

Rapid Impedance Spectrum Measurements for State-of-Health Assessment of Energy Storage Devices

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ABSTRACT

Harmonic Compensated Synchronous Detection (HCSD) is a technique that can be used to measure wideband impedance spectra within seconds based on an input sum-of-sines signal having a frequency spread separated by harmonics. The battery (or other energy storage device) is excited with a sum-of-sines current signal that has a duration of at least one period of the lowest frequency. The voltage response is then captured and synchronously detected at each frequency of interest to determine the impedance spectra. This technique was successfully simulated using a simplified battery model and then verified with commercially available Sanyo lithium-ion cells. Simulations revealed the presence of a start-up transient effect when only one period of the lowest frequency is included in the excitation signal. This transient effect appears to only influence the low-frequency impedance measurements and can be reduced when a longer input signal is used. Furthermore, lithium-ion cell testing has indicated that the transient effect does not seem to impact the charge transfer resistance in the mid-frequency region. The degradation rates for the charge transfer resistance measured from the HCSD technique were very similar to the changes observed from standardized impedance spectroscopy methods. Results from these studies, therefore, indicate that HCSD is a viable, rapid alternative approach to acquiring impedance spectra.

KEY WORDS: impedance spectra, rapid measurements, state-of-health, lithium-ion

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INTRODUCTION

Battery technologies and other energy storage devices (e.g., fuel cells and ultracapacitors) have become critical components to several industries including automotive, military, space, medical, electric utility, and consumer electronics. Since battery systems can be very expensive, successful manufacturing and implementation in the field benefit from accurate, in-situ monitoring techniques for state-of-health and remaining useful life assessment. State-of-health (SOH) is a measure of the battery's condition relative

to its rated condition at beginning of life; remaining useful life (RUL) is an estimation of the remaining battery usage given its previous history and present power and energy demand. Developing industry standards for SOH and RUL, however, has been challenging because of the complexity of the problem. Passive monitoring techniques (e.g., observing changes in voltage and current along with ambient temperature) can yield valuable information about battery behavior, but the acquired data generally only track changes in capacity and state-of-charge (SOC), which is the percentage of remaining capacity relative to the rated

capacity [1,2,3,4]. With increasing demands on the battery performance, however, simple passive monitoring is becoming an insufficient measure of SOH. Knowledge of resistance rise, and the corresponding power fade, is also critical for a complete assessment of battery health. Resistance is typically determined using a volt/ohm meter at a specific measurement frequency or with active pulse tests over an SOC range using a constant current signal and observing the voltage response [5,6,7,8], but these approaches are not well suited for rapid battery diagnostics (or other energy storage devices).

Recently, AC impedance has been emphasized as a useful measure of battery SOH and RUL [9,10,11,12,13,14,15,16]. Impedance spectra are acquired through low-level, charge-neutral input signals at various frequencies, and the data have been shown to correlate well with the corresponding resistance and power capability results that are determined from standardized pulse tests [9]. However, full-spectrum electrochemical impedance spectroscopy (EIS) typically requires expensive and delicate equipment operating in a laboratory environment. EIS measurements also tend to be sequential and can take a long time to complete (usually from ten minutes to an hour depending on settings), especially when very low frequencies are included [10]. As a result, most of the research to date has emphasized either offline EIS measurements or online measurements at only a small set of frequencies (e.g., sequentially measuring the impedance at a low, medium, and high frequency) to assess battery health and life [12,13,14,15,16].

BACKGROUND

ELECTROCHEMICAL IMPEDANCE SPECTROSCOPY

Electrochemical impedance spectroscopy testing generally consists of a small, charge-neutral AC voltage signal applied to the battery over a broad range of frequencies (e.g., 10 kHz to 10 mHz). The current response is then measured to determine the impedance at each desired frequency. Alternatively, an AC current signal is injected into the battery and the voltage response is measured. A typical EIS Nyquist curve measured at 60% SOC for a commercially available Sanyo SA cell (1.2 Ah) is shown in Figure 1. The point at which the high-frequency tail crosses the real axis is the ohmic resistance (R_O) within the battery, which includes the effects from the electrolyte, electrodes, tabs, etc. The semicircle in the mid-frequency region identifies the charge transfer resistance (R_{CT}). The charge transfer resistance is primarily influenced by the kinetic reactions at the electrodes due to an imposed constant voltage or current signal. The low-frequency tail on the right is known as the Warburg impedance that is caused by the diffusion of ions [17].

