



# Effects of carbon additives on the performance of negative electrode of lead-carbon battery



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## ABSTRACT

In this study, carbon additives such as activated carbon (AC) and carbon black (CB) are introduced to the negative electrode to improve its electrochemical performance, the negative electrode sheets are prepared by simulating the negative plate manufacturing process of lead-acid battery, the types and contents of carbon additives in the negative electrode sheets are investigated in detail for the application of lead-carbon battery. The electrochemical performance of negative electrode sheets are measured by chronopotentiometry, galvanostatic charge-discharge and electrochemical impedance spectroscopy, the crystal structure and morphology are characterized by X-ray diffraction and scanning electron microscopy, respectively. The experimental results indicate that the appropriate addition of AC or CB can enhance the discharge capacity and prolong the cycle life of negative electrode sheets under high-rate partial-state-of-charge conditions, AC additive exerts more obvious effect than CB additive, the optimum contents for the best electrochemical performance of the negative electrode sheets are determined as 0.5wt% for both AC and CB. The reaction mechanism of the electrochemical process is also discussed in this paper, the appropriate addition of AC or CB in negative electrode can promote the conversion of  $\text{PbSO}_4$  to Pb, suppress the sulfation of negative electrode sheets and reduce the electrochemical reaction resistance.

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## 1. Introduction

The environment problem will undoubtedly become one of huge challenges to human beings, thus the development of clean energy technology which was meeting the requirements of environmental protection was heavily pursued. As portable energy, batteries have become an important part of human life. Hybrid electric vehicles (HEVs) are developed to reduce vehicle emission due to environmental concerns. As a prospective candidate energy-storage system for HEVs applications, valve-regulated lead-acid (VRLA) battery has many advantages including low initial cost, well-established manufacturing base, distribution networks and high recycling efficiency [1,2] compared to other competitive technologies [3–5]. However, in HEVs, the battery is operated continuously in high-rate partial-state-of-charge (HRPSoC) cycling. In the HRPSoC mode, batteries experience short charge and discharge pulses with high

currents, high charging currents and exhaustive discharges cause an inhomogeneous acid density profile (acid stratification) of sulfuric acid used in the battery [6]. An early loss of capacity can be observed during a partial-state-of-charge mode because of the insufficient utilization of active mass and the discharged product,  $\text{PbSO}_4$ , which has not efficiently converted back to its original form, leading to progressive accumulation of irreversible  $\text{PbSO}_4$  on negative active material (NAM) [7–10]. The accumulation of  $\text{PbSO}_4$  reduces the effective reaction area of negative plates, and makes the charge and discharge processes of the negative plates difficult, finally leading to battery breakdown.

To address the abovementioned problems, many efforts have been made over the past few decades. Battery researchers and engineers have tried various innovative cell design solutions [11–13]. UltraBattery, a new technology, was developed by Commonwealth Scientific and Industrial Research Organization (CSIRO) Energy Technology. UltraBattery, which combines an asymmetric supercapacitor and a lead-acid battery in one unit cell, will reduce the cost and prolong the life of lead-acid battery in HEVs. These favorable parameters of the technology have been described in detail in the

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literatures [14,15]. Some studies also reported that the introduction of carbon materials to the negative plates of lead-acid battery can improve discharge capacity, charge acceptance ability and increase cycle life of the negative plates. Nakamura's experiment [16,17] proved that the addition of carbon black to the NAM can improve the charge acceptance significantly and retard the sulfation of negative plates during the simulated HRPSoC test of HEV batteries. Calabek et al. [18] established that the presence of carbon in NAM hinders the continuous growth of  $\text{PbSO}_4$  crystals, sustaining the formation of small crystallites with high solubility leading to efficient charge process. Some researchers [19–22] found that addition of carbon nanotubes to the NAM may enhance the charge acceptance, reduce energy losses, reserve capacity and cold-cranking performance of the battery. Pavlov [23] reported that electrochemical active carbon in the negative paste improves the charge efficiency and slows down sulfation of the negative plates. Saravanan et al. [24] reported that carbon from sugar into the NAM enhance the charge-discharge characteristic of the lead-acid cells. Moseley [25] reported that capacitive phenomena occur when Pb active mass contains considerable amounts of carbon. During charging with high currents, the electric double layer on carbon surface was charged first, and this process occurs at the potentials between the stationary electrode potential of Pb and the potential of hydrogen evolution. The electric double layer was discharged slowly at the expense of the reduction of  $\text{PbSO}_4$  to Pb. Thus, "the capacitive element can support charge and discharge events that occur at the highest rates, and the Faradaic part of the cell can cope with events that take place over a longer timescale." Moseley et al. [26] summarized the hypotheses proposed in the literature for the effect of carbons on the HRPSoC performance of the batteries as follows: (a) carbon enhances the overall conductivity of NAM [16]; (b) carbon facilitates the formation of small isolated  $\text{PbSO}_4$  particles that are easy to dissolve and restricts the growth of  $\text{PbSO}_4$  crystals [17]; (c) some carbon forms contain impurities which hinder the hydrogen evolution, thus improving the efficiency of charge [27]; (d) carbon acts as an electroosmotic pump that facilitates acid diffusion in the inner NAM volume at high rates of charge and discharge; high-surface area carbon materials particles exhibit a supercapacitive effect on NAM [28,29]. The use of carbon minimizes the uneven distribution of  $\text{PbSO}_4$  in the cross-section of negative electrode sheet, reduces discharge and charge current densities, and provides a conductive network to facilitate subsequent charging process [17,30]. Pavlov and coworkers [23] established that during the cycling of cells under HRPSoC conditions, the electrochemical reactions of charge at the negative plates occur not only on the Pb, but also on the surface of the carbon as well. They proposed a parallel electrochemical mechanism for charge and draw the conclusion that carbon should be incorporated and change the structure of NAM to enhance the battery performance under HRPSoC conditions [31]. Hollenkamp et al. [32] reported that the specific surface area (SSA) of NAM is an important parameter to the electrochemical performance of the negative plates. Pavlov et al. [33] reported that carbon added to NAM increase the SSA and decrease the median pore radius of NAM, which may change the electrochemical performance.

