Multi-strategy study of environmental degradation of ibuprofen: from chemical catalysis to biological treatment

Jinrui Wu[1, *], *Youyang* Zhang[2], and *Yuhan* Zhou[3]

[1]College of chemistry, Xiangtan University, China
[2]College of Medicine and Chemical Engineering, Taizhou University, China
[3]International Department, Jinan Xinhang Experimental Foreign Language School, China

Abstract. Ibuprofen, as one of the pharmaceuticals and personal care products, accumulates continuously in the environment with its increasing usage, posing pollution and potential threats to human health through various ways. Therefore, there is a pressing need to explore efficient methods for the degradation of ibuprofen. This paper primarily discusses current chemical and biological degradation approach of ibuprofen, explores the use of catalysts and degradation techniques in different degradation methods, and do comparative analysis of different degradation ways. The advantages and disadvantages of different methods in practice will be discussed and some problems which may encountered during the degradation process will be identified to provide references for further research on efficient degradation methods for ibuprofen.

1 Introduction

Emerging organic contaminants (EOCs) are man-made organic pollutants that do not naturally occur in the environment. Pharmaceuticals and personal care products (PPCPs) are classified as EOCs. Ibuprofen, as a nonsteroidal anti-inflammatory drug with an annual output of thousands of tons, is widely used around the world due to its excellent antipyretic and analgesic activities, ranking as the third most consumed drug globally [1]. Ibuprofen, as a PPCPs, mainly originates from hospitals, domestic sewage, urban landfills, animal feces and urine [2]. With its increasing usage, the accumulation of ibuprofen in soils and various aquatic systems poses numerous pollution hazards to the environment. Despite the awareness of the significant hazards of EOCs and the prohibition or restriction of certain EOCs, their relatively long half-lives enable them to enter the human body through respiration, diet, and skin contact, leading to reproductive developmental toxicity, endocrine disruption, neurobehavioral toxicity [3], and carcinogenic effects, greatly harm people's health. However, existing ibuprofen degradation methods still face limitations such as low degradation efficiency, high cost, and toxicity of by-products. Therefore, it's crucial to develop an efficient degradation method for ibuprofen.

Catalysts, as chemical reagents capable of increasing the rate of chemical reactions by reducing the activation energy, present a new approach to solve such issues. During the degradation process of ibuprofen, the use of suitable catalysts can enhance the efficiency of degradation, reduced degradation time, and lower energy consumption. By designing and synthesizing materials with specific catalytic activities, an efficient and environmentally friendly degradation of ibuprofen can be achieved. Therefore, the search and development of various novel catalysts to address different types of organic pollutant degradation problems are imperative. These catalysts may include metal oxides, nanomaterials, composite catalysts, etc., possessing unique structures and catalytic properties, providing strong support for the degradation of ibuprofen and other organic compounds. This paper will overview the research progress on ibuprofen degradation in recent years, with a particular focus on the important applications of catalysts in ibuprofen degradation. Firstly, we will review the pharmacological activity of ibuprofen as a phenylpropionic acid nonsteroidal anti-inflammatory drug and elucidates the process of ibuprofen entering the natural environment as a pollutant [4]. Simultaneously, this study explores novel efficient catalysts to enhance the degradation efficiency of ibuprofen, reduce energy consumption, and achieve environmentally friendly degradation methods. Currently, catalysts such as highly active Ag/Bi dual nanoparticles-modified BiOBr photocatalyst, Nb_2O_5 sol-gel catalyst, Nb_2O_5/MWCNT nanocomposite material, etc., demonstrate significant advantages in degrading ibuprofen. The paper discusses ibuprofen degradation methods from three different degradation types of systems, including physical degradation, biological degradation, and chemical degradation [5], among which chemical degradation includes photocatalytic degradation and hydrolysis. One method of hydrolysis involves the addition of H_2O_2 to subcritical water to generate OH· radicals, stabilizing the degradation of ibuprofen, with ibuprofen degrading by 88% within 105 minutes. The results indicate that subcritical

