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Experimental Study on Series-Parallel Grouping Inconsistency of Zinc–Nickel Single Flow Batteries

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This study experimentally investigated the effects of the charge/discharge current, initial state of charge *(SOC)*, internal resistance, and initial nominal capacity on the performance characteristics and efficiency of series-parallel battery to address the inconsistency problems of zinc–nickel single flow battery during the use of battery pack energy storage system. Experimental results show that the differences in charge/discharge current, initial *SOC*, and initial capacity change the inconsistency of battery pack. The difference in initial nominal capacity is the most influential factor, whereas the charge/discharge current is the least influential factor. Meanwhile, the inconsistency of internal resistance increases, which enlarges the energy loss of the battery pack and increases the temperature. The optimum operating current is below 30 A and the optimum deviation in initial *SOC* is within 10%, at which the inconsistency of the zinc–nickel single flow battery pack and the balance of the branch current are satisfactory.

Keywords: zinc-nickel single flow battery; series-parallel batteries; inconsistency; charge/discharge characteristics

1. INTRODUCTION

Battery energy storage systems are important in the present era of rapid growth of renewable energy sources. Zinc–nickel single flow battery (ZNB) exhibits outstanding merits, such as simple structure, long cycle life, high energy conversion efficiency, low cost of materials, flexible power increase or decrease, and no ion cross contamination; this material has potential application in energy storage of solar/wind energy systems, large emergency power supply system, and smart grid[1-5]. A large-scale energy storage system can improve the voltage and capacity by series-parallel connection of battery stack. Therefore, the difference in the initial performance of the single-cell stack and the inconsistency of the circulatory state increases the inconsistency of the series/parallel battery pack, and this difference is directly related to the life and reliability of battery pack[6]. Studying the consistency of the battery stack and predicting its performance parameters effectively during the ZNB energy storage system are common tasks in large-scale demonstration stage[7-9].

At present, research on the inconsistency of energy storage battery is always the key and difficult point. As early as in 1995, Peters[10] studied the battery voltage inconsistency of lead–acid batteries. The results showed that this factor seriously decreases the effectiveness of the battery pack. Conte et al.[11] analyzed the inconsistency in the internal resistance of lead–acid batteries on the basis of experiments on the actual running of a car. Shi et al.[12] verified that the unbalance current causes the capacity attenuation of the parallel battery group through modeling and experimental analysis of the equivalent circuit of large-capacity $LiFePO_4$ batteries. Chen et al.[13] proposed that the resistance difference is the main reason for the difference in the voltage of cell stack. The charging/discharging experiments were performed on 200 lithium batteries, and the equivalent model was established in accordance with the battery pack. Wang et al.[14] pointed out that the trend of voltage inconsistency in battery pack conforms to normal distribution on the basis of the statistical analysis of the power storage battery group for electric vehicle. Fan et al.[15] experimented on cycle life of lithium single-cell and battery pack. The single-cell model, which considers the degree of aging, was established and extended to the modeling for the inconsistencies of the battery pack. Guo et al.[16] explored the effect of battery voltage inconsistency on capacity attenuation for lithium battery energy storage system. He et al.[17] analyzed the influence of inconsistent factors such as open circuit voltage, internal resistance, and capacity of lithium ion battery during overcharging and underdischarging in series and hybrid battery pack by experiment. Obviously, study on the inconsistency of the storage battery pack is mainly focused on lead–acid batteries and lithium batteries, while research on ZNB is still blank.

In this study, the effects of the charge/discharge current, initial state of charge *SOC*, internal resistance, and initial nominal capacity on the performance characteristics of series-parallel battery are investigated. The experimental data of charging/discharging test for battery are compared, and the reasons for the inconsistency of single cell, which influences the characteristics and efficiency of series/parallel battery pack, are analyzed. As a result, a reference for sorting strategy, control system design, security management and system parameters, and performance prediction of ZNB energy storage system is provided.

