PERSIAPAN DAN KARAKTERISASI PALLADIUM BERBASIS ELEKTROLIS UNTUK SEL BAHAN BAKAR

PREPARATION AND CHARACTERIZATION OF ELECTROLYSIS BASED PALLADIUM FOR FUEL CELLS

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Abstrak

Penelitian ini memaparkan hasil penelitian mengenai preparasi dan karakterisasi elektrolit berbasis palladium (Pd) untuk aplikasi dalam sel bahan bakar. Dengan tujuan memanfaatkan sumber energi yang efisien dan ramah lingkungan, sel bahan bakar menawarkan solusi inovatif dengan mengkonversi energi kimia menjadi energi listrik tanpa proses pembakaran. Penelitian ini bertujuan untuk mengembangkan elektrolit yang memiliki kinerja tinggi, dengan fokus pada analisis sifat fisik dan kimia elektrolit yang dihasilkan. Variabel yang digunakan dalam penelitian ini meliputi komposisi elektrolit berbasis palladium (Pd), kondisi operasional (suhu, tekanan), dan luas permukaan elektroda. Metode yang digunakan meliputi sintesis elektrolit berbasis palladium (Pd) dan karakterisasi menggunakan teknik analisis lanjutan seperti X-ray diffraction (XRD), *Inductively Coupled Plasma Optical Emission Spectrometry* (ICP-OES), dan *Cyclic Voltammetry* (CV). Hasil penelitian menunjukkan bahwa elektrolit berbasis palladium (Pd) memiliki konduktivitas ionik yang lebih baik dan stabilitas yang lebih tinggi dibandingkan dengan elektrolit konvensional. Selain itu, ditemukan bahwa variasi kondisi operasional berpengaruh signifikan terhadap kinerja sel bahan bakar, dimana peningkatan luas permukaan elektroda secara efektif dapat meningkatkan produksi hidrogen. Sebagai kesimpulan, elektrolit berbasis Pd berpotensi meningkatkan efisiensi dan kinerja sel bahan bakar, mendukung pengembangan teknologi energi bersih yang berkelanjutan.

Kata kunci: Elektrolit berbasis Palladium (Pd), Sel bahan bakar, Stabilitas termal, X-ray diffraction (XRD), Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES), Efisiensi sel bahan bakar.

Abstract

This research explains the results of research on the preparation and characterization of palladium-based electrolytes (Pd) for applications in fuel cells. With the aim of utilizing efficient and environmentally friendly energy sources, fuel cells offer innovative solutions by converting chemical energy into electrical energy without the combustion process. This research aims to develop high-performance electrolytes, focusing on the analysis of the physical and chemical properties of the resulting electrolytes. Variables used in this study include the composition of palladium-based electrolytes (Pd), operational conditions (temperature, pressure), and electrode surface area. The methods used include the synthesis of palladium-based electrolytes (Pd) and characterization using advanced analysis techniques such as X-ray diffraction (XRD), Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES), and Cyclic Voltammetry (CV). Research results show that palladium-based electrolytes (Pd) have better ionic conductivity and higher stability compared to conventional electrolytes. In addition, it was found that variations in operational conditions have a significant effect on fuel cell performance, where increasing the electrode surface area can effectively increase hydrogen production. In conclusion, Pd-based electrolytes have the potential to increase the efficiency and performance of fuel cells, supporting the development of sustainable clean energy technology.

Keywords: *Palladium-based electrolyte (Pd), Fuel cell, Thermal stability, X-ray diffraction (XRD), Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES), Fuel cell efficiency.*

1. Introduction

This research on "Palladium-Based Electrocatalyst Preparation and Characterization for Fuel Cells" answers important needs in the field of renewable energy, especially improving the efficiency and performance of fuel cells. The motivation for choosing this research problem comes from the increasing demand for sustainable energy solutions and the limitations of expensive traditional platinum-based catalysts and their limited availability. Palladium (Pd) offers a promising alternative because of its unique catalytic properties and its potential to

reduce costs, making it an interesting subject to explore. This is very relevant because the hydrogen economy relies heavily on efficient catalysts to drive reactions such as hydrogen evolution and oxygen reduction, which are important for fuel cell operations (Sarkar, 2018).

