

# A Method for Preparing Gallium-nickel Alloy Solid Targets by Alternating Current Plating

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**Abstract:** In this paper, an alternating current electroplating platform is built. Gallium-nickel alloy plating is prepared on copper targets by using alternating current electroplating methods. The mixed solution of gallium and nickel metal ions is used as the electroplating solution. The differences of gallium-nickel alloy targets prepared by alternating current plating under different conditions of temperature, current density and pH of the plating solution were investigated, and a set of electroplating conditions for gallium-nickel alloy targets prepared by AC electroplating platform were summarized in this article. It is concluded that the gallium-nickel alloy coating with excellent surface quality can be obtained under the conditions of the temperature of the plating solution below 80°C, the current density of 25-50 mA/cm<sup>2</sup> and the pH of the plating solution of 1.75-2.25 by the analysis of a series of experimental results. At the same time, it seems that the quality of gallium-nickel alloy coating obtained by AC plating is better than that of DC plating under the same conditions through the characterization results. Therefore, the scheme of using an AC electroplating platform to prepare gallium-nickel alloy targets is feasible.

**Keywords:** AC electroplating; Gallium-nickel alloy; Solid target.

## 1. Introduction

Positron emission computed tomography (PET) can be used to study the diagnosis and treatment of various diseases such as tumors, cardiovascular, cerebrovascular diseases, and nervous system diseases<sup>[1]</sup>. PET can timely reflect the distribution range of diseased cells, changes in physiological functions, and other information<sup>[2]</sup>. And it is a relatively advanced clinical examination imaging technology in the field of nuclear medicine. PET uses annihilation radiation and positron collimation (or photon collimation) technology<sup>[3]</sup> to non-destructively and quantitatively measure the spatial distribution, quantity and dynamic changes of PET imaging agents or their metabolites in vivo<sup>[4]</sup>. It obtains molecular-level imaging information on the biochemical, physiological and functional metabolic changes caused by the interaction between PET imaging agents and targets in vivo<sup>[5,6]</sup>, which can provide important data for clinical research.

Commonly used positron-emitting radionuclides include <sup>18</sup>F<sup>[7]</sup>, <sup>11</sup>C<sup>[8]</sup>, <sup>64</sup>Cu<sup>[9]</sup>, <sup>89</sup>Zr<sup>[10]</sup>, <sup>68</sup>Ga<sup>[11]</sup>, etc. Among these positron nuclides, <sup>68</sup>Ga has become a hot research topic in recent years due to its excellent performance, and the <sup>68</sup>Ge/<sup>68</sup>Ga generator is an ideal generator for obtaining <sup>68</sup>Ga nuclides<sup>[12]</sup>. The half-life of the parent nuclide <sup>68</sup>Ge is relatively long, reaching 270.95 days, which makes the service life of the <sup>68</sup>Ge/<sup>68</sup>Ga generator up to about one year. The half-life of the daughter nuclide <sup>68</sup>Ga is 67.79 min, the decay mode is 89.1% β<sup>+</sup> decay, and the energy is 836.02 keV. The shorter half-life of <sup>68</sup>Ga matches the pharmacokinetics of small molecule drugs such as peptides<sup>[13]</sup>, making it easier to achieve high-quality dynamic imaging. Its lower decay energy can effectively protect healthy cells, making it an ideal positron nuclide for imaging. At the same time, the shorter half-life of <sup>68</sup>Ga supports a higher elution frequency without generating excessive unnecessary radiation doses for patients. The use of <sup>68</sup>Ge/<sup>68</sup>Ga generator allows users to prepare <sup>68</sup>Ge-labeled drugs without using an accelerator, extremely saving costs.

