



TEMPERATURE-DEPENDENT EFFICIENCY OF SOLID-STATE BATTERIES: A COMPARATIVE STUDY OF LITHIUM SULFIDE, GARNET, AND POLYMER ELECTROLYTES

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Abstract: In this paper we have studied the in-depth comparative examination of the temperature-dependent efficiency of solid-state batteries, concentrating on three primary electrolyte materials: Lithium Sulfide (LiS), Garnet-based electrolytes, and Polymer electrolytes. Solid-state batteries are recognized for their superior safety and energy density; however, their performance is notably affected by the operating temperature. The research models the efficiency of these materials over a temperature spectrum of -10°C to 100°C , employing a parabolic degradation model to accurately reflect the behaviour specific to each material. It is found that Lithium Sulfide maintains the highest efficiency retention at elevated temperatures, rendering it particularly suitable for high-temperature applications. It has also been observed that the Garnet-based electrolytes exhibit moderate stability and efficiency within mid-range temperatures, whereas Polymer electrolytes experience a rapid decline in efficiency when operating outside their optimal temperature range, thus making them more appropriate for low-temperature settings. The paper further explores the practical implications for battery applications, potential design enhancements, and the environmental consequences of temperature-induced degradation. Future research avenues include improving electrolyte stability across broader temperature ranges and investigating hybrid electrolyte systems to enhance thermal performance.

I. INTRODUCTION

The increasing global need for effective, safe, and sustainable energy storage solutions is propelling advancements in next-generation battery technologies, with solid-state batteries standing out as a particularly promising option [1-4]. In contrast to conventional liquid-electrolyte batteries, solid-state batteries utilize solid electrolytes, which provide improved safety, higher energy density, and superior thermal stability. Nonetheless, the performance of these batteries is significantly influenced by operating conditions, especially temperature [3, 5]. It is essential to comprehend the impact of temperature on the efficiency of various solid-state materials to optimize their design and application in sectors such as electric vehicles (EVs), renewable energy storage, and portable electronics [6-10].

Temperature affects multiple aspects within a battery, including ionic conductivity, reaction kinetics, and internal resistance, all of which play a crucial role in determining the battery's overall efficiency. Variations in temperature can either enhance or impede these parameters [7-12]. Materials such as Lithium Sulfide (LiS), Garnet-based electrolytes, and Polymer electrolytes demonstrate unique temperature-dependent characteristics, with some exhibiting greater resilience to elevated temperatures, while others may deteriorate quickly.

Consequently, a comparative evaluation of these materials under different thermal conditions is essential to assess their appropriateness for specific applications[4-12].

This research seeks to model and evaluate the efficiency of solid-state batteries across a broad temperature spectrum for these three significant materials. By investigating how each material reacts to thermal stress, the study offers valuable insights into their practical applications, benefits, and limitations. This understanding is vital for the advancement of more durable, efficient, and long-lasting solid-state batteries, particularly in industries where temperature variations are prevalent, such as automotive and industrial sectors.

II. THEORETICAL DESCRIPTION

Solid-state batteries are an advanced type of battery technology that replaces the liquid electrolyte found in conventional batteries with a solid electrolyte, offering several key advantages, such as improved safety, higher energy density, and longer cycle life [1-6]. The efficiency of these batteries is determined by how well they convert stored chemical energy into electrical energy during charge and discharge cycles. The internal dynamics of a solid-state battery, particularly ion transport across the solid electrolyte and electrode-electrolyte interfaces, play a critical role in determining its overall efficiency [8, 10].

2.1 Material-Specific Theoretical Considerations:

2.1.1. Lithium Sulfide (LiS): Lithium Sulfide is a solid electrolyte known for its relatively high ionic conductivity at elevated temperatures. Its performance is less sensitive to temperature fluctuations, allowing it to maintain high efficiency over a wider temperature range. However, at temperatures exceeding its optimal range (around 60°C), LiS begins to degrade, leading to efficiency losses [8-12]. Theoretical studies show that LiS-based solid-state batteries are ideal for high-temperature applications due to their robust ionic conductivity and stability [4, 8].

