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Electrical Method for Battery Chemical Composition Determination

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ABSTRACT Storage of electrical energy is one of the most important technical problems in terms of today's technology. The increasing number of high-capacity high-power applications, especially electric vehicles and grid scale energy storage, points to the fact that we will be faced with a large number of batteries that will need to be recycled and separated in the near future. Additionally multi-chemistry battery management systems that enables the collective use of superior features of different batteries with different chemical composition. Here, battery chemical composition determination emerges as a technical problem. In this study, an alternative method to the currently used methods for categorizing batteries according to their chemistry is discussed. As the foundation, batteries with four different chemical composition including Lithium Nickel Cobalt Aluminium Oxide, Lithium Iron Phosphate, Nickel Metal Hydride, and Lithium Titanate Oxide aged with a battery testing hardware. Fifth, is Lithium Sulphur battery which is simulated. Brand new and aged batteries are used in experimental setup that is consist of a programmable electronic DC load and a software developed to run the algorithm on it. According to the algorithm, batteries are connected to two different loads one by one and voltage-current data are stored. Collected data are pre-processed by framing them and framed data are processed with a separation function. Eventually, the determination problem is converted to a classification problem. In order to solve this, artificial neural network and classification tree algorithms are applied. Because the artificial neural network algorithm is applied in previous studies and the high computational cost of it is presented; classification tree algorithm is concluded to be more applicable especially on low-power microcontroller applications. Consequently, 100% accuracy for battery chemical composition determination is achieved and results are presented comparatively.

INDEX TERMS Batteries, battery management system, classification algorithms, electric vehicle, machine learning, optimisation, recycling.

I. INTRODUCTION

Electric Vehicle (EV) technologies and the innovations it brings are one of the leading issues that touch our lives today and a sustainable EV technology seems to become critical tool in next decades to overcome the environmental problems like global heating. Journey of developing EV technology can be taken as far as to discovery of magnetism. On this journey, the biggest obstacle to the realization of this technology was the energy-storage problem. It could only be realized by the achievement of proper and sufficient electrical energy storage capability. Despite the fact that studies on this subject began

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in the mid-18th century, the voltaic battery developed by Alessandro Volta at the end of the century, was the first significant development [1]. As a continuation of Volta's work French physicist Gaston Planté developed the first rechargeable battery in mid-19th century [2]–[4]. These achievements steps are followed by many scientist accomplish development of lithium batteries which lets the start of EV era [5]–[19].

EVs are consist of modules such as electric motor, motor controller, inverter etc. that performs different tasks. Within these modules, battery is the most crucial one because it provides the energy needed. Batteries are in need of a proper management in order to run safe and efficient. This system is called battery management system (BMS). There is no consensus on the definition however in the literature, BMS

is defined as a system that measures potential problems with an electric vehicle's battery, as well as mandatory equipment to monitor, control and balance the battery pack [20], [21]. From today's technical perspective, BMS can be defined as an integrated hardware and software body that enables the battery to operate within a specified safe/optimal operating envelope (in terms of parameters such as current, voltage, temperature, state of charge (SoC), state of health (SoH), state of energy (SoE), remaining useful life (RUL) etc.) and at the same time logging, processing and sending data via wireless connection to a cloud server, and/or sharing it with other connected devices and vehicles.

In order to do these complicated tasks, the BMS functions are also becoming complex and vary. At this point, one would expect to find a universal BMS but on contrary, in most cases the BMS software and hardware depend on the battery type/chemistry for which it is designed. However, BMS hardware, software, and functions differ depending on the application.

In this study, a battery chemistry determination function is developed as the successor of the authors' related past studies [22]–[28]. Battery chemical composition determination (BCCD) is practically difficult task for real-time applications on the other hand; it is a necessary function for specific applications such as recycling/second-life application of EV batteries and multi-chemistry BMSs.

