



Understanding the functions of carbon in the negative active-mass of the lead–acid battery: A review of progress

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ABSTRACT

The addition of supplementary carbon to lead–acid batteries that are intended for use in emerging automotive duties can provide improvement in two aspects of performance.

(i) In both hybrid electric and battery electric vehicles that are designed to preserve energy through the operation of regenerative-braking, conventional lead–acid batteries exhibit a rapid decline in the efficiency of the recuperative charging (which can involve rates up to 30C₇) and fail quickly as a result of an accumulation of lead sulfate on the negative plate. It has been widely reported that supplementary carbon — either intimately mixed with the negative active-material or included as a separate component attached to the plate — can enhance charge-acceptance.

(ii) Full-hybrid electric and battery electric vehicles employ high-voltage batteries composed of large numbers of cells connected in series. Consequently, when conventional lead–acid batteries are used in such configurations, the continuous cycling encountered in normal driving will almost certainly lead to divergence in the states-of-charge of the unit cells and thereby demand periodic equalization charges. In this application, it has been demonstrated that lead–acid batteries with supplementary carbon incorporated into the negative plate are rendered immune to the divergence problem and therefore operate without the need for an equalization charge.

The inclusion of supplementary carbon does, however, promote hydrogen evolution and failure due to the loss of water from the electrolyte solution. Current research efforts are directed towards methods by which this disadvantage can be mitigated without losing the benefits that the addition of carbon provides.

This review covers the extensive research that has been conducted to understand the mechanism by which the additional carbon operates, the additional studies that have sought to identify the best types and optimum amounts of carbon that should be used, together with the most effective manner for their deployment.

1. Recognition of extra carbon as a boost to dynamic charge-efficiency

For decades, negative plates in lead–acid batteries have been provided with a combination of carbon, barium sulfate and an organic additive, which is usually a wood extract, e.g., a lignosulfonate. These additives are collectively called an ‘expander’, although this term is often used purely for the organic component of the mix. During charge–discharge cycling, the expander serves to prevent individual crystals of lead from growing and combining into a dense structure with a low surface-area and, therefore, a low electrical capacity. The total amount of expander used in industrial valve-regulated lead–acid (VRLA)

batteries varies between 1.0 and 2.5 wt.% with respect to the weight of the oxide in the paste mix. The individual additions of carbon, barium sulfate and organic material vary according to the manufacturer’s specifications [1].

The benefits of including additional carbon in the negative active-mass beyond the level that is normal for the ‘expander’ function were first demonstrated by the work of Nakamura and Shiomi [2,3], who made negative plates that contained up to ten times the customary level of carbon. The actual amounts were not disclosed but, based on typical practice, were believed to be about 2.0 wt.% of the negative active-material loading. The trials, which were directed towards both electric vehicle and photovoltaic (PV) power applications, were conducted with

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Nomenclature*Acronyms and initialisms used in the text*

1BS	Monobasic lead sulfate ($\text{PbO} \cdot \text{PbSO}_4$)
3BS	Tribasic lead sulfate ($3\text{PbO} \cdot \text{PbSO}_4 \cdot \text{H}_2\text{O}$)
4B3	Tetrabasic lead sulfate ($4\text{PbO} \cdot \text{PbSO}_4$)
a.c.	Alternating current
AC	Activated carbon
AGM	Absorptive glass mat
ALABC	Advanced Lead–Acid Battery Consortium
BET	Brunauer–Emmett–Teller (technique for measuring specific surface-area)
BM	Ball mill
BP	Barton pot
BMS	Battery management system
BSS	Battery support system
CB	Carbon black
CSIRO	Commonwealth Scientific and Industrial Research Organisation
d.c.	Direct current
DCA	Dynamic charge-acceptance
DCE	Dynamic charge-efficiency
EAC	Electrochemically active carbon
EES	Electrical energy storage
EF	Fermi level
EG	Expanded graphite
FG	Flake graphite
GO	Graphene oxide
HER	Hydrogen evolution reaction
HEV	Hybrid electric vehicle
HRPSOC	High-rate partial-state-of-charge
LD	Donor level
OAN	Oil Absorption Number
PSOC	Partial state-of-charge
S	Siemens ($= \text{A V}^{-1}$ or Ω^{-1})
SoC	State-of-charge
SoCOT	Operational state-of-charge
VRLA	Valve-regulated lead–acid (battery)
XPS	X-ray photoelectron spectroscopy

