



Research Article

Environmental Footprint Assessment of Methylene Blue Photodegradation using Graphene-based Titanium Dioxide

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Received: 16th February 2023; Revised: 7th March 2023; Accepted: 8th March 2023
Available online: 12nd March 2023; Published regularly: March 2023



Abstract

To date, photocatalysis has received much attention in terms of the degradation of organic pollutants in wastewater. Various studies have shown that graphene-based photocatalysts are one of the impressive options owing to their intriguing features, including high surface area, good conductivity, low recombination rate of electron-hole pair, and fast charge separation and transfer. However, the environmental impacts of the photocatalysts synthesis and their photodegradation activity remain unclear. Thus, this report aims to identify the environmental impacts associated with the photodegradation of methylene blue (MB) over reduced graphene oxide/titanium oxide photocatalyst (TiO₂/rGO) using Life Cycle Assessment (LCA). The life cycle impacts were assessed using ReCiPe 2016 v1.1 midpoint method, Hierarchist version in Gabi software. A cradle-to-gate approach and a functional unit of 1 kg TiO₂/rGO were adopted in the study. Several important parameters, such as the solvent type (ultrapure water, ethanol, and isopropanol), with/without silver ion doping, and visible light power consumption (150, 300, and 500 W) were evaluated in this study. In terms of the selection of solvent, ultrapure water is certainly a better choice since it contributed the least negative impact on the environment. Furthermore, it is not advisable to dope the photocatalyst with silver ions since the increment in performance is insufficient to offset the environmental impact that it caused. The results of different power of visible light for MB degradation showed that the minimum power level, 150 W, could give a comparable photodegradation efficiency and better environmental impacts compared to higher power light sources.

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Keywords: Sustainability; Photodegradation; Graphene-based TiO₂; Environmental Impact; Energy

How to Cite: K. Kong, Y. Weng, W.H. Lam, S.Y. Lai (2023). Environmental Footprint Assessment of Methylene Blue Photodegradation using Graphene-based Titanium Dioxide. *Bulletin of Chemical Reaction Engineering & Catalysis*, 18(1), 103-117 (doi: 10.9767/bcrec.17450)

Permalink/DOI: <https://doi.org/10.9767/bcrec.17450>

1. Introduction

With rapid industrialization and the rising world population in recent decades, various environmental issues have become increasingly

important in our lives. Water pollution is one of the major environmental concerns as harmful and toxic substances are constantly discharged into our water systems. The large scale of industries have resulted in the abundant use and often unregulated discharge of partial or untreated chemicals, thus, could become a serious glob-

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al environmental issue if overlooked. Organic pollutants are toxicants that are capable of causing various diseases in humans if not monitored well and consumed exceeding the permissible limits. Industrial consumables such as detergent, organic dye, petroleum hydrocarbon, and plastic are vital sources of these organic pollutants. Moreover, the chemical complexity of these organic pollutants can cause prolonged harm to wildlife and long-term damaging effects on the environment [1]. For example, the lipophilic nature of PCBs, dioxin, and DDT allows them to be stored in fat cells and retained for years, thus, the effects could be long-lasting [2].

Various types of treatment technologies have been utilized to remove organic pollutants from wastewater, such as aerobic treatment, adsorption, coagulation, and flocculation [3,4]. These technologies are very effective but are limited by high operational costs, complicated techniques, long operation time, and so forth. Advanced Oxidation Process (AOP) is another type of water treatment technology that could complement the abovementioned shortcomings. To date, AOPs have received a lot of attention in terms of the degradation of organic pollutants in wastewater. Generally, AOPs utilize the reactivity of OH radicals to degrade the organic pollutants with a series of oxidation and reduction processes [5]. Photocatalysis, one of the AOPs, possesses great advantages such as mild operating conditions, low cost, low toxicity, good chemical stability, super hydrophilicity, and high efficiency as compared to conventional water treatment techniques [6]. In short,

photocatalysis accelerates photoreaction with the support of a photocatalyst.

Among the photocatalysts, graphene-based TiO_2 photocatalyst offers high quantum conversion efficiency, better solar energy utilization, and a larger surface area. Furthermore, graphene hinders the high recombination rate of the electron due to its high electron mobility in TiO_2 /graphene composite photocatalyst [7,8], consequently enhancing the photocatalytic degradation of pollutants. Figure 1 shows the number of publications and citations has increased over the last decade, with a peak of 906 publications in 2019 and citations of 71287 in 2021. Hence, the utilization of graphene-based photocatalysts has earned respect and is one of the most promising techniques.

Nevertheless, the environmental impact of photocatalytic activities and the synthesis of photocatalysts is still an important subject to ensure the viability of the process. For instance, a Life Cycle Assessment (LCA) of solar-based treatment was conducted to determine the environmental impact possessed by TiO_2 photocatalyst [9]. Besides, an LCA based on mono-doped TiO_2 catalysts with transition metal has investigated whether the improvement in photocatalytic activity can offset the environmental impacts of metal doping [10]. The results showed that mono-doped TiO_2 photocatalyst possessed limited environmental impact in the synthesis process and treatment of organic micropollutants [9,10]. However, reports on the LCA on the degradation of organic pollutants by graphene-based photocatalysts are limited. Hence, this research aims to exam-

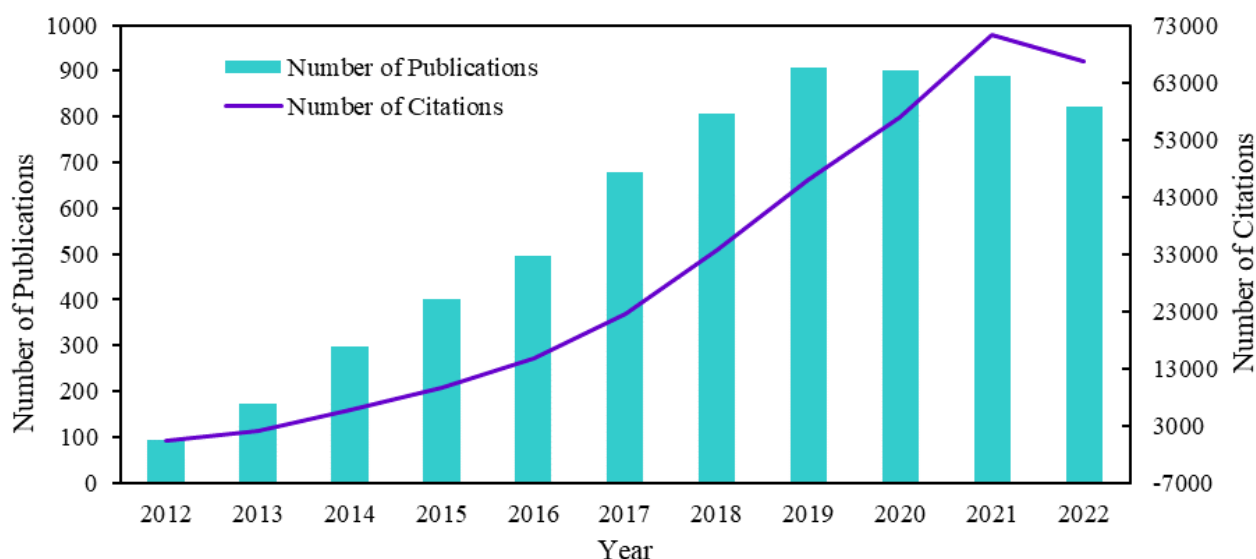


Figure 1. Number of publications and citations on graphene-based photocatalysts from 2012 to 2022, retrieved from the Web of Science database with the topic keywords “graphene” and “photocatalysis” on February 16, 2023.

ine the environmental impacts of TiO₂/rGO production. Additionally, the environmental benefits of different solvents (ultrapure water, ethanol, and isopropanol) and power consumption of visible light (150 W, 300 W, and 500 W) were evaluated.

2. Materials and Methods

2.1 Synthesis of Graphene Oxide (GO) and Titanium Oxide/Reduced Graphene Oxide (TiO₂/rGO) Photocatalyst

The data utilized in the LCA methodology was applied according to ISO standards. The data are retrieved from past experimental works [11–19].

2.1.1 GO synthesis route

Firstly, graphene oxides (GO) were first synthesized through a Hummers' process. Natural graphite powder (5 g) was added to a concentrated H₂SO₄ (250 mL) at room temperature and agitated for 30 minutes to initiate the reaction. The mixture was then cooled to 5 °C in an ice bath, and then KMnO₄ (30 g) was gradually added while keeping the temperature below 15 °C. The mixture was then agitated for 4 hours in an oil bath at around 40 °C. The mixture was placed in an ice bath once more and stirred until it reached 5 °C. After that, aliquots of deionized water (20 mL) were gently added, keeping the temperature below 50 °C, until the mixture was diluted to 1 L. Then, drop by drop, 30% H₂O₂ (25 mL) was introduced and agitated for 1 hour until the KMnO₄ reaction was completed. To remove the metallic ion, the final mix was rinsed twice with 10% HCl and 5 times using deionized water [11]. Finally, GO was synthesized. Reduced graphene oxide (rGO) was synthesized along with TiO₂ using a hydrothermal method, as described in Section 2.1.2.