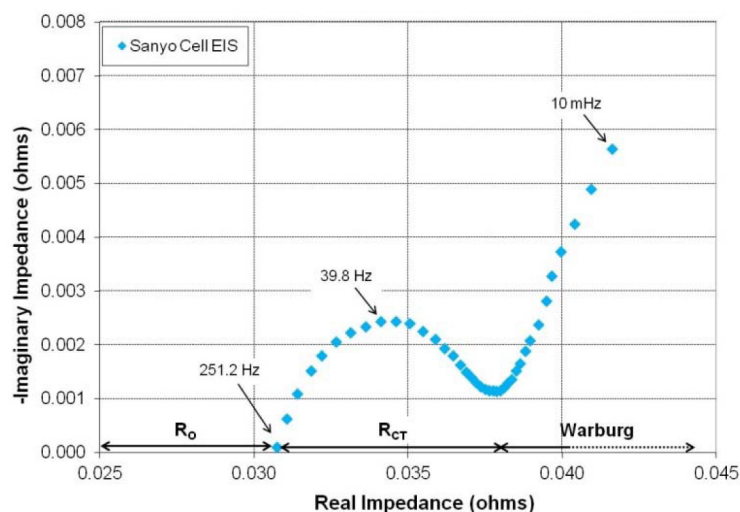


Figure 1. EIS Nyquist curve description.

Figure 2 shows EIS measurements for a Sanyo SA lithium-ion cell at different stages of life. It was subjected to more than 80,000 shallow cycles (i.e., approximately 1% Δ SOC swings) at a test temperature of 50°C, as described below. There is a small increase in the ohmic resistance, approximately 3 m Ω , which may come from the solid electrolyte interphase layer and other side reactions consuming lithium from the electrolyte as the cell is aged [18]. The majority of the impedance growth, however, occurs in the mid-frequency charge transfer resistance, where the semicircle grows in both height and width as a function of cell age; the growth in charge transfer resistance is approximately 6 m Ω . The low-frequency Warburg tail shifts to the right due to the increased cell resistance but generally retains the same slope and is otherwise unaffected by cell aging.

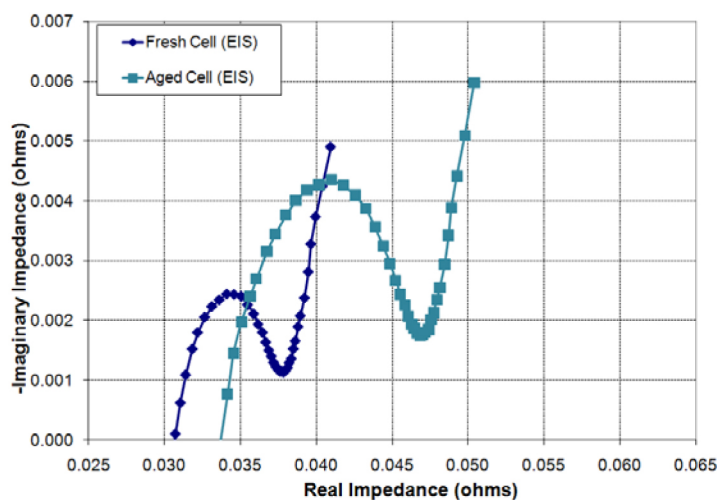


Figure 2. EIS Nyquist curves for an aging Sanyo SA cell.

The change in measured impedance as a function of aging has been shown to linearly correlate with the resistance growth and power fade that are determined from standardized pulse tests [9-10]. These observations have indicated that impedance measurements can be used as a benign alternative for SOH and RUL assessment [10-11]. Typical EIS measurements, however, must be performed offline in a laboratory setting, or online at a very small set of sequentially measured frequencies, and this is not practical for embedded, near real-time monitoring. Rapid, broadband impedance measurement techniques, including Compensated Synchronous Detection (CSD), have recently been developed for online, in-situ applications as a means of monitoring changes in resistance and power as the battery is being operated.

