

Post-Combustion Coal Desulfurization : Review

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Artikel histori :

Diterima 12 September 2020
Diterima dalam revisi 23 Oktober 2020
Diterima 29 Oktober 2020
Online 31 Oktober 2020

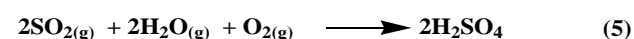
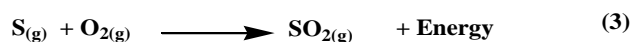
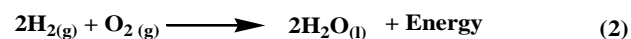
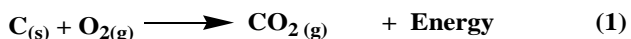
ABSTRACT: The sulfur content in coal ranges from 0.5 to 5%, and it is an impurity that must be removed before burning coal, due to the toxic effects and acid rain caused by SO₂ gas generated from sulfur oxidation. Sulfur cleaning technology can be carried out on coal before combustion (pre-combustion), and can reduce the inorganic sulfur content by up to 50%; however, it cannot reduce the organic sulfur content. The aim of the coal desulfurization review is to direct desulfurization research to be efficient and effective, as well as to be environmentally friendly. Sulfur in coal consists of organic sulfur and inorganic sulfur. Coal inorganic sulfur consists of pyrite (FeS₂), sulfate, and sulfide. Organic sulfur consists of thiophenic, aromatic sulfides, and disulfides. Inorganic sulfur can easily be separated from coal by means of flotation, microwave energy, magnetic forces, ultrasonic energy, and as well as microorganisms. Organic sulfur can be removed by chemical and biodesulfurization, however, this process is inefficient for industrial scale. The most recommended method is the absorption of SO₂ gas in the gas from coal combustion, or post-combustion desulfurization. Various methods have also been investigated to separate SO₂ gas, and more details will be described in this paper.

Keywords: acid rain; coal desulfurization; electrochemical desulfurization

1. Introduction

1.1 Effects of sulfur on the environment

Burning coal using oxygen aims to produce energy released by the reaction of the elements in coal, namely carbon (C), sulfur (S), hydrogen (H), Nitrogen (N), and Oxygen (O). The coal burning reaction are exothermic reaction. Gas from coal combustion contains components resulting from the oxidation reaction of the constituent elements of coal. The chemical reaction that occurs in the coal burning process is the oxidation reaction of the elements C, H, O, S, and N. The equation for the oxidation reaction of the elements in coal is shown in Equation (1) to Equation (5).



Equations (1) to Equation (5) show that the products of coal combustion are SO₂, NO_x, SO_x, CO_x, H₂O, and CO₂. The gas mixture will produce sulfuric acid when exposed to rain so that the pH reaches 3 to 4. All of these gases have

toxic effects on humans, as well as corrosive effects and irritation to equipment and skin, caused by sulfuric acid. Therefore, coal processing must be done before it is converted into energy or chemicals, especially to remove sulfur. The reduction or removal of sulfur content is known as the desulfurization process.

Desulfurization process can reduce the sulfur content of coal, however, on the other hand it can reduce the calorific value of coal. Therefore, the study of the desulfurization process, which can reduce sulfur levels and increase the calorific value of coal, needs to be continuously developed. It is necessary to figure out the forms of the chemical structure of sulfur in coal before explaining the various desulfurization processes, since it aims to determine the type of desulfurization process that is suitable.

1.2 Forms of sulfur in coal

Coal has a complex chemical structure and chemical-physical properties. The chemical-physical properties of coal are represented using several parameters that determine the quality of the coal (Russo, C., et al., (2019). These parameters are: Calorie Value, Total Moisture, Inherent Moisture, Total Sulfur, Ash Content, Fixed Carbon, and Volatile Matter. The elements of coal generally consist of C, H, O, N, S, and the composition varies depending on the type of coal or the source of the coal mine. The highest C content is found in Anthracite coal (93%), and the lowest is in peat coal (55%). There is carbon content in coal types of the

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Lignite (73%) and Bituminous (84%) (Maidi and Yunasril, 2020).

