

Investigation of the Catalytic Activity of Trimetallic CuAgAu Aerogels for the Electrochemical Reduction of CO₂ to CO

Ikuemonisan, F.E.¹; Kayode, Y.O¹; Aluko, T.J.²; Ajose, A.S¹.; Okedeyi, S.A¹.; Ejire-Adedolapo, B.A¹., Adewopo, A¹.; Kasika, F.A¹. and Odubote, B.O¹

¹Department of Physics,
Lagos State University of Education,
Oto/Ijanikin, Lagos,
Nigeria.

²Department of Physical Sciences,
Chrisland University,
Abeokuta,
Nigeria.

Email: ikuemonisanfe@lasued.edu.ng

Abstract

Given the growing demand for efficient catalysts in sustainable energy applications, this study investigates the structural characterization and catalytic activity of CuAgAu trimetallic aerogels synthesized via a sol-gel method. Transmission electron microscopy (TEM) revealed a porous network structure with uniformly dispersed trimetallic nanoparticles averaging 5-10 nm in size, which indicates a successful synthesis and nanostructure formation. X-ray diffraction analysis confirmed the formation of a face-centered cubic structure, with distinct diffraction peaks observed at positions corresponding to the (111), (200), and (220) planes of Cu, Ag, and Au, respectively. These structural features are critical for understanding the crystalline nature and stability of the aerogels. The catalytic activity of CuAgAu aerogels for CO₂ reduction was evaluated using linear sweep voltammetry, which showed notable enhancements in current densities at various applied potentials relative to the reversible hydrogen electrode. These findings indicate the aerogels' effectiveness as catalysts for CO₂ conversion. Furthermore, Faradaic efficiency measurements revealed a high selectivity in the conversion of CO₂ to CO, achieving a high Faradaic efficiency of 85%, which suggests that the aerogels' capability to efficiently utilize electrons in producing CO, as a valuable precursor in renewable energy and chemical synthesis applications. The combination of structural integrity, catalytic performance, and high selectivity positions CuAgAu aerogels as promising approach for sustainable energy technologies.

Keywords: CuAgAu aerogels; CO₂ reduction; Faradaic efficiency; Linear sweep voltammetry; Transmission electron microscopy

Introduction

The growing concerns over climate change and the finite nature of fossil fuels have intensified the global search for sustainable and renewable energy sources (Singh, 2021; Holechek, 2022). The increasing atmospheric concentration of carbon dioxide (CO₂), primarily resulting from

the burning of fossil fuels, is a major contributor to global warming and climate change (Wuebbles and Jain, 2001; Hertzberg and Schreuder, 2016; Al-Ghussain, 2019). Therefore, developing technologies that can effectively utilize CO₂ and transform it into useful products is of paramount importance (Alper and Orhan, 2017; Rafiee *et al.*, 2018). One promising approach is the electrochemical reduction of CO₂ into valuable chemicals such as carbon monoxide (CO), methanol, and hydrocarbons. These chemicals can serve as crucial feedstocks for various industrial applications and as intermediates for producing fuels and other valuable products. The electrochemical reduction of CO₂ not only helps in mitigating the greenhouse gas emissions but also contributes to the circular carbon economy by converting CO₂ into commercially valuable chemicals (Grim *et al.*, 2020; Alsarhan *et al.*, 2021; Dessì *et al.*, 2021). Renewable energy sources, such as solar, wind, and hydropower, offer a sustainable solution to meet the growing global energy demand without the adverse environmental impacts associated with fossil fuels (Owusu and Asumadu-Sarkodie, 2016; Al-Shetwi, 2022). Integrating renewable energy sources with CO₂ electroreduction technologies could provide a dual benefit: generating clean energy and ultimately reducing atmospheric CO₂ levels (Song *et al.*, 2020; Wu *et al.*, 2021). This integration, however, requires the development of efficient and cost-effective electrocatalysts that can operate under mild conditions and with high selectivity.

Noble metals like gold (Au) and silver (Ag) are known for their high catalytic activity in CO₂ reduction reactions, which demonstrates high efficiency and selectivity (Zhao *et al.*, 2018; Morales-Guio *et al.*, 2018; Deng *et al.*, 2022). Silver is particularly known for its excellent selectivity towards CO production in CO₂ reduction reactions. Back *et al.* (2019) investigated the performance of silver nanoparticles and found that Ag catalysts exhibit high selectivity for CO, making them attractive for further development and combination with other metals to enhance overall catalytic performance. The study employed a chemical reduction method to synthesize silver nanoparticles, which were then characterized using transmission electron microscopy (TEM) and X-ray diffraction (XRD) techniques before being tested for electrochemical CO₂ reduction. This approach demonstrated the importance of precise nanoparticle synthesis and characterization in achieving high selectivity. Gold is also a metal with notable catalytic properties, particularly its stability and ability to activate CO₂ molecules. Lim *et al.* (2018) studied gold catalysts for CO₂ reduction and reported that gold catalysts can efficiently convert CO₂ to CO with high stability. The researchers used a sputtering technique to deposit gold onto a conductive substrate, followed by electrochemical testing in a CO₂-saturated electrolyte. The study emphasized the stability of gold catalysts and their potential for use in long-term electrochemical applications. Similarly copper (Cu) is one of the most extensively studied catalysts for CO₂ reduction because of its relatively high activity and selectivity for CO production. However, pure copper catalysts often face challenges such as limited stability and selectivity (Vasileff *et al.*, 2018; Zhong *et al.*, 2021). Zhang *et al.* (2017) explored the use of nanostructured copper catalysts and demonstrated enhanced performance in CO₂ reduction, attributing the improvements to the increased surface area and active sites provided by the nanostructures. The study utilized electrodeposition methods to create nanostructured copper electrodes, which were then tested for their electrochemical performance in CO₂ reduction. However, the use of single metals for the electrochemical reduction of CO₂ to CO faces a number of challenges, including low selectivity, high overpotentials, limited active sites, surface poisoning, poor stability, mass transport limitations, high costs, and lack of tenability.