2. WORKING PRINICIPLE OF ZNB

ZNB use nickel oxide electrode as positive electrode, use alkaline saturated zinc-acid salt solution as electrolyte, and use inert conductor as negative electrode. During the charging process and discharging process, the electrolyte flows through between the stack and the tank reservoir by the pump (see Fig. 1)

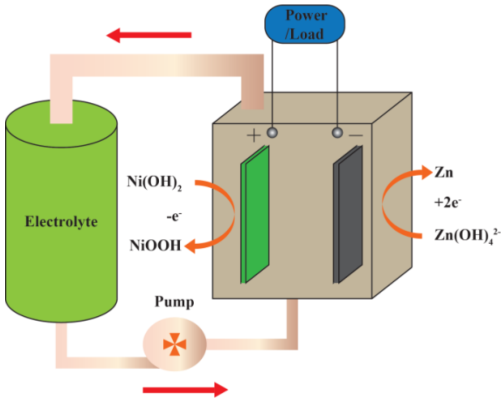

Figure 1. Working principle diagram of ZNB

The positive electrode reaction [18] is:

$$2NiOOH + 2H_2O + 2e^- \underset{charge}{\overset{discharge}{\rightleftarrows}} 2Ni(OH)_2 + 2OH^-, \varphi = 0.49V \qquad (1)$$

The negative electrode reaction is:

$$Zn + 4OH^- \underset{charge}{\overset{discharge}{\rightleftarrows}} Zn(OH)_4^{2-} + 2e^-, \varphi = -1.215V \qquad (2)$$

Thus, the total reaction is:

$$Zn + 2NiOOH + 2H_2O + 2OH^- \underset{charge}{\overset{discharge}{\rightleftarrows}} 2Ni(OH)_2 + Zn(OH)_4^{2-}, \varphi = 1.705V \qquad (3)$$

In the battery, two simultaneous reactions occur on the surface of both positive electrode and negative electrode. $NiOOH$ is changed to $Ni(OH)_2$ at positive and the zinc is oxidized to zincate on the negative electrode substrate when discharging. The reverse process occurs when charging.

3. EXPERIMENTAL

3.1 Experimental setup

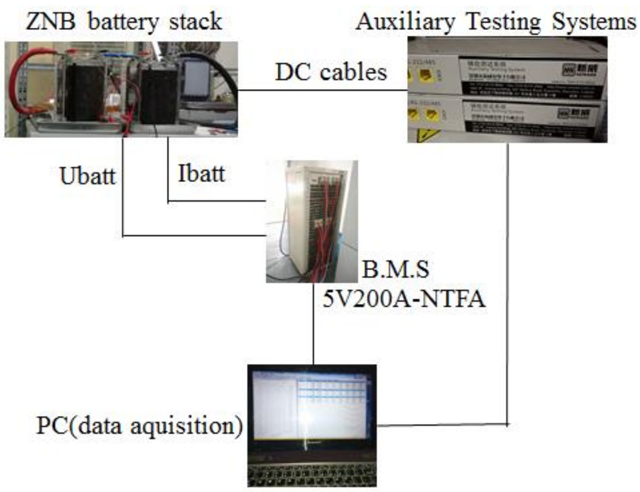

Figure 2. Principle of the experimental test device

The experimental object is a domestic company production of the second and third generation of ZNB, which use nickel oxide electrode as positive electrode, alkaline saturated zinc–acid salt solution as electrolyte, and inert conductor as negative electrode. During charging and discharging, the electrolyte flows between the stack and the tank reservoir by using the pump[19]. The principle of experimental test is shown in Fig. 2.

The basic parameters of a single cell are shown in Table 1.

Table 1. Basic parameters of the experimental object

Parameters	#1, #2, #3, #4, #5	#6
Maximum cut-off voltage (V)	2.05	2.05
Minimum cut-off voltage (V)	1.2	1.2
Rated voltage (V)	1.6	1.6
Rated capacity (Ah)	216	300
electrode pair	19	23
Temperature (°C)	−40 to 40	−40 to 40

The experimental equipment mainly includes the BTS7.5.X battery testing system, auxiliary testing system, 5V200A-NTFA detector produced by Shenzhen Xinwei company, and tong-type ammeter.

