Prognostics Health Estimation of Lithium-ion Batteries in Charge-Decay Estimation Uncertainties – A Comparative Analysis

Chinedu I. Ossai [1]

[1]*School of Information Technology & Mathematical Sciences, University of South Australia, Mawson Lakes, GPO Box 2471 Adelaide SA 5001, Australia*

chinedu.ossai@unisa.edu.au, ossaic@gmail.com

ABSTRACT

This study uses nonlinear mixed effect-based degradation modeling that considers the influence of uncertainties on the state-of-charge of lithium-ion batteries to determine the State-of-Health (SOH) of the batteries at different End-of-Life (EOL) failure thresholds. The results of the analysis obtained with lithium-ion batteries data from NASA Ames Centre repository, confirms that the SOH of the batteries is influenced by the uncertainties. This is because the random effects models show a better correlation with the experimental data than the fixed effects models that have not considered uncertainty. It is important therefore that battery prognosis is done in consideration of these parametric uncertainties, to forestall poor estimation of the SOH of the lithium-ion batteries at various stages of the lifecycle. Seeing that the presence of uncertainties could result in unwarranted failures of assets powered by the batteries, due to over-estimation of the remaining useful life (RUL) or capital loss, due to early decommissioning of efficient batteries when the RUL is under-estimated.

1. INTRODUCTION

One of the challenges in asset integrity management is the ability to estimate the reliability of the facilities, as longer lifecycles of assets resulting from advanced designs and manufacturing technologies make it more difficult to obtain failure results from failure tests analysis. Historical failure records from component life tests, which used to be the primary source of information for reliability estimation is not providing enough information for longtime decisions as failure trend data are becoming sketchier. This longevity of components, though a good thing, has unfortunately impacted on the validity and accuracy of existing SOH estimation methodologies as rudimentary life data analysis does not provide enough information for the study of different deterioration mechanisms such as crack, wear, fatigue, corrosion, oxidation, and decay (Wu & Shao 1999). I, therefore, have to make use of degradation trends and physical / phenomenological models of degradation as an alternative tool for the prognostics of components, sub-systems, and systems of assets.

Lithium-ion batteries that are used in many industrial applications, as the source of energy, have also been technologically revolutionized, with enhanced lifecycle duration that has made it challenging to estimate the SOH at different EOL failure thresholds. This challenge has inspired this research that aims to utilize degradation modeling technique of nonlinear mixed effects to establish the future status of the battery charge decay, vis-à-vis determining the SOH of the battery over time, considering the uncertainties. The research on lithium-ion battery SOH has intensified over the recent years with numerous authors working on different techniques that help to estimate the lifetime of the batteries using degradation modeling (Mo, Yu, Tang & Liu, 2016; Hu, Jain, Tamirisa & Gorka, 2014; Xing, Ma, Tsui & Pecht, 2013). For instance, the implementation of the degradation mechanism in the modeling of charge decay of lithium-ion batteries provided the medium for using the Dempster-Shafer theory and Bayesian Monte Carlo simulation to estimate the Remaining Useful Life (RUL) of the batteries (He, Williard, Osterman & Pecht, 2011). Researchers have used a data-driven approach to the estimation of battery RUL, by applying a support vector machine algorithm (Nuhic, Terzimehic, Soczka-Guth, Buchholz & Dietmayer, 2013) whereas relevance vector machine, which is also a machine learning tool was used by other authors (Wang, Miao & Pecht, 2013). Other researchers who have worked on lithium-ion battery degradation modeling using other techniques that were either meant for validation or completion of the prognostic modeling process have only done so by considering the fixed effect parameters, which have a global behavioral pattern that is assumed to be uninfluenced by any uncertainty. Other authors (Liu, Pang, Zhou, Peng & Pecht, 2013) combined Gaussian process functional regression with sigmoidal degradation model for lithium-ion battery SOH estimation while particle filter, Kalman filter, particle swarm optimization, auto-regression and the genetic algorithm have also been combined in one form or another by other researchers (Mo et al., 2016; Orchard, Hevia-Koch, Zhang & Tang, 2013; An, Choi & Kim 2013; Miao, Xie, Cui, Liang & Pecht, 2013; Xian, Long, Li & Wang, 2014; Long, Xian, Jiang & Liu, 2013) to predict the RUL of lithium-ion batteries. Despite numerous researches in lithium-ion battery prognosis, as exemplified by the literatures reviewed above, one of the most fundamental causes of flawed prognostic estimation – uncertainty, has not been considered by most of these studies. Given the fact that this omission could be a fundamental source of faulty prognosis and unreliable RUL estimation, it became imperative that this study explores the influences of uncertainties on the lifetime estimation of lithium-ion battery. This is via a comparative analysis that considered the charge decay estimation with both the fixed effects and random effects (parameters that are individually driven by uncertainties and have the potential of causing imbalance in the state of charge capacity decay). Uncertainties such as manufacturing defects, environmental conditions, experimentation measurement errors, physical and chemical characteristics of the defects in materials (He et al., 2011) can influence the charging and discharging pattern of the batteries. This action is through the electrolyte medium, which allows for transfer of the lithium ions (Li+) between the positive and negative electrodes during the diffusion process (Daigle & Kulkarni, 2016). This makes it imperative to incorporate the impact of these uncertainties in degradation modeling. Again, the SOH of the battery, which is a measure of the stored energy, is dependent on the electron transfer process that is also linked to the diffusion rate of the lithium-ions in the electrolyte.