2.1.2 TiO₂/rGO synthesis route

A hydrothermal technique was used to produce the TiO₂/rGO photocatalyst (P25/rGO). 2 g TiO₂ was suspended in 400 mL ultrapure water and agitated at 500 rpm for 30 min. The necessary amount of GO was then added, and the mixture was agitated for another 2.5 h to en-

sure that the GO sheets were evenly distributed. After that, the solution was transferred to a 600 mL Teflon-lined stainless steel autoclave reactor and hydrothermally treated for 3 h at 120 °C [12]. After that, the TiO₂/rGO photocatalyst was rinsed and centrifuged three times before being dried at 50 °C.

To compare the effect of different chemicals or substances used in the synthesis process, an emphasis has been focused on the solvent used in the synthesis of TiO₂/rGO photocatalyst. In the base system, ultrapure water was utilized while isopropyl and ethyl alcohol were chosen as the alternative routes [11,13].

After deciding on the solvent, another comparison will be done in terms of the necessity and the sustainability of doping photocatalyst with silver (Ag). For the process, silver nitrate will first be dissolved into deionized water and mixed with TiO₂/rGO solution afterwards for 30 min at 94 °C with vigorous stirring [14,15].

2.2 Photodegradation Activity

200 mg of TiO₂/rGO photocatalyst was used to degrade 200 mL methylene blue (MB) with a concentration of 15 ppm for 60 min [16]. In this section, different power levels of visible light sources of the photodegradation system were compared to evaluate their environmental impacts. The power levels of the visible light source used were 150 W, 300 W, and 500 W, respectively [17–19], as tabulated in Table 1.

2.3 Goal/Functional Unit

The goal of this study is to determine the environmental impacts associated with a graphene-based photocatalyst in terms of its synthesis and photodegradation process. In detail, the study is to compare the environmental impact possessed by different chemicals used in the synthesis process and the power consumption of visible light in the photodegradation process. The scientific community and wastewater treatment decision-makers, with the aim to prepare the photocatalyst in the most sustainable manner, are hence the designated audience for this publication.

The life cycle inventory for photocatalyst production and photodegradation was normalized per functional unit. In this study, the func-

Table 1. Comparison of alternative options.

Process	Option 1	Option 2	Option 3
Chemicals in synthesis process	Ultrapure water	Isopropyl alcohol	Ethyl alcohol
Doping process	With doping agent (Ag)	Without doping agent (Ag)	
Power consumption of visible light	150 W	300 W	500 W

tional unit used is 1 kg of TiO_2/rGO photocatalyst to treat MB solution. It defines the unit of service used as the basis when compares among different alternatives. The assumptions made for the LCA were: (a) The system is located in Malaysia which possesses a daily erythemal UV dose of 5518.13 J/m^2 . The minimum value is taken to obtain the minimum result [20]; (b) Chemicals that are unavailable in the GaBi database 2020 are replaced with similar chemicals that possessed related data in the software; and (c) Since the system is assumed to be located in Malaysia, LCI data for water

supplies and electricity are based on Malaysia's standards.

2.4 Scope and System Boundaries

This LCA study is based on a cradle-to-gate approach. Cradle-to-gate is an assessment of the life cycle of a particular product from the extraction of raw materials ('Cradle') to any stage in the life cycle before the product disposal ('Gate'). In this study, the system boundary was defined using the cradle-to-gate approach would include the extraction of raw materials

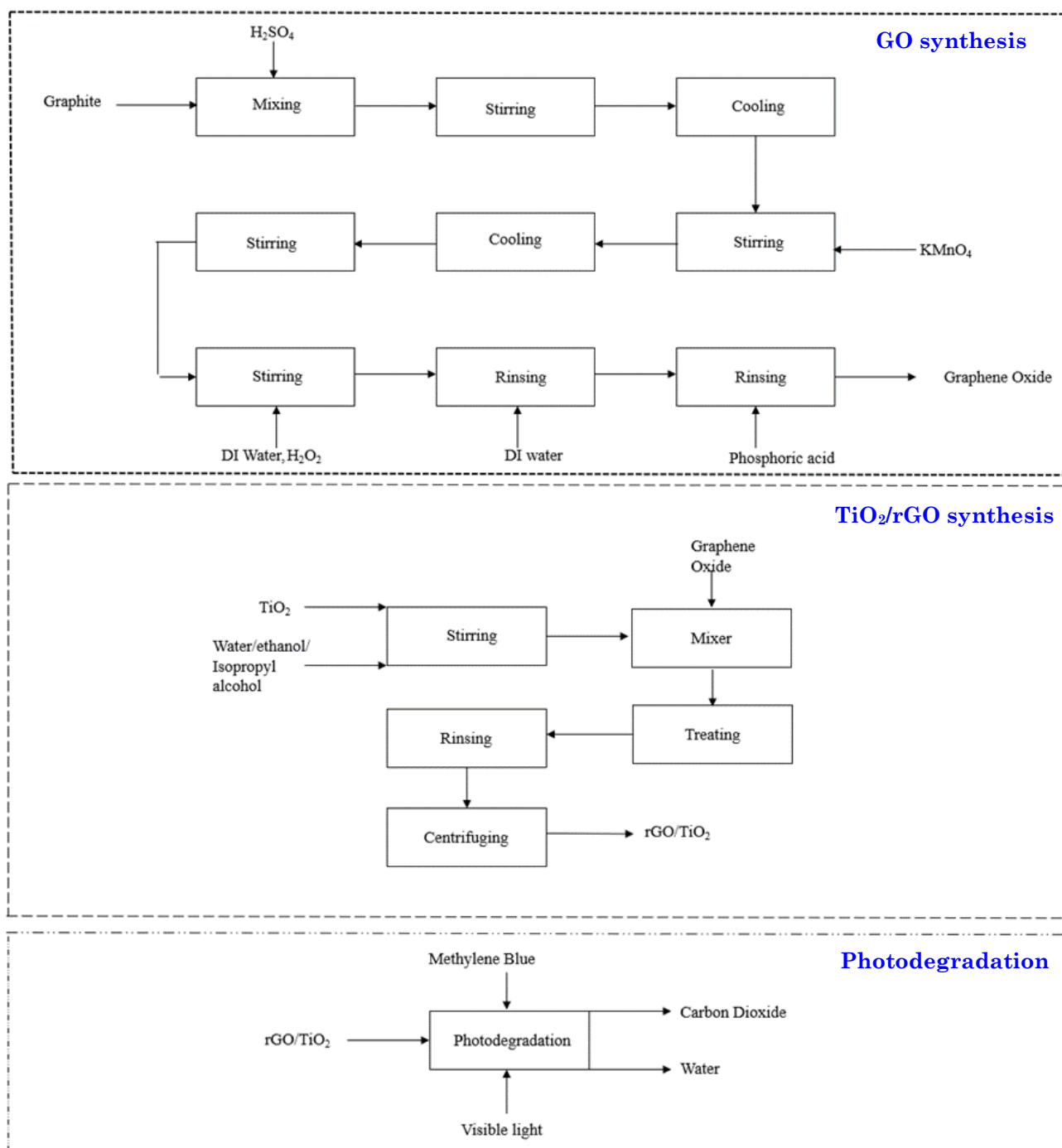


Figure 2. System boundaries.

to produce TiO₂/rGO photocatalyst, the photocatalyst synthesis process and the photodegradation process, excluding the disposal of the photocatalyst. Figure 2 shows the overall process flow diagram for the synthesis of GO and TiO₂/rGO photocatalyst and the photodegradation process with boundaries. Based on the process flow diagram, the input and output from each boundary are also indicated in the system boundaries diagram in Figure 2.

2.5 Inventory Analysis

The inventory data acquired from laboratory-scale methods included in the database was used to examine the environmental implications of TiO₂/rGO synthesis and photodegradation. The TiO₂/rGO production system's foreground system includes chemicals utilized as raw materials, as well as electricity usage in the stirrer, heat exchanger and autoclave reactor. All chemicals involved in the process are listed in Table 2. The process and chemicals involved in the system were modeled with the data available in the database (GLO: global; RER: regional market for Europe; and MY: Malaysia).

Table 2. Chemicals needed in the life cycle.

Use phase	Chemicals
Synthesis of graphene oxide	graphite powder
	H ₂ SO ₄
	KMnO ₄
	Deionized water
	HCl
Synthesis of TiO ₂ /rGO	H ₂ O
	TiO ₂
	Ultrapure water, isopropyl alcohol, ethyl alcohol (Comparison)
	DI-water, silver nitrate
Doping process	DI-water, silver nitrate
Photodegradation	Methylene Blue

Table 3. The chemicals and electricity used in GO production.