The background of this research is rooted in the need to develop an electrocatalyst that not only increases fuel cell performance but also reduces dependence on precious metals such as platinum. Previous studies highlighted various Pd-based catalysts, which showed their effectiveness in various types of fuel cells, such as Proton Exchange Membrane Fuel Cells (PEMFC) and Direct Alcohol Fuel Cells (DAFC). For example, recent advances show that Pd alloys can significantly increase catalytic activity by reducing problems such as CO poisoning, which is a common challenge in fuel cell operations. Previous researchers also emphasized how structural arrangements and alloy strategies can optimize Pd-based materials for better electrocatalytic activity (Li, 2021).

Furthermore, the novelty of this research lies in its potential to provide new insights into Pd-based catalysis mechanisms and identify optimal compositions that maximize performance while minimizing costs. The significance of overcoming this problem is underlined by its implications for sustainability and energy efficiency. As global energy demand increases, the development of effective electrocatalysts is not only beneficial but also important to advance fuel cell technology.

In reviewing the literature, several studies have contributed to the understanding of Pd-based catalysts over the past decade. For example, research has focused on advanced nanostructures and alloying strategies that improve oxygen reduction reactions (ORR) in fuel cells. In particular, incorporating various metals into Pd-based catalysts to improve their performance in formic acid oxidation has shown a significant increase in catalytic activity (Kankla, 2023). In addition, research has explored new fabrication techniques for polymeric electrolytic membranes that incorporate Pd-based catalysts, which further highlight their flexibility and innovation potential.

The challenges faced are not only interesting but also ripe to be explored due to the extensive availability of data from previous research efforts. This platform allows for more focused investigation into specific aspects of Pd-based catalysts, such as their structural properties and electrochemical behavior under various operational conditions. By connecting this finding with a broader trend in fuel cell research, this study aims to make a significant contribution to existing knowledge (Meng, 2015).

2. Experimental Methods

A. Materials

Several important material components are needed to achieve optimal results. Carbon Nanotubes (CNT) functions as a supporting material that provides a high surface area, increasing the catalytic properties of metal nanoparticles. Palladium(II) Chloride ($PdCl_2$) is used as a source of palladium metal, which is the main active component in catalysts. Sodium Hydroxide (NaOH) is added to adjust the pH of the solution to 11, creating an alkaline environment that supports the reduction process. Sodium Borohydride ($NaBH_4$) is used as an effective reducing agent. Deionized Water (DI Water) is required for initial ultrasonic treatment and additional washing measures, ensuring good CNT dispersibility and removing contaminants. Methanol is used to wash the final product to remove the remaining reagents. Hydrochloric Acid (HCl) and Nitric Acid (HNO_3) are needed to dissolve the metal component from the PdCu/CNT sample, allowing accurate quantification of the metal content in the catalyst.

B. Synthesis of Pd//CNT

First, use an ultrasonic shock of 0,04 g CNT in 8 mL of DI water for 5-10 minutes, and add 0,03 M solution of $PdCl_2$ solution and use ultrasonic for 5-10 minutes. Next, add 3 M NaOH to the mixed solution to adjust the pH to 11. Then, slowly pour $NaBH_4$ at a flow rate of 1 mL/minute for a reaction time of 20 minutes and use ultrasound for 5-10 minutes. Stir again for 1-2 hours. Separate into 4 centrifuge tubes, each containing 40 mL. Centrifuge at 10.000 rpm for 5 minutes. Wash the sample with DI water 3 times and methanol 2 times. Collect samples in Petri dishes. Dry on a hot plate at a temperature of 85°C. After drying, store the dried sample in a small bottle and label it.

C. Pd/CNT Analysis with XRD

The process begins by preparing a Pd/CNT sample in the form of a fine powder, which is then placed in a special container for analysis. XRD instruments are prepared by selecting the appropriate X-ray source and determining measurement parameters such as angle range (2θ) and scanning speed. After the instrument is turned on, X-rays are directed to the sample, and the detector records the intensity of X-rays reflected at various angles, producing a diffraction pattern that shows the relationship between the intensity and the angle of 2θ. This data is then analyzed using software to identify crystal phase, particle size, and lattice parameters, providing important insight into the catalytic properties of Pd/CNT and its application in chemical reactions.

