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WASTE-TO-ENERGY

POTENTIAL OF USING ACTIVATED CARBON AND RICE HUSK ASH IN PRIMARY CELLS

B.Sc. Engineering (Mechanical) Thesis

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NOMENCLATURE

AC	Activated Carbon
RH	Rice Husk
RHA	Rice Husk Ash
BRH	Burned Rice Husk
DC	Direct Current
Al-air battery	Aluminum-Air battery
CH	Coconut Husk
LH	Lignin-hemicellulose
GDL	Gas Diffusion Layer
CVD	Chemical Vapor Deposition
% Wt	Percentage of Weight
K	Kelvin
JFP	Jackfruit Peel
CMC	Carboxymethyl cellulose
EV	Electric Vehicles
OPC	Ordinary Portland Cement
LIB	Lithium-ion Battery
ORR	Oxygen Reduction Reaction
OER	Oxygen Evolution Reaction
HER	Hydrogen Evolution Reaction

ABSTRACT

Rapid growth of population has brought up some serious concerns, such as waste management and excessive use of fossil fuels causing pollution and negative impact on climate change. A large portion of the waste is organic waste, which can be turned into Activated Carbon (AC) and AC can be used to generate electricity by using it as a cathode catalyst in metal-air cell. Aluminum-air cell has the potential of becoming vital in energy-storage application in future because of its high theoretical energy density, even higher than commonly used lithium-ion batteries. However, it is not used widely because the cost of air cathode catalyst and metal anode is high. But if we replace the catalyst by activated carbon obtained from waste or a mixture of activated carbon with other waste (rice husk ash) and use recycled aluminum foil as anode then the production cost might be feasible for vast use of this type of cells. Our objective is to utilize some common and organic waste in the process of fabricating Al-air battery suitable for small and day-to-day usage reducing production cost and limitations. In this paper, we have focused on the feasibility of using activated carbon and rice husk mixture as air cathode catalyst of Al-air cell; and the observations were interesting. We prepared 11 samples of mixture of rice husk and AC in different ratios and initially found the best results to be from 0.68-0.72V, which increases by 8-20% measuring each sample after 3 days. In this study, graphite cathode of a dry cell is replaced too by mixture of AC and RHA. Voltage drop is quite negligible for the mixture of 10% RHA. And, voltage is similar to the new battery in case of 100% activated carbon as cathode. If considered the environmental effect, using recycled activated carbon and rice husk ash will decrease the pollution.

CHAPTER 1

INTRODUCTION

As human population has increased rapidly in a very short time, it is affecting every aspect of our daily life. Even management of waste has become a great problem because of this vast population. Poor waste management resulted in soil pollution, water contamination, inhabitable atmosphere[1]. Public health has been greatly affected due to pollution. For the developing countries like Bangladesh, it is very difficult to efficiently process the huge amount of waste it produces. Then again, we are in the peak of the improvement of technology. With the development of science and technology, various ways of waste recycling and management have been introduced.

As stated, we are in the peak of the improvement of technology, there needs to be production of immense amount of energy; And we cannot deny the fact that, we are producing harmful waste in this procedure by burning fossil fuels. A large amount of energy is consumed in the form of DC Cells. Various portable electrical devices use dry cells as energy source. Dry cell is a cell in which the electrolyte exists in the form of a paste, is absorbed in a porous medium, or is otherwise restrained from flowing. It's a type of electric battery, commonly used for portable electrical devices. It transforms chemical energy to electrical energy. A little moisture is present to allow the flow of current[2]. DC cells are used in many components such as calculator, cameras, clocks, watches, torches and in various portable devices. The global battery market is about \$50 billion US, of which roughly \$45.5 billion is allocated to non-rechargeable batteries[3]. Aluminum-air battery produces electricity from the reaction of oxygen in the air with aluminum. They have one of the most noteworthy energy densities of all dry cells. Using waste and recycled products as raw materials, we can reduce the cost of procurement. The aluminum-air battery is very much suitable and has the potential of becoming source of power for electric vehicles due to its high energy density; much higher than that of the lithium-ion batteries[4].

Our Primary objective has been producing a source of energy using recycled wastes as raw materials. Singapore has already established waste-to-energy plants where solid wastes are burnt to produce steam to generate electricity. Further studies and application are going on all around the world. But there has been very little work on producing DC cells from waste

materials. In our study, we have used recycled aluminum foil, activated carbon, rice husk ash, recycled tissue paper etc. solid wastes to procure functioning DC cells.

External covering of the rice grain is called the rice husk and it is extracted while milling process. Rice husk is very much available in rice producing countries and 30%–50% of it is organic carbon[5]. Rice Husk Ash is used in many sectors. RHA is used to improve the microstructure of the cement paste[6], silica is also extracted from RHA[7]. As it contains significant amount of carbon, it can be used as an alternative to graphite in cathode.

Various organic wastes can be turned into Activated Carbon. Many experiments have been conducted in this topic. Activated carbon can be produced from palm oil shell, durian shell, coconut shell, date stones, almond shell, jackfruit, rubber wood etc. organic wastes.

AC and RHA mixture can be used to generate electricity by using it as a cathode catalyst in metal-air cell. Aluminum-air cell has the potential of becoming vital in energy-storage application in future because of its high energy density. We even studied the possibilities of using recycled activated carbon as cathode in regular non rechargeable dry cells.

1.1 PROJECT OBJECTIVE

Our project is developed to work on waste-to-energy to contribute in waste management and reducing harmful environmental impact of used dry cells. By using recycled raw materials and removing harmful chemicals, we want to produce environment-friendly source of electro-chemical energy. We are studying two methods to replace conventional dry cell battery.

We studied Al-Air batteries, which is not used widely because the cost of air cathode catalyst and metal anode is high. But we replace the catalyst by activated carbon obtained from waste or a mixture of activated carbon with rice husk ash and used recycled aluminum foil as anode which decreased production cost.

As an alternative to graphite rod of primary DC cells, we tried to reduce the manufacturing cost by using a mixture of Rice Husk Ash and recycled Activated Carbon. To carry out this study, graphite cathode was replaced with our mixture and the result was analyzed to determine its feasibility.

1.2 THESIS OVERVIEW

In this book, we are discussing the details of our study and result briefly. Discussion of raw materials, their other uses, our methodology, previous works of our model, analysis of results, limitations and suggestions all are included in this book. We divided the book in seven (7) chapters according to topic.

In the first chapter, an introduction of our project is given. We have given an overview of our project and the book.

In the second chapter, we reviewed previous works related to our study. We discussed previous works on waste management technologies, preparation of activated carbon, use of activated carbon and rice husk ash and review of Al-air cell.

In the third chapter, the methodology of our experiment is described. Procedure of burning the rice husk, collecting activated carbon, preparing mixtures and the setup of our experiment are discussed.

In the fourth chapter, results from our experiment are presented. The results have been analyzed and shown in graphical and tabular form. We also have discussed the findings and feasibility of our project later in this chapter. We also discussed the potential and application of our experiment.

In chapter five, we discussed the future works that can be done based on our project. we discussed our limitations and challenges and how that should be avoided on future experiments. We also discussed the scope of improvements. Conclusion of our book is given by discussing the overview of our work and results.

Relevant figures and tables are presented throughout the book and their lists are indexed. References of the research papers which were necessary for our experiment are added at the end of the book.

CHAPTER 2

LITERATURE REVIEW

Literature review is generally surveys books, articles and other sources which are relevant to a particular topic, research area or theory. By doing so, it provides a brief description, short summary and evaluates these works relating to the specific research problem which is being investigated. Literature reviews provides an overview of sources that have been explored while researching. It also demonstrates to the readers how a research fits within a larger field of study.

This study contains a lot of areas which should be studied. During this experimental work, researchers needed to read about waste management techniques, procedure of preparing activated carbon, topics covering the sectors in which activated carbon and rice husk ash is being use and has the potential of being used.

2.1 PREPARATION OF ACTIVATED CARBON

Activated carbon or otherwise known as activated charcoal, which is actually crude form of graphite, substance which is commonly used as pencil lead. Besides activated carbon is widely used in dye removal and also has other applications. Activated carbon is widely used in air purification, energy storage, electrode materials for li-ion batteries biosensors, chromatography, hydrogen storage, immobilizing the biomolecules. It has high surface area, adsorption capacity, and high adsorption rates from the gas or liquid phases. So wide application of activated carbon is observed. Activated carbon is used as solvents recovery, in gas separation and sometimes as catalyst. Besides the application of activated carbon is seen in waste-water treatment plants which is used to remove the organic pollutants from the drinking water. By implementation of different chemical methods activated carbon is prepared from many resources for most of these applications. From raw carbon resources like lignite, unburnt coal, peat, and biomass waste such as sawdust, wood, coconut shell, sugar cane bagasse, rice husk, oil-palm stone, coffee beans Lignocellulosic waste materials, paulownia wood, pomegranate seeds, cattail, olive tree, jatropha hull, bamboo, orange peel, thevetia peruviana, ramie, grape stalk, pine apple waste biomass, and almond shell activated carbon is prepared. Again, by pyrolysis of physic nut waste activated carbon is also produced. Activated Carbon, prepared from all these sources, have high surface area, adsorption capacity, high adsorption rates for liquid gas separation, adsorption. Several Researchers have reported the preparation of AC from different local sources. These preparative methods are explained below.

2.1.1 Rice Husk

AC was prepared from rice husk (RH) [8], because RH gives better yield of AC with high surface area. The AC which was obtained from RH have critical conditions like temperatures 293.15 K, 303.15 K, 313.5 K and 323.15 K and pressure up-to 3.5 MPa. Firstly, RH was washed then dried properly. In the presence of nitrogen dried RH was carbonized at 1273K. Then, the RH was crushed, after that KOH was mixed with crushed RH. Then, the mixture was heated to 1073 K in presence of nitrogen. The activated product was constantly washed with distilled water, then this washed product was dried at 393 K in vacuum. As a result, AC was prepared.

Activated carbon can be prepared in many ways. Another method used to prepare AC is given. First of all, impurities of inorganic material of Silica (SiO_2) was removed by washing RH with hydro-fluoric acid (HF) [9]. To prepare AC, at normal temperature for about 12 hours 30g of RH were taken in 14mL of HF. Under Nitrogen atmosphere (100 mL/min) Raw RH (washed) was carbonized by heating it at the temperature of 673K for 4 hours to achieve the better results of activating agent during impregnation. Impregnation of all material was carried out at 333K. Impregnated Product was dried at 373K for 24h, in the presence of an activating agent; ZnCl_2 or NaOH, at the rate of 1/3 & 1/4 (RH/Activating agent). Finally, at the temperature of 873K activation was done in period of 4 hours in presence of Nitrogen and appreciated extent of AC was prepared. Different fractional quantity of mesopores, surface acids group, AC is obtained by varying the temperature and time.

2.1.2 Coconut Husk

AC obtained from coconut husk (CH) have much greater advantage because it was successfully optimized for basic dye adsorption[10], [11]. Locally collected raw material was used to prepare Optimized AC. First the raw material was washed to remove dirty particles from the surface. Then, at the temperature of 378 K the washed material was dried in oven. Then, it was cut into desired small pieces and loaded onto a vertical tubular reactor made of stainless steel and placed in tube furnace. The whole carbonization process was done in presence of nitrogen which was flown at $150 \text{ cm}^3/\text{min}$ rate. The char mixed with pellets of potassium hydroxide (KOH) within different impregnation ratios (IR). Mixture was dehydrated for removal of moisture content. Under the flow of nitrogen AC was cooled to 298K. Finally, AC was washed with warm water.

