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Non-Zero Intercept Frequency: An Accurate Method to Determine the Integral Temperature of Li-ion Batteries

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Abstract—A new impedance-based approach is introduced in which the integral battery temperature is related to other frequencies than the recently developed zero-intercept frequency (ZIF). The advantage of the proposed non-zero-intercept-frequency (NZIF) method is that measurement interferences, resulting from the current flowing through the battery (pack), can be avoided at these frequencies. This gives higher signal-to-noise ratios (SNR) and, consequently, more accurate temperature measurements. A theoretical analysis, using an equivalent circuit model of a Li-ion battery, shows that NZIFs are temperature dependent in a way similar to the ZIF and can therefore also be used as a battery temperature indicator. To validate the proposed method impedance measurements have been performed with individual LiFePO$_4$ batteries and with large LiFePO$_4$ battery packs tested in a full electric vehicle under driving conditions. The measurement results show that the NZIF is clearly dependent on the integral battery temperature and reveals a similar behavior to that of the ZIF method. This makes it possible to optimally adjust the NZIF method to frequencies with the highest SNR.

Index Terms—Electrochemical impedance spectroscopy, Integral battery temperature, Lithium batteries, Non-zero intercept frequency, Sensorless temperature measurement.

I. INTRODUCTION

The demand for battery-powered devices has increased tremendously in the last decade. Li-ion batteries are nowadays most widely applied to power these devices due to its high gravimetric and volumetric energy density. The increased functionality of many electronic devices and (Hybrid) Electric Vehicles ((H)EVs) put, however, some serious demands on the operation of modern Li-ion batteries. For example, users desire short charging times which invokes high charging currents. Consequently, this puts serious requirements on the functioning of these battery systems, especially with respect to heat generation.

It is well known that the temperature has a strong influence on the battery life-time and safety. High temperatures may lead to a decrease in battery performance, accelerate aging and introduce unsafe situations, like thermal runaway. Under high temperature conditions side reactions may occur at the electrode/electrolyte interfaces such as, for example, metal dissolution from the cathode [1]. On the other hand, low temperatures reduce the energy and power capability of Li-ion batteries [2]. Lithium plating may occur at the anode under these conditions, which leads to permanent loss of cyclable lithium [1] and the risk of short-circuiting [3]. For all these reasons it is essential to maintain the battery temperature within a well-defined range by making use of an advanced thermal management system.

It is, however, rather challenging to determine the battery (pack) temperature as an input parameter for thermal management systems as the temperature varies throughout the battery [4]–[13] and the thermal mass of batteries makes it difficult to detect internal temperature changes externally [8], [14]. In addition, measuring the temperature in large battery packs with many serial and/or parallel-connected cells applied in (H)EV becomes even more complex. Therefore, many researchers are investigating reliable and accurate methods to directly measure the battery (pack) temperature in the application.

Temperature determination can, for instance, be performed by implementing a thermal model to identify the internal battery temperature [6], [9], [11], [12], [15]–[22]. Such a model may be difficult to implement because of the multitude of required model parameters, some of which are difficult to quantify. Alternatively, surface-mounted temperature sensors can be used for each individual cell in battery packs. This approach suffers, however, from a significant heat transfer delay, and can be cost-prohibitive and challenging to implement in terms of positioning, thermal contacting and wiring. Another method is to make use of (multiple) intrusive temperature sensors [8], [23]–[25], having the advantage of monitoring non-uniform temperature distributions and hazardous hot spots. However, intrusive sensors must comply with stringent requirements, i.e. that (i) the transport of Li-ions in the electrolyte may not be affected by its presence, (ii) the sensors should be chemically inert and may not react with or dissolve into the electrolyte and
(iii) the sensors must be fully compatible with the assembly process [24]. Temperature measurements which do not make use of intrusive- or surface-mounted devices are more convenient. Methods based on electrical measurements, such as electrochemical impedance spectroscopy (EIS) are therefore much more attractive to apply, if accurate and reliable.

