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Parameter Improvement of Composite Sinusoidal Waveform Charging Strategy for Reviving Lithium-ion Batteries Capacity

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Keywords：lithium-ion battery, charging, composite sinusoidal waveform, capacity recovery state.

ABSTRACT

The current usage of lithium-ion batteries in many applications will cause environmental problems owing to the large number of batteries consumed. The composite sinusoidal waveform charging method can revive the capacity of aged batteries. However, the composite waveform parameters for charging each lithium-ion battery type can be different. In this study, a criterion for parameter adjustment is proposed. According to this criterion, aged batteries can recover their capacity and be reused.

INTRODUCTION

With the life conveniences brought about by the advancement of mobile device technology and growing global attention toward green energy, the demand for secondary batteries has dramatically increased. As the most important energy carrier, lithium-ion battery use continues to increase (Service, 2019; Zubi et al., 2018; Zeng et al., 2019).

In recent years, these batteries have been produced and consumed in large quantities for mobile devices, energy storage equipment, and electric vehicles.

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Therefore, with the significant increase in their global usage, battery repurposing is bound to be a difficult problem that needs to be addressed in the future (Gu et al., 2019; Liu et al., 2019; King and Boxall, 2019).

Many countries and large global companies regard lithium battery development as their primary strategic goal, owing to their advantages of higher capacity, longer service life, and shorter charging time. Most of these entities focus their research on electrode materials development (Pomerantseva et al., 2019), improvement of electrolyte formulation (Wu et al., 2019), and usage management conditions (Liu et al., 2017). However, less attention has been paid to the charging method (Monem et al., 2015; Li et al., 2020), even though a change in charging strategies can chemically reverse the problem of lithium battery aging caused by charge and discharge cycles.

The constant current–constant voltage (CC–CV) charging method has been widely used as a conventional charging method. At the end of the CC mode, the battery reaches full voltage and is switched to the CV mode. However, the charging current is very small, which increases the charging time. As a result, the CC–CV charging method cannot satisfy the user’s need for rapid charging. Furthermore, with this charging method, a solid electrolyte interphase (SEI) layer, which is a passivating film, can be easily produced on the negative electrode surface at the end of the CC mode (Birkl et al., 2017; Wang et al., 2018; An et al., 2016). This reduces the number of lithium ions involved in the battery charge/discharge cycle, resulting in battery capacity degradation. This is currently the primary reason for the limited lithium-ion battery life cycle and an obstacle to extensive applications (Barré et al.,2013; Palacin, 2018). Previously, Chen et al. (2013) proposed a sinusoidal waveform charging strategy for battery charging by conducting battery charging experiments at 1 Hz, 100 Hz, and 10 kHz, as well as at the minimum-ac-impedance frequency $(f_{R_{min}})$. The results showed that using $f_{R_{min}}$ for charging can shorten the cell charging time, reduce the cell temperature rise during charging, improve the charging efficiency, and extend the battery service life. In addition, a composite sinusoidal waveform charging strategy has been proposed, which can effectively solve the aging problem caused by lithium-ion battery charging (Chen et al., 2018; Chen et al., 2019). The key to this charging strategy is to introduce a negative voltage for discharging into the charging phase when the battery nears a full charge. The charging and discharging stages comprise two half-cycles of a sinusoidal waveform with different amplitudes, as shown in Figure 1.

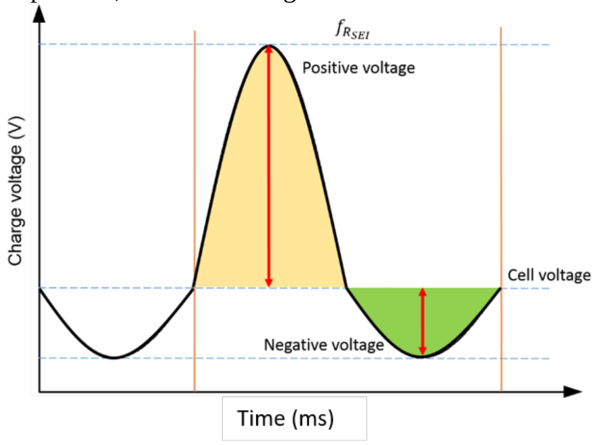


Fig. 1. Composite sinusoidal waveform for charging.