The types of sulfur in coal are described by Calkins (1994). According to Calkins, the chemical sulfur molecules in coal are in the form of inorganic sulfur and organic sulfur. The most inorganic form is in the form of pyrite (FeS_2) as the main inorganic sulfur contaminant in most of the coal. Apart from pyrite, there are also sulfur compounds, free sulfur, and sulfates. Currently, commercially available inorganic coal sulfur analysis and desulfurization methods are available.

From other literature, information is obtained that sulfur in coal can be divided into two categories, namely organic and inorganic sulfur. In general, sulfur in coal is mostly inorganic when the sulfur content in coal is $>2\%$, and organic when the coal sulfur content is $<0.5\%$. The Chinese National Standard classifies coal with a total sulfur content of $>3\%$ as high-sulfur coal.

Therefore, desulfurization of high-sulfur coal should be directed towards removing inorganic sulfur. Pyrite is the largest component of inorganic sulfur in coal. Although elemental sulfur and sulfur sulfate are also constituents of inorganic sulfur in coal, they are generally not considered in the calculation of inorganic sulfur due to their very-low content. Therefore, the desulfurization of high-sulfur coal is focused on removing the pyrite (Liu, F., et al., 2020).

The structure of organic sulfur in coal is mainly a component that has a macromolecular structure, and is uneasily separated from the coal's structure. Two types of methods have been developed to determine organic sulfur levels, namely destructive methods which include pyrolysis and catalytic reduction processes with their products, namely H_2S ; the product of the sulfur oxidation reaction is SO_2 . Meanwhile, the non-destructive method uses the X-ray method which can directly determine the type of sulfur (aliphatic, aromatic, or thiophenic).

There are three chemical structures in organic sulfur components, namely aliphatic, aromatic, and heterocyclic sulfur. Aliphatic types are less stable at high temperatures. At high temperatures in the pyrolysis process, aliphatic sulfur will break down to produce H_2S , and some of it becomes a more stable heterocyclic structure. Heterocyclic are typically thiophenic, that is, ring structures, and the number of rings varies from a single ring structure to six rings. Apart from sulfur Nitrogen is also present in the heteroatom structure of nitrogen and/or oxygen. Many of the heterocyclic sulfur structures contain alkyl substituents of varying sizes and complexity.

Aromatic sulfides and disulfides are present in high sulfur containing coal, yet usually in small amounts compared to heterocyclic sulfur compounds. Low-sulfur coal obtains sulfur mainly from the sulfur component in coal-forming mines. However, high-sulfur coal is now known to derive most of its sulfur from the reduction of sulfate ions to H_2S in the ocean or brackish water in coal seams through microbial processes. Hydrogen sulfide (H_2S) chemically enters peat during diagenesis. Hydrogen sulfide H_2S also reacts with iron ions in water to produce pyrite in coal. Organic sulfur is sulfur bound to the carbon structure of coal, therefore, it cannot be released in pre-combustion desulfurization.

Inorganic sulfur, which is usually in the form of FeS pyrite, is the sulfur that is easily separated in pre-combustion desulfurization using flotation, magnetic, microwave energy, or ultrasonic wave energy.

Another method for removing inorganic sulfur by pre-combustion desulfurization method uses commercial methods with coal washing, for example, using the flotation and oil agglomeration methods. Research to remove organic sulfur is continuing, however until now, there has been no economical desulfurization process to remove chemicals from all organic sulfur components of coal. One of the methods to remove aliphatic organic sulfur is by chemical and biological means, yet these two methods are inefficient because of the high investment costs.

Chemical methods can digest the chemical structure of aliphatic organic sulfur by reaction with very strong bases. In this reaction, the heterocyclic sulfur structure reacts selectively with reagents which can cause single electron transfer such as potassium naphthalene. Apart from chemical methods, certain species of microorganisms have been shown to consume organic sulfur in coal, and to selectively convert or change some of the sulfur components of coal (Calkins, W. H., (1994). An efficient method for removing organic sulfur is post-combustion desulfurization, which is the separation of toxic gases in the gas stream from burning coal before being discharged into the environment.

Post-combustion desulfurization is used in almost all industries that use coal as fuel. The processing method can use physical and chemical methods. Table 1 shows the post-combustion desulfurization method that has been carried out.

