

Long-Term Validation of Rapid Impedance Spectrum Measurements as a Battery State-of-Health Assessment Technique

Jon P. Christophersen
Idaho National Laboratory

John L. Morrison
Montana Tech. of Univ. of Montana

Chester G. Motloch
Motloch Consulting, Inc.

William H. Morrison
Montana Tech. of Univ. of Montana

ABSTRACT

The objective of this study was to assess the long-term capability and impact of a rapid, in-situ impedance measurement technique known as Harmonic Compensated Synchronous Detection. This technique consists of a sum-of-sines excitation signal that includes a targeted selection of frequencies and only requires one period of the lowest frequency. For a given frequency range of 0.1 Hz to approximately 2 kHz, the measurement duration would only be ten seconds. The battery response is captured and synchronously detected for impedance spectra measurements. This technique was compared to laboratory-based performance degradation measurements using commercially available lithium-ion cells. The cells were aged for 150,000 cycles at accelerated rates using temperatures of 40 and 50°C. Every 30,000 cycles, cycle-life testing was interrupted to gauge degradation at the reference temperature of 30°C. The results demonstrated that growth in the ohmic and charge transfer resistances during aging strongly correlate with the corresponding changes in discharge capacity, pulse resistance, and available power capability that were independently determined from standardized test methods. Additionally, the rapid impedance spectrum technique appears to be a benign measurement that does not impact battery aging. Consequently, this technique appears to be useful as an onboard sensor that, when combined with passive measurements, may be used for enhanced diagnostics, management, and control.

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INTRODUCTION

Batteries and other energy storage devices are becoming increasingly important for several industries, including automotive, electric utilities, and military applications. For example, electric, hybrid, and plug-in-hybrid electric vehicles are placing higher demands on the battery packs for engine-off modes, as well as enhanced power assist for motive function to reduce fuel consumption. Batteries are also experiencing increasing power and energy requirements for portable military and commercial electronics, such as radios and computer systems. Battery technologies must perform in

normal as well as severe environments, such as military combat missions that may include temperatures ranging from -40°C to 50°C in military aircraft [1], or have a 15-year calendar life, as in the case of U.S. commercial hybrid electric vehicles [2,3]. These performance requirements, combined with the high costs of energy storage devices, emphasize the need for improved battery management systems that accurately assess the overall state-of-health and remaining useful life in addition to optimized management, control, and safety.

Typical management systems focus on passive measurements of voltage and current as a function of time and temperature to gauge battery health [4,5,6]. A smart system uses the measured data to count coulombs while also compensating for degradation losses (e.g., using a weighting factor to compensate for efficiency losses). These can also require periodic full discharges and charges to assess overall remaining capacity as a function of use and to recalibrate the system [4]. Alternatively, the remaining capacity could be estimated based on equivalent circuit models that track the changes in the main charge storing capacitor element using a sliding-mode observer and Kalman estimator techniques [5]. Other methods include offline testing to establish lookup tables that can be used in combination with expert learning systems to assess the state-of-health (SOH) from passive measurements. For example, Okoshi et al. developed lookup tables of SOC versus direct current resistance for vehicle idling-stop-system applications, and incorporated Kalman filtering techniques to predict SOH based on direct current measurements [6]. However, simple passive monitoring provides an incomplete picture of health and this forces manufacturers to oversize their battery designs which increases costs. With higher demands on battery technologies, improved health assessment techniques are required for enhanced management systems that can extend life, increase performance capability, improve safety, and provide useful information for secondary use applications (e.g., an automotive battery at end of life that could be used for grid stabilization).

Electrochemical Impedance Spectroscopy (EIS) has been shown to be a valuable laboratory tool for battery diagnostics. The acquired spectra can reveal changes in the bulk behavior of the electrochemical processes as the battery ages, which can give an indication of deterioration in the electrode surface and diffusion layer [7]. Parameters extracted from impedance spectra, such as the ohmic and charge transfer resistances, have been incorporated into many modeling tools (physics-based, empirical, and semi-empirical models) for offline estimations of battery performance and overall life capability [7,8,9]. Although EIS has not generally been considered for online sensing (historically, the measurement time can be lengthy and the required equipment is typically bulky and expensive), rapid impedance spectra measurement techniques have been developed using a simplified hardware platform that could be designed as an embedded system [10-11]. These techniques could be implemented as an onboard sensor that, when combined with passive measurements, may be used for enhanced battery diagnostics, management, and control. A prototype SOH assessment architecture based on the rapid impedance measurement techniques for onboard battery management systems is presently under development [12].