A number of EIS-related temperature determination methods have been presented, for example, relating the internal temperature to the time constant of a particular RC-element in an equivalent circuit, representing the electrochemical battery impedance [26]. The values of the corresponding RC-element can then frequently be computed through impedance measurements. Srinivasan et al. [27], [28] as well as Schwarz et al. [29] related the internal battery temperature to the phase shift between an applied sinusoidal current and the resulting alternating voltage. The internal battery temperature has also been correlated to the real part [14] and imaginary part [30] of the impedance at a predefined frequency. The real-part method has subsequently been combined with surface temperature measurements and a thermal impedance model to estimate the temperature distribution inside cylindrical cells [31], [32]. Zhu et al. relate the internal battery temperature to either the magnitude or phase of the impedance in a certain frequency range [33]. Jointly selecting impedance parameters was done by Beelen et al. [34] in order to obtain a more accurate temperature estimation.

A number of above mentioned methods have been evaluated by Koch et al. [35], showing that the use of the imaginary part and phase are preferred above the real part and absolute value of the impedance. Recently, the present authors reported a new method in which the introduced zero-intercept frequency (ZIF) was used to determine the internal battery temperature [36], [37]. The advantage of this method is that it can make use of a relatively large frequency variation over practical temperature ranges. The temperature can therefore be determined with relatively high accuracy. Advantageously, it should be emphasized that an EIS-based temperature measurement method indicates the integral battery temperature rather than the maximum or minimum temperature. If there is a temperature gradient across the cell, a temperature close to the average will be indicated [32].

All EIS methods have been designed to determine the temperature under electronically non-disturbing conditions. In applications such as (H)EV the electric motor and other components draw, however, large currents from the battery (pack) over a wide range of frequencies, which may overlap with the frequencies of interest. In order to avoid dominating interfering currents in an electronically disturbing environment our previous work [36], [37] has been developed further by presenting a temperature measurement method which is based on a non-zero intercept frequency (NZIF). The NZIF is defined as the frequency at which the imaginary part of the battery impedance is equal to a non-zero constant, leading to higher or lower frequencies than the ZIF, i.e. in either the inductive or capacitive part of the impedance spectrum.

Moving from ZIF towards NZIF determination makes it possible to perform EIS measurements in a frequency range where the external interferences are relatively low. This offers great potentials to increase the signal-to-noise ratio (SNR) and, consequently, to determine the temperature much more accurately. In this contribution the EIS measurement interferences and the NZIF method are addressed in detail and supported by experimental battery data. To show the strength of the NZIF-method, experiments are performed on both individual batteries under laboratory conditions and large battery packs, powering an EV under real driving conditions.

II. (NON)-ZERO INTERCEPT FREQUENCY MODELING

Equivalent circuit models comprising inductive, resistive and capacitive elements can be used to simulate the battery impedance [38]–[40]. A simple, but very convenient equivalent circuit model for a theoretical analysis is shown in Fig. 1 [37]. In this circuit $L$ represents the inductance, $R_s$ the series resistance and $C_d$ the diffusion capacitance of the battery. The capacitance $C_{kin}$ in parallel to the resistance $R_{kin}$ is used to simulate the double layer capacitances, charge transfer kinetics and surface layers of the electrodes. Given this circuit, the total battery impedance ($Z_h$) can be written as

$$Z_h = j\omega L + R_s + \frac{R_{kin}}{1 + j\omega R_{kin} C_{kin}} + \frac{1}{j\omega C_d}.$$ \hspace{1cm} (1)

The imaginary part of (1) is derived as

$$Z_{im} = \omega L - \frac{(C_{kin} + C_d) R_{kin} R_{kin}^2 \omega^2 + 1}{C_d \omega (C_{kin} R_{kin}^2 \omega^2 + 1)}.$$ \hspace{1cm} (2)