D. Analysis of Pd/CNT with ICP-OES

Induced Plasma Optical Emission Spectrometry (ICP-OES) is an advanced analysis technique to detect and measure metal elements in various samples. The sample preparation process begins by weighing 5 mg Pd/CNT (palladium-copper composite/carbon nanotube) and adding a solution of hydrochloric acid (HCl) and nitric acid (HNO_3) in a ratio of 3:1, with a total of 10 mL. This mixture is then subjected to ultrasonic sound for 30 minutes to 1 hour to increase dissolution. After that, the mixture is heated on a hot plate at a temperature of 85°C for 1 to 3 days to ensure all metals dissolve. After the heating period, the sample is filtered to separate the solid residue from the liquid phase, and the filtered solid is mixed with 50 mL of deionized water (DI) for further analysis preparation.

Once prepared, the sample is ready for analysis using ICP-OES. In the ICP-OES system, the liquid sample is nebulized into an aerosol, which is then transported to a plasma torch where high temperature (about 10.000 K) causes the atoms in the sample to be excited. When these atoms return to their ground state, they emit light at a characteristic wavelength that is unique to each element. The light emitted is measured by a detector, which measures its intensity; this intensity correlates with the concentration of each element in the original sample. Thus, ICP-OES allows fast and simultaneous analysis of several elements with high sensitivity and accuracy.

E. Electrochemical Measurement

To perform a Cyclic Voltammetry (CV) analysis on a Pd/CNT sample, first weigh 5 mg Pd/CNT. Next, add 5 mL of ethanol to the container containing Pd/CNT as a solvent to disperse the catalyst. Then, add 50 L of 5% Nafion solution into the mixture, where Nafion acts as a binder to increase the stability of the catalyst film. Finally, use an ultrasonic machine for 30 minutes to ensure that all components are mixed well and produce a homogeneous dispersion, thus preparing the solution for the next electrochemical CV analysis.

Drop the prepared solution onto the catalyst paper, use a micropipette to apply 10 drops while heating the paper on the hotplate at a temperature of 85°C. After the drip process is complete, let the catalyst paper dry, then weigh the paper again with the applied solution. Next, place the catalyst paper on the electrode that has been prepared and run the computer to monitor the electrochemical result graph. Wait for about 2 hours to get optimal results.

3. Results and Discussion

A. X-Ray Diffraction (XRD)

X-Ray Diffraction (XRD) is an analysis technique used to determine the crystal structure of a material by utilizing the interaction of X-rays with atoms in the sample. In the context of the analysis of bimetallic nanoparticles such as Palladium (Pd) supported on carbon nanotubes

(CNT), XRD plays an important role. This technique not only helps identify the phase in the sample but also provides in-depth information about particle size, composition, and interaction between nanoparticles and substrates (Göksu, 2020).

One of the main reasons for using XRD in Pd/CNT analysis is its ability to identify the crystal structure of nanoparticles. By analyzing the resulting diffraction pattern, researchers can determine which phase exists, whether it is Pd or mixed bimetallic phase. Identifying these phases is very important because the physical and chemical properties of materials often depend on their crystal structure. For example, certain phases may be more active in catalytically than others, so having a proper understanding of structure can help in designing materials with optimal performance (Sánchez-Resa, 2021).

In addition to phase identification, XRD allows particle size measurement in Pd nanoparticles. Using the Scherrer equation, particle size can be calculated from the width of the peak of diffraction. This particle size is very important because, in many applications, including catalysis and electronics, the reactivity and conductivity properties of materials can be affected by particle size. Therefore, understanding the particle size and distribution can help optimize the synthesis conditions to achieve the desired material properties (Göksu, 2020).

$$FWHM = \frac{K\lambda}{L\ Cos\theta} \quad \text{..... (3.1)}$$

$$L = \frac{K\ \lambda}{(FWHM) \times\ Cos\theta} \quad \text{..... (3.2)}$$

Where :

FWHM = Full width at half maximum of the diffraction peak in radians. This represents the broadening of the peak due to the finite size of the crystallites.

K = Shape factor, a dimensionless constant that depends on the crystallite shape and the definition of FWHM. The image states that K ranges from 0,89 to 1,39. A value of 0,9 is often used as an approximation.

λ = Wavelength of the X-rays used in the diffraction experiment (given as 1,5418 Å in the image).

L = Average crystallite size. This is the quantity we want to determine using the Scherrer equation.

θ = Bragg angle, which is half of the angle between the incident and diffracted X-ray beams.