2. Literature Review

Post-desulfurization of coal is desulfurization to remove sulfur in the gas from coal combustion. The desulfurization process can be carried out physically, chemically, and a combination of physical-chemical methods. The physical method uses adsorbents, and the chemical method uses chemicals that can react with SO_2 gas to produce new components that can be separated from the exhaust gas mixture resulting from coal combustion. SO_2 gas in the gas from coal combustion is a gas produced by the oxidation reaction of organic sulfur in coal. Organic sulfur cannot be removed by using coal pre-combustion desulfurization processing since sulfur is bound to coal macromolecules, therefore, physical treatment cannot break the bonds between sulfur and coal macromolecules. Organic sulfur can be removed using alkaline chemicals or strong acids, however, it requires high temperature and pressure conditions so it is not economical. To overcome this problem, organic sulfur is separated after the coal is burned at high temperatures, namely at coal combustion temperatures that exceed 600°C . Sulfur bound to the macromolecule will be released and react with oxygen, producing SO_2 gas. Sulfur dioxide gas (SO_2) is a very dangerous, poisonous gas that causes acid rain. Acid rain has a very low pH, between 3 and 4, of course, it can kill fish, trees, and damage buildings and can damage historic

artifacts. SO₂ gas is the most toxic gas for humans, causing various diseases such as bronchitis, conjugative inflammation, and emphysema. Therefore, efforts to remove SO₂ gas from the gas from coal combustion are very important to save humans from respiratory diseases.

2.1. Organic sulfur desulfurization problems

The main problem with coal utilization is the presence of sulfur impurities, both organic sulfur and inorganic sulfur. Sulfur or sulfur in coal can be removed or controlled using three main methods, namely (1) removal of sulfur when coal is used, (2) adding sulfur-binding chemicals during the coal combustion process, and (3) processing or absorption of sulfur present in the flue gas after combustion. For the coke-making industry, desulfurization of coal is important because sulfur has a negative effect on the quality of the coke product, thus, if the coal is to be used to produce coke, pre-combustion of coal is very effective. Although cleaning sulfur after coal is burned can also remove organic sulfur and

inorganic sulfur, the desulfurization process requires expensive investment costs (Xia et al., 2018).

The advantage of post-combustion desulfurization is that it can remove both organic sulfur and inorganic sulfur. Pre-combustion desulfurization can be used to remove inorganic sulfur such as pyrite; the method used is physics. Pyrite is a type of sulfur integrated with minerals, while organic sulfur is bound to carbon. If coal is crushed past a certain size, the minerals that bind sulfur will be separated from coal and also from organic sulfur. Furthermore, inorganic sulfur can be separated by gravity with the aid of a magnetic force, or using a flotation method which has been proven to be used for desulfurization of inorganic sulfur. In other words, the gravity, magnetic, and flotation methods can clean coal inorganic sulfur, however, they are unable to separate organic sulfur.

In the last few decades, microwave energy has been widely used in the pre-combustion desulfurization process to remove inorganic sulfur. Later, other researchers used the microwave together with the addition of acid/alkaline chemicals.

Table 1. Post-combustion desulfurization

No	Method	Conclusion	Reference
1	The absorption method uses Iron oxide (FeO) assisted by microwaves	To remove H ₂ S from gas from coal combustion (syngas).	Feng et al. (2020)
2	Wet desulfurization uses water as a solvent to absorb SO ₂ and Hg gases	The wet desulfurization process uses solvents that can dissolve impurity gases in the gas from coal combustion, able to remove sulfur dioxide (SO ₂) and water-soluble mercury compounds, for example, HgCl ₂ from other impurity gases in flue gas originating from coal.	Heidel and Klein (2016)
3	Wet desulfurization using solvent I ₂	A new method of desulfurization of wet exhaust gas based on the Bunsen reaction is the sulfur-iodine (SI) thermochemical cycle. The I ₂ and HI absorption systems are used to remove SO ₂ from coal-fired exhaust gases. The SO ₂ removal efficiency was around 98.8% at a concentration of I ₂ = 25.6 mmol/L. The absorption products are H ₂ SO ₄ and HI, which can be easily separated by distillation. Compared to traditional wet flue gas desulfurization (FGD) by-products, such as gypsum or magnesium sulfate, H ₂ SO ₄ has better commercial value, and HI can be used as a feedstock for hydrogen production. Researchers claim that this study is a promising technology for the removal of SO ₂ from coal burning.	Zhu et al. (2017)
4	Dry desulfurization	Researchers claim that dry desulfurization is more efficient than wet desulfurization process. The disadvantage of the wet process is that it is more corrosive to equipment than the dry process. The second disadvantage is that the dissolved solvent and gas form lime slurry. This sludge is erosive or can erode the surface layer of the equipment so that the equipment used requires certain types of materials to avoid corrosion and erosion.	Gong and Yang (2018)
5	Desulfurization using water absorber	This method is a wet desulfurization method using a double rotation sprayer, aimed at increasing the efficiency of absorption of sulfur dioxide gas in coal combustion gas which has a high sulfur content.	Qin et al. (2019)
6	Calcium formate as additive in fuel gas desulfurization in power plants	Calcium formate is the salt used in the wet desulfurization process in order to avoid the formation of calcium carbonate in the desulfurization reactor and to increase the efficiency of the desulfurization. Because it can reduce the formation of lime sludge.	Li et al. (2017)