Equation (2) can be rearranged to an algebraic equation of degree 4 in terms of $\omega$. This results in

$$C_d \omega^2 (C_{kin} R_{kin}^2 \omega^2 + 1) - (C_{kin} + C_d) C_{kin} R_{kin}^2 \omega^2 - C_d \omega (C_{kin} R_{kin}^2 \omega^2 + 1) Z_{im} = 0.$$ \hspace{1cm} (3)

**Fig. 1.** Basic equivalent circuit model to simulate battery impedance.

If $Z_{im} = 0$, (3) can be rewritten as a bi-quadratic equation and then analytically resolved [37], providing the ZIF ($\omega_0$)

$$\omega_0 = \left( \frac{C_d L - (C_{kin} + C_d) C_{kin} R_{kin}^2}{2 L C_d C_{kin} R_{kin}^2} + \frac{1}{L C_d C_{kin} R_{kin}^2} \right)^{0.5}.$$ \hspace{1cm} (4)

For the analytical analysis of the NZIF it is convenient to define two functions $h(\omega)$ and $g(\omega)$ as
\[ h(\omega) = LC_d\omega \left( C_{kin}^2 R_{kin}^2 \omega^2 + 1 \right) - (C_{kin} + C_d) C_{kin} R_{kin}^2 \omega^2 + 1, \]  
\[ g(\omega) = C_d \omega \left( C_{kin}^2 R_{kin}^2 \omega^2 + 1 \right), \]

accordingly. Given (5a) and (5b), an implicit function \((H)\) can be defined from (3) as

\[ H(\omega, Z_{im}) = h(\omega) - Z_{im} g(\omega). \]  

A first-order Maclaurin series approximation can be used to solve the (N)ZIF \((\omega)\) as a function of \(Z_{im}\), according to

\[ \omega(Z_{im}) \approx \omega_0 + \frac{d\omega}{dZ_{im}} \bigg|_{Z_{im}=0} Z_{im}, \]  

where \(\omega_0\) is the ZIF defined by (4). The derivative term at the right-hand side of (7) can be determined by differentiating the implicit function in (6) and finally yields [41]

\[ \frac{d\omega}{dZ_{im}} \bigg|_{Z_{im}=0} = \frac{g(\omega_0)}{h'(\omega_0)}, \]

where

\[ h'(\omega_0) = 2\omega_0 L C_d \left( C_{kin}^2 R_{kin}^2 \omega_0^2 + 1 \right) + 2\omega_0^3 L C_d C_{kin}^2 R_{kin}^2 \]

\[ - 2\omega_0 (C_{kin} + C_d) C_{kin} R_{kin}^2 \]

which is the derivative of (5a) with respect to \(\omega\) calculated at \(\omega_0\).

The internal resistances of Li-ion batteries strongly depend on temperature and can be described by the Arrhenius relationship [42], [43], according to

\[ \frac{1}{R_{kin}} = A \exp \left( -\frac{E_a}{RT} \right), \]

where \(A\) is the pre-exponential factor, \(E_a\) is the activation energy, \(R\) is the gas constant and \(T\) is the absolute battery temperature. The used simulation values for these parameters are shown in Table I. Equation (10) can be substituted for \(R_{kin}\) in all above equations. Subsequently, (7) can be plotted as a function of temperature for various \(Z_{im}\) values. A typical example of such a plot is shown in Fig. 2. Since this is only an example to show the NZIF behavior, the y-axis scale is not an example to show the NZIF behavior, the y-axis scale is not

\[ \begin{array}{|c|c|c|c|}
\hline
Symbol & Quantity & Value & Unit \\
\hline
C_{kin} & Kinetic capacitance & 10 & mF \\
C_d & Diffusion capacitance & 1 & mF \\
L & Inductance & 7 & nH \\
A & Pre-exponential factor & 400,000 & \Omega^{-1} \\
E_a & Activation energy & 10 & kJ mol^{-1} \\
R & Gas constant & 8.314 & J mol^{-1} K^{-1} \\
T & Temperature & 253 \ldots 323 & K \\
\hline
\end{array} \]

