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Development of a Modified 2.4 GHz Microwave System for In-Liquid Plasma Generation and its Application in Aqueous Metal Sulfate Treatment

Pria Gautama[1,*,a], Shinfuku Nomura[2,b], Nurfiansyah Nurfiansyah[1,c], A. Ariputra[1,d], Arfandy Arfandy[1,e], Yiyin Klistafani[3,f], Tri Susilo[4,g] and Muhammad Firdan[5,h]

[1]Mechanical Department, Politeknik Negeri Ujung Pandang, Jalan Perintis Kemerdekaan, Makassar, 90245, Indonesia
[2]Heat and Mass Transfer Laboratory, Ehime University, 3 Bunkyo-cho, Matsuyama, Ehime, 790-8577, Japan
[3]Department of Mechanical Engineering and Materials Science, Washington University in St. Louis, One Brookings Drive, St. Louis, Missouri 63130, United States of America
[4]Department of Electrical Engineering, King Fahd University of Petroleum and Minerals, Dhahran, 31261, Saudi Arabia
[5]Mechanical Departement, Borneo Tarakan University, Tarakan, North Kalimantan, 77115, Indonesia

[*,a]pria_gautama@poliupg.ac.id, [b]nomura.shinfuku.mg@ehime-u.ac.jp, [c]nurfiansyah@poliupg.ac.id, [d]a.ariputra@poliupg.ac.id, [e]arfandy@poliupg.ac.id, [f]kyiyin@wustl.edu, [g]g202522670@kfupm.edu.sa, [h]firdan@borneo.ac.id

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Abstract

This study aims to develop a plasma generation system that can be effectively applied in liquid phase environments, specifically for the treatment of aqueous metal sulfate. The research was conducted through several sequential stages designed to streamline and clarify the experimental procedure: design, fabrication (assembly), testing, observation, and data analysis. The effectiveness of the device design was confirmed by the successful formation of plasma within the reactor. Experimental observations confirm that plasma treatment duration significantly affects the pH reduction in $NiSO_4$ and $FeSO_4$ solutions. $FeSO_4$ exhibits a steeper pH decline than $NiSO_4$ under the same exposure period, implying a stronger response to plasma-generated acidic species. The pH decreased from 6.14 to 3.6 for $NiSO_4$ and from 5.26 to 3.1 for $FeSO_4$.

1. Introduction

The unique properties of plasma, characterized by highly energetic charged and neutral species, make it a powerful tool for driving complex reactions. Recently, in-liquid plasma systems (ILP) have emerged as a promising technology for environmental and materials applications [1-3]. Microwave-induced plasma at 2.45 GHz is an attractive method for generating ILP due to its efficient energy transfer. Despite its potential, generating stable in-liquid plasma is challenging.

This study aims to develop a plasma generation system that can be effectively applied in liquid-phase environments, specifically for the treatment of aqueous metal sulfate solutions. The system is designed to investigate the influence of plasma exposure on pH variation and the chemical behavior of metal ions such as Ni and Fe in sulfate-based solutions. The findings from this study are expected to contribute to the development of environmentally friendly and energy-efficient plasma-assisted technologies for metal extraction and refinement, particularly from nickel-bearing minerals such as saprolite

Microwave-induced plasma is generated when electromagnetic energy at microwave frequency interacts with gas molecules or vapors, leading to their ionization. When a sufficiently strong electric field is applied, free electrons in the medium gain energy from the oscillating microwave field and collide with neutral atoms or molecules. These collisions transfer energy, resulting in the ionization of the gas and the creation of charged particles such as ions, electrons, and excited species. Once the number of charged particles reaches a critical threshold, a self-sustaining plasma discharge occurs [4].

Based on the theoretical description above, it can be concluded that the mechanism of in-liquid plasma generation from a microwave source involves several key components and coupled physical processes. A microwave source (2.45 GHz magnetron) generates electromagnetic waves, which are transmitted through awaveguide or resonant cavity to focus and couple energy into the reactor [5-6]. The quartz reactor or beaker containing the liquid serves as the dielectric medium where strong localized electric fields are formed. Within these regions, vapor bubbles are generated due to microwave heating and act as microreactors for plasma ignition through dielectric breakdown. The resulting plasma zone appears as small, luminous regions inside the bubbles where microdischarges occur. Furthermore, gas emission and bubble dynamics play a critical role in sustaining the discharge, showing continuous formation, expansion, and collapse of plasma bubbles under oscillating microwave fields.

