Design of Multifunctional Structural Batteries with Health Monitoring Capabilities

Purim LADPLI, Raphael NARDARI, Fotis KOPSAFTOPOULOS, Yinan WANG, Fu-Kuo CHANG

Department of Aeronautics & Astronautics, Stanford University, Stanford CA 94305, United States
E-mail: {pladpli, rnardari, fkopsaf, yinanw, fkchang}@stanford.edu

Key words: Multifunctional materials, Energy storage, Structural battery, Distributed sensors, Battery health monitoring

Abstract
This work presents the development of the first-generation Multifunctional Energy Storage (MES) Composites - a multifunctional structural battery which embeds li-ion battery materials into high-strength composites together with in-situ networks of sensors and actuators. MES Composites not only can supply electrical power but also serve as structural elements, capable of concurrently carrying mechanical loads. In addition, the built-in sensor/actuator networks can monitor the state and health of both the composite structure as well as the battery on a real-time on-demand basis. As part of this study, MES Composite batteries are fabricated and then undergo a series of characterization tests to evaluate the synergistic energy-storage and load-carrying functionalities. Data generated from built-in sensors are also used to characterize the battery state of charge and health, in comparison with results from the electrochemical reference performance tests, and quantify any non-catastrophic degradation in the electrochemical performance. The obtained results verify the multifunctional capabilities of MES Composites for developing a novel minimum-weight and efficient energy storage system.

1 INTRODUCTION

Recently, more and more advanced high-performance energy storage technologies are being developed to meet the requirements of various mobile and vehicle applications, particularly when related to high-energy lithium-ion batteries. Current approaches for electric vehicle (EV) energy storage systems focus primarily on increasing cell-level energy density, in order to reduce the energy-to-weight ratio, extend the range and performance, and reduce the cost [1,2]. However, for state-of-the-art EVs, the system-level functional weight can be as much as twice the weight of the cells due to the extensive mechanical protection and enclosures, as well as the additional monitoring and sensing systems required to sustain the useful life of the batteries [1,2]. These disparate protective components reduce the packing factor and greatly decrease the system-level effective energy density. Moreover, the advantages of high-energy-density cells are also largely offset by the complexity and cost of the more demanding system-level engineering requirements.

As it stands, the vehicle-level energy density of current EVs, as well as their system cost, is still not at an economically viable level, and has been a barrier against a widespread adoption of EVs and electric systems. Thus, the electric vehicle and battery community has recently put forth a research thrust seeking novel multifunctional energy storage designs [2-5] (Figure 1).

in the Proceedings of the 8th European Workshop on Structural Health Monitoring (EWSHM 2016), 5-8 July 2016, Bilbao, Spain (www.ndt.net/app.EWSHM2016)
A multifunctional energy storage design can significantly improve the system-level specific energy by freeing-up the EV energy storage from the need for the protection and monitoring systems, and enabling additional functionalities thus removing component redundancy. In essence, such a multifunctional design of the energy storage system needs to be able to substitute the functionalities of the three components previously mentioned: 1) the energy storage itself; 2) supporting structures and mechanical protection enclosures; and 3) monitoring and sensing systems.

The concept of using energy storage materials concurrently as a structural element, liberating the need for extra mechanical protection, has been discussed in the literature [6-10]. However, these structural batteries take either of the extreme forms: one requiring unnecessarily major modification to the material composition, or a holistic approach which is merely repackaging of the commercial batteries [3,11,12]. Furthermore, to the best of our knowledge, in-situ integration of monitoring and sensing capability to energy storage materials, let alone to structural energy storage, is novel and has not been widely presented in the literature.

Batteries can only operate safely and reliably in a narrow envelop, thus demanding accurate state-estimation, control and management systems. Obtaining an accurate estimation of the battery state of charge (SoC) during real operation has always been a challenge to the battery community [13]. State-of-the-art techniques use traditional voltage and current measurements (and temperature in some cases), and from the data time-history, the cumulative energy consumption is calculated via state-estimation models. The numerical time integration, the dynamic nature of the battery usage, and the measurement noise often lead to large cumulative errors making it difficult to obtain a satisfactory accuracy in SoC estimation. Additionally, the remaining useful life of a battery, or the state of health (SoH), degrades over time. The model parameters for the state-estimation models have to be continuously updated during operation to reflect battery degradation, such as the impedance growth, but can only be approximated due to the lack of precise laboratory equipment on-board [14].