However, the removal of organic sulfur from coal does not appear to be possible using flotation-based physical separation methods since organic sulfur is usually integrated into the structure of the organic macromolecular skeleton of coal. Therefore, to remove organic sulfur, one has to use chemical leaching, oxidation reactions, and bio-desulfurization. The bio-desulfurization process is very slow so it is suitable for laboratory scale, however, it is difficult to apply on an industrial scale.

Lignite coal is mostly processed into coke through the pyrolysis process since pyrolysis is considered an environmentally friendly process in utilizing low-calorie coal. Low-calorie coal pyrolysis is to obtain liquid or gaseous products, or to obtain chemicals or tar. Since low-calorie coal has a high ash content, high humidity, and high sulfur content, many types of processing must be done before coal is used, namely desulfurization, ash removal, and moisture reduction. To eliminate pre-processing, it is advisable to use a pyrolysis process so that organic carbon will decompose at high temperatures, resulting in a gas that is released from the macromolecular structure. Gu et al., (2017) found that under conditions of temperature and pressure of pyrolysis reactions, alkyl sulfide, thiophene, and aryl sulfide in coal, break down into sulfur-containing gases, and are released from coal when the pyrolysis temperature reaches 700 to 900 °C. Decomposition of pyrite occurs when the pyrolysis temperature exceeds 300 °C. In addition, Xia et al. (2018) found that the magnetism of pyrite also increased during the pyrolysis process. Therefore, the coal pyrolysis process, when combined with dry magnetic separation, can remove pyrite from coal effectively. The application of dry magnetic separation is beneficial and economical in coal cleaning since dry separation can save water resources. However, the combination of pyrolysis and dry magnetic separation is still unable to remove organic sulfur from coal.

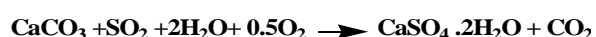
2.2 Organic sulfur separation from coal

2.2.1 Conventional post-combustion desulfurization

Research on the post-combustion desulfurization process is a conventional desulfurization with the aim of reducing levels of SO₂ and mercury metal in coal combustion gases before being discharged into the environment. There are two types of SO₂ absorbent media, namely dry media and wet media. The limestone-gypsum Wet Gas Desulfurization (WFGD) technology is the most widely used FGD technology in the world. Fuel Gas Desulfurization can control sulfur dioxide from coal-fired stations, and can also be effective in capturing volatile pollutants such as F, As, B, Cl, Se or Hg in the form of agase. Apart from limestone, there are other studies using zeolite, where the zeolite is mixed with coal before the coal is burned, then the gas produced from coal combustion is analyzed to determine the SO₂ content and other gases such as Hg (mercury), then the analysis results are compared with the conditions without zeolite. From the results of this study, it can be concluded that zeolite can absorb 6 to 8% SO₂; besides SO₂, zeolite can absorb Hg.

Post-combustion desulfurization can be classified into two, the single-use system and regeneration system, depending on the technique of handling the absorbent material after the desulfurization process. The single-use system treats used sorbents as waste or as a by-product. The regeneration system recycles or regenerates the SO₂ sorbent which is further processed to produce sulfuric acid (H₂SO₄) or SO₂ liquid. No waste is generated in the regeneration system. The two systems can be further subdivided into wet system or dry system, depending on whether the sorbent is aqueous or solid state. In the wet process, the flue gas leaving the FGD is saturated with water, and will form a slurry which can be used as a by-product.