Use phase		Input	Value	Unit
Graphene oxide production	Mixing	Graphite	0.0075	kg
		Sulphuric Acid	0.375	
	Stirring 1	Potassium Permanganate	0.045	kg
	Stirring 2	Hydrogen peroxide	0.0375	kg
		Deionized water	0.03	
	Agitating/Rinsing	Hydrochloric acid	0.01	kg
		Electricity	10.8	MJ

2.6 Impact Assessment

The possible environmental consequences were calculated using the inventory analysis findings using GaBi database. Life cycle assessment may convert all associated emissions and outputs into precise environmental scores acquired via the use of characterization elements in a specific technique. The following aspects of a life cycle impact assessment (LCIA) have been completed: (1) choice of impact categories, (2) allocation of inventory data to impact categories (classification), and (3) computation of category indicator results according to ISO 14042. (characterization). GaBi thinkstep has been selected as the primary software to conduct the LCA. The present LCA study is based on a cradle-to-gate approach, i.e. from the production of precursor materials to their usage but excludes the disposal part.

The LCIA method utilized was ReCiPe 2016 v1.1 midpoint method, Hierarchist version which includes Stratospheric Ozone Depletion, Global Warming, Ozone Formation, Terrestrial Ecotoxicity, Ionizing Radiation, Fine Particulate Matter Formation, Marine Eutrophication, Freshwater Eutrophication, Freshwater Ecotoxicity, Marine Ecotoxicity, Land Use, Human Toxicity, Mineral Resource Scarcity, Water Consumption, Freshwater Consumption, Photochemical Ozone Formation and Metal Depletion. The midpoint categories are represented in terms of the mass (in kg) of the reference material generating the harm (weighted impact), but the unit for water consumption and land use is the area and time of a land use, as well as the cubic meter, respectively.

3. Results and Discussion

3.1 LCA of Graphene Oxide (GO) Production

In terms of the LCA for the graphene oxide (GO) production, the amount of GO needed to produce 1 kg of TiO₂/rGO photocatalyst was assessed. 1 kg of photocatalyst meanwhile is the functional unit of this LCA study. Table 3 tabulates the quantity of the chemicals and elec-

tricity used for the production of GO, while Figures 3 and 4 show the environmental impact distribution derived from each chemical and electricity consumed in the production of GO. The chemicals analyzed were graphite, sulphuric acid (H_2SO_4), potassium permanganate (KMnO_4), hydrogen peroxide (H_2O_2), deionized (DI) water and hydrochloric acid (HCl). In total, electricity accounts for the majority of the environmental impacts since it is used in all processes which include mixing, stirring 1, stirring 2 and agitating. Before analyzing the chemicals, a few substitutions have been made in GaBi software. DI water and HCl have insufficient ReCiPe data. Thus, substitutions have been made to replace them with fresh water and phosphoric acid (H_3PO_4) in the software. DI water is commonly used in experimental works since the water quality is more predictable and can lead to more repeatable results. However, due to insufficient data, DI water was replaced with fresh water. H_3PO_4 has been chosen to replace HCl , since it can function similarly to HCl which is removing metal ions [21]. Some of the negative values in the figure

could be explained by the consumption of these chemicals in the processes, instead of disposing them to the environment. For example, H_2SO_4 was consumed in the process not disposed to the environment, thus it will not negatively impact the environment in terms of freshwater ecotoxicity.

First of all, graphene accounted for 75% of midpoint-mineral resource depletion, also

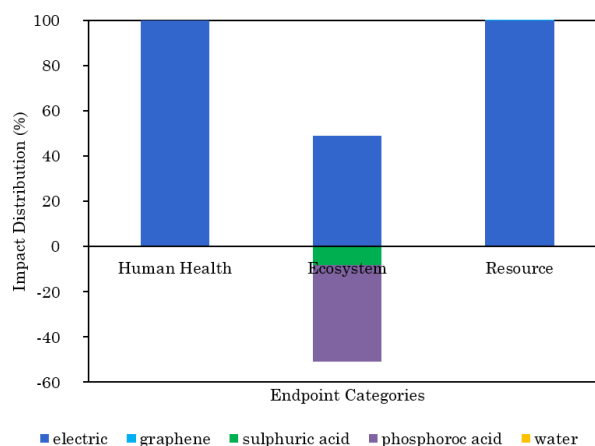


Figure 4. Endpoint data of GO production.

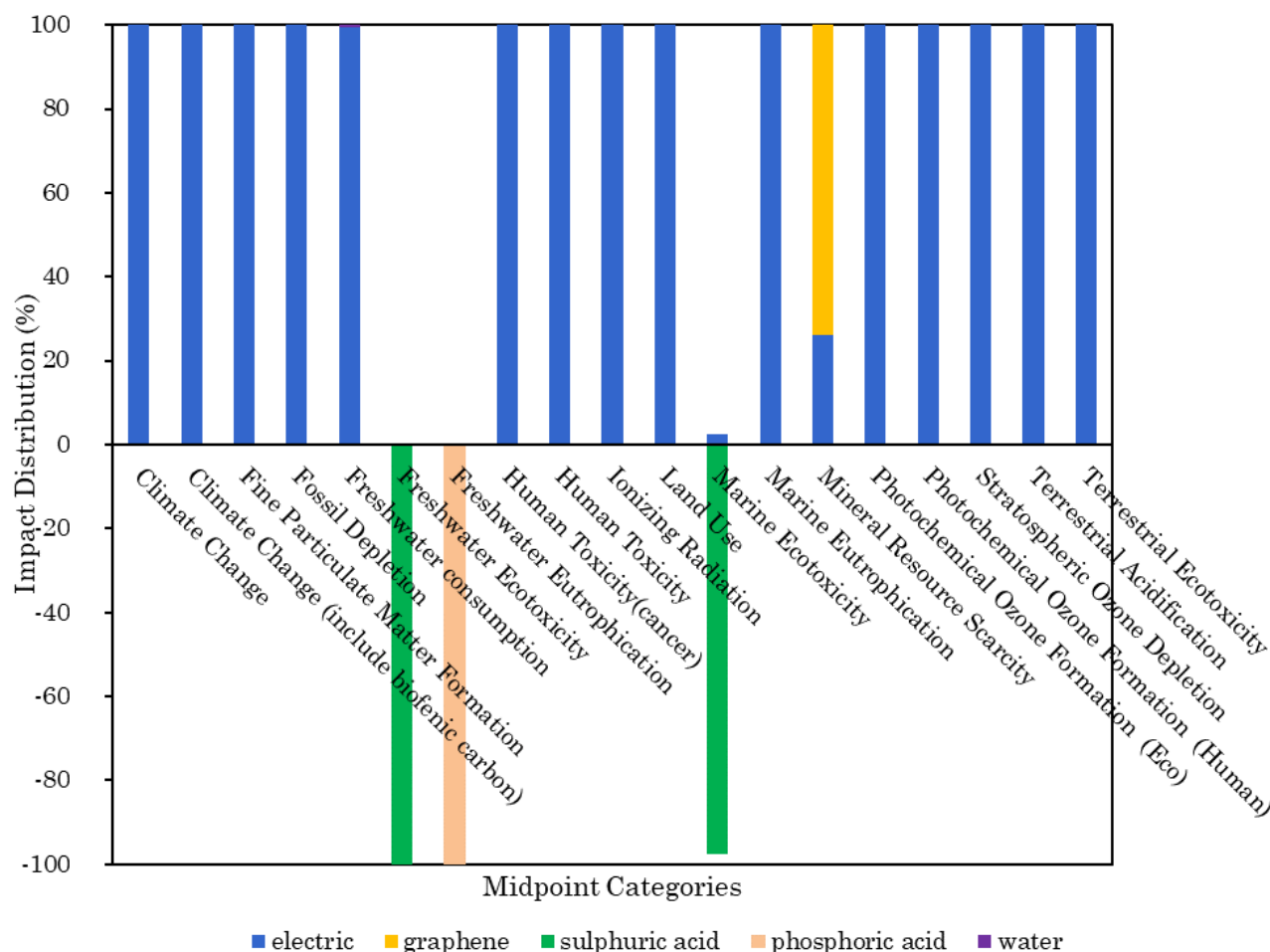


Figure 3. Midpoint data of GO production.

known as mineral resource scarcity, which has a value of 1.44×10^{-3} kg Cu eq. For this midpoint, the damage modelling process is separated into various parts for the impact category of mineral resource scarcity. A mineral resource's main extraction results in a global decline in ore grade, or the concentration of that resource in ores, which will increase the amount of ore produced per kilogram of the mineral resource extracted. This results in an average surplus ore potential, which is the midpoint indicator for this impact category, when combined with the anticipated future exploitation of that mineral resource. The potential for surplus cost will rise as the possibility for surplus ore rises. However, graphene is the main component in this process and this impact is inevitable.

In terms of H_2SO_4 , it mainly contributes to freshwater ecotoxicity and marine ecotoxicity. Freshwater is a special type of habitat for the environment and is also necessary for human life. Freshwater contamination can have an adverse effect on human health in addition to endangering the ecosystem. It is crucial to keep anthropogenic pollution below the level that would constitute a risk. It contributes a negative value of 57100×10^{-5} kg 1,4 DB eq. As mentioned, instead of disposing it, H_2SO_4 was consumed in this process, thus making the value negative. The same goes for marine ecotoxicity which possessed a negative value of 136×10^{-4} kg 1,4 DB eq [22,23].

