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Performance study of large capacity industrial lead-carbon battery for energy storage

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ABSTRACT

Electrochemical energy storage is a vital component of the renewable energy power generating system, and it helps to build a low-carbon society. The lead-carbon battery is an improved lead-acid battery that incorporates carbon into the negative plate. It compensates for the drawback of lead-acid batteries' inability to handle instantaneous high current charging, and it has the benefits of high safety, high-cost performance, and sustainable development. The recycling efficiency of lead-carbon batteries is 98 %, and the recycling process complies with all environmental and other standards. Deep discharge capability is also required for the lead-carbon battery for energy storage, although the depth of discharge has a significant impact on the lead-carbon battery's positive plate failure. This study optimizes and enhances the lead-carbon battery's positive plate, allowing it to perform both high-current charging (340.255 A) and deep discharge (70 % DOD) operations. Selecting acceptable lead alloys, improving the structure of the positive grid, and regulating the grid's curing and drying processes are all part of the optimization and improvement process. The upgraded lead-carbon battery has a cycle life of 7680 times, which is 93.5 % longer than the unimproved lead-carbon battery under the same conditions. The large-capacity (200 Ah) industrial lead-carbon batteries manufactured in this paper is a dependable and cost-effective energy storage option.

1. Introduction

Renewable energy is quickly gaining traction throughout the world as a vital part of achieving a low-carbon future [1–3]. Renewable energy generation, such as solar and wind, is intermittent and volatile, which may cause grid damage [4–7]. Grids can benefit from energy storage technologies to ease this issue. Energy storage is essential for load balancing, peak management, frequency regulation, damping energy oscillations, and improving power quality and reliability [8].

Electrochemical energy storage is one of the most popular technologies in the world because of its compact size, fast dynamic response, flexible transportation and deployment, scalability, ideal supporting equipment, and mature technology [9–12]. It is one of the most widely used energy storage technologies. Lithium-ion batteries, liquid flow batteries, sodium-sulfur batteries, nickel-hydrogen batteries, lead-acid batteries, and other electrochemical energy storage methods are often used. The lead-acid battery is the most affordable secondary battery, has a wide range of applications, and is safe [13]. The most crucial factor to remember is that lead-acid batteries are virtually entirely recyclable. Under the criteria of long-term growth, which is crucial. Every industrialized country has a well-organized closed-loop system for recycling lead, which includes battery manufacturing, battery recovery, recycling, and manufacturing new batteries from recycled materials. With almost 95 % of end-of-life batteries recycled in Western countries, lead has the greatest end-of-life recycling rate of any regularly used metal. Users do not have to pay for recycling since lead-acid batteries are recycled so effectively that about 98 % of the material may be recycled, and the recycling process is entirely consistent with environmental and other requirements [14]. Although there are collecting and recycling operations for lithium-ion batteries, there are extra costs connected with disposal, which are not cost-effective; there is presently no or the very limited large-scale collection and recycling infrastructure for other batteries. As a result, lead-acid batteries provide a dependable and cost-effective energy storage option [15–20].

Because of the high relative atomic mass of lead (207), which is one of the densest natural products, lead-acid batteries have low specific energy (Wh /kg). Lead-acid batteries' low specific energy costs some flexibility, but this isn't a problem for energy storage systems that

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prioritize cheap cost, high dependability, and safety.

There are two problems with the negative plate of lead-acid batteries used in energy storage systems. The first is that the negative plate cannot handle instantaneous high current charging. The second is that when the battery is running in a partial state of charge, the negative plate undergoes rapid sulfuration, resulting in shorter battery life. To reduce the workload of the negative plate in the battery, we introduce carbon into the negative plate of the lead-acid battery, which is called a lead-carbon battery. There are three different batteries available right now [21]. In the first type, 0.2–5 % mass fraction of carbon is added to the negative lead paste, and the carbon is used as an additive or component of the negative electrode or substance. Half of the negative plate of the second kind is made of a regular sponge lead plate, while the other half is constructed of a carbon supercapacitor, also known as a supercell. The third type of lead-carbon battery has a carbon supercapacitor in place of the typical lead negative plate.

