Effects of Lithium Sulfate and Zinc Sulfate Additives on the Cycle Life and Efficiency of Lead Acid Batteries

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ABSTRACT: The influence of lithium and zinc sulfate additives on the cycle life and efficiency of a 2 V/20 A H lead acid battery was investigated. Charging and discharging processes (cycle) were carried out separately for dilute sulfuric acid electrolyte, sulfuric acid–lithium sulfate electrolyte, and sulfuric acid–zinc sulfate electrolyte solutions for one (1) hour each. The voltage after 30 min of the charging process yielded 2.30 V for dilute HSO₄, 2.74 V for HSO₄ + LiSO₄, and 2.90 V for HSO₄ + ZnSO₄ solutions. A load of 6 V, 10.3 W incandescent lamp was applied, and after 30 min of the discharge process for each electrolyte, the voltage was 1.10 V for HSO₄, 1.90 V for HSO₄ + ZnSO₄, and 1.30 V for HSO₄ + LiSO₄. A calculated efficiency of 77% for HSO₄, 74% for HSO₄ + ZnSO₄, and 85% for HSO₄ + LiSO₄ solution was obtained. The cycle test is evidence that the addition of lithium sulfate salt improved the cycle life and efficiency of the 2 V/20 A H lead acid battery, while zinc sulfate offered no significant improvement. The cycle life of a battery increases with decrease in acid concentration, longer discharge time, and increase in efficiency.

1. INTRODUCTION

Lead acid batteries are the most popular secondary rechargeable batteries in the world with well-proven records of reliability. They are also of the best electrical energy storage systems. This is also a basis for distinguishing battery systems. It is an electrochemical device that has the potential of converting chemical energy into electrical energy. A normal lead acid battery consists of an anode, a cathode, an electrolyte, separators, and an external case. The materials used as the electrode and electrolyte determine the specific characteristics of lead acid battery systems. This is also a basis for distinguishing battery systems. The sulfuric acid electrolyte is an active material in the lead acid battery that is involved in the cell reactions. The concentration of such an electrolyte changes when the battery is discharged and charged. In lead acid batteries, the open circuit voltage is dependent on the concentration of the electrolyte according to the Nernst equation. The resistance and freezing point of such a battery are also dependent on the sulfuric acid concentration. Several studies have established the effects of high acid concentration and temperature on the cycle life of the lead acid battery. Some have tried to elucidate the fall in the battery cycle life on different basis. Despite these, manufacturers are still using high concentrations of sulfuric acid.

The significance of this research is to establish a cost-effective, durable, and less harmful lead acid battery for domestic and industrial use. The influence of lithium and zinc sulfate salts on the cycle life and efficiency of a 2 V/20 A H lead acid battery is used to investigate this. The charging and discharging cycles will be carried out separately on the dilute sulfuric acid electrolyte solution and on the two mixed electrolyte solutions.

This research work is aimed at enhancing the charge cycle and cathode stability of a lead acid battery using sulfuric acid–lithium and zinc sulfate salt as additives. Comparing the charging and discharging cycles of dilute sulfuric acid electrolyte solution and mixed (sulfate) electrolyte solution will be used ascertain if the salt additive yielded any significant improvement on the cycle life and efficiency of this 2 V/20 A H lead acid battery.

2. MATERIALS AND METHODS

2.1. Materials. For this study, a 2 V/20 A H refillable lead acid battery was used, which was sourced from the Arieria Market Aba in Nigeria, and 33 vol % of dilute sulfuric acid with specific gravity = 1.25 and lithium and zinc sulfate salt were used. A digital voltimeter was used for measuring the voltage, and a 6 V incandescent lamp acted as the load applied.

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basic lead acid chemical reaction in a sulfuric acid electrolyte, where the sulfate of the acid is part of the reaction, is as follows:

(Overall reaction)\(^6\)

\[
PbO_2 + 2Pb + 2H_2SO_4 \xrightarrow{\text{charge}} 2PbO_4 + 2H_2O
\]

(1)

The discharge reactions of materials are as follows,\(^6\)

Cathode reaction

\[
PbO_2 + HSO_4^- + 3H^+ + 2e^- \rightarrow PbO_4 + 2H_2O
\]

(2)

Anode reaction

\[
Pb + HSO_4^- \rightarrow PbSO_4 + H^+ + 2e^-
\]

(3)