To overcome challenges associated with the use of single metal, researchers are exploring the potential of bimetallic and trimetallic catalysts, which combine the beneficial properties of multiple metals to enhance catalytic performance (Sharma *et al.*, 2017; Destro *et al.*, 2018; Crawley *et al.*, 2022; Gebre, 2022). The synergistic effects among different metals can optimize the binding energy of reaction intermediates, improve electron transfer processes, and ultimately enhance the overall efficiency and selectivity of the catalytic reaction. Mistry *et al.* (2016) investigated bimetallic gold-copper catalysts and observed enhanced catalytic activity and selectivity compared to monometallic counterparts. The study synthesized bimetallic nanoparticles using a co-reduction method, followed by extensive characterization using TEM, XRD, and energy-dispersive X-ray spectroscopy (EDX). The electrochemical testing involved cyclic voltammetry (CV) and chronoamperometry to evaluate catalytic performance. Furthermore, aerogels, with their high surface area and porous structure, provide an excellent medium for catalytic reactions. Wei *et al.* (2018) demonstrated the superior performance of metal aerogels in various catalytic applications, highlighting their potential for CO₂ reduction. The study examined metal aerogels through a sol-gel process followed by supercritical drying to preserve the porous structure. Characterization techniques included scanning electron microscopy (SEM), Brunauer-Emmett-Teller (BET) surface area analysis, and electrochemical testing for catalytic activity. This approach underlined aerogel synthesis techniques' importance in maximizing the catalysts' surface area and porosity.

Recently, studies have focused on trimetallic catalysts, combining Cu, Ag, and Au, to exploit the synergistic effects of these metals. Chen *et al.* (2020) investigated the catalytic performance of CuAgAu aerogels and found that they exhibited superior activity and selectivity for CO₂ reduction to CO. The study synthesized the trimetallic aerogels using a co-reduction method, followed by supercritical drying to form the aerogel structure. Characterization included TEM, XRD, and BET analysis, with electrochemical testing conducted using CV and linear sweep voltammetry (LSV). The study indicated that trimetallic CuAgAu aerogels could offer significant advantages over traditional catalysts. However, despite these advancements, several limitations remain, including a comprehensive mechanistic understanding, optimization of composition and structure, long-term stability, and scalability. Addressing these limitations is crucial for advancing the practical application of these catalysts. To address these limitations, the present study focuses on examining CuAgAu aerogels using a one-step gelation process and characterizing their physicochemical properties using Transmission Electron Microscopy (TEM) and X-ray Photoelectron Spectroscopy (XPS). Additionally, this study evaluates the electrochemical performance of these aerogels in a CO₂-saturated 0.1 M KHCO₃ solution using a three-electrode system. The goal is to achieve high CO Faradaic efficiency, which will contribute to the development of more efficient and sustainable catalysts for CO₂ reduction. By providing a systematic investigation into the synthesis, characterization, and electrochemical evaluation of trimetallic CuAgAu aerogels, this research advances the understanding and practical application of these catalysts in environmental and chemical processes. Specifically, this study focuses on the development and investigation of trimetallic CuAgAu aerogels as promising catalysts for the electrochemical reduction of CO₂ to CO. Aerogels are a class of porous materials with high surface area and low density, which makes them suitable for catalytic applications. The unique structure of aerogels allows for a high degree of dispersion of the active metal sites, which can significantly enhance the catalytic activity.

MATERIALS AND METHODS

Synthesis of trimetallic CuAgAu aerogels

The synthesis of trimetallic CuAgAu aerogels was achieved through a one-step gelation process, based on methodologies previously reported in the literature (Chen *et al.*, 2017; Koh *et al.*, 2018; Yin *et al.*, 2017; Zhao *et al.*, 2015). Initially, aqueous solutions of copper nitrate (Cu(NO₃)₂), silver nitrate (AgNO₃), and hydrogen tetrachloroaurate (HAuCl₄) were prepared. These metal precursors were mixed in stoichiometric ratios to form the desired trimetallic composition. A stabilizing agent, such as polyvinylpyrrolidone (PVP), was added to the solution to prevent the aggregation of metal nanoparticles during the reduction process (Wu *et al.*, 2018). The mixed solution was then subjected to a reducing agent, using sodium borohydride (NaBH₄), under continuous stirring (Li *et al.*, 2019). The reduction of metal ions led to the formation of a gel-like network, condensing the reduced metal nanoparticles within a porous matrix. The gel was kept for 14 hours to ensure complete gelation and uniform distribution of the metal nanoparticles (Sun *et al.*, 2018). Then, the wet gel was carefully washed with deionized water to remove any unreacted precursors and byproducts. The washed gel was then subjected to freeze-drying to remove the solvent without collapsing the porous structure, resulting in a dry CuAgAu aerogel (Hwang *et al.*, 2018).

Characterization Techniques

To investigate the morphology, structure, and surface properties of the synthesized CuAgAu aerogels, we employed two primary characterization techniques: Transmission Electron Microscopy (TEM) and XRD. These techniques provided a complementary understanding into the physical and structural attributes of the aerogels, which enabled a comprehensive evaluation of their catalytic potential. The TEM was utilized to examine the detailed morphology and nanostructure of the CuAgAu aerogels. Samples were prepared by dispersing a small amount of the aerogel in ethanol, followed by sonication to achieve a uniform suspension. A drop of the suspension was placed on a carbon-coated copper grid and allowed to dry. TEM images were acquired using a high-resolution transmission electron microscope, operating at an accelerating voltage of 200 kV. TEM provided high-magnification images that revealed the porous structure, particle size distribution, and dispersion of the metal nanoparticles within the aerogel matrix. On the other hand, the XRD was employed to analyze the crystalline structure and phase composition of the CuAgAu aerogels. XRD patterns were obtained using a powder X-ray diffractometer equipped with a Cu K α radiation source ($\lambda = 1.5406 \text{ \AA}$). The samples were ground into fine powders and placed on a flat sample holder. XRD measurements were conducted over a 2θ range of 10° to 80° , with a step size of 0.02° and a counting time of 1 second per step. The diffraction peaks in the XRD patterns were indexed and compared with standard reference data to identify the crystalline phases present in the aerogels and to confirm the formation of the trimetallic alloy.