Obtaining <sup>68</sup>Ge is a key prerequisite for preparing <sup>68</sup>Ge/<sup>68</sup>Ga generators, and accelerator irradiation of gallium alloy targets is currently the main way to obtain <sup>68</sup>Ge. The existing methods for preparing gallium alloys are mainly electrochemical deposition<sup>[14]</sup> and melting method<sup>[15]</sup>. Compared with hot-melt alloys, electroplated alloys have higher hardness and lower ductility under the same metal composition, and it is easier to obtain alloys composed of high melting point and low melting point metals. Gallium, the metal used to prepare <sup>68</sup>Ge, is a metal with a low melting point. So that it's easy to melt under the irradiation of high-energy proton accelerators. Gallium alloys need to be prepared to increase the melting point of the target. Nickel is a low-priced metal with a high melting point (1455°C). It has stable chemical properties and can easily form an alloy with gallium, which can effectively increase the melting point of gallium-containing metal coatings. Therefore, this study used electrochemical deposition to prepare gallium-nickel alloy coatings. In this study, gallium-nickel alloy was electroplated in a sulfate solution system, and an AC electroplating platform was built to study the plating conditions of gallium-nickel alloy under different cathode current density, solution temperature, and solution pH. Through a series of studies, a better electroplating scheme for preparing gallium-nickel alloy solid targets by AC electroplating platform was given, and the feasibility of preparing gallium-nickel alloy targets through AC electroplating platform was studied.

## 2. Experiment

### 2.1. Preparation

Experimental equipment: WPS305B DC Power Supply. labck 500V/A adjustable AC power supply. Several high-power resistors with different resistance values. Current detector (digital display, direct reading). DF-101S heat collecting constant temperature heating magnetic stirrer (Shanghai Lichen Bangxi Instrument Technology Co., Ltd.). BS224S Electronic balance. Electroplating tank (self-

designed). Magnetic stirring bar. Pure platinum mesh anode (99.9%), NiSO<sub>4</sub>·6H<sub>2</sub>O (99.9%, Aladdin); Ga<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> (99.99%, HWRC CHEM Co., Ltd).

## 2.2. Target pretreatment

The copper target used in the experiment is a standard copper target ordered online and polished. Before using it in electrochemical deposition experiments, the target surface needs to be degreased, pickled and activated to eliminate the effects of oil stains, oxide layers or other metal impurities on the copper target surface on the electrodeposition process of the alloy layer. The process of copper target pretreatment is mainly as follows:

①The copper sheet cleaned with detergent was ultrasonically treated with alcohol for 10 min. This step is used to remove most of the oil stains on the surface of the copper target. The degreasing speed is fast and the efficiency is high. After the completion, the copper target was rinsed with deionized water and alcohol and dried.

②Prepare degreasing fluid: In experiments, the saponification reaction of alkaline solution is generally used to remove oil stains on the surface of the workpiece. The chemical degreasing formula is shown in Table 1. The chemical degreasing operation was carried out at a degreasing liquid temperature of 70°C and the degreasing time was 15 min. After chemical degreasing, the copper target was rinsed with deionized water and alcohol and dried.

③The degreased copper target was immersed in 8 mol/L sulfuric acid for pickling and activation for 10 min. After activation, the copper target was rinsed with deionized water and alcohol and dried.

After completing the copper target pretreatment, weigh and record the mass and thickness information of the target before electroplating. Then install the anode (platinum sheet) and copper target (cathode) in the electroplating tank and connect the power supply.

**Table 1.** Degreasing fluid formula

Drug	concentration
Na <sub>2</sub> CO <sub>3</sub>	20 g/L
Na <sub>3</sub> PO <sub>4</sub> ·12H <sub>2</sub> O	20 g/L

## 2.3. Plating solution preparation

The plating solution is prepared using two sulfates: NiSO<sub>4</sub>·6H<sub>2</sub>O (99.9%) and Ga<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> (99.99%). Weigh a certain amount of NiSO<sub>4</sub>·6H<sub>2</sub>O and Ga<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> and place them in a 500 mL beaker. Pour 300 mL of deionized water into the beaker and stir at room temperature until the metal salt in the solution is completely dissolved. Use dilute sulfuric acid to adjust the solution pH to between 1.0 and 2.5 to prepare the electroplating solution. The molar ratio of gallium ions (Ga<sup>3+</sup>) and nickel ions (Ni<sup>2+</sup>) in the plating solution is about 6:1.