XRD also plays an important role in analyzing the interaction between Pd and CNT nanoparticles as substrates. When CNT is used as a support for bimetallic nanoparticles, this

interaction can affect the stability and catalytic properties of the system. By studying changes in diffraction patterns before and after adding CNT, researchers can gain insight into how CNT affects the structure and distribution of nanoparticles. This is very important for applications where thermal and mechanical stability of nanoparticles is required (Fuchun Zhu, 2013).

During the synthesis or application process, Pd nanoparticles can undergo phase changes due to environmental conditions or special treatment. XRD allows real-time monitoring of these changes, allowing researchers to understand system dynamics and make necessary adjustments to achieve optimal results. The ability to monitor phase changes provides an additional advantage in developing new materials with targeted properties (Arulmani, 2019).

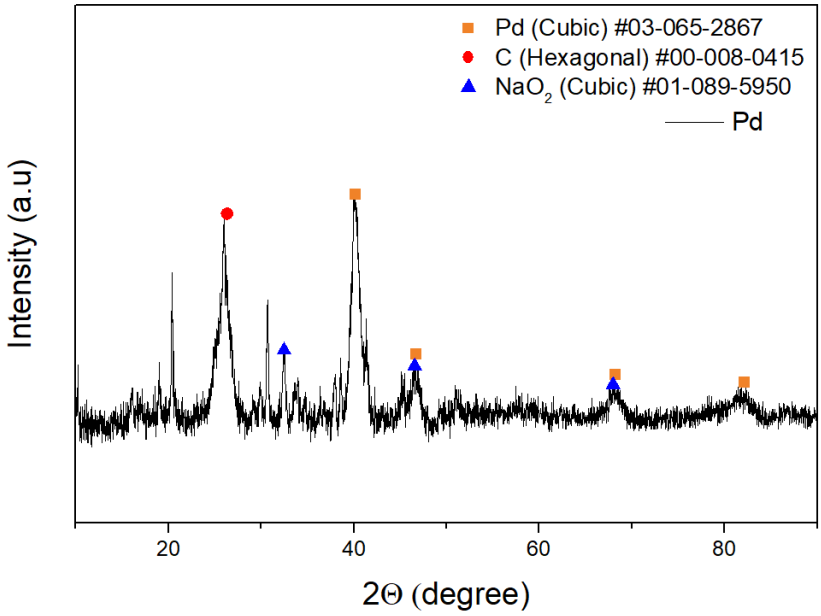

Figure 1. Pd graph using XRD

From the picture above, it can be seen that to read the graph, you need to pay attention to the highest peak in the form of an orange box, which shows palladium (Pd) with a theta angle of 40°. The red circle symbol represents carbon (C) with a theta angle of 26,02°. Meanwhile, the blue triangle symbol shows NaO_2 with a theta angle of 32.5°. On the other hand, there are three lower graphs on the right, which also show palladium (Pd) in the NaO_2 mixture. This can be interpreted as a mixture that has not been completely separated or considered as a remaining mixture that still exists in the palladium sample (Pd). The results obtained are as follows:

a. Palladium (Pd) Cubic Structure

The peaks indicated by orange squares correspond to Palladium (Pd) in a cubic crystal structure. The reference code #03-065-2867 is a unique identifier for this phase in crystallographic databases. The positions and intensities of these peaks are characteristic of the atomic arrangement specific to Pd, confirming its cubic phase.

b. Carbon (C) Hexagonal Structure

The red circles denote peaks associated with Carbon (C) in a hexagonal crystal structure. The reference code #00-008-0415 identifies this phase in the crystallographic database. The presence of carbon suggests either impurities or a carbon-containing phase within the PdCu sample, which may arise from the synthesis or processing conditions.

c. Sodium Peroxide (NaO_2) Cubic Structure

The blue triangles indicate peaks attributed to Sodium Peroxide (NaO_2) in a cubic crystal structure, with reference code #01-089-5950 confirming this phase. The detection of NaO_2 may imply contamination or a reaction product formed during sample preparation or analysis.