In this research, the influence of uncertainties in the SOH of the lithium-ion batteries with reference to the battery charge capacity decay will be studied with nonlinear mixed effect degradation model. The aim of which is to determine the influence of the random effects on the prognostics of the lithium-ion battery, by establishing the remaining useful life at 70%, 60% and 50% EOL failure thresholds. The SOH of the battery at these EOL thresholds will be estimated with fixed (uncertainties are not considered) and random effects (uncertainties are considered) and comparison of both will be done to establish the flaws associated with the prognostic health monitoring of lithium-ion batteries without the consideration of uncertainties. The consideration of random effects in prognostic will enhance the accuracy of the RUL estimation of the lithium-ion batteries.

2. DEGRADATION MODELING OF LITHIUM-ION BATTERY CHARGE DECAY

In this study, the lithium-ion battery charge capacity decay has been assumed to follow a physical degradation trend of sigmoidal model per previous researchers (Mo et al. 2016, He et al. 2011). The degradation model uses charge decay (units of Ah) as a measured physical characteristic, with the number of cycles (the period between successful charging and discharging) to develop a charge decay behavioral trend of the battery per Eq. (1),

$$Q(k) = P_1 e^{-r_1 k} + P_2 e^{-r_2 k} \quad (1)$$

where $Q(k)$ represents the charge decay at a given cycle k, P_1 and P_2 represent the constants that are related to the battery internal impedance whereas r_1 and r_2 are the charge decay constants that are related to battery usage. P_1, P_2, r_1, and r_2 are the fixed effect parameters.

Mixed effect modelling as a dynamic estimation concept is vital for predicting the response of parameters in a system that shows significant variabilities but has global behavioral pattern due to the accumulation of uncertainties that can be associated with multiple experimental observations of the battery capacity decay at various charging cycles. Hence, for different experimental replicates (at the same conditions), a multilevel nonlinear mixed effect model can be used to determine the time-dependent battery charge capacity decay at the charging cycles. Since the charge capacity decay observations can be treated as a multilevel nonlinear mixed effect model, it can be classified as a level 1 observation, which is nested to level 2 (charging cycles) that is nested to level 3, involving many observations of experimental replicates. The fundamental form of the responses (battery charge capacity) of parameters in mixed effect models are expected to be the same for some individual parameters (fixed effects) of the population, which in this study has been assumed to be associated with the sigmoidal model parameters shown in Eq. (1). The other variables have individually driven known and unknown effects that generally create an imbalance in the state of the system (random effects). The combination of the fixed and random effects in lithium-ion battery charge decay modelling will result in the inferential knowledge of the model parameters, their influences on the charge fade pattern and parametric interactions.

This work will be focusing on the general form of nonlinear mixed effect model that can be written per Eq. (2).

$$Q_{ij} = f\left(k_{ij}, P_1, r_1, P_2, r_2\right) + \varepsilon_{ij}, 1 \leq i \leq n, 1 \leq j \leq m_i \quad (2)$$

where ε_{ij} is the error, which is assumed to be independent and identically distributed *(iid)* Gaussian number with zero mean and unknown variance $\sigma^2 \rightarrow N\ (0, \sigma^2)$, n is the number of covariates, m_i is the number of observations of i^{th} covariate and f represents the degradation of the lithium-ion battery charge capacity, which is a nonlinear function of the fixed and random effects and Q_{ij} is the actual level of lithium-ion battery charge capacity observed at a given cycle k_{ij}.