## 2.4. Experimental methods

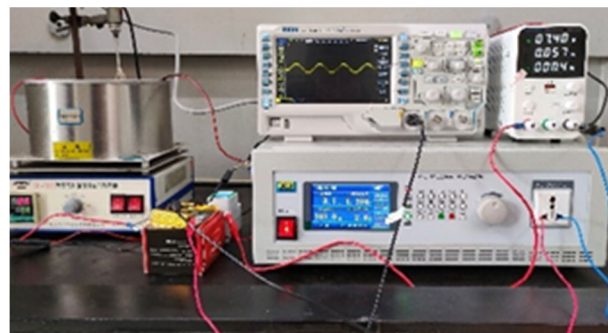
The electrochemical deposition steps involved in this study are as follows: Pretreatment of copper targets for electroplating (chemical degreasing, pickling sparks, weighing). Electroplating equipment assembly (cathode and anode assembly, plating solution filling, power supply connection). Gallium-nickel alloy coating electrodeposition. Post-plating target cleaning, weighing, data recording and post-processing (coating weight, coating quality, coating

composition determination, current efficiency calculation, etc.).

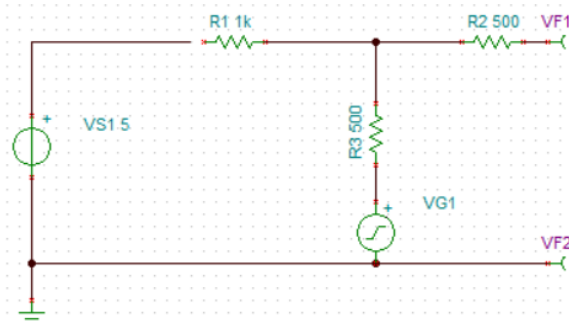
## 2.5. AC plating platform setup

During electroplating by alternating current, the cathode and anode are alternately biased positively and negatively. When the cathode is negatively biased, the gallium-nickel metal ions in the solution move toward the cathode surface under the action of the positive bias of the anode, and finally precipitate and deposit on the cathode surface. When the cathode is positively biased, the gallium-nickel alloy coating formed on the cathode surface dissolves under the action of positive voltage. The burrs or cracks on the coating surface will form a larger electric field, and the gallium-nickel alloy layer in these places will dissolve first. Therefore, when using AC voltage for gallium-nickel alloy electroplating, the voltage itself will modify the surface of the coating, and theoretically a coating with better surface quality can be obtained.

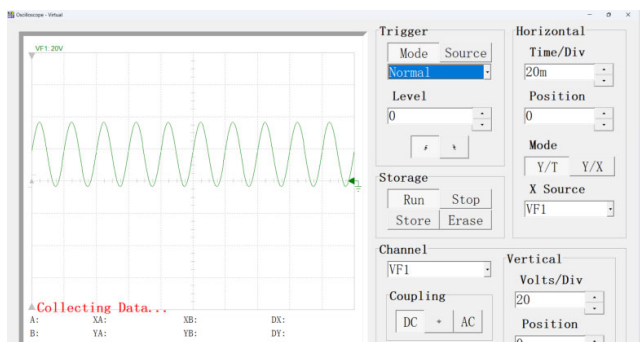
In order to achieve greater electroplating efficiency, the AC electroplating platform built in this experiment uses a DC power supply and high-power resistors of different sizes to raise the sinusoidal voltage output by the AC power supply, so that most of the sinusoidal voltage is in a positive bias and a small part is in a negative bias. During electroplating, the cathode to be plated is under negative bias voltage for a long time, receiving the deposition of gallium and nickel metal ions for more time, and is under positively bias for a short time to modify the morphology of the coating. Then, It can improve the electroplating efficiency while ensuring the quality of the coating. Based on the above situation, the physical picture of the AC electroplating platform designed in the experiment is shown in Figure 1, the circuit diagram is shown in Figure 2, and the voltage waveform output by the AC electroplating platform is shown in Figure 3.



