



Failure mechanism of the lithium ion battery during nail penetration

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ABSTRACT

Nail penetration is one of the most important methods to study the internal short circuit safety of lithium ion batteries (LIBs). A series of penetration tests on LIBs under different conditions are conducted. The effects of the states of charge (SOC), penetration positions, depths and speeds are analyzed. As for different penetration positions, thermal runaway reaction is more severe when the battery is penetrated at center due to the faster propagation of thermal runaway. The battery surface temperature is not positively correlated with penetration depth, and the temperature distribution becomes more nonuniform with the increasing of penetration speed. All batteries get into thermal runaway if their temperatures exceed 233 °C due to the shrinkage of separator and trigger of reaction between cathode and electrolyte. The fire behavior of penetrated batteries is exhibited in this work. “Micro short-circuit cell” structure is proposed to interpret the mechanism of internal short circuit induced by penetration.

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1. Introduction

Lithium ion batteries (LIBs) are widely used in consumer electronics products and electric vehicles for their excellent advantages, such as high working voltage, large specific capacity, long cycle life and friendly to environments [1–6]. However, fire and explosion accidents caused by LIB thermal runaway are also reported frequently, which lead to catastrophic damage to lives and property [7–9]. Many thermal management methods, such as on-and-off air cooling [10], carbon fiber composites with 2D microvascular networks [11], nanofluids [12], phase change material (PCM) [13] and heat pipe [14] are used to decrease the temperature of single batteries or packs in order to guarantee safety. In order to investigate the failure mechanism of LIBs, many test methods, such as penetration, mechanical loadings [15–19], external short circuit [20], overcharge [21] and external heating test [22] were employed. In addition, different kinds of calorimeters were used to study the thermal property of battery materials and full batteries at elevated temperature [8,23–27].

Among the abuse conditions of LIBs, internal short circuit is one of the most critical failure modes [19] where a current path develops within the battery. It can be caused by penetration, mechanical loads [15–19], separator failure or contaminants [28]. Safety testing laboratories employ very low indentation speeds to induce

internal short circuit, and the researchers in UL employed speeds of 0.01–0.1 mm s⁻¹ [19]. The surface temperature at the point near the indentation site, distance moved by the indenter, applied force of the indenter, and the open circuit voltage were measured in their tests. They also advanced the lithium ion battery standards and investigated the aging effects on lithium ion battery [19]. On the other hand, Dubaniewicz et al. [18] used a plastic wedge with different speeds to crush various batteries (different cathodes, different geometries) within a 20-L explosion-containment chamber filled with CH₄-air to see if the thermal runaway of battery could ignite the flammable gases or not, so as to simulate a potential CH₄ ignition hazard of underground mine. Some researchers set up a micro mechanical model [17] to investigate the sequence of internal short circuit failures induced by tension, compression or both. The internal configuration of prismatic LIBs after pinching was exhibited by Wang et al. [16], showing the extensive internal faulting which might break the separators and cause internal short circuit. Some novel devices [29,30] are designed to be embedded in the battery to form different kinds of internal short circuits, i.e. anode-cathode short, electrode-current collector short and collector-collector short, respectively. Generally, the critical component of the novel devices is an electrolyte-compatible PCM. When the battery is heated to above the melting temperature of PCM, PCM melts and a current path is created between positive and negative sides. In Ref. [29], a 25 μm diameter copper puck is used as the short circuit trigger.

Compared to the above methods to study internal short circuit, penetration tests are often conducted to simulate the insertion of

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foreign objects into battery body during service [31]. There have been various experiments studying the penetration of LIBs. Some specific short modes, such as aluminum-anode and cathode-anode short, were compared with nail penetration [30]. Kim et al. [32] conducted nail penetration tests on lithium polymer batteries with ceramic-coated separator, and proposed three different failure modes during nail penetration. In addition to experiments, simulation is also a functional method in the study of penetration. Chiu et al. [2] suggested a numerical electrochemical model to simulate penetration test, and the simulation of temperature variation matched test results well. Zhao et al. [33] constructed a 3D model to investigate the nail penetration process, and proposed a method to calculate the resistance and current during internal short circuit. Apart from temperature, some other important parameters involved in penetration, like thermal runaway time, temperature distribution and active material loss, could also be estimated by simulation as reported in Ref. [34]. Besides single cells, thermal runaway propagation process within battery modules induced by penetration was also investigated [35–38].

In addition to temperature variations, fire behavior is also a very important feature to characterize the property of flammable materials. Meanwhile, many fire precautions are taken based on it. The fire behavior of LIBs caused by external heating, like radiative heating, is investigated recently [7,9,22,39–41], but the fire behavior induced by nail penetration is rarely reported to the best knowledge of authors. In this work, we will exhibit the fire behavior of LIBs during penetration. The crush or nail penetration speeds identified in the literature vary significantly, from 0.01 mm s^{-1} to 100 cm s^{-1} , and the effects of the penetration speeds proposed by different researchers are also different [18]. Ichimura [42] suggested that lower penetration speed can produce more adverse results. In this study, a higher penetration speed was employed. On the other hand, some researchers have investigated the effects of penetration positions, speeds and property of nails by modeling [31,43,44], while experiments are rarely applied to investigate the influences of penetration depths and positions. In this work, penetration experiments were performed to show the effects of SOC, penetration positions, depths and speeds. And the inner structure of internal short circuit within jelly-rolls was presented by micro short-circuit cell model and “string of candied haws” structure. Finally, the failure mechanism of LIBs during penetration was proposed.