The waveform in the charging stage was designed according to the energy required for lithium ions to move at each charging process stage. The energy required for the process in which lithium ions move from the positive electrode to the negative electrode is estimated using first-principle calculations, and the energy required for each stage is estimated through a sinusoidal waveform, thus forming the positive half-cycle amplitude of the charging waveform. There are four composite sinusoidal waveform stages for charging. The first stage is the separation of lithium ions from the positive electrode, and the required energy is approximately 0.1–0.5 eV. The second stage is for lithium ions to pass through the SEI membrane, and the required energy is approximately 0.7–0.9 eV. The third stage is the energy required to insert lithium ions into the negative electrode, which is approximately 0.4–0.5 eV. The fourth stage is to disintegrate the lithium ions that have not completely passed through the SEI film during the charging process, and the required energy is approximately 0.1–0.2 eV. Based on the four-stage data, a preliminary charging waveform can be established and combined into a sine wave. This waveform will not cause the battery to have a momentary excessive pressure difference and can be charged with a smoother waveform. The minimum-ac-impedance frequency corresponds to the SEI layer impedance frequency ($f_{R_{SEI}}$) to deduce the time required by the lithium ions to move from the positive electrode to the negative electrode during charging. Most importantly, given the SEI layer produced by the lithium ions at the electrode–electrolyte interface, a discharge stage is specially designed to suppress the SEI layer formation to diminish the capacity decline (Chen et al., 2018). Experimental results show that the charging method can effectively extend the battery service life, decrease the temperature rise during charging, reduce internal battery impedance, and even recover an old battery’s capacity (Chen et al., 2019).

Although composite sinusoidal waveform charging has been proposed to suppress battery aging, parameter management of the composite sinusoidal waveform charging strategy is the key factor affecting the battery capacity recovery and charging efficiency. Parameter improvement for revising repurposing lithium ion batteries can solve the impending pollution problem caused by repurposing lithium ion batteries. Based on our previous research (Chen et al., 2018; Chen et al., 2019), this study focuses on further parameter adjustments of the abovementioned strategy. In particular, the key adjustment parameters include the positive and negative sinusoidal wave half-cycle amplitudes and charging frequencies. In addition, the parameters must be adjusted for different lithium-ion battery types. The composite sinusoidal waveform comprises two positive and negative sinusoidal waveform half-cycles. The positive half-cycle amplitude is chiefly designed based on the energy required for lithium ions to penetrate the SEI layer. However, as the positive half-cycle voltage is quite high for the battery, such a large potential difference introduces a very large charging current into the battery, causing a significant battery temperature increase. To avoid the damage caused by the temperature rise of the internal battery materials and to retain the energy required for the lithium ions to penetrate the SEI layer, we conducted experiments on batteries with a similar state of health (SOH) by adjusting the positive half-cycle amplitude. A back-electromotive force is specially introduced into the discharge end design of the negative half-cycle to reverse the unstable lithium ions from the complex, so as to reduce the SEI layer development and maintain the quantity of lithium ions involved in the cycle. Moreover, in the overall charging process, the negative half-cycle pertains to discharging, and its amplitude affects the full charging time. In this study, experiments were conducted on several batteries by adjusting the negative half-cycle amplitude. The charging time can be reduced if the negative voltage value can be reduced as much as possible. As for frequency, because the measured impedance frequencies of the SEI layer are different for different types of lithium-ion batteries, the charging frequency is adjusted according to the battery type. The charging method can be improved based on the battery charging time, degree of temperature rise, increase or decrease in impedance, and increase in the capacity to determine the better parameters of the composite waveform charging strategy for lithium batteries.

MATERIALS AND METHODS

The battery specifications and laboratory equipment

The experiment conducted in this study used a repurposed 40138 model lithium iron phosphate (LFP) battery from an electric vehicle (Phoenix Battery Co., Ltd., Hsinchu, Taiwan; denoted as PH) to improve the composite sinusoidal waveform parameters. Tests were also performed on an aged 18650 LFP battery (Pihsiang Energy Technology Co., Ltd., Hsinchu, Taiwan; denoted as BS) and a lithium polymer battery obtained from an iPhone 6s that had undergone normal use (Huapu Technology Inc., Changshu, China; denoted as Li-polymer). The specifications, battery models, and laboratory equipment used in this study are detailed in Table 1 to ensure repeatability by other research groups.