*Corresponding author: 202305600544@smail.xtu.edu.cn

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water degradation is a simple, rapid, and environmentally friendly method that does not produce secondary waste streams [4]. Regarding chemical degradation, this paper will discuss in detail the specific applications of different catalysts including metal oxides, nanomaterials, composite catalysts, etc., in photocatalytic degradation and hydrolysis. Additionally, this paper focuses on the research progress of different catalysts' production processes, costs, toxicity, etc. However, there are still several drawbacks to the use of catalytic degradation materials. Firstly, the use of expensive reagents to prepare composite materials through lengthy, multi-step procedures often renders their use non-reproducible and unsustainable. Furthermore, their environmental status is not always positive, as certain types have been demonstrated to possess ecotoxicity [4]. Therefore, exploring more efficient and environmentally friendly catalysts is particularly important. Finally, the development prospects of catalysts applied to ibuprofen degradation are discussed. In summary, the degradation of ibuprofen is related to global ecological security, and efficient degradation of ibuprofen using different catalysts is an important topic in modern ecological conservation.

2. Characteristics of ibuprofen

2.1 Pharmacological properties of ibuprofen

Ibuprofen is widely used for the treatment of various mild to moderate pains, such as headaches, toothaches, and joint pain. Additionally, it is employed to alleviate fever and inflammatory conditions, such as colds, flu, and viral infections. Not only due to its efficacy and relatively low side effects, but ibuprofen also possesses multiple pharmacological properties. Firstly, the analgesic mechanism of ibuprofen primarily involves the inhibition of cyclooxygenase (ibuprofen mainly inhibits both COX-1 and COX-2), leading to reduced synthesis of prostaglandins (PGs) [6], thereby decreasing stimulation of pain receptors to achieve a reduction in pain perception. Moreover, ibuprofen also affects neurotransmitters involved in pain transmission, further alleviating pain sensation. Ibuprofen's anti-inflammatory action involves the inhibition of the release of various inflammatory mediators, including histamine and kinins, during the inflammation process, thereby alleviating the inflammatory response. Additionally, ibuprofen can inhibit the aggregation and activation of white blood cells, reducing the infiltration of inflammatory cells, thus exerting an anti-inflammatory effect. Furthermore, ibuprofen is an effective medication for treating fever symptoms because it can inhibit prostaglandin synthesis in the anterior hypothalamus, exerting an antipyretic effect through the hypothalamic temperature regulation center [7], thereby reducing temperature and restoring normal body temperature. The majority of ibuprofen is administered orally, rapidly absorbed after oral administration, with a high plasma protein binding rate, primarily metabolized by the liver, and its metabolites excreted through the kidneys. Its moderate half-life maintains a stable blood drug concentration in the body, ensuring therapeutic efficacy. However, ibuprofen is not a panacea, and for certain specific conditions such as gastric ulcers and asthma, it should be used with caution or avoided. Although ibuprofen has good therapeutic effects, long-term use should be avoided as it may lead to gastrointestinal mucosal ischemia, decreased defense, and renal vasoconstriction [8]. Therefore, it should be used according to medical advice to avoid overuse.

2.2 Ibuprofen as a pollutant

In addition to its significant medical effects, ibuprofen has gradually attracted attention in recent years as an environmental pollutant. Ibuprofen is an important type of PPCPs, belonging to EOCs. Its ability to exist in water bodies not only affects the balance of aquatic ecosystems but also poses potential threats to human health. Therefore, understanding the characteristics of ibuprofen as a pollutant is crucial for environmental protection and human health. Ibuprofen exhibits the characteristics of "persistent" pollutants in the environment because it belongs to drugs with a long half-life, is not easily volatile, not easily adsorbed by sediment, and has little potential for natural chemical degradation. These characteristics make it difficult for ibuprofen to be removed by natural processes once it enters the environment, leading to its long-term presence. Its presence in the environment can cause toxicity to organisms and carries the risk of teratogenicity and mutagenicity. These impacts are not limited to aquatic organisms; humans and other higher organisms can also be affected through the food chain. Additionally, the presence of ibuprofen in surface water bodies and other types of water bodies disrupts the balance of aquatic ecosystems and affects the sustainable use of water resources. Given the characteristics of ibuprofen as a pollutant and its impacts on the environment and human health, effective management and control measures need to be implemented.

Overall, ibuprofen, as a typical PPCPs pollutant, cannot be overlooked for its characteristics as a pollutant. It has a wide range of sources, stable physical and chemical properties, strong environmental persistence, and toxic effects on organisms. Therefore, it is necessary to strengthen relevant research, develop more effective and scientific management measures, and use more efficient chemical materials to limit the harm of ibuprofen to the environment.

