

PhD Theses

**IONIZING RADIATION INDUCED DEGRADATION OF
PHENYLUREAS IN AQUEOUS SOLUTION**

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1. Introduction and objectives

Different pollutants may enter the environment due to human activities that can cause air, water and soil pollution via their different physical and chemical interactions. These pollutants present a global risk for the ecosystems. Concerning water pollution, trace amounts of pesticides and antibiotics represent the highest risks for our surroundings.

Physical, chemical and biological methods are applied for the removal of contaminants in waters during traditional wastewater treatment. However, these methods can be applied only with low efficiency for elimination of certain organic pollutants, thus the researchers focused on new techniques to be developed. These techniques are called Advanced Oxidation Processes (AOP) in which highly reactive radicals, mainly hydroxyl radicals ($\cdot\text{OH}$) decompose the organic contaminants. High-energy ionizing radiation treatment is studied as AOP in the Department of Radiation Chemistry (Hungarian Academy of Sciences, Centre for Energy Research), in this technique primary reactive radicals forming during water radiolysis ($\cdot\text{OH}$, e_{aq}^- and $\text{H}\cdot$) induce the chemical transformations of the solutes.

The phenylureas belonging to the class of herbicides are applied in large quantities mainly in the agricultural sector, therefore it is not surprising that they can be detected as micropollutants in the environment. Their degradability has already been investigated by a number of AOP, nevertheless, we have little information about the effects of high energy ionizing radiation on their aqueous solutions. In this work ionizing radiation induced decomposition of three phenylureas, fenuron, monuron and diuron was studied in dilute aqueous solutions.

Our main goals were the comparison of degradation efficiencies of these three compounds and following up of their degradation processes. In addition, the reactions of reactive radicals formed in water radiolysis were separately studied under different conditions. The relationship between the environmental impact of degradation products and H_2O_2 formed during water radiolysis was also investigated.

2. Background

The phenylurea herbicides are used as total and selective weedkillers in agricultural crops (citrus, asparagus, cotton, beans, maize, wheat, bushes and fruits) and non-agricultural fields like along railroads and highways. They have long lifetime in the environment and they can be detected both in surface waters and ground waters¹²³. The compounds studied, fenuron, monuron and diuron belong to this class. They are structurally similar compounds consisting of an aromatic ring and a carbamide sidechain. They differ in the number of chlorine atoms on the aromatic ring (Fig. 1).

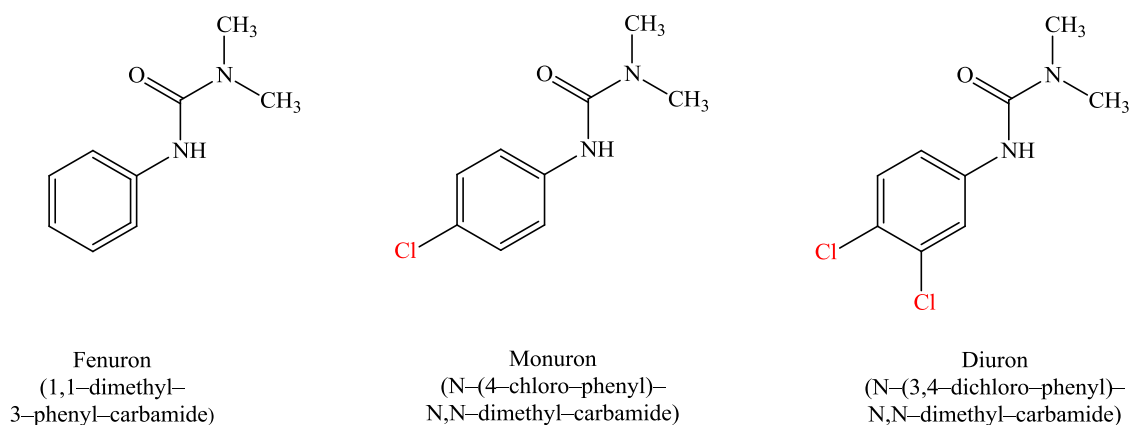


Figure 1: Structures of selected pesticides

The degradability of phenylureas have already been studied by some AOPs. During their chemical degradation isocyanates may form from zwitterion intermediers through a stepwise mechanism⁴. According to Salvestrini and his co-workers zwitterion forms which gives phenyl isocyanate derivative and dimethylamine⁵ by acid-base catalysis. The hydrolysis of phenyl isocyanate results in 3,4-dichloroaniline that is the only product detectable by HPLC technique. Hydrolysis is a very fast process and phenyl isocyanate derivatives cannot be detected by liquid chromatography due to their fast transformations.

