Electrochemical Conversion of CO_2 into Useful Chemicals and PKL Electricity

K. A. Khan, M. A. Mamun, Md. Israrul Adal, Sharif Mia, and M. Hazrat Ali

1 Introduction

The main theme should be the addressing global challenges in the new era through research, innovations, and strengthening connections (Hasan and Khan 2017, 2019, 2020). PKL electricity is produced mainly using Zn/Cu-based electrodes and PKL extract. The performances are gradually decreased within a few months (Hazrat Ali et al. 2019; Khan et al. 2016a, 2017). To remain sustain of this PKL electricity, several techniques have been developed like the use of nanoparticles, dynamic method, use of secondary salt, etc. The use of CO_2 is a new technique for PKL electrochemical cells (Khan et al. 2016b, c, 2018a, b). To keep it in mind, the electrochemical conversion of CO_2 into useful chemicals and PKL electricity has been designed and developed (Khan et al. 2019a, b). The PKL electricity is a new, renewable energy and innovative technique across the globe (Khan et al. 2019c). So that R&D work should be done regularly on PKL electricity. This research work will also be an excellent avenue for collaborative work for more innovative research for the uplifting of the community and industrial development. The nominal voltage of a galvanic cell is fixed by the electrochemical characteristics of the active chemicals used in the cell (Khan et al. 2020). The actual voltage generated at the cell terminals at a particular time depends on the load current and the internal impedance of the cell, and this also varies with the temperature, the state of charge, and with the age of the cell. A comparative study of the voltage of the normal voltaic/galvanic cell, traditional PKL electrochemical cell, and CO_2-based PKL electrochemical cell has also been conducted (Ruhane et al. 2017a). The PKL electrochemical cell depends on some parameters (Ruhane et al. 2017b) which have been discussed here. The production and the use of CO_2 is an

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innovative technology for an electrochemical cell or battery. The different parameters of the PKL electrochemical cells both with and without CO_2 have been studied.

2 Methodology

In this section, the methodology of CO_2 production has been discussed. The methodology of the preparation of PKL electrochemical cell has also been studied. Finally, the use of CO_2 for PKL Electricity has been designed and fabricated.

2.1 Experimental Procedure

The experimental setup of the CO_2 production device for practical utilization is shown (Fig. 1). The chemical reaction was $NaHCO_3 + HCl = NaCl + H_2O + CO_2$. The produced CO_2 goes out through a tube. Figure 1 also shows the collection of

Fig. 1 Experimental setup of CO_2 production device

Fig. 2 Experimental setup with a balloon for electrochemical conversion

CO_2 by a balloon. Figure 2 shows the utilization of CO_2 in a PKL electrochemical cell for getting more electricity for a long time duration.

2.2 *Application of CO_2 Gas into the PKL Electrochemical Cell: Preparation of CO_2 Gas*

About 2.0 g of powdered $NaHCO_3$ and 250 mL 0.1 M HCl solution were taken in a 500 mL conical flask. Then the mixture was heated very slowly, releasing CO_2 gas which was dried by passing through silica gel.

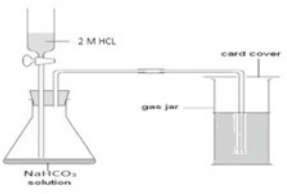
The methods of CO_2 gas preparation in the laboratory are shown in Fig. 3. Here, Fig. 3a shows the Schematic diagram of CO_2 gas preparation and Fig. 3b shows the Experimental setup of CO_2 gas preparation.

Instrumentation and Apparatus

Model CHI 660E electrochemical workstation, Graphite pencil electrode (GPE) as working electrode (Livo), GCE, modified GCE, carbon-based paper electrode and activated GPE electrode as working electrode, Pt wire auxiliary electrode, Ag/AgCl reference electrode, pH meter (Hanna), Digital Multimeter, Zn, Cu, Al, Ag, Fe electrodes, and Magnetic Stirrer.

Chemicals and Reagents.

Carbon dioxide (CO_2), Sodium bicarbonate solution ($NaHCO_3$), Hydrochloric acid (HCl), PBS solutions for various pH, Deionized water or double-distilled water, Graphite powder, Activated charcoal (Sigma Aldese), Nujol, Alumina powder, Silica gel, and Highly pure N_2 gas (Linde BD Ltd.).



