradation of NDMA. Under the UV irradiation, NDMA has a strong absorption band in the range of 225–250 nm, which breaks the N-N bond and generates degradation products such as dimethylamine (DMA) and nitrite [9–12]. However, the high energy consumption and difficulty to completely mineralize limits its further application. Among the few technically effective NDMA remove methods, advanced reduction processes (ARPs) have recently emerged as a promising option.

As a new type of water treatment technology, ARPs combine UV activation techniques with reductants to generate active reducing radicals for the efficient degradation of target pollutants. Li first reported a systematic study on the rapid removal of monochloroacetic using UV-activated sulfite [13] and Vellanki proposed the concept of ARPs in the following year [14]. It has been proven that hydrated electrons (e_{aq}^-) and hydrogen atoms ($\bullet\text{H}$) are the main activate products formed during the ARPs, while other radicals such as sulfite radical anions ($\bullet\text{SO}_3^-$) and sulfur dioxide radical anions ($\bullet\text{SO}_2^-$) can also be generated, depending on the various activation methods (photolysis, radiolysis and sonolysis) and reductants (sulfite, dithionite and iodide). ARPs have attracted widespread attention in a short period of time due to their excellent performance on degrading various contaminants such as vinyl chloride, perchlorate, bromate, nitrate, 2,4,6-trichlorophenol and polyfluoroalkyl substances (PFAS) [15–18].

The reducing radicals generated via ARPs have a low redox potential, making it possible to attack the N-N bonds in NDMA molecules and decompose them into small fragments for further degradation. However, a systematic investigation on the reductive degradation of NDMA via ARPs is still lacking so far. Little is known about the influencing factors, pathways and mechanisms involved in this process. The major objectives of this study are therefore to (1) systematically investigate the kinetics of the NDMA degradation using the chosen UV/sulfite ARP and the impacts of some of the key parameters of the reaction system (i.e., reductant dosage, initial reaction pH, UV intensity and dissolved oxygen) on NDMA degradation; (2) determine the intermediates and final products generated from the reductive degradation; and (3) propose a possible degradation pathway of NDMA via UV/sulfite ARP.

2. Materials and Methods

2.1. Reagents and Solutions

A standard solution containing 1000 µg mL⁻¹ N-nitrosodimethylamine in methanol was purchased from Sino-pharm Chemical Reagent Co., Ltd., Shanghai, China. The isotope internal standard substance (NDMA-D6) was purchased from Sigma-Aldrich Co., Ltd., Shanghai, China. HPLC-grade methanol and acetonitrile were obtained from Aladdin Co., Ltd., Shanghai, China. High-purity nitrogen and oxygen were purchased from Messer Co., Ltd., Klaeford, Germany. Deionized water was used for dilution and analysis.

2.2. Batch Experiments

The NDMA standard solution was diluted with deionized water to the concentration of 0.1 mM in a 1 L volumetric flask and then placed in a closed photochemical reactor. The photochemical reactor consists of a low-pressure mercury lamp capable of emitting 254 nm UV light, a reaction vessel, a magnetic stirring system, a cooling water circulation system and a temperature control system. The pH value of the solution was adjusted to a certain value using phosphate-buffered solution before the reaction began. The nitrogen saturation and oxygen saturation conditions were achieved by introducing high-purity nitrogen and oxygen before the reaction began. The reactor was airtight and without any head space in order to avoid the impact of air and volatilization. A total of 10 mL of sample was taken at different reaction time points (0, 5, 10, 15, 20, 30, 40, 50, 60 min). The sample was filtered through 0.22 μm nylon fiber membrane and pressed into a brown glass vial for subsequent detection.

2.3. Analytical Methods

High performance liquid chromatography with a diode array detector (HPLC-DAD, Shimadzu Corp., Tokyo, Japan) was used to analyze NDMA and its degradation intermediates (dimethylamine and methylamine). The mobile phase consisted of water and acetonitrile with a flow rate of 1 mL min⁻¹. The gradient elution procedure was as follows: 20% acetonitrile to 60% acetonitrile within the initial 4 min and held at 60% acetonitrile within 2 min, then 60% acetonitrile to 20% acetonitrile in 3 min and maintained at 20% acetonitrile to re-equilibrate the column until the end of the run time at 12.8 min. The injection volume was 100 μL . The detection wavelength of the diode array detector was 228 nm.