III. EXPERIMENTAL

In order to investigate the (N)ZIF as a function of temperature, laboratory experiments have been conducted with prismatic 90 Ah LiFePO4 (Thundersky TS-LFP90AHA) batteries. The impedance spectra were collected in the galvanostatic mode using printed-circuit boards (PCB) developed by NXP Semiconductors. A simplified circuit diagram of the PCB connected in parallel to a battery is shown in Fig. 3a. An alternating current is generated through the battery by opening and closing a switch (Fig. 3a) with a pulse-density modulated (PDM) signal of a sine wave (Fig. 3b). The AC voltage response of the battery can then be measured through a low-pass filter in order to calculate the complex impedance.

The impedances were measured at twenty five logarithmically distributed frequencies in the range of 10 to 5000 Hz.
with a sine-wave current of 100 mA RMS. The ZIF and NZIF are determined by interpolating the imaginary part of the impedance to $Z_{im} = 0$ and $Z_{im} > 0$, respectively.

All battery impedance measurements were conducted at a constant temperature of -20, -10, +10, +30 and +50°C using climate chambers (Maccor MTC-010). The State-of-Charge (SoC) has been controlled in all experiments at 20, 40, 60, 80 and 100%. Before the impedance measurements were started the batteries were allowed to equilibrate for 4 hours at the various temperatures. Automatic cycling equipment (Maccor 4200) was used to (dis)charge the batteries.

In order to investigate the (N)ZIF in a full EV under driving conditions [44]–[46], the EIS spectra for three selected 90-Ah prismatic LiFePO$_4$ batteries were recorded during test drives on a public road. The installed batteries were identical to the individual cells investigated under the temperature-controlled laboratory conditions described above. During the test drives impedances were measured at predetermined frequencies with a 1 s time interval. In addition to the impedance measurements the current flowing through the battery pack was measured simultaneously.

In order to analyze and quantify the interfering current resulting from the electric components in the EV more accurately, the battery pack current was recorded at a much higher sampling rate (100 kHz) on a vehicle test bench (dynamometer). A current sensor with a current range of ±500 A was used to record the pack current.

IV. RESULTS

A. Zero-Intercept Frequency for LiFePO$_4$ batteries

In previous work [36], [37] we have shown that the ZIF for small-size Li-ion batteries can be related to the integral battery temperature. Fig. 4 shows the ZIF ($f_0$) as a function of temperature for a large-capacity (90 Ah) LiFePO$_4$ battery measured under laboratory conditions. It can be concluded that $f_0$ decreases with increasing temperature and that this relationship is essentially independent of SoC, indicating that this specific battery characteristic is generic. Moreover, measurements have shown that it also holds for both small and large capacity Li-ion batteries and that it is aging independent [36], [37]. The only difference is that the present high-capacity LiFePO$_4$ batteries show significantly lower ZIF-values. This is shown in Fig. 5, where ZIF ($f_0$) measurements are plotted as a function of battery capacity. This figure illustrates that the ZIF is lower for high-capacity batteries than for low-capacity batteries.

B. Electric vehicle measurement setup

An electric research vehicle has been used to measure the battery impedance and pack current under driving conditions [44]–[46]. The battery pack of this EV is composed of 91 prismatic LiFePO$_4$ batteries in series. A photograph of one of the three battery modules is shown in Fig. 6, which in this case consists of a series assembly of 28 batteries. Each battery is monitored by a conventional cell board (Elithion). These conventional cell boards (see Fig. 6a) are all connected to the Battery Management System (BMS) of the EV.