EXPERIMENTAL

The purpose of this study was to assess the long-term capability and impact of a rapid, in-situ impedance measurement technique known as Harmonic Compensated Synchronous Detection (HCSD). The HCSD technique consists of a sum-of-sines excitation current signal that includes frequencies separated by octave harmonics so as to eliminate the effects of crosstalk interference. With no crosstalk error, the duration of the excitation signal needs to be only one period of the lowest frequency. Thus, given a frequency range of 0.1 to 1638.4 Hz with 15 frequencies included within the sum-of-sines excitation signal, the measurement duration would only be ten seconds long. Each of the harmonic frequencies would include multiple periods to fill the ten-second measurement duration (e.g., the first harmonic of 0.2 Hz would have two periods within the sum-of-sines, the next harmonic of 0.4 Hz would have four periods, etc.). The overall battery response to the excitation signal is captured and then synchronously detected at each frequency of interest to determine the impedance spectrum [10].

Long-term validation of HCSD was conducted on commercially available, high-power lithium-ion cells having a layered oxide cathode with Nickel Manganese Cobalt (NMC) and graphite anode [13]. The cells are 18650-size and have a rated capacity of 1.2 Ah. Several of these cells were subjected to aging protocols for the purpose of validating the Plug-in Hybrid Electric Vehicle (PHEV) testing methodology [3]. As part of that effort, 25 cells were characterized and then subjected to life aging at various temperatures using the test matrix shown in Table 1 [13]. Characterization testing was performed at a reference temperature of 30°C, beginning with a sequence of constant current discharges from a fully charged state. The first three discharges were performed at the one-hour rate (i.e., 1.2 A) to ensure cycling stability. This was followed by a constant power discharge test and a low-current Hybrid Pulse Power Characterization (L-HPPC) test. The constant power discharge test is a scaled, 10-kw discharge between the maximum and minimum voltages. The cells had a scaling factor of 1400, so the constant power test was performed at 7.14 W [13]. The L-HPPC test profile is shown in Figure 1, and consists of 10-s discharge and regen pulses with a 40-s rest at open circuit voltage in between [3]. This profile is repeated at each 10% depth of discharge (DOD) increment starting at 90% state-of-charge. After the L-HPPC, a 25-hour static capacity test was performed using a constant current of 48 mA (i.e., 1.2 A / 25) between the maximum and minimum voltages. Finally, the cells were characterized with an EIS measurement at 15% DOD over a frequency range of 100 kHz to 0.01 Hz with ten points per decade of frequency.

Table 1. Control group test matrix for PHEV validation testing.

Group	# of Cells	Temperature	SOC
1	10	30°C	60%
2	5	40°C	60%
3	5	50°C	60%
4	5	60°C	60%

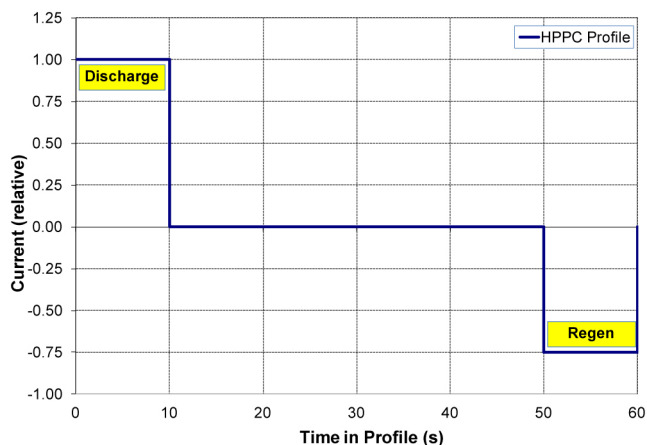


Figure 1. HPPC profile that is repeated at each 10% DOD increment [3].