Table 1. Specific data for the tank vehicle.

Name (Model)	Specifications	
PH: 40138 model LFP battery	Typical Rated Capacity	10 Ah
	Nominal Operating Voltage	3.2 V
	Charging Voltage	3.65 V
	Cut-off Discharge Voltage	2.5 V
	Max Charge Current	50 A
BS: 18650 model LFP battery	Typical Rated Capacity	1.45 Ah
	Nominal Operating Voltage	3.2 V
	Charging Voltage	3.65 V
	Cut-off Discharge Voltage	2.5 V
	Max Charge Current	1.5 A
Li-polymer: Lithium polymer battery	Typical Rated Capacity	1.715 Ah
	Nominal Operating Voltage	3.82 V
	Charging Voltage	4.35 V
	Cut-off Discharge Voltage	2.7 V
	Max Charge Current	2 A
Charge/discharge machine 17208M-6-30, Chroma ATE Inc., Taiwan	Maximum Voltage/Current	6 V/30 A
	Voltage Accuracy	±0.015%
	Current Accuracy	±0.02%
Thermal data logger 51101-64, Chroma ATE Inc., Taiwan	Temperature Scale	ITS-90
	Temperature Accuracy	±(0.01% of reading + 0.5) °C
Signal generator/DC power supply, N6785A, Keysight Technologies Inc., USA	Voltage Range	0–40 V
	Voltage Accuracy	0.03% + 6 mV
	Current Max	25 A
	Current Accuracy	0.04% + 4 mA
	Maximum Frequency	10 kHz
Electrochemical test equipment VersaSTAT 3 Potentiostat Galvanostat, AMETEK.Inc., USA	Measurement Frequency Range	10 μHz–1 MHz
	Applied Voltage Range	±10 V
	Applied Current Range	±2 A
	Voltage Accuracy	±0.2% of value ±2 mV
	Current Accuracy	±0.2% of reading

Experimental parameters of the composite sinusoidal waveform

Because different composite sinusoidal waveform parameters are required for different lithium-ion battery types, an analysis was conducted using the charging frequency and amplitudes of the positive and negative half-cycles of the composite sinusoidal waveform charging strategy as the parameters. The experimental steps consisted of three parts.

Step 1: To ensure the battery internal chemical balance and stabilize the battery SOH value, the SOH of each battery was measured before conducting the charging experiment. The battery was first charged at a CC of 0.5 C until it reached full voltage; then, the voltage was maintained constant at CV (full voltage) until the current reached a 0.05 C-rate value. The battery was allowed to rest for 15 min after charging completion, and then discharged at a CC of 0.5 C until its discharge cut-off voltage was reached, after which it was allowed to rest again for 15 min. The charge/discharge cycle was repeated five times to ensure the internal battery chemistry reached equilibrium.

Step 2: After determining the SOH, the electrochemical impedance spectroscopy (EIS) of each battery at zero state of charge (SOC) and the corresponding $f_{R_{SEI}}$ were measured using the EIS test equipment.

Step 3: After the battery's $f_{R_{SEI}}$ was confirmed, several lithium-ion batteries with similar SOH were selected for each parameter adjustment in the experiment to ensure reproducibility. Initially, the battery was charged at CC until it reached full voltage, after which the battery was charged at CV until the current reached a 0.05 C-rate value. After resting for 15 min, the battery was discharged at CC until the battery voltage dropped to its discharge cut-off voltage. Simultaneously, the battery capacity using CC–CV charging was recorded. After resting the battery for 15 min, the battery was charged using the composite sinusoidal waveform charging strategy until the current was less than 0.05 C-rate. Then, the battery was rested for 15 min and discharged at CC until the battery voltage dropped to its discharge cut-off voltage. The battery temperature and charging time, as well as the battery capacity improvement, were recorded.