2.1.3 Coconut Shells

In this section, the method of preparing AC using physically or chemically activation process of coconut shells[12] is discussed. For physical activation, raw materials were carbonized under inert-atmosphere. For chemically activating, raw materials were reacted with chemicals to help with initial dehydration. Chemical activation is commonly used as bio-material to obtain high production and more surface areas. During this process, lower energy values are used. H_3PO_4 solutions were produced in 1.0, 1.5, 2.0 impregnation ratios. The solution was defined as ratio of the dry weight of H_3PO_4 with respect to weight of coconut shell. To make the samples, 10g of coconut shells were used. The time of soaking was fixed, 12 hours. The activation temperatures were 673 and 773 Kelvin. The time of activation was set as 10, 20 and 30 minutes. At 673 Kelvin temperature, moisture and volatile materials were removed. Impurity of acid from AC was removed by washing them. Moreover, AC was washed with 100ml of distilled water. When pH reading was 6-7, it was acidic. When pH reading was 7-8 for washing, then the sample became neutral. Activated carbon was again washed using 0.1M NaOH solution and then finally washed with distilled water. After washing the sample, they were placed in an oven at 723 Kelvin temperature for drying.

Another way to activate carbon is, washing the coconut shells with deionized water for cleaning, dried and then browned using rural mill. And then sealed for the experiment [13]. The 1st step is carbonization of coconut shells. Coconut shell was placed in a stainless-steel pot and kept horizontally inside a tube furnace where it was heated electrically. The temperature inside was up-to the carbonization temperature. After finishing carbonization, sample was cooled to 298K, in the room temperature. In the 2nd step, the activation was done in self-made tubular microwave furnace. Approximately, 25g of fully carbonized compound was placed inside the reactor. After that, it was heated with N_2 -flow. There were three types of activation were involved. They are, steam activation, carbon di-oxide activation and Activation by steam with CO_2 .

There is another way to prepare AC from coconut shells [14]. Coconut shell was firstly dried at the temperature of 378K for a day (24 hours). After that, it was crushed using laboratory blender. Then it was grounded and sieved into the coarse granules. Zinc Chloride ($ZnCl_2$) & Carbon Di-oxide (CO_2) were used to activate the reaction, while Hydrogen Per-Oxide (H_2O_2) were used for oxidizing the products. These were pretreatment procedures which were performed in producing AC. The samples were then soaked within the Zinc Chloride ($ZnCl_2$)

solution in 1:1, 2:1, 3:1 ratio and then the samples were dried at the temperature of 378K. After the soaking process was done, the sample was refluxed with H_2O_2 . In that time, the temperature was held at 373K for 60 min to get solid product. By filtering, the solid material was separated from H_2O_2 solution and then it was washed using de-ionized water. After that, the final product was dried at 378K temperature. The samples obtained after filtrating were added to the reactor which was a PARR- 4848 autoclave at 473K. That reactor was allowed to cool to 298K and the final product were dried at 378K. The temperature was preferred for the treatment was 473K to deliver hydrochar having maximum Oxygenated Flouro Groups. The Hydrochar was obtained by using a hydrothermal treatment. During that process, 25g of sample was mixed with 125 mL water and $ZnCl_2$ to get the ratio of $ZnCl_2$ and Raw material as 2:1 or 3:1. The mixed samples were treated, at different temperatures (473K, 548) for 20 minutes in the reactor. Precursor was then activated by using a physio-chemical process. Those precursors were then loaded on alumina boats in furnace. Temperature was increased to 1073K at the $10^{\circ}C/min$ at 50mL/min flowrate of nitrogen. The final AC was dried and consumed for analysis.

2.1.4 Olive Tree Residue

Olive tree wood in ratio of 3:5:1 was impregnated with the acidic solution of H_3PO_4 [11],[15]. The samples were dried very rapidly. After that, they were stored in a desiccator. The impregnated samples were then carbonized inside a furnace. The furnace was equipped along with Euro-therm 904 temperature controllers and 1m long tubular ceramic insert. At first, the temperature of the furnace was calibrated. The carbonization was occurred at 623-723K temperature with 283K/min heating rate. At that time, a constant nitrogen flow of $100\text{ cm}^3/min$ was going on for 1hour. Then, the products were washed with distilled water inside vacuum to reach pH=6 in liquid residual. Using barium nitrate, the disappearance of phosphate ions in solution was determined. The resulted final products were dried in oven and further dried in closed container.

2.1.5 Bamboo

The bamboo were carbonized at 673K for 1hour[16], [17] in a furnace. The bamboo samples were cooled down at room temperature (298K). During the process of carbonization, nitrogen gas was flushed to maintain inert atmosphere. With the impregnation ratios of 0.5, 1, 1.5, 2, and 2.5, K_2CO_3 was mixed with solid carbonized. After finishing mixing process, all the samples were dried at 393 K inside an oven for about 12 hours. AC was produced when these samples were heated under the conditions of activation temperatures of 1073K, 1123K, 1173K,

1223K and 1273K, nitrogen flow rate of 200mL/min and activation times were 1 hours, 1.5 hours, 2 hours, 2.5 hours and 3 hours. Then with the continuous flush of nitrogen gas, AC was cooled inside furnace tube at room temperature with de-ionized water. The cooled solid was then washed and filtered to gain the neutral pH value of 7.

2.1.6 Rice Straw

First, fraction extraction of lignin-hemicellulose (LH) from rice straw was performed by treating it with NaClO₂ and then washing and drying it at the same time. [18]. To make hemi-cellulose, de-waxed rice straw (94.7%) powder was put in 1.4 Wt% NaClO₂ under acidic conditions (PH=5 by 10% CH₃COOH) at 15 mL/g liquid to solid ratio at 343K for 6 hours. The filtrate is treated with ethanol to produce alkal-soluble lignin and hemicellulose. The LH powder was dried in an oven, then put in a quartz tube with a 2cm inner diameter and dried in a furnace at 333K for 12 hours (5^oC/0.5h). The LH was then heated to 1073K with a flow rate of 100 ml/min of N₂. Finally, activated carbon was collected and cooled, and AC particles were washed in 5% HCl to eliminate impurities before being dried at 378K for half an hour.

2.1.7 Pineapple Waste Biomass

To extract impurities, the solid pineapple waste biomass was washed with tap water. [19]. It was then oven-dried at 383K until a moisture content of 5-10% was obtained. The samples were cut into small pieces of random shapes after drying. The chemical activation method was used to prepare the AC, which is given as follows. Raw material was added to the beaker (500ml) containing the 1:1 ratio activation agent and left to be immersed for 1 day at 298K by occasional stirring with a glass rod. Then the sample was dried at 383K for 24 hours by carbonization at 773K for 1 hour. It was then cooled to a temperature of 298K. After that, the activated sample was washed with hot distilled water (303-308K) to remove the unused activating agent and then oven-dried at 373K for a further 24-hour period.

2.1.8 Almond Shells

Almond shell was washed and dried for removing the moisture content at 378K for period of 24 hours [20]. The samples were then grounded and sieved to 1–2 mm of size. Both carbonization and activation of chars were carried out in an electrical furnace ‘Nabertherm’. A vertical stainless-steel reactor was placed inside it. During the process of carbonization, 15g raw materials were used for the preparation of the chars. From the beginning of the process, nitrogen gas at the rate of 150 cm³/min was flowing. The temperature of the furnace was increased at 5^oC/min rate from room temperature to 450^oC. The temperature was held for 1

hour. After pyrolysis process, the furnace was rapidly cooled to room temperature with nitrogen flushing through the sample. The charcoals were then activated by a CO₂ flow to prepare for the final product at fixed heating rate at 10⁰C/min.

2.1.9 Coal Pitch

The coal pitch and n-pentene were mixed with a ratio of 1:10 and stirred continuously. [21]. The mixture was stored at 298K for a period of 2 hours. After that, the soluble part of the sample was separated from the sample by vacuum filtration, which was done at least three times. After filtering three times, asphaltene was obtained and passed through the drying process. This resulted asphaltene was then used as solvent. It also had contaminants. As a result, to produce clean and fine product, it was filtered. Then, a mixture was created with cleaned asphaltene in 10grams and KOH within specific ratios of 0, 1:10, 1:4, 1:2 and 1:1. The resulting mixture was heated at different temperatures (673K, 723K, 773K, and 823K) for a period of 60 minutes under a purified nitrogen gas flow of 400mL/min. KOH was used to reduce the formation of tar and to enhance the carbonization process. After the carbonization reaction, the resulting char was washed with 1mol/L HCl solution and washed again with hot distilled water to reach pH=7 of the wash. After washing, the separation of the residues in solid form was carried out by vacuum filtration, then oven-dried at 373K for a period of 12 hours. After drying, the activation of the carbon was performed by steam with nitrogen at different temperatures for half an hour. In extreme temperature conditions, carbon and steam were treated to produce CO and H₂ with a large amount of pore produced. The gas carrying out the reaction was supplied with a flow rate of 400mL/min, while the amount of steam in the carrier gas ranged from 4g/hour to 16g/hour.

2.1.10 Jackfruit Peel Waste

The raw precursor to the production of AC was jackfruits. [22]. Then the jackfruit peel (JFP) was separated and the carpel fiber was removed to clean it. The JFP was repeatedly washed with distilled water to remove impurity. After that, the JFP was oven-dried at 378K to obtain a dried sample of constant weight. Then the dried sample was ground with the help of the JANKLE and KUNKEL micro hammer mill to reduce the size of the sample particles. After that, the dried JFP was placed in the desiccator as a precursor to the preparation of the AC. Precursor material was carried through an approximate testing to identify the other contents present in JFP, which were 4.22% of moisture content, 10.19% of ash, 50.17% of volatile matter while 35.42% of fixed carbons. Precursor materials of 20 grams along with phosphoric

acid and 85 %Wt concentration were impregnated with stirring. The concentration ratio of H_3PO_4 was fixed in order to bring the impregnation ratios to 1:1, 2:1, 3:1 and 4:1. In a desiccator, the resulted slurry was placed.

When the time period of 24 hours had passed, the slurry was ready to pass through two steps. In the first step, slurry was placed in the horizontal tubular shaped reactor. Then placed in the muffle furnace to proceed with the semi-carbonization process at 473K for about half-hour. Then the slurry became black, generating dark colored sticky dry powder. Upon completion of the semi-carbonization process, the powder passed through the heater to reach the activation temperature. Then carbonization process was carried out under the nitrogen flow rate of $100\text{cm}^3/\text{min}$ at standard temperature-pressure for around of 45minutes. After that, the powder was activated, which was converted to AC. The AC product was passed through the collection and then cooled in the desiccator, washed repeatedly with hot distilled water at 343K to neutralize the pH of the washing solution. In the end, the AC was vacuum oven-dried at 383K for a period of 24hours and stored in a desiccator for further analysis.

2.2 USE OF ACTIVATED CARBON

Porous carbon materials play an important role in adsorption technologies, particularly in new applications such as catalytic support, battery electrodes, capacitors and gas storage. Such applications require carbon materials to exhibit high surface areas along with high mesopore or macropore ratios to micropores, as many macromolecules and ions exceed the pore diameters of micropores and therefore have difficulty entering them. [23] .