Re-parameterization of the charge decay rates in Eq. (1) was done to ensure better fitting of the data, reduce error inherent in the data and increase the chances of obtaining optimal solutions. This can be seen in the expression shown in Eq. (3).

$$\begin{cases} Q_{ij} = P_1 e^{-\left(e^{\lambda_1 k_{ij}}\right)} + P_2 e^{-\left(e^{\lambda_2 k_{ij}}\right)} + \varepsilon_{ij} \\ \lambda_1 = \log(r_1)\,, \lambda_2 = \log(r_2) \end{cases} \tag{3}$$

The charge capacity decay over the charging cycles can now be expressed by Eq. (4).

$$Q_{ij} = f\left(k_{ij}, P_1, \lambda_1, P_2, \lambda_2\right) + \varepsilon_{ij}, 1 \le i \le n, 1 \le j \le m_i \tag{4}$$

If β represents the parameters such that $\phi_{ij} = \{P_{1i}, \lambda_{1ij}, P_{2i}, \lambda_{2ij}\}$ denotes the individual observation, i for cycle k_{ij}, then the relationship can be further expressed in Eq. (5).

$$\begin{cases} \phi_{1ij} = P_{1i} = \alpha_1 + \theta_{1i} \\ \phi_{2ij} = \lambda_{1ij} = \alpha_2 + \theta_{2i} \\ \phi_{3ij} = P_{2i} = \alpha_3 + \theta_{3i} \\ \phi_{4ij} = \lambda_{2ij} = \alpha_4 + \theta_{4i} \end{cases} \tag{5}$$

where $\theta_i = (\theta_{1i}, \ldots, \theta_{4i})^T$ represents the random effects vector of the parameters and $\alpha_i = (\alpha_{1i}, \ldots, \alpha_{4i})^T$ represents the fixed effect vector of the model. Hence, the charge capacity of the lithium-ion battery can thus be represented by Eq. (6) in consideration of the expression in Eq. (5).

$$Q_{ij} = f\left(k_{ij}, \alpha, \theta_i\right) + \varepsilon_{ij}, 1 \le i \le n, 1 \le j \le m_i \tag{6}$$

Since the vector combining the fixed and random effects, which are the known values (measured charge decay at the charge cycles) and the unknown values (random effects), is represented by ϕ, the relationship in Eq. (7) follows.

$$\phi_{ij} = A_{ij}\alpha + \beta_{ij}\theta_i \tag{7}$$

where A_{ij} is the matrix for combining the fixed effects while β_{ij} is the matrix for combining the random effects.
It, therefore, follows that the charge capacity decay at the charging cycles of the lithium-ion battery can be represented by Eq. (8).

$$\begin{cases} Q_{ij} = f\left(\phi_{ij}, k_{ij}\right) + \varepsilon_{ij}, 1 \le i \le n, 1 \le j \le m_i \\ \theta_i \underset{\sim}{iid} N(0, \psi) \end{cases} \tag{8}$$

where ψ is the covariance of the random effects.

To solve for the fixed and random effect parameters in Eq. (2), different techniques that include least square estimation, Bayesian hierarchical method and Maximum Likelihood Estimation (MLE) (Kuhn & Lavielle, 2005; Wu & Shao, 1999) can be adopted. In this research, the MLE technique has been used for estimating the values of the parameters because of the robustness and generality of the technique for solving problems with known and unknown values (Harter & Moore, 1965; Aslam, Kazim, Ahmad & Shah, 2014), since, the random effect represents the unknown values associated with the observed values of the charge decay at different cycles that represent the known values based on the sigmoidal equation (Eq. 1). If L represents the likelihood function for the charge capacity decay, it will be estimated by using the joint density of the known parameters and its unknown counterparts, as shown in Eq. (7) (Kuhn & Laville, 2005; Aslam et al., 2014).

$$L\left(\phi_{ij}, Q_{ij}\right) = \prod_{i=1}^{n}\prod_{j=1}^{m_i} P\left(Q_{ij}, \phi_{ij}\right)$$

$$= \prod_{i=1}^{n}\prod_{j=1}^{m_i} \int P\left(Q_{ij}, \theta_i, \phi_{ij}\right)\, d\theta \tag{7}$$

where $P\left(Q_{ij}, \phi_{ij}\right)$ represents the probability density function of the nonlinear battery charge decay denoted by f, which depends on the experimental observed lithium-ion battery charge capacity decay at different cycles and the unknown random effect parameters.

Since the maximum likelihood estimator for the parameter ϕ results in the maximization of the value of the likelihood function L, the solution of the MLE problem is obtained by determining the logarithmic value of L as per Eq. (8) (Harter & Moore, 1965; Aslam et al., 2014).

$$\frac{\partial \log(L)}{\partial \theta} = 0 \tag{8}$$

Due to the complexity of Eq. (7) and Eq. (8), a numerical approach that solves for the model parameters iteratively is utilized. For this study, stochastic approximation expectation and maximization technique that uses Markov Chain Monte Carlo (MCMC) simulation in a Metropolis-Hasting computational framework has been adopted.