COMPENSATED SYNCHRONOUS DETECTION

The CSD technique [19] consists of simultaneously exciting the battery with a set of charge-neutral current sinusoids at different frequencies that are separated logarithmically. This sum-of-sines signal, therefore, contains multiple frequencies spread over the specified range of interest. For accurate detection with minimal error, it is advantageous to include several periods of each discrete frequency. The minimum number of periods required for an accurate measurement depends on the number of frequencies included in the overall excitation signal. Studies have shown that a minimum of three periods of the lowest frequency is generally sufficient for this technique when about 12 frequencies are included in the input signal (e.g., a low starting frequency of 0.1 Hz, having a period of 10 seconds, would require a 30-second measurement) [19-20]. If, however there are additional noise sources that affect the response signal, or if more frequencies or interest are included in the excitation signal, more periods of the lowest frequency will be required to minimize the crosstalk interference.

The impedance magnitude and phase can be synchronously detected at each frequency within the sum-of-sines signal using the input current and measured voltage response. Since the duration of the measurement is not infinite, the input and response signals appear as sinc functions in the frequency domain. These sinc functions can overlap and cause crosstalk error in the impedance spectra. To minimize this error, the initial results are compensated by first reassembling all of the components except for the frequency of interest back into a sum-of-sines signal in the time domain. This reassembled time record is then subtracted from the original response signal and synchronously detected again with significantly reduced crosstalk error. This process is repeated for each frequency component to yield more accurate impedance spectra that are directly comparable to the EIS [19,20,21].

The advantage of CSD is that it is a simpler implementation that can be significantly faster compared to EIS. Additionally, the technique can be successfully implemented on a hardware platform that can be designed as an embedded system. However, it is still not very well suited to near real-time, onboard applications since it requires at least three periods of the lowest frequency to successfully resolve the impedance spectra.

HARMONIC COMPENSATED SYNCHRONOUS DETECTION

A variant of CSD is Harmonic Compensated Synchronous Detection (HCSD). The HCSD technique enables the detection of impedance magnitude and phase assuming a bandwidth-limited sum-of-sines input current signal with a harmonic frequency spread. With harmonic separation, the crosstalk error observed with the CSD approach is eliminated. This removes the need for compensation and enables a measurement to be completed within only one period of the lowest frequency. The derivation of the HCSD technique is based on the input sum-of-sines signal shown in Equation (1), where A_i , ω_i , and ϕ_{in_i} are the amplitude, radian frequency, and phase, respectively, of the i^{th} input sinusoid, and M is the number of sinusoids within the excitation signal [19-20]. For a logarithmic distribution, the radian frequency (ω_i) can be governed by Equation (2), where the i^{th} sinusoidal signal is an octave harmonic of the lowest frequency (ω_1). This input signal is injected into the battery, and the measured response is sampled as shown in Equation (3), where B_i is amplitude of the i^{th} output sinusoid, Δt is the sampling time step, ϕ_{out_i} is the phase response of the i^{th} output sinusoid, and N is the number of points in the captured time record [19,20,21].

$$x(t) = \sum_{i=1}^M A_i \sin(\omega_i t + \phi_{in_i}) \quad (1)$$

$$\omega_i = \omega_1 \cdot 2^{i-1} \quad i = 1 : M \quad (2)$$

$$y[j] = \sum_{i=1}^M B_i \sin(\omega_i (j-1)\Delta t + \phi_{out_i}); \quad j = 1 : N \quad (3)$$

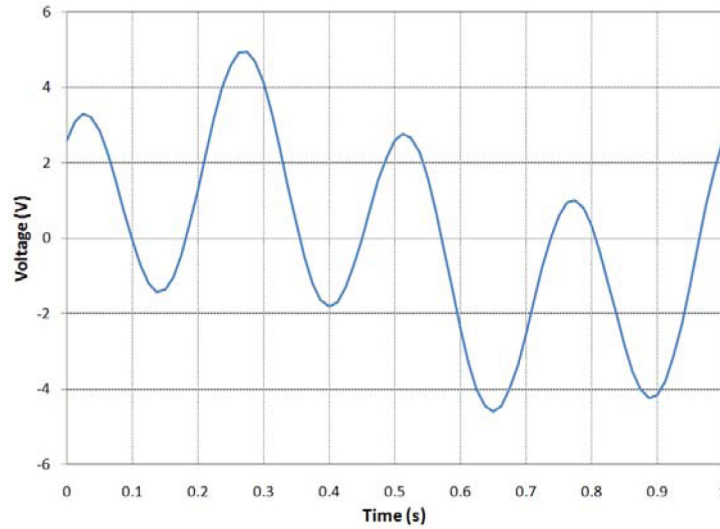


Figure 3. Sum-of-sines voltage response for one period from Equation (6).

