

Contribution to the Study of an Electric Copper —Tin Cell Using Sulphuric Acid as Electrolyte. Effect of the Acid Concentration

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How to cite this paper: Touré, S. (2026)
Contribution to the Study of an Electric
Copper. *Open Journal of Applied Sciences*,
16, 1006-1015.
<https://doi.org/10.4236/ojapps.2026.163059>

Received: February 7, 2026

Accepted: March 28, 2026

Published: March 31, 2026

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Abstract

In literature, some cells using Sn and Cu are described, but they usually use tin sulphate and copper sulphate as electrolytes. The concern of the present study is an electric copper-tin cell using sulphuric acid as electrolyser. The two electrodes, Cu (positive electrode) and Sn (negative one) are dipped in the sulphuric acid solution. Several cells were made, with different concentrations of the acid, in purified water. For each cell, the electromotive force E and the intensity I were measured. The output electric power P was calculated. The plotting of E against I and P against I showed the influence of the concentration on the curves. Then some selected intervals of E were chosen. For each interval, the average value of E was calculated. In addition, the average values of the corresponding I values of the intensity I were calculated. Those average values were used to make new plottings which showed less influence of the acid concentration on the curves. From those curves, the internal resistance r and the potential difference V of the cell at room temperature were determined. The experiments showed the presence of white tin sulphate and blue copper sulphate as deposits on the electrodes. Reactions occurring at the electrodes were discussed, including the hydrogen evolution reaction steps. Tin sulphate is interesting because it can be used as anode material for lithium secondary battery. Moreover, it can yield tin dioxide which is an n-type direct gap semiconductor, with an interesting band gap, and can be used as a material for photovoltaic conversion.

Keywords

Electric Copper-Tin Cell, Tin Sulphate, Hydrogen Evolution Reaction,
Electromotive Force, Photovoltaic Conversion

1. Introduction

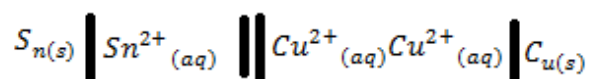
The world is facing today several challenges regarding the energy problem. As a

matter of fact, the world's population is always growing, resulting in the increasing of energy consumption. Such situation leads to environment pollution, greenhouse effect gas emissions and climate warming. Most of those damages are due to the fossil sources of energy. Hence new sources of energy are looked for that may allow the energy transition.

Several solutions are looked for, such as solar energy, wind energy, hydrogen energy, biomass energy, geothermal energy, etc. The development of electrical cells and batteries is also one of the ways that are looked for to get clean and sustainable energy.

Electrochemical cells are widely studied and developed, since the first electrical cell developed by Volta in 1799 [1] [2].

In literature, some cells using Sn and Cu are described, but they usually use tin sulfate and copper sulfate as electrolytes. Their chain is indicated below.



The structure of such cells is given in **Figure 1** [3].

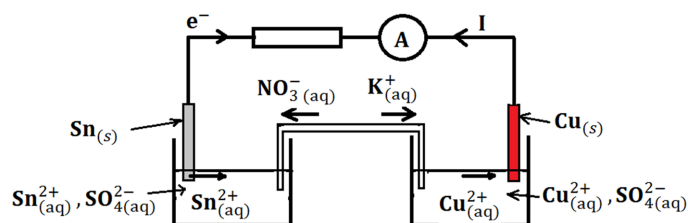


Figure 1. Structure of a classical Cu-Sn cell.

An alkaline bridge links the two electrolytes.

The standard electrochemical potential of this Cu-Sn cell is $E^0 = 0.48 \text{ V}$ [4].

The concern of the present study is an electric copper-tin cell using sulphuric acid as electrolyte.

2. Materials and Methods

Electrical cells were built, using sulphuric acid solution in water as electrolyte. The two electrodes are copper for the positive electrode and tin for the negative electrode. The structure of the cell is shown below in **Figure 2**.

A glass jar is used as a container of the sulphuric acid and the electrodes. Those electrodes are introduced in the jar through two insulated holes which are made in the cover of the jar opening. Insulated copper wires are used for the link between the electrodes and the multimeter.

The details about the construction of the cells are as follows:

- the electrode exposed area is 4 cm^2 .
- the electrode spacing is 3 cm.
- the immersion depth is 6 cm.

