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On the Theory of the Arrhenius-Normal Model with Applications to the Life Distribution of Lithium-Ion Batteries

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Abstract: Typically, in accelerated life testing analysis, only probability distributions possessing shape parameters are used to fit the experimental data, and many distributions with no shape parameters have been excluded, including the fundamental ones like the normal distribution, even when they are good fitters to the data. This work shows that the coefficient of variation is a replacement for the shape parameter and allows using normal distributions in this context. The work focuses on the Arrhenius-normal model as a life-stress relationship for lithium-ion (Li-ion) batteries and precisely derives the estimating equations of its accelerating parameters. Real and simulated lives of Li-ion batteries are used to validate our results.

Keywords: Li-ion; goodness-of-fit tests; ALT; coefficient of variation; Arrhenius model; MLE

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1. Introduction

Due to their high energy density, high power density, and declining costs, lithium-ion batteries are a promising technology for energy storage [1–3]. Lithium-ion battery life distribution predictions are necessary for the commercialization of batteries for a variety of uses [4,5]. A product's lifetime, warranty period, and many other essential measurements could be misjudged if a proper life distribution is not predicted or if its characteristics are incorrectly estimated [6,7].

In spite of its importance in statistics, as it describes many natural variables well, the normal distribution is less commonly used in life data analysis than other distributions because its left tail extends to negative infinity and because it lacks a shape parameter, making it hard to utilize in accelerated life analysis [8,9]. In accelerated life tests (ALT), products of interest are exposed to a number of harsher conditions than the use conditions in order to obtain immediate information regarding their life distribution [10,11]. These conditions are extrapolated to estimate the life distribution under use conditions using an appropriate life-stress relationship, such as the Arrhenius [12,13], Eyring [14,15], and inverse power law relationships [16,17]. The Arrhenius model has been frequently used when the accelerating stress is thermal, where it is integrated with an appropriate statistical life model. In ALT, the life model for Li-ion batteries is usually assumed to be Weibull or lognormal [18,19], where these distributions possess shape parameters, and the assumption that allows us to extrapolate the accelerated lives to the working life is that the shape parameter remains constant for several stress levels, ensuring that the failure mechanism remains the same.

While the theory of the Arrhenius-Weibull [20–22] and lognormal [23–25] models is well-established and has been addressed in numerous prior works, the theory of the Arrhenius-normal distribution has not been discussed even in the main references of accelerated testing. This prevents us from achieving precise estimates when the experimental data is, actually, normally distributed like the Li-ion batteries and forces us to adopt more complicated models like the three-parameter Weibull distribution [26–28] or the generalized gamma distribution [29,30]. Engineering data are frequently described using the normal distribution [31]. It is an excellent life model for fatigue [32,33], software reliability growth models [34], and general degradation models [35]. It is a symmetrical and easily understood distribution that is also known as the Gaussian distribution. Particularly for recently manufactured batteries, it has been discovered that several Li-ion batteries follow the normal distribution [36].

The primary goal of this work is to extend the Arrhenius presumptions to a variety of distributions, with a particular emphasis on the Arrhenius-normal model. The general assumptions of Arrhenius are reset in Section 2. Section 3 proposes the theory of the Arrhenius-normal model and the estimating equations of its accelerating parameters. The theory of the Arrhenius-normal model is applied to the life distribution of Li-ion batteries in Section 4. Section 5 serves as the paper's conclusion.

2. Generalizing the Arrhenius Model

Suppose that $X \sim F(\xi_X)$ and $Y \sim F(\xi_Y)$ are two random variables following the same statistical distribution F, and describing the life distribution of a product under two different stresses S_X and S_Y such that ξ_X and ξ_Y are the parameter spaces of X and Y, respectively. If it is further assumed that the stress S_Y is more severe than S_X, then there exists a scalar $A > 1$ such that:

$$X = A * Y. \tag{1}$$

The acceleration factor [37,38], or scalar A in (1), is a quantity that connects a product's life at stress S_X to its life at stress S_Y.