The designed PCBs to measure the impedance are rigidly connected to the terminals of three selected batteries, as indicated in the inset of Fig. 6. In order to accurately measure the impedance it is very important that the PCB is properly connected to the batteries and that a four-point measurement set-up is applied. The current flowing through the battery pack is measured by a similar PCB as used for the impedance measurements.
measurements. This PCB measures the voltage across a shunt resistance of 100 $\mu\Omega$ (Fig. 6e). The shunt is connected in series with the battery pack.

C. Zero-Intercept Frequency measurements in electric vehicle

To demonstrate the concept of measuring the ZIF in a battery-powered application, the impedance is sequentially measured at 300 and 400 Hz with a time interval of 1 s during a test drive on a public road. Since the batteries in the EV were at a temperature of about 20°C during the test drive, it is expected according to Fig. 4, that the ZIF is between 300 and 400 Hz. Therefore these two measurement frequencies were selected in this case. Fig. 7 shows the results obtained for a single cell in the battery pack for about 20 min. The pack current is shown in Fig. 7a, where a negative current refers to a discharge current upon driving and a positive current to regenerative braking, i.e. corresponding to the battery charging process. Fig. 7b shows the correspondingly measured imaginary parts of the impedance. The 300 Hz results (red symbols) generally generate positive values while the 400 Hz results (blue symbols) generally reveal negative values. However, obviously the results at both frequencies are highly scattered when the pack current is high. On the other hand, the impedance results show much less dispersion when the pack current is relatively low. The large spread in these impedance results cannot be attributed to temperature variations as the battery (pack) temperature hardly changes on this time scale, as confirmed by other measurements. These results therefore suggest that the pack current powering the EV significantly interferes with the impedance measurements.

The dispersion in impedance becomes even more apparent in Fig. 8 where the measured values are plotted in the complex plane. Occasionally large outliers and even unrealistic negative values of the real part of the impedance can be found. The largest outliers are found at 300 Hz and are even outside the scale of the figure, as indicated by the arrow. The extremely deviating values are indicated in the caption of Fig. 8.

The ZIF can be determined by interpolating the imaginary impedances at 300 and 400 Hz to $Z_{\text{im}} = 0$. The interpolated
$f_0$-values are shown in Fig. 7c and also reveal quite some scatter and large outliers even at unrealistic negative frequencies. Obviously, using these raw data will lead to inaccurate temperature measurements, unless sophisticated filtering techniques are applied. These results suggest that interferences generated by the electronics of the EV introduce large errors in the measured impedance and, consequently, also in the ZIF. To investigate the source of these interferences in more detail the battery pack current in the EV has been investigated at a significantly higher sampling rate.

### D. Battery-pack current spectrogram

In order to identify and quantify the battery pack current interferences in the EV, test-bench measurements have been performed under laboratory conditions. The tests, mimicking a driving situation, included three cycles of acceleration (to 50 km/h) and deceleration (regenerative braking). Fig. 9a shows a typical example of the resulting battery pack current profile measured for 2 min with a sampling frequency of 100 kHz. This sampling frequency is much higher than that reported for the test drive shown in Figs. 7 and 8. Negative and positive currents in Fig. 9a again refer to discharging (driving) and charging (braking), respectively. Discharge currents up to 200 A have been measured, which corresponds to a C-rate of approximately 2.2. The current levels shown in Fig. 9a are very similar to those shown in Fig. 7.

A spectrogram was calculated based on these current measurements by taking a Fast Fourier Transform (FFT) with a moving window of 1 s ($10^5$ samples) and an overlap of 75% (0.75 s). Since the measured data are not periodic, a Hanning window has been used. The spectrogram of Fig. 9b shows the current (indicated by colors) flowing through the battery pack as a function of time and frequency. Note that the time axis of Fig. 9a and Fig. 9b are the same. Obviously, when current starts to flow the current amplitude increases rapidly up to a frequency of 1 kHz. Beyond 1 kHz no significant increase is visible. The red semicircles found in Fig. 9b represent the electric motor speed, the lowest semicircle is the base frequency and the upper ones are the corresponding harmonics. The spectrogram shows very similar behavior for accelerating and decelerating, resulting in the characteristic symmetric semi-circles.