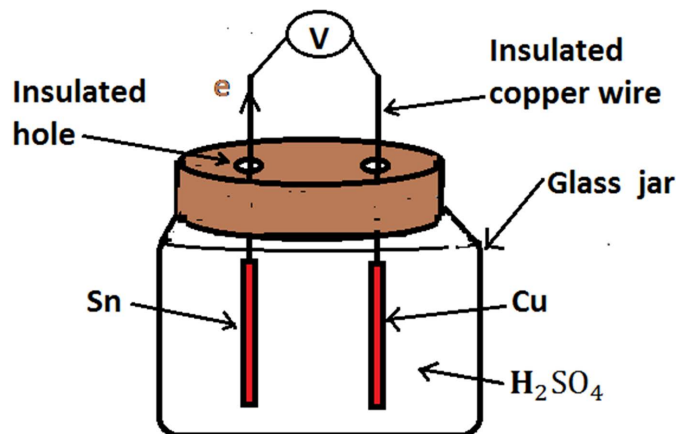


Figure 2. Structure of the copper-tin cell.

It should be noted that the solutions were not stirred.

The acid concentration varied by using various amounts of demineralized water, starting from a 28° sulphuric acid.

63 cells were assembled. The concentrations lie within the range 0.9 - 1.2 Kg/L.

A Metrix MX 430 multimeter was used for the electromotive and current intensity measurements.

The measurement uncertainty for the electromotive force and the current is 1.3%.

The electromotive and current intensity were measured in open-circuit condition. An external resistor would have an influence on the relation between the electromotive and the current.

The electromotive force (e.m.f) is theoretically linked to the concentration of ions. The potential difference E is of course linked to the Gibbs free energy by the following formula

$$\Delta G = -nFE \quad (1)$$

Under standard conditions, Equation (1) becomes

$$\Delta G^0 = -nFE^0 \quad (2)$$

It is also known that, at temperature T ,

$$\Delta G = \Delta G^0 + RT \ln Q \quad (3)$$

where Q is the reaction quotient.

Combining Equations (1)-(3), one gets the Nernst equation

$$E = E^0 - \frac{RT}{nF} \ln Q \quad (4)$$

By introducing C_{ox} the concentration of the oxidised ion and C_{red} the concentration of the reduced ion, E is expressed as

$$E = E^0 + \frac{RT}{nF} \ln \frac{C_{ox}}{C_{red}} \quad (5)$$

Equation (5) shows that the potential varies with the ions concentration.

One of the concerns of the present study was to see experimentally the effect of the acid concentration on the graph showing the variation of E with the current intensity, $E = f(I)$, and how the effect can be corrected.

It was also important to know the effect of the acid concentration on the variation of the output electric power with the current intensity.

The e.m.f. between the electrodes is a consequence of the redox reactions occurring in the cell.

The proposed electrode processes are the following. It is worth noting that in reference [3], it is indicated that, in sulphate medium, tin is oxidised.

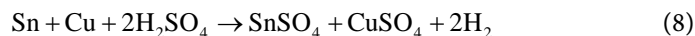
Here the anode is the tin electrode. At this electrode tin Sn is oxidised and SO_4^{2-} ion is reduced



At the cathode, which is the copper electrode, the proton H^+ is reduced



The overall cell reaction based on Equation (6) and Equation (7) is therefore



The reactions show a decrease in the acid concentration and also deposits of tin sulphate and copper sulphate. In addition, there is a hydrogen release.

But it should be noted that other reactions may occur as side reactions, such as hydrogen evolution reactions (HER) [5]. The mechanism of HER in acid media proceeds via three main steps.

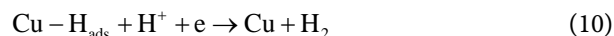
The mechanism occurs at the cathode surface which is here Cu. Two mechanisms may occur:

1-The Volmer-Heyrovsky mechanism with two steps which are:

- Volmer step (proton adsorption)

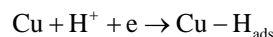


- Heyrovsky step (electrochemical desorption)



2-The Vomer-Tafel mechanism with also two steps:

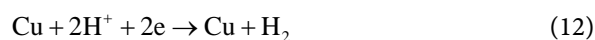
- Volmer step (proton adsorption)



- Tafel step (recombination)



Hence, HER leads to a hydrogen release and the overall reaction for the HER mechanism is



It is worth noting that Density Functional Theory calculations are often carried out for the three elementary steps of the HER [6]. For that, calculations of the binding free energy ΔG_H are performed

3. Results and Discussions

3.1. Electromotive Force, Potential, Intensity, Internal Resistance and Allowable Electric Power of the Cell

From the experimental values obtained, some plottings were made. The first one was the plotting of the e.m.f. E against the current intensity, as shown in **Figure 3**.