3.2 Experimental design

Table 2. Experimental design and steps

Group	Cell	Series			Parallel		
		Steps	Cut-off voltage	Rated capacity	Steps	Cut-off voltage	Rated capacity
A	#1 and #2	Constant current charge/discharge test at 30, 35, 40, 50, and 100 A	Maximum 4.1 V, Minimum 2.4 V	216 Ah	Constant current charge test at 30, 50, and 100 A	Maximum 2.05 V, minimum 1.2 V	432 Ah
B	#3 and #4	Charge the initial *SOC* to 20% and 30%, 20% and 50%, and 20% and 70% with constant current charge test at 50 A	Maximum 4.1 V, Minimum 2.4 V	162 Ah, 143 Ah, 111 Ah	Charge the initial *SOC* to 40% and 50%, 40% and 60%, and 40% and 80% with constant current charge test at 50 A	Maximum 2.05 V, minimum 1.2 V	238 Ah, 216 Ah, 172 Ah
C	#1 and #2	(1) 100A current charging for 2 minutes; (2) 5 min holding time; (3) Repeat step (1) and (2) until the battery reaches the cutoff voltage; (4)	Maximum 4.1 V Minimum 2.4 V	216 Ah	Constant current charging test at 50 A	Maximum 2.05 V, minimum 1.2 V	432 Ah

		discharge process similar to HPPC					
D	#5 and #6	Constant current charging test at 50 A	Maximum 4.1 V, Minimum 2.4 V	216 Ah	Constant current charging test at 100 A	Maximum 2.05 V, minimum 1.2 V	516 Ah

For the ZNB energy storage battery group under the long-term charge/discharge process, the differences in charge acceptance capacity, self-discharge rate, and capacity decay rate for the single cell increase the difference in the *SOC* of each cell. In particular, a divergence trend is observed, which expands the inconsistency of the battery group[20]. The effects are divided into several situations, such as short board effect, overcharge and overdischarge, performance degradation, and temperature[21]. To explore the influence factors of battery inconsistency, from the charge/discharge current, *SOC*, internal resistance, and initial nominal capacity, the charge/discharge experiments and the comparative analysis of the battery pack are carried out. The inconsistency of the battery pack is judged from two aspects of the open circuit voltage and the branch current.

To effectively evaluate the performance difference and external characteristics of the battery pack, the experiments and steps, as shown in Table 2, are designed.

On the basis of the above-mentioned experimental steps, the battery terminal voltage data of the series battery pack are collected through the auxiliary test system at 25 °C, and the current change of branch circuit is recorded with the ammeter.

4. RESULTS AND DISCUSSION

4.1 Different charge/discharge currents

Figure. 3 shows the experimental test results of the series batteries A. Although the charge and discharge conditions of the #1 and #2 are the same, the initial state of the #1 and #2 is as uniform as possible before the start of the experiment. However, in the actual charging and discharging process, due to the inconsistent degree of battery aging, the state of charge will vary with the progress of charging and discharging, and the magnitude of this difference is closely related to the current. The overall trend of the stack voltage of the each single cell in the ZNB series battery under different charge/discharge currents is similar. In the constant current charge/discharge of 30 A, the solid-line #1 and the dashed-line #2 present a good consistency in voltage variation. Within the safe cut-off voltage, different degrees of divergence are found at the beginning and end of charge/discharge, and the divergence increases with time.

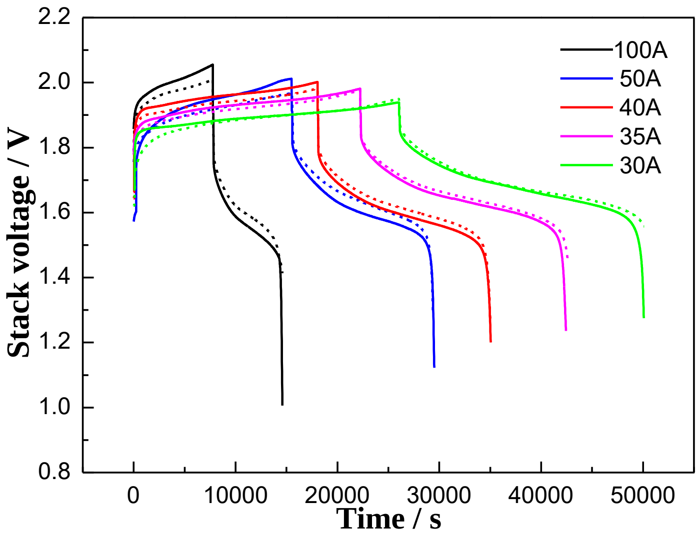

Figure 3. Voltage curve of each single cell in series batteries with different charge/discharge currents