(a): Schematic diagram of CO_2 gas preparation

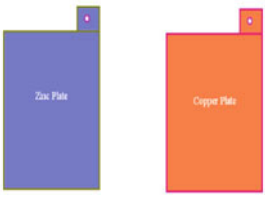
(b):Experimental set up of CO_2 gas preparation

Fig. 3 Methods of CO_2 gas preparation

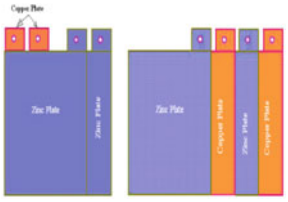
2.3 Zn and Cu Electrodes Preparation

To prepare a PKL (Pathor Kuchi Leaf) electrochemical cell, a cathode and an anode are needed. In our research article, it is shown that Zn (Zinc) plate is used as a cathode and the Cu (Copper) plate is used as an anode. The Zn and Cu plates are available in the local market. The price of the Zn plate is of low cost compared to the Cu plate. On the other hand, the longevity of the Cu plate is much more than the Zn plate. Moreover, the Zn plate is called a sacrificial element rather than the other metal plates. That is why the performance Zn/Cu-based PKL electrochemical cell is better than the other metals. The preparation technique of the Zn/Cu-based PKL electrochemical cell for practical utilization is shown in Fig. 4.

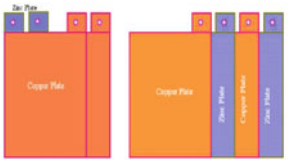
Figure 4a shows the Zn and Cu for a cell (Zn:Cu = 1:1). Figure 4b shows the keeping of Zn plate first (Zn:Cu = 2:2). Figure 4c shows the keeping of Cu plate



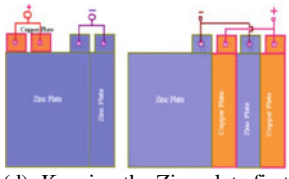
(a): Zn and Cu for a cell(1:1)



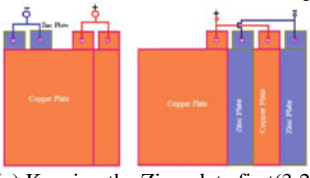
(b):Keeping Zn plate first (2:2)



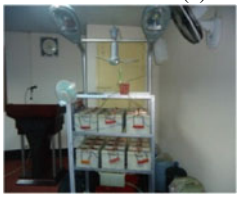
(c): Keeping Cu plate first(2:2)



(d): Keeping the Zinc plate first(3:2)



(e) Keeping the Zinc plate first(3:2)



(f): Design and fabrication of 1KW PKL Power

(g): Practical application of 1KW PKL Power

Fig. 4 Design and fabrication of Zn/Cu-based 1 KW PKL power for practical utilization

first (Zn:Cu = 2:2). Figure 4d shows the keeping of the Zinc plate first (Zn:Cu = 3:2). Figure 4e shows the keeping of the Zinc plate first (Zn:Cu = 3:23:2). Figure 4f shows the design and fabrication of 1 KW PKL Power and, finally, Fig. 4 g shows the Practical application of 1 KW PKL Power.

2.4 Chemical Reactions

Since Zn is a sacrificial element, we have

$$Zn = Zn^{2+} + 2e^- \tag{1}$$

$$2e^- + Cu^{2+} = Cu \tag{2}$$

Adding (1) + (2), $Zn + 2e^- + Cu^{2+} = Zn^{2+} + 2e^- + Cu$

$$\text{Or} \quad Zn + Cu^{2+} = Zn^{2+} + Cu \tag{3}$$

where Cu^{2+} = Reactant Ions and Zn^{2+} = Product Ions.

$$\text{Again we have} \quad Zn = Zn^{2+} + 2e^- \tag{4}$$

$$2e^- + H^+ = H = H_2 \tag{5}$$

Adding (4) + (5), $Zn + 2e^- + H^+ = Zn^{2+} + 2e^- + H_2$

$$\text{Or} \quad Zn + H^+ = Zn^{2+} + H_2 \tag{6}$$

where H^+ = Reactant Ions and Zn^{2+} = Product Ions.

$$NaHCO_3 + HCl = NaCl + H_2O + CO_2$$

Here, NaCl and H_2O are the useful chemicals and CO_2 is the conversion/production product of electricity.