3. Degradation methods of ibuprofen and applications of catalysts

3.1 Chemical degradation

Chemical degradation refers to the process of decomposing pollutants using chemical methods, which plays a crucial role in treating EOCs such as ibuprofen. Through chemical reactions, these recalcitrant organic compounds can be transformed into smaller, less toxic, or more easily biodegradable substances. Chemical

degradation methods mainly rely on the action of catalysts to accelerate the decomposition process of pollutants through catalytic reactions, achieving efficient and low-energy degradation. In the study of ibuprofen degradation, chemical degradation methods have shown their unique advantages, especially in rapidly treating large amounts of pollutants and reducing environmental hazards. Chemical degradation can be divided into various types, among which photodegradation and hydrolysis are two main strategies. Photodegradation relies on light (usually ultraviolet) activating specific catalysts, such as titanium dioxide (TiO_2), to generate free radicals capable of decomposing organic pollutants. This method is suitable for treating ibuprofen pollution in water bodies, with the advantage of high efficiency, but may require additional light sources. On the other hand, hydrolysis processes utilize water molecules to react with pollutants, breaking them down into smaller molecules. This method is very effective for degrading certain pollutants under specific conditions, but its degradation rate and applicability are limited by the chemical properties of the pollutants and environmental conditions. Chemical degradation provides an efficient solution for the environmental treatment of ibuprofen and other EOCs. Through research on different chemical degradation methods, not only can degradation efficiency be improved, and energy consumption reduced, but also the optimal pathway for environmentally friendly degradation can be found.

Ozone catalytic oxidation technology, persulfate catalytic oxidation technology, and photocatalytic oxidation technology have good degradation effects on pharmaceutical and domestic wastewater containing ibuprofen. Ozone catalytic oxidation method has a degradation efficiency of 75% to 99% for ibuprofen, photocatalytic oxidation method has a degradation efficiency of 80% to 95% for ibuprofen, and activated persulfate oxidation technology has a degradation efficiency of 70% to 100% for ibuprofen. Moreover, these methods have fast overall reaction rates, thorough reactions, no secondary pollution, and high development potential [9].

Many oxidation methods can degrade IBP efficiently in a short time, but whether they can become completely safe technologies for removing IBP still needs to consider the toxic side effects of degradation products [10].

3.1.1 Photodegradation

Photodegradation of ibuprofen can be achieved using various metal oxide catalysts such as TiO_2, ZnO_2, Fe_2O_3, etc. The main mechanism involves the oxidation and degradation of ibuprofen by hydroxyl radicals or ozone radicals under catalyst conditions. Samer Khalaf et al. investigated the degradation of ibuprofen by titanium dioxide powder and commercially coated glass active surfaces of titanium dioxide. They successfully reduced the ibuprofen concentration to below 60% within 30 minutes and found that the titanium dioxide catalyst on the glass surface is easy to recycle, making it environmentally friendly [11].

Hayati et al. utilized zinc oxide nanoparticles and activated carbon impregnated with iron oxides (AC-FO) as an effective Z-type photocatalyst (ZACFO). This composite catalyst can inhibit the recombination of charge carriers and accelerate the charge separation of light-induced e^-/H^+ pairs, showing great potential for the removal of ibuprofen, completely degrading 20 mg/l ibuprofen within 20 minutes [12].

Rebollo et al. studied the catalysis of ibuprofen by a hybrid system of hydrotalcite-titanium dioxide ($Ca_2Al(OH)_6Cl{\cdot}2H_2O$) under ultraviolet irradiation. Ibuprofen removal rates reached 96% under ultraviolet irradiation for more than 30 minutes [13].

Advanced oxidation processes (AOPs) involve the oxidation of water and organic pollutants using hydroxyl radicals. Photo-Fenton oxidation is one of the most widely used AOPs and an important reaction in the photodegradation of ibuprofen, achieving both high degradation rates and environmental friendliness [14]. Research on different composite catalysts for Fenton oxidation is at the forefront of ibuprofen photodegradation studies.

Sun et al. investigated a novel photocatalyst, HSO_3-MIL-^{53}(Fe), with acidity-modulating groups, finding that at pH 8 or 9, vis/H_2O_2/HSO_3-MIL-^{53}(Fe) achieved 100% photodegradation efficiency for ibuprofen [15].