In the cases of 35, 40, and 50 A, obvious inconsistencies are found in the two single cells. During the initial charging process, the voltage of #1 is gradually higher than that of #2. During the discharge process, the voltage of #1 is lower than that of #2, and #1 exhibits high maximum termination voltage and low minimum termination voltage. In the case of 100 A, the characteristics of inconsistency are more obvious. At this time, the maximum charge voltage and the minimum discharge voltage reach 2.06 and 1.01 V, respectively, thereby causing overcharge and overdischarge of #1. Table 3 shows the variation in termination voltage of each single cell in the series batteries with different charge/discharge currents.

Table 3. Termination voltage of single cell in series batteries

Cell	30 A		35 A		40 A		50 A		100 A	
	U_{max}/ V	U_{min}/ V	U_{max}/ V	U_{min}/V	U_{max}/ V	U_{min}/V	U_{max}/ V	U_{min}/ V	U_{max}/ V	U_{min}/ V
#1	1.950	1.464	1.9757	1.4465	1.982	1.2641	2.012	1.280	2.055	1.412
#2	1.901	1.275	1.981	1.236	2.002	1.2003	1.967	1.123	2.010	1.006

Table 3 shows that, as the charge/discharge current enlarges, the voltage difference between the cells in the non-platform period increases. The highest termination voltage and the lowest termination voltage of #1 gradually increase, and the lowest termination voltage of #2 gradually decreases. The experimental results show that, as the charge/discharge current increases, voltage inconsistency occurs in advance, and the impact on battery inconsistency increases. In accordance with the literature experimental conclusion, large charge/discharge current indicates decrease in voltage efficiency and obvious energy efficiency of the battery[22]. Several experiments show that the current value should not be too large. Thus, the optimum operating current is below 30 A, at which the inconsistency and energy efficiency of the series battery are satisfactory.

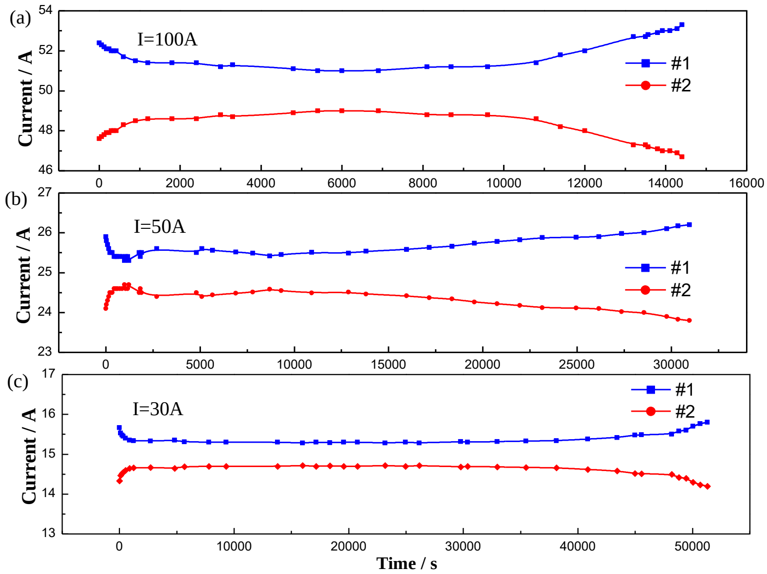

Figure 4. Current curve of each single cell in parallel batteries with different charge currents