When the battery packs on an electric vehicle do not meet the required performance criteria, and when they reach the end of their useful life, using them in areas other than automotive is defined as the second-life. Recycling EV batteries is a critical issue from economic and environment point of view because of environmental harm and the limited reserves of elements used in batteries. In the recycling process and in the sale of recycled products, it is necessary to classify and sort batteries according to their chemical composition [29]. The difficulty in determining the battery chemistry is mostly due to the lack of proper labelling, standard design, or clear marking on the package [30]. Currently, there are methods such as X-ray radiographic scanning, computer vision, artificial intelligence, and blockchain to sort and classify batteries according to their type [30]–[33].

Multi-chemistry BMSs enable to use superior properties of different battery chemistries in conjunction to make battery systems more efficient [34]–[38]. In such kind of application, BCCD function is found to be necessary. By this way, the BMS can automatically figure out the chemical composition of the connected battery so it can manage with the proper parameters.

In this study, proposed BCCD can also provide application flexibility to users and manufacturers who want to switch to a different type of battery chemistry. For all battery-powered devices or vehicles, it offers the opportunity to design without being dependent on battery chemistry. The crucial feature of the proposed method is that it allows applications to support battery chemistries that are likely to be released in the future. The biggest challenge in the process of BCCD is doing it

on-board. Because BMSs measure raw voltage/current data and these data overlap due to different chemical composition and state of the battery (such as SoC, SoH, SoE, RUL). To eliminate this problem, a separation function based on statistical significance is developed. With this developed function, it is concluded that the overlapping values can be successfully separated, resulting a categorization algorithm can determine the chemical composition of battery with 100% accuracy.

II. METHODOLOGY

Since the scope of the study includes both battery management systems and second life and/or recycling processes of batteries, the data of healthy batteries and batteries aged at different rates are discussed together.

The first four batteries used are Lithium Nickel Cobalt Aluminium Oxide (NCA), Lithium Iron Phosphate (LFP), Nickel Metal Hydride (NiMh), and Lithium Titanate Oxide (LTO) batteries, which are currently commercialized and widely used. Owing to the increasing number of patents and commercialization potential in recent years, the fifth battery chemistry has been determined to be lithium sulphide (LiS) [39]. The data of NCA, LFP, NiMh and LTO batteries are obtained from the experimental setup that is prepared for this study in a laboratory setting; the data of LiS batteries are obtained from a simulation prepared in MATLAB/SIMULINK.

All the batteries are aged with 1 C until their capacity is fade to 80% which is widely accepted as the usable life limit of lithium batteries especially for high power applications. Aging process took four months in total and Neware BTS4000 battery testing system. This system is rated for 5V and 12A.

A. DATA ACQUISITION

1) EXPERIMENTAL SETUP

Prodigit 3350F programmable electronic DC load is used for collecting current-voltage data for two different loads. This electronic load is rated for 1200W, 60V and 120A. DC load is controlled by a software, which is developed under LabVIEW. Five hundred thousand data samples are collected from battery cells with five different chemistries. The process took around twenty-three weeks in total. Minimum discharge and maximum charge voltages for the batteries are presented in Table 1.

Two different loads, whose values are determined by the software interface, are switched from one to the other during a specified period. Meanwhile, the current and voltage values are recorded at a frequency of 1 Hz. To stabilize the measured values at each load, the load is maintained for 10 seconds and then switched to the other load. The reason for such a design is to obtain current-voltage pairs for different loads under stress without allowing the battery to relax.

2) SIMULATION

Lithium-Sulphur ECN Model [40] is run using the same algorithm as that used for the experimental setup. Here in

TABLE 1. Data statistics.

Group		Min	1 st Q	Median	3 rd Q	Max
LFP	Voltage	2,7985	3,1546	3,2026	3,2309	3,6069
	Current	0,2930	0,3320	0,4610	0,6460	0,6980
LiS	Voltage	2,0364	2,0963	2,1020	2,1080	2,4245
	Current	0,2061	0,2236	0,2994	0,3987	0,4822
LTO	Voltage	1,3946	2,3194	2,3875	2,4737	2,8955
	Current	0,1550	0,2510	0,3210	0,4840	0,5720
NCA	Voltage	2,4966	3,4121	3,62325	3,8518	4,2417
	Current	0,2640	0,3770	0,5160	0,7270	0,8490
NiMh	Voltage	0,8683	1,2447	1,2612	1,2845	1,3913
	Current	0,1160	0,1370	0,1950	0,2620	0,2870

order to simulate the aging effect internal resistor values in the model are increased in a randomly order. Additionally, simulation temperature is set to 25 °C and initial SoC is taken as 100%.