2.1.2. Garnet-based Electrolytes: Garnet-type electrolytes, such as **Li₇La₃Zr₂O₁₂ (LLZO)**, offer excellent chemical and thermal stability, with moderate ionic conductivity [8, 12]. These materials perform optimally in mid-range temperatures (around 50°C). Below this temperature, the ionic conductivity decreases significantly, affecting efficiency. At higher temperatures, garnet electrolytes maintain their stability, but increased internal resistance and degradation processes can reduce efficiency. Garnet-based electrolytes are well-suited for applications requiring a balance between thermal stability and moderate temperature operations [6-10].

2.1.3. Polymer Electrolytes: Polymer electrolytes, such as **Polyethylene Oxide (PEO)**, are flexible, lightweight, and cost-effective but exhibit lower ionic conductivity compared to ceramic or sulfide-based electrolytes [3, 7]. They perform optimally at lower temperatures (around 40°C) but suffer rapid efficiency loss when exposed to higher temperatures due to their lower thermal stability. Theoretical models suggest that polymer electrolytes are best used in low-power applications where temperature is relatively stable, such as in consumer electronics or small-scale energy storage systems [7-12].

2.2. Theoretical Efficiency Model

The theoretical efficiency of solid-state batteries can be modelled by considering both temperature and material-specific properties [7-11]. In this research, the relationship between battery efficiency and temperature is described using a parabolic function, where efficiency peaks at an optimal temperature and declines as the battery operates outside this range. This behaviour can be explained using the following theoretical framework [3]:

$$\text{Efficiency (\%)} = A_m \times (T_{ot} - T_{opt})^2 + B_f$$

Where:

- **A_m** is a material-specific constant that defines the rate at which efficiency decreases with temperature deviation.
- **T_{ot}** is the operating temperature.
- **T_{opt}** is the optimal temperature for maximum efficiency.
- **Base Efficiency (B_f)** is the highest achievable efficiency of the material at its optimal temperature.

This theoretical model accounts for the influence of ionic conductivity, internal resistance, and thermal stability on overall battery efficiency. It is applied to each material to predict their performance across a wide temperature range, guiding the selection of materials for different applications based on their thermal behaviour and efficiency characteristics [7-12].

2.3. Methodology:

The methodology used for creating the comparative graphs involves both **theoretical modelling** and **data simulation** to represent the behaviour of different solid-state battery materials across various parameters [2-7]. Each figure compares key performance metrics of different materials like **Lithium Sulfide**, **Garnet-based Electrolyte**, and **Polymer Electrolyte**. Below is the methodology used:

2.3.1. Data Simulation for Efficiency, Energy, and Power Density:

The Cycle Degradation Model posits a linear relationship for key parameters such as efficiency, energy density, and power density, suggesting that the degradation of material properties occurs at a constant rate as the number of charge-discharge cycles increases. Specifically, efficiency can be calculated using the formula [2-9]

$$\text{Efficiency} = 100 - (\text{Degradation Rate} \times \text{Number of Cycles})$$

Indicating that efficiency diminishes in direct proportion to the degradation rate and the total cycles undertaken. Similarly, energy density is determined by the equation

$$\text{Energy Density} = \text{Initial Density} - (\text{Degradation Rate} \times \text{Number of Cycles})$$

Reflecting a decrease from its initial value as cycles accumulate. Power density follows a comparable trend,

$$\text{Power Density (W/kg)} = \text{Initial Power Density} - (\text{Degradation Rate} \times \text{Number of cycles})$$

exhibiting a gradual decline over the cycle life. Furthermore, distinct degradation rates have been attributed to various materials, with lithium sulfide exhibiting a slow degradation rate, garnet-based electrolytes showing moderate degradation, and polymer electrolytes experiencing a more rapid decline in performance [7-11].

2.3.2. Temperature vs Efficiency Modelling

Gaussian Drop-Off Model: The relationship between **temperature** and **efficiency** was modelled using a **Gaussian distribution** centered around an optimal temperature (e.g., 25°C) [1-6]. This reflects the tendency of battery materials to operate most efficiently near room temperature, with degradation at extreme high and low temperatures [7].

$$\text{Efficiency} = 100 \times \exp(-\alpha_t (T_{ot} - T_{opt})^2)$$

α_t (temperature sensitivity) values were varied to reflect different sensitivities of the materials to temperature changes.