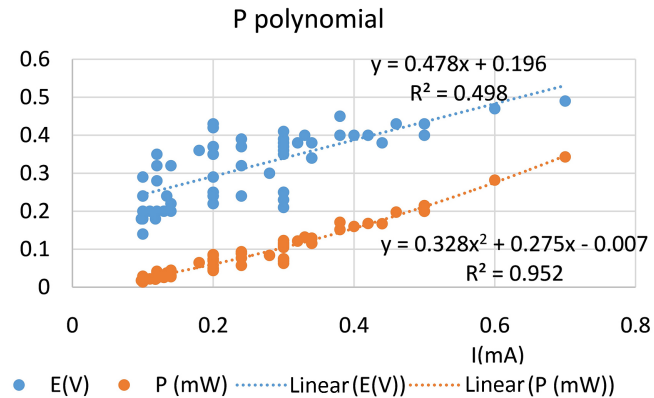


Figure 3. Plot of electromotive force E and the electrical power output P against the current intensity I in the case of a polynomial correlation for P .

It is worth noting that the theoretical variation of E with the current intensity I is given by the following Equation (13), because the measurements were made without any external resistor

$$E = V + rI \quad (13)$$

V is the potential difference between the devices; r is the internal resistance of the cell.

The linear correlation obtained from the plotting of E against the intensity I is

$$E = 0.1963 + 0.4781 \cdot I \quad (14)$$

The figure shows a great dispersion of the experimental points around the regression curve, resulting in the following not very high coefficient of determination.

$$R^2 = 0.4986.$$

This dispersion is due to the variation of the concentration from one point to another. From Equation (14), one gets

$$V = 0.1963 \text{ V and } r = 0.4781 \text{ k}\Omega.$$

Moreover, from Equation (13), one gets

$$EI = VI + rI^2 \quad (15)$$

$P = EI$ is the allowable electric power, *i.e.*, the maximum output power delivered by the device. VI is the useful power delivered by the cell.

Figure 3 also shows the plotting of P against the intensity I . Equation (15) suggests a polynomial correlation.

The equation of the polynomial correlation is:

$$P = 0.3287 \cdot I^2 + 0.2751 \cdot I - 0.0078 \quad (16)$$

The intercept, which is very weak, can be neglected, so that, according to Equation (15), an average value of the potential difference is $V = 0.2751$ V.

It should be noted that the polynomial correlation of P against I shows a very small dispersion of the experimental points around the regression curve, resulting in the following high coefficient of determination $R^2 = 0.9523$.

This high value of R^2 shows that the variation of the acid concentration has few effect on the output power P .

In a second step, we looked for a way to reduce the influence of the acid concentration on $E = E(I)$. For that, ten Intervals of E were selected, with their corresponding ten intervals of the intensity I . The ten intervals are the followings:

$0.1 < I < 0.13$; $0.14 < I < 0.20$; $0.21 < I < 0.24$; $0.25 < I < 0.30$; $0.31 < I < 0.35$; $0.36 < I < 0.40$; $0.41 < I < 0.45$; $0.46 < I < 0.50$; $0.51 < I < 0.60$; $0.66 < I < 0.70$.

For each interval, the average values of I and E were calculated. **Table 1** gives those values.

Table 1. Average values of the intensity, the e.m.f. and the power in the selected intervals.

Average values of intensity I (mA)	Average values of e.m.f E (V)	Allowable electric power P (mW)
0.1177	0.229	0.02695
0.178	0.315	0.05
0.24	0.338	0.08112
0.29846	0.31615	0.09436
0.3325	0.375	0.12468
0.38667	0.41667	0.1611
0.43	0.39	0.1677
0.48667	0.421	0.2049
0.6	0.47	0.282
0.7	0.49	0.346

New plottings were made using the values given in **Table 1**, as shown in **Figure 4**.

The plotting of E against the current intensity I was first considered.

The linear correlation obtained from the plotting of E against the intensity I is

$$E = 0.2216 + 0.4098 \cdot I \quad (18)$$

Here, the figure shows few dispersion of the experimental points around the regression curve, resulting in a very high coefficient of determination which is

$R^2 = 0.9068$. The values of V and r obtained from Equation (13), are $V = 0.2216$ V and $r = 0.4098$ k Ω .