The content of inorganic nitrogen (including nitrate and nitrite) and formate (HCOO^-) was analyzed using an anion chromatograph equipped with a conductivity detector (ICS-1000, DIONEX Co., Ltd., Sunnyvale, CA, USA). The content of ammonia was analyzed using salicylic acid spectrophotometry. The pH was detected using a pH Meter (PHS-3G, Leici Corp., Shanghai, China). The UV light intensities were measured and calculated using iodide/iodate spectrophotometry. The concentration of dissolved oxygen was determined via iodometry.

3. Results and Discussion

3.1. Comparison between Direct UV Photolysis and UV/Sulfite ARP

NDMA exhibits absorption peaks at 227 and 332 nm, which makes direct UV photolysis feasible for removing NDMA. However, the UV irradiation intensity required to achieve an acceptable removal efficiency of NDMA is considerable [19]. Preliminary experiments were conducted in order to compare the removal efficiency of NDMA between direct UV photolysis and the UV/sulfite ARP and the results are shown in Figure 1a. As can be seen, the removal efficiency of NDMA via the UV/sulfite ARP was much higher than that of direct UV photolysis under the same UV intensity (3 mW cm⁻²). A total of 94.40% of NDMA was removed via the UV/sulfite ARP at the end of the reaction, while only 45.48% of NDMA was removed via direct UV photolysis within the same reaction time. Moreover, increasing the UV intensity can significantly improve the removal efficiency of NDMA via direct UV photolysis. An NDMA removal efficiency of 93.29% was achieved via direct UV photolysis when the UV intensity increased to 12 mW cm⁻², which approaches the performance of the UV/sulfite ARP. The degradation of NDMA via direct UV photolysis and the UV/sulfite ARP followed pseudo-first-order kinetics, and the rate constants were calculated and are shown in Figure 1b. The k_{obs} of direct UV photolysis (3 mW cm⁻² UV intensity), direct UV photolysis (12 mW cm⁻² UV intensity) and UV/sulfite ARP were 0.0115 ± 0.002 , 0.0463 ± 0.010 and 0.0464 ± 0.011 min⁻¹, respectively, which was in accordance with the above discussion. Therefore, the degradation of NDMA via the UV/sulfite ARP may be a more effective and energy saving method than direct UV photolysis.

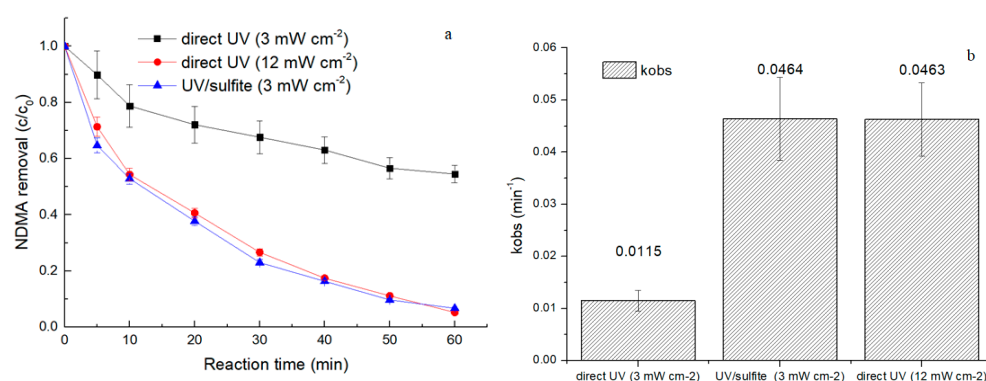


Figure 1. (a) The removal efficiency of NDMA via direct UV photolysis and UV/sulfite ARP. (b) Pseudo-first-order rate constant, k_{obs} . Experiment conditions: initial NDMA concentration = 0.1 mM; sulfite dosage = 0.2 mM; dissolved oxygen (DO) = 7.69 mg/L; and pH = 7.04.

