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Review
of the doctoral dissertation by Humam Ahmed, MSc (Eng)
entitled: “Degradation of selected drugs used in COVID-19 therapy in the aquatic environment by means of solar light driven processes”

1. Formal basis for the preparation of the review

This review of the doctoral dissertation by Humam Ahmed, MSc Eng, was prepared at the request of the Chair of the Discipline Council for Environmental Engineering, Mining and Power Engineering at the Silesian University of Technology, Prof. Krzysztof Labus, PhD (letter RIE-BD. 512.3.2026), based on the submitted manuscript written by Humam Ahmed, MSc Eng. “Degradation of selected drug used in COVID-19 therapy in the aquatic environment by means of solar light-driven processes".

2. Assessment of the relevance of the dissertation topic

The pharmaceutical industry is one of the most profitable and rapidly growing sectors in the world. Globally, there has been a significant increase in the consumption of antiviral drugs due to an ageing population, a rise in chronic diseases (HIV, HCV) and new threats such as COVID-19 and seasonal influenza. The rise in pharmaceutical consumption leads to increased leaching into the environment. Removing antiviral drugs from the aquatic environment poses a challenge, as conventional water treatment plants and sewage treatment works are often unable to remove them completely. Furthermore, emerging epidemics, such as COVID-19, contribute to the introduction of new antiviral drugs and an increase in their consumption. This necessitates research into their concentration in the environment, persistence, and potential for effective removal, as well as their impact on the environment and human health.

In this context, the subject matter of the doctoral dissertation by Humam Ahmed, MSc Eng. fits within the current research trend concerning the removal of pharmaceuticals from the aquatic environment. In my opinion, the subject matter of the dissertation is topical and highly significant from the perspective of environmental protection.

3. Assessment of the dissertation structure

The doctoral dissertation under review comprises 132 pages and contains 18 tables, 25 figures and 34 appendices in the form of screenshots of the chromatograph display. The bibliography includes 114 references. The dissertation begins with a table of contents, a list of key abbreviations, and an abstract in English and Polish.

The dissertation consists of twelve chapters – the first chapter is a literature review on the subject of the dissertation, followed by a chapter presenting the research hypothesis, the aim and scope of the

work. The subsequent chapters cover the research methodology, a discussion of the results obtained, a summary and conclusions, and directions for further research. The subsequent chapters comprise the bibliography, supplementary information, a list of figures and a list of tables. The dissertation concludes with a list of the PhD candidate's publications and a list of her other achievements. In my opinion, the adopted structure of the dissertation is clear and comprehensive.
Chapter 1 consists of nine sub-chapters, in which the PhD candidate discusses advanced oxidation processes, characterises antiviral drugs contaminating the aquatic environment, and describes the drugs under investigation in the dissertation.

Chapter 2 consists of five sub-chapters. In the first, the PhD candidate presented her research hypothesis:
The efficiency of the degradation of COVID-19-related pharmaceutical compounds in the aquatic environment can be significantly increased through sunlight-assisted photocatalysis, with the efficiency being largely influenced by the type of photocatalyst, the composition of the aqueous matrix and the process parameters.

In the following subchapter, the author presented three scientific problems that needed to be addressed. In the third subchapter, she discussed the main objective of the dissertation, which is to investigate the photodegradation process of selected antiviral drugs used in the treatment of COVID-19 in an aquatic environment under conditions of exposure to sunlight and artificial light, in the absence and presence of photocatalysts that assist the degradation process. In the next subsection, she presented nine specific research objectives. The fifth subsection discusses the scope of the doctoral dissertation.

Chapter 3 – *Materials and Methodology* – consists of three sub-chapters. In the first sub-chapter, the candidate listed the three antiviral drugs under investigation: isoprinosine, ritonavir and remdesivir, as well as the commercial photocatalysts used (TiO_2 P25, ZnO, SnO_2, ZrO_2 and $BaWO_4$) and those synthesised in the laboratory ($AgTiO_2$ and $BaTiO_3$), as well as the ionic compounds NaCl, Na_2SO_4, $CaCO_3$ and $NaNO_3$ used as sources of anions. In the following subchapter, the author discussed the aqueous matrices used: ultra-pure water, so-called Milli-Q, tap water and surface water collected from the Ostropka River. The first served as a reference matrix, free from contaminants, whilst the other two reflected actual environmental conditions. Subchapter 3 consists of 11 sub-sections, in which the methodology of the experimental studies is presented in detail. In these, the PhD student discussed the method of preparing standard solutions of the tested drugs, the method of determining the maximum absorption wavelength for the selected drugs, and the method of preparing calibration curves. In addition, the solar reactor used in the studies and the course of the photolytic, photocatalytic and H_2O_2-assisted studies were described. The final subchapter describes the kinetic studies conducted.