The averages of the differences ΔE before and after averaging the values of I and E , using Equation (14) in the non-averaging case and Equation (18) after averaging, are calculated. Four values of the current intensity I were selected.

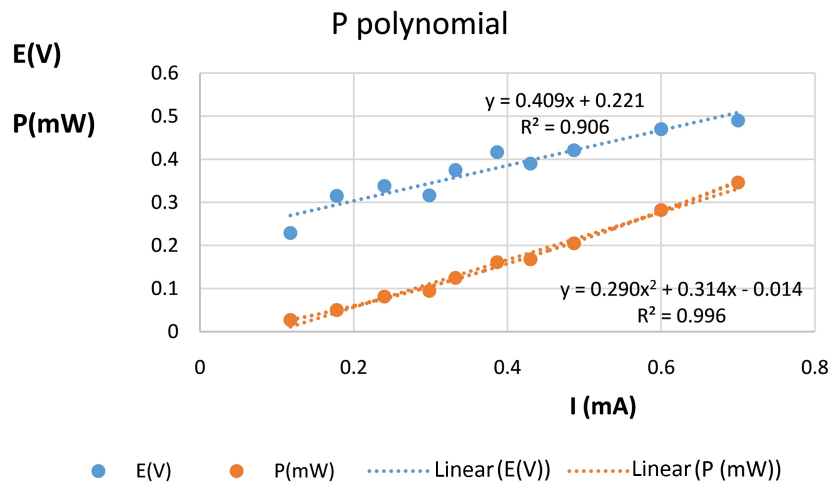


Figure 4. Plot of electromotive force E and the electrical power output P against the current intensity I using average values of I and E .

In the non-averaging case, for the given value of the current I , the e.m.f. E is calculated by using Equation (14). The differences between this value and the E values contributing to the selected interval are calculated then averaged to get ΔE_{na} .

Then, for the same current, Equation (18) is used to get the e.m.f. E calculated by using Equation (18). The difference ΔE_{av} between this calculated value and the value given in **Table 1** is calculated.

ΔE_{na} and ΔE_{av} are quantities that allow the estimation of the dispersion of the experimental points around the regression linear curves.

The results of those calculations are summarized in **Table 2**.

Table 2. Average differences ΔE before and after averaging the values of I and E .

Current intensities I (mA)	Average differences in the non-averaging case ΔE_{na} (V)	Difference after averaging ΔE_{av} (V)
0.24	0.072	0.018
0.298	0.064	0.027
0.43	0.012	0.008
0.487	0.0103	0.000

Table 2 shows that differences are very small after averaging, if compared with those obtained in the non-averaging case.

Hence, the technique used, consisting in averaging the E and I values into selected intervals, reduces highly the influence the concentration variation on the variation of the e.m.f. E with the current intensity, by reducing greatly the dispersion of the experimental points around the regression linear curve.

Then the plotting of the maximum output power P against the current intensity I was considered. The following polynomial correlation is found

$$P = 0.2901 \cdot I^2 + 0.3143 \cdot I - 0.0146 \quad (14)$$

Here again, the intercept, which is very weak can be neglected, so that, according to Equation (10), an average value of the potential difference is $V = 0.3143$ V and the average value of the internal resistance is $r = 0.2901$ k Ω .

We note again that the polynomial correlation of P against I shows a very small dispersion of the experimental points around the regression curve, resulting in the following very high coefficient of determination $R^2 = 0.9966$.

3.2. Discharge Behaviour of the Cell

A study of the discharge of one of the cells was carried out during five hours. **Figure 5** shows the evolution of the e.m.f. with time.

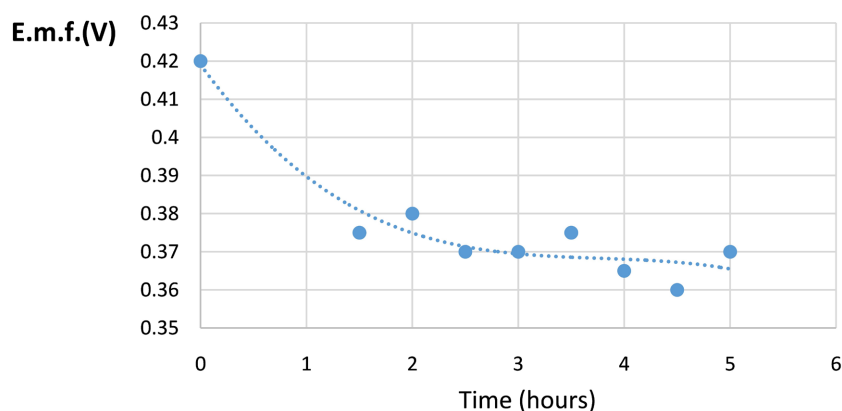


Figure 5. Evolution of the e.m.f., with time.

