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Lithium-Sulfur battery lifetime prediction, a reliability approach

Predicción de vida de batería Litio-Azufre, un enfoque de confiabilidad

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Abstract

The remaining useful life (RUL) phenomenon of lithium sulfur batteries is characterized by being a nonlinear phenomenon that implies difficulties to determine it; performance tests to evaluate and predict the RUL involve long times and a large sample data that, sometimes, is limited for an adequate characterization due to insufficient data. The present work addresses the problem through a reliability analysis considering a highly censored reduced sample, which allows characterizing is reliability key indicators and RUL through the Weibull distribution and the reduced bias adjustment (RBA) method, considering uncertainty in the estimation. The results show that the method is capable to predict RUL of lithium-sulfur batteries, as well as the usefulness of reliability indices.

Characterization, Reliability, Indices, Uncertainty

Resumen

El fenómeno de vida útil de baterías litio-azufre se caracteriza por ser un fenómeno no lineal que implica dificultades para determinarlo; las pruebas de desempeño para evaluar y predecir la vida útil esperada implican tiempos largos y una muestra amplia que, en ocasiones, se ve limitada para una adecuada caracterización por la insuficiencia de datos. El presente trabajo aborda el problema mediante un análisis de confiabilidad considerando una muestra reducida altamente censurada, lo que permite caracterizar sus indicadores clave de confiabilidad y vida útil mediante la distribución Weibull y el ajuste de sesgo reducido (RBA, por sus siglas en inglés), considerando la incertidumbre en la estimación. Los resultados muestran que el método es capaz de realizar predicciones adecuadas acerca de la vida útil de baterías tipo litio-azufre, así como la utilidad de los índices de confiabilidad.

Caracterización, Confiabilidad, Índices, Incertidumbre

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I. Introduction

The growing demand for energy, the energy crisis and the challenges of climate change provide a trigger for electrification and the development of energy storage systems. Lithium-ion batteries play an important role in these issues (Hu *et al*., 2020); however, this type of battery has reached its energy limit, leading to the need to develop new types of batteries, such as Lithium-Sulfur (Li-S), which has the characteristic of being higher energy density (Benveniste *et al.*, 2018). Several researchers have been working in the development of new practical batteries linking the effect between cathode and anode (Gao *et al*., 2023); others are working in how to deal with shuttle effect and dendrite formation (Yu *et al*., 2023), or looking to extend battery lifetime (Zhu *et al*., 2023).

Conventional methods of lithium-ion batteries to predict or estimate state of charge or coulomb counting (both related to battery lifetime) not effective for Li-S batteries (Brieske *et al.*, 2023).This implies that the current work on modeling and estimation of state and useful life is not sufficient for this battery chemistry and new models are necessary (Fotouhi *et al*., 2016). Additionally, the lifetime of Li-S batteries is characterized by having a non-linear behavior, divided into two regions, with a high plateau and a sudden drop in capacity (also called low plateau), which reinforces the need for the development of new modeling techniques, and estimation (Knap *et al*., 2018).

In this paper it is aimed to predict remaining useful life for a Li-S battery, considering a small sample and highly censored due to several constraints around the manufacturing of the battery. These constraints are related to its new chemistry (cathode fabricated with Sulfur coming from agave), manufacturing process of battery is under design, and the research group is working on optimizing a Li-S battery. Regarding these conditions, analysis Weibull is proposed to estimate battery RUL, reliability and risk.

The Li-S lifetime battery phenomenon is nonlinear, and it implies a difficult to use some existent prediction methods.

Enough data to characterize and predict lifetime is a key element for having a good model. Nevertheless, time to experiment with batteries sometimes require days or weeks and then it is not possible to have complete and enough data to include in the prediction model. With Weibull Analysis it is possible to predict time to failure for a Li-S battery considering possible censored data.

In the application of the present work was found that the analysis Weibull is useful because it is possible to model and predict the battery RUL under uncertainty conditions considering a highly censored data under a reliability approach.

II. Methods

In next sections the used methods at this work will be briefly explained; between them are included reliability and Weibull analysis, censored data, goodness-of-fit test, Weibull parameter estimation with censored data and reduced bias adjustment (RBA).

1. Reliability analysis

Reliability is defined as the probability that a product will last a specified time under specific conditions. One challenge in reliability is concerned with quantitative measures like mean time to failure (MTTF), which is one of the most widely used terms in reliability engineering. MTTF applies to Li-S battery since once failed it is not repairable and is defined as the number of charge-discharge cycles until failure.