Dekkiche et al. reported ibuprofen degradation under Fe(III)-citric acid iron catalysis. They found that with increasing hydrogen peroxide concentration, the kinetic curve resembled first-order decay, with efficiency and reaction rate increasing with increasing acidity [16].

Adityosulindro et al. proposed combining photo-Fenton oxidation with low-frequency ultrasound. Coupling Fenton (visible light, 360-740 nm, 150 watts) with ultrasound improved the degradation rate of the molecule at low Fenton reagent concentrations, but ultrasound had no beneficial effect on ferrous regeneration when the molar ratio of iron to ibuprofen approached [17].

Shi et al. reported significantly improved efficiency in degrading ibuprofen by doping FeOCl with rare earth elements (La and Y), with degradation constants of FeOCl/La and FeOCl/Y being 7.12 times and 5.21 times that of FeOCl, respectively [18].

3.1.2 Hydrolysis

The hydrolysis of ibuprofen involves a nucleophilic substitution reaction, wherein a nucleophilic group (hydroxide ion or water molecule) attacks the electrophilic group (X) in the compound (RX) and replaces the adjacent negatively charged electron-withdrawing group (X). The hydrolysis reaction can be represented by the following equation [2]:

$$RX + H_2O = ROH + HX \quad (1)$$

Under environmental conditions, ibuprofen has very low water solubility, which is a limiting factor for advanced oxidation. Temperature significantly affects the

solubility of nonpolar or moderately polar organic compounds in subcritical water. As the water temperature increases, the polarity of water decreases, leading to increased solubility of hydrophobic organic compounds. However, the solubility of hydrophobic organic compounds in water is also influenced by other water properties such as surface tension, diffusion rate, viscosity, and the structure of the solute. Complex molecular interactions, such as hydrogen bonding between water and solutes, may result in different solubility trends over a wide temperature range. The significant increase in ibuprofen solubility can be attributed to the development of hydrogen bonds between water and ibuprofen [4].

Recent progress has been made in catalytic degradation methods by Zhang et al., who proposed several methods, including hydrolysis under subcritical conditions. Subcritical water hydrolysis involves the oxidation of substances in hot compressed water (100-374°C). Under these conditions, the dielectric constant decreases, making water essentially nonpolar and thus a good solvent for nonpolar hydrophobic substances. $OH\cdot$ radicals generated during the process can effectively degrade ibuprofen, achieving an 88% degradation rate within 105 minutes [4].

Additionally, in the catalytic degradation of ibuprofen by sludge-based nano Fe_3O_4-MnO_2 catalysts, it was found that the optimal dosage of sludge-based catalyst was 0.4 $g\cdot L^{-1}$; H_2O_2 dosage was 3.33 $g\cdot L^{-1}$; and the pH of the degraded ibuprofen solution was 3.3. When the degradation time was 2.5 hours, the degradation rate of ibuprofen by sludge/Fe_3O_4-MnO_2 reached over 95%, while SAC/Fe_3O_4-MnO_2 achieved over 90% degradation. The removal of ibuprofen by sludge-based catalysts mainly relies on the strong oxidative action of hydroxyl radicals, partly derived from the reduction properties of nanomaterials and partly from the catalytic generation of hydroxyl radicals by sludge-based catalysts [19].

The hydrolysis process is relatively simple, safe, and effective for various pollutants, and environmentally friendly because it does not involve the addition of solid or harmful reagents. However, in this process, temperature and pH are crucial factors. Temperature affects the production of $OH\cdot$ radicals [4], while changes in pH alter the rate of drug decomposition and affect the generation rate of active radicals [20].

3.2 Biodegradation

In the process of biodegrading ibuprofen, microorganisms utilize ibuprofen as a source of carbon and energy in their metabolism to achieve ibuprofen degradation. The main degradation mechanism generally involves hydroxylation followed by decarboxylation. Among different microbial processes, aerobic microbial degradation pathways are predominant [21]. Studies have shown that the biodegradation removal rate of ibuprofen can reach over 70%, emphasizing that microbial pathways are the most probable routes for the degradation of pharmaceuticals and PPCPs [22].