By applying (1) to the expected value (E) and standard deviation (SD), we obtain

$$E(X) = A * E(Y), \tag{2}$$

$$SD(X) = A * SD(Y). \tag{3}$$

Now, dividing (3) by (2), we arrive at an equation without an acceleration factor.

$$\frac{SD(X)}{E(X)} = \frac{SD(Y)}{E(Y)}, \tag{4}$$

in which the left and right sides, respectively, are the coefficients of variation (c) [39,40] of X and Y.

$$c(X) = c(Y). \tag{5}$$

When a life model possesses a shape parameter, the primary assumption that validates using the Arrhenius or any other life stress relationship with the life model is that its shape parameter is free of the stress and assumed to be constant, as illustrated in many previous works. Among all we mention the solar cells [41] and light-emitting diodes [42], which are assumed to follow Weibull distribution, that were later shown to better follow lognormal distribution as in [43], respectively. See also [44] and along similar lines [45], in which the life of the lithium-ion battery is shown to be acceptably Weibull, and Ref. [46] compares the Weibull with lognormal and inverse normal models. Recently, Ref. [47] proves using standard statistical goodness of fit tests [48,49] that the normal and lognormal distributions achieve much higher p-values [50,51] than the Weibull distribution based on real lifetime data generated from a well-designed experiment.

This assumption is the key point that makes Arrhenius a legitimate life-stress relationship in addition to the usual assumption of Arrhenius model, which assumes that the life must be an Arrhenius function of a positive stress S, where, in this work, S denotes the thermal stress expressed by temperature, that is:

$$l(S) = e^{a+b/S} \text{ or } log l(S) = a + b/S \tag{6}$$

where l is any life measure, such as their mean life, typical life, median life, etc. The model acceleration parameters a, b are to be determined.

This work argues that the constancy of the shape parameter assumption can be replaced with a more general assumption, which assumes that the *coefficient of variation* (c) must remain constant and independent of the stress. That is, under normal work stress S_0 and various accelerated stresses $S_1, S_2, \cdots, S_m$ then:

$$c_0 = c_1 = c_2 = \cdots = c_m, \tag{7}$$

where c_j is the coefficient of variation at stress S_j.

Table 1 depicts some examples of the coefficient of variation for several commonly used life distributions in accelerated life testing analysis.

Table 1. The coefficient of variation for common life distributions that have shape parameters.

Distribution	Coef. of Var. (c)
Weibull (α : shape, β : scale)	$c(\alpha)=\sqrt{\Gamma(1+2/\alpha)/\Gamma^2(1+1/\alpha)-1}$
Lognormal (ρ : shape, γ : scale)	$c(\rho)=\sqrt{exp(\rho^2)-1}$
Gamma (k : shape, θ : scale)	$c(k)=1/\sqrt{k}$
Log-logistic (η : shape, υ : scale)	$c(\eta)=\sqrt{\eta tan(\pi/\eta)/\pi-1}$

It can be seen from Table 1 that the coefficients of variation are pure functions of the shape parameters and free of the other parameters. That is, the shape parameter is constant if and only if the coefficient of variation is constant. This means that assumption (7) completely agrees with the primary assumption of the Arrhenius model, but it is more general and can be extended to distributions that do not have shape parameters like the normal distribution.

3. The Arrhenius-Normal Model

In this section, the new assumption (7) is utilized to develop the Arrhenius-Normal model by providing the estimating equation of the accelerating parameters a and b, and derives the maximum likelihood estimator (MLE) of the general coefficient of variation of the model under this assumption.

If the life model is assumed to be normal with mean μ and standard deviation σ, where the mean μ is chosen as the life characteristic, then (6) will take the form

$$log\mu = a + b/S, \tag{8}$$

and under the assumption (7), the standard deviation will be:

$$\sigma = ce^{a+b/S}, \tag{9}$$

where $c = \sigma/\mu$ is the coefficient of variation.

Now, the target is to estimate the accelerating parameters a and b, and the coefficient of variation c from experimental data of an accelerated life test.