For lithium sulfate

\[
PbO_{2(s)} + 2Li_{(aq)}^+ + 2e^- \rightleftharpoons Li_2PbO_{2(s)}
\]

(4)

For zinc sulfate

\[
PbO_{2(s)} + 2Zn_{(aq)}^{2+} + 2e^- \rightleftharpoons Zn_2PbO_{2(s)}
\]

(5)

There is no lead acid battery that is 100% efficient in storing power, but an 85% efficiency is a realistic mark. The efficiency of a battery is dependent on the rate of charging or discharging and the state of charge.\(^7\) The higher the rate of charge or discharge, the lower the efficiency.

\[
\text{Voltage efficiency} = \frac{\text{discharge voltage}}{\text{charge voltage}} \times 100
\]

(6)

When a voltage is supplied to a battery, which is greater than the battery’s voltage, current flows through the battery in the reverse direction to when it is supplying current and the battery will charge.\(^7\) The rate of charge or current that will flow is dependent on the difference between the battery’s voltage and that supplied to it. While it is significant to the life and performance of a lead acid battery to be fully charged regularly, it is also important to not continue charging it when it has been fully charged, as this could damage the battery.\(^8\) When a lead acid battery is discharged, there is formation of lead sulfate crystals at both the negative and positive terminals and electrons are also released. Over a period, the sulfate gradually
accumulates, crystalizes, and clogs the plates to a stage where the battery can no longer accept any charge. This process is called sulfation and only occurs during discharge which eventually leads to scrapping. The deeper the discharging of the battery, the higher the level of sulfation which is an insulating film that covers the plate. A lead acid battery that requires more time to fully discharge has a better life and performance than the one that takes less time to completely discharge.

2.1.1. Lithium Sulfate. From eq 4, 2 mol is required, and therefore, mass of lithium sulfate required is

\[ 2 \times 127.96 \text{ g} = 255.92 \text{ g} \]

Mass of lithium sulfate required in 400 mL of dilute H₂SO₄ is

\[ 255.92 \times 0.4 = 102.37 \text{ g} \]

2.1.2. Zinc Sulfate. From eq 5, 2 mol is required, and therefore, mass of zinc sulfate required is

\[ 1 \times 287.54 \text{ g} = 287.54 \text{ g} \]

Mass of zinc sulfate required in 400 mL of dil H₂SO₄ is

\[ 287.54 \times 0.4 = 115.00 \text{ g} \]

2.2. Procedure. 2.2.1. Charge and Discharge Cycle for Dilute Sulfuric Acid Electrolyte Solution. In the experiment, 400 mL of dilute sulfuric acid (33 Vol % specific gravity of 1.25) was measured with a weighing cylinder and gently poured into the empty refillable 2 V lead acid accumulator with the help of a funnel. The voltage was read and recorded as the charge voltage at time 0 min. Connection of the regulated 5 V DC power supply was carried out for beginning of the charging cycle. The charging voltage was read with a digital voltmeter and recorded after 5 min. This reading and recording of charging voltage were also performed after 10, 15, 20, 25, and 30 min of the charging cycle. The power source was disconnected, and the voltage was read and recorded as the discharge voltage at time 0 min. A 6 V incandescent lamp was connected at both terminals of the cell for the discharge cycle process. After a discharge time of 5, 10, 15, 20, 25, and 30 min, the discharge voltages were all read and recorded. A moving coil voltmeter was used to check the accuracy of the digital voltmeter in reading the voltages.

2.2.2. Charge and Discharge Cycle for Mixed Electrolyte Solution. After emptying and cleaning of the refillable lead acid...
accumulator with distilled water, another 400 mL of dilute sulfuric acid was measured and gently poured into the battery. 102.4 g of lithium sulfate salt was weighed with the electronic scale and gently poured into the battery with the help of a spatula. The voltage at this time was read with a digital voltmeter and recorded under time 0 min. The regulated 5 V DC power source was connected for starting of the charging cycle. After 5 min, the charging voltage was read with a digital voltmeter and recorded. This charging cycle continued until 30 min. The charging voltages at 10, 15, 20, 25, and 30 min were also read and recorded. At the end of the charging cycle, the power supply was disconnected and the voltage was read and recorded. The 6 V incandescent lamp was connected at the positive and negative terminals of the lead acid accumulator with the help of connecting wires. After 5 min of discharge, the voltage was read and recorded as the discharge voltage after 5 min and this discharge cycle was continued till the 30th minute. The respective discharge voltages at 10, 15, 20, 25, and 30 min were all read and recorded as the discharge voltages after those time intervals.