2. Weibull analysis

Some works on prediction for batteries using Weibull model exists, applied to lithium-ion battery in renewable energy, electric vehicle and telecommunications (Eom *et al*., 2007), (Ganjeizadeh *et al*., n.d.), (Ossai & Raghavan, 2017) and (Schmalstieg *et al*., 2013). The Weibull distribution is a flexible model that can describe the probability distribution of the lifetime characteristics of a battery. Some studies have used the Weibull distribution to estimate the lifetime probability distribution of batteries based on experimental data (Chiodo *et al*., 2016)(Jinliang Cao, 2021). The Weibull distribution is also used to model lifetime and failure time data in reliability applications (Kumar & V, 2017)(Gu *et al*., 2011).

In some cases, a 2-fold Weibull mixture distribution is selected as the suitable distribution for the lifetime of the battery (Sultana, 2015). The three-parameter generalized inverse Weibull distribution is another form of the Weibull distribution that is mainly used in reliability lifetime data analysis (Li *et al*., 2019). The reliability of batteries can be evaluated based on the Weibull life distribution. Then, Weibull analysis is a useful tool for analyzing the lifetime characteristics of batteries.

Weibull distribution is often characterized by three parameters: location, scale, and shape. Weibull distribution can take on the characteristics of different distributions. And its primary advantage is that it can afford failure predictions with limited sample size (Vignarooban *et al*., 2016).

This paper will focus on the 2-parameter Weibull distribution. These parameters are scale (η) and shape (β). The shape parameter determines the failure distribution that best describes the data. Early failures occur when $0<\beta<1$. When $\beta=1$, it is considered that failures are random and when $\beta>1$ indicates wear-out failures. The scale parameter defines the characteristic lifetime for which the device will have failed. The probability density function (f), cumulative distribution function (F), reliability function (R) of a Weibull distribution is expressed as follows:

$$f(t) = \frac{\beta}{\eta}\left(\frac{t}{\eta}\right)^{\beta-1} e^{-\left(\frac{t}{\eta}\right)^{\beta}} \quad (1)$$

$$F(t) = 1 - e^{-\left(\frac{t}{\eta}\right)^{\beta}} \quad (2)$$

$$R(t) = 1 - F(t) = e^{-\left(\frac{t}{\eta}\right)^{\beta}} \quad (3)$$

Where η is scale parameter, β is shape parameter and t is time to failure.

Some of the main characteristics of Weibull Analysis is that it allows to work with censored data; this means that is possible to deal with short samples, special when these samples require long times to be evaluated and then it is necessary to stop experiments and include partial information to evaluate or to predict lifetime in any component or product.

3. Censored data

Censored data appears when, for a random variable of interest, the monitoring is stopped before measurement is complete (i.e. before the unit being monitored fails or when an experiment should be stopped before it ends) (Pham, 2006). Then, censored observation contains partial information about the variable of interest, or it is incomplete (Kleinbaum DG, 2010). During cycle test, charging/discharging cycles can be interrupted due to several conditions (power outages, operation mistakes or any other accident). These situations are difficult to prevent during experimentation. Such interruptions caused segments to be censored observations because the information is incomplete. Every dataset is important due to the long experimental time, and this survival analysis can manage datasets with interruption, so no dataset must be discarded (Lin *et al*., 2020).

4. Goodness of fit test with censored data

Goodness-of-fit test is used to determine whether a given sample of data follows a specific probability distribution. This kind of test is important in several fields, including reliability analysis, medical research, and environmental studies. One commonly used goodness-of-fit test is the Anderson-Darling (AD) test.

The AD test is nonparametric, measuring the discrepancy between the observed data and a specified theoretical distribution. It is particularly useful for detecting deviations in the tails of the distribution. It is based on the empirical distribution function and is calculated by comparing the observed cumulative distribution function with the expected cumulative distribution function under the null hypothesis of a specific distribution. It has been applied to various distributions and types of data, including censored data (Pakyari & Al-Hamed, 2023), (Afify & Mohamed, 2020), (Ding *et al*., 2017).

5. Weibull Parameter estimation with censored data

A common approach to estimate the parameters of a Weibull Distribution is via the method of maximum likelihood (ML), in which parameters are set to values that maximizes the log-likelihood of the data.