3. DATA ACQUISITION SETUP AND FAILURE CRITERIA DEFINITION

The fact that the use of lithium-ion batteries results in the loss of charge capacity over time of usage, due to aging (Daigle & Kulkarni, 2016; Broussely et al., 2005; Sarre, Blanchard & Broussely, 2004), which is related to the number of charging cycles, makes the degradation continuous and irreversible, thereby making $f\,(k_{ij}, \alpha, \theta_i)$ a decreasing function with the number of cycles. The failure of the battery is expected to occur when a certain critical threshold, f_c, of the degradation which has been taken as 70%, 60% and 50% of the original retained charge capacity of the lithium-ion battery, is reached. This End-of-Life (EOL) charge capacity corresponds to the expected failure charge cycle of the batteries and forms the basis for the comparative study of the battery SOH estimated with the fixed and random effect models.

Lithium-ion battery data from NASA® AMES research Centre (Saha & Goebel, 2007) was used for this study, considering four sets of battery data – B0025, B0026, B0027 and B0028, obtained via a three-stage operational profile of charging, discharging and impedance at 24ºC. The lithium-ion batteries were charged at constant current of 1.5A until the battery voltage reached 4.2V and the voltage was maintained until the charge current dropped to 20mA. The discharge was done at a constant current level of 2A until the battery voltages fell to 2.7V, 2.5V, 2.2V and 2.5V for batteries B0025, B0026, B0027 and B0028 respectively while the impedance measurement was carried out from 0.1Hz to 5kHz using the electrochemical impedance spectroscopy (EIS). The repeated charging and discharging accelerated the battery aging mechanism whereas the impedance measurement helped to understand the changing trend of the internal mechanisms with the progression of the battery deterioration. The experimental results of the four batteries are shown in Fig 1.

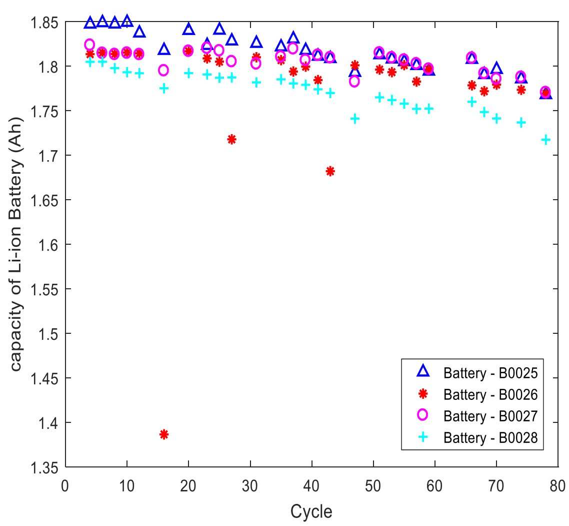

Fig 1: Experimental data of Lithium-ion battery charge capacity-decay for B0025, B0026, B0027 and B0028.

4. RESULTS AND DISCUSSION

In this study, the battery charge capacity at the cycles is used as an indicator of the state-of-health of the batteries because the stored energy in the battery cells reflects the charge capacity of the batteries (Hu et al., 2014). With the initial charge capacity of 2 Ah, it is expected that the EOL of the battery, which was assumed to be at 70%, 60% and 50% of the retained original charge of the battery, will be reached when the charge capacity of the batteries dropped to 1.4 Ah, 1.2 Ah and 1.0 Ah respectively for 70%, 60% and 50% of EOL. The parametric values of the fixed and random effects of the batteries were obtained by MLE using the stochastic approximation expectation and maximization algorithm (see Table 1).

The information in Table 1 was used to determine the fixed and random effects of the batteries, which was compared with the experimental data in Fig 2. This figure indicated that the random effect was a better fit to the experimental data than the fixed effect. To further validate this result, the Root Mean Square Error (RMSE) shown in Eq. (9) was used as a metric to compare the errors in describing the real data.

$$RMSE = \sqrt{\frac{1}{n_i}\sum_{i=1}^{n_i}\left(Q_{exp} - Q_{pred}\right)^2} \tag{9}$$

Table 1: Fixed and random effect parametric values of the lithium-ion battery charge capacity decay model.