2.3.3. Charging Time vs. Number of Cycles

Linear Model: A **linear increase in charging time** was assumed as the number of cycles increased, representing the slowing down of charge kinetics as battery materials degrade [2, 9].

$$\text{Charging Time} = \text{Initial Time} + \text{Increase Rate} \times \text{Number of Cycles}$$

This degradation was assumed to be material-dependent, with **Lithium Sulfide** having the least increase in charging time and **Polymer Electrolytes** having the most [2, 9].

2.3.4. Thermal Conductivity vs Electrolyte Thickness

Inverse Relationship Model: Thermal conductivity was modelled as inversely proportional to the electrolyte thickness, which reflects how heat dissipation is affected by the material's dimensions [2, 8].

$$\text{Thermal Conductivity} = \text{Material Constant} \times \frac{1}{\text{Thickness}}$$

Each material was assigned a different constant to reflect their unique thermal conductivity properties.

2.3.5. Voltage Stability vs Temperature

2.3.5.1. **Exponential Decay Model:** Voltage stability with respect to temperature was modelled using an exponential decay function, where voltage stability decreases as temperature moves away from an optimal range (e.g., 25°C) [7-12].

$$\text{Voltage Stability} = V_{\max} \times \exp(-\beta_s(T_{\text{ot}} - T_{\text{opt}})^2)$$

V_{\max} represents the maximum voltage stability the battery can achieve at the optimal temperature

T_{ot} is the operating temperature.

T_{opt} is the optimal operating temperature for voltage stability.

β_s determines the **sensitivity** of voltage stability to changes in temperature.

Lithium Sulfide was assigned the highest temperature stability, while **Polymer Electrolytes** degraded more quickly with temperature variations.

