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Investigating the Performance of Rechargeable Zinc-Air Fuel Cell

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Abstract

Zinc-air fuel cells (ZAFCs) are a promising energy source that could compete with lithium-ion batteries and perhaps proton-exchange membrane fuel cells (PEMFCs) for next-generation electrified transportation and energy storage applications. In the present work, a flow-type ZAFC with mechanical rechargeable was adopted, combined with an auxiliary cell (electrolyzer) for zinc renewal and electrolyte recharge to the main cell. In this work a practical study was performed to calculate the cell capacity (Ah), as well as study the electrolysis cell efficiency by current efficiency, and study the effective parameters that have an influence on cell performance such as space velocity and current density. The best parameters were selected to obtain the best performance for cell operation. The obtained cell capacity was 2.4Ah. The best performance of the electrolyzer was obtained with 0.6min^{-1} space velocity. At the same time, the best performance of the electrolyzer was when the value of the current density was 200A/m^2

Keywords: Fuel cell, zinc-air fuel cell.

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1- Introduction

Zinc Air Fuel Cell (ZAFC) could be a promising alternative energy source for secondary batteries, Proton exchange membrane fuel cell (PEMFC), or direct methanol fuel cell (DMFC) due to its high specific energy, low cost of zinc metal and air fuels, and environmentally friendly by-products [1,2]. Furthermore, it may employ low-cost metal oxides as catalysts and avoids the problems of storing and compressing explosive or volatile fuels.[3] The ZAFC is based on simple chemistry, with zinc (Zn) serving as a fuel and ambient air serving as an oxidant, respectively, and an aqueous alkaline solution serving as an electrolyte[4,5]. Zinc is converted to a Zn^{2+} cation after losing electrons to an anode current collector[6]. The electrons created on the anode go through an external circuit to an air cathode, where oxygen is reduced and hydroxyl ions (OH^-) are formed. The hydroxyl ions migrate through the electrolyte to the anode side, where they react with zinc cations to generate zinc oxides. The following is a summary of the chemical reactions that occur in a zinc air system [7].
Anode reactions:

$$Zn + 4OH^- \leftrightarrow Zn(OH)4^{2-} + 2e^- \quad (E_0 = -1.25V) \quad (1)$$

$$Zn(OH)4^{2-} \leftrightarrow ZnO + 2\,OH^- + H_2O \quad (2)$$

Cathode reactions:

$$\tfrac{1}{2}O_2 + H_2O + 2e^- \leftrightarrow 2OH^- \quad (E_0 = 0.4V) \quad (3)$$

Therefore, the overall reaction can be expresses as:

$$2Zn + O_2 \leftrightarrow 2ZnO \quad (E_0 = 1.65V) \quad (4)$$

A zinc-air fuel cell (ZAFC) can be recharged in two ways, mechanical and electrical recharge [8]. An electrically rechargeable Zn-air cell is recharged by supplying electricity directly to the cell [9]. During recharge, oxygen is generated at the air electrode whilst Zn metal is electrochemically regenerated at the Zn electrode. The production of dendritic Zn during recharge is a key issue with the Zn electrode. This is a severe problem in the Zn electrode because it causes direct contact between the cathode and the anode, stopping the cell. Moreover, it is hard to find necessary bifunctional catalysts for air cathode to support both oxygen reduction reaction (ORR) and oxygen evolution reaction (OER). Furthermore, during recharging, the cathode rapidly deteriorates due to the development of oxygen bubbles and air electrode corrosion [10,11].

There are two basic types of mechanically rechargeable Zinc-air fuel cells (ZAFCs): (1) ZAFCs that can be refueled. (2) reconstructable ZAFCs. After the discharge process, the Zn plates are replaced with new ones in the reconstructable type. In refuelable types, a flowing electrolyte system is used to regenerate or precipitate zinc particles through a co-cell (electrolyzer) and is returned to the main cell again as well as regeneration of the electrolyte [12]. In a ZAFC, the anode type (fuel) is a major component, and instead of Zn plates, Zn particles can be used as a fuel source in a flow mode. In recent decades, porous Zn plates have been used commercially as anodes for ZAFCs.