The fact that the charge and health state of an electrochemical cell is also physically coupled with the distribution and changes in the material properties, such as the density and modulus is well documented and studied [15]. Analogous to the prevailing concept employed by Structural Health Monitoring (SHM) [16-19], these material property changes induced electrochemically can also be treated as anomalies in comparison with a-priori known material
and structural properties and health states (baseline states). Hence, generalizing the concept to a multifunctional structural battery, both the structural and electrochemical states can be simultaneously determined, using in-situ distributed sensors that can sense the various underlying physical phenomena and external stimuli, in combination with advanced signal processing and system identification techniques.

Therefore, this work presents the development of Multifunctional Energy Storage (MES) Composites, a novel form of structural batteries with in-situ networks of sensors and actuators, capable of simultaneously storing energy, carrying mechanical loads, and providing real-time status on structural integrity and battery health. Some preliminary results focusing on the structural construction and the electrochemical-mechanical performance have been recently published by the authors [11,12]. However, this is the first time that the concept of combined structural and battery health monitoring for multifunctional structural energy storage is introduced.

2 PROBLEM STATEMENT AND METHOD OF APPROACH

To achieve the system-level energy density performance at an economically feasible level, a novel multifunctional design of the EV energy storage system is necessary. The design needs to combine functionalities of the three key components for a working electric system: 1) energy storage; 2) supporting structures and mechanical protection enclosures; 3) battery monitoring and sensing systems.

Hence, in this work, we present the development of a novel structural battery with integrated health monitoring capabilities (Multifunctional Energy Storage (MES) Composites), which integrates high-energy li-ion battery materials into high-strength composites together with in-situ networks of sensors and actuators (Figure 2). MES Composites can simultaneously store energy, serve as a structural element building-block, as well as have the capability of sensing and monitoring the structural and battery health on a real-time basis. This multifunctional energy storage building-block is highly scalable and can potentially deliver considerable weight and volume savings at the system level for various types of electric vehicles and electric systems, for instance, electric cars, aircraft, and space vehicles.

Figure 2: Multifunctional Energy Storage (MES) Composites concept - embedding li-ion battery materials inside high-strength carbon-fiber composites, together with in-situ networks of sensors and actuators.
The MES Composites’ novel architecture and multifunctionality are enabled by two important breakthroughs:

1. The unique **vertical material integration technique** allowing li-ion battery materials to be embedded into high-performance structural carbon-fiber composites while optimizing for electrochemical and mechanical performance. This aspect has been presented by the authors in [11,12], and will be described briefly herein.

2. **Enabling battery and structural health monitoring capabilities** through in-situ integration of sensor/actuator networks to structural batteries, allowing the collection of insightful data on structural integrity and battery condition and its use to predict the remaining useful life of the structural battery system (Figure 3). Special emphasis is given to this part in this study, showing preliminary feasibility results on using SHM-based ultrasonic inspection for battery health prediction, as well as providing a foundation for the structural and battery health monitoring framework for MES Composites.

### 3 VERTICAL MATERIAL INTEGRATION

State-of-the-art li-ion pouch cells comprise a stack of alternating anode and cathode layers, separated by thin micro-porous polymer separator membranes (Figure 4a). Li-ion cells are designed primarily to store energy, and as a result the mechanical load carrying capability and load transfer through the cells is minimal, if any [9]. The individual electrode layers are loose, therefore mechanical coupling is not present between the layers. Even minimal mechanical load exerted on a battery can thus cause excessive relative layer-slipping, resulting in battery degradation or even short-circuiting.

The material integration and functionalization concept for MES Composites has been introduced in [11,12] (Figure 4b). MES Composites encapsulate li-ion battery materials inside structural carbon-fiber-reinforced-polymers (CFRP) “facesheets”. The stiff structural CFRP facesheets are placed on both sides of the electrode stack, separated by the stack thickness, to carry the majority of the bending moment, similar to a sandwich structure.
However, without the interlayer shear resistance of the battery core, the thin battery layers will bend about their own individual neutral axis, and the structural contribution from the facesheets will be minimal. Therefore, MES Composites employ through-thickness polymer reinforcement pins, which extend through the perforations in the electrode stack, similar to the Lithylene technology [20]. The through-thickness reinforcement pins interlock the individual electrode layers, and mechanically link the two structural CFRP facesheets together (Figure 4b). The interlocking pins enable load transfer between the two facesheets and inhibit the relative slipping between the adjacent electrode layers, allowing the entire laminate to be able to bend about a common neutral axis. This approach significantly increases the stiffness and strength of MES Composites over traditional li-ion batteries.