RESULTS AND DISCUSSIONS

Better parameters of Composite Sinusoidal Waveform in a PH Cell

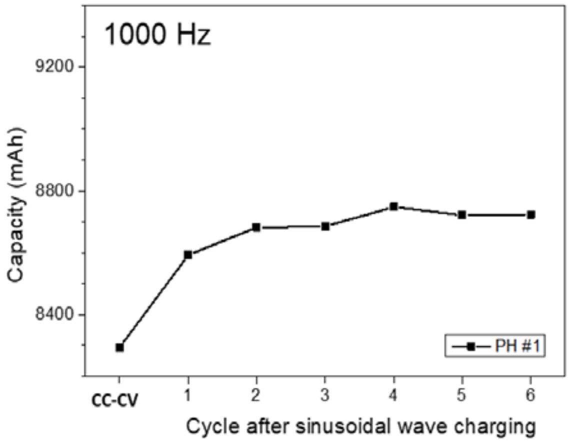
This experiment was conducted on a repurposed 40138 model LFP battery from an electric vehicle. Before performing the composite waveform charging on the battery, the internal chemical battery substances were stabilized. Following the two experimental process steps, the measurement frequency of the PH cell was set to 3000 – 0.01 Hz, at a current amplitude of 200 mA and $f_{R_{SEI}}$= 500 Hz. The electrochemical impedance spectra of the PH cell are shown in Figure 2. In a previous literature, a sinusoidal charging strategy is proposed that the minimum-ac-impedance frequency of Li-ion battery is the optimal charging frequency (Chen et al., 2018). The minimum-ac-impedance frequency is attributed to Li-ion migration time during charging. Experimental demonstrations had revealed improvements of the battery's lifetime, charging efficiency, maximum rising temperature compared with that of the conventional charging methods. However, the correlation between charging frequency and ion migration is still a hypothesis, thus there will be more evidence to discuss this correlation in the next article. After the $f_{R_{SEI}}$ of the battery was confirmed, several batteries with similar SOHs were selected for Step 3 of the experiment.

Fig. 2. PH cell electrochemical impedance spectroscopy.

Frequency adjustment of composite sinusoidal waveform

The composite sinusoidal waveform charging strategy with different charging frequencies of 1000 Hz and 500 Hz ($f_{R_{SEI}}$) was used to compare batteries with similar SOH in this study. Figure 3(a) shows that the power released using the composite waveform charging strategy at a frequency of 1000 Hz was ~5% (500 mAh) higher than that released using the CC–CV charging strategy. Moreover, the battery capacity stability could still be maintained after multiple battery charging cycles using the former strategy. As shown in Figure 3(b), when the charging frequency of the composite waveform charging strategy was set at $f_{R_{SEI}}$, the capacity was increased by ~3% (300 mAh) compared to that of a battery originally charged at 1 kHz, and there was even a difference of ~8% (800 mAh) compared with the CC–CV charging strategy.

(a)



(b)

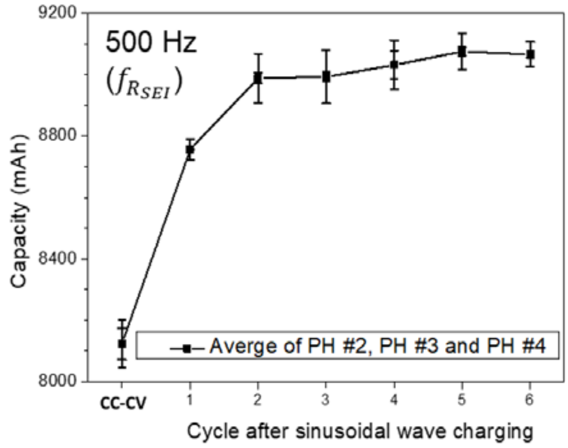


Fig. 3. Increase in the PH cell capacity charged at (a)1000 Hz and (b) 500 Hz ($f_{R_{SEI}}$).

Moreover, such differences are not coincidental. After multiple charge/discharge cycles, the battery capacity remained stable. The two charging frequencies had different effects on the recovering power capacity because the SEI layer impedance frequencies were different owing to changes in the battery SEI layer structure as the battery usage time increased. Note that the composite sinusoidal wave frequency is preferably in line with the $f_{R_{SEI}}$ and cannot be arbitrarily changed. Otherwise, the lithium ions cannot move from the positive electrode to the negative electrode using the charging strategy. In such a case, it would not be possible to recover the battery power or fully charge the battery.