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The porous structure has a high active surface ratio, which improves reaction efficiency by increasing electrolyte molecule encounters [13]. The Zn particles have a greater surface area than a Zn plate, allowing for improved contact between the active metal and the electrolyte [14]. As a result, compared to Zn plates, Zn particles have the following advantages: (i) Zn particles have a larger surface area for reaction, which is desired; (ii) The particle anode with flowing electrolyte is very important because it may prevent the hydrogen evolution process (HER) and so improve cell performance and power output [15].To keep the electrochemical processes running and create electricity, fuel cells require a constant supply of fuel.

The wasted Zn anodes will be removed using a circulating flow type electrolyte system, while Zn fuel may be delivered to the reaction tank on a regular basis. As a result, without mechanical exchange, the reactant discharge can still be successfully re-supplied [1].

The rechargeable system of Zinc-Air Fuel Cell contains two separate electro-chemical systems, the first subsystem is Zinc-Air Fuel Cell conventional electricity generation system. The second subsystem is a zinc regeneration electrolytic cell or electrodeposition cell as shown in Fig. **1** [16].

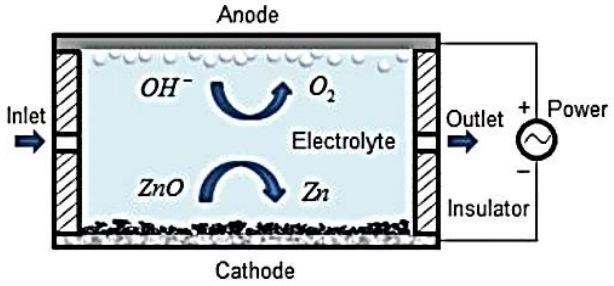


Fig. 1. Schematic view of zinc electrodeposition [16].

Zinc can be re-generated by several methods. However, Electrolysis or electrodeposition are the best procedures to utilize with Zn-air flow cells. The discharge products of the zinc-air fuel cell could be utilized directly in the electrolysis cell or electrolyzer as a reactant. An electrolyzer regenerates zinc from the discharge stream of Zn-air flow cells that contain zincate and zinc oxide. The fuel cell is then re-charged with the regenerated Zinc [11].

The charging current is provided to the zinc electrolysis cell (electrolyzer) incitement the inverse reactions of ZAFC. At the negative electrode of the electrolysis cell, as shown in equations (5),(6) Zinc is regenerated from the zincate solution resulting from the electrochemical processes that occurred during the discharge in the zinc-air fuel cell, whereas oxygen (O_2) will be formed at the positive electrode (anode) as shown in equation (7) [10]. The following are general electrolysis reactions:

At the cathode (negative electrode):

$$ZnO + 2\,OH^- + H_2O \leftrightarrow Zn(OH)4^{2-} \quad (5)$$

$$Zn\,(OH)_4^{2-} + 2e^- \leftrightarrow Zn + 4OH^- \quad (6)$$

At the anode (positive electrode):

$$2OH^- \leftrightarrow \frac{1}{2}\,O_2 + H_2O + 2e^- \quad (7)$$

General reaction

$$ZnO \leftrightarrow Zn + \frac{1}{2}\,O_2 \quad (8)$$

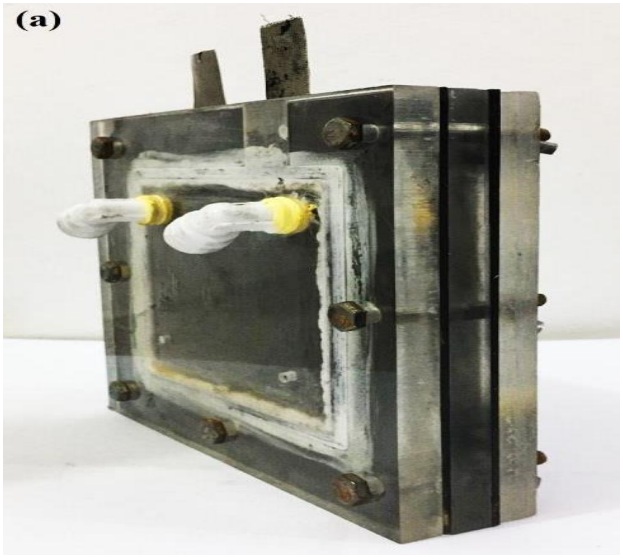
In this work, the ZAFC of a mechanical refuelling flow type was adopted, combined with a co-cell or electrolyzer for zinc regeneration and electrolyte recharge to the main cell. The operational conditions for zinc-air fuel cells were by choosing the best parameters obtained from our previous study, 40%wt. electrolyte concentration of KOH, 6 L/min airflow rate, 100 mL/min electrolyte circulation rate and 50°C electrolyte temperature [17].