3.2. Effect of pH

The pH value of the solution will affect the distribution of the reducing agent in water, as well as causing an interaction between free reducing radicals, thus affecting the degradation of the target pollutants via ARPs. Therefore, this study investigated the removal efficiency of NDMA via the UV/sulfite ARP under different pH conditions, and the results are shown in Figure 2.

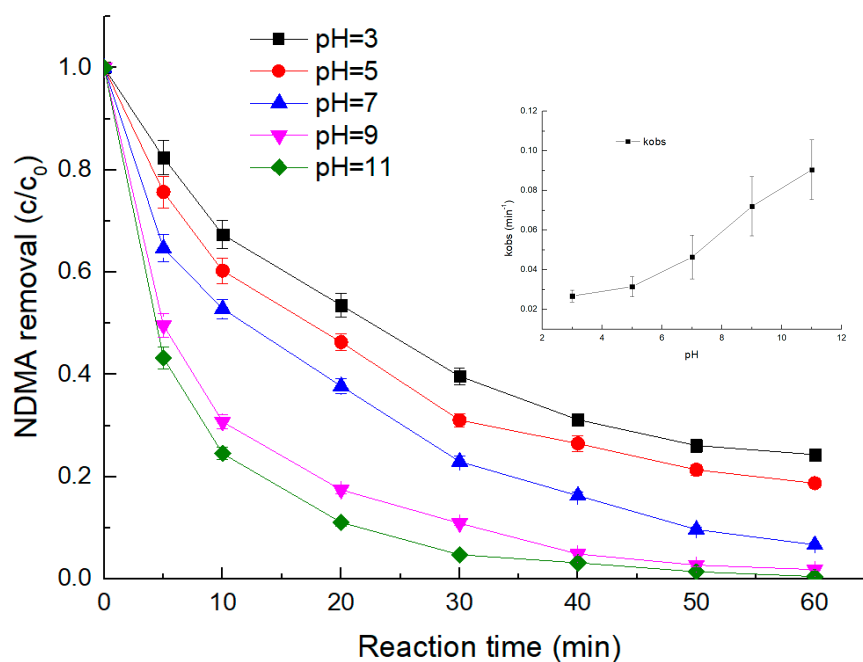


Figure 2. Effect of pH on NDMA removal efficiency and pseudo-first-order rate constant via UV/sulfite ARP. Experiment conditions: initial NDMA concentration = 0.1 mM, UV intensity = 3 mW/cm², sulfite dosage = 0.2 mM, and DO = 7.69 mg/L.

The results shown in Figure 2 indicated that the NDMA removal efficiency was favored under alkaline conditions by UV/sulfite ARP, while acidic conditions would inhibit the degradation of NDMA. The removal efficiency of NDMA increased from 21.57% to 66.79% within the initial 5 min of the reaction when the initial pH increased from 3 to 11. Moreover, NDMA was completely removed from the solution under alkaline conditions (pH = 9 or 11) after the 60 min reaction time, while only around 80% NDMA removal efficiency was achieved under acidic conditions at the end of the reaction. Generally, the distribution of sulfite species (SO_3^{2-} ; HSO_3^- and H_2SO_3) in the water changed with different pH.