As presented in [11, 12], despite the disruptive change in the cell construction compared to commercial pouch cells, minimal impact on the electrochemistry and electrochemical performance is achieved. Owing to the interlocking mechanisms, MES Composites show a significant increase in bending stiffness and no discernable degradation on the electrochemical performance upon applications of quasi-static and even cyclic fatigue loading.

4 ENABLING IN-SITU BATTERY AND STRUCTURAL HEALTH MONITORING

4.1 SHM and State-Awareness Demonstration of Sensor-Network-Integrated Systems

Recent progress in the SHM community has been towards maturing the real-time system-wide state-awareness by bridging the link between big data from sensors and algorithms for determining the health state and system operation [21]. The system reliability and robustness rely on the capability to deploy a large number of sensors of many different types over a large structural area, which had been technically prohibited until the recent breakthroughs.

The Structures and Composites Laboratory at Stanford University has demonstrated that the integration of distributed sensing systems with structures has tremendous potential in enabling condition-based maintenance based on SHM [16-19], which contributes to the enhanced vehicle safety and reliability. Distributed sensors enhance the SHM capabilities for damage detection, improving the reliability and accuracy of the structural diagnostics and prognostics.

Furthermore, the recent development of the micro-fabricated stretchable sensor networks
[22] allows a multitude of small-scale sensors, including all wiring, to be fabricated en-masse through standard micro-fabrication processes. After fabrication, these networks can be expanded to cover a structural area up to a few orders of magnitude larger than the initial fabrication real-estate. The minuscule footprint of the components allows the networks to be embedded into the host structure with minimal parasitic effects. Myriads of various sensor types tailored for SHM can be fabricated on to the sensor networks, including piezoelectric transducers which are widely used for ultrasonic damage detection, strain gauges for strain level measurement, temperature sensors for environmental condition monitoring, etc.

The introduction of the stretchable sensor network to autonomous systems [23-24] also has also shown significantly improved state awareness, allowing the system to sense the environment, operating condition and structural health state, and enabling the interpretation of sensing data to improve system performance and control characteristics.

4.2 Framework for Battery and Structural Health Monitoring of MES Composites Using Distributed Sensor Data

The stretchable sensor networks can be readily integrated to MES Composites and thus make the complete structural parts capable of generating data necessary for structural health estimation. Nevertheless, there is also a very advantageous opportunity to simultaneously use the same sensor data for determining the battery charge state as well as the battery health status.

In addition to the voltage and current data, the sensor networks can potentially provide rich supplementary information about the operating conditions and even the inherent physical changes in the cells during operation, which will be useful for battery SoC and SoH prediction and estimation.

- The distributed temperature sensors are used to measure the temperature distribution, based on which the state-estimation model parameters, such as the cell resistance and capacitance, can be updated. The temperature-induced acceleration in aging can also be estimated, even at localized areas of the cell, and used to improve the battery health prediction. The ability to measure the maximum temperature in real-time at discrete locations is also required for safety and prevention of thermal runaway.

- The strain gauges measure the stress levels in the cell due to external mechanical loads during operation, and therefore can be used to quantify the mechanically-induced degradation of the battery performance and preemptively detect non-critical degradation due to mechanical stress.

- The piezoelectric transducers are used in active ultrasonic-guided-wave-based methods to detect changes in the physical properties within the battery materials and correlate them with the SoC. The changes observed from charge/discharge (electrical) and loading cycles can also provide very useful information in determining mechanical degradation and the battery SoH.

Using ultrasonic information to correlate battery SoC and SoH is undoubtedly not trivial and is worth detailed investigation. Therefore, in this paper, emphasis is given to the results from a preliminary study in correlating ultrasonic signature with changes in battery materials, and potential applications for SoC and SoH determination. This will provide a foundation towards the construction of physics-assisted data-driven state-estimation algorithms for SoC and SoH. Coupled with the already established structural diagnostics and prognostics models, both the battery SoH and the structural integrity of MES Composites can be simultaneously determined using the shared distributed sensor data, as can be summarized by the framework presented in Figure 5.
4.3 Battery Charge and Health Monitoring with Ultrasonic Guided Waves Using Piezoelectric Transducers

While there have been recent efforts in correlating ultrasonic interrogation with battery SoC and SoH, such approaches rely on the bulk-wave-based method [25,26]. In those works, through-thickness transmitted and reflected waves are used to correlate the changes in materials with the charge state and mechanical degradation. However, their major drawback is the requirement of external bulky ultrasonic probes and equipment, extensive human intervention, and accurate baseline collection. The novelty of the currently presented approach lies in the introduction, experimental demonstration, and assessment of using permanently mounted thin piezoelectric disc transducers to generate ultrasonic guided waves for on-demand battery SoC and SoH estimation.