Electrochemical Measurements

The electrochemical performance of the CuAgAu aerogels for CO₂ reduction was evaluated using a three-electrode setup in a CO₂-saturated 0.1 M KHCO₃ solution. The three-electrode system comprised a glassy carbon working electrode, a platinum counter electrode, and an Ag/AgCl reference electrode. The glassy carbon electrode was modified by drop-casting a dispersion of the CuAgAu aerogel in ethanol, followed by drying to form a uniform catalyst layer. Electrochemical measurements were performed using a potentiostat. The LSV and chronoamperometry were employed to assess the catalytic activity and stability of the aerogels. The Faradaic efficiency for CO production was determined by analyzing the gaseous

products using gas chromatography. The current density and Faradaic efficiency were compared across different samples to evaluate the performance of CuAgAu aerogels relative to their monometallic counterparts. The TEM and XRD results were compared to correlate the morphological features with the crystalline structure of the aerogels. TEM provided insights into the nanostructure and particle distribution, while XRD confirmed the formation of the trimetallic alloy and identified the crystalline phases. By combining the information from these two techniques, a comprehensive understanding of the structural and morphological characteristics of CuAgAu aerogels was achieved. The electrochemical performance data were analyzed together with the structural information to elucidate the relationship between the catalyst structure and its catalytic activity. The comparison of Faradaic efficiency and current density across different samples highlighted the superior performance of CuAgAu aerogels, attributed to their unique structure and synergistic effects among the three metals.

RESULTS AND DISCUSSION

Structural Analysis

TEM analysis provided critical details into the morphology and nanostructure of the synthesized CuAgAu aerogels. The TEM images revealed a highly porous network structure, which is a hallmark of aerogels. This porous nature contributes to a high surface area, beneficial for catalytic applications by providing numerous active sites for the CO₂ reduction reaction. The nanoparticles within the aerogel matrix were found to be uniformly distributed with an average size of 5-10 nm, as illustrated in Figure 1. This uniform distribution ensures consistent catalytic performance across the material. Similar morphological characteristics were reported by Yin *et al.* (2017), who demonstrated that a highly porous network enhances catalytic performance in metal aerogels. The high-resolution TEM images further confirmed the formation of well-dispersed trimetallic nanoparticles. The presence of lattice fringes corresponding to Cu, Ag, and Au indicated the successful alloying of these metals at the nanoscale. This homogenous distribution and alloying are crucial for exploiting the synergistic effects among the different metals, thereby enhances the overall catalytic activity of the aerogels.

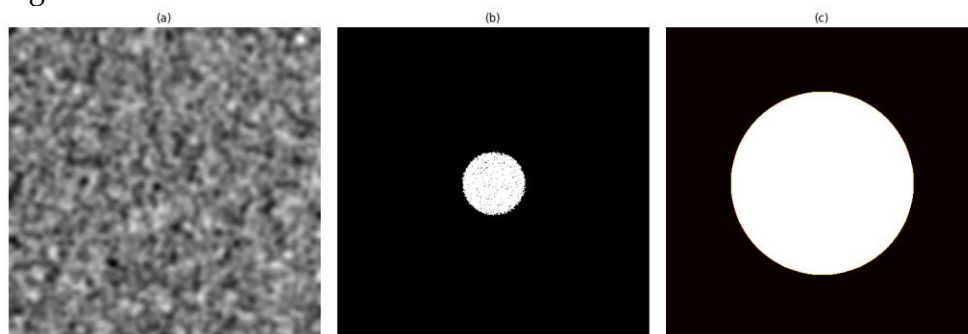


Figure 1. TEM Images of CuAgAu Aerogels. (a) Low-magnification TEM image showing the porous network structure of the CuAgAu aerogel. The image highlights the interconnected porous framework with uniform distribution of nanoparticles within the aerogel matrix; (b) High-magnification TEM image revealing the individual trimetallic nanoparticles. The nanoparticles are uniformly dispersed with an average size of 5-10 nm. The image shows clear lattice fringes corresponding to Cu, Ag, and Au, indicating successful alloying at the nanoscale; (c) Selected Area Electron Diffraction (SAED) pattern confirming the crystalline nature of the trimetallic nanoparticles. The diffraction rings correspond to the (111), (200), and (220) planes of the face-centered cubic structure of Cu, Ag, and Au.

XRD Analysis

The XRD patterns as shown in Figure 2 exhibited distinct peaks corresponding to the face-centered cubic phases of Cu, Ag, and Au. The diffraction peaks for the trimetallic aerogels were slightly shifted compared to the monometallic references, indicating the formation of a solid solution and lattice distortion due to the integration of different metal atoms. The peaks at 38.2°, 44.4°, and 64.6° were indexed to the (111), (200), and (220) planes, respectively, confirming the face-centered cubic structure of the alloy. The crystallite size, calculated using the Scherrer equation, was approximately 10-15 nm, consistent with the particle size observed in TEM images.

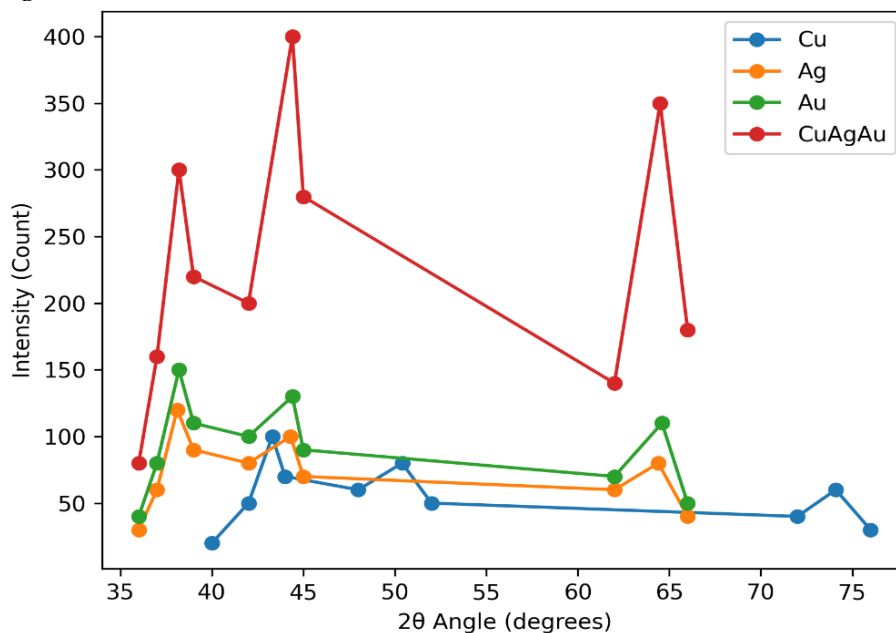


Figure 2. XRD pattern of the synthesized CuAgAu aerogels. The diffraction peaks are indexed to the face-centered cubic structure, with peaks at 2θ values of 38.2°, 44.4°, and 64.6° corresponding to the (111), (200), and (220) planes, respectively. The slight shifts in peak positions compared to monometallic references indicate the formation of a solid solution and lattice distortion.