B. PRE-PROCESSING

In its raw form, it is not always possible to determine the battery chemistry from the current and voltage data alone, owing to overlapping. Therefore, the data must be pre-processed. For this purpose, data are put into frames and a separation function is developed to separate the overlapping values.

1) FRAMING

The data obtained are converted into data frames; the current values of the first and second load (10 data points for 10 seconds, 1 Hz sampling frequency), and then the voltage values are combined side by side to form data frames, as shown in Figure 1. By this way, electrical information related to battery chemistry is combined to obtain results that are more accurate.

2) SEPARATION FUNCTION

The main goal of the separation function is to transform overlapping data frames into disjoint sets so that the classification problem can be solved with full accuracy by using a machine-learning algorithm. To achieve this, first it is

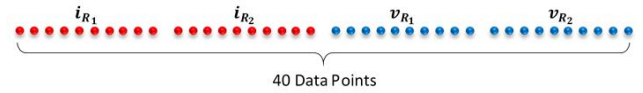


FIGURE 2. Data frame.

necessary to determine which parts of the data are statistically more significant. The data are summarized with minimum, 1st quartile, median, 3rd quartile and maximum statistics in Table 1. The conformity of the numerical variables to the normal distribution on the basis of the group is examined using the Shapiro-Wilk test. The Kruskal-Wallis H test is used to determine whether there is a statistically significant difference between the groups. After the Kruskal-Wallis H test, pairwise group comparisons are made using the Conover test [41].

According to the results obtained, it is concluded that the voltage values are statistically more significant. In this context, a function has been derived that will enable the separation of current values by amplifying the voltage in the data frames as in (1). By other means, the separation function separates the current values by using statistically more significant voltage values.

$$f : x_{i,j} \rightarrow y_{i,j}, \quad \forall x_{i,j} \in \mathbb{R} \text{ and } \forall y_{i,j} \in \mathbb{R}$$

$$f_s(x) = \begin{cases} n \leq \frac{n_{max}}{2}, & f(x) = \frac{(x + \varepsilon) \times \eta}{\beta} \\ n > \frac{n_{max}}{2}, & f(x) = \frac{(x + \varepsilon) \times \eta \times \rho}{\beta} \end{cases} \quad (1)$$

Here,

- n*: Row number
- n_{max}*: Maximum number data in a frame
- ε*: Offset
- η*: Upscaling factor
- ρ*: Significance factor
- β*: Downscaling factor

$$y_{j,k} = f_s(x_{j,k}) \quad (2)$$

$$Y = f_s(X) \quad (3)$$

where *j* is the sample count in a frame, *k* is the total frame count, *X* is representing the collected data, and *Y* is representing the processed data. These data form a matrix consisting of data frames in columns. Because the number of samples in a frame is 40, frames of 40 units consisting of current and voltage data can be written as columns, as in (4). This formation is used in the application of the separation function.

$$X = \begin{bmatrix} i_{1,1} & i_{1,2} & \dots & i_{1,k-1} & i_{1,k} \\ i_{2,1} & i_{2,2} & & i_{2,k-1} & i_{2,k} \\ \vdots & & & & \vdots \\ v_{39,1} & v_{39,2} & & v_{39,k-1} & v_{39,k} \\ & & \dots & & \\ v_{40,1} & v_{40,2} & & v_{40,k-1} & v_{40,k} \end{bmatrix} \rightarrow Y$$

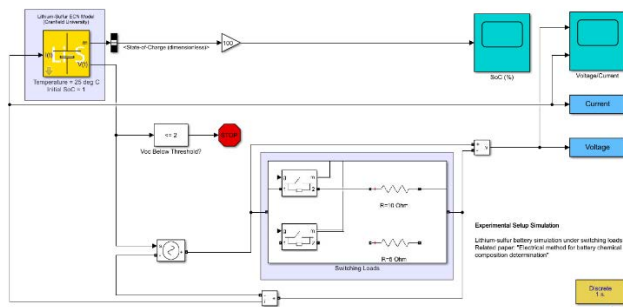


FIGURE 1. LiS battery simulation.

