

Destruction of amphetamine in aqueous solution using gamma irradiation



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ABSTRACT

Amphetamine-type stimulants are among the most prevalent and widespread commonly abused drugs. Amphetamine and its derivatives were detected in aquatic environment. This study aimed to demonstrate experimentally the ability of γ -irradiation combined with persulfate anions ($S_2O_8^{2-}$) to degrade and mineralize the amphetamine in aqueous solution. An initial amphetamine concentration of 125 μ M in distilled water was completely degraded by a γ -ray dose of 2.8 kGy. Generation of the sulfate radical ($SO_4^{\cdot-}$) from the fast reaction of added $S_2O_8^{2-}$ with hydrated electrons (e_{aq}^- ; $k_{e_{aq}^-/S_2O_8^{2-}} = 1.1 \times 10^{10} M^{-1} s^{-1}$) improved the efficiency of amphetamine degradation and mineralization. A γ -ray dose of 0.667 and 0.350 kGy in the absence and presence of $S_2O_8^{2-}$ anions degraded 90% of the amphetamine, respectively. For γ -ray/free O_2 and γ -ray/ $S_2O_8^{2-}$ systems, 11.5 and 7 kGy was required for 50% amphetamine mineralization, respectively. Addition of HCO_3^- anions lowered the amphetamine degradation yield, whereas N_2 gas, SO_4^{2-} , and Cl^- anions had a negligible effect.

1. Introduction

At present, the quality of water treated by conventional processes in wastewater treatment plants is often unsatisfactory, and additional processes are required for adequate decontamination, as demonstrated by the detection of emerging organic contaminants using recently developed analytical methods (Aneggi et al., 2017; Catalá et al., 2015; Choi et al., 2016; Esplugas et al., 2002; Gogate and Pandit, 2004; Parsons, 2004; Valcárcel et al., 2012). Among the most efficient chemical processes used for water treatment are Advanced Oxidation Processes (AOPs) consisting of ozonation (O_3), photocatalysis, and UV-photolysis (Andreozzi et al., 1999; Legrini et al., 1993). AOPs are characterized by in situ formation of highly reactive, powerful oxidizing species such as hydroxyl radicals ($\cdot OH$; $E^\circ = 1.9$ V; Wardman, 1989), which play the primary role in the decontamination of various classes of organic contaminants in aqueous media, such as phenol (Alkhurajji et al., 2017), endocrine disrupting chemicals (Choi et al., 2016), pesticides (Hossain et al., 2013), dyes (Sun et al., 2013), and pharmaceutical compounds (Zheng et al., 2011).

Amphetamine-type stimulants are among the most prevalent and widespread commonly abused drugs (UNODC, 2016). These drugs are emerging contaminants in environmental aqueous media (Repice et al., 2013). Due to its continuous uncontrolled consumption and incomplete elimination by wastewater treatment plants, these drugs can enter and persist in aquatic environments (Boles and Wells, 2010; Postigo et al.,

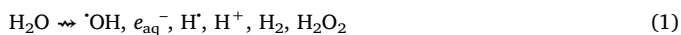
2011; Nuijs et al., 2011). Amphetamine and its derivatives have been detected in aquatic environments in ng/L to μ g/L concentrations. Although such low concentrations do not pose a significant direct threat to aquatic organisms (Peng et al., 2016), the effects of their bioaccumulation and their environmental and toxicological significance remain unknown (Catalá et al., 2015; Mendoza et al., 2014a, 2014b). However, these categories of organic contaminants can pollute aquatic systems including rivers and lakes (Mendoza et al., 2014a; Pal et al., 2013; Petrie et al., 2015), ground water (Jurado et al., 2012; Repice et al., 2013), and wastewater (Zuccato et al., 2009). Abused drugs have even been detected at very low concentrations (13–150 ng/L) in tap water (Catalá et al., 2015; Pal et al., 2013). However, the elimination of abused organic drugs by AOPs in aqueous solution has received little attention. Valcárcel et al. (2012) and Catalá et al. (2015) investigated the elimination of six different chemical classes of abused drugs, including amphetamine-type stimulants and cocaine, in natural fluvial waters by Photo-Fenton treatment. Meanwhile, Rodayan et al. (2014) highlighted the ability of ozone to remove abused drug residues from wastewater. UV_{254}/H_2O_2 processes were also tested for the removal of abused drugs (Russo et al., 2016), and chlorination of some amphetamine-type stimulants has been investigated in simulated and real drinking water (Huerta-Fontela et al., 2012).