Fixed effect Parameters		Random effect parameters				
	Value		B0025	B0026	B0027	B0028
P_1	1.8066	Ψ_{11}	1.8066	1.8066	1.8066	1.8066
λ_1	5.7847	Ψ_{22}	6.6627	5.1922	6.2691	4.6680
P_2	1.8117	Ψ_{33}	1.8366	1.7896	1.8210	1.7994
λ_2	-8.2961	Ψ_{44}	-7.9857	-8.6873	-8.2803	-8.0179

where Q_{pred} represents the predicted battery charge capacity with fixed or random effect, Q_{exp} represents the battery charge capacity obtained from experiment and n_i represents the number of samples.

The results of the RMSE of the fixed and random effects at 99% confidence interval, which is shown in Table 2, further confirm that the RMSE of the random effect models is smaller than those of the fixed effect model for the batteries. This is an indication that the random effect predicted the battery charge decay better than the fixed effects, a proof that the uncertainties associated with battery manufacturing, measurement and environmental conditions indeed have a notable influence on the battery charge capacities.

This result has ramifications for the prognostic health monitoring of lithium-ion batteries, since the estimated charge capacity at the EOL will deviate a little or significantly from the actual battery charge capacity, depending on the use of fixed effect or random effect models (Fig 3). However, the fact that random effect models have consistently shown better prediction across the battery data set is an indication that it is a better prediction model. Figs 3 also clearly showed the variation in the predicted future battery charge capacities at different charge cycles with battery B0027 having the least variation among all the battery sets. This could be attributed to the limited influence of uncertainties on the battery in comparison to batteries B0025, B0026 and B0028.