The design and fabrication of a 2.4 GHz microwave plasma generation system have been previously reported [7-8]. however, several shortcomings were observed in its practical application. These include significant electromagnetic wave leakage and a high level of water vapor entering the vacuum chamber. Such issues not only affect the operational stability of the experiments but also present potential safety risks to users due to electromagnetic radiation exposure.

This study builds upon previous designs by introducing key modifications to effectively contain electromagnetic waves, thereby creating a safer and more robust system. The simple, modified microwave system developed here is capable of sustaining stable plasma directly within an aqueous bulk, an achievement that has been limited in earlier, leak-prone setups. The system was tested on solutions of nickel sulfate ($NiSO_4$) and ferrous sulfate ($FeSO_4$) to assess its performance in treating metal sulfates. The principal novelty of this research is its demonstration of a commercially available microwave oven, repurposed with improved safety and containment, for direct in-liquid plasma generation and its subsequent application in decomposing metal sulfates.

2. Research Methodology

2. 1. Design

The research was conducted through several sequential stages designed to streamline and clarify the experimental procedure: design, fabrication (assembly), testing, observation, and data analysis. The design phase employed the Solid Works software to visualize and refine the model.

In this study, a Sharp R-21DO23L microwave oven with an output power of 450 W (Figure 1) was utilized as the primary source of microwave energy. The waveguide was constructed from a 3 mm thick aluminum plate with dimensions of 330 mm × 12 mm × 12 mm. The plasma reactor comprised a Teflon cylinder (diameter 68 mm, height 29 mm), an acrylic tube (diameter 61 mm, height 177 mm), and an aluminum cylinder (diameter 30 mm). Two tungsten electrodes (3 mm each), a manifold, a pressure gauge, and a heat-resistant O-ring were included to ensure proper sealing of the reactor system. The test liquids consisted of $NiSO_4$ and $FeSO_4$ with each concentration 0.0001 Molar.

FIGURE 1. Microwave Oven Sharp 2.4 GHz type R-21DO23L

Functional design of development the 2.45 GHz microwave plasma generator consisted of three main subsystems: a microwave oven serving as the source of electromagnetic energy, a waveguide for directing the microwaves toward the reactor, and a reactor chamber for plasma observation. Structural design of wave guide was modified by adding the wave guide to magnetron tunel as shown at Figure 2 and Figure 3. The waveguide, fabricated from aluminum, measured 330 mm x 120 mm × 120 mm, with an 8 mm electrode inlet hole positioned 122 mm from the magnetron, corresponding to one wavelength (λ) at 2.45 GHz [7,8].

The reactor was built from an acrylic tube (inner diameter 61 mm, height 177 mm) with teflon covers on both ends, sealed by an O-ring to prevent leakage (Figure 4). Final design of microwave plasma generator by assembling three major components: microwave, wave guide and reactor (Figure 5).

The present design introduces several improvements compared to the previous version of the microwave plasma generator [7,8]. A new reactor holder component was fabricated from aluminum with an outer diameter of 110 mm and an inner diameter of 61 mm, featuring an internal thread structure for secure attachment. This holder was mounted on the upper surface of the waveguide to provide mechanical stability and to facilitate easy removal of the reactor during liquid replacement or maintenance. Another notable modification involves the microwave casing design. The upper cover was fitted with a precisely machined aperture sealed with an acrylic window, effectively minimizing electromagnetic radiation leakage to a level well within the safety limits for human exposure. These improvements not only enhance operational safety but also ensure better reproducibility and convenience during experimental procedures.