The addition of carbon materials to NAM can improve the performance of the negative plates, indeed. However, most researchers studied the influence of carbon additive on the negative plates of lead-carbon battery by an actual battery. On one hand, the battery performance in an actual battery depends on not only negative plate but also positive plate, so the problem focused on carbon additive in the negative electrode becomes more complicated in an actual battery; On the other hand, the preparation of an actual battery consumes lots of electrode materials. Furthermore, the optimum type and quantity of carbon additives and the mechanism for improved HRPSoC cycling

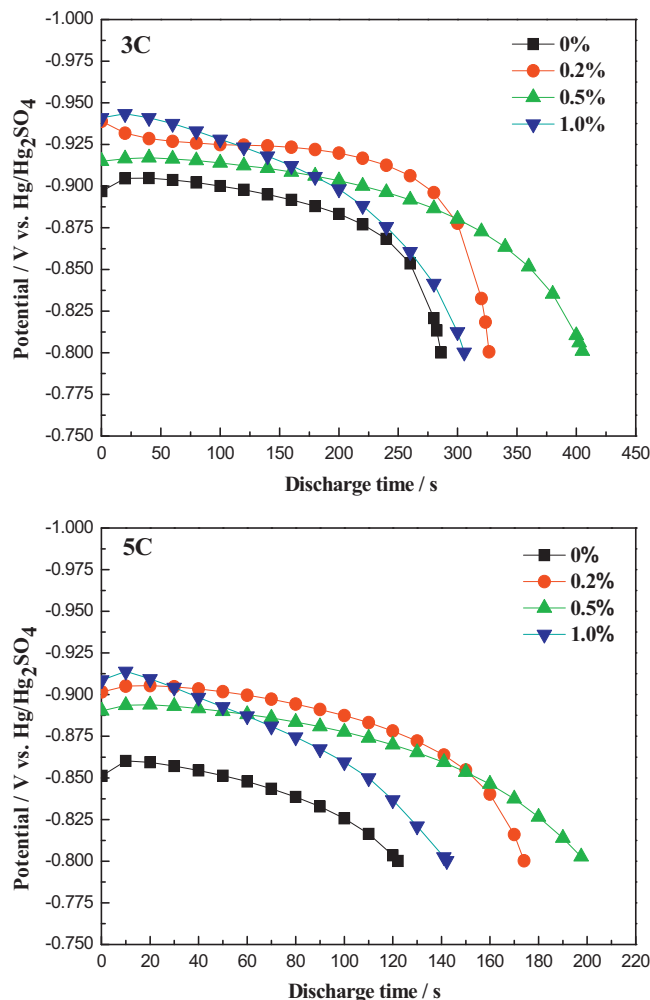
**Table 1**

Characteristics of the carbon additives added into NAM.

Symbol	Type of material	Conductivity (S cm)	BET surface $\text{m}^2/\text{g}$	Particle size
AC	Activated carbon	0.35	1800–2000	5–10 $\mu\text{m}$
CB	Carbon black	70	254	30 nm

performance of lead-carbon battery are still unclear and need further investigation.

In this paper, the negative electrode sheets were prepared by simulating the negative plate manufacturing process of lead-acid battery, the active mass in the negative electrode sheets was only about 0.2 g for a three-electrode system and 1.0 g for simulated flooded test cells, two types of commercially available carbon materials (activated carbon (AC) and carbon black (CB)) were selected as the additives for the negative electrode sheets. The discharge capacity and electrochemical impedance spectroscopy (EIS) of lead-carbon negative sheets were evaluated in a three-electrode cell, and the HRPSoC cycling performance was measured in the simulated test cells. The effect of AC or CB on negative plates and their reaction mechanism were discussed according to the experimental results.



**Fig. 1.** The discharge time results for negative electrode sheets with different contents of activated carbon at 3C and 5C discharge rates.

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