$$= \begin{bmatrix} y_{1,1} & \dots & y_{1,k} \\ \vdots & \dots & \vdots \\ y_{40,1} & \dots & y_{40,k} \end{bmatrix} \quad (4)$$

3) PARAMETER OPTIMISATION

After obtaining the separation function in matrix form, the primary issue for its application is to determine the parameters used in the function. Because the voltage parts of the overlapping data are found to be statistically significant, ρ has to take a value other than one. For appropriate and realistic parameter selection, an optimisation algorithm is used.

In this case, the ‘‘Firefly Optimisation’’ metaheuristic optimisation algorithm is chosen, which is first proposed by Xin-She Yang [42], [43]. As it is widely stated in the literature, metaheuristic algorithms like particle swarm optimisation (PSO) or its variants can be used in such kind of parameter determination problem because of its applicability [44]. In this study, the cost function presented in (6) is used in the firefly optimisation algorithm to determine the parameters of the separation function. Here, the data of five types, namely batteries with five different chemistries, are used separately. For this, it is designated as Type 1, Type 2, Type 3, Type 4, and Type 5 LiS.

$$\mu = \frac{1}{k} \sum_{i=1}^k Y_{n,k} \quad (5)$$

$$f_{Cost} = \left[10^6 - \sum_{i=1}^{40} \left(\sqrt{(\mu_{Type1}(i) - \mu_{Type2}(i))^2} - \sqrt{(\mu_{Type4}(i) - \mu_{Type3}(i))^2} - \sqrt{(\mu_{Type2}(i) - \mu_{Type5}(i))^2} \right)^2 \right] \quad (6)$$

C. CLASSIFICATION

Owing to the data processed with the separation function, the problem of determining the battery chemistry has been transformed into a classification problem. Many algorithms have been proposed to solve this problem. In this study, artificial neural networks (ANNs) and decision tree algorithms are used among machine learning algorithms. Because ANN is widely used for on-board (on-chip) applications which is run on a microcontroller. Considering the computational cost of ANN for the solution of BCCD problem is presented in detail in previous studies [22]. According to the results, ANN has great computational cost for on-chip applications. The reason why decision tree algorithm is chosen in this study is its low computational cost and on-chip applicability. Obtained results are presented comparatively.

For the design of the ANN, the number of hidden neurons and output neurons are fixed at 10 and 5, respectively. Input neuron numbers are 40 as data count in a frame. In addition, the classification tree algorithm, among machine learning algorithms, is used for the same purpose. The term classification tree is first used by Breiman *et al.* in 1984 [45].

Classification trees predict answers to questions posed to the data, also known as decision trees. Therefore, decisions in the tree are tracked from the beginning (root) node to a leaf node containing the response to predict a response. Classification trees give boolean responses such as ‘true’ or ‘false’. According to these responses, they either branch to other leaf nodes or arrive at a conclusion that includes the data in a category.

Figure 3 presents the workflow of the study. The pre-processing and classification sections are fully conducted in MATLAB 2021b. The simulation part of the data acquisition section is conducted in SIMULINK 2021b. The experimental study part of the data acquisition section is conducted with a programmable electronic DC load controlled via LabVIEW software.

III. RESULTS AND DISCUSSIONS

The data collected from the experimental and simulation studies are presented in Figure 4 in the form of current-voltage pairs. Here, more than 500000 measurements are seen that the data of batteries with different chemistries and aged at different rates from 25 to 2500 cycles overlap with each other in certain areas. In particular, the data of NCA and LFP batteries and LTO and LiS batteries overlap. Because the data are collected for two different loads, a pattern separated by two sharp linear lines can be seen.