In the dry process, dry sorbent is mixed with coal to be burned. The dry process produces sorbent waste that has absorbed SO₂, and is used as a by-product. The resulting gas is dry or unsaturated gas. The cost required for one process is lower than the regeneration process, however, the regeneration process is chosen if there is no land available to dispose of waste. The results of literature search show that the separation of coal organic sulfur is more effective using post-combustion desulfurization. The following sections describe several methods for removing organic sulfur. Single-use WFGD technology is the most widely used technology worldwide, accounting for 95% of installed capacity. WFGD is often further categorized based on the type of reagent used as an absorbent material. Limestone is commonly used as the reagent of choice for WFGD. However, the WFGD system can also be designed for different reagents, including lime, magnesium-lime, ammonia, sodium carbonate, and seawater. In addition, the WFGD system is successfully used for a wide variety of coal types such as lignite, bituminous, anthracite, and sub-bituminous. In a conventional WFGD system that uses limestone as an absorber, i.e., a stack, the gas enters the spray tower where the limestone slurry is sprayed to absorb SO₂, and produces gypsum for disposal or reuse as a byproduct. The overall reaction is written as:



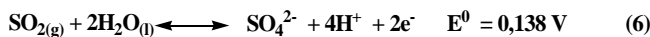
One example of DFGD (Dry Fuel Gas Desulfurization) is a dry method using Fe (and assisted by magnets). Organic sulfur in coal is difficult to remove using gravity separation, oil agglomeration, and flotation. Although chemical leaching/oxidation and bio-desulfurization by bacteria are considered efficient methods of removing organic sulfur from low-rank coal, these chemical and biological desulfurization technologies are usually difficult to quickly implement in the coal cleaning industry (Xia et al., 2018).

The desulfurization method using the dry method has been carried out with the help of magnets. In this research, iron (Fe) powder is reduced, then added in order to act as a promoter to increase coal desulfurization. The coal is preheated with Fe powder, and the separation of the Fe which has bound the organic sulfur is continued to dry magnetic separation. Organic sulfur in coal can bind with Fe, and forms a magnetic compound bound by Fe which can be effectively separated by dry magnetic separators (Xia et al., 2018).

Organic sulfur in coal can bind with Fe and form Fe-related magnetic compounds which can be separated effectively by dry magnetic separators. Hence, Fe powder acts as a promoter to enhance the removal of organic sulfur from coal. This method is effective for application in coal cleaning, especially for desulfurization of coal with high sulfur. Compared to the chemical method using acid or base or the bio-desulfurization method for the separation of organic sulfur, the dry magnetic method is more environmentally friendly because the addition of acids or bases not only requires the cost of chemicals, e.g., acids, alkalis, etc., but also increases the environmental load because chemical liquid waste also needs to be processed. For coal bio-desulfurization, suitable microorganisms need to be found, and usually the bio-desulfurization efficiency is not good enough for industrial applications (Ye et al., 2018).

2.2.2 Electrochemical post-combustion desulfurization

Several post-combustion desulfurization methods have been developed not only for desulfurization of gas from coal combustion in the power generation sector, but have now been developed to clean exhaust gases from the combustion of vehicle fuels from diesel, gasoline and aviation fuel; this technology is known as CO₂ capture. Various technologies were also developed specifically to remove SO₂ gas, including using electrochemical methods and using membranes. The principle of the electrochemical method can be seen in Figure 1. The SO₂ gas that dissolves in water can be converted to H₂SO₄ through the Reaction Equation (6):



The first step of electrochemical desulfurization is the adsorption of SO₂ into water to become a soluble SO₂ solution in water. Furthermore, SO₂ will be reduced to sulfate ions. The sulfate ion reacts with water to give sulfuric acid. The solubility of gases in water is limited by equilibrium at a certain temperature. As in the Reaction Equation (6), that the reaction of SO₂ with water is an equilibrium reaction. To change the equilibrium so that the reaction shifts to the right, it is necessary to reduce the product concentration by separating the product (H⁺) and electrons. Efforts to separate or reduce the concentration of the product can be done electrochemically using electrochemical cells. The electrochemical cell consists of two compartments, namely the anode side and the cathode side. Between the anode and cathode there is an electrolyte membrane known as the proton exchange membrane, which is a solid, negatively charged form (See Figure 1). The function of the anode is to reduce sulfur dioxide, and with the help of water molecules, it will turn into SO₄²⁻ ion, protons (H⁺), and electrons. The positively charged protons will pass through the electrolyte membrane which is negatively charged, while the electrons cannot pass through the membrane and leave the cell as a source of DC electricity. On the cathode side there will be an electrochemical reaction between the H⁺ and the electrons, producing hydrogen gas (H₂).