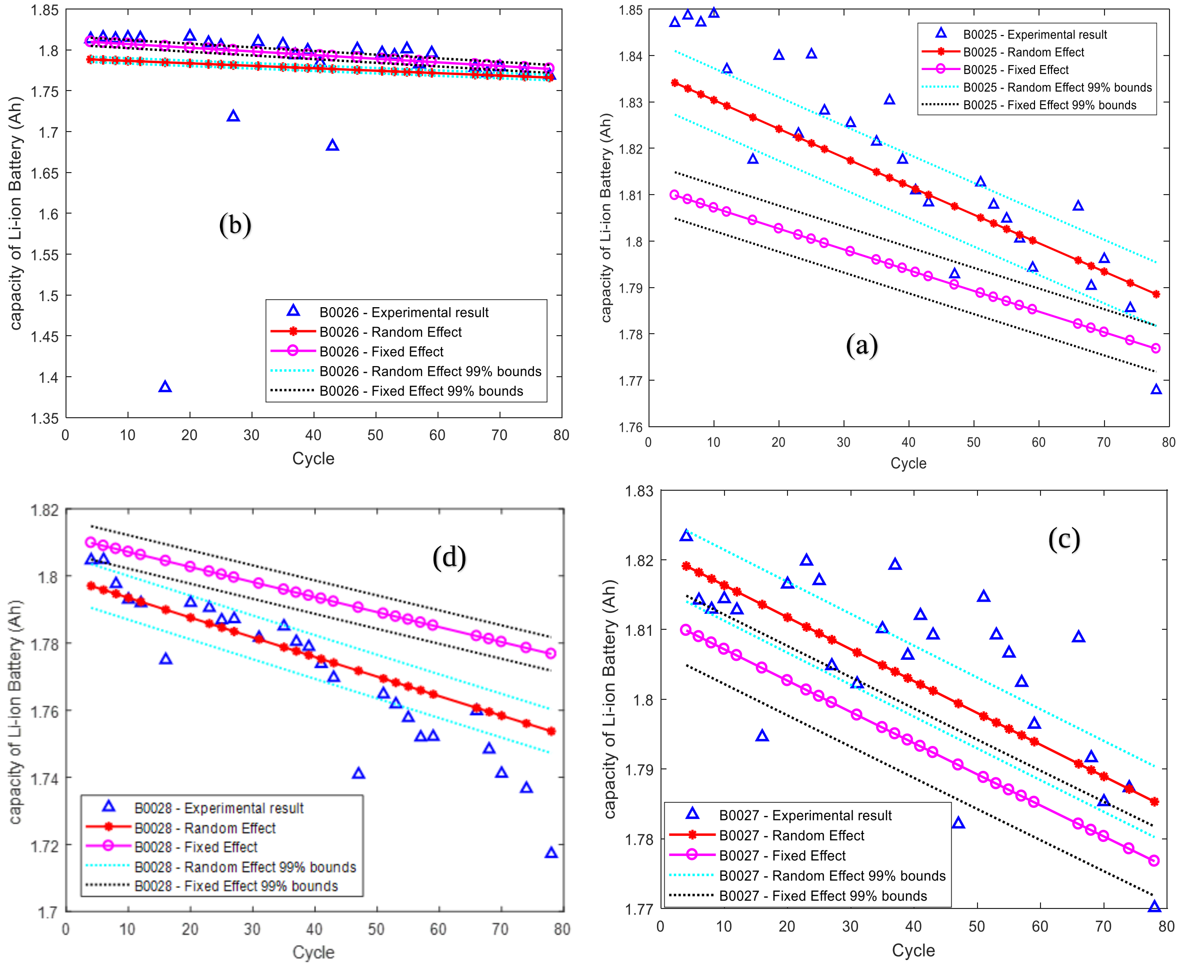

Fig 2: Comparison of the Random and Fixed effects estimations of battery charge capacity decay with the experimental results for batteries – (a) B0025, (b) B0026, (c) B0027 and (d) B0028

Table 2: RMSE of the batteries obtained at 99% confidence interval considering fixed and random effects.

Battery	Fixed effect			Random effect		
	Lower bound	Mean	Upper bound	Lower bound	Mean	Upper bound
B0025	0.1471	0.1608	0.1741	0.1016	0.1042	0.1223
B0026	0.2899	0.2916	0.2938	0.287	0.2871	0.2874
B0027	0.1063	0.121	0.1368	0.0993	0.0987	0.1095
B0028	0.1496	0.1628	0.1758	0.1063	0.111	0.1271

The Probability Density Function (pdf) of the charging cycles of the batteries is exemplified at 70% EOL (Fig 4). The figures distinctively showed the variation in the predicted EOL cycles at the failure thresholds. Again, B0027 can be clearly seen to have limited variation at the failure thresholds, when compared with the other batteries.

The information about the EOL charging cycles at the failure thresholds in Fig 4 provides an additional indication of the expected flaws in the prognostic health estimation of the batteries when the random effect is not considered. Table 3 shows the estimated EOL cycles of the lithium-ion batteries at 70%, 60% and 50% failure thresholds for the fixed and random effect models. The percentage variation of the EOL cycles of the random effects (K_{RE}) and fixed effects (K_{FE}) model was determined with Eq. (10).

$$Variation\ (\%) = \frac{K_{RE} - K_{FE}}{K_{RE}} * 100\% \tag{10}$$

Since the RMSE of the random effect model is smaller than that of the fixed effect model (Table 2), it can be inferred from Table 3 that predicting the state-of-health of the batteries with fixed effects model will result in prognostic health estimations that will vary considerably with those predicted by random effects model. This scenario will have significant ramification for cost and reliability of assets that depend on the batteries for energy. For instance, at 70% EOL threshold, battery B0025 is expected to fail 30% earlier with the random effect model prediction than when it will fail with fixed effect modeling. This can result in unplanned breakdown and the associated consequences if fixed-effect model was used for the prognosis. Moreover, decommissioning of the battery due to shorter EOL cycle prediction with fixed effect model can also occur. This is evident with battery B0026 that will work 29% more, when prognostics is done with random effects model than when fixed effect model is used.