¹ Thurman, E.M., Bastian, K.C., Mollhagen, T., 2000. Occurrence of cotton herbicides and insecticides in playa lakes of the high Plains of West Texas. *Sci. Total Environ.* **248**, pp. 189–200.

² Gerecke, A.C., Canonica, S., Müller, S.R., Schaerer, M., Schwarzenbach, R.P., 2001a. Quantification of dissolved natural organic matter (DOM) mediated phototransformation of phenylurea herbicides in lakes. *Environ. Sci. Technol.* **35**, pp. 3915–3923.

³ Gerecke, A.C., Tixier, C., Bartels, T., Schwarzenbach, R.P., Müller, S.R., 2001b. Determination of phenylurea herbicides in natural waters at concentrations below 1 ng l⁻¹ using solid-phase extraction, derivatization, and solid-phase microextraction gas chromatography–mass spectrometry. *J. Chromatogr. A* **930**, pp. 9–19.

⁴ Williams, A., Jencks, W.P., 1974. Urea synthesis from amines and cyanic acid: kinetic evidence for a zwitterionic intermediate. *J. Chem. Soc. Perkin Trans. 2*, pp. 1753–1759.

⁵ Salvestrini, S., Di Cerbo, P., Capasso, S., 2002. Kinetics of the chemical degradation of diuron. *Chemosphere* **48**, pp. 69–73.

Sunlight and UV induced photochemical decomposition of phenylureas have been also studied in the presence of oxygen⁶. The main degradation products were phenol type molecules in aqueous media. Biphenyl type compounds with a very low yield⁷ formed also during sunlight induced photolysis of chlorine-containing phenylureas via the coupling of two molecules.

In O₃ treatment⁸ of fenuron, monuron and diuron the values of the initial degradation rate coefficients changed between 0.6 (± 0.02) and 10.6 (±0.2) × 10⁸ mol dm⁻³ s⁻¹. This method was the most effective in the case of fenuron. Relationship was established between the number of electron-withdrawing chlorine atoms and the efficiency of this method: the efficiency decreased with increasing chlorine atoms. Similar conclusion was obtained during H₂O₂/O₃ induced decomposition of halogenated phenylureas⁹.

Degradation of diuron and monuron has been also studied by photo-Fenton, as well as the efficiency of this method was compared with other AOPs^{10,11}. The amount of Fe³⁺ and H₂O₂ was very important in the treatment because their dosage can influence the efficiency in the degradation of monuron and diuron: the reaction can be inhibited above a given concentration of additives. The technique was more effective compared to UV photolysis and H₂O₂ photolysis.

The high energy ionizing radiation induced degradation (or radiolysis) as an AOP has the advantage that there is no need for chemical additives. The highly reactive species from water radiolysis are responsible for the chemical transformations in the solution. Radiolysis proved to be very effective for removal of several organic pollutants in the environment (e.g. halogenated hydrocarbons, aromatic hydrocarbons etc.)¹².

⁶ Kotzias, D., Korte, F., 1981. Photochemistry of phenylurea herbicides and their reactions in the environment. *Ecotoxicol. Environ. Saf.* **5**, pp. 503–512.

⁷ Tanaka, F.S., Hoffer, B.L., Wien, R.G., 1985. Detection of halogenated biphenyls from sunlight photolysis of chlorinated herbicides in aqueous solutions. *Pest Manag. Sci.* **16**, pp. 265–270.

⁸ Kovács, K., Farkas, J., Veréb G., Arany, E., Simon, G., Schrantz, K., Dombi, A., Hernádi, K., Alapi, T., 2016. Comparison of various advanced oxidation processes for the degradation of phenylurea herbicides. *J. Environ. Sci. Health, Part B* **51** (4), pp. 205–214.

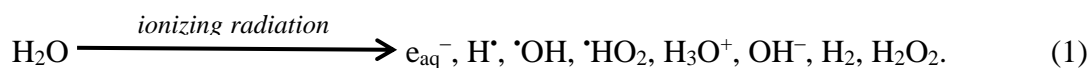
⁹ Tahmasseb, L.A., Nélieu, S., Kerhoas, L., Einhorn, J., 2002. Ozonation of chlorophenylurea pesticides in water: reaction monitoring and degradation pathways. *Sci. Total Environ.* **291**, pp. 33–44.