Figure. 4 shows the variation in the imbalanced currents of the single cell in parallel battery A. Although the initial state of #1 and #2 was kept as consistent as possible before the start of the experiment, the internal resistance must be different due to factors such as battery aging. During the charging process, the current of #1 and #2 must be different due to the difference in the initial resistance, which will bring about the difference in the state of charge of #1 and #2, and further exacerbate the inconsistency of the internal resistance. Under the control of the battery test system, the total current of the two branches in Figure. 4(a) is kept at 100 A during the charge process, and the current value of #1 is greater than that of #2. As charging progresses, the current difference between the two batteries decreases first and then gradually increases. The reason is that the inconsistent internal resistance of single cells leads to differences in operating current distribution. Among them, the current value of #1 slowly decreases first and then slowly increases. The value reaches 53.3 A at the end of charging, which deviates by 6.6% from the predicted current. Figures. 4(b) and 4(c) show the experimental curves for 50 and 30 A, respectively. The difference in the absolute current value of the two batteries is smaller than that at 100 A, and the branch current distribution gradually stabilizes. As the charging current increases, the imbalance current of ZNB branch gradually increases. Compared with the LiFePO4 battery, a reverse deviation in the branch current is observed after one crossing [12]; however, no crossover occurs in ZNB. One reason may be that the impact of connectors on the branch current distribution loss and additional connector resistance is ignored. ZNBs are accompanied by side reactions at the end of charge/discharge, which is counteracted with the accumulation of *SOC*.

4.2 Different initial SOCs

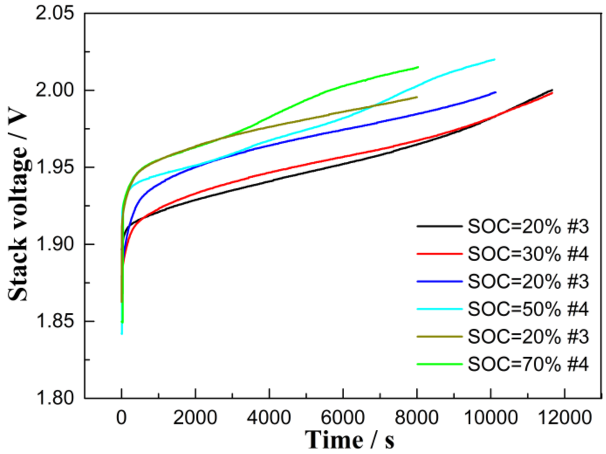

Figure 5. Voltage curve of each single cell in series batteries with different initial *SOC*s

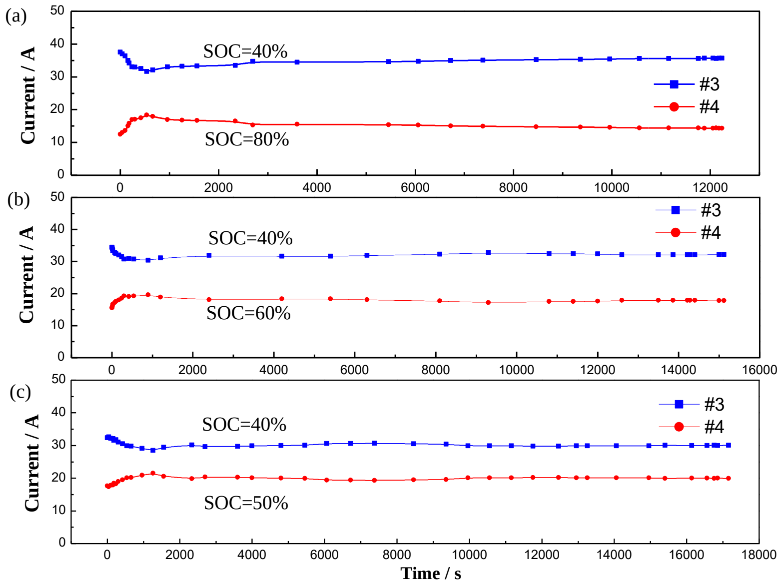

Figure 6. Current curve of each single cell in parallel batteries with different initial *SOC*s

Figure. 5 shows the experimental test results of the series batteries B with different initial *SOC*s. In the charging process of two batteries, the stack voltage increases with time. Large total *SOC* of the battery group indicates short time to reach the cut-off voltage or full state. The initial *SOC*s are 20% and 30%, and the *SOC* is relatively close. The terminal voltage is uniform, and the deviation is small. The initial *SOC*s are 20% and 50%. As the difference in *SOC* increases, the operating state deviation of the two batteries rises. The battery with large initial *SOC* is filled first, whereas the battery with small initial *SOC* is not fully charged. As the charging progresses, the fully charged battery continues to charge, and the terminal voltage rises suddenly. As shown in Figure. 5, voltage protrusion occurs later in the charging platform, thereby causing the battery to overcharge. The experimental observation shows that, as the battery continues to overcharge, the dendrite growth between the plates at the end of the charging period becomes progressively severe. When the number of cycles reaches more than 50 times, a short circuit occurs between the positive and negative plates of a single cell in the stack.