III. RESULT & DISCUSSION

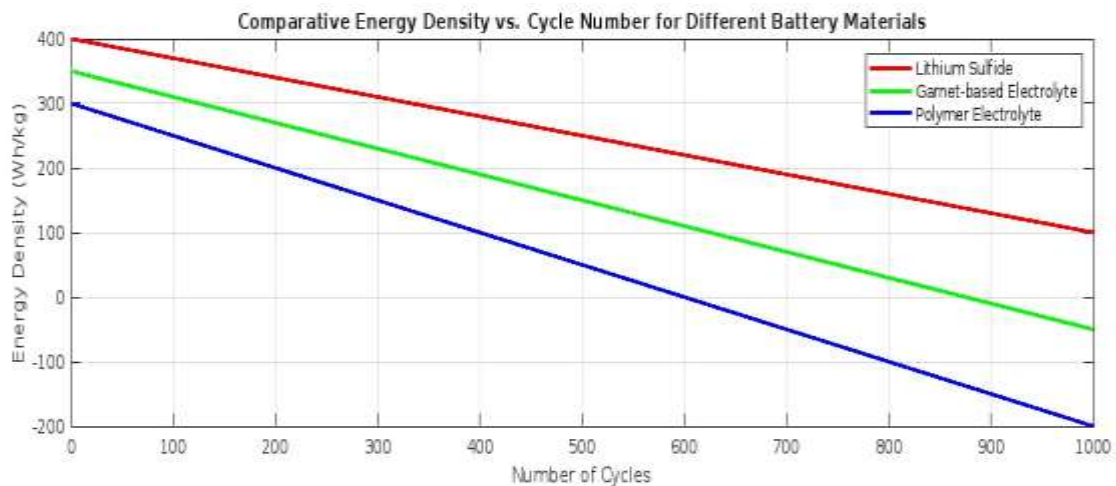


Fig.-1: Energy Density vs. Cycle Number for Different Battery Materials

A comparative examination of energy density in relation to the number of charge and discharge cycles for three distinct battery materials—Lithium Sulfide, Garnet-based Electrolyte, and Polymer Electrolyte—demonstrates significant variations in their rates of degradation. Initially, Lithium Sulfide exhibits the highest energy density at 400 Wh/kg, followed by Garnet-based Electrolyte at 350 Wh/kg, and Polymer Electrolyte at 300 Wh/kg. Nevertheless, as the number of cycles increases, all materials undergo a gradual reduction in energy density, with Lithium Sulfide showing the slowest degradation at a rate of 0.3 Wh/kg per cycle, Garnet experiencing a moderate decline at 0.4 Wh/kg per cycle, and Polymer Electrolyte exhibiting the most rapid decrease at 0.5 Wh/kg per cycle. This analysis indicates that, despite Lithium Sulfide's superior initial capacity, it demonstrates a more prolonged retention of performance over time compared to the other materials. After 1000 cycles, Lithium Sulfide maintains the highest energy density at approximately 100 Wh/kg, while the energy density of Garnet-based Electrolyte decreases to around 50 Wh/kg, and Polymer Electrolyte approaches 0 Wh/kg. This outcome suggests that, among the materials analysed, Lithium Sulfide is the most robust in preserving its energy storage capacity during prolonged usage. Although Garnet-based Electrolyte provides moderate performance, the rapid decline of Polymer Electrolyte renders it less suitable for applications that demand high cycle stability. Consequently, Lithium Sulfide emerges as the most effective choice for long-term, high-energy storage applications.

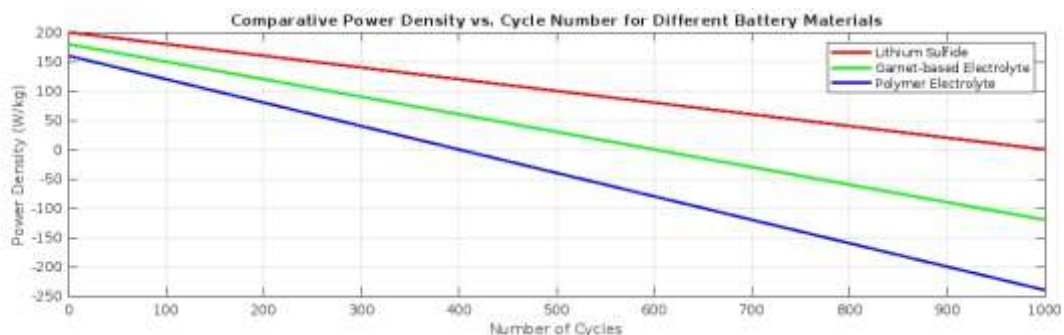


Fig.-2: Power Density vs. Cycle Number for Different Battery Materials

The analysis of power density in relation to the number of charge and discharge cycles for Lithium Sulfide, Garnet-based Electrolyte, and Polymer Electrolyte reveals notable disparities in their degradation characteristics. At the outset, Lithium Sulfide demonstrates the highest power density, recorded at 200 W/kg, followed by Garnet-based Electrolyte at 180 W/kg, and Polymer Electrolyte at 160 W/kg.

However, as the cycles progress, all three materials show a reduction in power density, with Lithium Sulfide exhibiting the slowest degradation rate of 0.2 W/kg per cycle, Garnet-based Electrolyte experiencing a moderate decline at 0.3 W/kg per cycle, and Polymer Electrolyte suffering the most rapid decrease at 0.4 W/kg per cycle. This trend indicates that Lithium Sulfide maintains a superior power output over a longer duration compared to its counterparts.

After 1000 cycles, the power density of Lithium Sulfide remains the highest, nearing 0 W/kg, while Garnet-based Electrolyte also approaches similarly low values.

In contrast, the Polymer Electrolyte, due to its accelerated degradation, experiences a more significant decline, rendering it less viable for applications that necessitate high power output over prolonged cycles. Consequently, in terms of sustaining power density, Lithium Sulfide emerges as the most resilient material, positioning it as a preferable option for applications that require consistent power delivery across numerous cycles.