On the other hand, the main contributor to photochemical ozone formation is electricity. Ozone is created via the photochemical interactions between NO_x and Non-Methane Volatile Organic Compounds, not by direct emission into the atmosphere (NMVOCs). Ozone can irritate airways and harm the lungs, endangering human health. Ozone concentrations cause respiratory distress in people, such as asthma and chronic obstructive pulmonary disease, to occur more frequently and severely (COPD). Ozone can also harm plants by slowing down growth and seed generation, accelerating the ageing of leaves, and making them less resilient to environmental shocks.

Lastly, water only contributes a tiny fraction of freshwater consumption, about 0.52%. Hence, it can be deemed negligible. Here, the usage of water consumption is the basis for all water-related problems. Water consumption refers to the use of water in ways that cause it to evaporate, be incorporated into goods, be moved to other watersheds, or be dumped into the ocean. Therefore, used water is no longer available to ecosystems or humans in the original watershed.

Figures 3 and 4 indicate that electricity is by far the biggest contributor to environmental damage. Both of which have negative effects on human health and the environment [24]. Even while coal and natural gas must be used in the production of power, the effect on the resources is less severe here than it is for ecology and human health. In addition, coal mining can contaminate water. Changes in groundwater flow brought on by mining operations frequently expose previously unpolluted streams to specific mineral substances that leach from the soil and result in acid mine drainage. Another by-product of several forms of energy use is solid waste. Along with coal, mining for coal necessitates the removal of a lot of earth [25]. As also seen in the endpoint result, the majority of the impact is contributed by electricity. Hence, it is important to decrease energy usage when possible. In this section, it may be possible to combine both stirring processes to reduce energy usage which can be an interesting idea. As also seen in the endpoint result, the majority of the impact is contributed by electricity.

3.2 LCA of TiO_2/rGO production

In terms of the LCA of the production of TiO_2/rGO photocatalyst, the amount of GO produced in the previous section has been utilized in this section. As mentioned, 1 kg of TiO_2/rGO photocatalyst is the functional unit. Table 4 shows the amount of chemicals and electricity used in this section. However, Table 4 only includes the newly added chemicals, such as ethanol, isopropanol, TiO_2 , etc., but excludes the aforementioned chemicals in section 3.1.

Table 4. The amount of resources used in TiO_2/rGO production.

Use phase		Input	Value	Unit
Production of TiO_2 -rGO	Stirring (Route 1)	Ethanol	198	kg
	Stirring (Route 2)	Isopropanol	198	kg
	Stirring (Route 3)	Ultrapure water	198	kg
	Hydrothermal process	TiO_2	0.99	kg
		Electricity	19	MJ
Additional doping	Doping process	Silver nitrate	0.02	kg
		Deionized water	0.2	kg

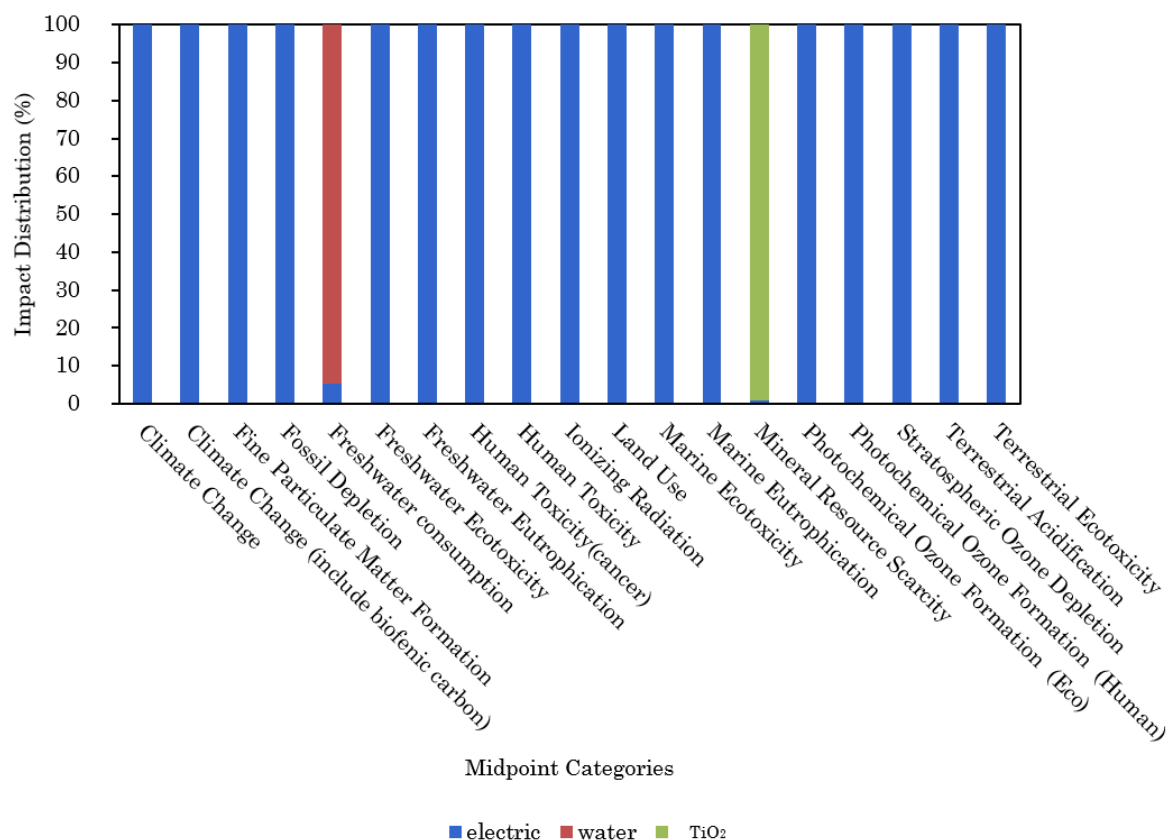


Figure 5. Midpoint data of TiO₂/rGO production using water as a solvent.

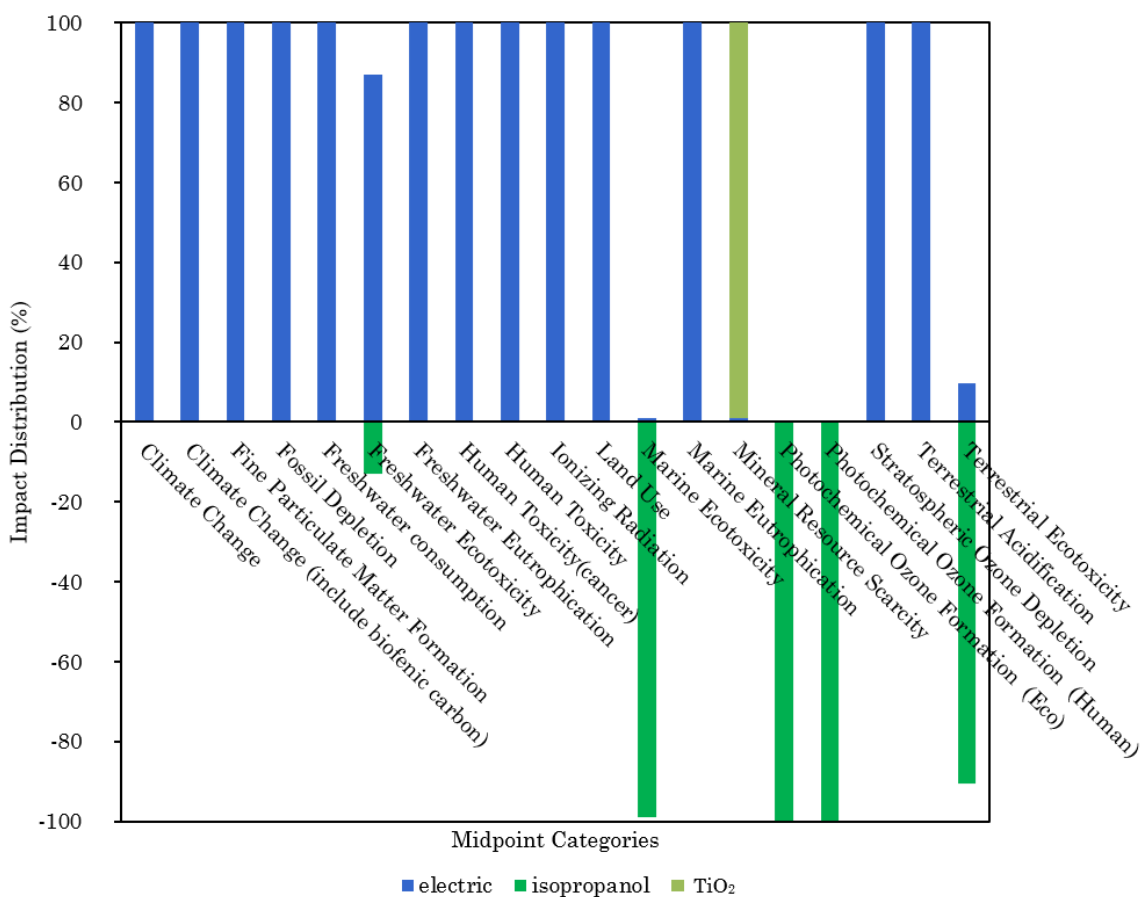


Figure 6. Midpoint data of TiO₂/rGO production using isopropanol as a solvent.