Mathematically speaking, suppose that an accelerated test is conducted at m constant stresses, $S_1, S_2, \cdots, S_m$ on m groups of items until the failure of all of them. This would produce m samples of failure times, of sizes, say $n_1, n_2, \cdots, n_m$, respectively. If we denote by $t_1^{(j)}, t_1^{(j)}, \cdots, t_{n_j}^{(j)}$ the failure times at the jth stress S_j, the likelihood function of the sample at this stress can be written as:

$$f_j\left(t_1^{(j)}, t_1^{(j)}, \cdots, t_{n_j}^{(j)}\right) = \prod_{i=1}^{n_j} f\left(t_i^{(j)}\right), \tag{10}$$

where $f\left(t_i^{(j)}\right)$ is the probability density function (PDF) of the normal distribution with mean μ_j and standard deviation σ_j associated with stress S_j. This PDF is given by:

$$f\left(t_i^{(j)}\right) = \frac{1}{\sqrt{2\pi}\sigma_j} exp\left\{\frac{-1}{2\sigma_j{}^2}\left(t_i^{(j)} - \mu_j\right)^2\right\}. \tag{11}$$

Consequently, the likelihood of the j^{th} sample has the form

$$L_j = \prod_{i=1}^{n_j} f\left(t_i^{(j)}\right) = \left(\frac{1}{\sqrt{2\pi}\sigma_j}\right)^{n_j} exp\left\{\frac{-1}{2\sigma_j{}^2}\sum_{i=1}^{n_j}\left(t_i^{(j)} - \mu_j\right)^2\right\}. \tag{12}$$

Therefore, the likelihood of the whole experiment under all stresses $S_1, S_2, \cdots, S_m$, is given by:

$$= \prod_{j=1}^{m} L_j = \left(\frac{1}{\sqrt{2\pi}}\right)^{\sum_{j=1}^{m} n_j} \prod_{j=1}^{m} (\sigma_j)^{-n_j} exp\left\{\sum_{j=1}^{m}\frac{-1}{2\sigma_j{}^2}\sum_{i=1}^{n_j}\left(t_i^{(j)} - \mu_j\right)^2\right\}. \tag{13}$$

By assumption (7), since the general coefficient of variation c is constant and independent of stress, then for all j, we have:

$$c = \sigma_j/\mu_j. \tag{14}$$

In view of (14), when σ_j is replaced in (13) by $c\mu_j$, the general log-likelihood becomes:

$$\psi = log(L) = constant - \sum_{j=1}^{m} n_j log\left(c\mu_j\right) - \frac{c^{-2}}{2}\sum_{j=1}^{m}\frac{1}{\mu_j{}^2}\sum_{i=1}^{n_j}\left(t_i^{(j)} - \mu_j\right)^2. \tag{15}$$

Setting the partial derivative of ψ in (15), with respect to c, yields the maximum likelihood estimate of the square of the coefficient of variation:

$$c^2 = \frac{\sum_{j=1}^{m}\frac{1}{\mu_j{}^2}\sum_{i=1}^{n_j}\left(t_i^{(j)} - \mu_j\right)^2}{\sum_{j=1}^{m} n_j}. \tag{16}$$

It is a very well-known fact that the best estimates of the mean $\left(\hat{\mu}_j\right)$ and standard deviation $\left(\hat{\sigma}_j\right)$ of the normal distribution are the sample mean and standard deviations. That is, for the j^{th} stress:

$$\hat{\mu}_j = \frac{\sum_{i=1}^{n_j} t_i^{(j)}}{n_j}, \tag{17}$$

$$\hat{\sigma}_j^2 = \frac{\sum_{i=1}^{n_j}\left(t_i^{(j)} - \hat{\mu}_j\right)^2}{n_j}, \tag{18}$$

Accordingly, the estimating equation of c is immediately obtained by replacing the parameters μ_j and σ_j in (16) with their estimates in (17) and (18), that is:

$$\hat{c} = \sqrt{\frac{\sum_{j=1}^{m} n_j \hat{c}_j^2}{\sum_{j=1}^{m} n_j}}, \tag{19}$$

where, $\hat{c}_j = \hat{\sigma}_j/\hat{\mu}_j$ is the j^{th} sample coefficient of variation.