¹⁰ Bobu, M., Wilson, S., Greibrokk, T., Lundanes, E., Siminiceanu, I., 2006. Comparison of advanced oxidation processes and identification of monuron photodegradation products in aqueous solution. *Chemosphere* **63**, pp. 1718–1727.

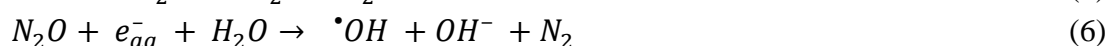
¹¹ Oturan, M.A., Oturan, N., Edelahe, M.C., Podvorica, F.I., El kacemi, K., 2011. Oxidative degradation of herbicide diuron in aqueous medium by Fenton's reaction based advanced oxidation processes. *Chem. Eng. J.* **171**, pp. 127–135.

¹² Getoff, N., 2002. Factors influencing the efficiency of radiation-induced degradation of water pollutants. *Radiat. Phys. Chem.* **65**, pp. 437–446.

Reactive species and molecular products form during the radiolytic decomposition of water (1). Hydroxyl radical ($\cdot\text{OH}$), hydrated electron (e_{aq}^-) and hydrogenatom ($\text{H}\cdot$) have contribution to the degradation processes.



These highly reactive species can be converted to so-called secondary radicals by adding appropriate additives (scavengers) so the individual radical reactions can be investigated easily. In aerated solutions e_{aq}^- and $\text{H}\cdot$ react with dissolved oxygen forming perhydroxyl/superoxide radical pair ($\text{HO}_2\cdot/\text{O}_2^{\cdot-}$) (2–3), thus the main reactive agents are $\cdot\text{OH}$ and the $\text{HO}_2\cdot/\text{O}_2^{\cdot-}$ radical pair. To study the reactions of the $\text{HO}_2\cdot/\text{O}_2^{\cdot-}$ radical pair, sodium formate is added to the sample and it is bubbled by O_2 (4–5). In deoxygenated media $\cdot\text{OH}$ and e_{aq}^- are the main reaction partners when the solution is saturated by N_2 . Under oxidizing conditions the reactions of $\cdot\text{OH}$ can be followed in N_2O saturated solutions ($c(\text{N}_2\text{O}) = 2.5 \times 10^{-2} \text{ mol dm}^{-3}$). Both e_{aq}^- and $\text{H}\cdot$ can be transformed to $\cdot\text{OH}$ (6–7). $0.2\text{--}1 \text{ mol dm}^{-3}$ *tert*-butanol is added to the samples when the e_{aq}^- reactions are studied. In reaction (8) $\cdot\text{OH}$ can be converted to a radical (2,2-dimethyl-2-hydroxy-ethyl radical), that does not react with the most organic compounds (8).



3. Methods

Decomposition of three phenylureas (fenuron, monuron and diuron) was studied using high-energy ionizing radiation. The transient intermediates were followed up by pulse radiolysis, γ -radiolysis experiments were performed to study the end-products. The experiments were carried out at room temperature.

The pulse radiolysis experiments were conducted by Tesla Linac LPR-4 type linear electron accelerator (Hungarian Academy of Sciences, Centre for Energy Research, Department of Radiation Chemistry) which supplies 800 ns pulses of 4 MeV energy electrons. The reactions of target compounds were studied with $\cdot\text{OH}$ and e_{aq}^- . The absorption spectra of the transient intermediates were calculated under different conditions and the second order rate coefficients were measured.

γ -radiolysis experiments were done by a panoramic type Co^{60} γ -source (SLL-01, (Institute of Isotopes Co. Ltd., Budapest). The efficiency of the decomposition was studied by sum parameters often applied in water and wastewater characterization. The end-products were followed up by UV spectrometry observing the effect of each radical in the degradation processes. Chemical oxygen demand (COD), total organic carbon (TOC) and total nitrogen (TN) contents were measured as the function of increasing dose. Adsorbable organic halogens (AOX) and chloride release measurements were carried out under different conditions in the cases of diuron and monuron to study dechlorination processes. The AOX experiments were conducted in the Laboratory of Research Group of Environmental Chemistry, Faculty of Science and Informatics, University of Szeged. The environmental impact of the starting molecules and products was analyzed by *Vibrio fischeri* testorganism. The H_2O_2 content of the samples was measured by Spectroquant[®] H_2O_2 -test. The separation and identification of end-products were performed by HPLC-MS/MS.