When the data is used in current-voltage pairs, the artificial neural network design consists of 2 input neurons, 10 hidden neurons and 5 output neurons configuration. With this configuration, the results presented in Figure 5a are obtained. Here, the distribution of the datasets is as follows; training 70%, verification 15% and testing 15%. Similar results, presented in Figure 5b, are obtained using the decision tree algorithm. When Figure 5a and 5b are examined together, a result confirming the overlap in Figure 4 is observed. Here, it is seen that, for NCA and LFP batteries, with artificial neural network algorithm 6406 data and with the decision tree algorithm, 6851 data points are incorrectly predicted. For LTO and NCA, 619 data with the artificial neural network algorithm and 474 data with the decision tree algorithm are incorrectly predicted. Likewise, for LTO and LiS batteries, 1950 data with the artificial neural network algorithm and 5769 data with the decision tree algorithm are predicted incorrectly. At this point, it is observed that the artificial neural network algorithm yields results that are more accurate for NCA-LFP and LTO-LiS pairs. While the artificial neural network algorithm made 9064 wrong predictions, the decision tree algorithm made 13212 wrong predictions in total. With ANN configuration, no matter how much fine tuning the network parameters are made, the result cannot be improved; due to overlapping data.

By bringing small pieces of information, reflecting the electrical and chemical properties of the battery together, precise results can be obtained. To achieve this, the data are framed as presented in Figure 1, as the first step of the pre-processing.