Replacing all or part of the negative lead plate with a carbon plate is not workable [21]. Only the double electric layer on the carbon surface is charged during the charging process. The charging capacity is limited and depends on the surface area of the carbon electrodes. Furthermore, the first lead-carbon battery's manufacture is extremely similar to that of existing lead-acid batteries, which are easily industrialized. As a result, the first type of lead-carbon battery is the focus of this study.

2. Lead-carbon battery optimization

Lead-carbon batteries work similarly to conventional lead-acid batteries, with PbO_2 as the positive active material, spongy lead as the negative active material, and dilute sulfuric acid as the electrolyte. The overall reaction equation of lead-carbon battery discharge is:

$$Pb + PbO_2 + 2H_2SO_4 = 2PbSO_4 + 2H_2O \tag{1}$$

The overall reaction equation of lead-carbon battery charge is

$$2PbSO_4 + 2H_2O = Pb + PbO_2 + 2H_2SO_4 \tag{2}$$

On the negative plate of a lead-carbon battery, the electrochemical reaction occurs not only on the surface of lead but also on the surface of carbon [22]. Two electrical systems operate in the negative active material: a capacitive system, which includes an electrical double layer for charge-discharge reactions, which primarily occurred on the carbon surface; and an electrochemical system, which includes electrochemical and chemical reactions that oxidize lead to lead sulfate and then reduce it to lead.

The following are the functions of carbon additions and their impact on performing lead-acid batteries [23–27]:

The carbon improves the negative active material's conductivity.

Carbon suppresses the sulfation of the negative plate by limiting the development of lead sulfate crystals.

Carbon particles with a large surface area have a super capacitive effect on negative active materials.

Carbon can make the negative active material more porous.

Carbon can increase the diffusion and fast migration of electrolyte ions in the negative plate during high-speed charge and discharge as an electroosmotic pump.

Impurities in some carbons obstruct the hydrogen evolution process, improving charge efficiency.

The negative plate carbon material of a lead-carbon battery is usually used with activated carbon and another carbon material [28], with the activated carbon increasing the reaction surface area and the other carbon material boosting the conductivity. Activated carbon and carbon nanotube are used as negative additives in this study.

The negative plate's performance was increased by adding carbon, while the positive plate's performance remained unchanged. If high-current charge-discharge and deep-discharge cycles are conducted, the positive active material of the lead-carbon battery may soften and flake off, resulting in positive plate failure and a decrease in battery cycle life. As a result, we change the positive plate.

To begin with, boosting the positive plate's conductivity allows it to handle greater currents. The conductivity of the positive plate can also be enhanced by adding carbon. However, since the carbon fibers oxidize, the increased performance is rapidly lost. As a result, this is not the best option. The grid is the conductor of the active material. Through the grid, the active material's power is equally input and output. By increasing the amount of lead alloy in the positive grid, the current density of the grid may be reduced and the current acceptance capacity increased. As a response, the positive grid is thickened and the number of ribs on the positive grid is increased (Fig. 1). The negative grid of the lead-carbon battery has a thickness of 2.8 mm and a mass of 150.7 g in this study, whereas the positive grid has a thickness of 4 mm and a mass of 302.0 g, which is almost twice that of the negative grid.

The depth of discharge is a crucial functioning parameter of the lead-carbon battery for energy storage, and it has a significant impact on the lead-carbon battery's positive plate failure [29]. The deep discharge will exacerbate the corrosion of the positive grid, resulting in poor bonding between the grid and the active material, which will cause the active material to fall off in lumps. Severe shedding can cause a short circuit, that can be fatal [30].

This study proposes three optimization points to overcome this issue. To begin, use a corrosion-resistant lead alloy like Pb-Ca-Sn. Although the Pb-Ca alloy has strong corrosion resistance, which can improve grid creep resistance and deep cycle performance, Pb-Ca alloys used as positive grids have premature capacity loss [31–33]. The addition of Sn to the Pb-Ca alloy inhibits thermal and storage passivation [34]. In addition, Al is added to the lead alloy to prevent oxidation of the Ca in the Pb-Ca-Sn alloy during grid casting.

Second, deep discharge causes the active material to expand and contract more, putting more stress on the corrosion layer and grid, resulting in deformation and even fracture, which has a detrimental effect on the battery's performance. Although the lead-calcium alloy grid has high mechanical strength, it will distort when thoroughly charged and discharged with a big current. A tiny tab is added to the bottom of the positive (negative) grid (the top right corner of the grid in Fig. 1) in this work, and the newly added tab has a diagonal structure with the positive (negative) terminal. To prevent permanent grid distortion, a deformation space is prepared for the grid on another diagonal line in this manner.