2.2.1 Activated Carbon as an Adsorbent

The word adsorption refers to the processes in which the solid surface of the substance is concentrated from its gaseous or liquid environment. The history of carbon adsorption in water purification goes back to ancient times [24]. Adsorption of porous carbon was mentioned as early as 1550 B.C. in Egyptian papyrus, and later in Hippocrates and Pliny the Elder, mainly for medicinal uses. Eventually, the adsorption phenomenon has been observed in scientific records by C.W. Scheele. [25], [26] in 1773 for gases exposed to carbon. This was followed by observations made by Lowitz in 1785 concerning the reversible removal of dye and odor produced by wood-charcoal substances from water. It is now common practice to understand the different types of adsorption: physical adsorption (physisorption) and chemisorption [29]. However, carbon adsorption efficiency depends on the different raw material sources, the history of its preparation and treatment situations, like pyrolysis temperature and activation

time. Several other factors can also influence adsorption capability under same sorption conditions as surface chemistry (heteroatomic content), surface loading as well as pore structure [27].

With the expansion of humanity, society, science, technology, our human race is hitting new horizons, but the price which we will pay in the coming years will surely become too massive. Researches have already shown that activated carbons are promising tools for the removal of various forms of pollution in particular, but their use is sometimes limited due to higher costs.

In light of the above, the potential of the rice husk, the abundant disposal of waste from the rice milling industry, has recently been shown to be prominent in the wastewater treatment process, not only to remove different types of dye [27], but also other organic or inorganic pollutants, such as metal ions, phenols, pesticides, chlorinated hydrocarbons; ideal adsorbents for gold-thiourea refining [28] and as air purifier in cleaning of atmospheric contaminants[29]. The major components of the rice husk that may be responsible for sorption are carbon and silica [29], [30]. Silica is composed of $\text{SiO}_4 \cdot 4\text{H}_2\text{O}$ in which each oxygen atom is shared between two adjacent tetrahedrons. The Si–O bond is about 50% ionic due to a large difference in the electro-negativity of oxygen and silicon. The ionic structure of silica thus provides the ability to adsorb polar molecules. [31].

2.2.2 Electrodes for Battery and Capacitor

As porous carbons are mostly amorphous in nature, a little presence of sp^2 carbon structures enhances the possibility of using these carbon materials for wider applications involving electrical conductivity[32], [33]. The sp^2 carbon sites in the carbon materials predominantly control the electronic and transport properties[34].

The porous carbon composite material, contains carbon and SiO_2 as components of the material. As SiO_2 is well known to be an insulator, the conduction property of rice husk activated carbon may be assumed to be due to the carbon component of the material[35]. The electron transport properties of the carbons prepared are investigated in detail along with their microstructure by Kennedy. The analysis of the variation of conduction as a function of temperature showed two different conduction processes operating respectively at moderate temperatures $T_b 150^\circ\text{C}$ and high temperatures $150 < T_b < 250^\circ\text{C}$. The first process that is due to a variable range hopping mechanism in the π localized states around E_F satisfying Mott's law and the second one due to a thermally activated process associated with an energy gap. The X-ray diffraction pattern of the sample confirmed the presence of the graphitic structure. The results

of electrical conductivity indicated that even though the tested samples were acid or water washed, substantial amounts of water-soluble minerals remain in the carbons[36].

The EDLC (electric double layer capacitors) or super-capacitors of the carbon/graphite composites in KCl can achieve value as high as 210 Fg^{-1} . It was found that the specific capacitance was not linearly proportional to the surface area. The EDLC system for high capacity is strongly dependent on the pore structure. Compared to commercial grade carbons, rice husk-based porous carbons could give higher double layer capacitance and can be used as electrode materials for EDLC manufacture[37]. Moreover, the electronic property of the porous carbons from hydro-chars by H_3PO_4 activation was also tested. A specific capacitance of 130 Fg^{-1} was achieved by using the porous carbon, indicating that the porous carbon prepared by this route has good electrochemical performance, and the localized graphitic nature of the porous carbon was proved by X-ray diffraction pattern [34].

2.2.3 Catalytic Supports

For catalytic applications, the carbon support should possess not only a high surface area but also high meso-porosity. Some people have confirmed that mesopores significantly enhance the catalytic activity of materials by allowing macromolecules to penetrate them and get easily adsorbed on the surface of the catalyst [38]–[40]. Lu et al. [41] prepared porous carbons from rice husks by activation with CO_2 .

Cobalt catalysts were supported on the porous carbons by excess-solution impregnation, and were used to carry out reactions with different constituents such as $\text{NO}+\text{CO}$, toluene, $\text{NO}+\text{toluene}$, and $\text{NO}+\text{CO}+\text{toluene}$ in the presence of 6% O_2 at 250°C to evaluate the activity of porous catalysts. The BET analytical results revealed that RHCO_2 (rice-husk-derived carbon by using CO_2) hold of higher mesoporous content. More high mesoporous content carbons were advantageous for catalyst preparation by conventional impregnation method; and exhibited good active-site distribution, and the active phases were nanosized. During toluene oxidation, RHCO_2 -catalysts showed higher conversion efficiency (100%) for deep oxidation at 250°C than other catalysts.

2.2.4 Hydrogen Storage

Hydrogen is an ideal alternative to fossil fuels from an environmental point of view because its combustion does not generate pollutants such as particles, nitrogen oxides, sulfur oxides, hydrocarbons, and carbon monoxide[42]. Today, many scientists and engineers, some

companies, governmental and non-governmental agencies and even finance institutions are convinced that hydrogen's physical and chemical advantages will make it an important synthetic fuel in the future[43]. From an economic point of view, hydrogen storage is an essential prerequisite for the widespread deployment of fuel cells, particularly in transport[44]. This interest has led to the so-called “hydrogen economy”. None of the known storage methods (compression, liquefaction, or storage as metal hydrides), however, can meet these targets drawn up by the US Department of Energy. It is quite clear that these targets cannot be met by systems that store hydrogen at high pressure or as a liquid at cryogenic temperatures.

Lillo MA et al.[42] studied the possibility of storing hydrogen using activated carbon. In their research, hydrogen storage has been studied in a large variety of activated carbons and activated carbon fibers and a wide range of pressures (up to 70 MPa). The experimental technique used has good reliability, and the experiments performed have a small error and high reproducibility. This seems to be essential to get trustworthy conclusions. In these samples, we have not found large amounts of hydrogen adsorbed. In any case, an activated carbon derived through a simple preparation method provides hydrogen storage values at 10MPa close to 1wt %. Experimental results have been compared with theoretical work found in the literature, and an important agreement can be observed. From this study, we conclude that the optimum pore size for hydrogen storage is that which can hold two layers of adsorbed hydrogen. This work also considers practical aspects related to hydrogen storage in activated carbons and activated carbon fibers.

2.3 USE OF RICE HUSK ASH

Rice husks are the hard-protective coverings of rice grains that are separated from the grains during the milling process. Rice husk is an abundantly available waste material in all rice-producing countries, and it contains about 30%50% of organic carbon. In the course of a typical milling process, the husks are removed from the raw grain to reveal whole brown rice which upon further milling to remove the bran layer will yield white rice. Current rice production in the world is estimated to be 700 million tons. Rice husk constitutes about 20% of the weight of rice and its composition is as follows: cellulose (50%), lignin (25%-30%), silica (15%-20%), and moisture (10%-15%). In the Rice Husk Ash, it is possible to find approximately 20% silica in the amorphous form [45]. In addition, it consists of 60-65% volatile matter, 10-15% fixed carbon, and 17-23% ash [46].

2.3.1 Research on RH Application

Rice husk is used for a variety of purposes depending on its physical and chemical properties, such as ash content and silica content. Rice husk is used directly as a strong fuel in power plants. It's also used as a starting material for the production of silica and silicon compounds. Rice husks are used in a variety of industries and domestic settings.

Rice husk is used as a fuel for processing paddy and producing process steam. Combustion and gasification generate heat energy. Rice husk is used as a fuel in low-capacity boilers in small-scale process industries. 1 ton of rice husk is needed to produce 1 MWH (million-watt-hour) of electricity. It's also used as a source of renewable energy in the home. [47].

Rice husk is used as a raw material for the production of activated carbon because it contains a high amount of cellulose and lignin. Because of their complex microporous structure, activated carbon is an important adsorbent [48]. Rice husk composted slowly due to its high lignin content, so earthworms were used to speed up the process. Rice husk can be processed into fertilizer in four months using the vermicomposting technique. Boiled rice husk is used as a substrate or medium for gardening, including hydroculture in some cases [48]. Rice hulls are the rice's outermost covering and are available in organic and natural varieties. Rice hulls are a low-cost by-product of human food production that can be used as a source of fiber and filler in low-cost pet foods. Rice husk contains 20% silica, making it an excellent raw material for producing silicon compounds such as silicon nitride, zeolite, silica, and pure silicon [50]. Bricks are made from rice husks. The higher the percentage of rice husk in the brick, the more porous the brick will be and the stronger the thermal insulation will be.[49].

Fapohunda et al. [52] discovered that the workability of concrete declines as the percentage of cement replaced by RHA increases, unless appropriate admixtures are used. Zhang MH et al. [53] proposed using RHA concrete with a low water–cement ratio and admixtures, and showed that RHA contains a lot of silica, which helps to increase the toughness of concrete when used in the right proportions with cement. When exposed to a magnesium sulphate solution, RHA concrete expands less than standard OPC concrete (control concrete). When exposed to magnesium sulphate solution, RHA concrete had a 2% lower strength reduction than OPC concrete. RHA in concrete, according to Chindaprasirt et al. [54], improved chemical resistance. According to Zarrei et al. [55], incorporating RHA with cement helped to reduce chloride ion penetration and water absorption According to Nicole PH et al. [56], raising the RHA replacement level increases chloride penetration resistance. According to Christopher et

al. [52], the use of RHA in concrete reduces the possibility of concrete corrosion in coastal areas due to chloride penetration.

Rice husk is used to make xylitol, furfural, ethanol, acetic acid, and other products. In the metal and computer industries, it is used as a cleaning and polishing agent. It's also used as a construction material and an industrial raw material, for example, as an insulating board material, a plastics filler, a filling material, and a material for producing panel board.[50].

2.4 ALUMINUM – AIR CELL

The aluminum-air battery is considered to be a promising source as a power source for electric vehicles (EVs) due to its high theoretical energy density, which is significantly higher than that of state-of-the-art lithium-ion batteries (LIBs). However, a few other technical and scientific difficulties preventing the large-scale development of al-air batteries still have not been resolved. Throughout this review, we express the fundamentals, obstacles and recent advances in al-air battery technology from aluminium anode, air cathode and electrocatalysts to electrolytes and inhibitors. Initially, aluminum alloys with transition metal elements are reviewed and shown to reduce Al's self-corrosion and improve battery performance. In addition, extensive studies of electrocatalytic materials for the reduction/evolution of oxygen, including Pt and Pt alloys, non-precious metal catalysts and carbonaceous air cathode materials, are highlighted for the cathode. In addition, the application of aqueous and non-aqueous electrolytes in al-air batteries is discussed for the electrolyte. In the meantime, the addition of inhibitors to the electrolyte to enhance electrochemical performance is also being explored.

As an alternative to conventional dry cells, we will produce aluminum-air batteries using recycled aluminum foil, rice husk ash, activated carbon waste, etc. These batteries produce electricity from the reaction of oxygen to aluminum in the air [51]. They had some of the most remarkable energy densities in all of the dry cells. However, when using conventional electrolytes, they are not generally used for issues of high anodyne cost and by-product expulsion. We can reduce the costs of procurement by using waste and recycled products as raw materials. The aluminum-air battery is very suitable and has the potential to become an electric vehicle power source due to its high energy density; much higher than that of the lithium-ion batteries [52]. Compared to other metal-air batteries, Al-air is very suitable for large-scale energy applications in the future due to its low cost and high specific capacity,

which is only the second highest in terms of lithium and much higher than that of magnesium and zinc [53], [54].