Amplitude adjustments in the negative half-cycle

The key function in designing the negative half-cycle is to break the unstable lithium complex.

According to theoretical calculations, the required amplitude is only 0.2–0.05 V (Chen et al., 2019), which may have a direct impact on the battery charging time and the power that can potentially be recovered. This section shows a comparison between the battery capacities with an amplitude of 0.2 V and adjusted amplitudes of 0.15, 0.1, and 0.05 V, to determine the most negative half-cycle amplitude.

Table 2 shows that adjusting the negative half-cycle amplitude did not significantly help increase the battery capacity. At first glance, the 0.05 V amplitude was the best of the three. However, it was found in subsequent experiments that this effect originating from the original waveform has already increased the battery capacity. The 0.1 V amplitude did not have a similar phenomenon. Thus, it can be inferred that 0.1 V is a critical value for increasing the battery capacity in the negative half-cycle. In addition, Figure 5 shows that the charging time can be minimized when using a 0.05 V negative half-cycle amplitude.

Table 2. Comparison of amplitude adjustments in the negative half-cycle.

Battery Number	Negative Peak Amplitude (V)	Capacity (mAh)	Negative Peak Amplitude (V)	Capacity (mAh)
PH #5	0.2	9409.84	0.15	9397.39
PH #6		9488.88	0.1	9495.88
PH #7		9503.57	0.05	9459.23

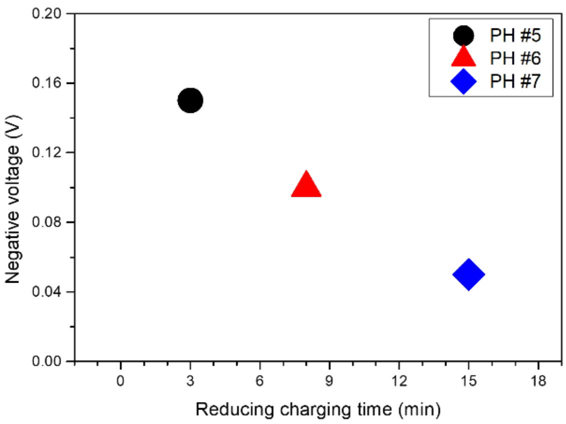


Fig. 4. Effects of different negative half-cycle amplitudes on reducing charging time.

This is because the use of a smaller amplitude reduced the discharging portion during the charging process, resulting in a shorter overall charging time. Combined with the above experimental results, 0.1 V is recommended as the most negative half-cycle amplitude for the composite waveform charging strategy.

Amplitude adjustments in the positive half-cycle

The amplitude of the positive half-cycle of the composite waveform charging strategy was estimated to fall in the range of 0.6–0.9 V, based on theoretical calculations (Chen et al., 2019). This section shows the experiments conducted on batteries using positive peak amplitudes of 0.6, 0.7, 0.8, and 0.9 V, to obtain the most positive half-cycle amplitude. As shown in Table 3, the change in the positive half-cycle amplitude varied the battery capacities. It had a significant impact on the battery charging time and degree of temperature rise. While the charging time was reduced with positive peak amplitudes of 0.8 and 0.9 V, the cell temperature rise was 10 °C or higher during the process, as shown in Figure 5.

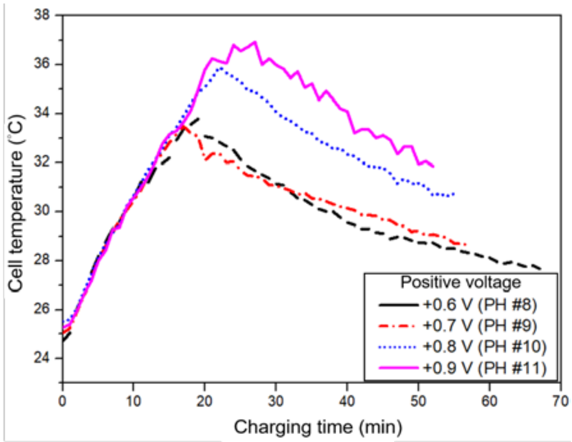


Fig. 5. Effects of different positive half-cycle amplitudes on temperature rise.