During the last decade, there has been increasing interest in the $SO_4^{\cdot-}$ radical ($E^\circ = 2.43$ V; Wardman, 1989) as a powerful oxidizing agent, which can be efficiently used for decomposition of organic

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pollutants in aqueous solutions (Criquet and Karpel Vel Leitner, 2011). The $\text{SO}_4^{\cdot-}$ radical can be generated by combining $\text{S}_2\text{O}_8^{2-}$ anions with an AOP in aqueous solution i.e. photolysis (Criquet and Karpel Vel Leitner, 2009), ultra-sonic irradiation (Chen and Su, 2012), and high-energy radiation (Alkhuraji et al., 2017; Alkhuraji and Karpel Vel Leitner, 2016). Moreover, numerous studies have demonstrated the high efficacy of the $\text{SO}_4^{\cdot-}$ radical in aqueous solutions for remediation of pesticides and refractory compounds (Bao et al., 2016; Khan et al., 2013), as well as pharmaceutical compounds and endocrine disruptors (Huang et al., 2005; Roshani, Karpel vel Leitner, 2011). Furthermore, ionizing radiation processes have been adopted efficiently for water remediation as one of the AOPs (Han et al., 2012). The ionizing radiation generated from an electron beam accelerator or emitted from CO-60 absorbed by water, leads to water radiolysis, where $\cdot\text{OH}$ radical is formed (Eq. (1); Buxton et al. (1988)).



In this context, the main objective of this research was to evaluate for the first time the potential of γ -radiation as an AOP for the destruction and mineralization of amphetamine in aqueous solutions, as a model compound representing commonly abused drugs detected in environmental aqueous systems.

2. Experimental

2.1. Chemicals and solutions

Amphetamine ($\text{C}_9\text{H}_{14}\text{NCl}$; 98.5%) was purchased from Lipomed AG (Switzerland) and supplied by the Naif Arab University for Security Sciences. Methanol (HPLC plus-gradient grade) was purchased from AppliChem Panareac (Germany). Sodium persulfate ($\text{Na}_2\text{S}_2\text{O}_8$; 98%) was purchased from Acros organic (USA). Sodium chloride (NaCl , 99%), sodium bicarbonate (NaHCO_3 , 99%), sodium sulfate (Na_2SO_4 , 98%), sodium nitrate (NaNO_3 , 99%), acetic acid, and orthophosphoric acid (85%) were purchased from Sigma-Aldrich (USA). All chemicals used in this research were used as received without further purification. A Milli-Q device (Elix Technology) inside Integral 5 Millipore was used for the preparation of stock and samples solutions.

2.2. Ionization source

Sample solutions of amphetamine were irradiated over the range 0.025–70 kGy using a Co-60 cell 220 (MDS-Nordion, Canada) at a dose rate of 3.9 kGy/h. The source installed at the King Abdulaziz City for Science and Technology (KACST), and was calibrated using aqueous ferrous sulfate (Fricke solution; ASTM Standard E1026, 2013).

2.3. Analytical methods

Initial and residual concentrations of amphetamine were optimized and determined by ultra-performance liquid chromatography (UPLC) using a Nexera X2 (Shimadzu). UV detection was performed at 210 nm using a Raptor Biphenyl column (5 μm , 50 \times 2.1 mm; Restek) at a flow rate of 0.3 mL/min, an injection volume of 30 μL , and a mobile phase containing 20% methanol and 80% ultra-pure water acidified with 0.1% orthophosphoric acid. The TOC content was determined using a Shimadzu TOC analyzer (TOC-L) connected to an ASI-L auto-sampler system.