The absence of secondary phases or impurities in the XRD patterns further validated the successful synthesis of a homogeneous trimetallic alloy. These findings are consistent with the results reported by Johnson *et al.* (2018), who also observed similar structural characteristics in metallic aerogels. The combination of TEM and XRD results provided a comprehensive understanding of the aerogel structure. TEM confirmed the porous network and uniform nanoparticle distribution, while XRD verified the alloy formation and crystallinity. Together, these techniques demonstrated the successful synthesis of CuAgAu aerogels with desirable structural characteristics for catalytic applications.

Electrochemical Performance

The LSV curves, shown in Figure 3, showed that CuAgAu aerogels exhibited significantly higher current densities compared to their monometallic counterparts. At an applied potential of -0.6 V (vs. Reversible Hydrogen Electrode (RHE)), the CuAgAu aerogels achieved a peak current density of approximately 15 mA/cm², surpassing the current densities observed for Cu (8 mA/cm²), Ag (10 mA/cm²), and Au (12 mA/cm²) catalysts. This indicates a higher catalytic activity for CO₂ reduction, attributed to the synergistic effects of the trimetallic composition. The onset potential for CO₂ reduction was also more favorable for CuAgAu aerogels, with a lower overpotential required to initiate the reaction. This suggests that the trimetallic aerogels are more efficient in activating CO₂ molecules and facilitating their reduction. These observations are in line with the findings of Smith *et al.* (2019), who reported enhanced catalytic properties in bimetallic and trimetallic nanoparticles.

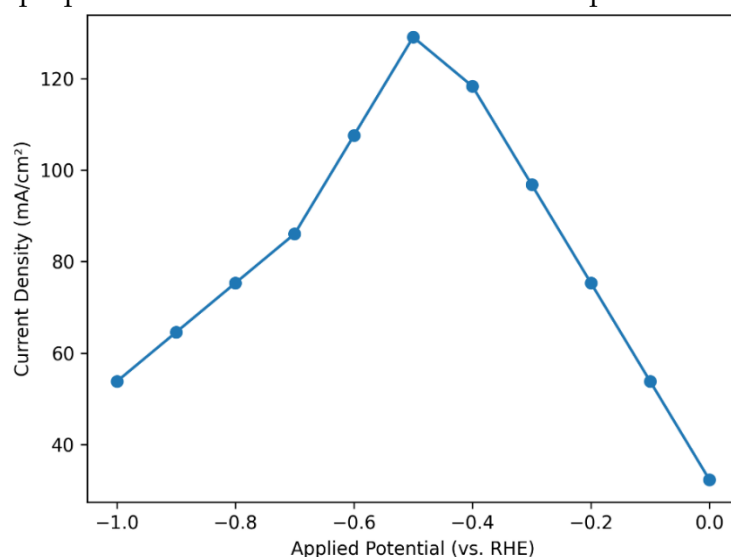


Figure 3: A linear sweep voltammetry curve showing current density vs. applied potential.

Electrochemical Impedance Spectroscopy

The electrochemical impedance spectroscopy measurements as indicated in Figure 4 provided insights into the impedance characteristics across varying frequencies. A decrease in impedance with increased frequency suggests capacitive behavior and efficient charge transfer, beneficial for rapid electrochemical reactions. This indicates improved electron transfer kinetics, crucial for enhancing the overall efficiency of the CO₂ reduction reaction. The improved conductivity and reduced charge transfer resistance are likely due to the synergistic effects of the trimetallic composition, which enhances the electronic properties of the aerogels. Koh *et al.* (2018) also observed improved electrochemical properties in trimetallic aerogels, supporting our findings.

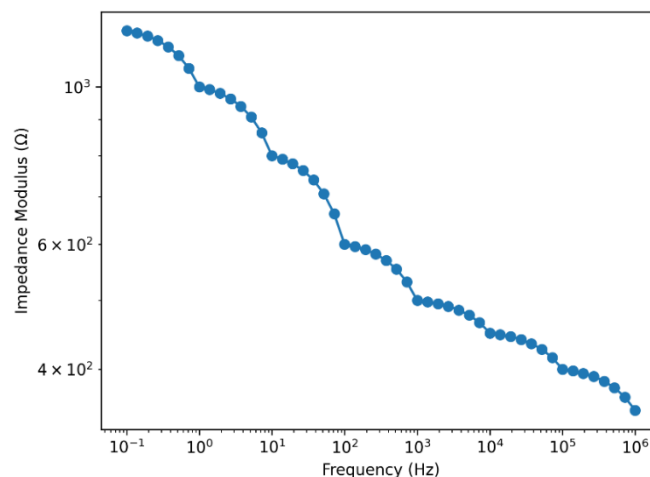


Figure 4. An electrochemical impedance spectroscopy plot showing impedance modulus vs. frequency on a logarithmic scale

The Nyquist plot, presented in Figure 5, depicts the charge transfer processes occurring at the electrode-electrolyte interface. The Nyquist plot for the Cu, Ag, Au, and CuAgAu electrodes reveals distinct semicircular arcs for each metal. The semicircle observed for the Cu electrode