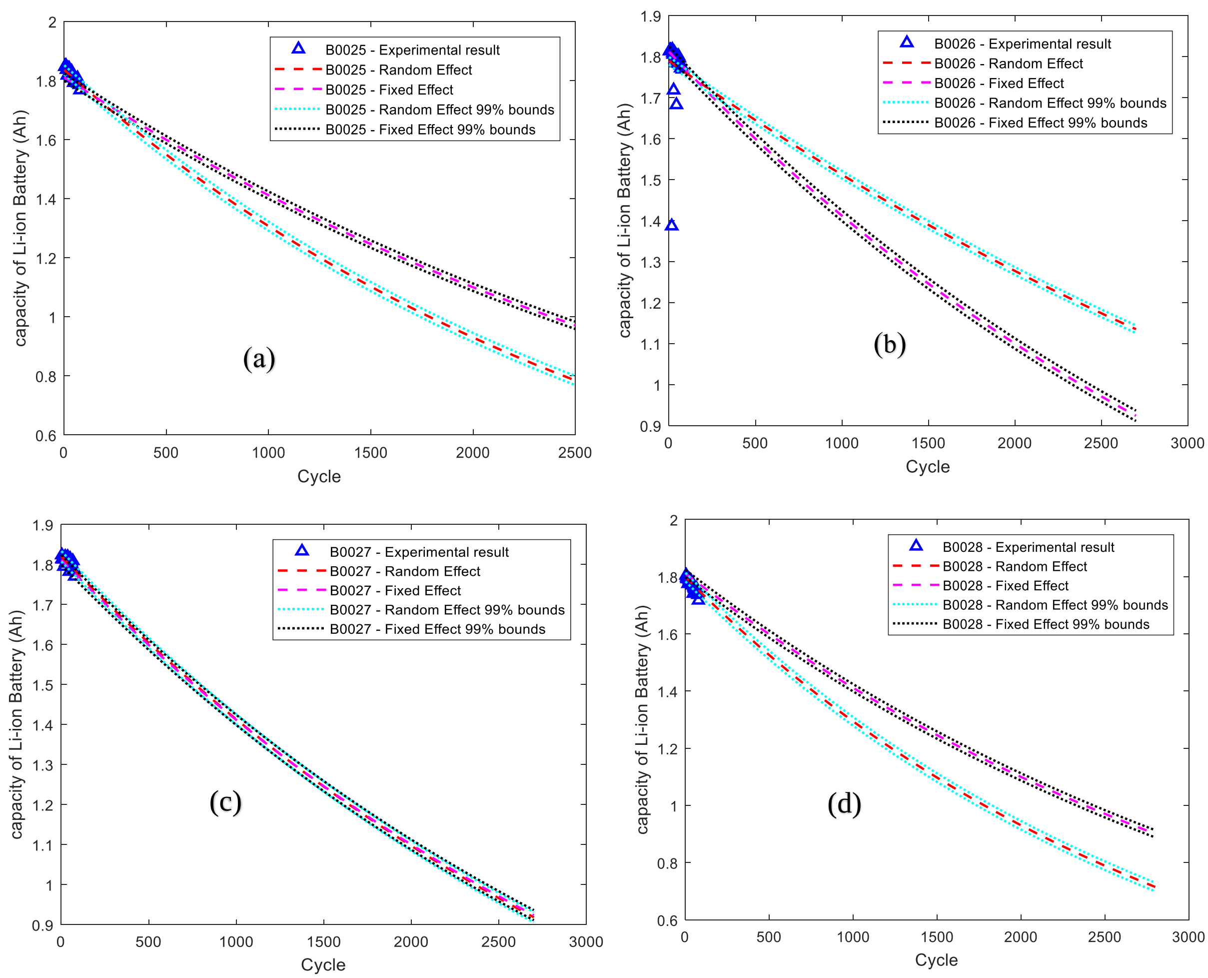

Fig 3: Estimated Fixed and Random effects models predicted future charge decay pattern of batteries for batteries: (a) - B0025, (b) - B0026, (c) - B0027 and (d) - B0028

Table 3: Variation of the estimated EOL cycle of the batteries with Fixed and Random effects models

EOL Criteria (%)	70%	60%	50%
	B0025		
Random effect	797	1250	1786
Fixed effect	1033	1651	2381
Difference	-236	-401	-595
% Variation	-30%	-32%	-33%
	B0026		
Random effect	1455	2368	2700
Fixed effect	1033	1651	2381
Difference	422	717	319
% Variation	29%	30%	12%
	B0027		
Random effect	1037	1645	2364
Fixed effect	1033	1651	2381
Difference	4	-6	-17
% Variation	0%	0%	1%
	B0028		
Random effect	761	1229	1782
Fixed effect	1033	1651	2381
Difference	-272	-422	-599
% Variation	-38%	-34%	-34%

5. CONCLUSIONS

This study used degradation modelling of battery charge capacity decay at different cycles to determine the reliability of lithium-ion batteries, by considering the uncertainties associated with the charge decay, using the nonlinear mixed effects model. This comparative study of the fixed and random effects models, for remaining useful life estimation of the lithium-ion battery was intended to show the influence of uncertainties, which can originate from the manufacturing process, measurements and operational environmental conditions of the battery.

After using maximum likelihood estimate (MLE) to determine the likelihood value of the nonlinear function of the fixed and random effects, stochastic approximation, expectation and maximization algorithm was used for determining the parametric values of the fixed and random effects. Lithium-ion battery datasets of four batteries – B0025, B0026, B0027 and B0028 obtained from NASA AMES research Centre were used to test the models and it was observed that the random effect models had smaller Root Mean Square Errors (RMSEs) than the fixed effect models; an indication that uncertainties inherent in the battery charge capacity decay, influenced the state of charge of the batteries, vis-à-vis the reliability estimated at the End-of-Life (EOL) thresholds that were taken to be 70%, 60% and 50% of the retained original battery charge capacity of 2.0Ah. The uncertainties in the battery charge capacity decay influenced the reliability of the batteries in diverse ways because of the level of the accumulated uncertainties inherent in the battery charge with some resulting in overestimation and others ending up with a conservative estimate of the remaining useful life.