Roman letters

A	Ampere; also real electrode surface-area (in m^2)
Å	Angstrom (10^{-10} m)
AC	Activated carbon
Ah	Ampere-hour
c	Molar concentration (mol L^{-1})
cm	Centimetre
°C	Degree Celsius
C	Coulomb ($= 1 \text{ A s}$)
C	Capacitance ($\text{in F} = \text{A s V}^{-1}$)
C_s	Specific-area capacitance (F cm^{-2})
C_m	Specific capacitance (F g^{-1})
C_{diff}	Ionic diffuse-layer capacitance (F)
C_H	Helmholtz double-layer capacitance (F)
C_{sc}	Space charge capacitance (F)
C_X/t	Discharge rate ($X = \text{hour rate}$; $t = \text{specified discharge}$

	time, usually in hours)
d	Thickness of double-layer (m)
e	Mathematical constant $= 2.71828$
e^-	Electron (negative charge: $1.602 \times 10^{-19} \text{ C}$; mass: $9.109 \times 10^{-31} \text{ kg}$)
E_F	Fermi level energy
E_V	Energy of the valence band
E_C	Energy of the conduction band
F	Farad ($= \text{C V}^{-1}$)
F	Faraday constant ($= 96\,485 \text{ C mol}^{-1}$)
g	Gram
i_o	Exchange-current density (A cm^{-2})
I	Current (A)
I_{bat}	Current (A) through the parallel connection of a capacitor and a charge-transfer component (a Faradaic element)
I_C	Current (A) through a capacitor
I_{CT}	Current (A) through a charge-transfer component (a Faradaic element)
J	Joule ($1 \text{ J} = 1 \text{ W s}$)
k	Kilo (10^3)
km	Kilometer
kW	Kilowatt
kWh	Kilowatt–hour
K	Kelvin (a measure of absolute temperature)
m	Metre; also milli (10^{-3})
mg	Milligram
min	Minute
ms	Millisecond
m	Mass (kg)
pm	Picometre (10^{-12} m)
Q	Electric charge ($\text{in C} = 1 \text{ A s}$)
R	Ohmic resistance ($\text{in } \Omega$), real part of impedance
R_{CT}	Resistance ($\text{in } \Omega$) of a charge-transfer component (a Faradaic element)
R	Universal gas constant ($8.3145 \text{ J mole}^{-1} \text{ K}^{-1}$)
s	Second
S	Surface-area (m^2)
T	Absolute temperature (K)
U	Volt (V); symbol commonly used in German forums; SI symbol is V
V	Voltage; also volume (m^3)
W	Watt ($1 \text{ W} = 1 \text{ J s}^{-1}$)
Wh	Watt–hour
wt.%	Percentage by weight

Greek letters

ϵ_0	Permittivity of vacuum ($98.854 \times 10^{-12} \text{ F m}^{-1}$)
ϵ_r	Relative permittivity of the dielectric
μ	Micro (10^{-6})
ρ	Density (kg m^{-3})
τ	Time constant (s), $\tau = RC$
ϕ_m	Work function
ϕ_{B0}	Schottky barrier height
χ	Electron affinity
Ω	Ohms

AGM-VRLA batteries that operated under PSOC conditions to lessen overcharge effects. Batteries with standard levels of carbon failed quickly due to the build-up of lead sulfate in the negative plate. By contrast, the companion positive plate was fully-charged. Batteries with extra carbon enjoyed appreciably longer operating lives. Recently, it

has been reported [4] that the additional carbon results not only in an increased cycle-life but also in greater specific energy at high rates.

To date, the prime aim of the work on carbon addition has been to support 12-V automotive batteries in micro-HEVS with regenerative-braking where the shortcomings of ‘dynamic charge-acceptance’ (DCA)

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