Once Equation (17) estimates the mean μ_j at each stress S_j, we can apply the usual least squares method (LSM) to estimate the two accelerating parameters a and b from Equation (8), for the paired data $\left(\frac{1}{S_j}, \, log\hat{\mu}_j\right)$ for $j = 1, 2, \cdots, m$. The LSM estimating equations are:

$$\hat{b} = \frac{m\sum_{j=1}^{m} log\hat{\mu}_j/S_j - \sum_{j=1}^{m} log\hat{\mu}_j \sum_{j=1}^{m} 1/S_j}{m\sum_{j=1}^{m}(1/S_j)^2 - \left(\sum_{j=1}^{m} 1/S_j\right)^2}, \tag{20}$$

$$\hat{a} = \frac{\sum_{j=1}^{m} log\hat{\mu}_j - \hat{b}\sum_{j=1}^{m} 1/S_j}{m}. \tag{21}$$

On the other hand, after estimating σ_j from Equation (18), $\hat{\sigma}_j$ and $\hat{\mu}_j$ together yield an estimate for the coefficient of variation at each stress S_j through the relationships

$$\hat{c}_j = \hat{\sigma}_j/\hat{\mu}_j, \text{ for } j = 1, 2, \cdots, m, \tag{22}$$

which, in turn, estimate the general coefficient of variation c by $\hat{c}$. Where $\hat{c}$ is obtained after plugging the $\hat{c}_j$ for $j = 1, 2, \cdots, m$ in Equation (19).

4. The Life Distribution of Li-Ion Batteries

This section revisits the experimental life test data set of Li-ion batteries discussed in [47], which was originally introduced in [45]. This censored data [52] was proved in [47] to follow the normal distribution using the Lilliefors [53], Chi-Square [54], Cramer-von Mises [55], and Jarque-Bera [56] goodness of fit tests, taking the censoring effect into consideration [57]. On the other hand, the efficiency of the censored sample of the normal distribution is proven to be high when tested using the efficiency function [58]. Each cell cycled in the experiment at a temperature of 25 °C with an Arbin BT2000. The cells were charged in a constant current, constant voltage mode at 1 C (4.4 A) constant current up to 4.35 V. This was followed by a constant voltage charge until the current dropped below C/40. Each cell was then discharged at a constant current of 10 C (44 A) until the terminal voltage fell to 3 V. The descriptions of the tested batteries are shown in Table 2, and their failure cycles are shown in Table 3.

Table 2. The description of the commercial Li-ion batteries tested.

Type of Battery	Li-Ion
Nominal capacity	4.4 Ah
Active material of the anodes	synthetic graphite
Active material of the cathode	LCO (Li Cobalt Oxide)
Number of cells tested	24 cells
Temperature	25 °C
Discharge rate	10 C

Table 3. Failure Cycles for the 20 observed batteries.

255	379	497	541
301	408	509	560
326	409	515	
338	430	518	
340	449	537	
341	475	541	

These 24 batteries were placed in an ALT. Only four batteries were still operating after the test's 593 consecutive cycles of charge and discharge; as a result, 20 lives out of

24 batteries were recorded. This scenario is usually called in statistics censoring [59], and the 20 observed failures form a censored sample.

The estimates of the mean and standard deviation of this sample of Li-ion batteries' lives at a use condition of 25 °C are obtained from [52] as:

$$\mu_{25} = 470.4 \text{ cycles}, \quad (23)$$

$$\sigma_{25} = 119.3 \text{ cycles}. \quad (24)$$

Accordingly, the probability distribution function of this type of battery at use conditions of 25 °C is:

$$f_{25}(t) = \frac{1}{119.3\sqrt{2\pi}} e^{-\frac{1}{2}\left(\frac{t-470.4}{119.3}\right)^2}, \quad (25)$$

where t is the number of cycles.

What is going to happen to the life if the thermal working conditions are higher than 25 °C. Recent studies show that an increase of 10 °C to the working temperature would roughly decrease the life to half, see, for example, [60,61]. Hence, according to the proposed theory in this work, both the mean life and standard deviation would decrease by half. This allows us to simulate some data under different stresses in order to validate the theory and provide a heuristic example to readers and practitioners. Here, we use the three thermal stresses of S_{35}, S_{45} and S_{55} where the index denotes the temperature in degrees Celsius. To be consistent with the experimental data, we generate $n_{35} = n_{45} = n_{55} = 20$ lives in cycles for each stress, as shown in Table 4.