This process was repeated for sulfuric acid-zinc sulfate electrolyte solution with 115.00 g of zinc sulfate salt used for the same duration of cycle process. The molecular weight of the additive and the mole fraction in the reaction equation were the parameters that made up the mass of additive used. A moving coil voltmeter was used to determine the accuracy of the digital voltmeter in reading the charge and discharge voltages.

3. RESULTS AND DISCUSSION

3.1. Dilute Sulfuric Acid Electrolyte Solution. Figure 1 shows the results of the charge and discharge cycle, respectively, for dilute sulfuric acid electrolyte solution of a 2 V lead acid battery. As seen from the charge cycle graph, the charging process of the lead acid battery using dilute sulfuric acid electrolyte solution had a constant charging voltage between the 0–5 min and had a slight increase after 5 min (0.15 V). There was no further voltage increase after 10 min of the charging cycle, which led to consistent drop to 2.30 V, while the charging voltage after 25 and 30 min was constant. The electrolyte solution of dilute sulfuric acid is used as a reference base for the other electrolyte solutions containing additives to check for improvement. As expected, for the discharge cycle after the application of the 6 V incandescent...
lamp, the discharge time increases as the voltage keeps dropping steadily. The voltage drop was more after 5 min of discharge cycle (0.30 V) and less after 25 min (0.10 V). The quantity of voltage discharged after 15 and 25 min, accounted for 58 and 92%, respectively, of the total discharged voltage of the discharge cycle process. This resulted in a total of 1.20 V that was discharged during the 30 min discharge cycle, and this value indicates that there was significantly no improvement on the life of this lead acid accumulator. The drop during the discharge cycle was not consistent.

3.2. Mixed Electrolyte Solutions (H₂SO₄ + Li₂SO₄ and H₂SO₄ + ZnSO₄). Figure 2 and 3 show the result of the charge and discharge cycles for the electrolyte solutions of lithium and zinc sulfate additives, respectively, of a 2 V lead acid battery. For the lithium sulfate additive mixed electrolyte solution, as the time increased, the voltage also increased, but the change was constant between the 10−15 min mark and has a total increase in voltage of 0.26 V, which resulted in a final charge voltage of 2.74 V after 30 min. The highest increase was between 15 and 20 min and 20−25 min (0.07 V) of the charging cycle, while the lowest increase was between 5 and 10 min (0.02 V). The total increased voltage after 20 min (0.15 V) and 25 min (0.22 V) accounted for approximately 58 and 85% of the total increased voltage of the entire charging cycle, respectively. There were inconsistent increases and decreases in the charging voltage during the charging process for zinc sulfate additive solution. The charging voltage increased after 10 min (3.20 V), followed by a decrease after 15 min but remained unchanged after 20 min (3.15 V). There was further decrease after 25 min and 30 min, as was shown. Thus, the lead acid accumulator of electrolyte additive solution of zinc sulfate in dilute sulfuric acid yielded no significant improvement to that of dilute sulfuric acid electrolyte solution.

The application of a 6 V incandescent lamp initiated the process for the discharge cycle for the additive electrolytes. For lithium sulfate additive solution, the voltage decreased from 2.10 to 1.3 V with more decrease between discharge times of 5−10 min and 10−15 min (0.20 V) of the discharge cycle, while the other discharge times had the same number of voltages discharged (0.10 V). The quantity of voltage discharged after 15 min (0.50 V) and 25 min (0.07 V) accounted for approximately 63 and 88% of the total discharged voltage of the discharge cycle, respectively. There was a significant and consistent increase in the charge cycle voltage and a steady decrease in the discharge cycle voltage, which explain an improvement to the lead acid battery with the lithium sulfate additive electrolyte solution.

The discharge voltage for zinc sulfate solution decreased as the discharge time increased with a total of 0.4 V discharged during the cycle. The highest voltage decrease occurred between 0 and 5, 20−25, and 25−30 min (0.10 V), while the lowest was between 5 and 10 min and 10−15 min of the discharge cycle process. 0.2 and 0.3 V were discharged after 10 and 25 min of the discharge cycle, which are responsible for 50 and 75% of the total discharged voltage, respectively (Table 1).