4. Results

Ionizing radiation induced reactions of three compounds (fenuron, monuron and diuron) were studied in dilute aqueous solutions. The efficiency of decomposition was compared using sum parameters. Degradation mechanisms were suggested based on the results obtained by pulse radiolysis and HPLC–MS/MS. Finally, relationship was established between H_2O_2 formed and the toxicity of the samples. Air saturated samples were irradiated in most of the experiments, so the effects of ionizing radiation were investigated on pesticides studied under real conditions. The individual radical reactions were also studied under different conditions, thereby, a comprehensive picture was obtained about the decomposition processes.

Pulse radiolysis measurements showed similar results for three pesticides both under oxidizing and reducing conditions which are depicted on the example of diuron (Fig. 2). The transient intermediates were examined under oxidizing conditions on the A and B parts and under reducing conditions on the C part of Fig. 2. Under oxidizing conditions a broad absorption band was observed between 300 and 400 nm that can refer to formation of hydroxy cyclohexadienyl type radicals. Based on these results it can be assumed that the main reaction is hydroxylation.

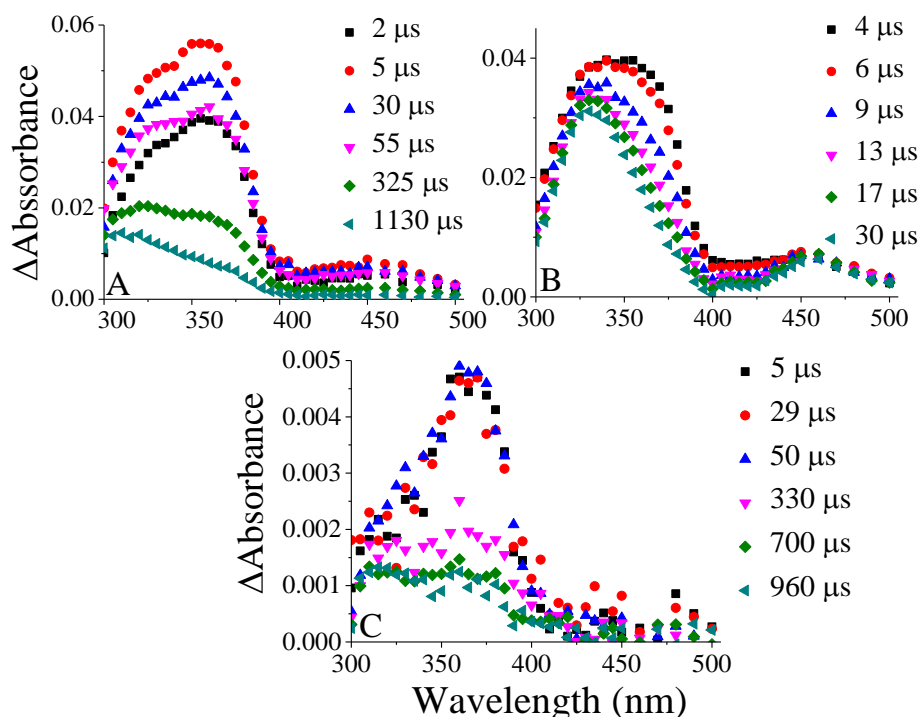


Figure 2.: Transient absorption spectra obtained in the reaction of diuron (A) with $\cdot\text{OH}$, (B) with $\cdot\text{OH}$ and $0.1 \times 10^{-3} \text{ mol dm}^{-3} \text{ K}_3[\text{Fe}(\text{CN})_6]$, (C) with e_{aq}^- ; $c_0 = 0.1 \times 10^{-3} \text{ mol dm}^{-3}$; dose/pulse: 20 Gy.

The rate coefficients calculated decreased with increasing number of chlorine atoms: fenuron > monuron > diuron. In e_{aq}^- reactions the transient intermediate has absorption band between 300 and 400 nm that can imply the presence of cyclohexadienyl type radicals. Based on the rate coefficients, the presence of electron-withdrawing chlorine atoms attached to aromatic ring increased significantly the rate of the nucleophile reaction.