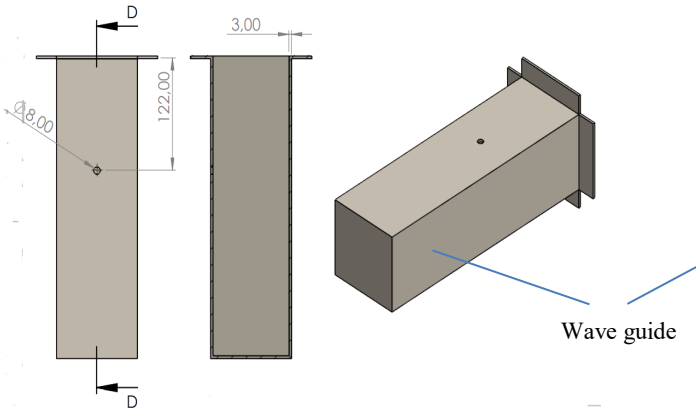

FIGURE 2. Design of plasma waveguide

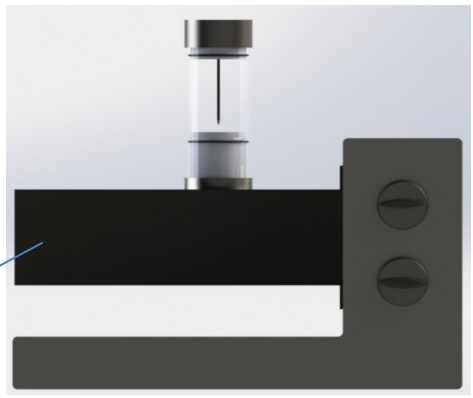

FIGURE 3. Waveguide installed to megatrone

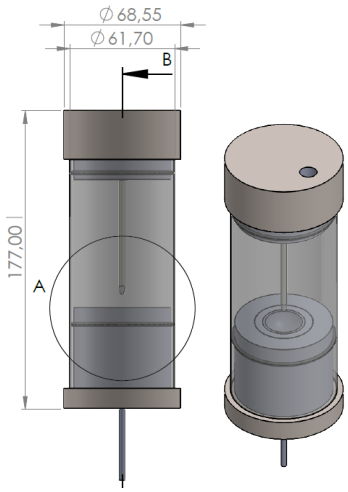

FIGURE 4. Plasma reactor design

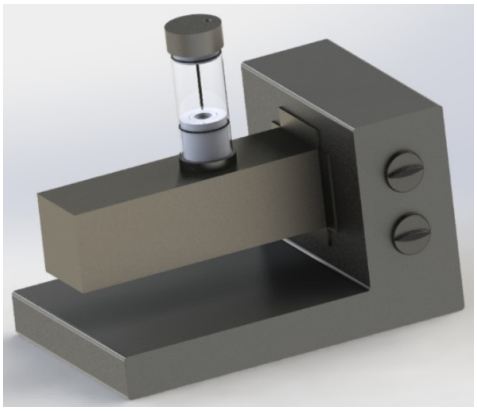

FIGURE 5. Design of a 2.45 GHz Microwave Plasma Generator

2. 2. Method

The experimental preparation involved setting up the 2.45 GHz microwave plasma system along with liquid samples consisting of $NiSO_4$ and $FeSO_4$ solutions at a concentration of 0.0001 M. Prior to testing, the plasma generator was calibrated to ensure efficient transfer of electromagnetic energy into the plasma reactor. The experiments were then performed by varying the plasma exposure times of 60, 90, and 120 seconds for each solution. The overall experimental setup and schematic of the 2.45 GHz microwave plasma apparatus are shown in Figure 6. Observations and data collection focused on monitoring the onset of plasma formation and measuring the pH values of each solution before and after plasma treatment. The collected data were subsequently analyzed to evaluate the effect of plasma exposure time on the chemical properties of the solutions.

The experimental arrangement consists of a plasma reactor, microwave plasma generator, pressure gauge, valve, and liquid sample. In each trial, 100 mL of 0.0001 M $NiSO_4$ solution was introduced into the reactor, which was mounted on the waveguide. The system pressure was reduced to –30 kPa using a vacuum pump before applying microwave power at the high setting. Plasma initiation was observed and recorded, and after exposure durations of 60, 90, and 120 seconds, both the microwave source and vacuum pump were turned off. The pH of each treated solution was subsequently measured to evaluate the effect of plasma interaction.