Current research on ibuprofen degradation has found that high doses of ibuprofen have an inhibitory effect on microorganisms. When ibuprofen concentration increases, the relative abundance of Firmicutes significantly increases from 0.33% to 40.13%, indicating that Firmicutes can coexist with high ibuprofen concentrations. According to RNA results, characteristic bacteria capable of degrading certain organic compounds can be detected. In the study, phyla of proteobacteria, actinobacteria, bacteroidetes, and ascomycota were detected. Among them, the relative abundance of Proteobacteria was over 90%, indicating its key role as a microorganism [21]. Experiments by Tran et al. showed that the glucose utilization efficiency in the upstream one-third of the wetland (i.e., COD removal significantly) promoted the proliferation of various bacteria. Among these bacteria, some non-ibuprofen-degrading bacteria may tend to produce enzymes or auxiliary factors for the cometabolic degradation of ibuprofen into intermediates such as 2-OH ibuprofen [23]. Similarly, in areas strongly influenced by high dissolved oxygen in influent water, ibuprofen may be oxidized by enzymes or auxiliary factors produced by glucose metabolism bacteria (or the oxidation of glucose metabolites). This may greatly contribute to the significant removal of COD (glucose) and ibuprofen in that area [24]. Therefore, it is believed that the biodegradation of ibuprofen depends on the potential of microorganisms to produce specific enzymes for ibuprofen degradation [5].

Furthermore, wastewater pharmaceutical concentrations can also be reduced through biological adsorption and bioremediation, which offer various advantages such as simple process design, environmental friendliness, low initial cost, high energy efficiency, cost-effectiveness, and low sludge generation. Bioremediation is a process of treating contaminated media by altering environmental conditions to stimulate the growth of microorganisms (bacteria, fungi, and algae) for the degradation of target pollutants. Biological adsorption is a process in which substances are adsorbed onto the surface of a biological entity through covalent, electrostatic, or molecular forces. Various adsorbents and their modified forms have been used to remove ibuprofen from wastewater, including wood apple shell, coconut shell, olive stones, mung bean husk, sugarcane bagasse, and date stone seeds derived sorbents [25].

4. Comparative analysis of ibuprofen degradation methods

In analyzing the chemical degradation process of ibuprofen, especially concerning methods involving the use of metal catalysts, it is essential to consider their environmental and economic impacts comprehensively. Although metal catalysts effectively accelerate the chemical reactions of ibuprofen, they often generate significant amounts of metal residues in practical applications. This not only increases the cost of catalyst recovery but also poses long-term pollution risks to the environment. For instance, certain metal catalysts require special treatment after use to prevent heavy metal contamination, a process that is both complex and costly.

Ozone oxidation has garnered attention as an efficient ibuprofen degradation method due to its high degradation rate and environmentally friendly reaction products. This method involves the reaction of ozone with ibuprofen, producing small molecules that are easy to handle or low in toxicity, thereby reducing environmental impact. However, the drawback of this method lies in its direct correlation between ibuprofen removal rate and ozone input. Additionally, the cost of catalysts is relatively high, especially when large-scale applications are necessary. In contrast, Fenton oxidation is widely researched due to its cost-effectiveness. This method utilizes iron salts and hydrogen peroxide to generate hydroxyl radicals in acidic conditions, effectively breaking down the chemical structure of ibuprofen. While Fenton reaction demonstrates high efficiency in rapidly treating large quantities of pollutants, it has strict pH requirements and typically operates under acidic conditions, which may limit its application in certain environments. Furthermore, pH adjustment and control also increase operational complexity and costs.

In the process of photocatalytic degradation of ibuprofen, metal oxide catalysts such as TiO_2, ZnO_2, and Fe_2O_3 exhibit varying catalytic activities. Specifically, TiO_2, under ultraviolet light irradiation, can effectively generate free radicals, promoting the photodegradation of ibuprofen. Experimental results show that TiO_2 can reduce the ibuprofen concentration in solution by 80% within 60 minutes. Additionally, while ZnO_2 and Fe_2O_3 can also catalyze the decomposition of ibuprofen, their degradation efficiencies under the same conditions are 65% and 50%, respectively, indicating the superiority of TiO_2. However, photocatalytic degradation typically relies on specific wavelengths of light, such as ultraviolet light, implying that the efficiency of photocatalytic degradation may significantly decrease in the absence of natural or artificially effective light sources. If artificial light sources, such as UV lamps, are used, the photocatalytic degradation process may consume a large amount of energy, increasing both processing costs and environmental energy burdens. Furthermore, commonly used photocatalysts in photocatalytic degradation, such as titanium dioxide, may face challenges in their recovery and reuse. Catalysts may experience reduced activity due to repeated use or difficulty in complete recovery from the reaction system during processing.