Figure. 6 shows the variation in the imbalanced currents of the single cell in parallel battery B. As shown in Figure. 6(a), the difference in the branch current compared with the initial charge gradually decreases, and the platform period of the charging process stabilizes. The battery current of #3 is large, whereas that of #4 is small. Thus, an imbalanced current value of approximately 18 A exists. The reason is that a difference in the initial inherent voltage of the cell is found due to the different initial *SOC*s. To maintain the same voltage in parallel circuit, the current of the battery with an initial *SOC* of 40% is always high. Figures. 6(b) and 6(c) show that, as the difference in initial *SOC* decreases, the imbalance current in the platform period gradually decreases by nearly 13 and 9 A. Thus, the parallel battery pack is extremely vulnerable to the accumulation of imbalanced current owing to the different parameters of the single cell. The cumulative difference in *SOC* between two batteries increases with time. Even if a small circulating current is generated, achieving *SOC* self-balancing is difficult. The instability of system operation caused by large difference in initial *SOC* should be avoided during the use of series-parallel battery. The experimental results show that the deviation in the initial *SOC* of the battery does not exceed 10%, and the consistency of the ZNB battery pack and the balance of the branch current are satisfactory.

4.3 Different internal resistances

From the curves of charge-discharge and holding time of zinc-nickel single-flow battery, it can be seen that the terminal voltage has a jump at the moment of loading current and withdrawing current, which is similar to the voltage response when current is loaded on a pure resistance. That is, the ohmic internal resistance R_0 in the corresponding model.

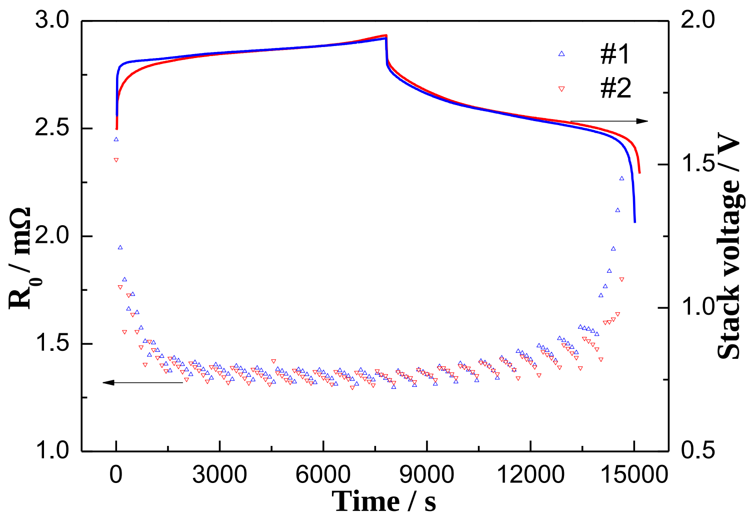

Figure 7. Curve of each single cell in series batteries with different internal resistances

When the battery is stationary for 5 minutes after discharge, after a sudden change in the battery voltage, the slow change will gradually stabilize. That is the polarization resistance R_{act} in the corresponding model. Therefore, by using parameter identification equation to analyze the change of pulse charge-discharge voltage response curve, we can identify the change characteristics of internal resistance of zinc-nickel single-flow battery with operating state. The specific method can be referred to our group's literature [19]. In reference [19], the process of parameter identification is described in detail and the analytical formulas of each parameters of the model are obtained by fitting. There are many specific contents, so we will not start the narration in this article. Therefore, analyzing the changes in pulse response curve can identify the variation characteristics of ZNB internal resistance [23], such as the scatter curve in Figure. 7.