Table 3. Amplitude adjustments in the positive half-cycle.

Battery Number	CC–CV Charging Strategy		Composite Waveform Charging Strategy				
	Power Released (mAh)	Maximum Temperature Rise (Celsius)	Positive Peak Amplitude (V)	Power Released (mAh)	Power Increased (%)	Maximum Temperature Rise (Celsius)	Charging Time(min)
PH #8	8573.39	12	0.6	9147.9	5.7	7	67
PH #9	8512.53	13	0.7	9106.4	5.9	7	57
PH #10	8602.97	13	0.8	9198.6	6.0	10	55
PH #11	8612.11	14	0.9	9223.7	6.1	11	52

Although a temperature rise of 10 °C appears normal in a single battery, it will cause considerable concern for user safety if applied to a battery pack or system. The charging time with a positive peak amplitude of 0.7 V was ~5 min longer than that with 0.8 and 0.9 V amplitudes. The temperature rise was ~7 °C, which is consistent with the rise with a 0.6 V amplitude. While the temperature rise was comparatively low (~7 °C) when the positive peak amplitude was 0.6 V, the overall charging time was extended by nearly 10 min compared to the time with other amplitudes. Combining the experimental results in the previous sections and this section, it can be concluded that the battery capacity increase with the composite waveform charging strategy was largely influenced by the positive half-cycle amplitude and was comparatively independent of that of the negative half-cycle. However, adjusting the positive half-cycle amplitude directly affects the charging time and temperature rise of the battery. Therefore, in this study, 0.7 V was considered the most positive half-cycle amplitude for the composite waveform charging strategy.

Battery Diversity Test in BS cell

In this section, diversity tests were performed on the 18650 model LFP battery, which has been widely used in electric vehicles, to confirm whether the better parameters of PH cell in the composite waveform charging strategy have the same effects on LFP batteries with different dimensions. To simulate the usage conditions in an electric vehicle, charging and discharging were performed at a high C-rate. Following the two experiment process steps, the BS cell measurement frequency was set to 2000–0.01 Hz, at a current amplitude of 200 mA and $f_{R_{SEI}}$= 1000 Hz. The BS cell electrochemical impedance spectra are shown in Figure 6. After the battery $f_{R_{SEI}}$ was confirmed, two batteries with similar SOHs were selected for Step 3 of the experiment. The battery was charged using a composite waveform where the positive and negative half-cycle amplitudes were 0.7 and 0.1 V, respectively. The experimental results are shown in Figure 7.

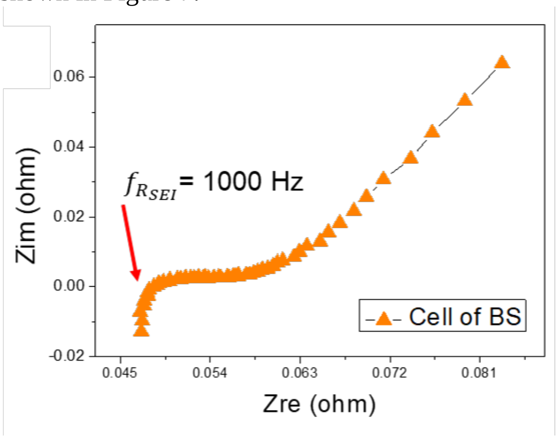


Fig. 6. BS cell electrochemical impedance spectroscopy.

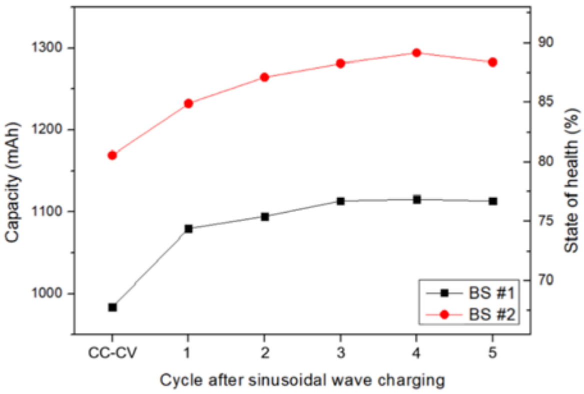


Fig. 7. Increase in the BS cell capacity.