Table 4. Simulated failure cycles for batteries at several stress levels.

S_{35}	123	151	167	180	191
	200	208	216	224	232
	239	246	254	262	270
	280	290	303	319	346
S_{45}	62	76	84	90	95
	100	104	108	112	116
	120	123	127	131	135
	140	145	151	159	174
S_{55}	31	38	42	45	48
	50	52	54	56	58
	60	61	63	65	67
	70	72	75	80	86

For the three datasets of Table 4, the estimated values of means, standard deviations, and coefficients of variation are obtained using (17), (18), and (22), respectively, and the results are listed in Table 5.

Table 5. The estimated means, standard deviations, and coefficients of variation of failure cycles for batteries at several stress levels.

Stress	n_j	$\hat{\mu}_j$	$\hat{\sigma}_j$	$\hat{c}_j$
S_{25}	20	470.4	119.3	0.2536
S_{35}	20	235.4	57.7	0.2451
S_{45}	20	118.0	28.7	0.2432
S_{55}	20	58.7	14.2	0.2419

In Table 5, the estimates from the experimental data that appear in Equations (23) and (24) are also added, so we have estimates at 4 stress levels in total.

Substituting the values shown in Table 5 into (20) and (21), one can obtain $\hat{a} = 2.62$ and $\hat{b} = 91.55$. Hence, the accelerated Equation (8) becomes:

$$log\mu = 2.62 + 91.55/S \text{ or } \mu = e^{2.62+91.55/S} \tag{26}$$

An accelerated life curve that might be called a battery's life characteristics curve is depicted in Figure 1, which has been developed using the Mathematica 12.3 package. The correlation coefficient is 0.9735 and is very nearly 1, demonstrating excellent curve-fitting. The outcome also demonstrates that the accelerated model is completely consistent with the Arrhenius model.

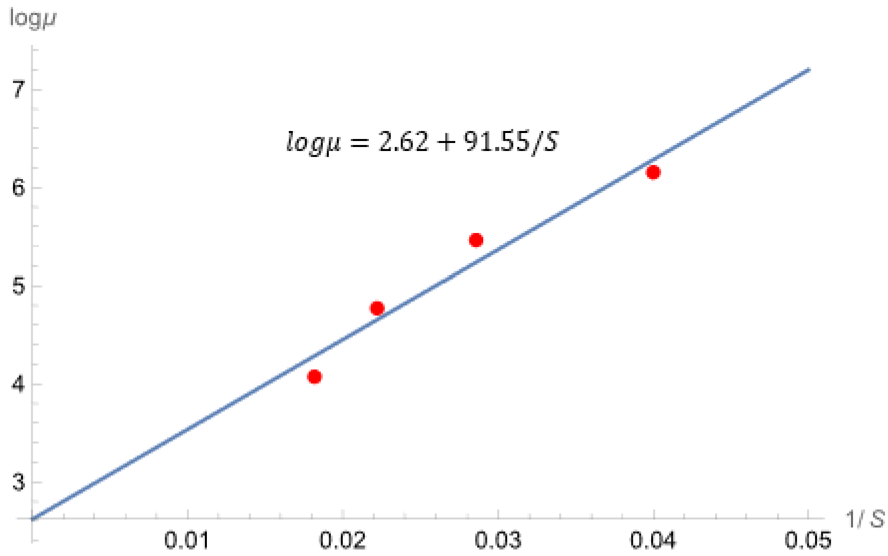

Figure 1. Life characteristic curve.

It can be seen from Table 5 that the mean and standard deviation decrease with the stress, whereas the estimated coefficients of variation $\hat{c}_j$ are almost the same for the different stresses, in agreement with the theory. The general coefficient of variation c can be estimated through (19), which provides the MLE of this quantity:

$$\hat{c} = 0.2460. \tag{27}$$

Note that this value is equal to the usual average of the four values of $\hat{c}_j$ mentioned in Table 5 because the four sample sizes are equal.