The performance of series battery is related to the voltage and internal resistance of a single cell, and the current passing through the series circuit is the same. The voltage rise or drop during charge/discharge of a single cell is proportional to its internal resistance. As shown in Figure. 7, the internal resistance of single cell increases dramatically at the beginning and end of the charge/discharge. The reason is that the side reaction of the electrode causes the electrolyte transport resistance to increase in the electrode porous material, thereby increasing the polarization resistance of the battery[24]. The side reaction is oxygen evolution reaction of positive electrode. The internal resistance of the charging/discharging platform is stable, whereas the internal resistance of #1 is slightly higher than that of #2. The smooth curve in Figure. 7 is the single voltage curve of the series battery. After the 1500s and 13000s, the internal resistance greatly increases, which is due to the conductivity of cathode activity substantial being reduced when the power is low[25,26]. As time progresses, the inconsistencies in the internal resistance of the single cells increase, thereby resulting in significantly different battery voltages.

During charging and discharging, the energy released by the battery is $W=IR^2$, and the current I in the series group is the same. In the case of $R_1 \neq R_2$, internal resistance of large batteries occurs, energy loss is large, large heat is generated, and the temperature rises quickly. As the number of cycles increases, the difference in the resistance and the temperature of the shell increase and the discharge capacity gradually attenuates.

Figure. 4(b) shows the variation in the imbalanced currents of the single cell in parallel battery C, which is in the constant current charging of 50 A. Equations (4)–(6) show that the current distribution among the single cells in the parallel battery pack is affected by several factors.

$$U_1 = U_2 = U, I_1 + I_2 = I \quad (4)$$

$$I_1 = \frac{U_{oc2} - U_{oc1}}{R_1 + R_2} + \frac{R_2}{R_1 + R_2} I \quad (5)$$

$$I_2 = \frac{U_{oc1} - U_{oc2}}{R_1 + R_2} + \frac{R_1}{R_1 + R_2} I \quad (6)$$

Where U_1 and U_2 are the stack voltages of the single cell, U_{oc1} and U_{oc2} are the open circuit voltages, $I_1$and I_2 are the currents, and R_1 and R_2 are the resistances. One part of the branch current is formed by the difference in open circuit voltage, and the other part is determined by the current allocated by the internal resistance. The change in branch current is affected by the internal resistance of branch battery, and the internal resistance of battery changes with the change in the *SOC* of battery.

4.4 Different initial capacities

Battery capacity generally refers to the maximum available capacity of battery, that is, the amount of battery discharged under full condition. This parameter is important in measuring the performance of the battery. One part of the single cells is overcharged and overdischarged, thereby resulting in performance failure or capacity attenuation during the long-term use of the battery pack. Considering the echelon use of the old batteries, the difference in battery capacity is important in measuring the imbalance of voltage.

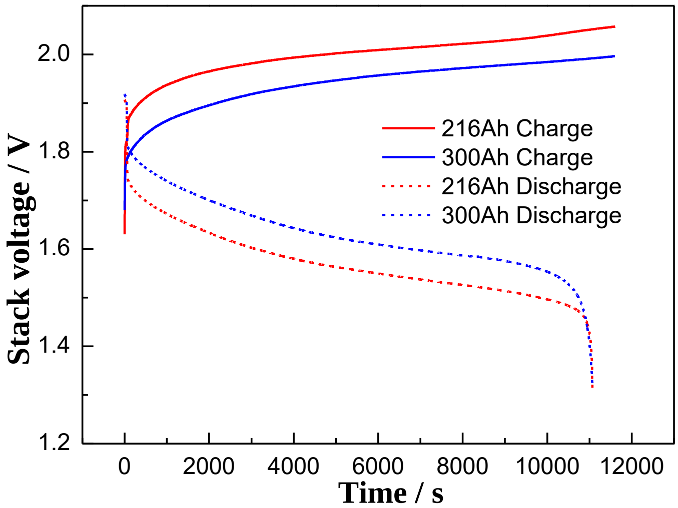

Figure 8. Voltage curve of each single cell in series batteries with different initial nominal capacities