Finally, an appropriate plate curing procedure is devised. The grid alloy undergoes an oxidation reaction during curing, resulting in a corrosion layer that is tightly coupled with the solid lead paste. The thickness of the lead paste applied to the positive grid grows as the grid thickness increases, resulting in a decrease in the oxygen diffusion rate

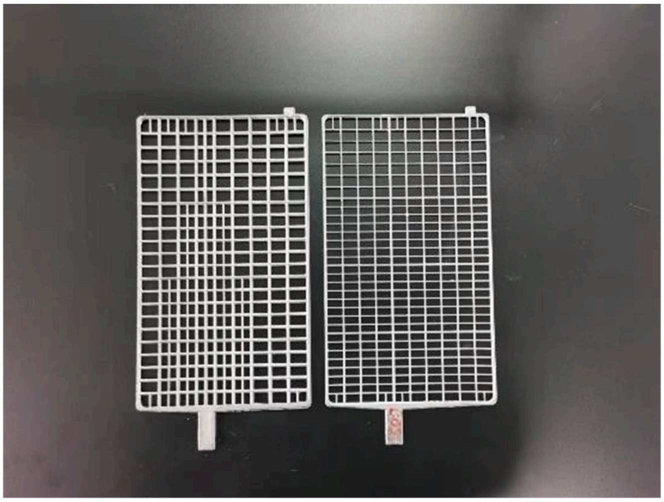

Fig. 1. Positive grid (left) and negative grid (right) of a lead-carbon battery.

in the lead paste and a weakening of the corrosion layer, making the original curing and drying process ineffective. Because the coupling of the Pb-Ca alloy grid and the active material is more difficult than that of the Pb-Sb alloy, the enhanced positive grid's curing and drying method must be changed.

After a lot of tests, the curing and drying process suitable for the positive grid in this paper was eventually determined, as shown in Table 1. The solidified and dried positive and negative plates (as shown in Fig. 2) have regular shapes, flat surfaces, no cracks, and high hardness. The plates are free to fall from a height of 0.8 m, and there are no cracks in the lead paste. This shows that the plates produced by the new method are of high quality.

3. Lead-carbon battery performance testing

The lead-carbon battery produced has a rated capacity of 200 Ah (charge/discharge rate and capacity decay are calculated using this capacity), a rated discharge current of 20 A, a rated charging current of 100 A, the rated working temperature is 25 °C, the rated working voltage is 2.0 V, the charging saturation voltage is 2.45 V, the discharge cut-off voltage is 1.8 V, the rated depth of discharge is 70 %, and the initial mass is 14,440 g.

Because the cell formation rate is around 85 %, the rate will progressively grow to 100 % as the number of uses increases. As a result, the true capacity of the battery is greater than the rated capacity. Fully charge a battery with a 100 % formation rate before discharging it at a constant current of 20 A. The discharge is halted when the battery voltage drops to the cut-off voltage. The real capacity is the quantity of power released during this time. In four steps, the lead-carbon battery is fully charged: First, discharge the battery at constant current (CC) using the rated current until the voltage drops to the cut-off value. Second, charge the battery with the rated current until the voltage reaches 2.45 V. Third, charge the battery with half of the rated current until the voltage reaches 2.45 V. Finally, charge for 3 h at 2.45 V constant voltage (CV). According to the foregoing technique, the real capacity is 239.904 Ah/482.509 Wh, with a specific energy of around 33.41 Wh/kg.

3.1. Discharging test

At a constant temperature of 25 °C, a high current discharge test was performed on a lead-carbon battery with a current range of 20 A-200 A.

When performing a high-current discharge test, first fully charge the battery, then discharge at a constant current until the battery voltage drops to the cut-off value. The cut-off voltage is critical for the battery management system's architecture. The high current CC discharge cut-off voltage refers to the voltage at which the electrolyte density decreases to 1.08 g/cm^3, which is often evaluated by testing, as illustrated in Fig. 4.

Fig. 2. Positive (left) and negative (right) plate after curing and drying.