Chapter 4, *Results and Discussion*, constitutes a substantial part of the dissertation. In the first subchapter, the PhD candidate discussed the ion profiling of the aqueous matrices used in the studies. In the following subchapter, she presented the results of the studies on photolysis using sunlight. In order to determine the degradation efficiency of selected antiviral drugs (isoprinosine, ritonavir and remdesivir), the research was conducted in three aqueous matrices (ultra-pure water, tap water and surface water) at a radiation intensity of 500 W/m². In the third subchapter, the PhD candidate discussed the results of research into the photocatalytic removal of selected antiviral drugs. The research was begun by determining the efficiency of the photocatalytic degradation of isoprinosine in ultrapure water using seven commercial photocatalysts and two synthesised as part of separate research projects. The photocatalysts were used at concentrations of 5.0 mg/L (TiO_2 P25 and ZnO only), 10.0 mg/L and 20.0 mg/L at a radiation intensity of 500 W/m². To determine the degradation efficiency in real-world environmental matrices (tap water and surface water), the author selected the most effective photocatalysts. The photodegradation efficiency of isoprynosine in tap water was determined using three photocatalysts (TiO_2 P25, ZnO and SnO_2), selected in preliminary studies, at concentrations of 10.0 and 20.0 mg/L under a radiation intensity of 500 W/m^2. For surface water, the candidate used two photocatalysts, TiO_2 P25 and ZnO, at a dose of 20.0 mg/L. To determine the effectiveness of photocatalytic removal of ritonavir in three aqueous matrices, she used three photocatalysts: TiO_2 P25, ZnO and SnO_2:ZnO (2:1) at a constant dose of 10 mg/L and a radiation intensity of 500 W/m^2. The PhD student investigated the photodegradation efficiency of remdesivir using two photocatalysts (TiO_2 P25 and ZnO) at a concentration of 20.0 mg/L. To verify whether the reduction in the concentration of the pharmaceutical contaminants under investigation in the aquatic environment resulted from actual degradation or merely from adsorption, experiments were conducted in the dark. In the case of isoprinosine, dark tests were performed using seven different photocatalysts (TiO_2 P25, ZnO, ZrO_2, $BaWO_4$, Ag-TiO_2, $BaTiO_3$, SnO_2), at a concentration of 10.0 mg/L in ultrapure water. Dark tests for ritonavir were carried out with three photocatalysts (TiO_2 P25, ZnO and SnO_2:ZnO (2:1)), and for remdesivir with two (TiO_2 P25 and ZnO). In the following sub-section, the candidate described the results of studies on the susceptibility of the tested drugs to degradation via H_2O_2-assisted photolysis. She used three volumes of H_2O_2: 125 µL (108.8 mg/L), 250 µL (217.5 mg/L) and 500 µL (435 mg/L) in the presence of sunlight (500 W/m^2) in ultrapure water. In addition, she conducted a test in the dark to confirm the effectiveness of H_2O_2 (500 µL) in removing selected pharmaceuticals. In subsequent chapters, the author investigated the influence of selected anions (SO_4^{2-}, Cl^-, NO_3^-, CO_3^{2-}) on the photodegradation of the studied antiviral drugs in ultrapure water and tap water. In the final sub-chapter, the PhD candidate investigated the effectiveness of sunlight-induced mineralisation of the organic compounds of the antiviral drugs under study in three aqueous matrices.

The substantive part of the dissertation concludes with Chapter 5: *Summary and Conclusions*. Chapter 6 outlines directions for further research, which, according to the candidate, should include, amongst other things, the identification and characterisation of intermediate products formed in photolytic and photocatalytic processes, as well as the potential for reuse and recycling of the photocatalysts employed.

The subsequent chapters are the *Bibliography*, comprising 114 references, 96 of which were published in the last ten years, listed in alphabetical order, and the chapter *Supplementary Information*. The dissertation concludes with a *List of Figures*, a *List of Tables*, a *List of Publications* and a *List of Other Academic Activities*.

4. Substantive Assessment

The subject of the research, which forms the basis of the doctoral dissertation by Humam Ahmed, MSc Eng, was the assessment of the degradation potential of selected antiviral drugs (isoprinosine, ritonavir, remdesivir), used in the treatment of COVID-19, via photolysis, photocatalysis and H_2O_2-assisted photolysis. The dissertation analysed the influence of environmental and process parameters, such as: water composition, light intensity, catalyst dose, contaminant concentration and reaction time.

I highly commend the choice of research topic and the comprehensive approach to the problem of determining the degradation efficiency of selected antiviral drugs. It should be emphasised that the title of the dissertation, its objectives and theses have been correctly formulated, and the research questions detailed by the author on pages 29 and 30 enabled the achievement of the objectives set out by the PhD candidate.