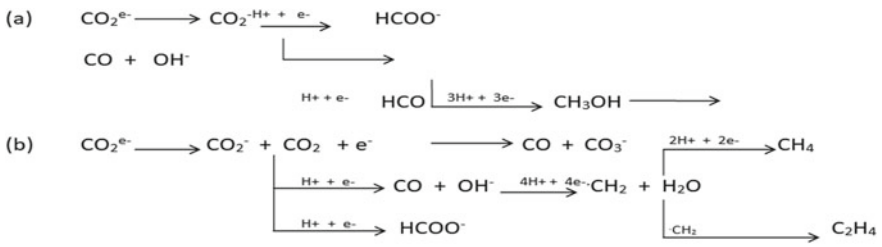
From Eq. (7), it is shown that the produced CO_2 can be used in the dead PKL electrochemical cell. The pH of the dead PKL electrochemical cell decreased than that of starting pH of the PKL electrochemical cell. When the produced CO_2 is applied to the dead PKL electrochemical cell, then the following chemical equation has occurred:

$$H^+ + CO_2 = H_2CO_3 \tag{8}$$

The HCO_3 will be produced in the dead PKL electrochemical cell. It is shown that the pH of the dead PKL electrochemical cell has been increased and then the dead PKL electrochemical cell has become reactive to generate electricity. In this case, H_2 gas will be produced as a byproduct.

2.5 Advantages of Electrochemical Reduction of CO_2

It is an easy process and the rate of electrochemical reduction of CO_2 is so much higher than the chemical reduction of CO_2. The advantages are as follows: (1) In presence of CO_2, it can easily convert into HCOOH (formic acid), CO (carbon monoxide), CH_3OH (methanol), $(COOH)_2$ (oxalic acid), C_2H_4 (ethylene), CH_4 (methane), etc. (which are useful products). (2) During the electrochemical conversion of CO_2 without aqueous catholyte (electrolyte), other's reagent is not necessary as the chemical conversion of CO_2 into the same product. (3) Higher temperature and pressure is not necessary as the chemical conversion of CO_2:



(4) In this process very low current (micro amp level) is needed to get the product. Because in this process, an electron is necessary (which is available here and obtained easily) for reduction. So this process is not expensive. (5) In this process, the reaction is carried out in the solution phase. Generally, the solution phase reaction is more easy than gas phase reaction.

2.6 Application of CO_2 Gas into the PKL Electrochemical Cell

During the preparation of the PKL electrochemical cell, the PKL extract was mixed with H_2O. In this case, O_2 was dissolved in water. So that O_2 should be neutralized before being applied to CO_2 gas into the PKL electrochemical cell.

3 Results and Discussion

3.1 Comparative Studies on Performance of Traditional Electrolytic Cells/Batteries

The variation of the voltages for five different typical electrolytic cells/batteries has been studied. It is also shown the comparison of the variation of the voltages with the variation of percentage of capacity discharged among the five different typical electrolytic cells/battery like Zn/MnO_2, Ni–Cd, Ni–Zn, Lead Acid, and Lithium-Ion battery.

Figure 5 shows that the variation of the voltages with the variation of percentage of capacity discharged for Zn/MnO_2 electrolytic cells/battery. The change of voltage was (1.50–1.10 V) 0.40 V up to 100% of Capacity Discharged. Figure 6 shows that the voltages vary with the percentage of capacity discharged for Ni–Cd electrolytic cells/batteries. The change of voltage was (1.48–1.15 V) 0.33 V up to 100% of capacity discharged.

Figure 7 shows that the voltages vary with the variation of percentage of capacity discharged for Ni–Zn electrolytic cells/batteries. The change of voltage was (1.60–1.55 V) 0.05 V up to 100% of Capacity Discharged. Figure 7 shows that the voltages vary with the percentage of capacity discharged for lead acid electrolytic cells/batteries. The change of voltage was (2.01–1.73 V) 0.24 V up to 100% of Capacity Discharged (Figs. 8 and 9).

Figure 7 shows that the voltages vary with the percentage of capacity discharged for lithium-ion electrolytic cells/batteries. The change of voltage was (4.10–3.80 V) 0.30 V up to 100% of Capacity Discharged.