It was found that although the experiment was conducted on batteries with different SOHs, the charging method could still increase the capacity of aged batteries by ~10%. Therefore, it can be deduced that the battery dimensions do not influence the composite waveform charging strategy performance. Hence, this new waveform for reviving batteries can be used in different sized lithium batteries. Moreover, the 18650 LFP battery has a wide range of uses, from 3C products to electric vehicles, thus enlarging the scope of impact and increasing this study's new waveform charging strategy contributions.

Battery Diversity Test in Li-polymer cell

In today's modern society, it is common for every person to have a smartphone. The cumulative number of iPhones sold to date has already reached 1.2 billion. We selected the most representative iPhone battery for the charging waveform diversity test. In this test, the current battery SOH was first measured using the conventional CC–CV charging strategy. Following the two experiment process steps, the measurement frequency of the Li-polymer cell was set to 10000–0.01 Hz, at a current amplitude of 200 mA and $f_{R_{SEI}}$= 8000 Hz. The Li-polymer cell electrochemical impedance spectroscopy is shown in Figure 8. After the $f_{R_{SEI}}$ of the battery was confirmed, two batteries with similar SOHs were selected for Step 3 of the experiment.

As shown in Figure 9, a new waveform charging strategy was used to revive the iPhone batteries. A reviving effect was observed for both batteries. One of them had its power recovered from ~846.7 mAh to 1205.2 mAh, which is a nearly 20% increase. Currently, the user demand for mobile phone battery power has increased. Considering that mobile phones are going to enter 5G networks, the battery discharge demand has also increased. The experiment conducted in this study can effectively increase the battery power and maintain its revival level, which has a significant impact and value to the industry.

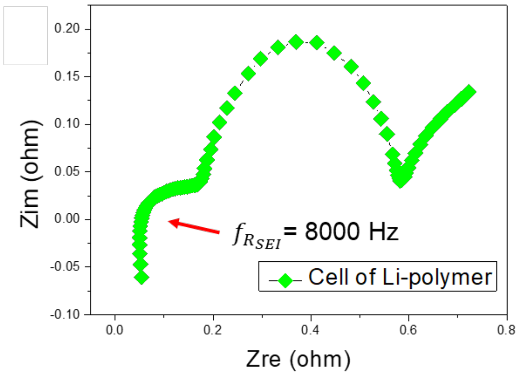


Fig. 8. Li-polymer cell electrochemical impedance spectroscopy.

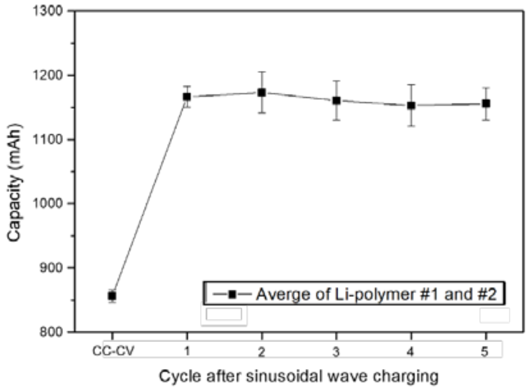


Fig. 9. Increase in Li-polymer cell capacity.

CONCLUSIONS

This study developed a charging waveform that has a greater impact on battery charging by adjusting various composite sinusoidal waveform parameters based on battery charging time, degree of temperature rise, increase or decrease in impedance, and capacity increase. The most positive and negative half-cycle amplitudes of the composite sinusoidal waveform were recommended to be 0.7 and 0.1 V, respectively. Note that the composite sinusoidal wave frequency should preferably be in line with the $f_{R_{SEI}}$. Otherwise, the lithium ions will not transfer from the positive electrode to the negative electrode under the charging strategy.

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恢復鋰離子電池容量的複合正弦波充電策略參數改進

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摘 要

由於生活中需要儲電的產品需求持續攀升，老化與廢棄的鋰離子電池將會對地球環境帶來極嚴重的衝擊。複合正弦波波形充電方法提供一種恢復舊電池容量的可能性。然而，適合各類型鋰離子電池所需充電的複合波形的參數並不相同。本研究提出了一種調整參數的準則。根據該標準，老化的電池可以恢復其容量並可以重複使用。