Table 1: Various metal - air battery parameters[4].

Batteries	Theoretical Voltage (V)	Theoretical Specific Capacity (Ah/kg)	Theoretical energy density (kWh/kg)	Practical Operating Voltage (V)
Li – air	3.4	1170	13.0	2.4
Zn – air	1.6	658	1.3	1.0 – 1.2
Mg – air	3.1	920	6.8	1.2 – 1.4
Na – air	2.3	687	1.6	2.3
Al – air	2.7	1030	8.1	1.2 – 1.6

Al–air batteries were first proposed by Zaromb et al. [55][56] in 1962. As a result, efforts have been made to apply them to a variety of energy storage systems, including EV power sources, unmanned aerial (and underwater) vehicle applications and military communications. [57][58][59][60]. In 2016, researchers have shown that the EV can drive more than 3000 km with the use of an al–air battery weighing ~100 kg. Despite this, Al–air batteries still have many challenges ahead of their practical application, such as the accumulation of by-products that can suppress the electrochemical reactions of the al–air battery. Based on all of this, this summary will discuss the challenges related with Al–Air battery components, including electrolytes, anodes and air cathodes, in creating better Al–Air batteries.

2.4.1 Electrolyte

The electrolyte is a core ingredient of Al-air batteries, and this section presents aqueous electrolytes used in these systems as well as additives that may influence the properties of the batteries. As for ionic liquid and deep eutectic solvent-based electrolytes for al-air batteries. Water-based aqueous electrolytes can be classified as acidic, neutral and alkaline depending on their pH values. Alkaline solutions such as potassium hydroxide (KOH) and sodium hydroxide (NaOH) solutions are the most common electrolytes developed for use in al–air batteries, and KOH-based electrolytes are preferable to NaOH due to their higher ion conductivity, lower viscosity, higher oxygen diffusion coefficient and faster reaction kinetics. [4] in which an Al–air battery using combination of the KOH alkaline electrolyte and the silver manganate nanoplate air cathode has recently been reported to be able to provide large specific cell capacities[61]. NaCl, a salt water electrolyte, has indeed been studied extensively for use as an Al–air battery electrolyte due to its abundance and safety wherein, if used, it could provide a battery potential of ~0.65–1.1 V for pure Al anodes. Previous studies also mentioned the effects of the concentration of NaCl and the operating temperature of the battery on the redox

potential of Al anodes [62]. However, studies have also proven that NaOH and KOH aqueous solutions may produce higher redox potentials in conjunction with Al anodes, which is one advantage of using alkaline solutions instead of salt water solutions (NaCl) [63]. In addition, researchers have shown that dendrite formation on anodes and by-product formation on cathodes can be suppressed by the use of acidic electrolytes such as hydrochloride and sulphuric acid [64]. Sadly, using acidic solutions may also lead to problems with the corrosion of al–air batteries.

Aqueous electrolytes face critical problems involving dendrite formation, electrode corrosion and the evolution of hydrogen gas. Corrosion and hydrogen gas inhibitors, in which the basic mechanism of corrosion inhibitors is the adsorption of inhibitor molecules on Al anode surfaces to suppress corrosion reactions, can be used to address these issues. On this basis, various electrolyte additives have been actively studied. Wang et al. [65] showed that the corrosion rate of Al anodes can be suppressed by mixing polyethylene glycol and ZnO additives with KOH aqueous electrolytes and Liu et al. [66] reported that the addition of CMC (carboxymethyl cellulose) and ZnO can also suppress the corrosion rate of Al anodes in which the self-corrosion of Al anodes in a NaOH alkaline electrolyte solution can be delayed. In this, these researchers suggested that the CMC carboxyl group could strongly bind to anode surfaces and interact with Zn^{2+} ions to enable the formation of a complex surface film of the anode, thereby stabilizing it. Studies have also reported that some inorganic additives may effectively prevent Al anode corrosion, although other studies have shown that inorganic chemicals such as chromates, vanadates, borates and hexafluoride silicates do not have significant inhibitory effects. [67]. In addition, organic additives can also effectively prevent the corrosion of Al anode [68]. Three dicarboxylic acids, including succinic acid, adipic acid and sebacic acid, were tested [69] as additives for alkaline electrolytes and found that these three acids reduced the evolution of hydrogen and confirmed that adipic acid was the most effective anode corrosion inhibitor. Studies have also proven that some aromatic carboxylic acids may also be effective in suppressing Al-corrosion in alkaline solutions by adsorption of carboxylic groups to Al surfaces and that similar effects may be observed in other metals under acidic conditions [70]. Furthermore, Mahmoud et al. [68] reported the influence of imidazole derivatives on the inhibition of Al corrosion in a 0.5 M HCl solution and suggested that the N atoms of the imidazole group can bind to Al surfaces to suppress corrosion. Interestingly, Wang et al. [71] suggested that amino acid and rare-earth metal complexes such as l-cysteine and cerium nitrate can also suppress Al anode corrosion and hydrogen gas evolution by forming a complex film

on anode surfaces in a 4 M NaOH alkaline solution and Wang et al. [72] applied a KOH solution in water with methanol as an electrolyte for comparison and found that the methanolic KOH solution delayed anode corrosion and that corrosion rates increased as the solution water content increased. Kang et al.[73] also reported that calcium oxide and l-aspartic can suppress the corrosion of Al anodes in NaOH alkaline media. Overall, all of these results suggest that various types of Al corrosion and hydrogen gas evolution inhibitors exist and can have practical use. More recently, Hopkins et al. [74] also created a new Al–air battery system using non-conducting oil instead of a liquid electrolyte during battery standby and reported significant Al anode corrosion suppression effects.

2.4.2 Anode Material

Naturally, pure aluminum has been chosen as an anodic material for Al-air batteries in virtue of its excellent electrochemical properties. Thermodynamically, a pure aluminum anode exhibits a potential of -1.66 V (vs. Hg/HgO) in saline and -2.35 V (vs. Hg/HgO) in aqueous solution. However, the practical open-circuit potential of the aluminum electrode is significantly higher, which is attributed to the competitions between the considerable electrode processes that occur on the Al surface including A.A. Mohamad[75], [76]:

- (i) the formation and/or dissolution of an initial Al_2O_3 and subsequent $\text{Al}(\text{OH})_3$ layer
- (ii) a three-electron charge transfer process producing Al^{3+} species
- (iii) the formation of corrosion products, $\text{Al}(\text{OH})_4^-$ and $\text{Al}(\text{OH})_3$
- (iv) a parasitic corrosion reaction on the aluminum surface releases hydrogen[77]

This side reaction causes corrosion and passivation of the aluminum surface, ultimately leading to the failure of Al-air batteries. A major problem with Al-air batteries is the thermodynamically favorable protective oxide film that forms spontaneously on the aluminum surface when exposed to air and aqueous solutions. This surface passivation leads to a positive shift of the corrosion potential of the aluminum electrode and a considerable slow-down of aluminum activation[78]. The self-corrosion is detrimental to the capacity of Al-air batteries and can reduce the discharge efficiency. Thus, many studies have focused on reducing the rates of corrosion and hydrogen evolution. Recently, the battery performance of 2N5 commercial grade Al with 99.5% purity has been studied in 4 M NaOH electrolyte. The performance of 2N5 Al in Al-air battery was found to be inferior to that of 4 N high pure (99.99%) Al due to the complex impurity layer reducing the average discharge voltage[79]. Lu et al. [80] investigated the electrochemical properties and battery performance of polycrystalline Al, Al

(001), (110) and (111) single crystals. The study revealed that Al (001) single crystals displayed lower corrosion rate and higher capacity density due to the low surface energy. However, high rate of corrosion and parasitic hydrogen evolution on the surface of the pure aluminum make it infeasible for application of Al-air batteries as energy sources.

As pure aluminum is unstable when used as an anode for Al-air batteries, the most common method to prolonging the battery operation time and decreasing the corrosion rate is through the use of Al alloys. A considerable number of alloying elements such as Ga, Tl, In, Sn, Zn, Bi, Mn and Mg have been adopted. The outstanding performance of Al alloys in Al-air batteries can be attributed to the comprehensive effect of each individual alloying component. At present, the most commonly used anode materials in Al-air batteries are Al-Zn, Al-In, Al-Ga and Al-Sn [81]–[85] alloys. Zinc (Zn) is well known to diminish hydrogen evolution on Al anodes by enhancing the HER potential and thus relieving the anode degradation [86]. Indium (In) is responsible for a positive shift of the anode potential and an increase of the hydrogen evolution overpotential [87]. In addition, gallium (Ga) has been proven to limit the oxide film passivation in chloride solutions by activating the surface sites of aluminum [88]. Furthermore, tin (Sn) can increase the dissolution rate of Al in aqueous solutions and reduce the corrosion rate. The corrosion behavior and performance of Al-0.5Mg-0.1Sn-0.02In-0.1Si and Al-0.5Mg-0.1Sn-0.02Ga-0.1Si alloys in Al-air batteries were investigated in 2 M NaCl and 4 M NaOH[89]. The results indicate that the Al-0.5Mg-0.1Sn-0.02Ga-0.1Si alloy exhibits a better electrochemical performance in the 2 M NaCl solution, while the Al-0.5Mg-0.1Sn-0.02In-0.1Si alloy shows a better electrochemical performance in the 4 M NaOH solution, this is due to the addition of Ga which can reduce the self-corrosion rate of Al-0.5Mg-0.1Sn-0.02Ga-0.1Si alloy in the NaCl solution and the addition of In can remove the passive film on the Al-0.5Mg-0.1Sn-0.02In-0.1Si alloy by the alkali. The addition of Si as an alloying component to Al-

0.5Mg-0.1Sn decreases the self-corrosion rate and increases the anode utilization. Boughrara et al. [90] studied the corrosion behavior of Al, Al-Zn and Al-Zn-Sn in 3 wt.% NaCl solution. The improved activity of Al-Zn and Al-Zn-Sn can be attributed to an inductive behavior of the intermediates leading to intergranular corrosion. Furthermore, the corrosion rate and discharge activity of Mg-6%Al-3%Zn and Mg-6%Al-3%Zn-(1%, 1.5%, 2%) In alloys were also investigated [91]. The Al-In alloy exhibited a lower self-corrosion rate and higher anodic efficiency than pure Al. These types of sacrificial anodes based on aluminum alloys are preferred in Al-air batteries due to the alloying element's ability to remove the passivating layer on the aluminum surface.

2.4.3 Air Cathode

The air cathode is one of the essential components of an Al-air battery, which is generally composed of a gas diffusion layer, current collector, and catalytic active layer. The gas diffusion layer is composed of a carbon material and a hydrophobic binder such as polytetrafluoroethylene (PTFE), making the diffusion layer permeable only to air and preventing the permeation of water [92]. Current collectors are typically made of a Ni metal mesh which can be connected to the external circuit and enhance the electron transfer processes [93]. The catalytic active layer consists of an electrocatalyst, carbon material, and binder, and is the location where the oxygen reduction reaction (ORR) takes place [94]. With regards to the air electrode, the sluggish efficiency of the oxygen reduction reaction is the critical barrier for the application of workable Al-air batteries. Additionally, there are other problems leading to the inefficiency of the air electrode. *Table 2* exhibits the related problems of air electrodes and the solutions to each aspect.

Table 2: Overview of air electrode related problems and solutions.