The sulfite was the dominant species in alkaline conditions, while the bisulfite was the dominant species in acidic conditions [20]. The differences in the UV absorption ability of various sulfite species led to the generation of different reducing radicals, which affected the NDMA degradation via a radical–reaction mechanism. It is reported that bisulfite does not absorb considerable amounts of UV irradiation in the range of 225–300 nm. This absorbance behavior suggests that bisulfite does not generate reducing radicals like sulfite under the UV irradiation in this study (254 nm). As a result, the degradation mechanism of NDMA in acidic conditions is just direct UV photolysis. On the other hand, sulfite can produce more hydrated electrons and sulfite radicals by UV excitation in alkaline conditions, which promotes the degradation of NDMA. Previous studies have confirmed that sulfite radicals and hydrated electrons are the main products of the sulfite photodegradation. In acidic conditions, the concentration of hydrated electrons is 4–5 orders of magnitude lower than in alkaline conditions, which could explain how the pH affects the rate constant for NDMA removal [21]. In addition, the relationship between rate constants and pH also showed that there was a significant increase in the k_{obs} when the initial pH increased from 3 to 11. The k_{obs} for the initial pH of 3, 5, 7, 9 and 11 was 0.00267 ± 0.001 , 0.0315 ± 0.005 , 0.0464 ± 0.011 , 0.0720 ± 0.015 and $0.0905 \pm 0.015 \text{ min}^{-1}$, respectively. This result is consistent with Jung's study, which investigated the effect of pH on bromate degradation via the UV/sulfite ARP and found that the k_{obs} increased from 0.025 min^{-1} to 0.10 min^{-1} when the pH ranged from 3 to 11 [16].

3.3. Effect of Sulfite Dosage

According to the Beer–Lambert law, changing the dosage of the reducing agent will affect the absorption of UV irradiation, which results in a variation in the radical generation rate in ARPs. Thereby, the effect of the sulfite dosage on the removal efficiency of NDMA via the UV/sulfite ARP was investigated in this study. The results from Figure 3 showed that the removal efficiency of NDMA increased from 35.39% to 67.73% within 5 min of the reaction time when the sulfite dosage increased from 0.2 mM to 1 mM. The k_{obs} of different sulfite dosages (0.2 mM, 0.5 mM and 1.0 mM) were 0.0464 ± 0.011 , 0.0789 ± 0.014 and $0.0978 \pm 0.018 \text{ min}^{-1}$, respectively. According to Botlaguduru's research, the degradation of pollutants via ARPs included two pathways: (1) direct photolysis and (2) decomposition based on radical reactions [15]. If the direct photolysis is the dominant reaction pathway for the degradation of NDMA via ARPs, the higher sulfite dosage means that most of the UV will be absorbed by sulfite under certain UV intensities, which results in a corresponding reduction of UV used for the NDMA photolysis. But, if the radical reaction is the dominant reaction pathway, the higher sulfite dosage could accelerate the generation rate of reducing radicals and promote the degradation of NDMA. The results shown in Figure 3 indicate that the radical reaction is likely to be the dominate mechanism for the degradation of NDMA via the UV/sulfite ARP. In addition, much of the literature reported that an excessive sulfite dosage could decrease the UV light transmission, thus potentially inhibiting the removal of NDMA. However, at the level of sulfite dosage in this study, the transmittance of the solution is greater than 90%, which prevented any negative impact on the high sulfite dosage.

3.4. Effect of UV Intensity

The removal efficiency of NDMA via the UV/sulfite ARP under three levels of UV intensity (3 mW cm^{-2} , 6 mW cm^{-2} and 12 mW cm^{-2}) was investigated, and the results are shown in Figure 4. It can be seen that 35.39% of NDMA was removed within 5 min of the reaction via the UV/sulfite ARP under 3 mW cm^{-2} UV intensity, while 61.63% of NDMA removal efficiency was achieved within the same reaction time when the UV intensity increased to 12 mW cm^{-2} . The linear increase in the k_{obs} shown in Figure 4 indicated that an increase in UV light intensity leads to a proportional increase in the NDMA removal rate. According to Lee's study [9], the increase in UV intensity would lead to a proportional increase in the photolysis rate of NDMA and the radical formation rate from

sulfite. Therefore, the increase in the total removal rate of NDMA should be proportional to the increase in the UV intensity. A similar trend was found by Botlaguduru, who investigated the bromate removal via UV/sulfite ARPs [15]. It is worthwhile considering that increasing the UV intensity will increase the energy consumption and ultimately increase the treatment cost. However, almost 90% of NDMA removal efficiency can be achieved under all three of the UV intensity levels after 60 min of the reaction in this study. Therefore, it is necessary to choose an appropriate UV intensity to balance the NDMA removal efficiency and the treatment cost.