After a short transient period characterized by a decrease of the e.m.f. during about 1.5 hours, a rather constant phase is observed. Hence, the voltage is rather stable over the period of experiment, after the transient period.

The average value of the e.m.f. during the constant phase is 0.368 ± 0.004 V.

The same discharge study was made regarding the current intensity I . **Figure 6** shows the evolution of the intensity with time.

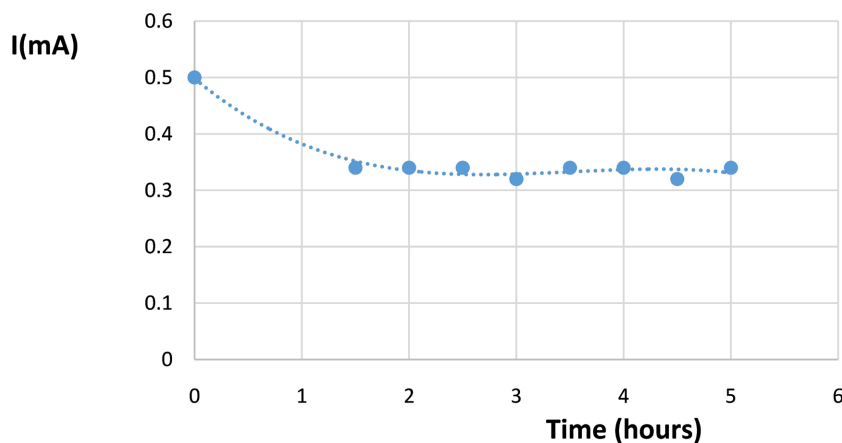


Figure 6. Evolution of the current intensity with time.

It is also observed that after a short transient phase characterized by a decrease of the intensity during about 1.5 hours, a constant phase is observed during which, the intensity is stable. The average value of the current intensity during the constant phase is 0.335 ± 0.007 mA.

3.3. Deposits on the Electrodes

The experiments showed deposits of white tin sulphate SnSO_4 at the anode and blue CuSO_4 at the cathode as consequences of Equation (6) and Equation (7).

Let SnSO_4 be considered. Tin sulphate is interesting because it can be used as anode material for lithium battery [7]. It is also interesting because at 378°C , it is decomposed into SnO_2 and SO_2 . This tin dioxide SnO_2 is an n-type direct gap semiconductor, which may be polycrystalline or amorphous. The crystalline SnO_2 has a band gap which lies in the range 2.85 - 3.3 eV [8].

4. Conclusions and Perspectives

The electric cell studied here is interesting for electricity production. It is also interesting because one of the products of the redox reactions at the cathode is hydrogen. The release of hydrogen is of course interesting because this gas can be used in PEM Fuel Cells to produce electricity. The electrochemical processes occurring at the electrodes are investigated, including the HER different steps. Further investigations can be performed by making Density Functional Theory calculations to assess the HER activity. The plotting of the electromotive force E and the electrical power output P against the current intensity showed a great influence of the acid concentration on E . This influence is weak for the electrical power output. We used a technique that reduces highly the influence of the concentration variation on the e.m.f. E . The average value of the potential difference is $V = 0.3143$ V and the average value of the internal resistance is $r = 0.2901$ k Ω . Moreover, a stable behaviour of the electrical cell is observed, after a short transient period. Apart from the hydrogen release, some deposits are observed, namely tin sulphate SnSO_4 and copper sulphate CuSO_4 . One of the deposits, SnSO_4 , is interesting for battery production and can yield SnO_2 which is useful for photovoltaic conversion of solar energy.

Further investigations are to be carried out, in order to optimise the allowable output power of the cell. The influences of the electrode spacing, the electrode exposed area, and the immersion depth are to be investigated.

All the experiments have been made at room temperature. Therefore, it would be relevant to investigate the influence of temperature on the cell electrical output.

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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