From the previous results it can be concluded that the main reaction type is hydroxylation, in these reactions hydroxylated derivatives of the starting molecules are produced. Formation and decay of these products were confirmed by UV spectrometry and HPLC-MS/MS measurements. On the UV spectra the new absorption band that appeared during radiolysis can refer to the presence of these products under all conditions when the oxidizing $\cdot\text{OH}$ has contribution to degradation reactions. Singly and double hydroxylated products can form during hydroxylation in the cases of the three compounds. Besides the aromatic ring, $\cdot\text{OH}$ may attack also the terminal methyl group on carbamide sidechain. Reductive dechlorination may also happen in the cases of chlorinated compounds. The degradation mechanism of the pesticides studied is shown on the example of diuron (Fig. 3) based on pulse radiolysis and HPLC-MS/MS results.

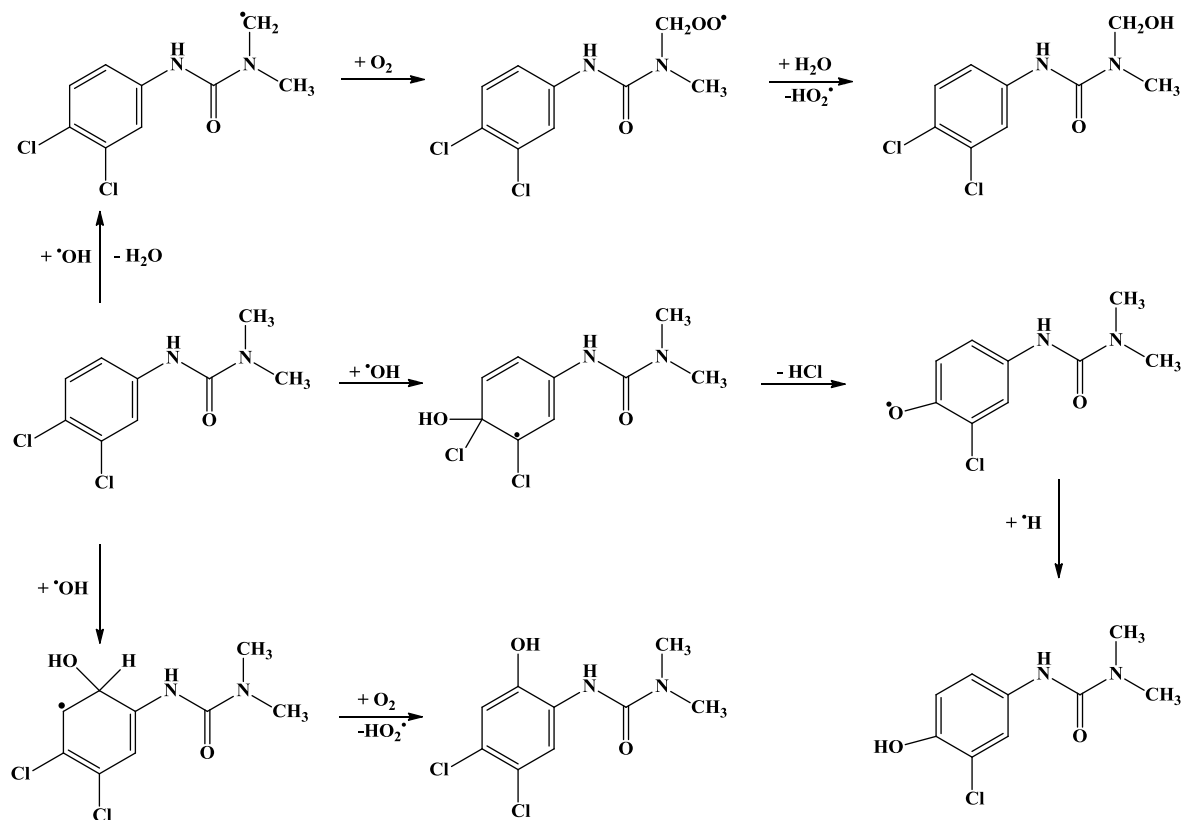


Figure 3.: Initial degradation reactions of diuron

The rate of chloride release and changes in the adsorbable organic halids (AOX) were measured to characterize the dechlorination reactions. The same results were obtained by the two methods. In chloride release experiments the effect of each radicals was also analyzed under different conditions the degree of the alteration was the most intensive in aerated solutions (0.15 és 0.10 $\mu\text{mol J}^{-1}$ for diuron and monuron, respectively). The e_{aq}^- as reducing agent has some contribution to dechlorination reactions: the presence of two electron-withdrawing chlorine atoms resulted in fourfold increase in the yield in diuron containing samples (0.08 $\mu\text{mol J}^{-1}$) compared to monuron containing solutions (0.02 $\mu\text{mol J}^{-1}$).