Similar to active-sensing guided-wave-based SHM techniques, the ultrasonic signals are collected in pitch-catch mode, where one transducer acts as an actuator generating the guided waves, and the arriving waves are measured at the other sensing transducers. Analogous to the guided-wave-based SHM, the piezoelectric signals can provide very useful information for estimating the remaining useful life of batteries. The signals collected from fresh batteries will serve as the baseline data, to which the data from subsequent charge/discharge cycles will be compared. Shifts in the guided-wave signal features as compared to the baseline, in combination with the rudimentary voltage and current information, can be used to estimate the battery SoH.

4.3.1 Variation in Guided Waves Signals with Battery State of Charge

Experimental Method

An experiment is performed on a 4000mAh MES Composite sample (160x110x5mm – the active battery stack is 90x90x3.5mm located in the center of the sample). Four 6.35mm-diameter disc piezoelectric transducers (PZT-5A) in the SMART Layer format (Acellent Technologies Inc.) are attached on the surface of the battery using Hysol E20HP structural epoxy adhesive, according to the schematics shown in Figure 6a.
The piezoelectric transducers are actuated with standard five-peak Hanning-windowed tone bursts, using a 64-channel ultrasonic data acquisition system (ScanGenie model; Acellent Technologies, Inc.). The peak-to-peak amplitude of the actuation signals is 75V. The center frequencies of the signals span between 100 to 200 kHz, and are selected in such a way to obtain clear wave packets in the sensor response. Measurements are taken every 1 minute for a total duration of approximately 12 hours.

Electrochemical cycling is performed at constant temperature (23°C) using an eight-channel battery analyzer (BST8-3, MTI Corporation). The cycling tests are done at a constant current rate of 800mA (approximately C/5), between 4.2V and 3.0V. The battery analyzer is time-synchronized with the ultrasonic data acquisition system.

Results and Discussion

Figure 6b shows representative piezoelectric sensor signals at two different SoC (at terminal voltage of 3.9V and 3.6V), obtained during the discharge phase. The signals shown are collected from the P1-P3 transducer pair with a center frequency of 150 kHz. The time of flight (ToF) and signal amplitude data are extracted from the signals throughout the cycle and plotted in Figure 6c. The signal amplitude is defined as the maximum amplitude of the signal envelope (Hilbert transform of the sensing signals). The ToF refers to the time between the maximum amplitude of the actuation (pitch) and response (catch) signals.

As the battery is charged, the signal amplitude slightly decreases, followed by a gradual increase until the cell potential of approximately 4.05V is reached. Near the end of the discharge, the signal amplitude decreases again (see Figure 6c). As the charge current is removed, the signal amplitude relaxes gradually to an equilibrium value. During the discharge process, the signal amplitude fluctuates slightly at the beginning of the discharge (<3.8V), followed by a gradual decrease except for the region near the end of discharge when the cell potential is less than 3.5V. This agrees with the findings reported in [25]. Signal attenuation is found to be generally decreasing during discharge and increasing during charge, with an exception regions near the beginning and end of the charge and discharge phases. During these two extreme regimes, it was hypothesized that the phase transformation in the cathode overshadows the global effect from the graphitic anode [25]. During charging, the ToF of the first wave packet generally decreases in a monotonic fashion. As the battery is discharged, the ToF increases monotonically until the minimum cut-off voltage.

It can be seen that there is a strong correlation between the signal features of ultrasonic guided waves and the battery SoC. Changes in the SoC of the battery are reflected in the changes in the density and elastic modulus of the anode and cathode materials, which in turn affect the behavior of the guided waves. Moreover, variations in the slopes of the signal amplitude and ToF versus time can be seen at different instances throughout the charge and discharge processes. Besides the effect from the cathode phase transition, these non-linearities are primarily induced by the intercalation staging in the graphitic anode.