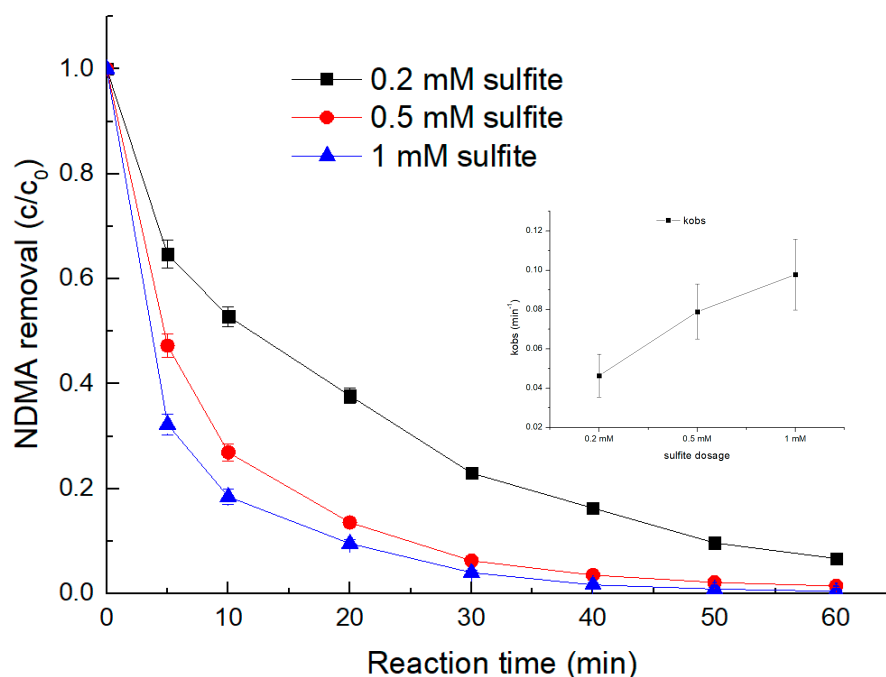


Figure 3. Effect of sulfite dosage on NDMA removal efficiency and pseudo-first-order rate constant via UV/sulfite ARP. Experiment conditions: initial NDMA concentration = 0.1 mM, UV intensity = 3 mW/cm^2 , pH = 7.04, and DO = 7.69 mg/L .

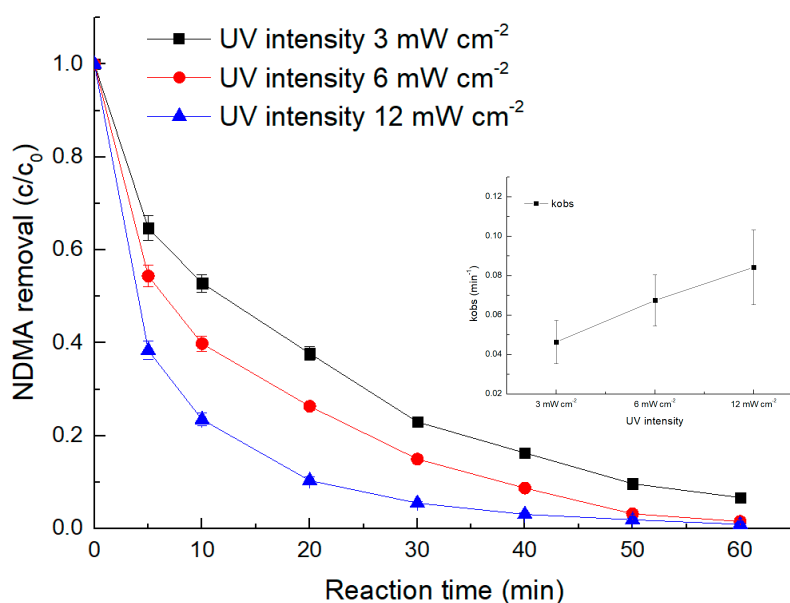


Figure 4. Effect of UV intensity on NDMA removal efficiency and pseudo-first-order rate constant via UV/sulfite ARP. Experiment conditions: initial NDMA concentration = 0.1 mM, sulfite dosage = 0.2 mM, pH = 7.04, and DO = 7.69 mg/L .