Supercritical water degradation technology has been proven to have efficient degradation capabilities for both single pollutant model solutions and complex actual wastewater containing organic pollutants. Under supercritical conditions, the physical and chemical properties of water change, enhancing its solvent capabilities, while generating highly active free radicals such as hydroxyl radicals, which effectively break down organic pollutants like ibuprofen. This technology is not only applicable to laboratory single pollutant systems but has also been proven highly effective in microbial degradation processes for treating ibuprofen and other pharmaceuticals and PPCPs. By utilizing microorganisms to metabolize organic compounds into inorganic substances, this method exhibits high removal rates in artificial wetland environments.

However, the process of microbial degradation is not without risks. Some strains may produce metabolites during the degradation of ibuprofen that have toxic effects similar to ibuprofen, which may pose new ecological risks in aquatic environments. Therefore, improving the efficiency and safety of microbial degradation is a key focus of current research. To overcome these challenges, research needs to identify or develop more efficient and ecologically safe microbial strains capable of degrading ibuprofen efficiently without producing harmful by-products. Additionally, integrating physical and chemical methods to pretreat pollutants is a promising direction, which can reduce the pollutant load entering the biological treatment system through adsorption, filtration, and other means, thereby increasing overall degradation efficiency. Furthermore, developing new bio-chemical combined degradation technologies, such as integrating bio-adsorption with microbial metabolic processes, can not only increase treatment speed but also enhance system stability and treatment range.

In summary, by optimizing microbial selection and enhancing system design, the capability of artificial wetland systems to treat pollutants such as ibuprofen can be significantly improved while reducing ecological risks, thereby advancing this technology towards broader applications. Overall, each of these degradation methods has its advantages and limitations, and the selection of the appropriate method depends on specific application scenarios, economic budgets, and considerations of environmental impact. Future research can focus on developing more economical and environmentally friendly catalysts or improving the recovery and reuse techniques of existing catalysts to enhance the overall sustainability and practicality of these chemical degradation methods.

5. Conclusion and outlook

This paper comprehensively explores various techniques for ibuprofen degradation, with a focus on analyzing the characteristics and differences between chemical and biological degradation technologies in terms of efficiency, cost, and environmental impact. Chemical degradation, utilizing metal oxide catalysts such as TiO_2, FeO, as well as nanomaterials and composite materials, demonstrates its ability to rapidly treat pollutants. However, it also faces challenges related to catalyst cost and post-treatment environmental burdens. Photodegradation and Fenton reaction exhibit efficient degradation performance, yet they require energy consumption and dependence on operational environments, necessitating the development of new technologies to address these issues.

Biological degradation offers an environmentally friendly solution by utilizing microorganisms to convert ibuprofen into harmless substances. Particularly, certain bacteria, such as Proteobacteria, demonstrate high efficiency in microbial degradation. However, the potential risks of toxic metabolites produced under specific conditions cannot be ignored. Biological adsorption and remediation techniques, employing natural organisms and biochar as adsorbents, provide new

directions for low-cost and sustainable pharmaceutical treatment, although further research on ecological risks is warranted.

Future research should focus on optimizing existing methods and exploring new degradation technologies to achieve more efficient and environmentally friendly ibuprofen degradation strategies. For instance, research should aim to develop catalysts that are low-cost, efficient, and have minimal environmental impacts, especially which can operate effectively under natural environmental conditions. By integrating technologies, such as combining chemical degradation with biological treatment and leveraging the rapid reactivity of chemical methods and the low-cost advantage of biological methods, we can improve overall efficiency in degradation and economy.

Through these studies, a better understanding and utilization of these technologies for treating ibuprofen and similar persistent organic pollutants can be achieved to provide solid support for environmental protection and public health. In summary, future research directions should focus on achieving the commercialization of these technologies, ensuring their sustainable application, and eventually achieve a win-win situation of environmental and economic benefits.

Authors contribution

All the authors contributed equally and their names were listed in alphabetical order.

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