Long-term aging for these cells consisted of cycle-life testing using the 50-Wh PHEV charge-sustaining test profile that is shown in Figure 2. This constant power profile is scaled to meet the cell requirements (using a factor of 1400 in this case) and was repeated continuously at the designated temperature and state-of-charge (SOC) from Table 1. Cycling was interrupted every 30,000 cycles for reference performance tests (RPTs) to gauge cell degradation as a function of aging. Reference performance tests consisted of a scaled, 10-kW constant power discharge and L-HPPC test at 30°C. Every fifth RPT, a 25-hour static capacity test and an EIS measurement at 15% DOD were also included [13].

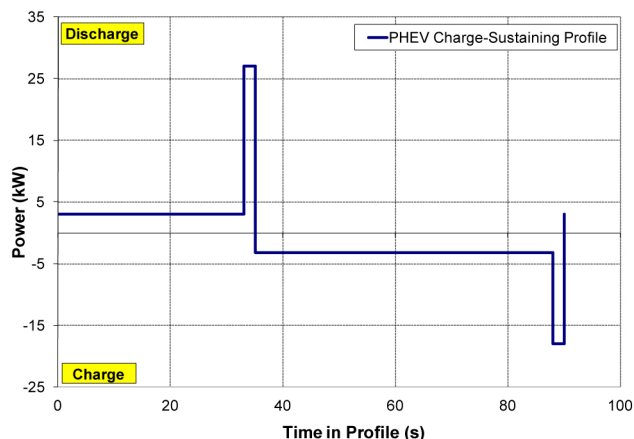


Figure 2. Charge sustaining cycle-life test profile [3].

The acquired test data from Table 1 have been discussed by Belt et al. [13] and were used as a control group for this HCSD validation study. Portions of the control group matrix in Table 1 were duplicated using twelve fresh cells with the addition of HCSD measurements at each RPT. The test matrix is shown in Table 2, and consists of accelerated aging at 40 and 50°C. As with the control group, the HCSD study cells were cycled at 60% SOC based on the PHEV charge-sustaining profile (see Figure 2) with RPTs every 30,000 cycles. Groups A and C in Table 2 were intended to verify the effectiveness of HCSD measurements under no-load conditions. Six cells (i.e., three cells per temperature group) were subjected to the same testing regime as the control group, but with the addition of an HCSD measurement at 60% SOC at characterization and every RPT immediately prior to the start of each cycle-set. Groups B and D in Table 2 included HCSD measurements every RPT as well, but were also subjected to ten thousand HCSD measurements while under active cycling to determine the impact of impedance measurements under load.

Table 2. Test matrix for HCSD validation study.

Group	# of Cells	Temperature	SOC	Comment
A	3	40°C	60%	No-Load Only
B	3	40°C	60%	Under Load
C	3	50°C	60%	No-Load Only
D	3	50°C	60%	Under Load

RESULTS AND DISCUSSION

The cells were aged for 150,000 cycles at the designated test temperature and SOC, resulting in seven HCSD measurements under no-load conditions as the cells degraded (characterization, end of life, and every RPT in between). The input sum-of-sines current for the rapid impedance spectra measurements covered a frequency range of 0.1 to 1638.4 Hz in octave harmonics with one period of the lowest frequency (i.e., 10 seconds) using an excitation current of 0.5 A_{RMS}. The cells with measurements under load (i.e., Groups B and D in Table 2) were each subjected to a total of 10,000 HCSDs during life testing as well. Impedance spectra results from the under load studies will be discussed in a separate publication.

Performance Degradation

Figure 3 shows a comparison of the average pulse resistance growth (determined from the L-HPPC test) between the HCSD study cells and the corresponding control groups at 60% SOC. The average control group results are shown by the open symbols, and the HCSD groups are in solid symbols. At first glance, these data appear to indicate that the HCSD cells have less degradation than the corresponding control cells. However, the average resistances for the HCSD cells are also lower at Characterization and RPT0 (i.e., the RPT immediately prior to first cycle set). This indicates the presence of manufacturing variability for the