Problems	Cause	Effect	Proposed Solution
Sluggish oxygen reactions	High overpotential required for driving oxygen reduction reaction (ORR) and oxygen evolution reaction (OER)[93]	Limited energy/power density and energy efficiency	Explore efficient, durable electrocatalysts [94]–[96]
Air electrode flooding	Electrolyte penetration into the pores of the air cathode	Reduced oxygen accessibility	Treat carbon electrode with wax [97]; Proper optimization of the gas diffusion layer (GDL) [98]
Carbonate precipitation	Atmospheric CO ₂ reacts with alkaline electrolyte, producing carbonate precipitates	Decreased electrolyte conductivity and air cathode activity	Supply pure O ₂ to reduce CO ₂ concentration [99]
Electrolyte drying out	Water evaporation to open air	Reduced battery life	Proper optimization of the gas diffusion layer (GDL) [99]

The electrocatalyst in the air electrode plays an important role in determining the electrode performance and maximization of energy density. However, the reaction kinetics of the ORR is often sluggish, and occurs with a large overpotential [100]. Thus, one of the key opportunities to improving the ORR efficiency and reduce the overpotential is to explore suitable electrocatalysts. A diverse selection of electrocatalytic materials have been utilized as cathode catalysts, including precious metals and alloys, transition metal oxides/chalcogenides, metal

macrocyclic compounds and carbonaceous materials[101]. In spite of the previous general reviews on the ORR electrocatalysts [102]–[108]. Recently attempts have been made to fabricate novel Pt-based core-shell nanomaterials as highly efficient catalysts[109]–[111] to maximize the exposed catalytical active sites. The core-shell structure is designed to bring catalysts both high activity and stability for metal-air systems by tuning the electronic structure and lattice parameters of Pt shell layers. For Pt-based alloy systems, it is necessary to develop novel synthetic approaches for PtM nano catalysts with controllable composition, shape, and size. Despite extensive research in exploring non-Pt electrocatalysts due to the high cost of Pt, Pt based electrocatalysts are still the most promising nanomaterials with superior catalytic performance in metal-air batteries. Although Pt has been regarded as the best ORR catalyst, the commercial market of Al-air batteries may still be hampered by the limited resources, high cost and unsatisfactory durability of the conventional Pt-based catalysts. Thus, searching for available alternatives to substitute Pt by efficient nonprecious metal catalysts has obtained intensive interest.

2.4.4 Cathode Catalyst

Non-precious catalysts such as transition metal oxides [112], spinel-type metal oxides[113], perovskite[113], metal-N complexes on carbon matrixes[114], and carbonaceous materials for ORR catalysis in metal-air batteries are discussed in this sections. Among the extensive research efforts dedicated to developing advanced ORR electrocatalysts, carbonaceous nanomaterials have been demonstrated as promising metal-free catalysts with satisfactory activity and durability towards ORR. On one hand, the carbonaceous catalysts have relatively wide stabilization potential windows and outstanding electronic conductivity. On the other hand, the attractiveness also depends on the structural diversity, including zero-dimensional (0D) fullerene, one-dimensional (1D) carbon nanotubes, two-dimensional (2D) graphene and various three-dimensional (3D) nanostructured carbon materials. Carbon nanomaterials, including graphite, graphene, and carbon nanotube, etc. are usually utilized as catalysts or supports due to their high electronic conductivity, large specific surface area, environmental acceptability and corrosion resistance[115]–[118]. Dai et al. [119] supplied an exhaustive review on nanocarbons materials and their recent development and challenges of metal-free catalysts based on intermolecular charge-spin redistribution for ORR/OER/HER and fuel cell. To avoid duplicating the present reviews, this section of this review will mainly focus on reviewing the most recent progresses on graphene-based and CNT-based nanocomposites, heteroatom doped carbon nanostructures, three-dimensional (3D) carbon nanomaterials, and

carbon quantum dots as electrocatalysts for ORR. Graphene, the one-atom-thick planar sheets of sp²-bonded carbon atoms that are densely packed in a honeycomb crystal lattice, is rapidly being explored as an alternative material for ORR catalyst supports. It has the following noteworthy advantages[116]:

- (1) The flexibility and immobilization of graphene nanosheets can provide a large space to accommodate the catalysts, and prevent their agglomeration;
- (2) The good superficial characteristics of graphene increase the solid-air contact efficiency, resulting in a large amount of oxygen adsorption on graphene;
- (3) The structure of graphene strengthens its electrical conductivity and the electron transfer rate on the surface of graphene;
- (4) The structural defects on single layered graphene provide more active sites to stimulate the electrocatalytic activity.

In recent years, significant improvements have been achieved in the extensive studies of low-Pt or non-noble metal nanomaterials dispersed on graphene with low-cost, excellent electrocatalytic activity, high stability and methanol tolerance [116], [120]–[124]. Dai et al. [125] reported an advanced bifunctional N-doped graphene supported Co₉S₈ nanoparticles (Co₉S₈/graphene) as an electrocatalyst for ORR and OER prepared by simultaneously etching and N-doping with NH₃-plasma treatment. The hybrid shows excellent ORR and OER activity, which is close to that of the commercial Pt/C catalyst. The cobaltite oxide nanosheets/graphene hybrid with a face centered cubic crystalline structure is prepared and developed as an effective cathode catalyst in high-performance Li-air batteries[126]. The excellent electrocatalytic performance of the Co₃O₄/rGO hybrid is attributed to the rapid electron transport kinetics and high electrocatalytic activity for ORR from the synergetic effects of cubic Co₃O₄ nanosheets and graphene. Bag et al. successfully prepared the Mn₃O₄/N-rGO hybrid composed of nitrogen doped reduced graphene oxide (N-rGO) and Mn₃O₄ by the one-step in-situ reduction using hydrazine[127], and the hybrid material exhibits high electrocatalytic activity towards ORR under alkaline condition and favors the four-electron pathway for ORR. The synergistic effect of N-rGO and Mn₃O₄ enhances the overall performance of the hybrid catalyst.

For practical applications, 3D nanostructured cathode materials having a large specific surface area, good mechanical integrity and facilitated ion diffusion/charge transfer paths are highly desirable. M. Pumera et al. [128] showed that mechanically rigid 3D graphene deposited on nickel template can act as efficient catalyst for ORR. Huang and co-workers[129] developed

novel 3D sulfur-nitrogen co-doped carbon foams with hierarchical pore structures. Such excellent catalytic activity and stability outperforming commercial Pt/C catalyst can be attributed to the synergistic effect and high electron transfer supplied by 3D continuous networks. Li et al. [130] reported similar work on the nitrogen and phosphorus dual-doped porous carbon foams as efficient electrocatalysts for ORR in basic, neutral, and acidic media, which exhibit better catalytic activity and much higher stability than those of the commercial Pt/C catalyst. N, S co-doped porous carbon synthesized from ordered mesoporous carbon and trace ionic liquids (ILs) as an ORR catalyst exhibits comparable catalytic activity and better stability than Pt/C catalyst in alkaline solution [131]. Additionally, Wong et al. [132] demonstrated that 3D nitrogen-doped graphene prepared by pyrolysis of graphene oxide with polypyrrole displayed excellent catalytic activity for ORR and OER and high stability compared with the commercial Pt/C catalyst. 3D B, N co-doped graphene foams were prepared by a modified chemical vapor deposition (CVD) method as highly efficient ORR catalysts, which exhibited much better than their undoped counterparts [133]. In another paper, an erasable-promoter-assisted hydrothermal method coupled with pyrolysis was developed to prepare N-CNT aerogels, which exhibited remarkable ORR activity in alkaline medium with a four-electron transfer pathway[134]. As can be seen from the above discussions, heteroatom doping of 3D carbon nanomaterials has been verified as a simple and effective approach to enhance the electrocatalytic activity for ORR. Furthermore, 3D nanocarbon has also been considered as an alternative support to enhance electrocatalytic performance. Sun et al. [135] reported a spinel ZnCo_2O_4 /Ndoped carbon nanotubes (ZnCo_2O_4 /NCNT) composite which showed highly efficient catalytic activity via a four-electron pathway in alkaline solution, and superior durability over commercial Pt/C catalyst.

CHAPTER 3

METHODOLOGY

In this chapter, experimental setup and analysis has been conducted to verify the effectiveness of the performance of mixture of rice husk and activated carbon inside a cell to produce electricity. Experimental set up and procedure of procuring an Al-Air battery along with graphite rod replacement of AA battery have been discussed briefly in this section.

3.1 COLLECTING ACTIVATED CARBON

Activated Carbon is a very common material. The beauty of Activated Carbon (AC) is, it can be extracted from many abandoned sources. Use of activated carbon can be seen in, reducing air pollution [136], Arsenic removal [137], cleaning vegetables and fruits[138], water treatment plants [139], Groundwater treatment [140], inhouse purification of drinking water [141], effluent gas streams purification [142], mercury vapors removal [143], NO_x and SO_x removal [144], phenols and phenolic compounds removal [145][146], etc.

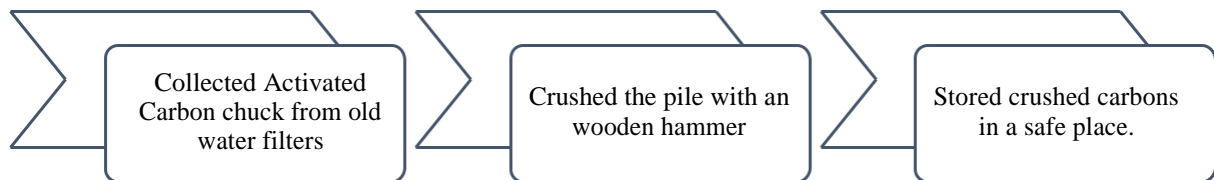


Figure 1: Flow Chart of Activated Carbon Collection

Waste activated carbon can easily be collected from water filters. They have the surface area of at least 1000m² for one gram weight [147]. According to the EPA, activated carbon is the only material that filters all 12 herbicides which are identified and 14 pesticide; and also, 32 identified organic contaminants[148]. That is why activated carbon is vastly used in water treatment process. *Figure 1* represents the process chart of the procedure of activated carbon collection from old water filters.



Figure 2: Activated Carbon (AC)

At first, we collected the chunk of activated carbon from old filter. Then we crushed it using a wooden hammer, as they were in bonded form. After the collecting, they were stored safely in a zip lock bag till further use. *Figure 2* shows raw activated carbon collected from the source.

3.2 BURNING RICE HUSK

RHA may be created by burning rice husk either in open field or under any special temperature-controlled incineration conditions. The open burning production of RHA has a high carbon content which has adversely affected the properties of the concrete and has also caused a highly crystalline form in the structures. Researchers have used various incineration processes [149]–[152]. RHA quality is controlled by an amorphous incineration process, which is essential for structural concrete [153]. Based on some study [154]–[156], it was found that the highest amorphous silica could be produced by burning the rice husk at temperatures between 500 and 700 °C and that the specific surface area up to 150 m²/g would be at the maximum temperature.



Figure 3: Burned Rice Husk (BRH)

3.3 PREPARING MIXTURES

After collecting activated carbon and rice husk ash, we used a precise digital weight machine to measure required amount of mixture. The machine is capable of measuring 0.01g to 500g and it has the function of automatic calibration. The values of the measurement taken is given in Table 3. Total weight of the mixture was 1g.