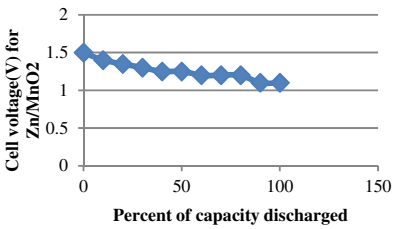


Fig. 5 Cell voltage (V) for Zn/MnO_2 with percentage capacity of discharged

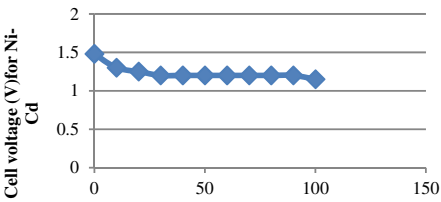


Fig. 6 Cell voltage for Ni–Cd with percentage capacity of discharged

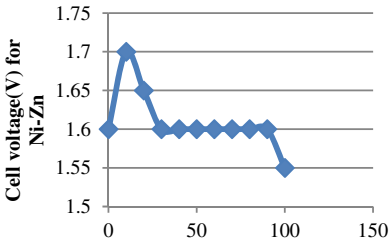


Fig. 7 Cell voltage (V) for Ni–Zn with percentage capacity of discharged

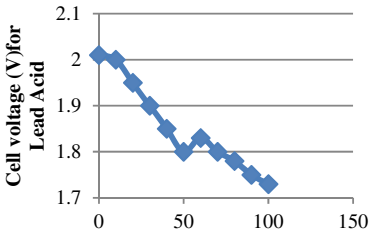


Fig. 8 Cell voltage for Lead Acid with percentage capacity of discharged

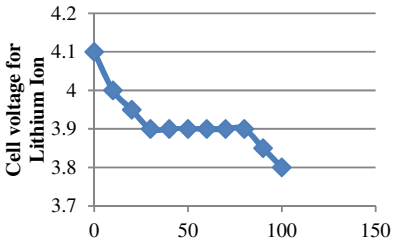


Fig. 9 Cell voltage for Lithium Ion with percentage capacity of discharged

Figure 10 shows that the Cell voltages (V) vary with the percentage capacity of discharged for Lithium-Ion, Lead Acid, Ni–Zn, Ni–Cd, and Zn/MnO_2 electrolytic cells together.

3.2 Discharge Characteristics with Load for PKL Electrochemical Cell

The discharge characteristics of PKL cell/module has been measured carefully. The fabricated PKL module was 6 V, which was used as a load for practical utilizations. A Light Emitting Diode (LED) lamp was used as a load of 6 V. For simplicity, it has been shown that the PKL module system is as shown in Fig. 11.

The observed values of load voltage, load current, and load power are tabulated. Taking the load tests for 120 h has been continued. It may be pointed out here that

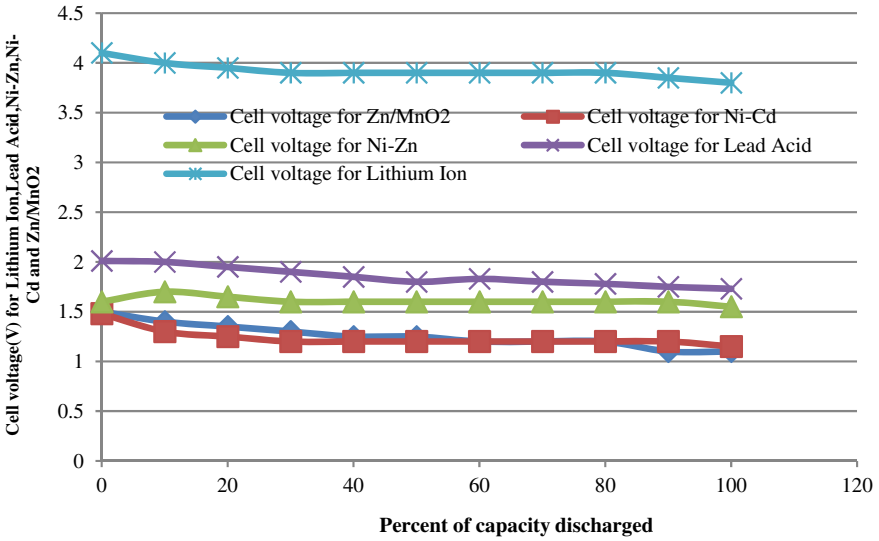


Fig. 10 Cell voltage for Zn/MnO2, Ni–Cd, Ni–Zn, Lead Acid, and Lithium-Ion with percentage capacity of discharged