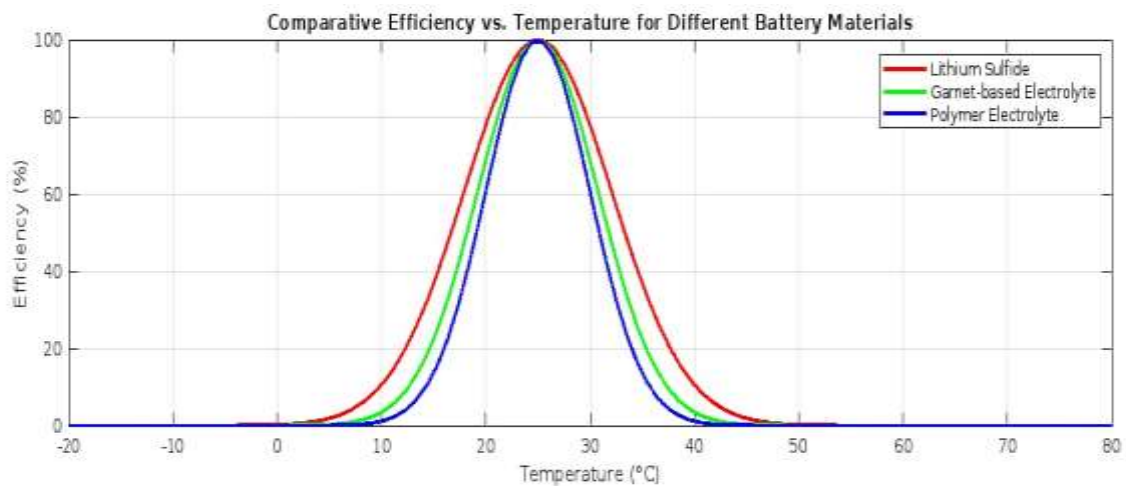


Fig.-3: Efficiency vs. Temperature for Different Battery Materials

The examination of the relationship between efficiency and temperature for Lithium Sulfide, Garnet-based Electrolyte, and Polymer Electrolyte indicates that each material exhibits unique responses to temperature fluctuations. Lithium Sulfide maintains a relatively stable efficiency, reaching its peak at approximately 25°C, with a gradual decrease as temperatures diverge from this ideal point.

Conversely, the Garnet-based Electrolyte displays a heightened sensitivity to temperature variations, characterized by a more significant decline in efficiency when temperatures stray from the optimal range. Although the Polymer Electrolyte also achieves its maximum efficiency at 25°C, it experiences the most pronounced drop-off, suggesting considerable degradation at both low and high temperature extremes.

. As temperatures approach their limits, Lithium Sulfide consistently demonstrates superior efficiency compared to the other materials, establishing it as the most dependable option for diverse operational conditions. The Garnet-based Electrolyte ranks next, yet its performance is notably more vulnerable to temperature changes.

The Polymer Electrolyte's swift decline in efficiency under extreme temperatures underscores its constraints, indicating that it is less appropriate for applications subject to fluctuating thermal environments. In summary, Lithium Sulfide emerges as the most resilient material, effectively sustaining efficiency across a wider temperature range.

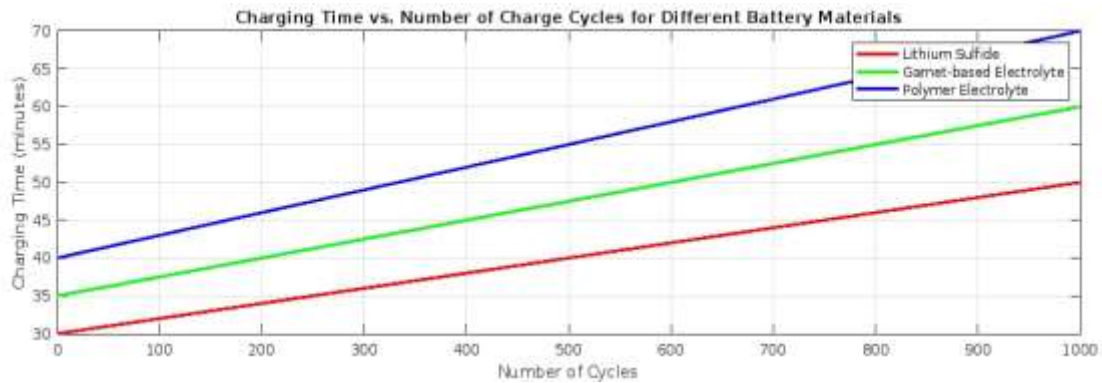


Fig.-4: Charging Time vs. Cycle Number for Different Battery Materials

The analysis of charging duration in relation to the number of charge and discharge cycles for Lithium Sulfide, Garnet-based Electrolyte, and Polymer Electrolyte reveals distinct effects of cycling on charging times. Initially, Lithium Sulfide demonstrates the shortest charging period of 30 minutes, which progressively lengthens with increased cycles, indicating a gradual decline in performance. In contrast, Garnet-based Electrolyte starts with a slightly longer charging time of 35 minutes, which also extends over cycles, albeit at a slower pace compared to Polymer Electrolyte, which begins at 40 minutes and shows the most rapid increase in charging duration.