Table 3: Sample name with different mixture proportion

Sample Name	% of RHA	% of AC	Weight of RHA	Weight of AC
0RHA100AC	0%	100%	0g	1g
5RHA95AC	5%	95%	0.05g	0.95g
10RHA90AC	10%	90%	0.1g	0.9g
20RHA80AC	20%	80%	0.2g	0.8g
30RHA70AC	30%	70%	0.3g	0.7g
40RHA60AC	40%	60%	0.4g	0.6g
50RHA50AC	50%	50%	0.5g	0.5g
60RHA40AC	60%	30%	0.6g	0.4g
70RHA30AC	70%	30%	0.7g	0.3g
80RHA20AC	80%	20%	0.8g	0.2g
90RHA10AC	90%	10%	0.9g	0.1g
100RHA0AC	100%	0%	1g	0g

In this *Figure 4*, samples of raw materials, which is the mixture of AC & RHA are shown.

These samples are of different ratios of RHA & AC and each sample contains 1 gm of mixture.

We can see from the figure different amounts of raw materials were measured for each sample.



Figure 4: Samples after measuring

3.4 EXPERIMENTAL SETUP

3.4.1 Al-Air Cell

We collected used aluminum foil, tissue paper, wire, salt water. Then we cut off the aluminum foil and tissue paper 10 cm square block. In a single unit cell, we used 1 aluminum foil and 2 tissue paper and a single piece of copper wire. *Figure 5* shows the raw materials needed in this experiment.

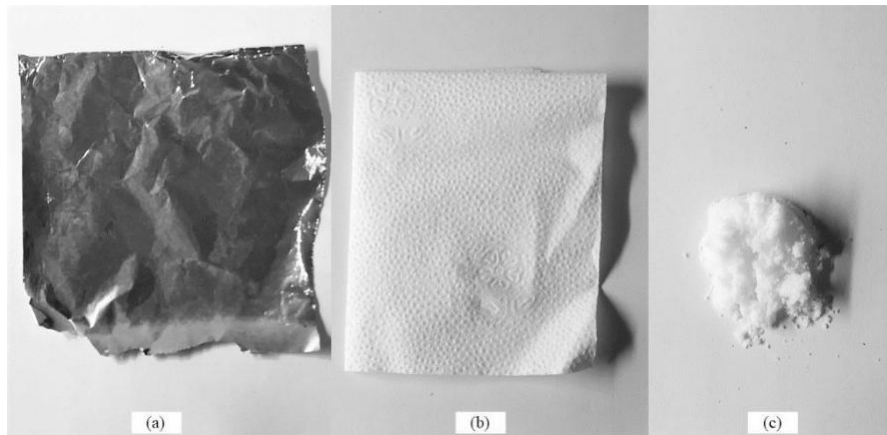
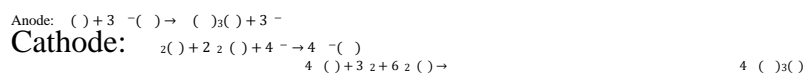


Figure 5: Raw materials for Al-Air cells

We used salt water mixture as an ion exchanger. This solution was made using 200ml water and 20g of common salt (NaCl).



This type of DC Cells relies on Reduction and Oxidation reactions. Oxidation of Aluminum (Al) happens at the anode and reduction of Oxygen (O₂) occurs at the cathode; which, generate electrical energy. The half and overall reactions equations are given below,



We began the experiment by cleaning every required material including the place of the experiment. We placed a tissue paper over an aluminum foil. Then we evenly distributed 5ml of salt water over the tissue paper. We sprinkled the activated carbon and rice husk ash mixture evenly on the wet tissue. We placed the copper wire 5cm from the corner.

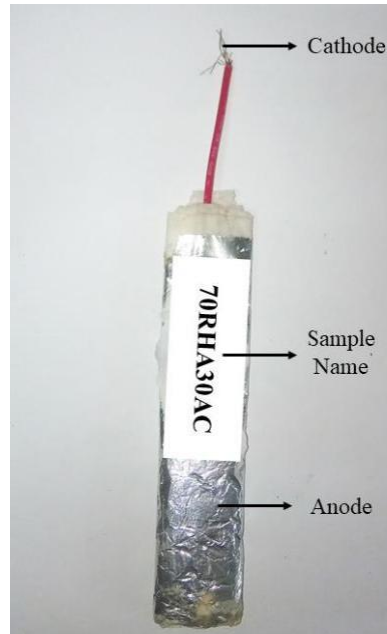


Figure 6: Structure of an Al-Air Cell

One end of the wire was revealed outside which acted as a cathode junction. Then we put another tissue paper over it and evenly distributed 5ml of salt water mixture again. Finally, we convoluted the whole set up.



Figure 7: All Al-Air cell samples

Figure 6 represents a sample, in which Anode and Cathode of the cell is shown. And in the Figure 7, all the samples are shown, which were made in the same method with different cathode catalyst mixtures.

3.4.2 Graphite Cathode Replacement

The experiment was initialized after the industrial made battery was arranged from local shop. Activated carbon was taken from used water filter; rice husk ash was collected as waste. After collecting the required components test were run on it.



Figure 8: AA Battery after cathode replacement

Voltage reading of the industrial battery (primary cells) was taken with a multimeter at initial condition. Then the battery was disassembled, the plastic covering was taken off along with the plates on both ends. *Figure 8* represents a cell after the replacement of graphite cathode.

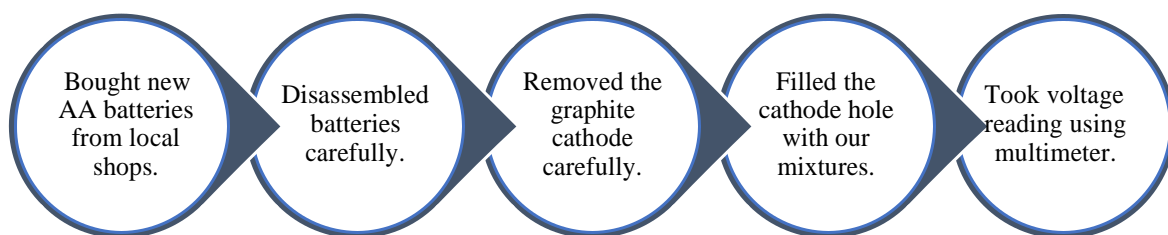


Figure 9: Steps of graphite cathode replacement

The cathode rod was taken off using a narrow plyer carefully so that apparatus is not damaged in anyway. Activated carbon and rice husk ash mixture was used instead of carbon rod. The mixture was inserted in place of the rod with required pressure. Eleven samples were made with different ratio of activated carbon and rice husk ash. Voltage readings were taken carefully for different samples after a specific interval of time using a multimeter. *Figure 9* shows a complete flow chart of the whole procedure.

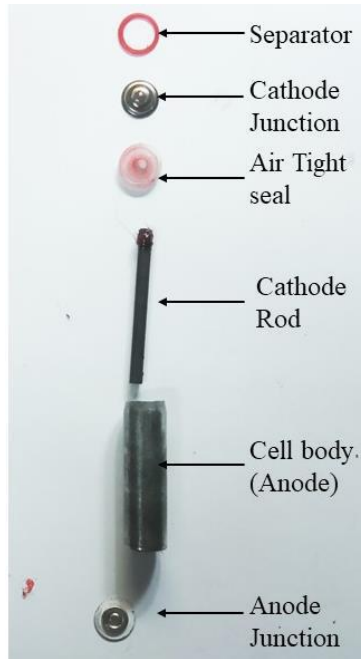


Figure 10: Disassembled AA cell

Figure 10 is the disassembled photograph of the cell. In a disassembled cell there were components like separator, cathode junction, an air tight seal to save internal components, a graphite cathode rod, anode cell body and an anode junction.



Figure 11: All samples with graphite replaced

The picture of all prototypes in Figure 11 is the picture taken after implementing the same method on eleven different cells for each sample. In the prototypes separators and seals were not used. And the graphite rod was replaced by our mixture.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 RESULTS

In this section, the experimental results and their evaluation is discussed. Also, many figures are included in this part of thesis, which is explained individually. In the last section, there is a final discussion part which concluded the obtained results from the experiment.

4.1.1 Aluminum-Air Cell

After preparing the samples, we collected voltage reading from each sample using multimeter. We collected reading for the first seven days to analyze the voltage characteristics of the foil cell samples. *Table 4* shows the first day voltage of each sample along with Rice Husk Ash and Activated Carbon percentage.

Table 4: Voltage Reading of Al-Air Cell at Day 1

Sample Name	Percentage (%)		Voltage (V)
	Rice Husk Ash (RHA)	Activated Carbon (AC)	
0RHA100AC	0	100	0.72
10RHA90AC	10	90	0.71
20RHA80AC	20	80	0.64
30RHA70AC	30	70	0.61
40RHA60AC	40	60	0.63
50RHA50AC	50	50	0.63
60RHA40AC	60	40	0.62
70RHA30AC	70	30	0.62
80RHA20AC	80	20	0.6
90RHA10AC	90	10	0.6
100RHA0AC	100	0	0.55

Table 4 also shows that, the increasing percentage of rice husk content results in reduction of voltage output. From the sample 30RHA70AC, the voltage reading starts to fluctuate heavily; So, average values were taken. Sample 0RHA100AC to 20RHA70AC (*Table 4*) were quite stable and ideal for constant voltage supply.

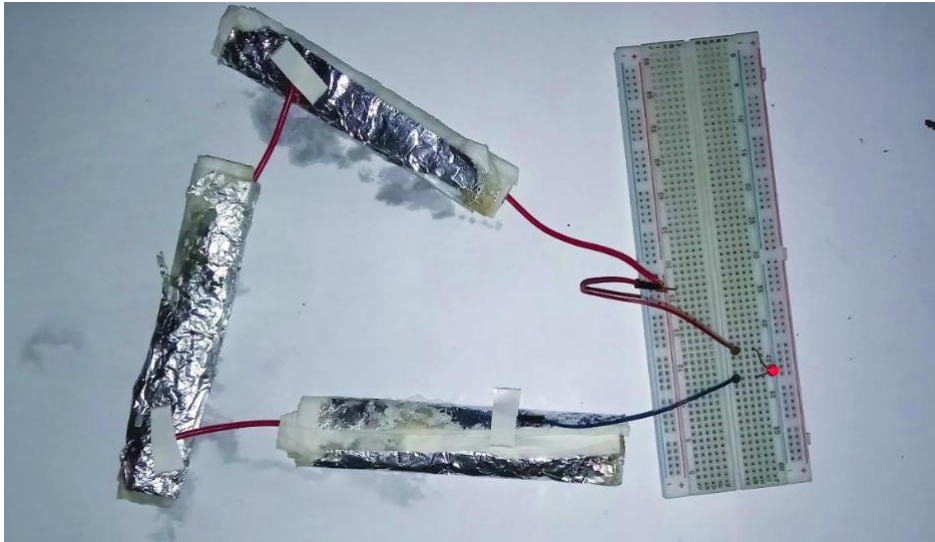


Figure 12: Series connection of Al-Air cells

We series connected three cells to form a battery, which gave us around 2 volt and it was enough to lit a 3mm Red LED (Figure 12). After pressing the cells, it produced little more voltage and more current was flowing; because of this the LED was glowing more than before.