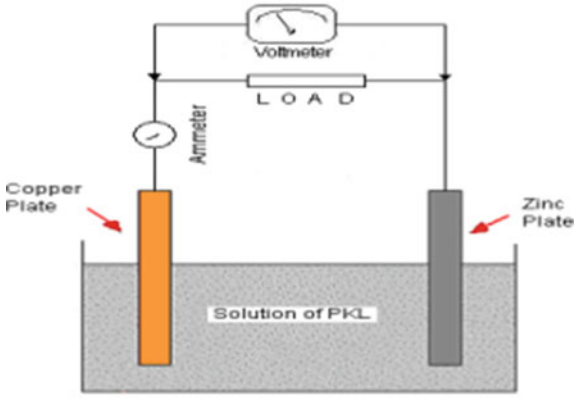


Fig. 11 Experimental setup of discharge characteristics

initially when we connected the load, a substantial voltage drop occurs and this drop is 0.48 V which is around 8% of system voltage.

Voltage, Current, and power of PKL module under load conditions have been studied. The data was taken up to 120 h. The data has been tabulated very carefully. The discharge characteristics of a PKL electrochemical cell with the load are also shown. The Voltage, Current, and power without CO_2-based PKL module under load condition. It is shown that the change of load voltage for 120 h is 0.34 V, that the change of load current for 120 h is 0.32 A, and that the change of load power for 120 h is 1.9 W (Fig. 12).

Fig. 12 PKL electricity without CO_2-based module

Figure 13 shows that the load voltage varies with the time duration. As per this graph, it is shown that the load voltage is reducing gradually as time duration. If we compare it with other cells, we find this change is relatively rapid. Figure 14 shows the variation of load current with the variation of time duration. As per this graph, it is seen that the load current is reducing gradually as time passes. If we compare it with other traditional cells, we find this change is relatively rapid.

Figure 15 shows the load power graph for the same system. According to the definition of load power, we have Power, P = V × I Watts, where V = Voltage (in

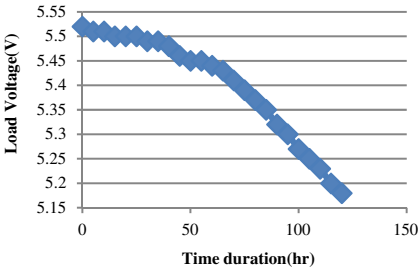


Fig. 13 Load voltage versus time duration plot

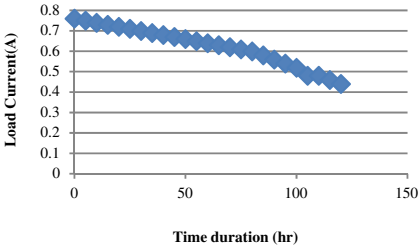


Fig. 14 Load current versus time duration curve

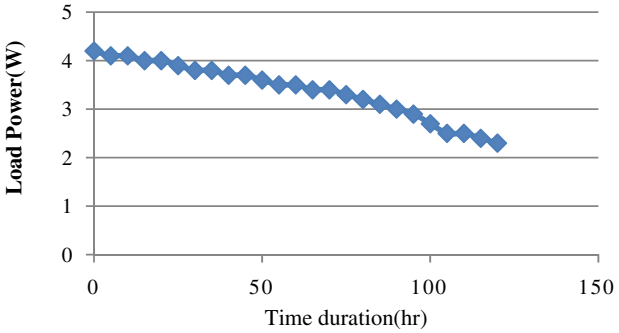


Fig. 15 Load power (W) versus time duration (h) plot

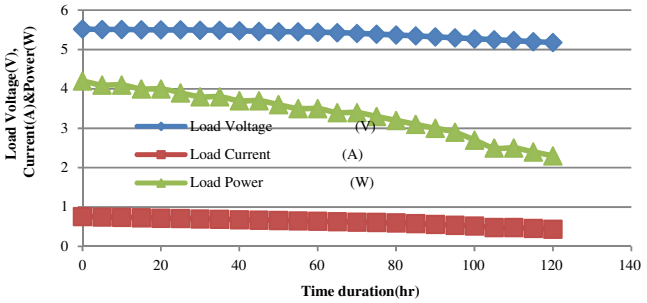


Fig. 16 Load voltage (V), load current (A), and load power (W) versus time duration (h) plot

volts) and I = Current (in Amp). Since voltage and current are reducing with time duration, therefore, as per this graph, it is seen that the load power is also reducing gradually as time passes.

Figure 16 shows the observed results which have been plotted for three characteristics of PKL cells together. The variations of load voltage, load current, and load power with time duration have been discussed.