The procedure for determine Weibull parameters for censored data using ML estimators is as follows:

a. Order data in a table so that all failures precede censored data.

b. For this case, assume that location parameter is equal to zero.

c. Determine the value of the shape parameter, β, so that both sides of equation 4 are equal.

d. Use equation 5 to determine the value of the scale parameter, η.

$$\sum_{i=1}^{r}\frac{\ln t_i}{r} = \left(\sum_{i=1}^{n} t_i^{\beta}\ln t_i\right)\left(\sum_{i=1}^{n} t_i^{\beta}\right)^{-1} - \left(\frac{1}{\beta}\right) \quad (4)$$

$$\eta = \left(\sum_{i=1}^{n} t_i^{\beta}/r\right)^{\frac{1}{\beta}} \quad (5)$$

6. Reduced Bias Adjustment

The ML estimate of the Weibull distribution scale parameter η has negligible bias, even for relatively small sample sizes. In contrast, the ML estimate of the shape parameter β is known to be strongly biased for small sample sizes (Makalic & Schmidt, 2023).

A method to adjust the shape parameter is the reduced bias adjustment (RBA), proposed in (Dodson, 1994); this method suggest making the correction multiplying the ML β by $C_4^{3.520}$ factor, eliminating the median bias in the ML beta (see equation 6.)

$$\beta_{unbiased=}\beta(RBA_{factor)} = \beta(C_4^{3.52}) \quad (6)$$

III. Application and results

A set of four identical prototypes CR2032 Li-S batteries were evaluated. The cathode was fabricated with carbon from agave bagasse waste; composition of cathode was 10% of carbon black, 70% of a combination of sulfur and carbon from agave, and 10% of PDVF (agglutinant). The characteristics of battery can be seen in table 1 and it can be appreciated that has 3 volts (V) and 3.3 mAh of nominal capacity.

Characteristics of battery	
Type	CR2032
Cathode Material	Li-S
Nominal Capacity	3.3 mAh
Nominal Voltage	3 volts

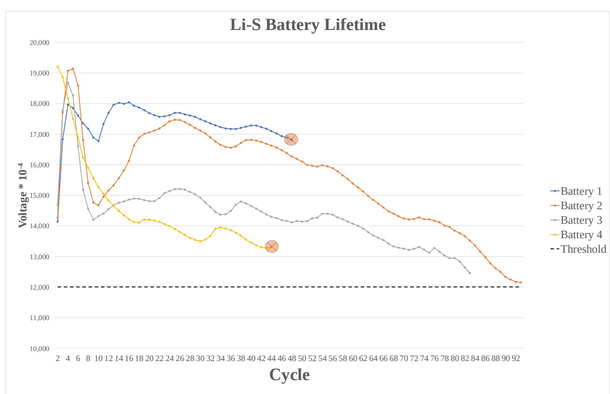
Table 1 Battery characteristics

The battery test was performed involving continuous cycling of a Li-ion battery. The initial charging conditions were set to 45 min up to reach 3v; the cycling protocol was set to a period of 1 min in rest, 45 min in a step of charging voltage constant of 3v, 1 min rest and then a step of constant resistant discharge to 5 kΩ. The end of life of the battery was considered when voltage v<1.2 (threshold line).

A summary of the four tested batteries is shown in table 2. In this table can be observable the time to failure (cycles) of each battery; if the experiment was stopped, then the data is censored. A value of 1 implies that the data is censored and a value of 0 implies that the experiment was complete, and the battery has reach its end of life (EOL).

Battery	Time to failure (cycles)	Censored 1 = Yes 0 = No
Battery 1	48	1
Battery 2	93	0
Battery 3	83	0
Battery 4	45	1

Table 2 Time to failure for batteries



Graphic 1 Li-S Battery lifetime (cycles) for each battery

In graphic 1 can be appreciated, graphically, the lifetime of each evaluated battery (in cycles). The experiments of batteries 1 and 4 had to be stopped before the end of life, while batteries 2 and 3 achieved their EOL.

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In order to estimate the RUL for each battery, the next 3 step procedure was employed.

1. Application of goodness-of-fit test to determine the probability distribution of data set.

2. Estimation of the two parameters of Weibull distribution with censored data (shape and scale parameters).

3. Reduced Bias Adjustment for shape parameter β.

4. Estimation of reliability key battery indices for reliability analysis.

Step 1 – Goodnes-of-fit test.

Using software Minitab to determine if the mean time to failure of battery data set fits to Weibull probability distribution was performed. In figure 1 can be observed that four most commons distributions were proposed (Weibull, Normal, Lognormal, Exponential) and according to section 4 data were evaluated using the Anderson-Darling statistic. Since the Anderson-Darling statistics for the Weibull distribution has a value of 5.389 and there is no significant difference between the four-distribution proposed, it is possible to determine that the MTTF of batteries follows a Weibull distribution and then the analysis Weibull can be used to estimate parameters and to make reliability analysis.