The greater the discharge current, the lower the battery voltage, and the smaller the constant current discharge capacity of the battery (Fig. 3). The lead-carbon battery can be discharged at a constant current of 200 A for 41min25s, releasing a total power of 138.077 Ah and a depth of discharge of 69.04 %. This shows that lead-carbon batteries are capable of both high-current discharge and deep discharge. Fig. 4 depicts the discharge capacity at various discharge currents.

3.2. Charging test

Conduct 60 A and 100 A CC charging tests on lead-carbon batteries, and the test is ended when the voltage reaches the saturation voltage of 2.45 V. In the first 30 min of 100 A CC charging, the charging voltage is almost the same as that of 60 A CC charging (Fig. 5), indicating that the lead-carbon battery can easily “absorb” 100 A current during charging. The battery voltage climbs fast after that. The charging lasts for 118.27mins with a charging capacity of 197.153 Ah or 98.58 % of the rated capacity.

Table 1
Positive plate curing and drying process.

No.	Temperature/°C	Humidity/%	Heating mode	Phase time /h	Steam humidification	Atomized water humidification	Airflow /%	Stage
1	40	99	1	5	1	1	20	Placing
2	48	88	1	2	1	1	30	Curing
3	48	88	1	1	1	1	30	Curing
4	56	85	1	8	1	1	30	Curing
5	56	88	1	24	1	1	30	Curing
6	52	80	1	1	1	1	40	Curing
7	52	80	1	3	1	1	40	Curing
8	45	60	1	1	1	1	80	Curing
9	45	60	1	5	1	1	80	Curing
10	45	50	0	4	0	0	100	Drying
11	65	10	0	1	0	0	100	Drying
12	65	10	0	14	0	0	100	Drying

The following is a description of the process parameter settings.
Heating mode: 0 shows radiator heating; 1 shows the heating mode automatically shifts between radiator and steam injection mode based on humidity.
Steam humidification: 0 shows that it is turned off; 1 shows that it is turned on and regulated by temperature and humidity.
Atomized water humidification: 0 shows that it is disabled; 1 shows that it is enabled and regulated by humidity.
When the air volume is set to 100 %, the air volume is = 800 m^3/h.

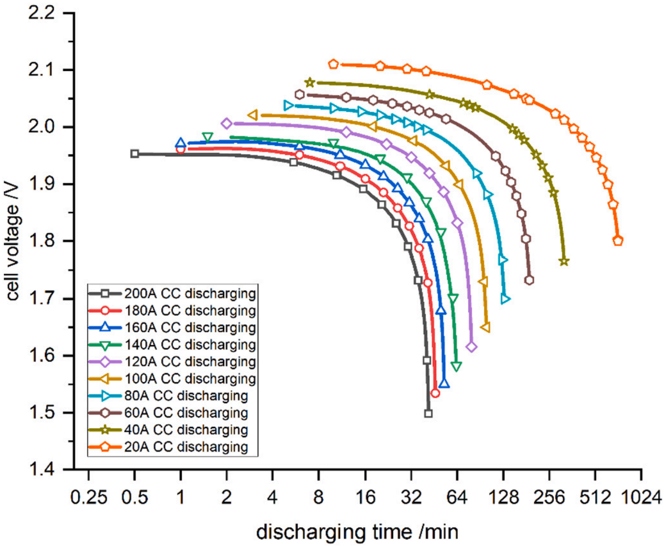

Fig. 3. Constant current discharging test with a high current.

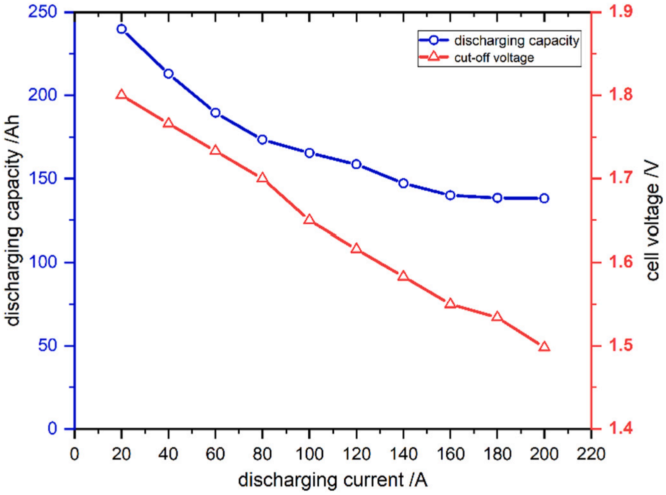

Fig. 4. Constant current discharging capacity and cut-off voltage with a high current.