3.3 Discharge Characteristics with CO_2-Based PKL Electrochemical Cell for Load

Figure 17 shows the methods of CO_2 Production for the PKL module. Figure 18 shows the method of CO_2-based PKL module for electricity production. Figure 19 shows the Measurement technique of pH of the PKL extract of a CO_2-based PKL unit cell. Figure 20 shows the appliance of CO_2-based PKL electricity with the load.

The Voltage, Current, and power of CO_2-based PKL (*Bryophillum pinnatum* Leaf) module under load conditions have been studied. It is shown that the change of load

Fig. 17 Production of CO_2 for PKL module

Fig. 18 CO_2-based PKL extract of a CO_2-based PKL unit cell

Fig. 19 Measurement of pH of PKL module for electricity production

Fig. 20 CO_2-based PKL electricity with load

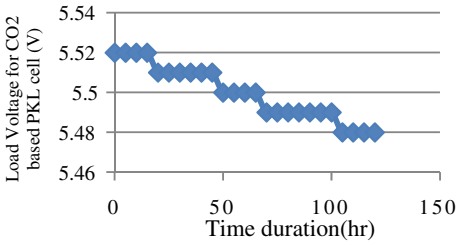


Fig. 21 Load voltage for CO_2-based PKL cell (V) versus time duration (h)

voltage for 120 h is 0.04 V, that the change of load current for 120 h is 0.02 A, and that the change of load power for 120 h is 0.14 W.

Figure 21 shows that the Load voltage (V) for CO_2-based PKL cell varies with the time duration (h). It is shown that the load voltage was constant up to 15 h and then it decreased up to 0.01 V up to 45 h and then after it decreased 0.01 V up to 65 h and then it decreased 0.01 V up to 100 h, and finally, it decreased 0.01 V up to 120 h. Figure 22 shows that the Load current (A) for CO_2-based PKL electrochemical cell varies with the time duration (h). It shows that the load current was constant up to 35 h and then it decreased up to 0.01 A up to 100 h and finally, it decreased 0.01 A up to 120 h.

Figure 23 shows that the Load power (W) for CO_2-based PKL electrochemical cell varies with the time duration (h). It shows that the load power was constant up to 15 h and then it decreased up to 0.06 W up to 35 h and then after it decreased

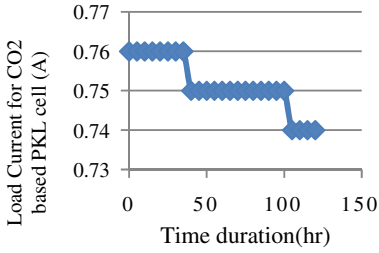


Fig. 22 Load current for CO_2-based PKL cell (A) versus Time duration (h)

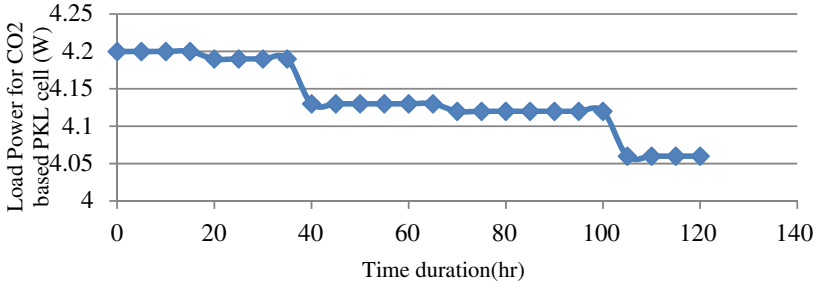


Fig. 23 Load power for CO_2-based PKL cell (W) versus time duration (h)

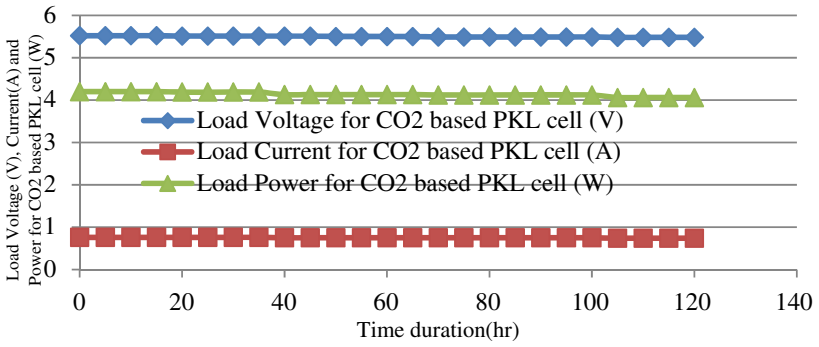


Fig. 24 Load voltage (V), current (A), and power for CO_2-based PKL cell (W) versus time duration (h)

0.01 W up to 65 h and then it decreased 0.06 W up to 100 h, and finally, it decreased 0.01 W up to 120 h.

Figure 24 shows the variation of the voltage, current, and power of CO_2-based PKL module under load conditions. It is shown that the change of load voltage is 0.04 V, that the change of load current for 120 h is 0.02 A, and that the change of load power for 120 h is 0.14 W for 120 h.