The rates of oxidation and mineralization characterized by COD and TOC, and the oxidation efficiencies were calculated. The oxidation efficiency is expressed by the ratio of the number of moles of O_2 incorporated into the products and that of the number of mols of $\cdot\text{OH}$ injected into the solutions. Both in oxidation and in mineralization processes decline was observed with increasing dose. The rate of oxidation and mineralization, as well as the oxidation efficiency increased with increasing numbers of chlorine atoms in the molecules. Investigating COD-dose dependence, the deviation from linearity was noticed at 1 kGy or higher dose. It can imply that small molecular mass organic acids form following ring-opening fragmentation. The oxidation of these organic acids proved to be a more difficult task. Comparing COD and TOC results, the degree of decrease was higher in the case of oxidation as the oxygen-to-carbon ratio increased considerably already at low doses when TOC-content changed only slightly. The total nitrogen content (TN) of the solutions decreased slowly compared to TOC, the nitrogen remained in liquid phase in the oxidized samples.

Table 1.: Oxidation and mineralization of compounds studied

	Fenuron	Monuron	Diuron
$\Delta\text{KOI}/\text{dose} \text{ (mg dm}^{-3} \text{ kGy}^{-1}\text{)}$	6	10	12
<i>Oxidation efficiency</i>	0.7	1.1	1.3
$\Delta\text{TOC}/\text{dose} \text{ (mg dm}^{-3} \text{ kGy}^{-1}\text{)}$	1.39	1.33	1.28

Relationship between the toxicity of each sample and H_2O_2 formed during the treatment was established. The results are illustrated on the example of fenuron (Fig. 4). H_2O_2 concentration monotonously increased in the function of the dose, the maximum concentration measured was $\sim 3 \times 10^{-4} \text{ mol dm}^{-3}$ at 2.5 kGy. This high H_2O_2 concentration can induce 50 % inhibition on *Vibrio fischeri* testorganism. The toxicity results showed almost 100 % inhibition in irradiated samples, which can be the consequence of end-products and H_2O_2 . Accordingly,

the environmental impact of the end-products can be examined with difficulty: H_2O_2 removal is a very important sample preparation step prior to toxicity tests. Catalase was applied for H_2O_2 removal, then H_2O_2 -content and toxicity of the samples were measured. Low concentration of H_2O_2 remained in the solutions after adding catalase, but its inhibitory effect was negligible in ecotoxicological test. Thus, the toxic effect of the organic end-products was examined after adding catalase. The results indicated that compounds with toxicity higher than that of the starting compounds formed at low doses. The toxicity disappeared at higher doses.

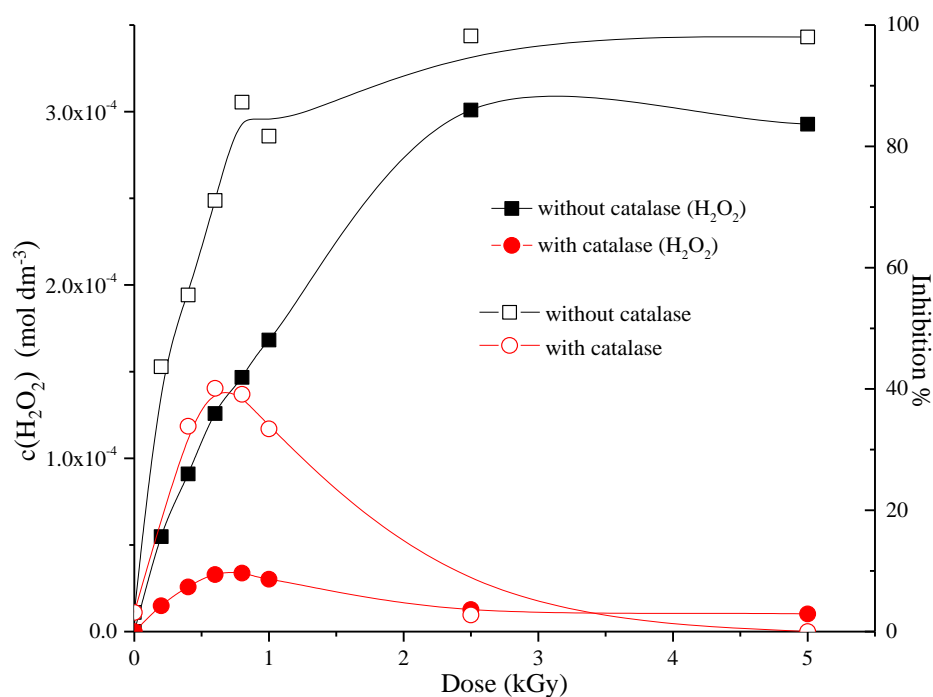


Figure 4.: Absorbed dose– H_2O_2 concentration and absorbed dose–inhibition curves in $0.1 \times 10^{-3} \text{ mol dm}^{-3}$ fenuron solutions.