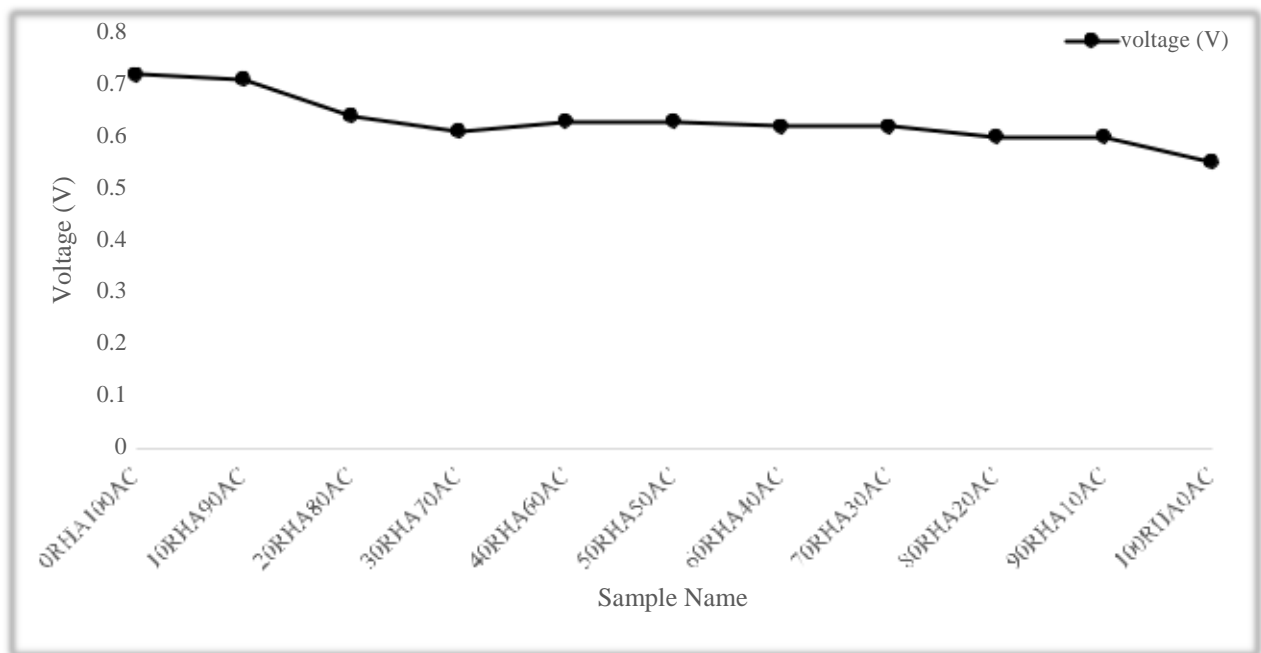


Figure 13: Voltage reading of Day 1 for the Al-Air cells

From Figure 13, we can see that voltage range of the samples goes from 0.72V to 0.55V at the first day of reading. Minimum voltage is obtained from 100RHA0AC sample.

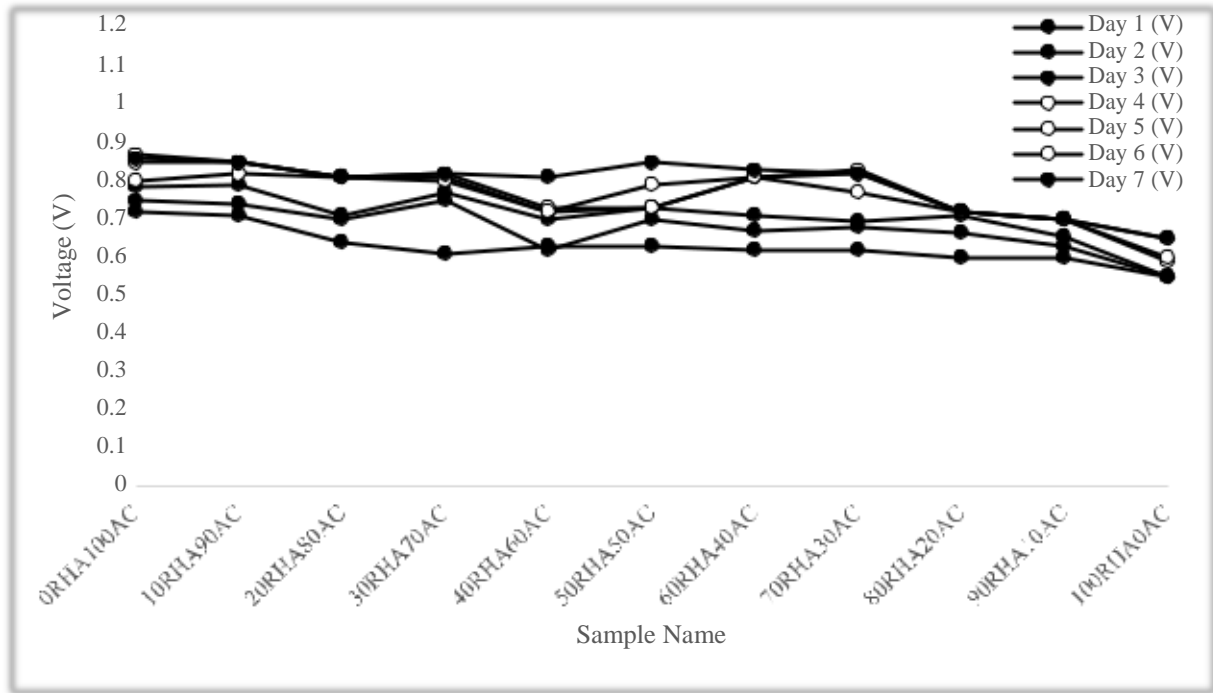


Figure 14: Voltage reading of consecutive 7 days for Al-Air cells

Figure 14 shows the change of voltage of each sample over seven days. It represents that, the changes of voltage over the days are not constant. Voltage outputs have drastically increased at Day 3 and Day 4 as the tissue paper adsorbs the electrolyte (NaCl) properly overtime and rate of increment reduces after that. After Day 3, voltage increases 8 – 20% for ten samples.

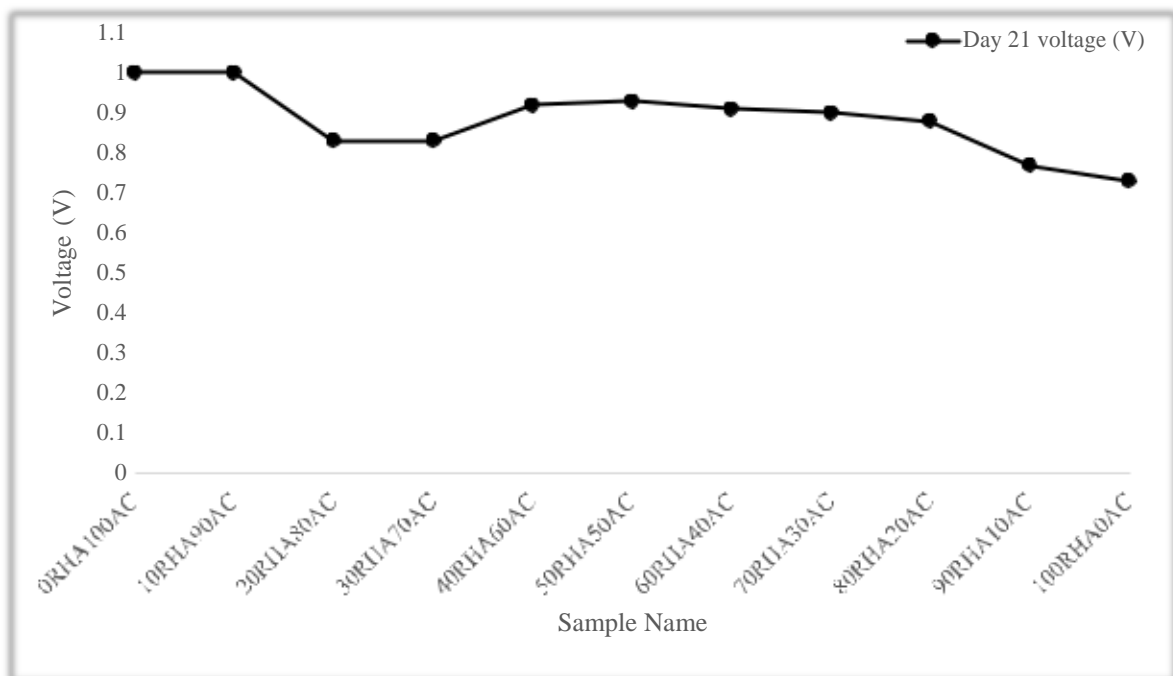


Figure 15: Voltage reading of Al-Air cell after 21 days

100RHA0AC sample is an exception as it remains constant for the first 3 days. The average increment of voltage after Day 3 is around 14%. Fluctuation of voltage reduces with time and

the dry cell samples become more stable. We have taken minimum 0.8 volt as a standard of usable sample and from the readings we suggest that from 0RHA100AC to 30RHA70AC samples are usable after 4 days of procurement.

We have also tested the durability of the samples. We have taken voltage reading after 21 days and found the samples functioning. *Figure 15* represents the voltage reading from the multimeter after 21 days. Though it shows more voltage, it was dry. This time, we needed to press the cells as before. The salt water we used as ion-exchanger was fully evaporated. As a result, flow of electron was stopped and no current was provided by the cells.

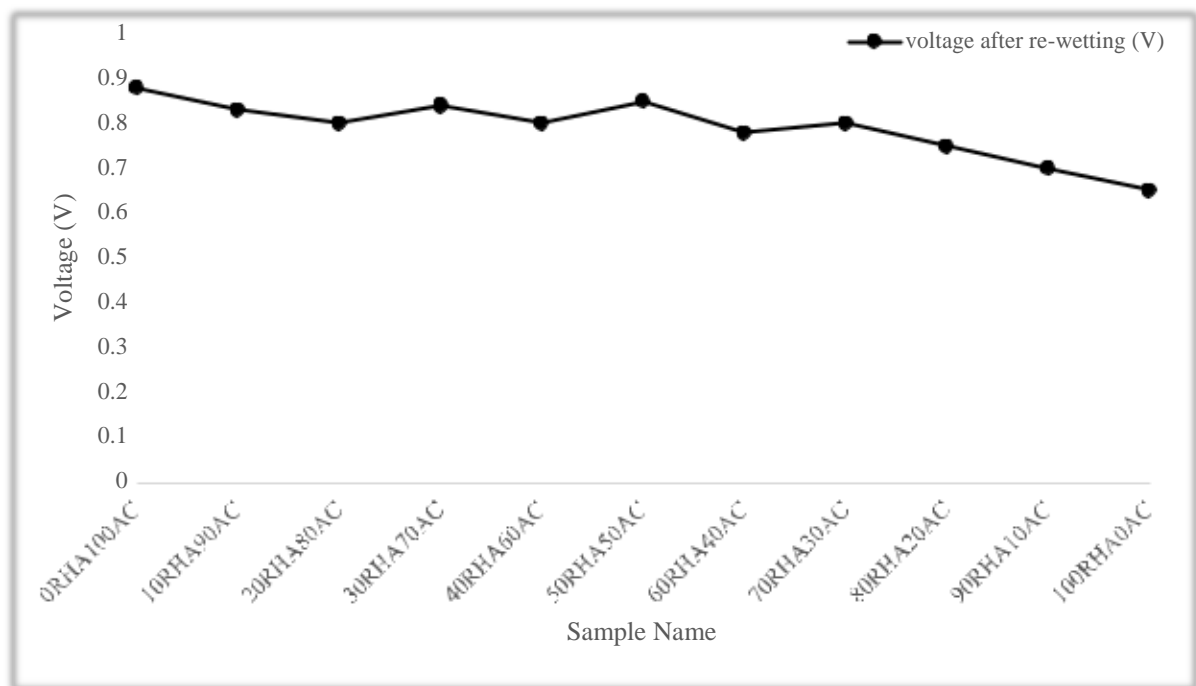


Figure 16: Voltage reading of Al-Air cells after re -wetting

After that, we added water in the cells and stored it shadowed area for 24 hours. On the following day, voltage reading was taken again, which is shown in *Figure 16*. Voltage has dropped compared to day 21, but it was supplying enough current again.