3.4 Self-discharge Characteristics of a CO_2-Based PKL Electrochemical Cell

Self-discharge characteristics of a CO_2-based PKL electrochemical cell are found. Using the Self-discharge characteristics, some data for open-circuit voltage (V_{oc}), Short circuit current (I_{sc}), maximum power (P_{max}), and internal resistance (r_{in}) has been collected and then tabulated carefully.

Figure 25 shows that the open-circuit voltage V_{oc} (V) varies with and without CO_2-based PKL electrochemical cell with the time duration (h). It is found that the open-circuit voltage (Voc) is better with CO_2-based PKL electrochemical cell than without CO_2-based PKL electrochemical cell. Furthermore, the open-circuit voltage is more steady with CO_2-based PKL electrochemical cell than without CO_2-based PKL electrochemical cell. So that it can be said that the longevity has been increased for use in CO_2 with the PKL extract.

Figure 26 shows that the short circuit current I_{sc} (A) with and without CO_2-based PKL electrochemical cell varies with the time duration (h). It is found that the short circuit current (I_{sc}) is better with CO_2-based PKL electrochemical cell than without CO_2-based PKL electrochemical cell. Furthermore, the short circuit current (I_{sc}) is more with CO_2-based PKL electrochemical cell than without CO_2-based PKL electrochemical cell. So that it can be said that the performance has been increased for use in CO_2 with the PKL extract.

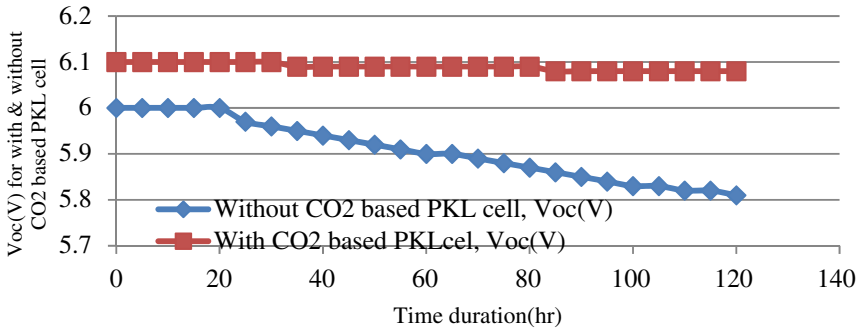


Fig. 25 Variation of Voc (V) with and without CO_2-based PKL electrochemical cell with the variation of time duration (h)

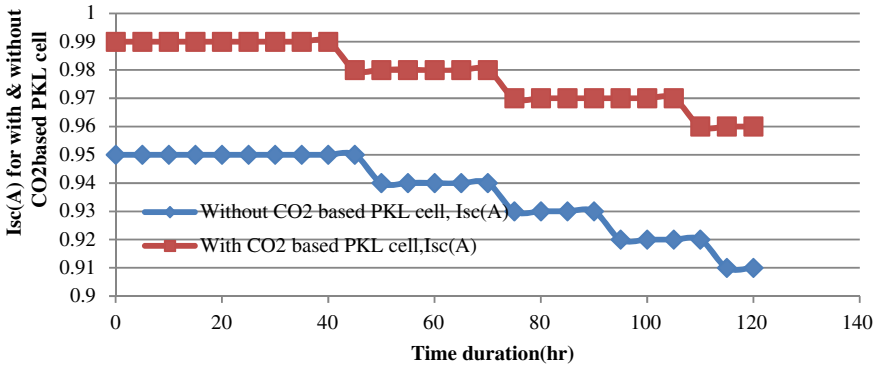


Fig. 26 Variation of Isc (A) with and without CO_2-based PKL electrochemical cell with the variation of time duration (h)

Figure 27 shows that the maximum power P_{max} (W) with and without CO_2-based PKL electrochemical cell varies with the time duration (h). It is found that the maximum power P_{max} (W) is better with CO_2-based PKL electrochemical cell than without CO_2-based PKL electrochemical cell. Furthermore, the maximum power Pmax (W) is more with CO_2-based PKL electrochemical cell than without CO_2-based PKL electrochemical cell. So that it can be said that the performance has been increased for use in CO_2 with the PKL extract.