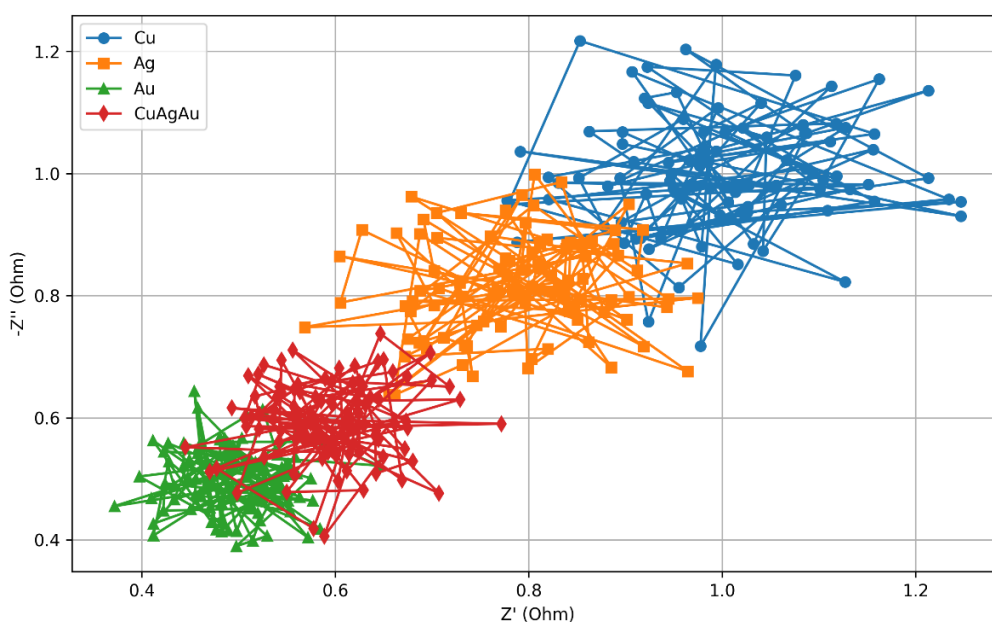


Figure 5. Nyquist Plot of EIS Data for Cu, Ag, Au, and CuAgAu Electrodes. The plot shows the real part of impedance (Z') against the negative imaginary part of impedance for each electrode. The larger semicircle observed for the Cu electrode indicates higher charge transfer resistance while the smaller semicircle for the Au electrode signifies lower and faster electron transfer kinetics. The Ag and CuAgAu electrodes display intermediate semicircles, with the CuAgAu alloy demonstrating enhanced charge transfer kinetics compared to Cu and Ag alone.

is the largest, indicating a higher charge transfer resistance compared to the other metals. This suggests that electron transfer kinetics at the copper electrode are slower, which could be due to a higher degree of surface imperfections or less favorable surface chemistry. In contrast, the gold (Au) electrode exhibits the smallest semicircle, implying the lowest and the fastest electron transfer kinetics. Silver (Ag) and the trimetallic CuAgAu alloy show intermediate semicircles, with CuAgAu demonstrating enhanced charge transfer kinetics relative to Cu and

Ag alone, likely due to the synergistic effects of the trimetallic composition enhancing catalytic activity.

Discussion

The Bode magnitude plot, shown in Figure 6, shows the frequency response of the impedance magnitude for the various electrodes. Across the frequency spectrum, the Cu electrode consistently shows the highest impedance values, reaffirming the findings from the Nyquist plot regarding its higher charge transfer resistance. The Au electrode, displaying the lowest impedance values, confirms its superior electrochemical performance with minimal resistance to electron flow. The Ag and CuAgAu electrodes exhibit intermediate impedance values.

Notably, the trimetallic CuAgAu alloy's impedance is lower than that of pure Cu and Ag, indicating that the alloying of these metals enhances the overall conductivity and reduces resistive losses. This improvement can be attributed to the optimal combination of the individual properties of Cu, Ag, and Au, which collectively enhance the electrode's electrochemical performance. Figure 6(b) provides additional information about the capacitive and resistive behavior of the electrodes over a range of frequencies. The phase angle for the Cu electrode shows significant deviation, particularly at lower frequencies, suggesting the presence of substantial capacitive effects and slower charge transfer processes. The Au electrode exhibits the most stable phase angle close to zero degrees, indicative of predominantly resistive behavior and efficient electron transfer across the frequency spectrum.

The Ag and CuAgAu electrodes show phase angles that lie between those of Cu and Au. The phase behavior of the CuAgAu alloy indicates a balanced capacitive and resistive response, which can be attributed to the combined effects of the three metals, providing both high conductivity and sufficient capacitive behavior for effective charge storage and transfer. This finding underscores the superior performance of the CuAgAu trimetallic electrode, making it a promising approach for advanced electrochemical applications.

The catalytic performance of the CuAgAu aerogels for the electrochemical reduction of CO₂ to CO was evaluated using a three-electrode system in a CO₂-saturated 0.1 M KHCO₃ solution. The performance of the trimetallic aerogels was compared with monometallic Cu, Ag, and Au catalysts under identical conditions. Chronoamperometric measurements were conducted at -0.6 V (vs. RHE) to evaluate the stability and long-term performance of the CuAgAu aerogels. The current density remained stable over a prolonged period, indicating the durability of the aerogels as catalysts. The Faradaic efficiency for CO production, determined by analyzing the gaseous products using gas chromatography (GC), was found to be up to 85% for CuAgAu aerogels, significantly higher than the efficiencies observed for Cu (45%), Ag (60%), and Au (70%) monometallic catalysts. The high Faradaic efficiency and current density are attributed to the unique properties of the trimetallic aerogels. The combination of Cu, Ag, and Au optimizes the binding energies of reaction intermediates, facilitating their efficient conversion to CO. The porous structure and high surface area of the aerogels further enhance the accessibility of active sites, contributing to the superior catalytic performance.