The second acceleration Equation (9) follows from (27) and becomes:

$$\sigma = 0.2460\ \mu. \tag{28}$$

The estimates appearing in Table 5 for the mean and standard deviation allow us to pictorially present the battery life distributions for several thermal stresses, as shown in Figure 2. The four curves correspond from left to right to S_{55}, S_{45}, $S_{35,}$ and S_{25}, respectively.

It can be clearly seen that the life distribution is shifted to the right and gets more dispersed when the thermal stress decreases, and vice versa. On the other hand, the acceleration Equations (26) and (28) can predict the battery distribution at stresses that have not been tested experimentally. For example, to predict the life distribution at stress S_{40}, we substitute $S = 40$ in the two equations to obtain the estimates for $\mu_{40} = 135.5$ cycles and $\sigma_{40} = 33.3$ cycles. These estimates are consistent with the trend in Table 5, as 40 lies in the stress range 25–55 used in the table. However, the estimates may significantly vary when it is conducted outside the experimental range.

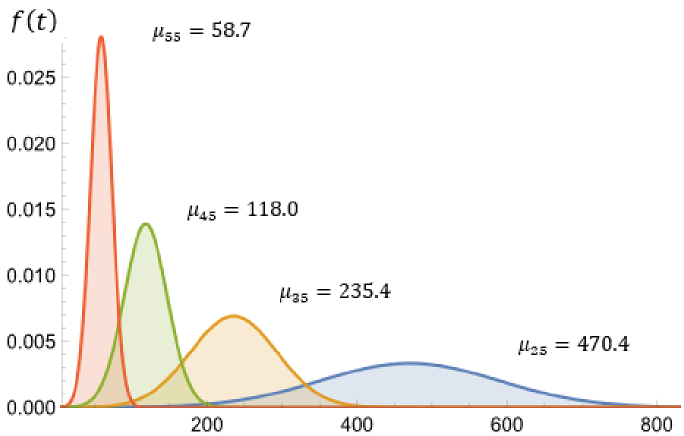

Figure 2. Battery Life Distributions Under Several Thermal Stresses.

Finally, it must be mentioned that the range of possible outcomes $T = t$ of the normal distribution is from $-\infty$ to ∞. Life must, of course, be positive. Thus, the fraction of the distribution below zero must be sufficiently small. This means that the mean μ must be at least three times as great as the standard deviation σ. It can be checked that, for any stress S, the fraction of the distribution with negative life solely depends on the coefficient of variation c and equal to:

$$P(T < 0) = P\left(Z < \frac{-1}{c}\right) \approx 2.4 \times 10^{-5}. \quad (29)$$

which is negligible, where Z is the standard normal random variable.

5. Conclusions and Recommendations

To recap, this paper introduced the Arrhenius-normal model, which was not addressed in previous works to the best of the author's knowledge. The model can be used to extrapolate the life distribution at use conditions from accelerated experimental data. It predicts how time-to-fail varies with temperature and describes the failure mechanisms when the failure-accelerated data is actually normally distributed. In addition to the normality of the experimental data, the key assumption of the Arrhenius-normal model is that, under a use stress and various accelerated stresses, the coefficient of variation keeps unchanged. The model does not invoke any further arbitrary assumptions. The maximum likelihood estimate of the general coefficient of variation is precisely derived and shown to be an explicit function of the estimates of the coefficients of variation at each stress. Although the focus of this work was on the normal distribution, the theory proposed in this work is applicable to any statistical distribution with a defined coefficient of variation even if the desired distribution does not have a shape parameter, as in classical accelerating test analysis practices.

We recommend that researchers apply the proposed theory to many products and materials that are normally distributed, such as light-emitting diodes and solar cells. In addition, study other life-stress relationships like inverse power laws and the Eyring model with normal distribution. Finally, replace the normal distribution with other distributions that do not have shape parameters, such as Gumbel and logistic distributions. The approach presented in this work can also be used to predict the life distribution of other types of batteries, such as all-solid-state sodium [62] and rechargeable aqueous zinc (Zn) batteries [63].

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