Figure 28 shows that the internal resistance, r_{in} (ohm) with and without CO_2-based PKL electrochemical cell varies with the time duration (h). It is found that the internal resistance, $\mathbf{r_{in}}$ (ohm) is better with CO_2-based PKL electrochemical cell than without CO_2-based PKL electrochemical cell. Furthermore, the internal resistance, rin (ohm) is less with CO_2-based PKL electrochemical cell than without CO_2-based PKL electrochemical cell. So that it can be said that the performance has been increased for using CO_2 gas with the PKL extract .

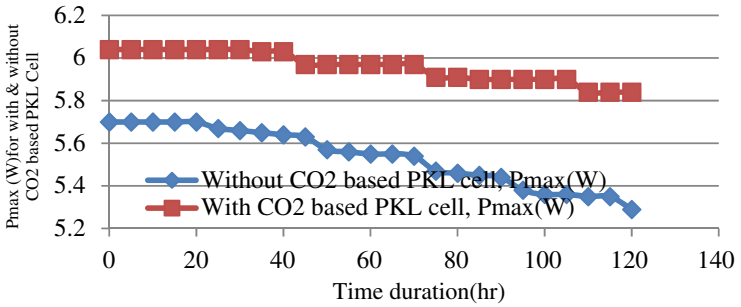


Fig. 27 Variation of P_{max} (W) with and without CO_2-based PKL cell with the variation of time duration (h)

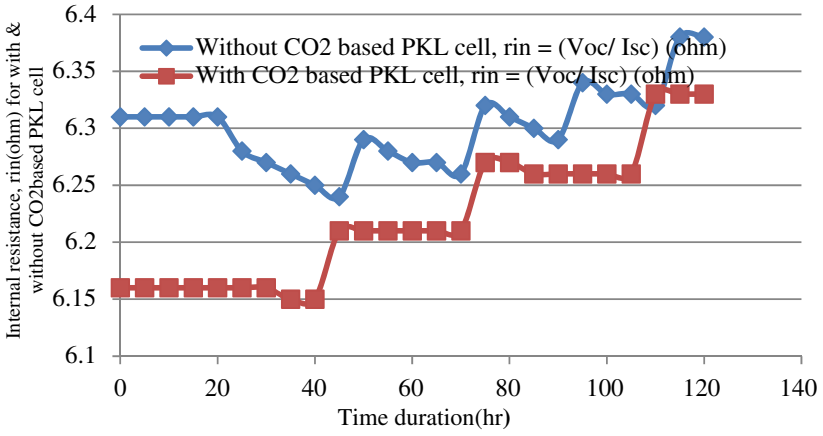


Fig. 28 Variation of internal resistance, r_{in} (ohm) with and without CO_2-based PKL electrochemical cell with the variation of time duration (h)

4 Conclusions

There are a lot of research papers on the electrochemical conversion of CO_2 into useful chemicals. But there was no use of CO_2 for electricity generation. This time the produced CO_2 has been used for electricity production. It is found that the performance of PKL electrochemical cell during electricity generation has been increased for using the produced CO_2. The variation of the voltage, current, and power of CO_2-based PKL module under load conditions has been studied. It is shown that the change of load voltage is 0.04 V, that the change of load current for 120 h is 0.02 A, and that the change of load power for 120 h is 0.14 W for 120 h. The Voltage, Current, and power without CO_2-based PKL module under load condition were found. It is shown

that the change of load voltage for 120 h is 0.34 V, that the change of load current for 120 h is 0.32 A, and that the change of load power for 120 h is 1.9 W. Finally, it is also found that the performances have been increased after adding CO_2 with the PKL extract. The life cycle of the PKL electrochemical cell has been increased. For a bigger size of the PKL electric plant, a CO_2 gas cylinder is needed. By using this CO_2 gas technology, the dead PKL electrochemical cell will get life for the generation of electricity. In this research work, the produced H_2 gas was a byproduct. Mallic acid increases during night time, Performances are better for night time collected PKL than the day time collected leaf. Reactant ion ($H+$) increases in the night time.

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