The literature review provides a logical introduction to the issues addressed in the dissertation.

The chapter *Materials and Methodology* provides a detailed discussion of the antiviral drugs selected for the study, the photocatalysts, the aqueous matrices, the experimental methodology, the research apparatus, and the models used in the kinetic studies. I consider the selection of research methods to be appropriate and sufficient for achieving the dissertation's objective.

The PhD student achieved a 9% degradation efficiency of isoprinosine in ultrapure water after 60 minutes of irradiation (500 W/m²). After 120 minutes, the efficiency was 14% in tap water and 8% in surface water, respectively. In the case of ritonavir, degradation after 90 minutes of irradiation in ultrapure water was only 15%, and after 120 minutes the author achieved a degradation efficiency of 38% in tap water and 51% in surface water. The degradation efficiency of remdesivir after 120 minutes of irradiation was 15% in ultrapure water, 27% in tap water and 23% in surface water. The PhD student demonstrated that solar photolysis leads only to limited degradation of the tested drugs, which confirms their high photostability and the insufficient effectiveness of photolysis as a standalone purification method.

The use of photocatalysts led to an increase in the degradation efficiency of antiviral drugs. The use of TiO_2 P25 and ZnO at a dose of 5.0 mg/L in the photocatalytic process of isoprinosine in ultrapure water resulted in degradation efficiencies of 100% after 30 minutes and 88% after 120 minutes, respectively. Increasing the dose to 10.0 mg/L resulted in an increase in the degradation efficiency of isoprinosine to 98% for ZnO after 120 minutes of irradiation. The highest degradation efficiency of isoprynosine in tap water was achieved at a dose of 20.0 mg/L for the three photocatalysts studied, amounting to 93%, 88% and 38% for ZnO, TiO_2 P25 and SnO_2, respectively. In the case of surface water, the PhD student used a dose of 20.0 mg/L for TiO_2 P25 and ZnO and achieved isoprinosine degradation efficiencies of 23% and 22%, respectively. For the degradation of ritonavir, the PhD student used a photocatalyst dose of 10.0 mg/L and found that ritonavir was completely adsorbed (100%) onto the surface of ZnO and SnO_2:ZnO (2:1) within 30 seconds of the reaction in all tested matrices, whilst TiO_2 P25 exhibited 72% adsorption after 120 minutes of irradiation. The author investigated the photocatalytic removal of remdesivir using a dose of 20.0 mg/L of TiO_2 P25 and ZnO. In ultrapure water, she achieved 98.92% adsorption efficiency for TiO_2 P25 and 81% for ZnO after 120 minutes of irradiation. The lowest adsorption efficiency was observed in surface water, at 5% and 1% for TiO_2 P25 and ZnO, respectively. Studies of the photocatalytic degradation of selected antiviral drugs used in COVID-19 therapy showed that TiO_2 P25 and SnO_2:ZnO exhibited the highest photocatalytic activity under sunlight. TiO_2 P25 exhibited greater stability in complex aqueous matrices, whilst ZnO provided a stronger, albeit less reversible, binding under controlled conditions.

The use of H_2O_2-assisted photolysis influences the efficiency of antiviral drug degradation. The PhD student found that isoprinosine undergoes 100% degradation after 30 minutes of reaction for H_2O_2 doses of 125 µL and 250 µL. However, at a dose of 500 µL of H_2O_2, complete degradation was achieved after 120 minutes of irradiation. In the case of ritonavir, 100% degradation was achieved within 45 minutes for all tested H_2O_2 concentrations. In contrast, the degradation efficiency of remdesivir after 120 minutes was 55%, 53% and 83%, respectively, for H_2O_2 doses of 125 µL, 250 µL and 500 µL.

Furthermore, the PhD student investigated the influence of inorganic ions (SO_4^-, Cl^-, NO_3^-, CO_3^-) present in natural waters on the degradation of isoprinosine and ritonavir. She demonstrated that these ions affected the efficiency of the process by scavenging reactive oxygen species or modifying the surface charge of the catalyst.

The *Discussion* presented by the PhD student is sound and supported by arguments based on the results presented in the relevant literature. The *Summary and Conclusions* chapter contains a list of the most significant achievements of the dissertation.

Alongside the positive assessment of the dissertation, the following questions arise:

1. On what basis was a solar radiation intensity of 500 W/m² adopted in the research?

2. What were the criteria for selecting the photocatalysts used, and which of their physicochemical properties determined the choice?
3. What conditions must be met to enable the use of photodegradation of the antiviral drugs under investigation in the wastewater treatment process?
4. What, in the author's view, is the potential toxicity of the intermediate products formed during the photodegradation of isoprinosine, ritonavir and remdesivir, and what impact might they have on aquatic organisms?
5. Why were different models used in the kinetic studies (pseudo-first-order, Langmuir-Hinshelwood, two-phase adsorption-desorption model), and what might be the implications of this when designing pharmaceutical removal processes on an industrial scale?