A practical study was performed to study the electrolysis cell efficiency, and study some parameters that have an influence on cell performance such as space velocity and current intensity, the best parameters were selected to obtain the best performance for cell operation. After selecting the best parameters that improved the cell's performance, it is necessary to know the amount of energy produced by the cell.

2- Experimental Section

2.1. System Design

The configuration of a single Zinc-air fuel cell (ZAFC) is shown in Fig. **2**, it consists of three plates of perspex material as the mainframe because displays efficient chemical resistance against the corrosive electrolyte and oxidation and also facilitates frequent disassembling. Apertures were made on the back and front ends of the cell structure, respectively, and are used as electrolyte and air inlet and exhaust gates. Each Perspex plate had the following dimensions: 15*15*1cm (length, breadth, and thickness).



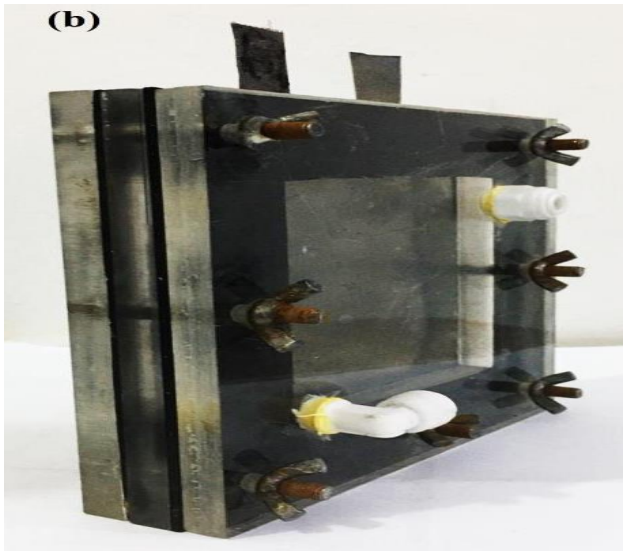


Fig. 2. Photograph of the Zinc-Air Fuel Cell (a) left side, (b) right side.

One of these plates opened from the inside with a size of (10*10 cm) to be electrolyte storage. The area (active area) of the air cathode was (100 cm^2), which was fixed to the left side of the Perspex plate. A non-woven fabric separator is placed between the anode and cathode to purpose as a separator to separate the cathode and anode. The air electrode was fixed to the left plate by an epoxy resin material. However, a rubber gasket has been positioned between the acrylic plates to prevent the electrolyte from leakage. Seven bolts secure all sides of the perspex plates, holding the cell together.

100 g Zinc particles with a diameter of about 0.5mm were placed within a 100-mesh stainless steel pouch that functioned as a current collector for the Zn anode. The current collector has a 100 cm^2 surface area.

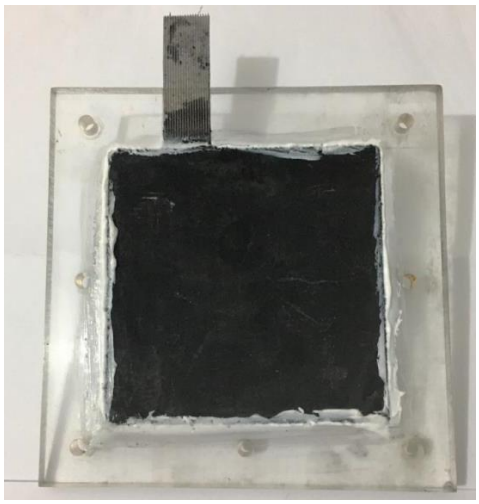


Fig. 3. Photograph of the air cathode plate fixed to the Perspex plates

The electrolyte is potassium hydroxide (KOH). It was prepared (electrolyte concentration of 40%wt.) by dissolving potassium hydroxide (Powder) in distilled water with continuous stirring for (5) minutes.

The cathode (air cathode) plate as shown in Fig. **3** contained three layers: a catalyst layer, a gas diffusion layer and cathode current collector. 100-mesh stainless steel was working as a current collector of the cathode.