However, particularly for li-ion batteries and MES Composites, the propagation substrate is mixed-media, with high anisotropy and inhomogeneity. The charge/discharge rate can also affect the rate at which the modulus and density change, as well as the acousto-elastic effect from the film stress developed from intercalation or phase change of the active materials. Therefore, an analytical relationship between the material property distribution, and the resulting signal attenuation and wave velocity cannot be derived in a trivial manner. Currently under investigation is the use of numerical techniques for guided wave propagation such as PESEA [27] to simulate the changes in waveforms due to the changes in material properties.
4.3.2 Deviation from Ultrasonic Baseline Signals in Aged Li-ion Batteries

Experimental Method

To demonstrate the feasibility of the proposed concept, an accelerated aging experiment is performed on production li-ion pouch batteries, where ultrasonic signals are collected and analyzed. The samples are 3650mAh li-ion pouch cells (135x45x5mm) – similarly, with four surface-mounted PZT-5A SMART layers (Figure 7a). The ultrasonic data collection is the same as previously described. The cells are cycled at an elevated temperature 45°C. The cycling is carried out at a higher current rate to accelerate the electrochemical degradation. The charge/discharge current is 3000mA (approximately 0.8C), between 4.2V and 2.75V. A constant-current-constant-voltage (CCCV) profile is used for the charging phase, with a cutoff current of 182.5mA (C/20).
Results and Discussion

The capacity retention with the number of charge/discharge cycles is shown in Figure 7b, with the values normalized with respect to the first cycle capacity. The accelerated capacity fading of the cell is approximately 0.25%/10 cycles. At Cycle 25 and Cycle 50, the capacity of the cell is approximately at 99.2% and 98.6% of the first cycle respectively.

The ToF and signal amplitude variation during Cycles 1, 25 and 50 is shown in Figure 7c. As can be seen, the cyclic ToF and signal amplitude of Cycles 25 deviate quite significantly from those of Cycle 1, which correspond to the baseline signals. The deviation at Cycle 50 is even more prominent, after the battery ages for another 25 cycles. As a general observation, the ToF overall appears to shift to a lower value as the battery ages, i.e. the wave speed is slower. This is accompanied by an overall higher signal amplitude as the electrical degradation progresses.
Such degradation-induced variation in the signal features provides very useful information for estimating the remaining useful life of the battery. As mentioned previously, the cyclic signature in the ToF and signal amplitude data is mainly due to the distribution and re-distribution of the material properties, mainly the modulus and density. Physical insights into the electrically-induced mechanical degradation can help build a physics-based relationship, which correlates the electrical degradation to the varying ultrasonic signature. Therefore, instead of relying on a pure data-driven model, one can establish a physics-assisted model that can be used in correlation with the machine-learning-based data-driven state-estimation algorithms for accurately predicting the battery SoC and SoH.

5 CONCLUSIONS

In this work, we have presented the concept and characterization results of first generation Multifunctional Energy Storage (MES) Composites and the pathway to incorporating in-situ sensing for battery and structural health monitoring applications. The preliminary results have illustrated the following key findings:

• The vertical material integration method allows MES Composites to concurrently store electrical energy and used as structural elements to carry mechanical loads in static and dynamic environments. The interlocking pins inhibit the deformation inside the li-ion battery electrodes, thus preventing degradation on electrochemical performance from mechanical loading.

• The feasibility of incorporating embeddable networks of micro-sensors/actuators into MES Composites for structural and battery health monitoring has been illustrated. The preliminary work explores the technical viability of using various sensors for sensing physical changes in the battery during electrical cycling. Guided-wave-based ultrasound techniques based on piezoelectric transducers have shown promising results in improving the accuracy of battery SoC estimation, and SoH identification.

Further work is ongoing in the areas of improving fabrication processes, design optimization, establishing prediction models based on sensor data, and incorporating the stretchable sensor network in MES Composites.

6 ACKNOWLEDGEMENT

The work was supported by the Advanced Research Projects Agency - Energy (U.S. Department of Energy) through the ARPA-E Award No. DE-AR0000393. The authors highly appreciate support from Farasis Energy Inc., particularly Dr. Keith Kepler, Dr. Hongjian Liu, and Dr. Michael Slater, for providing access to electrochemical testing facilities, and valuable feedback and suggestions throughout the fabrication process. The authors would also like to thank the Structures and Composites Lab’s graduate students Raunaq Rewari, Anthony Bombik, and Fabian Rothmaier for their significant contribution in sample preparation and testing.

REFERENCES