Additional measurements without regenerative braking have
also been performed. These measurements revealed that the current levels in the low frequencies (\(f < 10\ Hz\)) are lower compared to those found with regenerative braking. At 110 s the power to the electric system of the car has been disconnected which causes a relatively high current for a short period of time up to a frequency of 400 Hz. This event is also visible in the small peak current in Fig. 9a. After this moment the current is zero.

In addition to the measurement shown in Fig. 9 a similar test-bench run has been performed by accelerating the EV to a speed of 100 km/h with subsequent regenerative braking to standstill. The corresponding pack current is shown in Fig. 10a. In this test-bench run the speed is twice as large and the pack current reaches somewhat higher values than in the previous run. In Fig. 10b the calculated spectrogram of the pack current is shown. In this spectrogram one characteristic semicircle is visible during accelerating and decelerating the EV. Despite the high motor speed and current, the amplitude of the pack current does not increase significantly beyond a frequency of 1 kHz. Around 80 s the power to the electric system of the EV was disconnected for 4.5 s and subsequently reconnected again. At 88 s the power has been disconnected permanently. These events are causing three peaks in current amplitude up to 400 Hz which are clearly visible in Fig. 10b.

From the results shown in the spectrograms of Figs. 9 and 10 it can be concluded that the current extracted from or supplied to the battery pack in an EV are highly dynamic over a wide frequency range, which is caused by the accelerating and regenerative braking process. In order to visualize this in more detail, a current spectrum has been taken from Fig. 9 in the time interval between 14 and 15 s. The resulting Fig. 11 shows that the current decreases about two orders of magnitude between 100 Hz and 1 kHz. The average current hardly changes anymore at frequencies beyond 1 kHz.

The EIS measurements presented in Fig. 7 and Fig. 8 were taken at 300 and 400 Hz. These frequencies are indicated by two paired horizontal (black) and vertical lines in the spectrogram in Fig. 9b and Fig. 11, respectively. From these figures it can be concluded that relatively high currents, of the order of 10 mA, are present at these two frequencies, as dictated by the driving process of the EV. If for example a 100 mA EIS-excitation signal is applied for the impedance measurements, this 10 mA current will give rise to considerable interferences, resulting in the poor SNR found in Fig. 7. It is therefore to be expected that much more accurate impedance results can be obtained in a frequency range where the current interferences are relatively small. Considering Figs. 9b and 10b, selecting 1600 and 2000 Hz (see horizontal lines) as measurement frequencies is therefore expected to be much more favorable. The current spectrum of Fig. 11 indeed reveals that the interfering current is only about 1 mA under these frequency measurement conditions, which is one order of magnitude lower in comparison to the lower frequency measurements. From Fig. 10b can be seen that selecting a set of frequencies lower than 1600 Hz would not result in a complete interference-free region. Therefore 1600 Hz is the minimum frequency to select in this particular application. At the high frequencies the impedance shows a straight line in the inductive area in the Nyquist plot, which is advantageous for linear interpolation to the NZIF. Therefore the new selected frequency window can be larger, i.e. 1600-2000 Hz, in comparison to the previous one, which was only 300-400 Hz.

Obviously, measuring the impedance characteristics at higher frequencies will not lead anymore to the NZIF where the imaginary part of the impedance is per definition zero. Therefore the NZIF method is introduced in this contribution.