A slurry combination of 7 g carbon black and 7 g polytetrafluoroethylene (PTFE) was cast in binder dissolved solvent to create the gas diffusion layer. The solvent was prepared by dissolving 3 g poly-styrene co-butadiene as a binder in 50 ml toluene and leaving it to dissolve completely for 24 hours.

The slurry mixture coated mesh stainless steel was then heat-pressed at 110 ◦C for 15min using a manual press tool (employing two steel layers as a press means) in an oven.

On the other side of the mesh stainless steel, the catalyst layer was created by dissolving 3 g MnO_2 and 7 g carbon black in the binder dissolved solvent. The solvent was prepared by dissolving 3 g poly styrene-co-butadiene as a binder in 50ml toluene and leaving it to dissolve completely for 24 hours. The catalyst layered cathode was heat-pressed at 110°C for 15min using a manual press tool in an oven.

The body structure of the electrolyzer is similar to a zinc-air fuel cell in the design. As shown in Fig.4 it consists of 4 plates of perspex plate with dimensions 15*15*1cm for each perspex plate, 2 of these plates opened from the inside with a size of (10*10 cm) to be electrolyte storage.

Also, apertures were formed on one side of the cell, these apertures are considered as gates to inlet spent electrolyte and outlet the fresh electrolyte. The electrolyzer included a stack arrangement with a cathode plate, and anode. The anode plate is made from stainless steel 100-mesh (0.149 mm) with an active area of 100 cm2. The cathode was made of a zinc plate with an area of 100 cm^2. Seven bolts secure all sides of the perspex plates, holding the cell together. The zinc-air fuel cell (ZAFC) is integrated with the electrolyzer as a complete system as a rechargeable zinc-air fuel cell (ZAFC). A regenerative fuel cell system requires on-site zinc fuel regeneration following the discharge cycle, which sets it apart from other systems that do not.

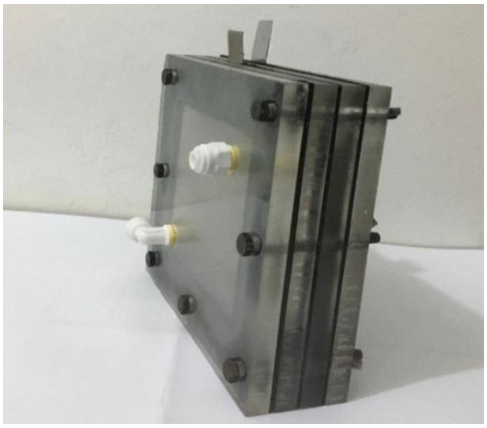


Fig. 4. Photograph of the Zinc regeneration electrolytic cell (electrolyzer)

As shown in Fig.5, mechanically rechargeable Zinc-air fuel cell this system contains a zinc-air fuel cell that is hydraulically connected to the zinc regeneration cell. Where the spent electrolyte comes out of the fuel cell and enters the regeneration cell. The electrolyzer precipitates zinc and reactivates the spent electrolyte. The fresh electrolyte leaves the regeneration cell and again it enters the zinc-air fuel cell to continue the cell's reactions and the transfer of electrons, thus generating electricity. A pump circulates the electrolyte between the cell and the additional electrolytic cell (electrolyzer), while flow meters monitor and control the flow.

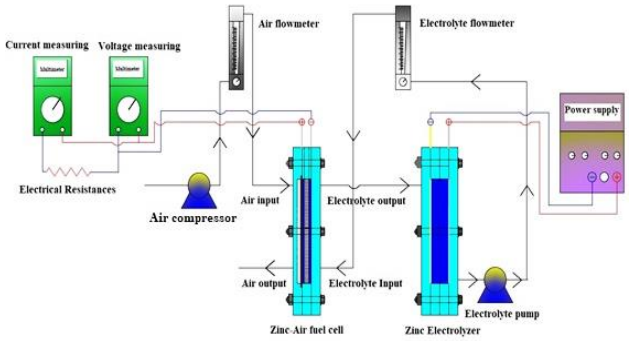


Fig. 5. Schematic diagram of Zinc-air fuel cell combined with zinc electrolysis cell (electrolyzer)

Air compressor was used to provide the cell with air. Airflow meters were used to monitor and control the air entering the cell. The system is also equipped with two digital multimeters, one of which is connected to the zinc-air fuel cell electrodes to monitoring and record the voltage readings of the cell, and the second is connected between the load and one of the cell electrodes to monitor and record the current. The system is also equipped with a DC power supply to supply the electrolysis cell with the energy needed to regenerate zinc and electrolyte.