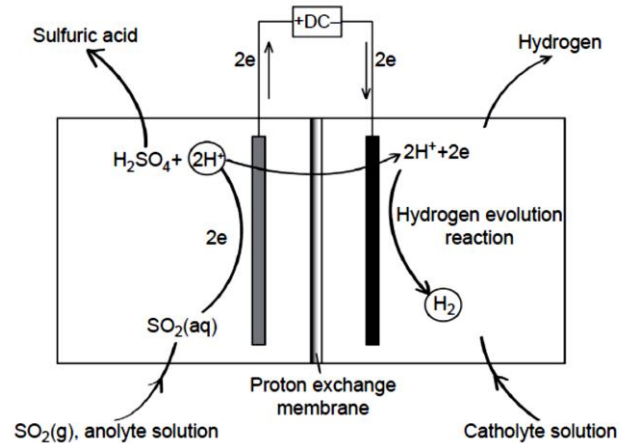


Figure 1. SO₂ desulfurization system using electrochemical cells (Yang and Yang, 2015)

Thus, the result of the SO₂ desulfurization process is a solution of sulfuric acid and hydrogen gas. Hydrogen can be used as fuel in fuel cells, and sulfuric acid solutions can be used as an intermediate chemical. The reaction at the anode and cathode can be written as the Reaction Equations (6) and (7). The comparison of desulfurization using conventional methods and electrochemical methods can be seen in the Table 1 below.

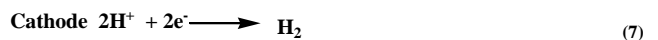
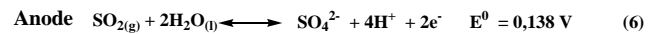


Table 2. Comparison of the desulfurization process using traditional methods and electrochemical methods

Conventional method	Electrochemical method
Advantages:	
1 Wet gas desulfurization and Dry Gas Desulfurization	Produces little or no secondary pollutants
2 SO ₂ removal percentage up to 98%.	Processing results are reusable
3 The product may be usable	Only takes a little energy because it produces usable hydrogen and sulfuric acid, and has a fairly high economic value
4 Adsorbents are inexpensive and easy to obtain	Operating conditions at low temperature and pressure, as well as low maintenance costs.
Disadvantages:	
1 Expensive investment costs, especially operating and maintenance costs	Can be installed without having to replace existing tools
2 Formation of fouling in absorber towers and downstream equipment	Can be designed for automation process
3 Expensive costs for residue disposal	Simple tool design

3. Conclusion and Suggestion

3.1 Conclusion

- Coal contains inorganic sulfur and organic sulfur. Inorganic sulfur is predominantly pyrite (FeS₂), sulfate, free sulfur, and sulfides. Inorganic sulfur can be separated physically by means of flotation, with the help of microwaves, ultrasonic vibrations, and magnets, using the pre-combustion desulfurization technique. Inorganic sulfur can also be removed using chemical methods and the help of microorganisms, however, these two processes are not economical on industrial scale.
- Coal organic sulfur cannot be removed using the method used in the inorganic sulfur removal process since organic sulfur is bound to coal macromolecules in the form of thiophene molecular structures, aromatic compounds, long-chain aliphatic, and hetero-cyclic compounds, making it difficult to separate from coal.
- The method currently used in the coal industry is post-combustion desulfurization using conventional methods and modern methods. The conventional methods are WFGD and DFGD; the most widely used is WFGD.
- The weaknesses of the conventional process can be overcome using a modern method, namely the electrochemical method. This method does not require chemicals, does not require large amounts of energy, and does not produce waste.
- The advantage of the electrochemical FGD process is that it can produce hydrogen gas and sulfuric acid as intermediate chemical.

3.2 Suggestion

For further research, it is recommended to develop electrochemical method due to its efficiency, effectiveness, and is environmentally friendly.

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