**Figure 1.** AC electroplating platform



**Figure 2.** The circuit diagram (VF1 and VF2 are outputs)



**Figure 3.** Theoretical voltage waveform output by the AC electroplating platform

## 2.6. Experimental setup

The experiment mainly explored the quality comparison of gallium-nickel alloy coatings prepared by AC electroplating platform under different electroplating conditions. The main purpose is to compare the coating quality obtained under different cathode current density, different plating solution temperature, and different plating solution pH conditions, and to explore the electroplating stability of the AC electroplating platform. Finally, the quality difference between the gallium-nickel alloy coating obtained by using the AC plating platform and the gallium-nickel alloy coating obtained by using the DC plating platform under the same electroplating conditions is compared.

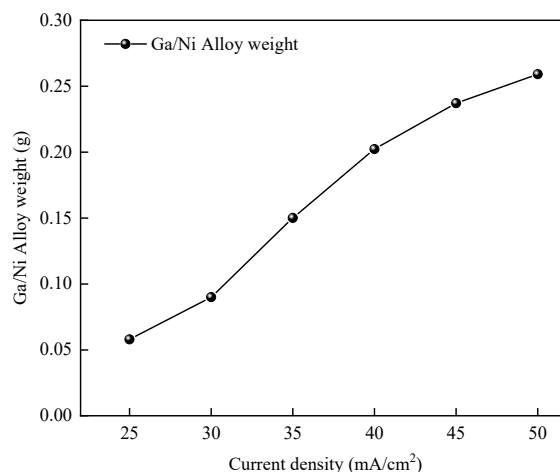
## 3. Results and Discussion

### 3.1. Electroplating performance under different cathode current density conditions

This experiment mainly explored the electroplating of gallium-nickel alloy targets on the AC electroplating platform under different cathode current densities. During the electroplating process, the anode and cathode areas remain unchanged and equal, and other electroplating conditions such as the plating solution temperature, the distance between the anode and cathode, the stirring speed, and the pH of the plating solution remain unchanged. The experiment investigated the quality differences of gallium-nickel alloy coatings obtained under current densities of 25, 30, 35, 40, 45, and 50 mA/cm<sup>2</sup>. After electroplating, weigh the target to calculate the coating weight.

The experimental results show that when using an AC electroplating platform for gallium-nickel alloy electroplating, electroplating at all different current densities can obtain gallium-nickel alloy coatings with good surface quality, and as the current density increases, more gallium-nickel alloy will be deposited on the copper substrate (Figure 4 shows the trend of the weight of the gallium-nickel alloy coating as the current density increases). However, as the current density increases to 50 mA/cm<sup>2</sup>, the excessive current density may lead to an extreme lack of ions to be plated, H<sup>+</sup> discharges and a large amount of hydrogen is released, and a rough and porous sponge-like coating is formed at the edge of the coating where the current density is the largest, and even causes the alloy coating to fall off to the point where the Cu substrate can be observed. Considering the electroplating efficiency and coating quality, it is believed that the cathode current density should be controlled within 50 mA/cm<sup>2</sup> when

using the AC electroplating platform to prepare gallium-nickel alloy targets.