2.2. Testing Methods

a. Fuel Cell Capacity Measurement

The purpose of this test is to check the capacity of the zinc-air fuel cell to produce power. The capacity of the battery is defined as the ability of the battery to store electrical energy, or it can be defined as the amount of electric current that the battery can deliver in a certain period of time, and it is expressed in units of Ampere hours (Ah), for example, a battery with a 2Ah theoretically gives a current of 1 ampere for 2 hours or a current of 2 amperes for one hour. Battery capacity can also be defined as the amount of electricity a battery gives off until its voltage drops rapidly. The operational conditions for this experiment were by choosing the best parameters obtained from our previous study [17], 40%wt. electrolyte concentration of KOH, 6 L/min airflow rate, 150 mL/min electrolyte circulation rate and the electrolyte temperature at 26°C.

The method of the test was by connecting the cell electrodes to the battery tester. The test device contains a constant load (electrical resistance) with a value of 5Ω, after adjusting the minimum voltage then pressing the "OK " button to start testing, the device automatically discharges the cell and after the test is started, the tester will display on the device screen the cell data, current, voltage, and fuel cell capacity, ampere-hours (Ah), until it reaches the minimum set voltage. After reaching the minimum voltage of the cell, the cell discharge will stop and the capacity of the cell will appear on the screen in units of (Ah), which now displays the actual capacity of the zinc-air fuel cell is discharging capacity.

b. The Space Velocity Effect on Current Efficiency for Zinc Regeneration

One of these parameters that are expected to have an impact on the performance of the zinc electrolysis cell (electrolyzer) is the space velocity. In chemical engineering, space velocity is defined as the ratio of the reactant's entering volumetric flow rate to the reactor volume, which shows how many reactor volumes of feed may be handled in a given length of time. the Space velocity of the electrolyte inside the electrolysis cell was examined. Six different electrolyte flow rates (0, 0.2, 0.4, 0.6, 0.8 and 1 min^{-1}) were tested to study their effect on current efficiency (CE) for zinc regeneration. At each flow rate, the cell will be discharged for two hours and then charged for one hour using a charging current of 500mA. The weight of the plate was measured before discharge a zinc plate was used as the anode in the zinc-air fuel cell. The plate was measured again to determine the amount of reactive or dissolved zinc inside the electrolyte. A laboratory digital DC power supply was used to supply the electrolysis cell with the necessary current. Operational conditions for this experiment where the operating temperature was at room temperature, electrolyte concentration 40%wt. of KOH. The current efficiency was calculated by the equation (9) [8]:

$$\text{Current efficiency of electrolyzer} = \frac{nF(N_{Zn,f} - N_{Zn,0})}{i^{\text{cell}} A_{elec.Zn}\, tf} \quad (9)$$

Where: n is zinc valence which is 2, F is Faraday constant (96،485.3365 C/mol.), tf is the total operating time in s, $N_{Zn,0}$ is initial mole of Zn and $N_{Zn,f}$ is final mole of Zn, i^{cell} Current density, A/cm^2 , $A_{elec.\,Zn}$ is active surface area of Zn electrode, cm^2.

Space velocity can be expressed mathematically by the equation (10) [18]:

$$\text{Space velocity (SV)} = \frac{V_0}{V} \quad (10)$$

Where: Vo represents the volumetric flow rate of the electrolyte entering the electrolyzer and V represents the volume of the electrolyzer.

c. Effect of the Current Density on Current Efficiency for Zinc Regeneration

The current density (CD) has a significant impact on the performance of the electrolysis cell, so it is necessary to study this parameter and study the best conditions that give the best performance of the cell.

In this experiment, seven different values of current density (50, 100, 150, 200, 250, 300 and 350 A/m^2) were studied and examined the effect of each value on the current efficiency. The cell was discharged for two hours and then charged for one hour according to the charge current density.

The operation conditions for this experiment were (T=28°C) at room temperature, the electrolyte concentration was 40%wt. of KOH and the space velocity 0.6 min^{-1}. A zinc plate electrode was used in the zinc-air fuel cell during discharge; it is weighed before and after the discharge to know the amount of zinc decomposed.

In the regeneration cell, a zinc plate was used as the negative electrode and a stainless-steel mesh as the positive electrode. A laboratory digital DC power supply was used to supply the electrolysis cell with the necessary current.