From the results of the XRD graph above, it can be seen that each analyzed elemental particle exhibits different characteristics. Palladium (Pd) with a cubic structure has a theta scale of 40,0007 degrees and a crystal size of 16,30848 nm. Meanwhile, Sodium Peroxide (NaO_2), also with a cubic structure, shows a theta scale of 32,5054 degrees and a larger crystal size of 24,56001 nm. On the other hand, Carbon (C), which has a hexagonal structure, is recorded with a theta scale of 26,0203 degrees and the smallest crystal size of 9,519821 nm. From this data, it can be concluded that the crystal sizes of Pd and NaO_2 are larger compared to the crystal size of C, indicating differences in the structure and physical properties of each element.

B. Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES)

Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) is a highly effective analytical technique for detecting and measuring the concentrations of chemical elements, particularly metals, in various types of samples. This technique utilizes plasma generated from argon gas heated to very high temperatures, reaching around 10.000 °K. This process causes the atoms in the sample to become excited, resulting in the emission of light at specific wavelengths. The emitted light is then measured to determine the concentrations of these elements. With this sophisticated working principle, ICP-OES has become one of the most widely used analytical methods in modern chemistry laboratories.

One of the main advantages of ICP-OES is its ability to perform simultaneous multi-element analysis. This allows users to analyze multiple elements in a single test, thereby enhancing efficiency and reducing analysis time. Additionally, this technique has high sensitivity, with the capability to detect element concentrations ranging from 0,0002 to 1000 ppm. The speed of analysis is also a crucial factor, ICP-OES can analyze up to 40 elements per minute, making it an ideal choice for laboratories that require high throughput (Douvris, 2023).

One of the primary reasons for using ICP-OES in the analysis of Pd/CNT is its ability to provide highly accurate data regarding metal composition. In the context of catalysts, the metal ratio is vital as it can influence the catalytic properties and effectiveness of the material. For instance, the ratio between palladium and copper can determine the catalyst's performance in specific reactions, such as glycerol oxidation. By utilizing ICP-OES, researchers can ensure that the metal concentrations align with experimental designs and research objectives (BES Group, 2024).

In addition to determining composition, ICP-OES also serves as a tool for analyzing catalyst quality. During the synthesis of Pd/CNT, there is often a possibility of variations in metal ratios due to factors such as reaction conditions or starting materials used. By performing analysis using ICP-OES, researchers can evaluate whether the metal proportions in the catalyst meet expectations. This is important to ensure that the produced catalyst has optimal activity and stability.

The use of ICP-OES also plays a role in validating the synthesis methods applied to produce Pd/CNT. By comparing ICP-OES analysis results with theoretical or target values, researchers can assess the efficiency of the synthesis process. If there are significant discrepancies between analysis results and theoretical values, this could indicate material loss or contamination during production. Thus, ICP-OES not only aids in material characterization but also provides valuable insights for developing better synthesis methods.

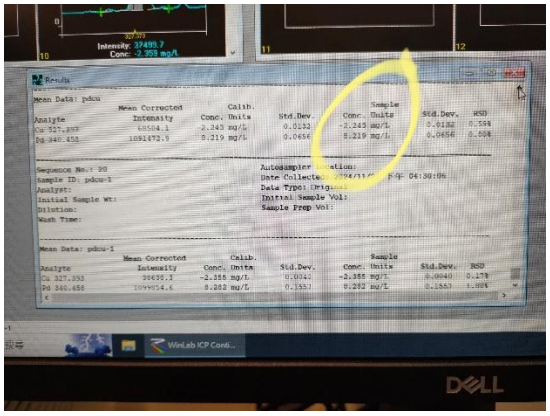

Figure 2. Results of analysis with ICP-OES

The results of the analysis of Pd/CNT samples using the ICP-OES method showed that the concentration of palladium (Pd) in the sample was 8,219 mg/L. From this analysis, the concentration of palladium in the solution was measured at 8,219 mg/L. Based on the calculation, the weight percentage (wt%) of palladium (Pd) in the sample was around 0,82%. This result is in accordance with the initial calculation before the sample preparation treatment for ICP-OES analysis, where the weight percentage for palladium (Pd) was 0,816%.

4. Conclusions

Based on the results of research and discussion, it can be concluded that Pd-based electrolytes show great potential for applications in fuel cells. Its superior properties, such as better ionic conductivity and higher stability compared to conventional electrolytes, make it a promising candidate for improving fuel cell performance. In addition, this study highlights the importance of optimizing operating conditions, such as increasing the surface area of the electrode, to further increase hydrogen production.

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