In this production process, the main objective is to compare the environmental impact generated by solvents and determine the one that contributes the least impact to the environment. The chosen solvents were ethanol, isopropanol and ultrapure water. However, in terms of ultrapure water, there is insufficient data in the database, thus fresh water was utilized to replace ultrapure water. Besides this, other chemicals are also discussed in this section. First of all, TiO_2 mainly contributes to metal depletion in the midpoint category and possessed a result of 0.0992 kg Cu eq. TiO_2 certainly contributes to metal depletion since it is a natural resource. One way to reduce the consumption of TiO_2 from the environment is to recycle existing products in the production of TiO_2/rGO photocatalyst. For instance, sintering the TiO_2 -containing waste allowed for the recycling of TiO_2 waste from the paint manufacturing industry [26]. Furthermore, recyclability is also a significant factor that should be considered in the utilization of photocatalyst to reduce environmental impact. For instance, according to Pesqueira *et al.* [9], maximum environmental impact comes with fresh photocata-

lyst being used in each photodegradation. Hence, the environmental impact could be reduced if the photocatalyst is reused.

In terms of electricity, it has dominated most of the impact categories. In this process, the amount of electricity needed was 19 MJ which almost doubled the value of the graphene oxide production process. Hence, it is recommended to provide some solution to further decrease the electricity needed in this process. In this case, the system was conducted at 120 °C for 3 h for the hydrothermal treating process. Hence, the sol-gel method which is a more chemical method can be utilized to reduce energy consumption. It has a relatively low reaction temperature (60–80 °C) and shorter heating duration as compared to the hydrothermal method. This will result in lower electric consumption which means environmental impacts are expected to be reduced [27,28].

3.2.1 Comparison of Solvents

Figures 5-7 depict the environmental impacts associated with the different solvents used. As mentioned, the compared solvents

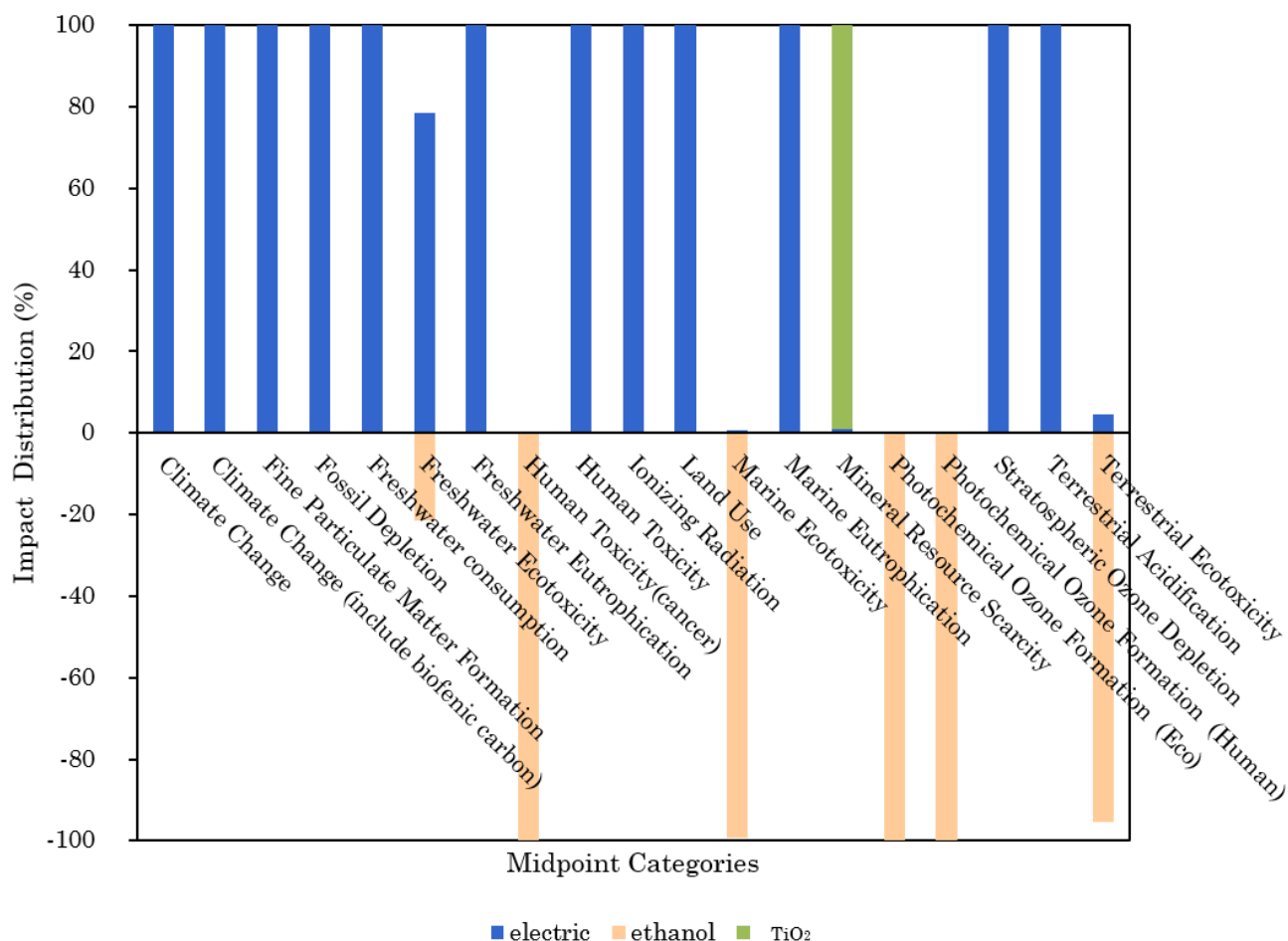


Figure 7. Midpoint data of TiO_2/rGO production using ethanol as a solvent.

were ultrapure water, isopropanol and ethanol. Although ultrapure water is preferable in a laboratory setting, it is more frequently employed in the semiconductor and pharmaceutical industries. Due to its level of filtration, it can be used in a variety of extremely delicate applications. However, ultrapure water was not in the GaBi database, thus it was substituted with fresh water regionalized in Malaysia. As seen in Figure 8, three solvents were compared without adding any other chemicals. Ethanol and isopropanol would first be compared since they contributed to similar impact categories. In categories like freshwater ecotoxicity, marine ecotoxicity, photochemical ozone formation, and terrestrial ecotoxicity, ethanol outweighed all the categories and contributed to an extra impact category which is human toxicity (cancer). Hence, the obvious choice here is isopropanol since it contributed less to every impact category that it shared with ethanol. Another solvent that seems to be the wisest choice is ultrapure water, it only contributed to freshwater consumption. However, since the solvents were not considered in the disposal stage, the generated data for isopropanol and ethanol will be in negative value. Hence, in our system, isopropanol will be the better choice

due to its negative environmental impact. In real world situation, ultrapure water will be the better choice if the disposal stage is considered since it will only contribute to freshwater consumption as compared. Hence, ultrapure water should be considered the better solvent in this case.

3.2.2 Comparison on TiO_2/rGO and $\text{TiO}_2/\text{rGO}/\text{Ag}$

Once the solvent was selected, the environmental impacts of doping the TiO_2/rGO photocatalyst with silver (Ag) were considered. The extra chemicals or substances added were silver nitrate (AgNO_3) and deionized water. As mentioned before, deionized water was substituted with fresh water regionalized in Malaysia due to data scarcity. The reason to dope the photocatalyst with Ag is that doped photocatalysts show better photocatalytic efficiency in terms of degrading organic pollutants [29]. As usual, water only contributed to freshwater consumption while silver nitrate contributed to freshwater ecotoxicity, human toxicity(non-cancer) and marine ecotoxicity. In terms of the value, they were -9.44 kg 1,4 DB eq, -13.8 kg 1,4 DB eq and -10.4 kg 1,4 DB eq respectively.