A constant-current and constant-voltage charging test was done to further examine the high-current charging performance of lead-carbon batteries. Charging with constant-current and constant-voltage has two stages: first, charging with constant current until the voltage reaches 2.45 V; second, charging with 2.45 V constant voltage, whereas the current gradually falls. The charging stops when the accumulative charging power reaches the real capacity.

The test has a 5-second sample interval, and the voltage has reached 2.45 V at the fifth second, indicating that the constant-current and constant-voltage charging has advanced to the second stage. In the first 4 min and 50 s, the charging current jumped from 187.191 A to 340.255 A (Fig. 6), owing to the gradual transformation of the battery plates from lead sulfate with relatively poor conductivity to positive and negative active material, also known as the gradual transformation of the plate from a non-conductor to a conductor. As a result, the lead-carbon battery's maximum permitted charging current is 340.255 A, demonstrating that it has a high current charging capacity. In all, the constant-current and constant-voltage charging process takes 73 min and 50 s, with a charging current of 194.25 A on average.

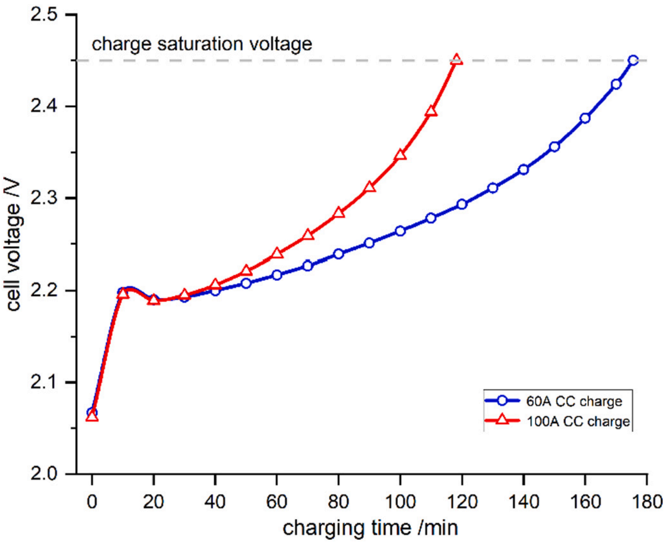

Fig. 5. Constant current charging at 60 A and 100 A.

3.3. *Cycle life test*

The total number of standard charge and discharge cycles when the battery discharge capacity has degraded to 80 % [35] (70 % [36] is also specified) of the rated capacity is referred to as battery cycle life. A cycle is a term referring to each completed charge and discharge. The conventional discharge procedure is a 20 A constant current discharge with a 70 % depth of discharge and a test temperature of 25 °C.

Testing the cycle life of a lead-carbon battery is a time-consuming and difficult process. There are various sped-up battery cycle life test techniques available now, mostly to enhance the cycle test temperature and depth of discharge. According to studies, every 10 °C increase in temperature reduces battery cycle life by 50 %, based on a temperature of 25 °C [37]. The depth of discharge has a major impact on battery cycle life, with cycle life at a depth of discharge of 100 % being around half that of a depth of discharge of 70 % [38–40].

The life cycle test conditions employed in this study are: The life cycle test is sped up by around 16 times when the ambient temperature is 55 °C and the depth of discharge is 100 %, and the next cycle charge capacity equals the last cycle discharge capacity throughout the life cycle test. The end of battery life occurs when the capacity of the battery drops to 80 % of its rated capacity during the test.

The enhanced lead-carbon battery was put through an 8720-hour cycle life test. The battery life is estimated to be around 7680 times after the 480th accelerated cycle test (Fig. 7). According to public information, lithium-ion batteries have a cycle life of roughly 4500 times [9], whereas Axion Power's lead-carbon batteries may be charged and discharged 1600 times [41]. In general, the depth of discharge of a battery can reach 70 % or even close to 100 %. However, it cannot be considered deep discharge capability if it is done at the cost of the battery's cycle life.