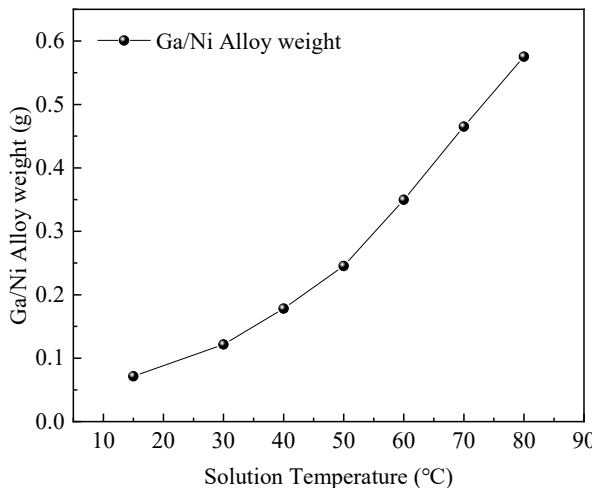


**Figure 4.** Coating weight at different current densities

### 3.2. Electroplating performance under different solution temperatures

This experiment mainly explores the electroplating of gallium-nickel alloy targets under different plating solution temperatures on the AC electroplating platform. During the electroplating process, the cathode and anode areas remain unchanged and equal, and other electroplating conditions such as cathode current density, the distance between the anode and cathode, stirring speed, and pH of the plating solution remain unchanged. The experiment investigated the differences in the quality of the gallium-nickel alloy coatings obtained at bath temperatures of 15, 30, 40, 50, 60, and 80°C. After electroplating, weigh the target to calculate the coating weight.

The experimental results show that within the temperature range of 15-80°C involved in the test, as the temperature of the plating solution increases during the electroplating process, the weight of the coating increase significantly (Figure 5 shows the change in the weight of the gallium-nickel alloy coating with temperature). At the same time, a higher solution temperature can optimize the quality of the coating. The increase in solution temperature is conducive to increasing the diffusion rate of ions and accelerating the dehydration rate of ions, thereby reducing concentration polarization, enhancing the activity of metal ions and the cathode surface, and enhancing the surface quality of the coating. However, as the solution temperature increases to 80°C, the evaporation rate of the solution accelerates during the electroplating process, resulting in serious loss of solution. The solution needs to be replenished at any time, increasing the workload. Based on the above results, it is considered that it is more appropriate to control the plating solution temperature to no more than 80°C when using an AC plating platform for electroplating in an opening environment. Considering the rapid loss of plating solution during high-temperature electroplating, this study does not involve electroplating at higher temperatures.

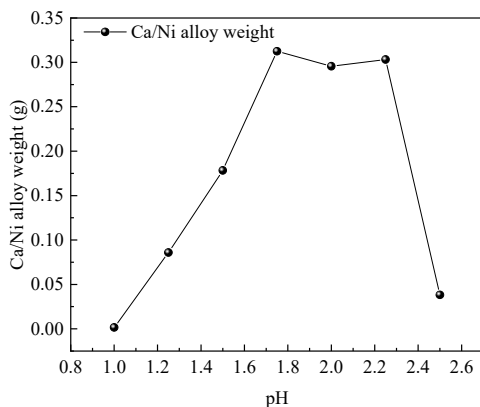


**Figure 5.** Weight gain of gallium-nickel alloy coating at different solution temperatures

### 3.3. Effect of Plating Solution pH on Coating Quality

The experiment mainly explored the electroplating of gallium-nickel alloy targets on the AC electroplating platform under different plating solution pH conditions. During the electroplating process, the cathode and anode areas remain unchanged and equal, and other electroplating conditions such as cathode current density, the distance between cathode and anode, stirring speed, and plating solution temperature remain unchanged. The experiment studied the coating conditions under the conditions of plating solution pH 1.0, 1.25, 1.5, 1.75, 2.0, and 2.5. After electroplating, weigh the target to calculate the coating weight.

The experimental results show that in the acidic plating solution system, the plating solution pH is too high (pH=2.5) or too low (pH=0.5) is not conducive to the deposition of gallium-nickel alloy coating. In these two cases, the electroplating efficiency is too low and the coating weight gain is small (Figure 6 shows the weight change of gallium-nickel alloy coating obtained under different plating solution pH conditions). The pH of the plating solution is between 1.75-2.25, which can obtain a gallium-nickel alloy coating with high electroplating efficiency and good coating quality.