E. Non-Zero-Intercept Frequency measurements

Fig. 12 shows the temperature dependence of the (N)ZIF of a 90-Ah LiFePO\(_4\) battery measured in various frequency ranges. The temperature of individual batteries has been accurately controlled. For comparison the \(f_0\) (dark blue) curve, representing the ZIF interpolated at \(Z_{im} = 0\), is also included in Fig. 12. The NZIF, interpolated at other values \((i)\) of the imaginary part of the impedance, is denoted by \(f_i\). For example, \(Z_{im} = 0.2\ m\Omega\) refers to \(f_{0.2}\) (light blue curve) and \(Z_{im} = 0.8\ m\Omega\) to \(f_{0.8}\) (magenta curve in Fig. 12). Every line shows the average of five different SoC-values, essentially independent on SoC. It can be seen that higher interpolation values lead to higher intercept frequencies as already expected from the theoretical analysis in Section II. The temperature dependencies of all NZIFs are very similar to that of the ZIF method. From this experimental verification it can be concluded that in case we have significant interfering signals the NZIF method is an excellent alternative to determine battery temperatures at less noisy frequencies.

In line with previous investigations [37], additional EIS measurements have also been conducted with Cobalt-based Li-ion batteries. The (N)ZIF results gave similar dependencies as found with the LiFePO\(_4\) system. Other EIS measurements confirmed that this also holds for batteries with smaller storage capacities and for aged batteries.
E. Non-Zero-Intercept Frequency Measurements in Electric Vehicle

In order to investigate the accuracy of the proposed NZIF-method under real driving conditions, another test drive has been performed on a public road at the preselected optimal frequencies of 1600 and 2000 Hz. The measurements at these two frequencies have been performed in alternating intervals of 1 s. The results are shown in Figs. 13 and 14. Fig. 13a shows that the dynamics in current are of the same order of magnitude as shown in Fig. 7a, which indicates that both measurements are comparable. Fig. 13b shows that the pack current hardly has an influence on the imaginary values of the impedance. The results are now much more stable and all values measured at 1600 Hz are lower than 0.65 mΩ and at 2000 Hz the values are all higher than 0.65 mΩ. Furthermore no outliers can be detected. This is also demonstrated in Fig. 14, where the impedance is plotted in the complex plane. The results indeed show much less distribution in real and imaginary part in comparison to the results presented in Fig. 8. No negative real impedances and large outliers can be detected, essentially indicating that the interfering currents do not influence these NZIF measurements.

The NZIF has been determined by interpolating the imaginary values of the impedance between both frequencies to $Z_{im} = 0.65$ mΩ. Fig. 13c shows these interpolated NZIF ($f_{0.65}$) results. These results clearly indicate that the present NZIF-method generates much more stable and hence accurate results and does not show any outliers. From these results it can be concluded that it is indeed useful to perform EIS measurements at frequencies where the battery pack current amplitudes and, consequently, the interferences are relatively low. As a result the SNR will become higher and the temperature estimation more accurate. Obviously the accuracy of the final temperature indication is also dependent on the signal processing of the raw data, the measurement device accuracy and the magnitude of the battery impedance. Without any additional signal processing a 30 times improvement has.
been achieved in terms of standard deviation by adopting the NZIF method instead of the ZIF method in this particular application.

G. Non-Zero-Intercept Frequency measurements in electric vehicle under dynamic temperature conditions

In Sections IV-C and IV-F measurement results of the ZIF and NZIF are shown and discussed from test drives with an EV on a public road. Due to the relatively short test periods in which the battery temperature is more or less constant these results are very well suited to investigate the differences between the ZIF and NZIF. In order to see changes in battery temperature another test drive was performed of which the duration was 5.5 times longer in comparison to the test drives shown in Figs. 7 and 13. For a fair comparison the y-axes in Fig. 15 are the same as in Fig. 13. In the test drive shown in Fig. 15 the same set of optimal frequencies (1600 Hz and 2000 Hz) were used since it was favorably concluded that these are promising for an accurate temperature indication, in contrast to the 300 Hz and 400 Hz frequencies.