3- Results and Discussions

a. Zinc-Air Fuel Cell Capacity Measurement

A battery tester device was used to automatically discharge the cell; the fuel cell was operated at room temperature (26 °C), and 40% wt. KOH concentration, 6 L/min airflow rate and 150 mL/min electrolyte circulation rate.

The cell examination continued for 26 hours. The cell voltage reading was taken every hour and the voltage readings were as shown in Fig. **6**.

After the cell reached the minimum voltage set in the device, the examination process was stopped and the device gave a reading of the cell capacity, which was 2.4 Ah.

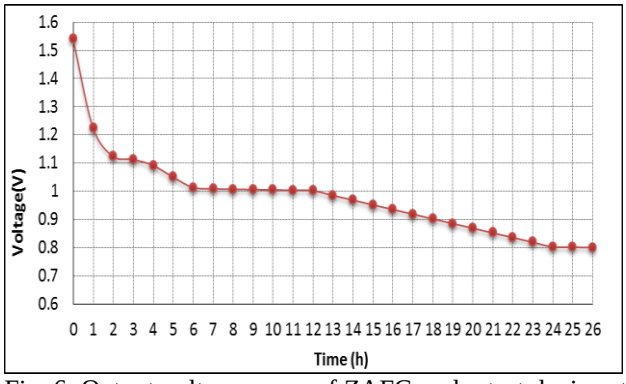


Fig. 6. Output voltage curve of ZAFC under test device at temperature (26 °C), and 40% wt. KOH concentration, 6 L/min airflow rate and 150 mL/min electrolyte circulation rate

3.2. Effect of the Space Velocity on Current Efficiency for Zinc Regeneration

After examining the parameters that have an impact on the performance of the zinc-air fuel cell, the parameters that have an effect on the performance of the electrolyzer must be examined. One of these parameters that is expected to have an impact on the performance of the electrolyzer is the space velocity. In chemical engineering, space velocity is defined as the ratio of the reactants' entering volumetric flow rate to the reactor volume, which determines how many reactor volumes of feed can be handled in a given length of time. The Space velocity of the electrolyte inside the electrolysis cell was examined.

Six different space velocity values (0, 0.2, 0.4, 0.6, 0.8 and 1 min$^{-1)}$ were tested to examine their effect on the performance of the electrolytic cell by calculating the current efficiency (CE). The operation conditions for this experiment were at room temperature (T=28°C) and the electrolyte concentration 40%wt. of KOH. At each value of Space velocity, the cell will be discharged for two hours and then charged for one hour using a charging current of 500mA.

The test results are shown in Fig.7. There was an increase in current efficiency from no flow state (stagnant) to flowing state at 0.2 min^{-1}velocity; the performance of the electrolyzer with flowing electrolyte was better than that of the electrolyzer with non-flowing electrolyte.

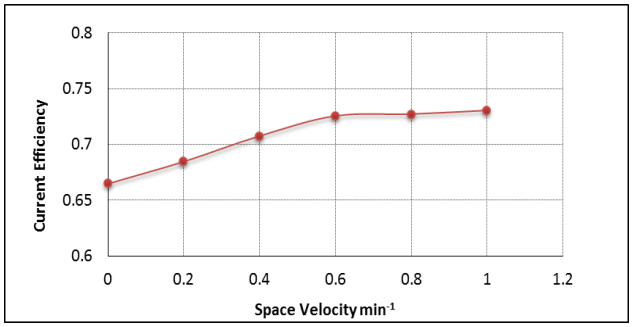


Fig. 7. Space velocity vs. Current efficiency the other parameters T=28°C and 40%wt. electrolyte concentration of KOH

When the space velocity was increased from 0.2 min^{-1} till 0.6 min^{-1} the current efficiency increased, after that, continuing to increase the space velocity from 0.6 min^{-1} to 1 min^{-1} did not lead to a significant increase in the performance of the electrolyze. Electrolyte hydrodynamics helps to increase ion diffusion and gas bubbles removed in electrolyze.

The high flow rate was favored because it lowered the thickness of the zincate ion's diffusion layer. As a result, Zn reduction was encouraged [8,19].