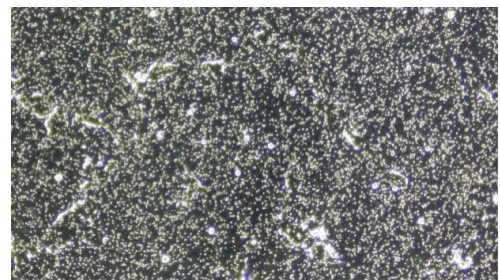


**Figure 6.** Gallium-nickel alloy coating weight under different plating solution pH

### 3.4. Effect of different power supply modes on coating quality

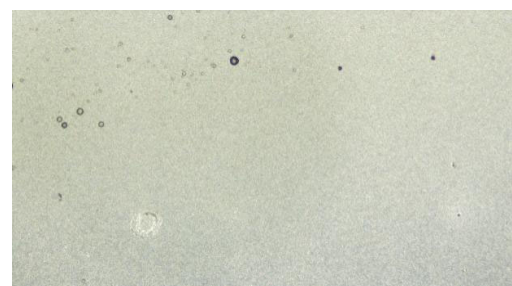
The experiment compared the gallium-nickel alloy electroplating effect of the DC electroplating platform and the AC electroplating platform. During the experiment, the electroplating conditions such as current density, solution temperature, solution pH, and anode-cathode distance were all controlled at the most suitable parameters and the control was consistent. Only the anode-cathode power supply method was different. The following conclusions were drawn:

①DC electroplating: Use a DC power supply to power the anode, the cathode is grounded, and the DC power supply is output in a steady current mode. The cathode current density can be controlled according to the size of the cathode surface area. The DC electroplating efficiency is high. After 6.5 h of electroplating, a gallium-nickel alloy layer of about 80 mg/cm<sup>2</sup> is obtained on the cathode surface. The coating thickness is about 80-90 μm, and the electroplating efficiency is as high as 37.65%. However, DC plating will leave a large number of tiny bubbles on the surface of the coating as shown in Figure 7, and the quality of the coating surface needs to be improved.



**Figure 7.** The coating surface obtained by the DC electroplating platform (130x microscope)

②AC plating: The AC plating platform was used for electroplating under the same conditions. After 6.5 h of electroplating, a gallium-nickel alloy layer of about 50 mg/cm<sup>2</sup> was obtained on the cathode surface. The coating thickness was about 50-60 μm, and the plating efficiency could reach 23.58%. The overall electroplating efficiency can reach about 60% of DC electroplating. If the output frequency of AC power and the proportion of positively bias are increased, greater electroplating efficiency can be achieved. Under the existing conditions, the gallium-nickel alloy coating obtained by the AC plating platform is shown in Figure 8. Compared with the coating obtained by the DC plating platform shown in Figure 7, the gallium-nickel alloy layer obtained by AC plating rarely has bubbles on its surface, which means that it has better density.



**Figure 8.** The coating surface obtained by the AC plating platform (130 times microscope)

## 4. Conclusion

Based on the above electroplating experimental results, it is believed that the use of an AC electroplating platform can obtain gallium-nickel alloy coatings with the best surface quality under a wide range of electroplating conditions, such as a solution temperature below 80°C, a current density of 25-50 mA/cm<sup>2</sup>, and a solution pH of 1.75-2.25. The electroplating efficiency of the AC electroplating platform built in this article can reach about 60% of that of the DC electroplating platform under the same current density. The electroplating efficiency can be further increased by increasing the AC frequency and the forward bias ratio. At the same time, the quality of the gallium-nickel alloy coating obtained by AC electroplating is better than that of the coating obtained by DC electroplating under the same conditions. In summary, the AC electroplating platform used in this paper can electroplate gallium-nickel alloy targets with good surface quality, which is feasible for subsequent irradiation experiments.

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