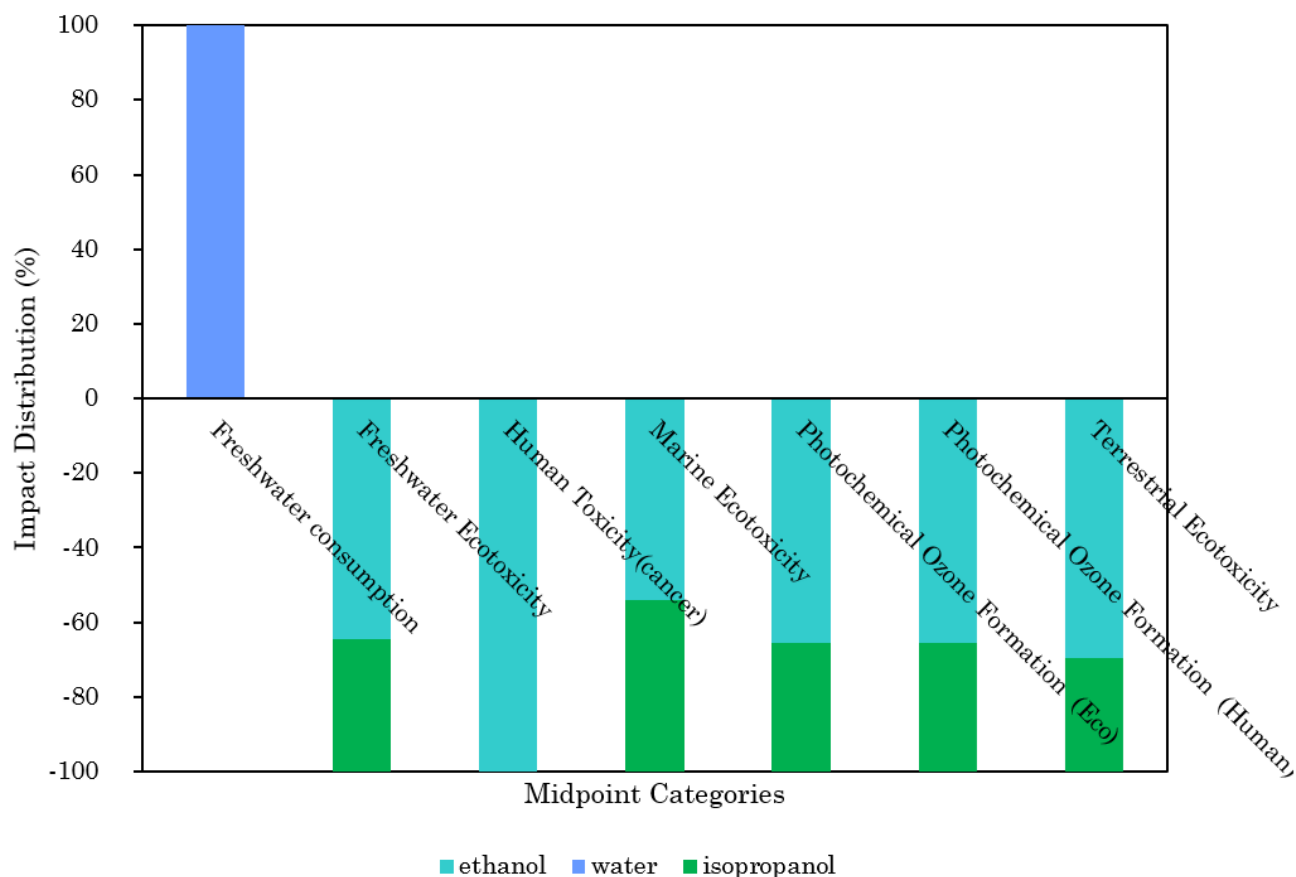


Figure 8. Midpoint data of comparison of solvent in TiO_2/rGO .

As shown in Figures 9 and 10, doping the photocatalyst with Ag certainly contributed a lot to the environment negatively. AgNO₃ contributed more than 90% in all three categories that it is involved in. It has been demonstrated that the composite catalyst's photocatalytic activity was significantly increased by the incorporation of Ag into the TiO₂/rGO structure. The combinatorial effects of greater absorptivity, higher charge separation, decreased charge recombination, and wide-range light absorption may be responsible for this improvement [30]. However, the environmental impact that came with the doping process should not be understated. Hence, to mitigate the environmental came with it, identifying the trade-off between photodegradation efficiency, doping agent composition and operating condition is crucial [31].

3.3 LCA of Photodegradation

Lastly, the LCA of utilizing different power levels of visible light in the photodegradation process is compared and is shown in Figure 11 and Table 5. Compounds with weak bonds that

are vulnerable to UV photodegradation can undergo productive photoreactions when exposed to visible light. Additionally, visible light photoreactions do not require specialized UV photoreactors because they may be carried out

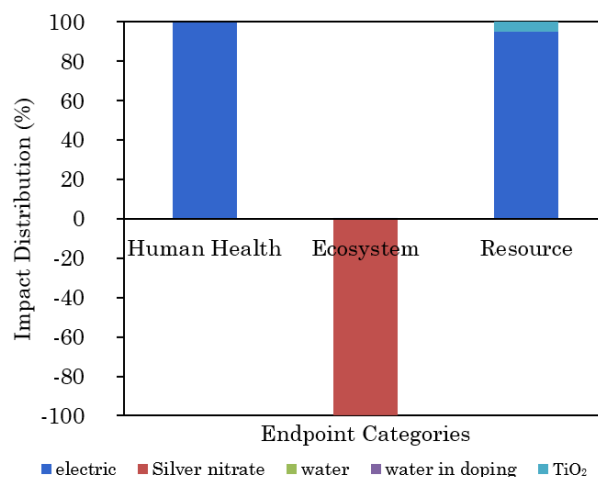


Figure 10. Endpoint result of TiO₂/rGO/Ag production.

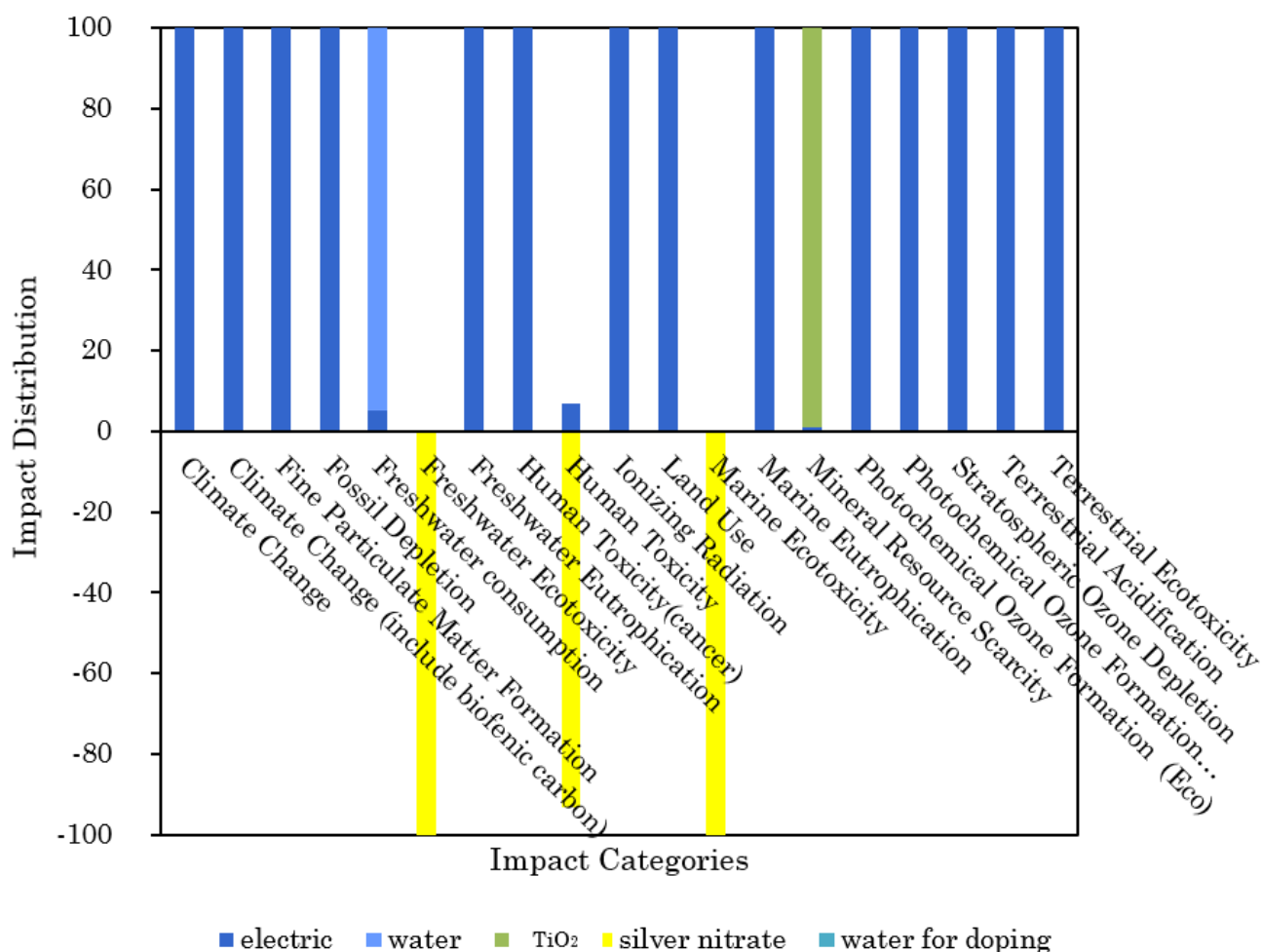


Figure 9. Production of TiO₂/rGO/Ag.

with almost any white light source. For instance, the current development of LED opens up new opportunities for photocatalytic degradation with less power usage and provides a fresh alternative to conventional ultraviolet sources [32]. However, in this study, only the power levels of visible light are considered, which are 150, 300 and 500 W respectively. Commonly, the environmental impacts will increase when the power level increases since the required electricity is higher, considering the same duration. The same can be described in this scenario where 150 W possesses the least environmental impact while 500W possesses the most environmental impact. Hence, the decision came down to the minimum power level to maintain the photodegradation efficiency of the photocatalyst.

Figure 12 also depicts that the environmental impact of three endpoint categories increases when the power level increases. As long as the photodegradation is above average and acceptable range, the lowest possible power consumption should be utilized to reduce the environmental impact contributed by light sources to the furthest extent in terms of human health, ecosystem and resource.