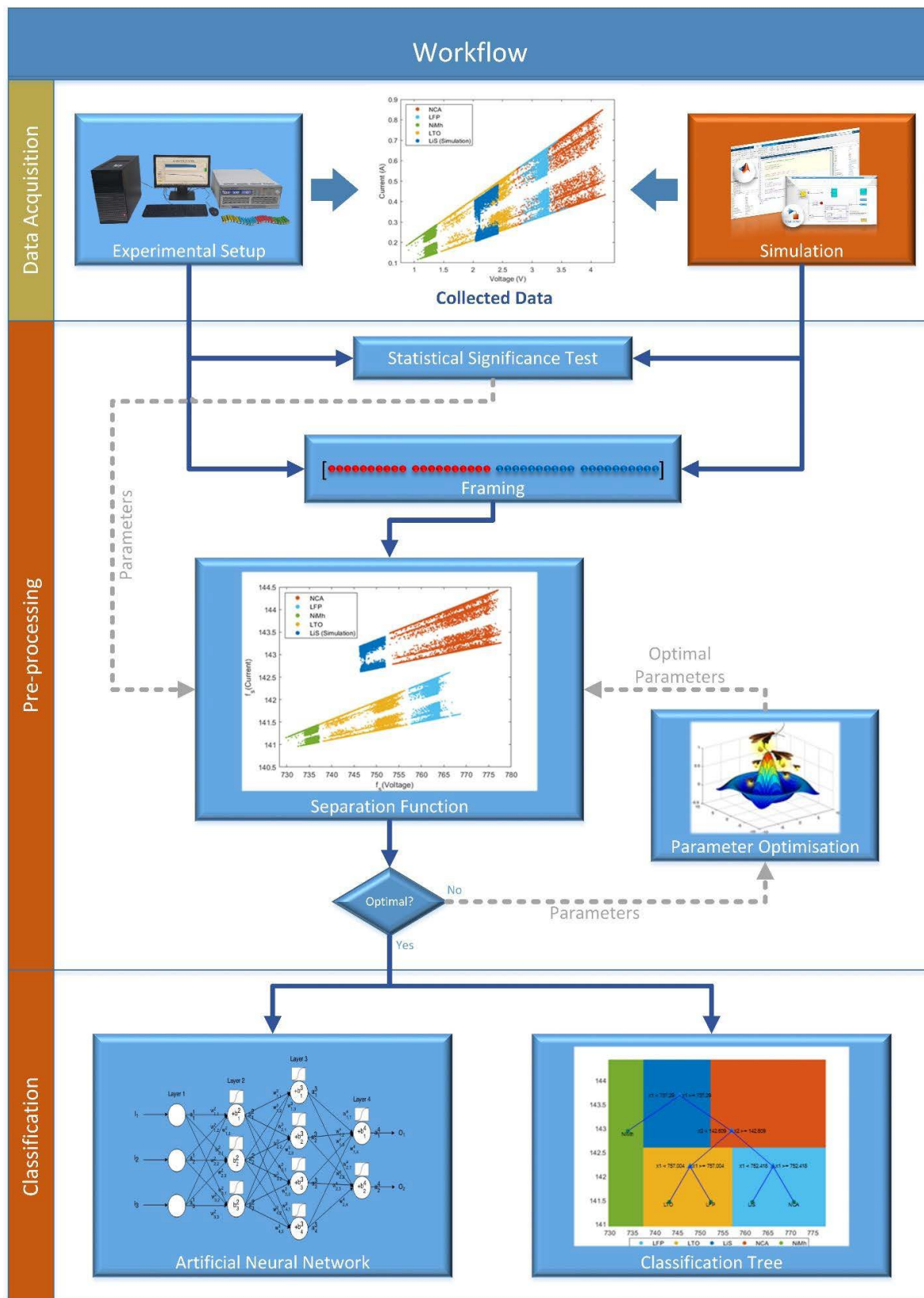


FIGURE 3. Workflow of the study.

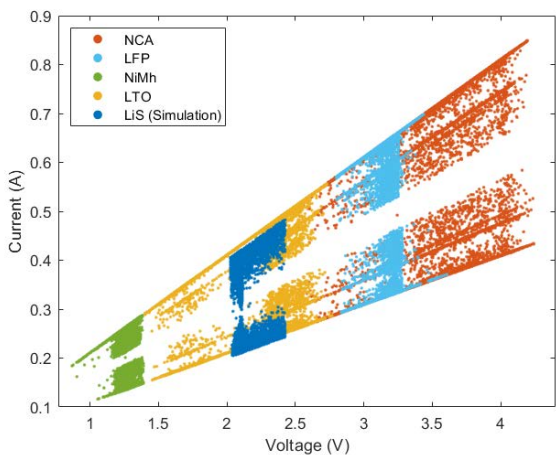


FIGURE 4. Raw data.

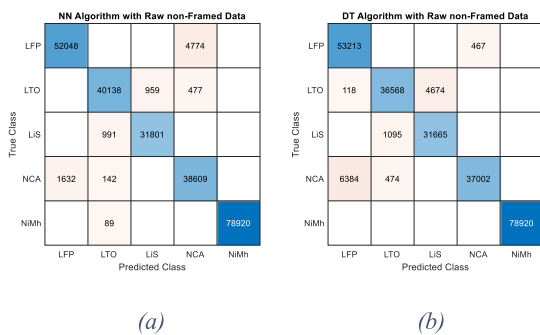


FIGURE 5. Neural network and decision tree confusion matrix for binary couples.

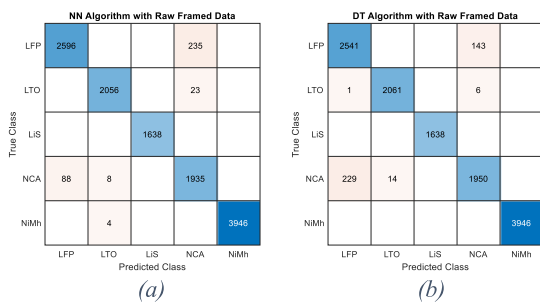


FIGURE 6. Neural network and decision tree confusion matrix for data frames.

When framed data are used, the results obtained by the artificial neural network algorithm are presented in Figure 6a, here, the distribution of the datasets is as follows; training 70%, verification 15% and testing 15%. Obtained results by the decision tree algorithm are presented in Figure 6b. The artificial neural network algorithm made 100 and the decision tree algorithm made 393 incorrect predictions in total.

In both cases where the data are used in pairs and frames, the results obtained from the artificial neural network and decision tree algorithms show that battery chemistries cannot be determined with 100% accuracy. However, when the data are evaluated as frames, it is observed that the algorithms

made relatively more accurate predictions. In order to improve the prediction accuracy, pre-processing the framed data with a separation function is proposed.