5. Theses of PhD dissertation

1. **In aqueous solutions the pesticides studied, fenuron, monuron and diuron can be decomposed by radiolysis with the highest efficiency in the presence of dissolved O₂.** In $\cdot\text{OH}$ reactions carbon centered radicals form that transform to organic peroxy radicals reacting with dissolved oxygen. The further reactions result in the formation of peroxides, phenols, etc, but ring fragmentation also proceeds. The hydrated electron and hydrogen atom, forming also during water radiolysis, in reacting with dissolved oxygen are converted to superoxide/perhydroxyl radical pair. These species increase both the rate of oxidation and the efficiency of the decomposition. The yield of chloride release was the highest in aerated solutions. (Publications 1, 2 and 3)
2. **The reaction between the substrate and the hydroxyl radical has the highest contribution to the degradation as it was shown in pulse radiolysis experiments.** This result was confirmed based on the identified products as well. (Publications 1, 2 and 3)
3. **In $\cdot\text{OH}$ reactions mainly hydroxy-cyclohexadienyl radicals form in the first step.** On the transient absorption spectra the absorption band of the produced species appeared between 300 and 400 nm with $\sim 3000\text{--}5000 \text{ dm}^3 \text{ mol}^{-1} \text{ cm}^{-1}$ maximum molar absorption coefficient, which is characteristic to hydroxy-cyclohexadienyl radicals. (Publications 1, 2 and 3)
4. **Pulse radiolysis results confirmed that the presence of chlorine atoms on the aromatic ring increases the rate of nucleophile attack in hydrated electron reactions: two times higher rate coefficients were measured for monuron and ten times higher for diuron, than for fenuron.** Additionally, in chloride release measurements the calculated yield was four times higher in diuron containing samples due the higher electron-withdrawing effect of two chlorine atoms than in samples with monuron. (Publications 2 and 3)
5. **During radiolysis of chlorinated compounds the decrease of adsorbable organic halids content and the increase of chloride release take place with the same rate in**

aerated solutions. The released chlorine atoms stabilize in the form of chloride ion.
(Publications 2 and 3)

6. **Hydrogen peroxide (H₂O₂) formed during radiolysis is found to disturb the ecotoxicological tests, so H₂O₂ removal, e.g. by catalase, is a necessary step before the tests. The toxicity of the degradation products can be measured after H₂O₂ removal.** At low doses some of the products are more toxic than the starting molecule, the toxic properties of the samples disappear at high doses. (Publication 1)

6. Publications

a) Publications that form the basis of the PhD theses

1. Kovács, K., Mile, V., Csay, T., Takács, E., Wojnárovits, L., 2014. Hydroxyl radical–induced degradation of fenuron in pulse and gamma radiolysis: kinetics and product analysis. *Environmental Science and Pollution Research* **21**, pp. 12693–12700.
IF: 2.76 Citation: 5 (independent 1)
2. Kovács, K., He, S., Mile, V., Csay, T., Takács, E., Wojnárovits, L., 2015. Ionizing radiation induced degradation of diuron in dilute aqueous solution. *Chemistry Central Journal* **9**, DOI:10.1186/s13065-015-0097-0.
IF: 2.552 Citation: 4 (independent 1)
3. Kovács K., He, S., Mile, V., Földes, T., Pápai, I., Takács, E., Wojnárovits, L., 2016. Ionizing radiation induced degradation of monuron in dilute aqueous solution. *Radiation Physics and Chemistry* **124**, pp. 191–197.
IF: 1.207 Citation: 2