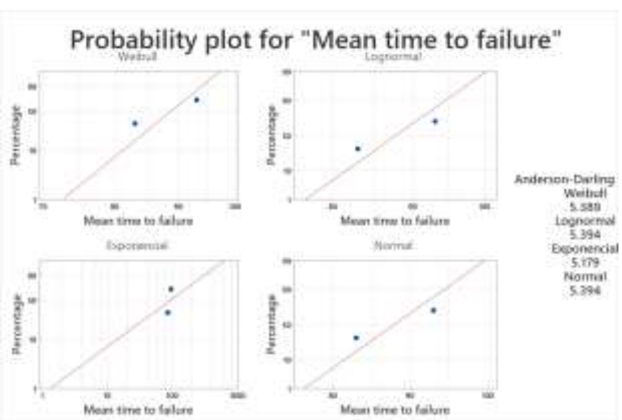

Figure 1 Probability plot for MTTF

Step 2 – Estimation of the two parameters of Weibull Distribution

Since the exponential distribution is a particular case of Weibull distribution (with shape parameter β=1), in this work we will approach the analysis in a general way through the Weibull analysis.

Assuming that MTTF follows a Weibull Distribution and according to equations 4 and 5, and the procedure of section 5, the ML method was used to estimate the two-parameters of the Weibull Distribution (shape and scale parameters); as a result, the scale parameter η=90.3649, meaning that the expected characteristic lifetime of evaluated batteries is around 90 cycles; meanwhile the shape parameter β=21.0918. According to the value of β, it can be interpreted that the battery EOL is caused by wear-out failures.

Step 3 – Reduced Bias Adjustment for shape parameter β

The ML estimate of the shape parameter β is known to be strongly biased for small sample sizes, then it is necessary to adjust according to equation 6 having the next values:

$$\beta_{unbiased=}\beta(RBA_{factor)} = 21.0918(0.797885)^4 = 9.5263$$

Step 4 – Reliability analysis

A reliability analysis has been conducted to obtain key indicators that will estimate some expected characteristics of lifetime based on the sampled tested.

Parameter estimation for Weibull Distribution		
	β	β_{unbiased}
	23.5734	9.5263
η	90.6387	
Reliability Indicators		
Mean time to failure (cycles)	88.57	86.05
Reliability	0.4407	0.4564
Failure probability	0.5593	0.5436
Variance (MTTF)	21.9039	117.4504
Standard deviation (MTTF)	4.6802	10.8375

Table 3 Reliability key indices of Li-S battery based on tested sample

In table 3 is observed that a summary of shape and scale parameters of Weibull Distribution for battery data set; in one side is the shape parameter β without RBA, and in the other side is the b parameter with RBA, $\beta_{unbiased}$ (calculated based on equation 6). Based on shape and scale parameters it is possible to calculate the expected MTTF, which is 88.57 cycles considering normal parameters and 86.05 cycles when unbiased shape parameter is considered.

Taking these values, it is possible to obtain another reliability key indicators such as reliability (estimation based on equation 3), meaning this as the probability that the battery will function at the point of the expected MTTF; another indicator is the probability to fail, calculated according to equation 2. And finally, it is possible to find the variance and the standard deviation even for estimation without RBA and the one with RBA. As it can be observed, there is not a notorious difference in MTTF, reliability, failure probability. Even though, there is a significant difference in the values in variance (21.9039 vs 117.4504) and consequently in standard deviation (4.6802 versus 10.8375). This mean that the application of RBA to β parameter in the estimation is adjusting those values because of the small and highly censored sample.

Conclusions

The estimation of RUL in Li-S batteries implies difficulties to determine it, because of the lack of data and time to evaluate experimentally sufficient batteries to obtain information about their life. In the present work it was possible to predict the lifetime phenomenon of four Li-S batteries which cathode was fabricated with carbon coming from agave bagasse waste.

Through the Weibull distribution and the RBA method is possible to predict RUL of batteries, their characteristic lifetime, expected MTTF, probability to fail and probability to perform consistently well (reliability) and then estimate some uncertainty information such as variance and standard deviation.

Notwithstanding the aims of this work is predict battery lifetime as an estimation (a confidence interval), there is no difference between the two means (biased and unbiased); however, in the variance there is, so it becomes necessary to include the confidence interval for the MTTF considering variance of both β estimators to make a comparison.

As it can be appreciated the application of RBA method in those calculations has an effect in its variance/standard deviation of expected MTTF due to the highly censored reduced sample. But at the end, these considerations in the way how the reliability indices are estimated, is possible to consider the available information (from a highly censored sample) in a capable method that predicts RUL.

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