As an example, consider a sum-of-sines input current signal, $x(t)$, consisting of two frequencies, ω_1 and $a\omega_1$, respectively (where a is an even integer) as shown in Equation (4). The resulting voltage response, $y[j]$, is shown in Equation (5), where the sine waves have been affected by unknown phase shifts, ϕ_{out1} and ϕ_{out2} . A sample voltage response, represented by Equation (6), for one period of the lowest frequency is shown graphically in Figure 3, where $B = [2, 3]$, $\omega_i = [2\pi, 8\pi]$ with $a = 4$ (i.e., $a\omega_1 = 4.2\pi$), and $\phi_{out_i} = [0, \pi/3]$.

$$x(t) = A_1 \sin(\omega_1 t + \phi_{in_1}) + A_2 \sin(a\omega_1 t + \phi_{in_2}) \quad (4)$$

$$y[j] = B_1 \sin(\omega_1 (j-1)\Delta t + \phi_{out_1}) + B_2 \sin(a\omega_1 (j-1)\Delta t + \phi_{out_2}); \quad j = 1:N \quad (5)$$

$$y[j] = 2 \sin(2\pi(j-1)\Delta t) + 3 \sin\left(8\pi(j-1)\Delta t + \frac{\pi}{3}\right); \quad j = 1:N \quad (6)$$

The voltage magnitude of this signal in the frequency domain, $|Y(\omega)|$, is shown in Figure 4. The resulting sinc functions are centered at the frequencies of $\pm\omega_0$ and $\pm a\omega_0$ (i.e., ± 1 and ± 4 Hz since $\omega_1 = 1$ rad and $a = 4$), as expected. Furthermore, crosstalk interference from the other sinc functions converge to zero at ± 1 and ± 4 Hz since the two input sinusoidal waves in Equation (6) are separated by an integer step factor.

This voltage response signal can then be synchronously detected for the in-phase and quadrature components. The in-phase response, S_P , at the k^{th} radian frequency (ω_k) is shown in Equation (7). This reduces to the real component of the

complex impedance as shown in Equation (8), where $\phi_{detected}$ is the detected phase for the k^{th} frequency of interest. Similarly, the quadrature component, S_Q , is shown in Equation (9), and it can be reduced to the imaginary component of the complex impedance as shown in Equation (10).

$$S_{P_k} = \frac{1}{N} \sum_{j=1}^N \left\{ \frac{A_k B_k}{2} \left[\cos(\phi_{in_k} - \phi_{out_k}) - \cos(2\omega_k (j-1)\Delta t + \phi_{in_k} + \phi_{out_k}) \right] + \sum_{i \neq k=1}^M \frac{A_k B_i}{2} \left[\cos((\omega_k - \omega_i)(j-1)\Delta t + \phi_{in_k} - \phi_{out_i}) - \cos((\omega_k + \omega_i)(j-1)\Delta t + \phi_{in_k} + \phi_{out_i}) \right] \right\} \quad (7)$$

$$S_{P_k} = \frac{A_k B_k}{2} \cos(\phi_{detected_k}) \quad (8)$$

$$S_{Q_k} = \frac{1}{N} \sum_{j=1}^N \left\{ \frac{A_k B_k}{2} \left[\sin(\phi_{out_k} - \phi_{in_k}) + \sin(2\omega_k (j-1)\Delta t + \phi_{in_k} + \phi_{out_k}) \right] + \sum_{i \neq k=1}^M \frac{A_k B_i}{2} \left[\sin((\omega_k + \omega_i)(j-1)\Delta t + \phi_{in_k} + \phi_{out_i}) - \sin((\omega_k - \omega_i)(j-1)\Delta t + \phi_{in_k} - \phi_{out_i}) \right] \right\} \quad (9)$$