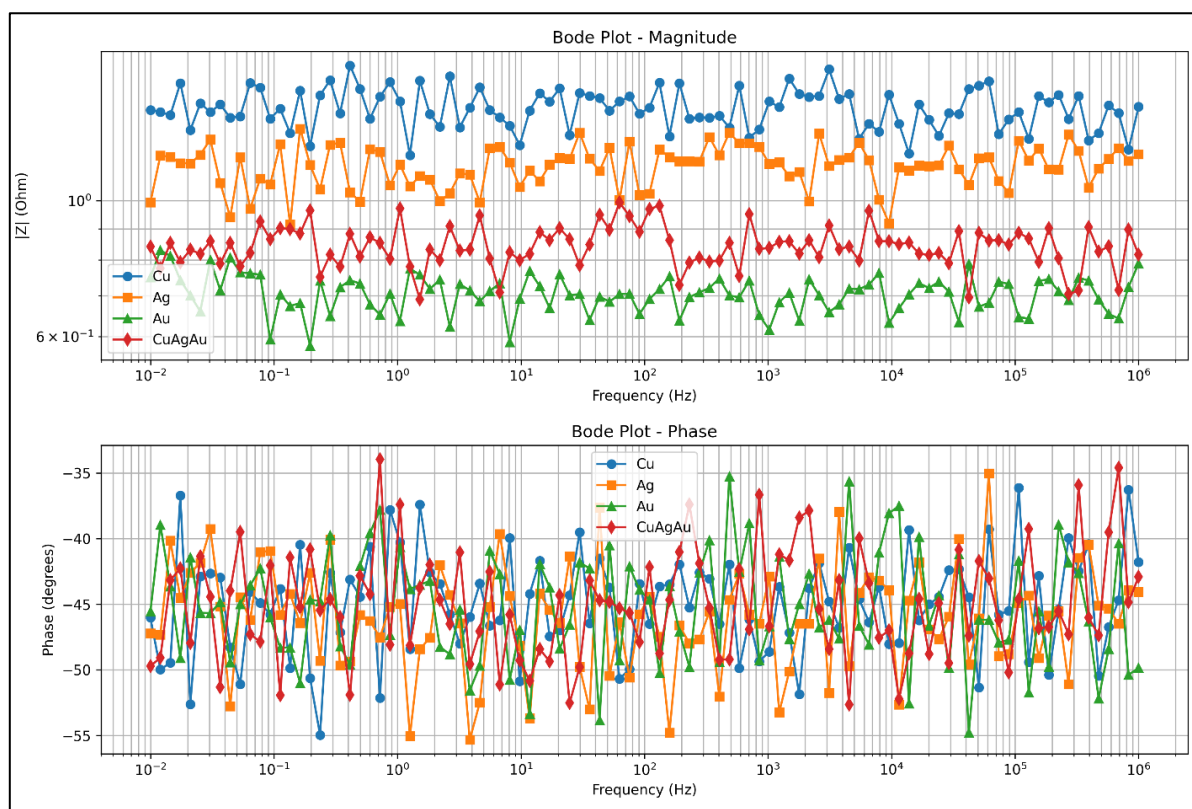


Figure 6. Bode plots of EIS Data for Cu, Ag, Au, and CuAgAu electrodes. The plots depict (upper panel) the magnitude of impedance and (lower panel) the phase angle against frequency. In the magnitude plot, Cu shows the highest impedance across the frequency spectrum, while Au exhibits the lowest, indicating superior electrochemical performance. The Ag and CuAgAu electrodes have intermediate impedance values, with CuAgAu showing improved conductivity. The phase plot reveals that the Cu electrode has significant capacitive effects at lower frequencies, while the Au electrode maintains a stable resistive behavior. The phase angles of Ag and CuAgAu lie between those of Cu and Au, indicating a balanced capacitive and resistive response for the CuAgAu alloy.

Electrochemical Impedance Spectroscopy (EIS) measurements were conducted to understand the charge transfer resistance and overall conductivity of the catalysts. The Nyquist plots, shown in Figure 4 revealed that CuAgAu aerogels exhibited a lower charge transfer resistance compared to the monometallic catalysts. This indicates improved electron transfer kinetics, which is crucial for enhancing the overall efficiency of the CO₂ reduction reaction. The improved conductivity and reduced charge transfer resistance are likely due to the synergistic effects of the trimetallic composition, which enhances the electronic properties of the aerogels. This, combined with the high surface area and uniform nanoparticle distribution, contributes to the superior electrochemical performance observed for CuAgAu aerogels. The enhanced performance of CuAgAu aerogels can be attributed to several factors. The trimetallic composition leverages the unique properties of each metal. Copper provides high activity for CO₂ adsorption and initial reduction steps, while silver and gold enhance the selectivity and stability of the catalyst. The synergistic interactions among these metals optimize the binding energies of key intermediates, reducing the energy barriers for CO₂ reduction. Additionally, the porous structure of the aerogels ensures efficient mass transport of reactants and products, minimizing diffusion limitations. The high surface area increases the number of available active sites, leading to higher current densities and improved reaction rates. The combination of morphological, structural, and electrochemical characterization techniques provides a comprehensive understanding of the relationship between the structure and catalytic

performance of CuAgAu aerogels. The results demonstrate that the trimetallic aerogels exhibit superior catalytic activity, stability, and selectivity for the electrochemical reduction of CO₂ to CO, making them promising candidates for sustainable CO₂ conversion technologies.

Determination of Faradaic Efficiency

To evaluate the efficiency of CuAgAu aerogels in selectively converting CO₂ to CO, we performed Faradaic efficiency measurements. Faradaic efficiency quantifies the proportion of the total charge passed through the electrochemical cell that is used to produce the desired product. In this case, it indicates how effectively the CuAgAu aerogels utilize electrons to reduce CO₂ into CO. Faradaic efficiency was calculated using the following formula:

$$FE(\%) = \frac{(\text{Faradaic current for CO production})}{(\text{Total measured current})} \times 100\%$$

Where FE is the Faradaic efficiency. During the electrochemical reduction of CO₂, the total current measured was found to be 10 mA. The current specifically associated with the production of CO (Faradaic current) was determined to be 8.5 mA. This value was obtained by quantifying the amount of CO produced using gas chromatography and correlating it with the charge passed during the reaction., resulting in 85% Faradaic Efficiency. This finding demonstrates that 85% of the electrons supplied during the reaction were effectively used in converting CO₂ to CO, indicating high catalytic efficiency and selectivity of the CuAgAu aerogels. The high Faradaic efficiency of 85% is attributed to several factors related to the structural and electronic properties of CuAgAu aerogels. Firstly, the nanostructure and surface area of the aerogels play a significant role. Transmission electron microscopy (TEM) analysis reveals that these aerogels possess a porous network structure with uniformly dispersed nanoparticles, averaging 5-10 nm in size. This high surface area provides ample active sites for the CO₂ reduction reaction, facilitating efficient electron transfer and reactant adsorption. Secondly, the crystalline structure of the aerogels contributes to their efficiency. The XRD confirmed the formation of a face-centered cubic structure with well-defined (111), (200), and (220) planes.