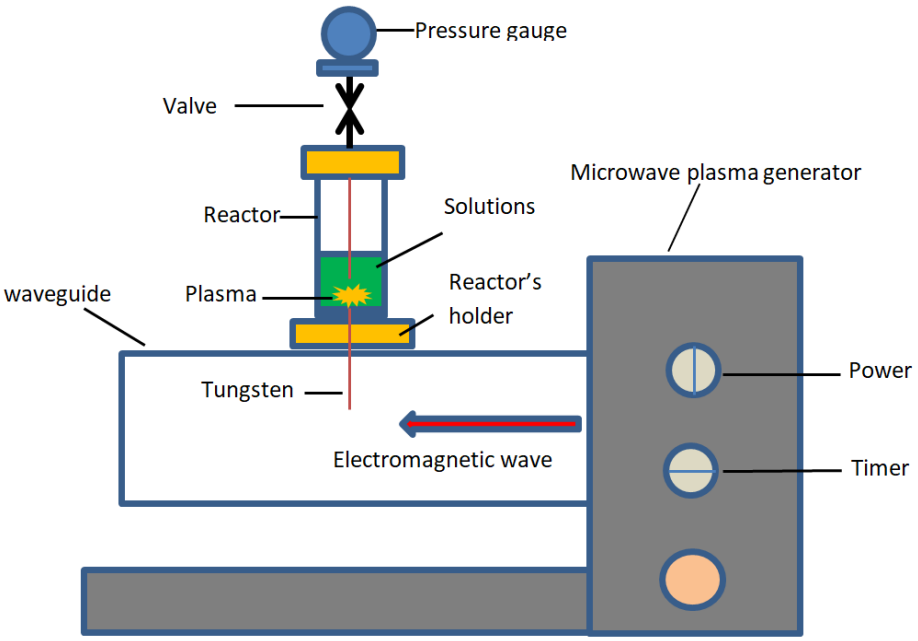

FIGURE 6. Setup of the in-liquid microwave plasma experiment

3. Results and Discussion

3. 1. Results

Results of the design and fabrication of the in-liquid plasma generator shown at Figure 7.

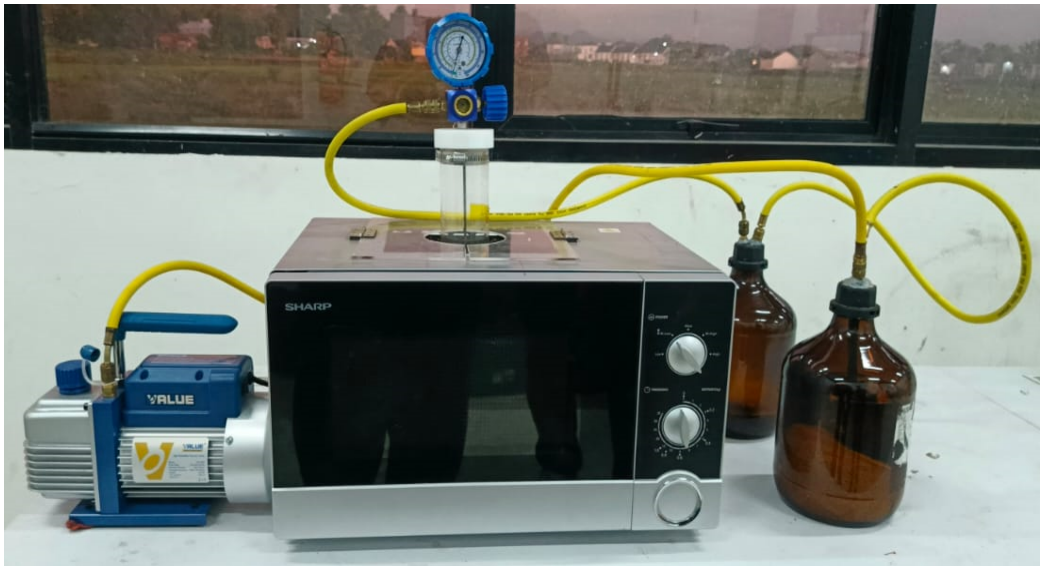

FIGURE 7. Modified Microwave Plasma Generator 2.45 GHz

The effectiveness of the device design was confirmed by the successful formation of plasma within the reactor. Plasma treatment of $NiSO_4$ and $FeSO_4$ solutions at a concentration of 0.0001 M with exposure durations of 60, 90, and 120 seconds exhibited a consistent reduction in pH values. Specifically, the pH of $NiSO_4$ decreased from 6.14 to 3.6, while that of $FeSO_4$ declined from 5.26 to 3.1 following plasma exposure. The characteristics of plasma behavior in liquid are summarized in **Table 1.**

TABLE 1. Results of characteristics plasma in liquid

		Solutions					
		$NiSO_4$, $6H_2O$			$FeSO_4$, $7H_2O$		
Concentration (M)	Time (s)	Ignition (s)	pH (before)	pH (after)	ignition	pH (before)	pH (after)
0.0001	60	3.35	6.14	4.59	2.66	5.26	3.4
	90	2.53	6.14	3.93	2.55	5.26	3.36
	120	2.77	6.14	3.6	2.62	5.26	3.1

3. 2. Discussion

The plasma treatment of $NiSO_4 \cdot 6H_2O$ and $FeSO_4 \cdot 7H_2O$ solutions at a concentration of 0.0001 M with exposure durations ranging from 60 to 120 seconds revealed two key phenomena: plasma ignition behavior and pH reduction. In both solutions, plasma ignition occurred within 2.53 to 3.35 seconds.