3.5. Effect of Dissolved Oxygen

The effect of dissolved oxygen on the removal efficiency of NDMA via the UV/sulfite ARP was investigated. The results from Figure 5 showed that 33.58% of NDMA was removed within 60 min of the reaction via the UV/sulfite ARP under oxygen saturation conditions, while almost 98.82% of NDMA removal efficiency was achieved within the same reaction time under nitrogen saturation conditions. In general, the presence of DO can substantially decrease the removal efficiency of contaminants in most ARP systems. This inhibitory effect was mainly due to the strong oxidizing superoxide radicals ($\bullet\text{O}_2^-$) generated by dissolved oxygen under the excitation of UV light, which could easily capture the reducing active radicals such as hydrated electrons, hydrogen radicals and sulfite radicals during the reaction. As mentioned above, the radical reaction was the dominant reaction pathway for NDMA degradation via the UV/sulfite ARP. The reduction of the reducing active radicals would inevitably affect the removal efficiency of NDMA. However, the opposite result was found; the presence of dissolved oxygen could increase the removal efficiency of the target pollutants. The promoting effect was attributed to the formation of oxidizing active radicals (such as $\bullet\text{OH}$ and $\bullet\text{SO}_4^-$) by the reaction between dissolved oxygen and sulfite radicals. The degradation of the target pollutants was achieved through the advanced oxidation pathways [20].

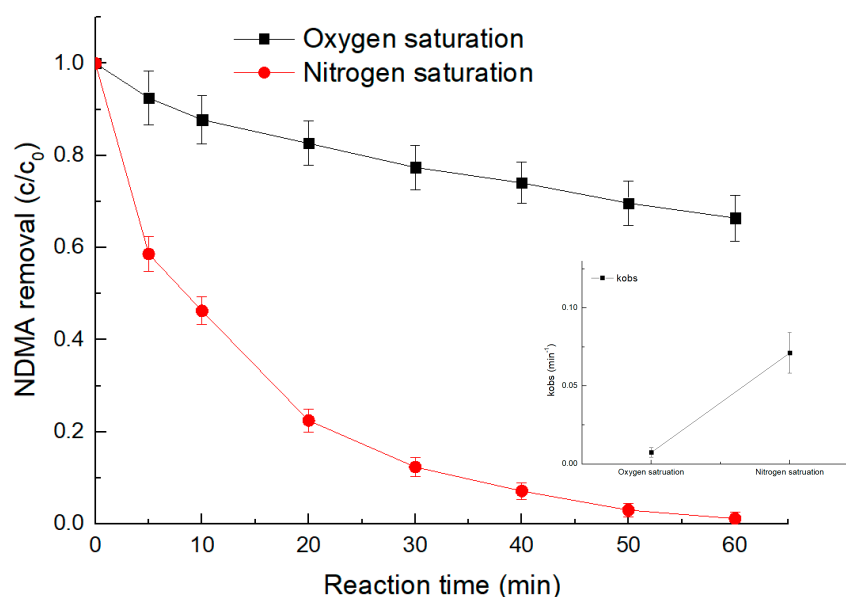


Figure 5. Effect of dissolved oxygen on NDMA removal efficiency and pseudo-first-order rate constant by UV/sulfite ARP. Experiment condition: Initial NDMA concentration = 0.1 mM, sulfite dosage = 0.2 mM, pH = 7.04, UV intensity = 3 mW/cm².