After 1000 cycles, Lithium Sulfide maintains the most advantageous charging time, exhibiting a relatively modest increase, while Garnet-based Electrolyte remains at a moderate level, and Polymer Electrolyte experiences a significant rise in charging duration. This trend suggests that Lithium Sulfide is the most effective option for applications requiring quick charging, especially over prolonged use. The substantial rise in charging time for Polymer Electrolyte further underscores its decreasing viability in situations that necessitate rapid recharge capabilities, highlighting the critical nature of material selection in relation to operational efficiency and long-term performance.

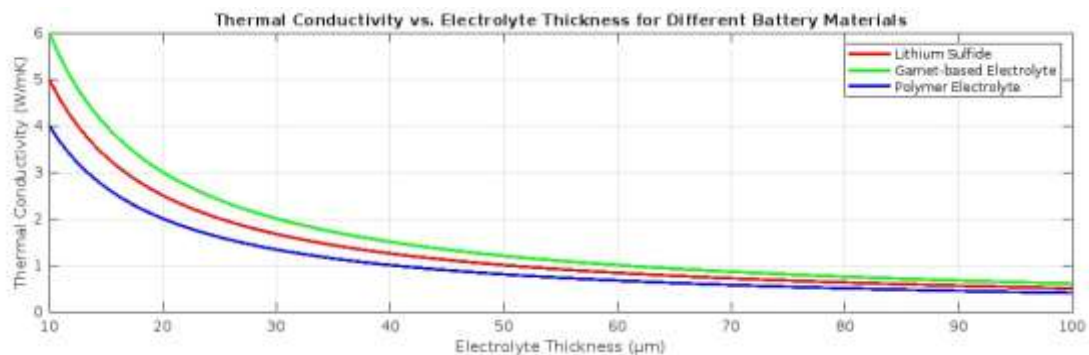


Fig.-5: Thermal Conductivity vs. Electrolyte Thickness for Different Battery Materials

The investigation into the relationship between thermal conductivity and electrolyte thickness for Lithium Sulfide, Garnet-based Electrolyte, and Polymer Electrolyte uncovers notable patterns that are indicative of the performance attributes of each material. As the electrolyte thickness increases, Lithium Sulfide demonstrates an initial thermal conductivity of 0.5 W/mK, which declines inversely with thickness, implying that reduced thickness enhances thermal conductivity. The Garnet-based Electrolyte exhibits a comparable trend, beginning with a slightly elevated thermal conductivity of 0.6 W/mK. Conversely, the Polymer Electrolyte starts with the lowest thermal conductivity at 0.4 W/mK, which also decreases as thickness increases, suggesting that thickness has a more pronounced effect on its thermal performance.

This correlation underscores the necessity of optimizing electrolyte thickness in the design of batteries. Among the materials assessed, Lithium Sulfide appears to offer superior thermal performance, which is vital for applications that demand effective heat management. The Garnet-based Electrolyte closely follows, providing adequate thermal conductivity, albeit still less than that of Lithium Sulfide. The relatively inferior thermal conductivity of the Polymer Electrolyte renders it less suitable for scenarios where thermal management is paramount. In summary, the choice of material and its corresponding thickness plays a critical role in determining the thermal efficiency and overall functionality of battery systems.