2. Materials and methods

2.1. Battery samples

Battery samples used in this study were 18650-type batteries, which were 18 mm in diameter and 65 mm in length. The cathode

material contains 98% $\text{Li}(\text{Ni}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3})\text{O}_2$ and 2% LiMn_2O_4 . And the anode is natural graphite. The solvent of the LiPF_6 -based electrolyte is the mixture of ethylene carbonate (EC), ethyl methyl carbonate (EMC) and dimethyl carbonate (DMC). The separator of the cells in this study is a ceramic-coated separator and $16.3 \mu\text{m}$ in thickness, and the main material of the ceramic layer is Al_2O_3 . The morphology of the separator and the X-ray diffractometry (XRD) pattern of the separator’s burning residue are shown in Section 3.5. The battery sample had a nominal capacity of 2000 mA h and a nominal voltage of 3.7 V. A battery cycler (Neware BTS-6V10 mA, Shenzhen) was used to charge test batteries to desired SOC. The chosen SOC were 0, 50, 75 and 100% in this study.

2.2. Apparatus and experimental procedures

Penetration tests were conducted using the battery penetration machine (Beier Experiment Apparatus Co., Ltd.), which looked like a big fire-proof chamber with a battery fixed tool and a stainless-steel nail. There are three alternative diameters for the nail, i.e. 3, 5 and 8 mm, respectively, which are all bigger than the ISC device reported in Ref. [29] (Cu puck $25.4 \mu\text{m}$). And the battery in this study is only 18 mm in diameter, so the diameter of the nail was chosen as 3 mm to simulating the insertion of objects from outside.

As shown in Fig. 1, the cylindrical battery was mounted horizontally, and the nail penetrated it vertically along the diameter. As shown in Table 1, different SOC batteries (0%, 50%, 75% and 100%) were penetrated in Test 1–4 so as to investigate the effects of SOC. In addition, three alternative penetration positions, namely center (P2), close to the negative pole (P1) and close to positive pole (P3), represented by three red arrows in Fig. 1, were chosen to study the influences of penetration positions in Test 4–6. The distance between the neighboring penetration sites is 16.25 mm. With respect to the impacts of penetration depths, lengths of the nail inserting into batteries were adjusted to 6, 9, 12 mm and pierced in Test 7–9 and 4. The number of the shorted electrode layers changes with the penetration depth.

The speed range of the nail is $10\text{--}40 \text{ mm s}^{-1}$ for our penetration machine, which is higher than the low indentation speeds reported in Ref. [18,19]. On the other hand, the purpose of this penetration test is simulating the external objects inserting into LIB during the high-speed transportation or real accidents, so the penetration speeds should be larger. In order to find the effects of penetration speeds, the speeds were changed from 20 to 40 mm s^{-1} in Test 10–11 due to our test purpose and limitation of apparatus. The temperature variations of the battery were recorded by 5 K-type thermocouples (1 mm in diameter) represented by five yellow cross-marks as shown in Fig. 1. The distance between the neighboring thermocouples is 16.25 mm. And the fire behavior was monitored by a video camera.

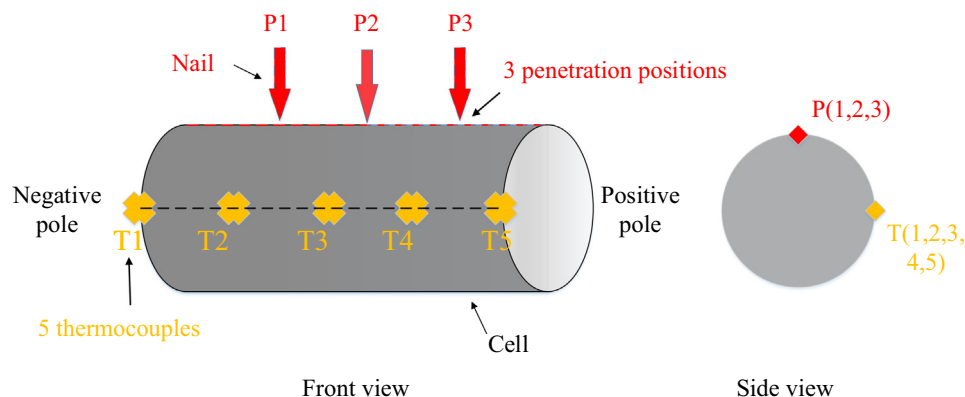


Fig. 1. Schematic diagram of penetration tests and thermocouple setup.

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