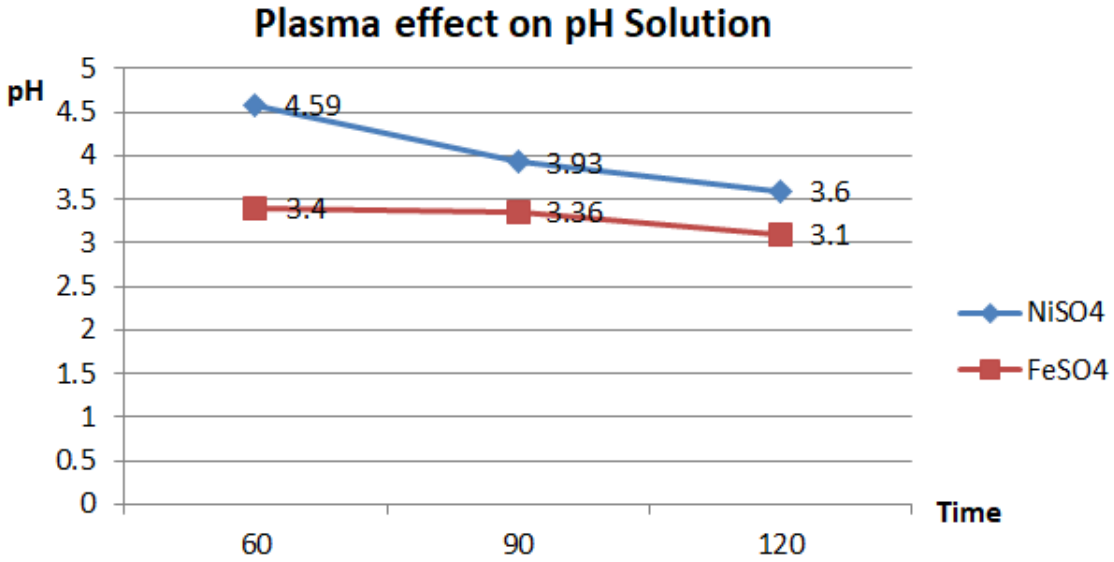

FIGURE 8. Effect of plasma on pH solutions

A consistent and notable finding was the decline in pH following plasma exposure. For $NiSO_4$ solutions, the pH decreased from an initial range of 6.14–3.6, while $FeSO_4$ solutions exhibited a reduction from 5.26–3.1. These pH changes for both solutions are illustrated in Figure 8.

The observed decrease in pH can be attributed to the formation of reactive oxygen species (ROS) and reactive nitrogen species (RNS) during plasma discharge, including hydroxyl radicals (•OH), hydrogen peroxide (H_2O_2), and nitric and nitrous acids. These reactive species significantly increase the acidity of the treated solution through oxidation and ionization reactions [1,3,6]. The acidification effect became more pronounced with longer plasma exposure times (90–120 seconds), confirming that extended treatment enhances the generation of radicals and accelerates proton release processes [9,10].

Furthermore, $FeSO_4$ solutions exhibited a slightly greater pH reduction compared to $NiSO_4$ under the same treatment conditions. This behavior is likely related to the higher redox activity of Fe^{2+} ions, which react more readily with plasma-generated species, facilitating oxidation and subsequent proton formation [10,12]. Such enhanced redox interactions have also been reported in plasma–liquid systems containing transition metal ions, where Fe-based solutions demonstrated faster acidification rates than Ni-based ones [9,11].

4. Conclusion

A device capable of generating plasma using a microwave-based system has been successfully developed. Experimental observations confirm that plasma treatment duration significantly affects the pH reduction in $NiSO_4$ and $FeSO_4$ solutions. $FeSO_4$ exhibits a steeper pH decline than $NiSO_4$ under the same exposure period,

implying a stronger response to plasma-generated acidic species. The pH decreased from 6.14 to 3.6 for $NiSO_4$ and from 5.26 to 3.1 for $FeSO_4$.

Future investigations should aim to assess the performance of in-liquid plasma on complex saprolite matrices containing diverse metal and mineral constituents, in order to approximate real operational environments. Understanding the relationship between plasma-induced pH shifts and the selective precipitation or separation of Fe and Ni remains a critical area of study. In addition, optimization of key plasma variables including intensity, frequency, and exposure period will be necessary to maximize separation efficiency. These efforts could contribute to the advancement of sustainable, low cost in liquid plasma technologies for the processing of saprolite as a nickel source.

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Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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