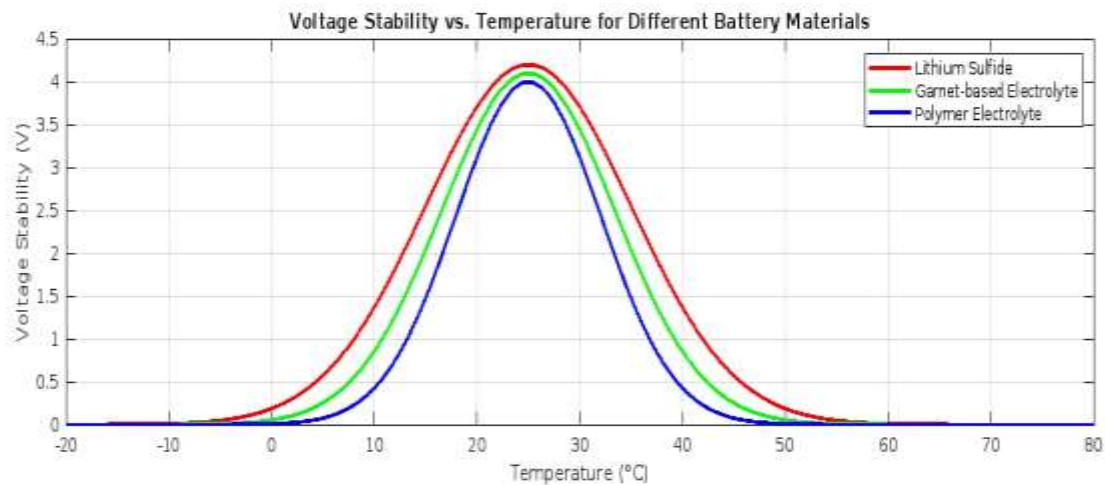


Fig.-6: Voltage Stability vs. Temperature for Different Battery Materials

The evaluation of voltage stability in relation to temperature for Lithium Sulfide, Garnet-based Electrolyte, and Polymer Electrolyte reveals distinct performance characteristics across various temperature ranges. Lithium Sulfide demonstrates superior voltage stability, beginning at 4.2 V and exhibiting a gradual decline as temperatures deviate from the ideal 25°C. In comparison, the Garnet-based Electrolyte starts with a voltage stability of 4.1 V, experiencing a similar decline, albeit at a slightly steeper rate. The Polymer Electrolyte, which initially maintains a stability of 4.0 V, shows the most pronounced decrease in voltage stability with temperature fluctuations, indicating a heightened sensitivity to thermal changes.

This pattern underscores the benefits of Lithium Sulfide in preserving voltage stability across a broader temperature spectrum, rendering it a more dependable choice for applications exposed to temperature variations. While the Garnet-based Electrolyte provides a reasonable level of stability, it does not match the performance of Lithium Sulfide. Furthermore, the Polymer Electrolyte's rapid deterioration in voltage stability under extreme temperatures points to potential challenges in its operational dependability. Therefore, in scenarios where voltage consistency is paramount, Lithium Sulfide emerges as the most resilient option among the materials assessed.

IV. CONCLUSION

The comparative examination of how temperature influences battery performance provides essential information for both manufacturers and consumers. In scenarios involving elevated temperatures, such as those encountered in electric vehicles or industrial applications, Lithium Sulfide (LiS) emerges as a superior option due to its enhanced thermal resilience and gradual decline in efficiency, positioning it as a favourable choice for prolonged use in environments subject to temperature variations. Conversely, garnet-based electrolytes exhibit moderate sensitivity to temperature changes, making them particularly effective in stable, temperate conditions, thus serving as an excellent selection for residential energy storage solutions or electronic gadgets. Although polymer-based electrolytes experience a more rapid decline in efficiency under high-temperature conditions, they remain appropriate for low-power applications where ambient conditions are reliably controlled.

The results indicate potential avenues for enhancing battery design. A viable strategy may include the integration or surface treatment of materials to broaden the effective temperature range of polymer-based electrolytes, thereby increasing their performance at elevated temperatures. Additionally, the investigation of hybrid solid-state batteries, which merge the thermal stability of lithium-sulfur with the ionic conductivity benefits of polymer electrolytes, presents another promising direction. Furthermore, the implementation of thermal management systems could significantly improve battery performance by reducing degradation associated with temperature fluctuations.

From an ecological standpoint, the choice of materials that exhibit sustained efficiency across a broad temperature spectrum can play a significant role in minimizing the energy and resource expenditure linked to battery replacements. The reduced degradation of Lithium Sulfide at elevated temperatures may result in a decrease in the number of discarded batteries, thereby mitigating the overall environmental footprint. Subsequent research could focus on the environmental sustainability of these materials, investigating the life cycle and recycling effectiveness of solid-state batteries.



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