3.3. Voltage Efficiency. Figure 4 shows the calculated voltage efficiency of the 2 V lead acid battery for each electrolyte fluid solution. The impact of the lithium sulfate additive can be seen on the efficiency improvement of the 2 V lead acid battery. When a lead acid battery is partially charged, the efficiency reduces with each charge, and if the battery never reaches full charge, the life of the battery may be affected.

| Additives (salt) | Molar Mass (g/mol) | Moles Reqd | Mass (g) | Mass in 400 mL of Dilute H₂SO₄ (g) | Mass Used in 400 mL of Dilute H₂SO₄ (g) |
|-----------------|-------------------|------------|----------|----------------------------------|---------------------------------|
| Zinc sulfate    | 127.96            | 2          | 255.92   | 102.37                           | 102.40                          |
| Lithium sulfate | 287.54            | 1          | 287.54   | 115.00                           | 115.00                          |

3.4. Statistical Analysis for the Discharge Cycle. A one-way analysis of variance (ANOVA) was performed, as shown in Table 2 for the charge cycle and Table 3 for the discharge cycle of both dilute sulfuric acid electrolyte and for the mixed (H₂SO₄ + Li₂SO₄ and H₂SO₄ + ZnSO₄) electrolyte solutions. The analysis shows that there was no significant interaction in the charge cycle but there was significant interaction in discharge voltage of the three electrolyte solutions. Charge cycle, F(2,18) = 0.484, p = 0.624 and discharge cycle, F(2,18) = 4.553, p = 0.025. Statistically, it also means that there is not enough evidence to conclude that the mixed electrolyte solution has higher or better charge cycle than the dilute sulfuric acid electrolyte, but based on the discharge voltages, there was improvement with the use of the additives, especially for sulfuric acid–lithium sulfate electrolyte solution.

3.5. DISCUSSION

A lead acid battery made up of dilute sulfuric acid electrolyte solution is the standard, and this was compared to that of the mixed electrolyte solution involving sulfate additives (H₂SO₄ + Li₂SO₄ and H₂SO₄ + ZnSO₄). As a voltage is applied to the lead acid battery which is often greater than the battery’s voltage, a current will flow through the lead acid battery in the reverse direction to when it is supplying current and this helps the battery to start charging. The rate of charge or current that will flow depends on the difference between the battery’s voltage and the voltage that is applied to it. Ideally, during the charge cycle, as the charge time increases, the charge voltage is also expected to increase for an improvement on the lead acid battery to be established. Similarly, for the discharge cycle of a typical lead acid battery, the discharge voltage decreases as the discharge time increases, which causes the formation of lead sulfate crystals at both the negative and positive terminals. When it takes a longer time to fully discharge a lead acid battery, there is an improvement in the life of such a battery. The charging cycle process for the dilute sulfuric acid solution of 2 V lead acid battery was not consistent in terms of increase in the voltage as shown, but it was better for the sulfuric acid–lithium sulfate mixed solution, where the increase in voltage during the charge cycle showed more consistency. The charging cycle process of sulfuric acid–zinc sulfate solution was inconsistent compared to that of lithium sulfate. In addition, the increase in the total quantity of voltage during the charge cycle indicates the quality of the cycle life of such a lead acid battery and therefore prolongs the life of the battery. This implies that after the charge cycle, the lithium sulfate additive electrolyte delivered increased ampere-hour rating and made a significant improvement to the 2 V lead acid battery.

For the discharge cycle process in which a 6 V incandescent lamp was applied with a result as shown, the decrease in voltage was more consistent in the sulfuric acid–lithium sulfate mixed solution than in both dilute sulfuric acid electrolyte and
zinc sulfate solutions, as illustrated in the graphs above. The total quantity of voltage discharged in the lead acid battery after 30 min of the discharge cycle for dilute sulfuric acid was more than that for the mixed electrolyte solutions. The mixed additive solution polarized much less compared to the sulfuric acid electrolyte solution and generated improvement with less discharged voltage after a 30 min discharge cycle. This means that a lead acid battery made up of dilute sulfuric acid electrolyte solution will discharge faster during the discharge cycle than that by the mixed electrolyte solutions.