3.6. Degradation Products of NDMA by UV/Sulfite ARP

The degradation products of NDMA via UV/sulfite ARP were investigated and the results are shown in Figure 6. As NDMA decomposed using UV/sulfite ARP, dimethylamine (DMA) and nitrite were the main intermediates generated first, with a maximum concentration of 29.7 μM and 35.7 μM , respectively. Afterwards, DMA underwent further degradation and its concentration decreased continuously until it was completely removed at the end of the reaction. Methylamine (MA) was the intermediate product of DMA with a maximum concentration of 30.9 μM after 20 min of the reaction. Similarly to DMA, MA also underwent further degradation and was completely removed at the end of the reaction. However, formate and ammonia were the final degradation products of DMA and MA. Different to DMA or MA, their concentrations constantly increased with the prolongation of the reaction time. The concentrations of formate and ammonia were 186.5 μM and 88.9 μM at the end of the reaction. Moreover, it is worth noting that no formaldehyde and methanol

were detected during the whole reaction process, which was probably due to their higher reaction rate coefficients with radicals [19]. Similarly to DMA, nitrite underwent further degradation and was completely removed at the end of the reaction. Nitrogen (N_2) was likely to be the final degradation product of nitrite according to other researchers. However, it was difficult to quantitatively detect the formation concentration of nitrogen under our laboratory conditions. Moreover, nitrate was not detected in solution, which indicated that the reduction of nitrite was the predominant pathway.

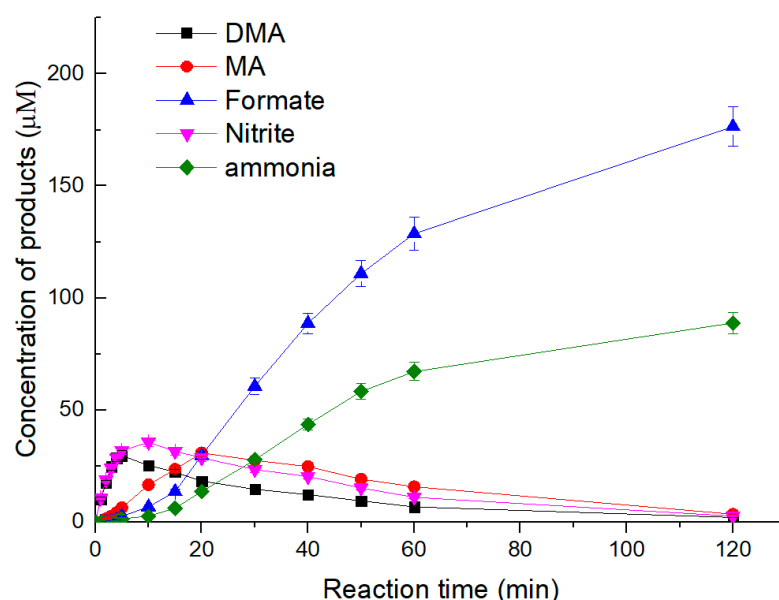
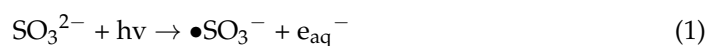


Figure 6. The concentration of products generated from the reductive degradation of NDMA via UV/sulfite ARP. Experiment conditions: initial NDMA concentration = 0.1 mM; sulfite dosage = 0.2 mM; UV intensity = 3 mW/cm²; DO = 7.69 mg/L; and pH = 7.04.

3.7. Reaction Mechanism

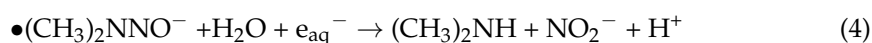
The generation of reducing radicals via the UV/sulfite ARP has been described in detail by many researchers. Ultraviolet radiation of sulfite generated sulfite anion radicals ($\bullet SO_3^-$) and hydrated electrons (e_{aq}^-) (Equation (1)). The hydrated electrons reacted with hydrogen ions to generate hydrogen radicals (Equation (2)). Hydrated electrons, sulfite radicals and hydrogen radicals were all reducing active substances that could participate in the degradation of NDMA, but hydrated electrons played a more critical role because of their higher standard reduction potential (−2.77 V) [22].



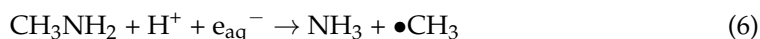
On the other hand, the ground state NDMA molecule in the solution was excited by UV and generated a transient adduct anion ($\bullet(CH_3)_2NNO^-$) (Equation (3)).