4. Conclusions

This study has assessed the environmental footprints of the photodegradation of methylene blue using a graphene-based TiO_2 photocatalyst via LCA. The determination of the environmental impacts allows the photocatalyst to serve as a sustainable option in the treatment of organic pollutants. The LCA was con-

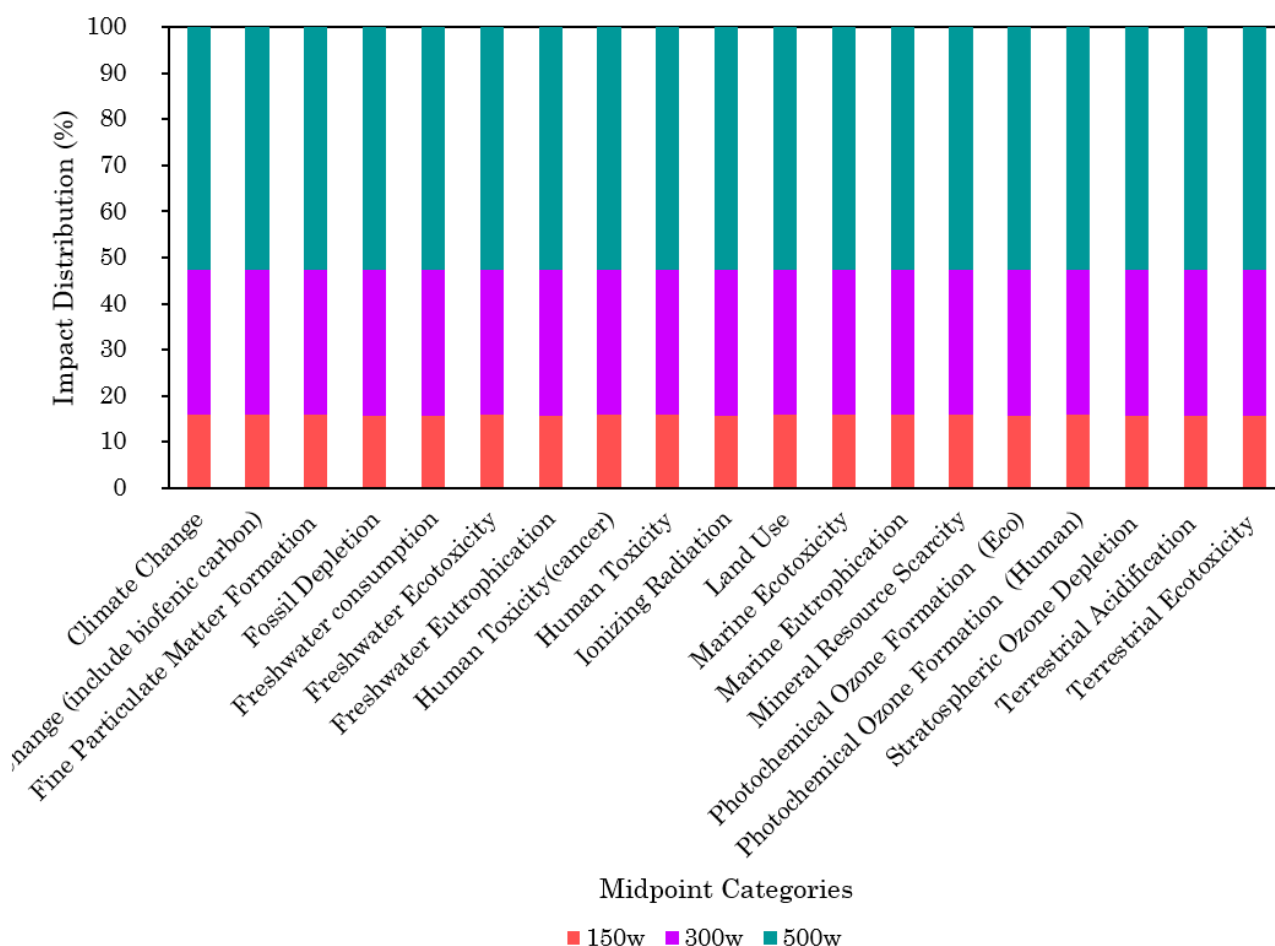


Figure 11. Midpoint data photodegradation using different power levels of light sources.

Table 5. Different power consumption of visible light used in photodegradation process.

Use phase		Input (W)	Value	Unit
Photodegradation (Visible light)	Route 1	150	0.54	MJ
	Route 2	300	1.08	MJ
	Route 3	500	1.8	MJ

ducted through a cradle-to-gate method, which is targeting the synthesis of photocatalyst and photodegradation process. Different chemicals, such as ultrapure water, isopropyl alcohol, and ethanol has been chosen to evaluate their environmental benefits in the synthesis of TiO₂/rGO photocatalyst. In terms of the solvent, ultrapure water is certainly a better choice since it contributed the least to the environment according to the data generated. Also, it is not advisable to dope the photocatalyst with silver since the increment in performance is not enough to offset the environmental impact that came with it. Different powers of visible light were compared, and the result showed that the decision came down to the minimum power level to maintain the photodegradation efficiency of the photocatalyst.

Although the objectives of this study have been met, two recommendations were recommended for this study. Firstly, more chemicals can be compared to gain further insights into the life cycle of the photocatalyst. For instance, oxidizing agents such as H₂SO₄ or H₂O₂ can possibly be replaced by other chemicals like acetic acid and sodium peroxide and could be compared to provide alternative solutions to the photocatalyst synthesis process. Secondly, it is recommended to couple LCA with Artificial Neural Networks (ANN). The coupling allows these tools to work synergistically, whereby the assessment of environmental impacts could be included in an ANN that selects the optimal operating conditions and predict the optimum photodegradation performance. Integrating the strengths of both tools could optimize the TiO₂/rGO performance while minimizing the environmental burdens.

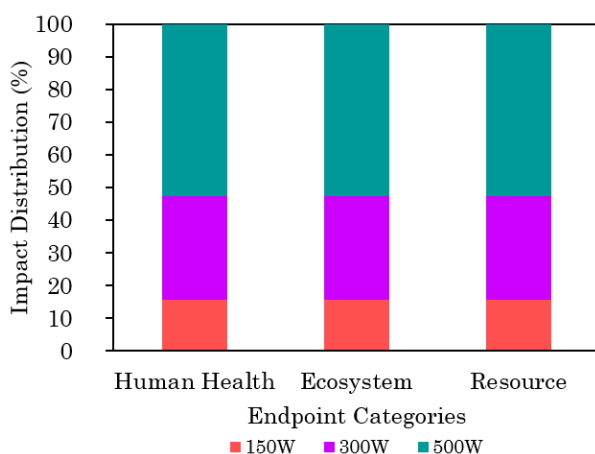


Figure 12. Endpoint data for different power consumption of visible light.

Acknowledgments

The work is supported by Xiamen University Malaysia Research Fund (grant number: XMUMRF/2019-C4/IENG/0019 and XMUMRF/2020-C5/IENG/0029) and Hengyuan International Sdn. Bhd. (grant number: EENG/0003).

CRediT Author Statement

K. Kong: Methodology, Investigation, Formal Analysis, Writing – Original Draft; Y. Weng: Investigation, Validation, Visualization, Writing – Original Draft; W.H. Lam: Conceptualization, Methodology, Investigation, Formal Analysis, Writing – Review and Editing; S.Y. Lai: Conceptualization, Investigation, Validation, Supervision, Project Administration, Funding acquisition, Writing – Review and Editing. All authors have read and agreed to the published version of the manuscript.

References

- [1] Geetha, D., Nagarajan, E.R. (2021). Impact and Issues of Organic Pollutants. In: Singh, P., Hussain, C.M., Rajkhowa, S. (Eds.) *Management of Contaminants of Emerging Concern (CEC) in Environment*, pp. 93–126. DOI: 10.1016/B978-0-12-822263-8.00003-8.
- [2] Schell, L.M., Knutson, K.L., Bailey, S. (2012). Environmental Effects on Growth. In: Cameron, N., Bogin, B. (Eds.) *Human Growth and Development*, pp. 245–286. DOI: 10.1016/B978-0-12-383882-7.00010-6.
- [3] Wang, Y., Pan, T., Yu, Y., Wu, Y., Pan, Y., Yang, X. (2020). A novel peroxymonosulfate (PMS)-enhanced iron coagulation process for simultaneous removal of trace organic pollutants in water. *Water Research*, 185, 116136. DOI: 10.1016/j.watres.2020.116136.
- [4] Drout, R.J., Robison, L., Chen, Z., Islamoglu, T., Farha, O.K. (2019). Zirconium Metal–Organic Frameworks for Organic Pollutant Adsorption. *Trends in Chemistry*, 1(3), 304–317. DOI: 10.1016/j.trechm.2019.03.010.
- [5] Raizada, P., Sudhaik, A., Singh, P. (2019). Photocatalytic water decontamination using graphene and ZnO coupled photocatalysts: A review. *Materials Science for Energy Technologies*, 2(3), 509–525. DOI: 10.1016/j.mset.2019.04.007.
- [6] Zhu, D., Zhou, Q. (2019). Action and mechanism of semiconductor photocatalysis on degradation of organic pollutants in water treatment: A review. *Environmental Nanotechnology, Monitoring & Management*, 12, 100255. DOI: 10.1016/j.enmm.2019.100255.