Firefly optimisation algorithm is used to determine the optimal values of the offset, significance, upscaling, and downscaling factor parameters in the separation function (1). The optimisation algorithm is applied with the cost function given in (6), which is chosen to maximize the difference between the mean values of the data sets. The parameters calculated according to the optimisation results are $\eta = 38.03$, $\rho = 5.051$, $\beta = 1.3523$, and $\varepsilon = 50.502$. The graph of the data processed using the separation function is presented in Figure 7. After pre-processing, it is easy to predict the battery type with classification algorithm because of disjoint values. Figure 7 shows that the overlapping data of LTO-LiS and NCA-LFP batteries are clearly separated. This demonstrates the effectiveness of the separation function.

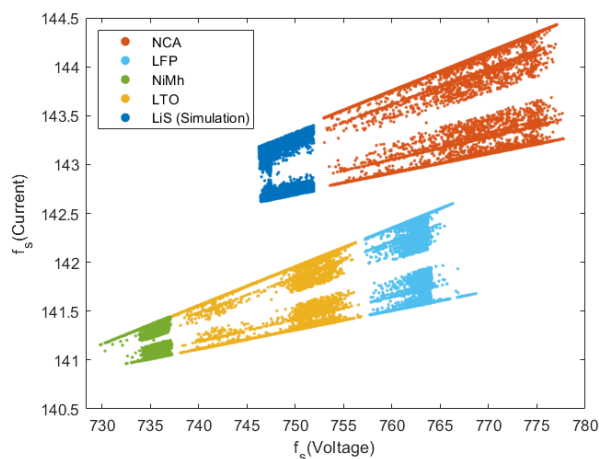


FIGURE 7. The effect of separation function.

To determine the battery chemistry with 100% accuracy, which is the aim of the study, it can only be achieved by pre-processing the data by both framing and separation function. With the pre-processed data, both the artificial neural network and decision tree algorithm are applied separately. With both, the battery chemistry can be determined with 100% accuracy. The results are shown in Figure 8.

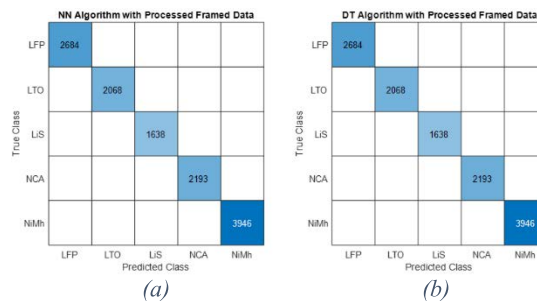


FIGURE 8. ANN algorithm and decision tree algorithm results after pre-processing data frames with the separation function.

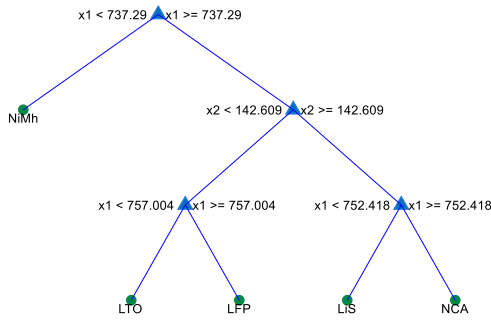


FIGURE 9. Decision tree algorithm.

Graphic description of the decision tree algorithm, which can detect battery chemistry with 100% accuracy and less computational cost, is presented in Figure 9. The pseudo code of the algorithm is as follows.

Here $x1: f_s$ (Voltage) } representing;
 $x2: f_s$ (Current)

node 1 if $x1 < 737.29$ then NiMh else if $x1 >= 737.29$ then node 3
 node 3 if $x2 < 142.609$ then node 4 else if $x2 >= 142.609$ then node 5
 node 4 if $x1 < 757.004$ then LTO else if $x1 >= 757.004$ then LFP
 node 5 if $x1 < 752.418$ then LiS else if $x1 >= 752.418$ then NCA

As it can be seen from the pseudo code of the decision tree algorithm, this algorithm can be easily implemented on any hardware, even with integrated circuits with low processing power. The manner in which the decision tree algorithm categorizes the data processed with the separation function is presented in Figure 10. Figure 10a shows the effect of the separation function. Owing to pre-processing, it is seen that current-voltage pairs with the same voltage value have different f_s (Current) values, although they have the same f_s (Voltage). Thus, classification areas of the batteries' chemical composition can be clearly predicted, as shown in Figure 10b.