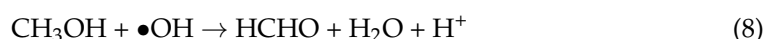
The attack occurred between the hydrated electron and $\bullet(CH_3)_2NNO^-$ and resulted in the cleavage of the N-NO bond. This reaction was confirmed as one of the key steps of NDMA degradation (Equation (4)) [23]. DMA and nitrite were the major intermediates in this reaction.



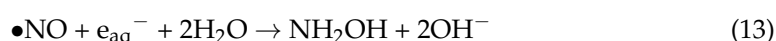
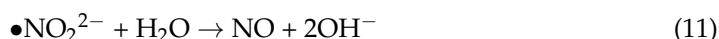
DMA could not absorb photons released by UV to undergo further decomposition due to the lack of nitroso functional groups. But, in this study, both DMA and nitrite were further degraded via the UV/sulfite ARP. Hydrated electrons attacked one of the C-N bonds in DMA molecules and caused its breakdown, thus producing methylamine and methyl radicals ($\bullet\text{CH}_3$) (Equation (5)). MA was continuously attacked by hydrated electrons, causing the destruction of the remaining C-N bond and the generation of ammonia and methyl radicals (Equation (6)).



As an intermediate product, the methyl radical was unstable and easily combined with a hydroxyl radical ($\bullet\text{OH}$) to form methanol, formaldehyde and ultimately a formate ion (Equations (7)–(9)). These processes could have occurred because of the higher reaction rate coefficients of methyl and methanol with a hydroxyl radical than a hydrated electron.



Simultaneously, nitrite was firstly attacked by a hydrated electron to form $\bullet\text{NO}_2^{2-}$, and then $\bullet\text{NO}_2^{2-}$ underwent hydrolysis to form nitric oxide (NO). Nitric oxide was further attacked by a hydrated electron to form a nitric oxide anion radical ($\bullet\text{NO}^-$). Part of $\bullet\text{NO}^-$ was reacted with a hydrated electron to form NH_2OH and then ammonia, while the rest of $\bullet\text{NO}^-$ was converted to $(\text{N}_2\text{O}_2)^{2-}$ through a dimeric reaction which became N_2O afterwards. N_2O could be further reacted with a hydrated electron to form N_2 as the final product (Equations (10)–(16)).



4. Conclusions

The UV/sulfite ARP was chosen to degrade NDMA and the effects of process variables including solution pH, sulfite dosage, UV intensity and dissolved oxygen on NDMA removal efficiency were investigated in this study. The following conclusions can be draw from this research:

- (1) The UV/sulfite ARP was an efficient and energy saving method for the reductive degradation of NDMA. An NDMA removal efficiency of 93.29% was achieved via the UV/sulfite ARP, while only 45.48% of NDMA was removed via direct UV photolysis within the same reaction condition.

- (2) The degradation of NDMA via the UV/sulfite ARP was favored under alkaline conditions. The removal efficiency of NDMA increased from 21.57% to 66.79% within the initial 5 min of the reaction when the initial pH increased from 3 to 11.
- (3) The degradation of NDMA via the UV/sulfite ARP followed pseudo-first-order kinetics. Both increasing the UV light intensity and sulfite dosage led to a proportional increase in the NDMA removal rate.
- (4) The presence of dissolved oxygen substantially decreased the removal efficiency of NDMA due to the formation of oxidizing superoxide radicals, which competed with NDMA by capturing the reducing active radicals during the reaction.
- (5) The final degradation products of NDMA via the UV/sulfite ARP were formate, ammonia and nitrogen. Some refractory intermediates such as DMA, MA and nitrite were completely decomposed via the UV/sulfite ARP.

Author Contributions: X.Z.: Investigation, visualization and writing original draft. S.W.: Chemical analysis and mathematical calculation. D.Z.: Formal analysis and review and editing. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data presented in this study are available on request from the corresponding author.

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

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