Therefore, a 2 V lead acid battery of sulfuric acid–lithium sulfate mixed electrolyte solution (H₂SO₄ + Li₂SO₄) offers better performance compared to a battery made of only sulfuric acid electrolyte solution or zinc sulfate additive. It is vital to note that this lithium additive had an interface with the electrochemical reaction of the lead acid battery without creating any obstruction. The individual compounds also have varying molar mass in g/mol, and there is a need for more charge and discharge cycles for complete compound absorption. A similar work by Willis¹¹ discovered that addition of copper sulfate, aluminum sulfate, cobalt sulfate, and magnesium sulfate mixed into four different parts with deionized water and allowed for about 20 min improved the performance and life of a 12 V lead acid battery. There was significant difference between the treated and untreated two identical 12 V lead acid batteries after they were subjected to similar charge–discharge cycles with some high-level charging and discharging which was repeated in the presence of well-supervised conditions.¹¹ The addition of discrete carbon nanotubes to both electrodes also enhanced and improved the performance and cycle life of a lead acid battery, according to Sugumaran.¹² These carbon nanotubes were incorporated to increase the acceptance and cycle life with no change to paste density and without impeding the manufacturing process. The nanotube-treated lead acid battery showed little change to reserved capacity, improved cold cranking, and enhanced overall system efficiency.¹² The development of a fast lead acid battery charging system by Cheng,¹³ using a multistate strategy, was established, which gave the same improvement results as the previous two discussed methods. The help of large steady current with minimal temperature rise created floated charge when the battery reached the stage at which it has been overcharged and remains in small quantity of current.¹³ The qualities of titanium dioxide and carbon additives were successfully used in enhancing the performance of lead acid battery, as investigated by Rekha.¹⁴ The titanium dioxide in the positive active material yielded benefits in improving the tetra basic lead sulfate which eventually improved the lifespan of the lead acid battery.¹⁴–¹⁶

On terminal corrosion, it was visually observed that dilute sulfuric lead acid battery appeared to be emitting more quantity of gas as compared to the small quantities emitted from the additive electrolyte solution used and had the least gas emission. Too much gassing with acid spray is a general cause of terminal corrosion on lead acid accumulators. The significant decrease in gassing in sulfuric acid–sulfate mixed electrolyte solution can effectively address the problem of terminal corrosion on lead acid batteries.

Based on hazardous gases, there was significant reduction of distilled water consumption in lead acid batteries of the lithium sulfate additive electrolyte solution compared to that of dilute sulfuric electrolyte solution. This is similar to gas and spray emissions which indicate a very distinctive improvement. With respect to maintenance reduction which is synonymous to gas and spray emission explained above, less gas emission represents reduction in the maintenance. This is because gas emission involves waste of energy, release of hazardous gas mixtures, and fine sulfuric acid mists. Based on these, it will be
cost-effective to maintain a lead acid battery made of sulfuric acid—lithium sulfate electrolyte solution.

The efficiency of a lead acid battery is the ratio between the output and the input of a cell, which can be expressed as percentage of per unit value. The voltage efficiency calculated for the mixed electrolyte solution (lithium sulfate) was greater than that of the sulfuric acid solution (85—77%), as shown in Figure 4. Hence, a lead acid battery made of sulfuric acid—lithium sulfate electrolyte solution will have a prolonged life than that made of only sulfuric acid electrolyte solution. The normal efficiency for a lead acid battery is estimated at 67%, and this increase with lithium sulfate additive goes a long way to improve the life of the 2 V lead acid battery. The lithium sulfate additive introduction into the battery cell due to the cohesive nature of the product resulted in particle shedding which in the traditional dilute sulfuric acid electrolyte always causes difficulty in the separators and electron effluents. The input and output qualities are improved and reduced in the heat produced.

4. CONCLUSIONS

In terms of the voltage efficiency, there was a distinctive improvement in the efficiency of a lead acid battery when using lithium sulfate additive electrolyte solution compared to that of dilute sulfuric acid solution (77—85%). The zinc sulfate offered no significant improvement to dilute sulfuric acid electrolyte as discovered from the experiments. For the life of lead acid battery, the one cycle test carried is quite short compared to the overall life of a battery to make a clear statement, but the life improvement can be inferred from the length of time it took to fully discharge. This implies that lithium sulfate additive in lead acid battery will prolong its life.

Finally, based on this experiment on the lead acid accumulator, it is safe to say that only lithium sulfate additive offered an improvement to the charge cycle, cathode stability, life, efficiency, maintenance, and corrosion of a normal lead acid battery made of dilute sulfuric acid electrolyte.

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Notes

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