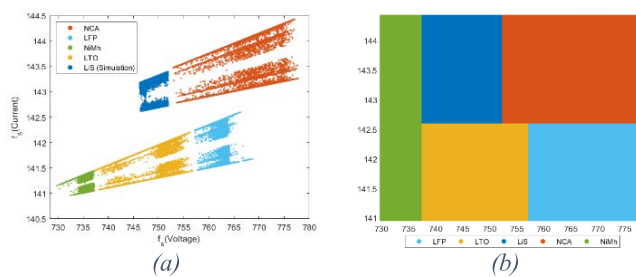


FIGURE 10. Pre-processed data and how it is classified by decision tree algorithm.

BCCD is especially designed for a new generation function for BMSs or for different commercial applications; one of the important factors is computational cost. For this reason, the

method is designed to be run on electronic hardware with a simple microcontroller. It is concluded that categorizing the data processed with the separation function using the decision tree algorithm is much more applicable because of its low computational cost despite the fact that it has 100% success on BCCD. The proposed method is applied to a multi-chemistry BMS which is patent pending [27]. Additionally, the method also applied to a system that determines the chemical composition of battery and calculates state of health. This system is also patent pending [28].

IV. CONCLUSION

Meeting the increasing energy need for electrical systems in parallel with the developing technology, storing electrical energy in sufficient density is a problem that has been going on for decades and waiting to be solved. Great steps toward solving this problem are taken in the 20th century. The development of lithium-based batteries has made high-power applications such as electric vehicles and grid-scale batteries. Due to increasing environmental awareness, legislative changes, incentives, and critical decisions made in line with the policies of countries to reduce carbon emissions, the trend toward electric vehicles has increased significantly. In addition, renewable energy systems, green energy, and large-scale storage systems used in grids have also increased the demand for batteries. However, this increase has revealed the problem of using end-of-life batteries in other applications or processing them in recycling centres. The separation of batteries according to their chemistry in these processes is a technical problem. To solve this problem, costly, error-prone, difficult to implement, and time-consuming methods such as computer vision, block chain, artificial intelligence, and X-rays are used. In addition, studies are conducted in the field of multi-chemistry battery management systems, which enable the joint use of the superior properties of different battery chemistries in order to make battery systems more efficient. Moreover, battery management systems must have next-generation features to support future battery chemistries. Determining the battery chemistry is a technical challenge.

A robust determination method is developed in this regard, presented in this study, applied to a new generation battery management system developed under a Project supported by Scientific and Technological Research Council of Turkey and Inonu University research fund. Additionally, patent applications have also been made [24], [25]. In this method, the battery chemistry is determined by measuring the current and voltage parameters of the battery for only 20 s. It differs from existing methods in terms of using only basic electrical parameters and ease of application. The main factors that make this possible are framing the collected data and pre-processing it with the proposed separation function. Because of pre-processing, the current values corresponding to the same voltage value of batteries with different chemistries are separated from each other. In this way, the problem of battery chemistry determination has become a categorization problem. Artificial neural networks and decision tree

algorithms are applied to solve this problem. In terms of computational cost, the decision tree algorithm is considerably more advantageous than the artificial neural network algorithm. It is concluded that it would be more appropriate to use this method with the decision tree algorithm in terms of both computational cost and ease of implementation so that it can be used even in complex or high cost applications with low computational power. As a result, thanks to this novel approach, 100% success is achieved by using basic electrical parameters for five most preferred commercial battery types.

It is foreseen that adding the proposed method-based determination function to battery management systems will provide flexibility so that both end user and BMS manufacturers can design without being dependent on battery chemistry. Considering the rapidly increasing use of electric vehicles worldwide, it will be possible to choose traction batteries that have completed their useful life, regardless of battery chemistry. In general, it allows the use of all existing battery types in the design and production of all battery-powered systems, and the use of new battery chemistries that will be released to the market by determining them. In addition, a low-cost, easy-to-apply, and fast alternative solution works with basic electrical parameters so that batteries can be separated according to their chemistry during the recycling process.

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Related patent pending 2021/005464.

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