The cycle life of a battery when it is deeply discharged can evaluate the battery's deep discharge capability. The unimproved lead-carbon battery has a cycle life of roughly 3968 times, which is just 51.2 % of the enhanced lead-carbon battery. This demonstrates that the optimization strategy boosted the deep discharge capacity of lead-carbon batteries significantly.

3.4. *Safety performance test*

At 70 °C, the battery was charged 600 Ah (3 times its rated capacity) with a constant current of 100 A. The battery is bulging at the end of the experiment, but the battery shell is unharmed, there is no electrolyte

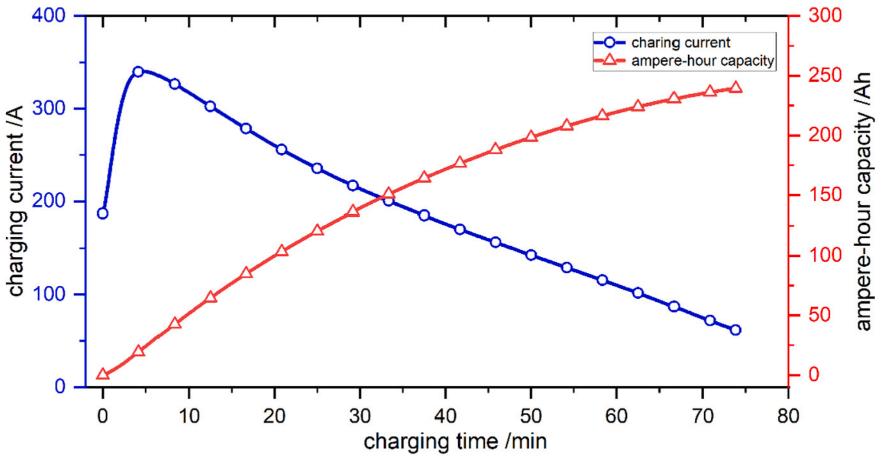

Fig. 6. Constant-current and constant-voltage charging.

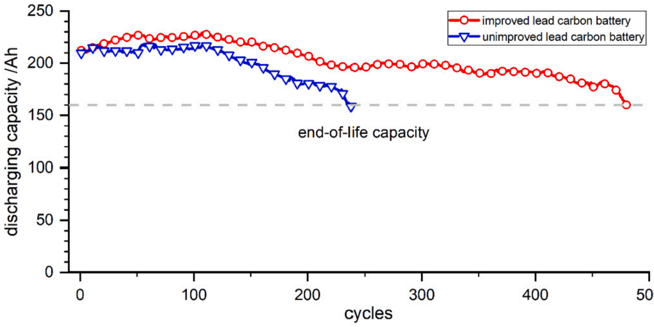

Fig. 7. Life cycle test (16 times acceleration).

leakage, and the battery has no harmful phenomena such as explosion or fire (Fig. 8), demonstrating that lead-carbon batteries have a good safety performance. However, the overcharged battery may still charge and discharge, but its capacity has been decreased to around 120 Ah.

4. Conclusion

In this study, activated carbon and carbon nanotube were added to the negative plate of a lead-acid battery to create an industrial lead-carbon battery with a nominal capacity of 200 Ah. When compared to lead-acid batteries, the maximum allowable charging current has increased from 0.3C to 1.7C (340 A). By thickening the positive grid, adding a tab, and refining the plate curing process, the cycle life of the lead-carbon battery has been enhanced during deep discharge. At 70 % depth of discharge, the cycle life is 7680 times, which is 93.5 % longer than an unimproved battery. This shows that the cost of a lead-carbon battery has been decreased to around RMB 0.55–0.6 per watt-hour, which may be further reduced to RMB 0.275–0.39 per watt-hour if the recycling price is between 35 % and 50 % of the purchase price.

CRediT authorship contribution statement

Zhide Wang (First Author): Conceptualization, Methodology, Validation, Formal Analysis, Investigation, Data Curation, Writing - Original Draft, Writing - Review & Editing, Visualization.

Xinpeng Tuo: Investigation, Validation, Data Curation.

Jieqing Zhou: Methodology, Resources, Project administration.

Gang Xiao (Corresponding Author): Conceptualization, Resources, Writing - Review & Editing, Supervision, Project administration, Funding Acquisition.

Fig. 8. Safety performance test.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors are unable or have chosen not to specify which data has been used.

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