- [7] Hu, G., Tang, B. (2013). Photocatalytic mechanism of graphene/titanate nanotubes photocatalyst under visible-light irradiation. *Materials Chemistry and Physics*, 138(2–3), 608–614. DOI: 10.1016/j.matchemphys.2012.12.027.
- [8] Chen, D., Cheng, Y., Zhou, N., Chen, P., Wang, Y., Li, K., Huo, S., Cheng, P., Peng, P., Zhang, R., Wang, L., Liu, H., Liu, Y., Ruan, R. (2020). Photocatalytic degradation of organic pollutants using TiO₂-based photocatalysts: A review. *Journal of Cleaner Production*, 268, 121725. DOI: 10.1016/j.jclepro.2020.121725.
- [9] Pesqueira, J.F.J.R., Pereira, M.F.R., Silva, A.M.T. (2021). A life cycle assessment of solar-based treatments (H₂O₂, TiO₂ photocatalysis, circumneutral photo-Fenton) for the removal of organic micropollutants. *Science of the Total Environment*, 761, 143258. DOI: 10.1016/j.scitotenv.2020.143258.
- [10] Fernandes, S., da Silva, J.C.G.E., da Silva, L.P. (2020). Life cycle assessment of the sustainability of enhancing the photodegradation activity of TiO₂ with metal-doping. *Materials*, 13(7), 1487. DOI: 10.3390/ma13071487.
- [11] Garrafa-Gálvez, H.E., Alvarado-Beltrán, C.G., Almaral-Sánchez, J.L., Hurtado-Macías, A., Garzon-Fontecha, A.M., Luque, P.A., Castro-Beltrán, A. (2019). Graphene role in improved solar photocatalytic performance of TiO₂-RGO nanocomposite. *Chemical Physics*, 521, 35–43. DOI: 10.1016/j.chemphys.2019.01.013.
- [12] Tolosana-Moranchel, Á., Manassero, A., Satuf, M.L., Alfano, O.M., Casas, J.A., Bahamonde, A. (2019). Influence of TiO₂-rGO optical properties on the photocatalytic activity and efficiency to photodegrade an emerging pollutant. *Applied Catalysis B: Environmental*, 246, 1–11. DOI: 10.1016/j.apcatb.2019.01.054.
- [13] Gonçalves, B.S., Silva, L.M.C., de Souza, T.C.C., de Castro, V.G., Silva, G.G., Silva, B.C., Krambrock, K., Soares, R.B., Lins, V.F.C., Houmard, M., Nunes, E.H.M. (2019). Solvent effect on the structure and photocatalytic behavior of TiO₂-RGO nanocomposites. *Journal of Materials Research*, 34(23), 3918–3930. DOI: 10.1557/jmr.2019.342.
- [14] Wang, P., Han, L., Zhu, C., Zhai, Y., Dong, S. (2011). Aqueous-phase synthesis of Ag-TiO₂-reduced graphene oxide and Pt-TiO₂-reduced graphene oxide hybrid nanostructures and their catalytic properties. *Nano Research*, 4(11), 1153–1162. DOI: 10.1007/s12274-011-0165-2.
- [15] Doluel, E.C., Kartal, U., Dikici, T., Yurddaskal, M. (2020). Effect of Ag Content on Photocatalytic Activity of Ag@TiO₂/rGO Hybrid Photocatalysts. *Journal of Electronic Materials*, 49(6), 3849–3859. DOI: 10.1007/s11664-020-08102-0.
- [16] Garrafa-Gálvez, H.E., Alvarado-Beltrán, C.G., Almaral-Sánchez, J.L., Hurtado-Macías, A., Garzon-Fontecha, A.M., Luque, P.A., Castro-Beltrán, A. (2019). Graphene role in improved solar photocatalytic performance of TiO₂-RGO nanocomposite. *Chemical Physics*, 521, 35–43. DOI: 10.1016/j.chemphys.2019.01.013.
- [17] Vinesh, V., Shaheer, A.R.M., Neppolian, B. (2019). Reduced graphene oxide (rGO) supported electron deficient B-doped TiO₂ (Au/B-TiO₂/rGO) nanocomposite: An efficient visible light sonophotocatalyst for the degradation of Tetracycline (TC). *Ultrasonics Sonochemistry*, 50, 302–310. DOI: 10.1016/j.ultsonch.2018.09.030.
- [18] Yu, L., Wang, L., Sun, X., Ye, D. (2018). Enhanced photocatalytic activity of rGO/TiO₂ for the decomposition of formaldehyde under visible light irradiation. *Journal of Environmental Sciences*, 73, 138–146. DOI: 10.1016/j.jes.2018.01.022.
- [19] Yu, M., Yu, T., Chen, S., Guo, Z., Seok, I. (2020). A Facile Synthesis of Ag/TiO₂/rGO Nanocomposites with Enhanced Visible Light Photocatalytic Activity. *ES Materials & Manufacturing*, 7, 64–69. DOI: 10.30919/esmm5f712.
- [20] Tan, K.C., Lim, H.S., Mat Jafri, M.Z. (2018). Study on solar ultraviolet erythral dose distribution over Peninsular Malaysia using Ozone Monitoring Instrument. *The Egyptian Journal of Remote Sensing and Space Science*, 21(1), 105–110. DOI: 10.1016/j.ejrs.2017.01.001.
- [21] Rahman, M.M., Adil, M., Yusof, A.M., Kamaruzzaman, Y.B., Ansary, R.H. (2014). Removal of Heavy Metal Ions with Acid Activated Carbons Derived from Oil Palm and Coconut Shells. *Materials*, 7(5), 3634–3650. DOI: 10.3390/ma7053634.
- [22] Freshwater Ecotoxicity as an environmental impact category to guide the selection of chemical-based products - ECETOC. <https://www.ecetoc.org/task-force/freshwater-ecotoxicity-as-an-environmental-impact-category-to-guide-the-selection-of-chemical-based-products/>. Accessed 11 Nov 2022.
- [23] Falkenmark, M., Rockström, J., Savenije, H. (2008). *Balancing Water for Humans and Nature: The New Approach in Ecohydrology*. Routledge. pp. 1–268.

- [24] Energy Commission (2015). *Malaysia Energy Statistic Handbook*. Suruhanjaya Tenaga (Energy Commission) No. 12, Jalan Tun Hussein, Presint 2, 62100 Putrajaya, Malaysia.
- [25] environmental impact of energy — European Environment Agency. <https://www.eea.europa.eu/help/glossary/eea-glossary/environmental-impact-of-energy>. Accessed 11 Nov 2022.
- [26] Bilici, Z., Bouchareb, R., Sacak, T., Yatmaz, H.C., Dizge, N. (2021). Recycling of TiO₂-containing waste and utilization by photocatalytic degradation of a reactive dye solution. *Water Science and Technology*, 83(5), 1242–1249. DOI: 10.2166/wst.2020.606.
- [27] Siong, V.L.E., Tai, X.H., Lee, K.M., Juan, J.C., Lai, C.W. (2020). Unveiling the enhanced photoelectrochemical and photocatalytic properties of reduced graphene oxide for photodegradation of methylene blue dye. *RSC Advances*, 10(62), 37905–37915. DOI: 10.1039/D0RA06703B.
- [28] Modan, E.M., Plăiașu, A.G. (2020). Advantages and Disadvantages of Chemical Methods in the Elaboration of Nanomaterials. *The Annals of “Dunarea de Jos” University of Galati Fascicle IX, Metallurgy and Materials Science*, 43(1), 53–60. DOI: 10.35219/mms.2020.1.08.
- [29] Wang, P., Han, L., Zhu, C., Zhai, Y., Dong, S. (2011). Aqueous-phase synthesis of Ag-TiO₂-reduced graphene oxide and Pt-TiO₂-reduced graphene oxide hybrid nanostructures and their catalytic properties. *Nano Research*, 4(11), 1153–1162. DOI: 10.1007/S12274-011-0165-2.
- [30] Doluel, E.C., Kartal, U., Dikici, T., Yurddaskal, M. (2020). Effect of Ag Content on Photocatalytic Activity of Ag@TiO₂/rGO Hybrid Photocatalysts. *Journal of Electronic Materials*, 49(6), 3849–3859. DOI: 10.1007/S11664-020-08102-0.
- [31] Costamagna, M., Ciacchi, L., Paganini, M.C., Calza, P., Passarini, F. (2020). Combining the highest degradation efficiency with the lowest environmental impact in zinc oxide based photocatalytic systems. *Journal of Cleaner Production*, 252, 119762. DOI: 10.1016/j.jclepro.2019.119762.
- [32] Jo, W.K., Tayade, R.J. (2014). New generation energy-efficient light source for photocatalysis: LEDs for environmental applications. *Industrial and Engineering Chemistry Research*, 53(6), 2073–2084. DOI: 10.1021/ie404176g.