In summary of the substantive assessment of the work, I confirm my positive opinion of the doctoral dissertation by Humam Ahmed, MSc Eng. The dissertation contains extensive experimental material, organised and presented in a comprehensible manner. The considerable amount of work and time involved in conducting the research, and above all its applied nature, should be emphasised.

5. Specific comments

The dissertation is fairly well-edited, though the author has not managed to avoid certain errors.

The reference list contains several entries that are repeated twice or even three times. This applies to the following articles: Ahmed, H., Felis, E. (2023) *Drugs used in COVID-19 therapy and their effects on the environment. Desalination and Water Treatment*, 301, 52–62; Ahmed, H., Siddiqui, B., Felis, E. (2025) *Adsorption and photodegradation as processes enabling the removal of antiviral drug ritonavir from the aquatic environment*. Journal of Ecological Engineering, 26(5), 195–202; Domingo-Echaburu, S., Irazola, M., Prieto, A., Rocano, B., Lopez de Torre-Querejazu, A., Quintana, A., Orive, G., Lertxundi, U. (2022) *Drugs used during the first wave of COVID-19 in Vitoria-Gasteiz (Spain) and their presence in the environment.* Science of The Total Environment, 820, 153122; Gwenzi, W., Selvasembian, R., Offiong, N. A. O., El Din, A. (2022) *COVID-19 drugs in aquatic systems: a review. Environmental Chemistry Letters*, 20(2), 1275–1294; Kang, Y. M., Kim, T. K., Kim, M. K., Zoh, K. D. (2020) *Greenhouse gas emissions from advanced oxidation processes in the degradation of bisphenol A: a comparative study of the H_2O_2/UV, TiO_2/UV, and ozonation processes*. Environmental Science and Pollution Research, 27(11), 12227–12236; Mansouri, F., Chouchene, K., Roche, N., Ksibi, M. (2021) *Removal of pharmaceuticals from water by adsorption and advanced oxidation processes: State of the art and trends*. Applied Sciences, 11(14); Miklos, D. B., Remy, C., Jekel, M., Linden, K. G., Drewes, J. E., Hübner, U. (2018) *Evaluation of advanced oxidation processes for water and wastewater treatment - A critical review*. Water Research, 139, 118–131; Nugnes, R., Orlo, E., Russo, C., Lavorgna, M., Isidori, M. (2024) *Comprehensive eco-geno-toxicity and environmental risk of common antiviral drugs in aquatic environments post-pandemic*. Journal of Hazardous Materials, 480, 135947; Zhou, C., Chen, J., Xie, Q., Wei, X., Zhang, Y. nan, Fu, Z. (2015) *Photolysis of three antiviral drugs acyclovir, zidovudine and lamivudine in surface freshwater and seawater*. Chemosphere, 138, 792–797.

One reference (Araújo et al., 2018 – p. 60) is missing from the reference list.

In my opinion, the structure of an academic dissertation should be divided into no more than three levels (chapter, subchapter, section), which are listed in the table of contents. For example, I consider the inclusion of sub-sections within sections 3.3.5 and 3.3.6 to be unnecessary. Furthermore, in Chapter 4, the PhD candidate has unnecessarily introduced sub-section 4.1, the title of which is a repetition of the chapter title; this has additionally resulted in there being only one sub-section in this chapter, which is inconsistent with the principles of academic writing.

6. Summary and Conclusion

The doctoral dissertation submitted for assessment constitutes a valuable scientific study, and the research findings significantly expand the state of knowledge regarding the potential for removing antiviral drugs used in COVID-19 therapy through photocatalytic degradation.

I conclude that the doctoral dissertation by MSc Humam Ahmed, entitled “Degradation of selected drug used in COVID-19 therapy in the aquatic environment by means of solar light-driven processes”, meets the requirements defined in Article 187 of the Act of 20 July 2018 on Higher Education and Science (Journal of Laws 2018, item 1668, as amended). The dissertation presents an original solution to a scientific problem, and the PhD candidate has demonstrated theoretical knowledge in the field of environmental engineering, mining and power engineering, as well as the ability to conduct independent scientific research.

I therefore request the Discipline Council for Environmental Engineering, Mining and Power Engineering at the Silesian University of Technology to admit Ms Humam Ahmed, MSc Eng, to the subsequent stages of the qualification procedure for the award of the degree of Doctor of Engineering and Technology in the discipline of environmental engineering, mining and power engineering.

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Prof. Dr. Eng. Joanna Rodziewicz