b) Further articles

4. Illés, E., Tegze, A., Kovács, K., Sági, Gy., Papp, Z., Takács, E., Wojnárovits, L., 2017. Hydrogen peroxide formation during radiolysis of aerated aqueous solutions of organic molecules. *Radiation Physics and Chemistry* **134**, pp. 8–13.
IF: 1.207 Citation: 0
5. Sági, Gy., Kovács, K., Bezsenyi, A., Csay, T., Takács, E., Wojnárovits, L., 2016. Enhancing the biological degradability of sulfamethoxazole by ionizing radiation treatment in aqueous solution. *Radiation Physics and Chemistry* **124**, pp. 179–183.
IF: 1.207 Citation: 3 (independent 3)
6. Mile, V., Harsányi, I., Kovács, K., Földes, T., Takács, E., Wojnárovits, L., 2017. Hydroxyl radical induced transformation of phenylurea herbicides: A theoretical study. *Radiation Physics and Chemistry* **132**, pp. 16–21.
IF: 1.207 Citation: 0
7. Alapi, T., Simon, G., Veréb, G., Kovács, K., Arany, E., Schrantz, K., Dombi, A., Hernádi, K., 2015. Toxicology aspects of the decomposition of diuron by advanced oxidation processes. *Hungarian Journal of Industry and Chemistry* **43 (1)**, pp. 25–32.
IF: 0 Citation: 0

8. Alapi, T., Simon, G., Kovács K., 2015. Endokrin hatású fenil-urea peszticidek eltávolítása kiegészítő víztisztítási módszerekkel. *Magyar Kémikusok Lapja* **70**:(10) 314–318.

IF: 0

Citation: 0

9. Kovács, K., Farkas, J., Veréb, G., Arany, E., Simon, G., Schrantz, K., Dombi, A., Hernádi, K., Alapi, T., 2016. Comparison of various advanced oxidation processes for the degradation of phenylurea herbicides. *Journal of Environmental Science and Health, Part B* **51** (4), pp. 205–214.

IF: 1.57

Citation: 2 (independent 1)

c) Conference proceedings

10. Sági Gy., Kovács K., Bezsenyi A., Csay T., Takács E., Wojnárovits L., 2015. Enhancing the biological degradability of sulfamethoxazol by ionizing radiation treatment in aqueous solution. *In: Szentmiklósi László (szerk.), Őszi Radiokémiai Napok 2015. Konferencia helye, ideje: Balatonszárszó, Magyarország, 2015.10.19–2015.10.21. Budapest: Magyar Kémikusok Egyesülete (MKE), pp. 47–52. ISBN:978-963-9970-59-5*

11. Kovács K., Takács E., Wojnárovits L., 2014. Fenilkarbamid herbicidek lebontása vizes oldatban nagyenergiájú sugárzással. *In: Szentmiklósi L (szerk.), Őszi Radiokémiai Napok. Konferencia helye, ideje: Balatonszárszó, Magyarország, 2014.10.13–2014.10.15. Budapest: Magyar Kémikusok Egyesülete (MKE), pp. 44–48. (ISBN:978-963-9970-50-2)*

12. Kovács K., Mile V., Takács E., Wojnárovits L., 2013. Fenuron lebontása vizes oldatban nagyenergiájú sugárzással. *In: Szentmiklósi L (szerk.), XIX. Őszi Radiokémiai Napok. Konferencia helye, ideje: Eger, Magyarország, 2013.10.16–2013.10.18. Budapest: Magyar Kémikusok Egyesülete (MKE), pp. 91–95. (ISBN:978-963-9970-42-7)*

13. Kovács K., Alapi T., Dombi A., 2012. Nagyhatékonyságú oxidációs eljárások fenilurea peszticidek vizes oldatainak kezelése során. *In: Endrődi Balázs (szerk.), XXXV. Kémiai Előadói Napok. Konferencia helye, ideje: Szeged, Magyarország, 2012.10.29–2012.10.31. Szeged: JATEPress Kiadó, pp. 176–180. (ISBN:978-963-315-099-3)*

14. Arany E., Kovács K., Méreg A., Alapi T., Farkas J., Dombi A., 2012. Nagyhatékonyságú oxidációs eljárások alkalmazása peszticid tartalmú vizek

utókezelésére. In: Jurecska L, Kiss Á (szerk.), Környezettudományi Doktori Iskolák Konferenciája 2012. Konferencia helye, ideje: Budapest, Magyarország, 2012.08.30-2012.08.31. Budapest: Eötvös Loránd Tudományegyetem, 2012. pp. 184–191. (ISBN:[978-963-284-242-4](#))

Total IF: 11.387

Number of citations: 16 (independent 6)