Figure. 8 shows the experimental test results of the series batteries D with different initial nominal capacities. The battery D is charged at a constant current of 50 A to a capacity of 216 Ah. As shown in Figure. 8, large nominal capacity of the single cell indicates low voltage during the platform. The reason is that, in the end of the charging process, the battery with a small capacity is gradually filled, the battery with a large capacity is only charged to 72%, and the depth of charge/discharge of #6 fails to achieve the optimum efficiency. In the discharge process, the battery with low capacity has a lower state of charge than the battery with high capacity. When the power of a low-capacity battery is completely discharged, the battery with high capacity still has a certain amount of electricity, and the low-capacity battery becomes the load in the circuit. Under the same test conditions, the two batteries have different charging and discharging depths due to the difference of the initial nominal capacity, which causes the cell voltage inconsistency to always exist. From the above analysis, it can be seen that low-capacity batteries will enter a vicious cycle of repeated overcharge/over-discharge during charging and discharging, which will lead to premature damage and accelerate the decay of normal batteries, thus affecting the performance and service life of the whole battery system.

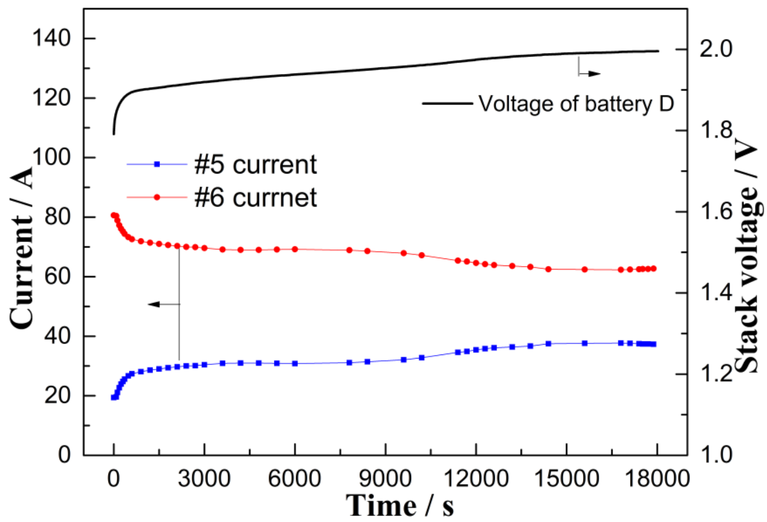

Figure 9. Curve of each single cell in parallel batteries with different initial nominal capacities.

Figure. 9 shows the voltage of parallel battery D and the variation in the imbalanced currents of the single cell in parallel battery D with different initial nominal capacities. As shown in Figure. 9, a large difference exists in the branch current in the early stage of charging. As charging progresses, the current value of #6 increases, whereas that of #5 decreases. The difference between the two branch currents narrows and balances, but a gap of 10 A still appears. The reason is that the internal resistance of the battery with large initial nominal capacity is large owing to the charging/discharging capabilities of the battery. Therefore, in the large-scale use of ZNB, batteries of the same type should be used in groups to avoid large differences in nominal capacity.

5. CONCLUSIONS

In this study, the effects of the charge/discharge current, initial *SOC*, internal resistance, and initial nominal capacity on the performance characteristics and efficiency of series-parallel battery are experimentally investigated. The conclusions are obtained as follows:

(1) As the discharge current increases, the voltage inconsistency of ZNB and the unbalance degree of parallel battery current increase. Under the cumulative cycle, the battery capacity attenuation occurs early. As the charge/discharge current increases, the voltage inconsistency of ZNB and the imbalance degree of parallel battery current increase. Under the accumulation of the cyclic process, the battery capacity attenuation occurs early. The optimum current value of series-parallel battery pack is below 30 A, at which the efficiency and inconsistency of the battery group are satisfactory.

(2) As the difference in initial *SOC* of the single cell increases, the inconsistency of the voltage increases gradually, and the deviation in the branch current also rises. When the optimum deviation in initial *SOC* is within 10%, the consistency of the ZNB pack and the balance of the branch current are satisfactory.

(3) The inconsistency in internal resistance greatly influences the series and parallel batteries. This parameter is not static, and it changes with the change in *SOC* of battery. At the beginning and end of charging and discharging, the difference in internal resistance of individual cells is obvious. As the difference increases, the energy loss enlarges and the temperature increases.

(4) The difference in initial nominal capacity leads to different charge/discharge depths of battery. The battery with low capacity enters a vicious cycle early, and it decays and damages in advance. The same type of battery is used to avoid large difference in initial nominal capacity.

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