4.1.2 Graphite Cathode Replacement

In this part of the study, an experiment was done by replacing the graphite rod of AA battery to find out if it works or not. In this section, the result of that experiment is discussed.

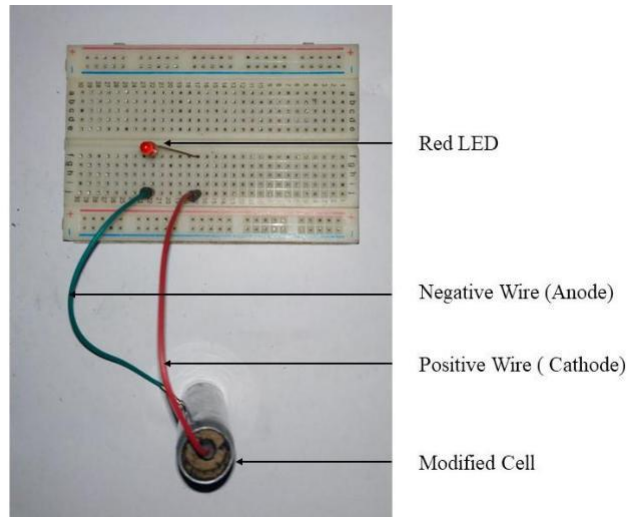


Figure 17: Current flow check after graphite rod replacement

After finishing the procurement of the prototypes, voltage reading was taken from each one using multimeter. *Figure 17* ensured the flow of electron in the circuit. *Table 5* shows the voltage of each sample along with Rice Husk Ash and Activated Carbon percentage.

Table 5: Voltage of AA batteries before and after graphite rod replacement

Sample Name	Percentage (%)		Voltage (v)		Voltage Drop (v)
	Rice Husk Ash (RHA)	Activated Carbon (AC)	Before replacing	After replacing	
0RHA100AC	0	100	1.683	1.682	0.001
10RHA90AC	10	90	1.68	1.673	0.007
20RHA80AC	20	80	1.687	1.663	0.024
30RHA70AC	30	70	1.686	1.66	0.026
40RHA60AC	40	60	1.68	1.51	0.17
50RHA50AC	50	50	1.685	1.52	0.165
60RHA40AC	60	40	1.685	1.2	0.485
70RHA30AC	70	30	1.685	1.00	0.685
80RHA20AC	80	20	1.684	0.8	0.884
90RHA10AC	90	10	1.683	0.71	0.973
100RHA0AC	100	0	1.685	0.7	0.985

Figure 18 shows that, the increase of rice husk in samples results in voltage reduction. The sample 30RHA70AC and afterwards, the voltage starts to fluctuate heavily. In that case, average voltage was considered. Sample 0RHA100AC to 20RHA70AC (Table 2) were quite stable and ideal for constant voltage supply.

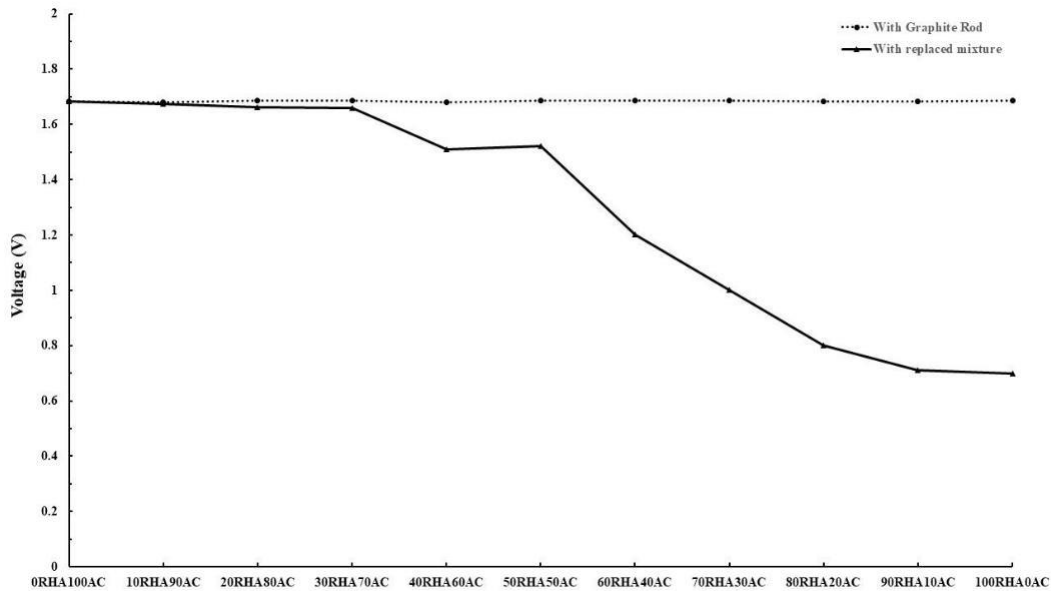


Figure 18: Voltage comparison of AA batteries before and after graphite rod replacement

From Table 5 and Figure 18 it can be concluded that, for 100% activated carbon there is none to negligible voltage drop. And for 10RHA90AC, the voltage drop is negligible. And, if we set a benchmark of 1.6V for these cells, then 30RHA70AC is also useable.

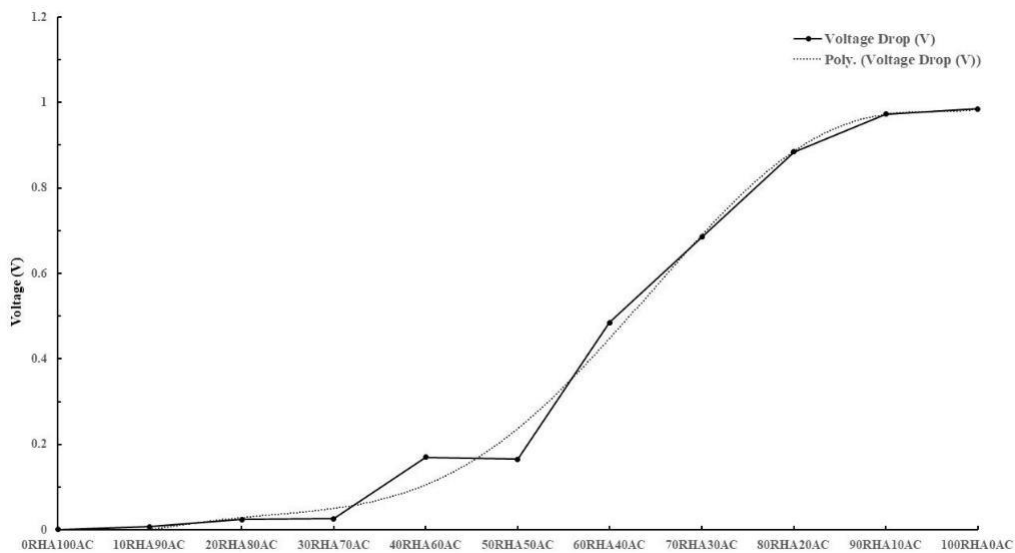


Figure 19: Voltage drop and its trend line after graphite rod replacement

4.2 DISCUSSION

As discussed before, our study actually consists of two projects - Al-air cell with rice husk ash and activated carbon mixture and replacement of graphite cathode with the same mixture. Methodology and results are already discussed. Prototypes of our model were examined to determine their characteristics. The result has been satisfactory. Now, we are going to analyze the results.

First, we worked with aluminum foil cell and analyzed its result. We found some inconsistent results and fluctuating values. Though we performed the experiment carefully, human error might be present. Aluminum foil quality might be deteriorated, number turns of each cell might not be same, NaCl solution amount might slightly differ cell to cell. There are many potential sectors that can be further studied to make a conclusive decision. We suggest that, durability test should be performed with loads. According to our study we found consistent result up-to the mixture containing 30 percent rice husk and 70 percent activated carbon. More samples from 0 to 30 percent rice husk should be studied to get a finer curve. The best result from this experiment is gained after re-wetting the cells on 21st day. This study provides insights on potential of easily procurable, cheap and eco-friendly DC cells from waste.

We also replaced the graphite cathode of a primary DC cell AC-RHA mixture. We have successfully used waste rice husk and recycled activated carbon to procure DC Cells. It is possible to replace the graphite cathode with recycled activated carbon in primary cells. Mixtures containing more than 30% Rice Husk Ash make the voltage unstable. So, it is suggested to use 10% RHA for best result and not more than 30% RHA for stability.

Our objective was to utilize and recycle waste in DC cells and produce energy. We procured two prototypes and our result and analysis show that, they are functional and effective. We faced some challenges and limitations in this study which need to be mitigated. There are also scope of future study and development. These will be discussed in the next chapter of the book.

CHAPTER 5

CONCLUSION AND SUGGATION

Throughout the whole experiment, we tried to be as cautious as possible. There have been many challenges in the whole procedure. Many of the challenges were overcome but some of them could not be mitigated. We are going to discuss the challenges we faced. In this study, obtained result was quite satisfactory to achieve our objectives. We have successfully used waste rice husk and recycled activated carbon to procure DC Cells.

After the experiment, it can be concluded that, it is possible to replace the graphite cathode with recycled activated carbon in primary cells. Mixture of activated carbon and rice husk ash were also functional as the replacement of graphite cathode. Voltage drop is between only 0.5-1.5% till 30% mixture of rice husk ash. Mixtures containing more than 50% Rice Husk Ash makes the voltage unstable.

Using the mixture of Activated Carbon and Rice Husk Ash in Al-Air batteries as cathode catalyst has been found functional as well. Our result goes as per with the standard average reading of this type of Al-air cell. Implementation of this mixture will reduce the manufacturing cost, which is one of the biggest problems for this type of cells. Mixtures containing more than 30% Rice Husk Ash makes the voltage unstable.

In this book, we provided detailed overview of our study and methodology. We analyzed and discussed the results and if they are suitable with our objectives. We discussed the challenges we faced and how to overcome them in future studies. Future potential of the study is also discussed. This project should be further studied and developed for the purpose of better waste management and energy harvesting.

5.1 CHALLENGES AND MITIGATION OF LIMITATIONS

After collecting the activated carbon, there was not any suitable equipment available for grinding them properly. We used hand tools to crush the activated carbon. We also could not procure the external body of the DC cells due to lack of equipment and materials. Collecting recyclable aluminum foil, tissue paper was also a challenge. We had to be cautious while choosing the raw materials. Rice husk ash was also needed to be put in a suitable storage to maintain suitable moisture

During cathode replacement of a primary DC cell, two challenges arose; making the dry cell cathode and putting it inside the battery. Since, any binder or thermosetting resin or pitch was not available we could not procure a rigid cathode body. The mixture was measured and then pushed inside to use it as a cathode.

5.2 SCOPE OF IMPROVEMENTS

As we mentioned in the challenges, some of the challenges could not be overcome which needs to be overcome in the next study. Activated carbon needs to be crushed properly. Cathode consisting of AC-RHA mixture should be procured using a suitable binder. External body of a primary DC cell be procured and used with the prototype.

There are many rooms for development as well. Durability test of all the prototypes should be carried out. Multiple prototypes for each mixture should be prepared and analyzed to understand the characteristics of our setups. Also, more sample of mixtures should be used to make the prototypes. A financial study should be done to take this project to industrial level.

Still some new challenges may arise in more studies. A scheduled and detailed development and research work of this models should be carried out to make this an alternative source of DC cells.

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