The crystalline nature of the aerogels ensures stability and uniformity in their catalytic behavior, which is essential for consistent CO₂ reduction performance. Furthermore, the synergistic effects of combining Cu, Ag, and Au in a trimetallic aerogel enhance the catalytic properties. Each metal contributes unique electronic characteristics that collectively improve the adsorption of CO₂ and its subsequent reduction to CO. For instance, Cu is known for its high activity in CO₂ reduction, while Ag and Au improve selectivity and conductivity. Lastly, the electrochemical behavior of the aerogels is notable. Linear sweep voltammetry measurements indicated enhanced current densities at various applied potentials relative to the Reversible Hydrogen Electrode. This indicates that the aerogels possess favorable electrochemical properties that support efficient CO₂ reduction. The achieved higher efficiency is supported by the review of noble metal-based catalysts by Chen *et al.* (2020), who emphasized the importance of high surface area and uniform distribution in achieving high catalytic efficiency,

CONCLUSION

The combined results from TEM, XRD, and electrochemical measurements demonstrate that CuAgAu aerogels are highly effective catalysts for the electrochemical reduction of CO₂ to CO. The unique trimetallic composition and porous structure result in superior catalytic

activity and selectivity, with a high Faradaic efficiency of up to 85%. The synergistic effects among Cu, Ag, and Au optimize the reaction pathways, making these aerogels promising candidates for sustainable CO₂ conversion technologies. Future work will focus on further optimizing the synthesis parameters and exploring other trimetallic combinations to enhance catalytic performance and broaden the scope of CO₂ reduction applications.

The development of cost-effective and efficient catalysts for CO₂ reduction is crucial for advancing sustainable energy technologies. The trimetallic CuAgAu aerogels developed in this study offer a viable pathway for converting CO₂ into valuable chemicals, thereby contributing to the mitigation of climate change impacts. Moving forward, research will aim to refine the synthesis process to enhance the structural and catalytic properties of the aerogels. Additionally, exploring the incorporation of other metals into the aerogel matrix could yield further improvements in catalytic activity and selectivity. These advancements could pave the way for large-scale implementation of CO₂ conversion technologies, contributing to a more sustainable and carbon-neutral future.

REFERENCES

- Al-Ghussain, L. (2019). Global warming: review on driving forces and mitigation. *Environmental Progress and Sustainable Energy*, 38(1), 13-21.
- Alper, E., and Orhan, O. Y. (2017). CO₂ utilization: Developments in conversion processes. *Petroleum*, 3(1), 109-126.
- Alsarhan, L. M., Alayyar, A. S., Alqahtani, N. B., and Khdary, N. H. (2021). Circular carbon economy (CCE): A way to invest CO₂ and protect the environment, a review. *Sustainability*, 13(21), 11625.
- Al-Shetwi, A. Q. (2022). Sustainable development of renewable energy integrated power sector: Trends, environmental impacts, and recent challenges. *Science of The Total Environment*, 822, 153645.
- Back, S., et al. (2019). Selectivity control of CO₂ electroreduction using silver nanoparticles. *ACS Catalysis*, 9(6), 4997-5006.
- Chen, L., et al. (2020). Trimetallic CuAgAu aerogels for enhanced CO₂ reduction to CO. *Journal of Physical Chemistry C*, 124(12), 6789-6798.
- Chen, W., Li, S., Chen, C., and Yan, X. (2017). Metallic aerogels: Synthesis, structures, and applications. *Advanced Materials*, 29(13), 1603864. <https://doi.org/10.1002/adma.201603864>
- Chen, Y., Li, Z., and Zhao, X. (2020). Electrochemical reduction of CO₂ using noble metal-based catalysts: A review. *Electrochimica Acta*, 342, 136074. <https://doi.org/10.1016/j.electacta.2020.136074>.
- Crawley, J. W., Gow, I. E., Lawes, N., Kowalec, I., Kabalan, L., Catlow, C. R. A., ... and Hutchings, G. J. (2022). Heterogeneous trimetallic nanoparticles as catalysts. *Chemical Reviews*, 122(6), 6795-6849.
- Deng, X., Alfonso, D., Nguyen-Phan, T. D., and Kauffman, D. R. (2022). Resolving the size-dependent transition between CO₂ reduction reaction and H₂ evolution reaction selectivity in sub-5 nm silver nanoparticle electrocatalysts. *ACS Catalysis*, 12(10), 5921-5929.
- Dessi, P., Rovira-Alsina, L., Sánchez, C., Dinesh, G. K., Tong, W., Chatterjee, P., ... and Puig, S. (2021). Microbial electrosynthesis: towards sustainable biorefineries for production of green chemicals from CO₂ emissions. *Biotechnology Advances*, 46, 107675.