In Fig. 15a the drive current is plotted as a function of measurement time. Negative and positive currents represent driving and braking, respectively. At time \( t = 0 \) driving was started from the garage in which the EV was parked. After approximately 60 min the EV was parked back in the garage for 6 min. Subsequently the EV went back on the road again for 20 min of driving. At \( t \approx 90 \) min the EV has been connected to a power plug in the garage in order to charge the battery pack with a current of 10A (0.1 C-rate). Fig. 15b shows the imaginary impedances measured at 1600 Hz and 2000 Hz. The NZIF, shown in Fig. 15c, is interpolated at \( Z_{im} = 0.65 \, \text{m} \Omega \). As expected, the imaginary impedances and NZIF measurement results are stable and not showing large outliers, in spite of the highly dynamic pack current (Fig. 15a) generated by the electric components of the EV.

Fig. 15d shows the temperature developments as a function of driving time: (i) the impedance-based temperature \( T_{EIS} \) which is calculated from \( f_{0.65} \), (ii) the temperature on the negative terminal of the battery \( T_{Neg} \) and (iii) the temperature at the bottom surface of the aluminum container in which the battery pack is assembled \( T_{Container} \). The two latter temperatures were measured by TSP-TH NTC thermistors from Thorlabs. The temperature of all components at \( t = 0 \) is about 22°C. This is the ambient temperature in the garage where the vehicle was parked. After starting driving \( T_{EIS} \) and \( T_{Neg} \) increase due to the heat generated inside the batteries. However, the container temperature decreases since the outdoor temperature was 6°C lower than in the garage. After about 60 min of driving the container temperature quickly increases because the EV was parked back in the garage for 6 min. After this resting period the drive was continued. At approximately 90 min the EV has been parked back in the garage. This increased the container temperature rapidly to a level similar to \( T_{EIS} \) and \( T_{Neg} \). After parking the EV no considerable changes in temperature evolution can be seen anymore. That means that the 10A charge current does not increase the battery temperature further.

It can be concluded that \( T_{EIS} \) follows the development of \( T_{Neg} \) closely in this experiment. Due to the low average current (0.3C) and the large thermal mass of the battery pack, the temperature increases slowly and therefore temperature gradients in the cell core are expected to be relatively small. The integral battery temperature \( T_{EIS} \) is generally somewhat lower than \( T_{Neg} \), which is measured locally at the negative battery terminal. The reason for a lower \( T_{EIS} \) is that the relatively cold container cools down the bottom of the battery pack. This cannot be detected by a temperature sensor at the top of the pack where the negative terminal is located. That also explains the local peak in \( T_{Neg} \) at 85 min, corresponding to driving with a large current. More heat is developed at the terminals since the current density there is relatively high. Obviously, the final temperature indication is dependent on the post processing of the measurement data. A standard deviation of ±1°C in the final indicated temperature has been achieved by using a simple moving average filter with a window length of 36 samples.

V. CONCLUSIONS

It has been found that the zero-intercept frequency (ZIF) can be considerably influenced by interfering currents flowing through the battery pack under operating conditions of, for example, an electric vehicle (EV). This can make temperature determination based on the proposed ZIF-method inaccurate.
The interfering currents may disturb the impedance measurements in such a way that it is complicated to apply the interpolated ZIF-method for temperature determination without sophisticated filtering of the measurement data.

Based on the experiments performed with an EV it has favorably been concluded that the battery pack current at frequencies beyond 1 kHz are about two orders of magnitude smaller than at lower frequencies. This observation opened the way to propose the modified non-zero intercept frequency (NZIF) method to measure the impedance with significantly higher signal-to-noise ratios. The NZIF-method shows a much more accurate temperature dependence in this application in comparison to the ZIF-method and is therefore a more reliable temperature indication method. Moreover, dependent on the electronic application and conditions, the NZIF-method can be adapted to an optimal frequency range where the battery pack current is low and hence improving the SNR.

Mathematical modeling and experiments with 90-Ah LiFePO4 batteries revealed that the NZIF show similar behavior in comparison to the ZIF as a function of temperature. The proposed NZIF method is therefore very suitable to measure the battery temperature under operating conditions in various electronic applications.

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REFERENCES


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