3.3. Effect of the Current Density on Current Efficiency for the Zinc Regeneration Process

The current density is an important parameter that has an impact on the zinc regeneration process. In this experiment, several values of current density were examined to find out its effect on the regeneration efficiency, which was expressed through current efficiency as shown in equation (9). Seven different values of current density (50, 100, 150, 200, 250, 300 and 350 A/m^2) were tested to examine the changes in current efficiency. The operation conditions for this experiment were at room temperature (T=28°C), the electrolyte concentration was 40%wt. of KOH and the space velocity 0.6 min^{-1}. The cell will be discharged for two hours and then charged for one hour according to the charge current density; the current efficiency was calculated as in Equation (9) was used to evaluate the Zn electrolyzer performance.

Fig. **8** shows the significant increase in the current efficiency, when the current density is increased from 50 to 200A/m^2, the current efficiency increased from 79% to 94.4%. After that, the increase in the current density did not lead to a significant improvement in the efficiency of the current, and the increase became useless, as when the current density was increased from 200 to 350 A/m^2, the efficiency of the current increased from 94.4 % to 96.1%, this increase is considered relatively slight for the amount of current applied. The internal resistance of the electrolyte leads to a decrease in the density of the current flowing through it and therefore the voltage decreases, this is referred to as the IR drop. These results agreed with [20].

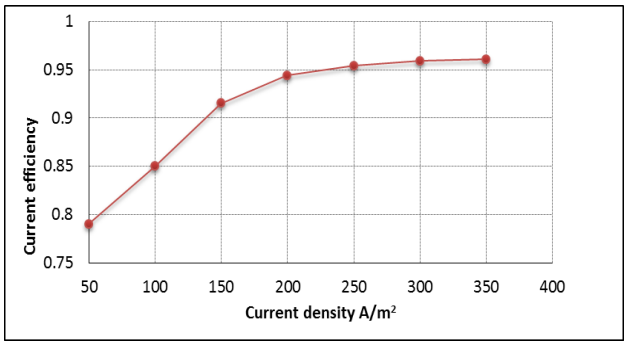


Fig. 8. Current density vs. Current efficiency the other parameters T=28°C and 40%wt. electrolyte concentration of KOH, space velocity 0.6 min^{-1}

4- Conclusions

The cell capacity test, as well as the parameters that affect the zinc regeneration process, were studied. The results and conclusions of this study were as follows:

1- After testing the capacity of the zinc-air fuel cell by a battery tester, the cell performance was good relatively to the test period of 26 hours, and the cell capacity can be determined by 2.4Ah.
2- There was an increase in current efficiency from state no flow conditions (stagnant) to flow state. The best performance of the electrolyzer was when the space velocity was 0.6 min^{-1}.
3- When the current density is increased from 50 to 200A/m^2, the current efficiency increased from 79% to 94.4%. The best performance of the electrolyzer when the current density value was 200A/m^2.

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دراسة كفاءة الاداء لخلية وقود الزنك – هواء القابلة لاعادة الشحن

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الخلاصة

تعتبر خلايا وقود الزنك والهواء (ZAFCs) مصدر طاقة واعدًا يمكن أن ينافس بطاريات الليثيوم أيون وربما خلايا وقود غشاء تبادل البروتون (PEMFCs) للجيل التالي من تطبيقات النقل وتخزين الطاقة. في هذا العمل ، تم اعتماد ZAFC من النوع المتدفق مع إمكانية إعادة الشحن ميكانيكيًا ، باستخدام خلية مشتركة (خلية التحلل الكهربائي) لتجديد الزنك وإعادة تجديد الالكتروليت واعادته إلى الخلية الرئيسية. في هذا العمل تم اجراء دراسة عملية لحساب سعة الخلية (Ah) ، وكذلك دراسة كفاءة خلية التحلل الكهربائي من حيث كفائة التيار، ودراسة بعض المتغيرات التي لها تأثير على أداء الخلية مثل ال(Space velocity) وكثافة التيار ، و اختيار افضل العوامل للحصول على أفضل أداء لتشغيل الخلية. كانت سعة الخلية التي تم الحصول عليها 2.4امبير–ساعة. كان أفضل أداء لخلية التحلل الكهربائي عندما كانت ال 0.6 (Space velocity) دقيقة $^{-1}$. أفضل أداء لخلية التحلل الكهربائي هو عندما تكون قيمة كثافة التيار 200امبير/م^2

الكلمات الدالة: خلايا الوقود, خلايا وقود الزنك–هواء