2.4. Experimental procedures

Samples of aqueous amphetamine solution ($125 \pm 6 \mu\text{M}$) to be irradiated were placed in 40 mL or 2 mL vials with airtight caps and Teflon-based silicon septa. Measurements were conducted immediately after irradiation. The initial concentration of dissolved oxygen (γ -ray/

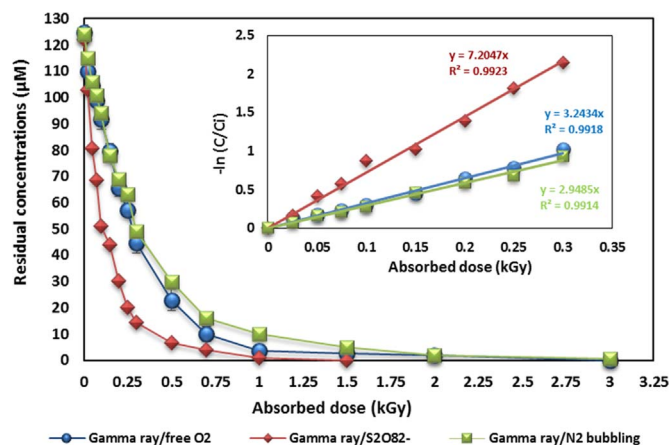


Fig. 1. Degradation of amphetamine following the absorption of different doses at $\text{pH}_i = 4.8$ and $[\text{O}_2]_i = 7.6 \text{ mg/L}$.

free O_2) was $7.6 \pm 0.9 \text{ mg/L}$ (if not stated otherwise), and the pH was unadjusted ($\text{pH}_i \approx 4.8$). To remove dissolved oxygen, samples were bubbled with nitrogen gas (N_2). To ensure reproducibility and reliability, γ -ray/free O_2 experiments were performed in triplicate.

2.5. Dose constant and the radiation chemical yield

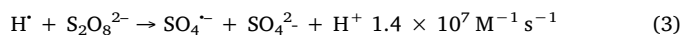
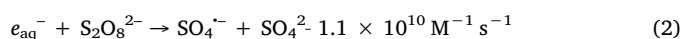
Radiation chemical yield (G-value, $\mu\text{mol/J}$), dose constant (k , kGy^{-1}), and absorbed dose for 50% and 90% elimination (D_{50} ; D_{90}) are useful parameters for assessing the effectiveness of the radiolytic degradation of organic contaminants, and were determined as described previously (Khan et al., 2015).

3. Results and discussion

3.1. Radiolytic destruction of amphetamine

Aqueous solutions of amphetamine ($125 \pm 6 \mu\text{M}$) were subjected to γ -irradiation at an absorbed dose of 0.025–3 kGy in the absence or presence of $\text{S}_2\text{O}_8^{2-}$ anions ($1500 \mu\text{M}$). Fig. 1 shows the destruction of amphetamine as a function of absorbed dose.

The degradation of amphetamine was initiated by the main primary products of water radiolysis ($\cdot\text{OH}$, H^{\cdot} , $e_{\text{aq}}^{\cdot-}$), notably $\cdot\text{OH}$ radical, which is in accordance with previous results reported for other aromatic organic contaminants (Alkhuraji et al., 2017; Sánchez-Polo et al., 2009). The residual concentration of amphetamine decreased immediately with increasing absorbed dose. Amphetamine was completely degraded in water at an absorbed dose over 2.8 kGy. However, the absorbed dose required to completely remove amphetamine was reduced by approximately 50% when the $\text{SO}_4^{\cdot-}$ radical was generated by the rapid reaction between added $\text{S}_2\text{O}_8^{2-}$ anions and reducing species (H^{\cdot} , $e_{\text{aq}}^{\cdot-}$; Eqs. (2) and (3); Buxton et al. (1988)). This improvement could be attributed to the simultaneous presence of the two radicals $\cdot\text{OH}$ and $\text{SO}_4^{\cdot-}$ (Eqs. (1) and (2)).



Based on the G-value of H^{\cdot} produced from water radiolysis and the reaction kinetics (k , $\text{M}^{-1} \text{ s}^{-1}$) for H^{\cdot} radical with $\text{S}_2\text{O}_8^{2-}$ anions compared with the G-value and k for $e_{\text{aq}}^{\cdot-}$, the reaction between $e_{\text{aq}}^{\cdot-}$ and $\text{S}_2\text{O}_8^{2-}$ anions appeared to be predominate in the radiolytic system. In addition, the results indicated no significant difference in amphetamine removal efficiency when solutions were bubbled with N_2 . This could indicate that there is no direct effect of $e_{\text{aq}}^{\cdot-}$ on the elimination of amphetamine. As shown in the inset of Fig. 1, the degradation kinetics of amphetamine (the relationship between the absorbed dose and $-\ln$

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