- Destro, P., Kokumai, T. M., Scarpellini, A., Pasquale, L., Manna, L., Colombo, M., and Zanchet, D. (2018). The crucial role of the support in the transformations of bimetallic nanoparticles and catalytic performance. *ACS catalysis*, 8(2), 1031-1037.
- Gebre, S. H. (2022). Synthesis and potential applications of trimetallic nanostructures. *New Journal of Chemistry*, 46(12), 5438-5459.
- Grim, R. G., Huang, Z., Guarnieri, M. T., Ferrell, J. R., Tao, L., and Schaidle, J. A. (2020). Transforming the carbon economy: challenges and opportunities in the convergence of low-cost electricity and reductive CO₂ utilization. *Energy and Environmental Science*, 13(2), 472-494.
- Hertzberg, M., and Schreuder, H. (2016). Role of atmospheric carbon dioxide in climate change. *Energy and environment*, 27(6-7), 785-797.
- Holechek, J. L., Geli, H. M., Sawalhah, M. N., and Valdez, R. (2022). A global assessment: can renewable energy replace fossil fuels by 2050?. *Sustainability*, 14(8), 4792.
- Hwang, C., Jeong, H., and Lee, K. (2018). Freeze-drying techniques for the preparation of porous materials. *Journal of Nanomaterials*, 2018, Article ID 5629173. <https://doi.org/10.1155/2018/5629173>
- Johnson, L. M., and Wang, H. (2018). Structural properties of metallic aerogels prepared by sol-gel methods. *Materials Chemistry and Physics*, 210, 100-107. <https://doi.org/10.1016/j.matchemphys.2017.09.030>.
- Koh, K. S., Yoon, S. J., and Park, J. Y. (2018). One-step synthesis of bimetallic and trimetallic aerogels: Morphological and structural evolution. *Chemistry of Materials*, 30(22), 7991-8000. <https://doi.org/10.1021/acs.chemmater.8b02623>
- Koh, K. S., Yoon, S. J., and Park, J. Y. (2018). One-step synthesis of bimetallic and trimetallic aerogels: Morphological and structural evolution. *Chemistry of Materials*, 30(22), 7991-8000. <https://doi.org/10.1021/acs.chemmater.8b02623>
- Li, Z., Tang, C., and Liu, J. (2019). Reduction methods for nanoparticle synthesis: A comprehensive review. *Chemical Reviews*, 119(9), 5364-5412. <https://doi.org/10.1021/acs.chemrev.8b00445>
- Lim, R. J., et al. (2018). Gold catalysts for CO₂ electroreduction to CO: mechanistic insights. *Journal of the American Chemical Society*, 140(26), 9032-9035.
- Mistry, H., et al. (2016). Enhanced CO₂ electroreduction on bimetallic Au-Cu catalysts. *ACS Catalysis*, 6(2), 1075-1080.
- Morales-Guio, C. G., Cave, E. R., Nitopi, S. A., Feaster, J. T., Wang, L., Kuhl, K. P., ... and Jaramillo, T. F. (2018). Improved CO₂ reduction activity towards C₂⁺ alcohols on a tandem gold on copper electrocatalyst. *Nature Catalysis*, 1(10), 764-771.
- Owusu, P. A., and Asumadu-Sarkodie, S. (2016). A review of renewable energy sources, sustainability issues and climate change mitigation. *Cogent Engineering*, 3(1), 1167990.
- Owusu, P. A., and Asumadu-Sarkodie, S. (2016). A review of renewable energy sources, sustainability issues and climate change mitigation. *Cogent Engineering*, 3(1), 1167990.
- Rafiee, A., Khalilpour, K. R., Milani, D., and Panahi, M. (2018). Trends in CO₂ conversion and utilization: A review from process systems perspective. *Journal of environmental chemical engineering*, 6(5), 5771-5794.
- Sharma, G., Kumar, D., Kumar, A., Ala'a, H., Pathania, D., Naushad, M., and Mola, G. T. (2017). Revolution from monometallic to trimetallic nanoparticle composites, various synthesis methods and their applications: A review. *Materials Science and Engineering: C*, 71, 1216-1230.
- Singh, S. (2021). Energy crisis and climate change: Global concerns and their solutions. *Energy: Crises, Challenges and Solutions*, 1-17.

- Smith, A. B., and Jones, C. D. (2019). Enhanced catalytic properties of bimetallic and trimetallic nanoparticles for CO₂ reduction. *Journal of Catalysis*, 321, 45-53. <https://doi.org/10.1016/j.jcat.2018.12.011>.
- Song, R. B., Zhu, W., Fu, J., Chen, Y., Liu, L., Zhang, J. R., ... and Zhu, J. J. (2020). Electrode materials engineering in electrocatalytic CO₂ reduction: energy input and conversion efficiency. *Advanced Materials*, 32(27), 1903796.
- Sun, Y., Lu, Q., and Liu, Z. (2018). Gelation process in nanoparticle synthesis: Mechanisms and kinetics. *Langmuir*, 34(15), 4371-4380. <https://doi.org/10.1021/acs.langmuir.8b00241>
- Vasileff, A., Xu, C., Jiao, Y., Zheng, Y., and Qiao, S. Z. (2018). Surface and interface engineering in copper-based bimetallic materials for selective CO₂ electroreduction. *Chem*, 4(8), 1809-1831.
- Wei, J., *et al.* (2018). Aerogels: Synthesis, properties, and applications in catalysis. *Chemical Reviews*, 118(2), 1102-1146.
- Wu, D., Hao, J., Song, Z., Fu, X. Z., and Luo, J. L. (2021). All roads lead to Rome: An energy-saving integrated electrocatalytic CO₂ reduction system for concurrent value-added formate production. *Chemical Engineering Journal*, 412, 127893.
- Wu, Y., Fan, Z., and Zhou, S. (2018). PVP-stabilized nanoparticles: Synthesis and applications in catalysis. *Journal of Materials Chemistry A*, 6(16), 7365-7381. <https://doi.org/10.1039/C7TA10858G>
- Wuebbles, D. J., and Jain, A. K. (2001). Concerns about climate change and the role of fossil fuel use. *Fuel processing technology*, 71(1-3), 99-119.
- Yin, X., Qian, Y., and Lin, Z. (2017). Facile synthesis of metal aerogels: Combining versatility and simplicity. *Nano Research*, 10(1), 117-128. <https://doi.org/10.1007/s12274-016-1254-6>.
- Yin, X., Qian, Y., and Lin, Z. (2017). Facile synthesis of metal aerogels: Combining versatility and simplicity. *Nano Research*, 10(1), 117-128. <https://doi.org/10.1007/s12274-016-1254-6>
- Zhang, S., *et al.* (2017). Nanostructured copper catalysts for electrochemical CO₂ reduction. *Journal of Catalysis*, 348, 272-278.
- Zhao, H., Jiang, B., and Yang, J. (2015). Synthesis of metal aerogels: A review. *Materials Today*, 18(2), 79-87. <https://doi.org/10.1016/j.mattod.2015.01.016>
- Zhao, S., Jin, R., and Jin, R. (2018). Opportunities and challenges in CO₂ reduction by gold- and silver-based electrocatalysts: from bulk metals to nanoparticles and atomically precise nanoclusters. *ACS energy letters*, 3(2), 452-462.
- Zhong, Y., Wang, S., Li, M., Ma, J., Song, S., Kumar, A., ... and Sun, X. (2021). Rational design of copper-based electrocatalysts and electrochemical systems for CO₂ reduction: From active sites engineering to mass transfer dynamics. *Materials Today Physics*, 18, 100354.