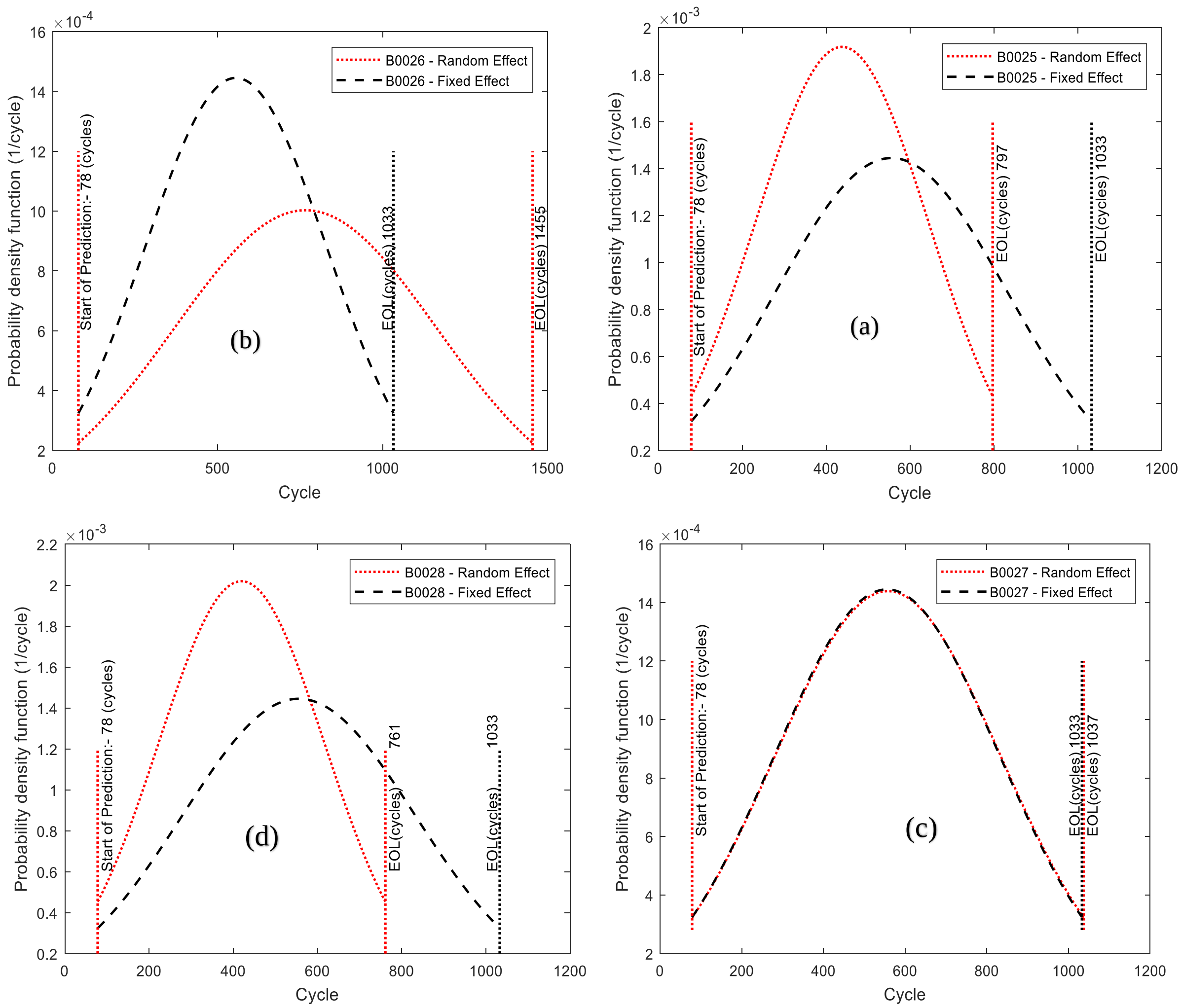

Fig 4: Probability Density Function of battery charging cycle at 70% End-of-Life for: (a) - B0025, (b) - B0026, (c) - B0027 and (d) - B0028

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BIOGRAPHIES

C. I. OSSAI obtained his PhD in Mechanical Engineering from Curtin University, Australia. Dr. Ossai is a member of Engineers Australia and has research interest in the application of predictive analytics and machine learning to the estimation of assets integrity, modelling of nanodevices physics of failure, uncertainty quantification, prognostics & system health management.