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New dry and wet Zn-polyaniline bipolar batteries and prediction of voltage and capacity by ANN

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Abstract

Chemically synthesized polyaniline doped with perchlorate ion was used as the electroactive material of the cathode in the construction of bipolar rechargeable batteries based on carbon doped polyethylene (CDPE) as a conductive substrate of the bipolar electrodes. A significant improvement in the originally poor adherence between the polymer foil and electroactive material layer of the anode was achieved by chemical pretreatment (etching) and single-sided metallization of the polymer foil with copper. A thin layer of optalloy was electroplated onto the surface of the copper-coated polymer foil to increase the battery overvoltage. A mixture of 1 wt% electrochemically synthesized optalloy, 92 wt% electrochemically synthesized zinc powder, 2 wt% MgO, 4 wt% ZnO and 1 wt% sodium carboxymethyl cellulose (CMC) was used as the anode mixture. Then, the electroactive mixture of the anode was coated onto the metallized surface of the CDPE. Graphite powder was used to coat the other side of the CDPE at 90 °C at 1 t cm^{-2} pressure This side was coated with a cathode mixture containing 80 wt% polyaniline powder, 18 wt% graphite powder and 2 wt% acetylene black. The battery electrolyte contained 1 M $Zn(ClO_4)_2$ and 0.5 M NH_4ClO_4 and 1.0×10^{-4} M Triton X-100 at pH 3.2. Both 3.2 V dry and wet bipolar batteries were constructed using a bipolar electrode and tested successfully during 200 charge–discharge cycles. The battery possessed a high capacitance of 130 mAh g^{-1} and close to 100% columbic efficiency. The loss of capacity during charge–discharge cycles for the wet bipolar battery was less than that for the dry bipolar battery. Self-discharge of the dry and wet batteries during a storage time of 30 days was about 0.64% and 0.45% per day, respectively. An artificial neural network (ANN) was used to model the voltage and battery available capacity (BAC) only for the dry bipolar battery at different currents and different times of discharge.
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Keywords: Rechargeable bipolar battery; Zn-polyaniline; Carbon doped polyethylene (CDPE); Battery available capacity (BAC); ANN modeling; Simultaneous prediction

1. Introduction

Recently, considerable interest has been paid to the study of polyaniline as a cathode in rechargeable batteries [1–6]. The majority of previously reported polyaniline-based rechargeable batteries are of the wet-type, in which hydrochloric acid or sulfuric acid have been used as a dopant acid and the corresponding electrolyte solutions contain $ZnCl_2$ and NH_4Cl at pH 4 and higher [7–11].

Recently, the use of bipolar systems in the construction of batteries has increased [12–16]. An important parameter in high-energy batteries is the thickness of the electrodes. Bipolar, flat plate electrodes demonstrate the importance of this parameter. The application of a bipolar cell design offers several advantages [13] and, therefore, this structure is used in the design principle for modern batteries. The design principle is strongly influenced by the choice of the electrochemical couple of the storage system [13].

Various substrates including stainless steel, lead, silver and carbon-polymer composite foils have been used in the construction of bipolar electrodes [14,17–22]. The aggravating disadvantages in the use of plastics as carriers for electroactive materials include low conductivity and lack of adherence to the electroactive layers. Usually, the electroactive materials are deposited onto the surface of a conductive

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carrier (metal or conductive polymer). A significant improvement of the originally poor adherence of the polymer foil to the electroactive layers can be achieved by mechanical (surface roughening) or chemical (etching) pretreatments [18–20].

The use of plastics with conductive fillers, like graphite and/or soot incorporated into the polymer demands an intermediate layer between the carbon-filled polymer and the zinc to prevent the formation of hydrogen gas by anodic corrosion, which is enhanced in the presence of carbon in any modification. The use of intrinsically conductive polymers like polypropylene and polyethylene as carrier materials requires special precautions and manufacturing processes because most of these polymers are insoluble and brittle. Thus, further treatment is hardly practicable. The advantage of intrinsically conductive polymers compared with filled plastics is that an intermediate layer between the carrier and the zinc is unnecessary. The achievable resistance of both plastics with conductive fillers and intrinsic conductive polymers are not comparable to that of metal carriers. However, the achievable resistance is sufficient for thin foils in bipolar arrangements.

Another promising method to form extremely thin electrodes, with a thickness in the range of some ten micrometers, is as follows:

- A metallic layer, serving as a current collector, is deposited onto one side of a thin, porous polymer foil such as polyethylene or polypropylene [18].
- The electroactive material is electrolytically or mechanically deposited onto the surface of a metallized plastic [19].

The flexibility of the polymer has a positive influence on the volume change of electroactive materials because the plastic provides flexible ‘mechanical struts’. The plastic acts as a binder and, therefore, prevents an increase in the internal resistance due to contact problems.

In previous studies, we employed polyaniline in the construction of a rechargeable battery [23], and showed that the polyaniline doped with perchlorate acts as a more suitable cathode material than chloride doped polyaniline for use in dry rechargeable batteries [24]. Recently, we successfully designed and constructed a rechargeable Zn-polyaniline dry battery based on stainless steel bipolar electrodes [12].

In all types of batteries, battery voltage is the first and most important factor. Modeling and prediction of voltage at various times and currents of discharge for each type of battery are of special interest. Because of the non-linear relationship between battery voltage and time of discharge and also voltage and discharge currents, artificial neural networks (ANNs) seem to be the most suitable for such modeling processes. In recent years, multivariate methods and ANNs were used for the prediction of battery characteristics such as capacity [25–30].

In order to fully employ the stored energy of the batteries, the accurate determination of battery available capacity (BAC) is very important. To the best of our knowledge, there are no reports about the modeling and prediction of polyaniline batteries. Most reports regarding modeling and prediction of battery characteristics were about lead-acid batteries [25–30]. For lead-acid batteries, the Peukert equation approach, which describes the relationship between the BAC value and the discharge current (I_d), is most commonly adopted. The Peukert equation is expressed as [31,32]:

$$\mathrm{BAC} = \frac{k}{I_d^{(n-1)}} \tag{1}$$

where k and n are constants determined from the discharge data of the lead-acid battery. However, the accuracy of the calculated BAC from Eq. (1) decreases drastically at both low and high discharge currents. Recently, a multi-level Peukert equation has been developed to improve the calculated accuracy, where two sets of constants k and n were specifically adopted for the calculation of the BAC under low and high discharge currents [27]. Nevertheless, Peukert-based equation approaches are only valid for lead-acid batteries and are ill suited to other batteries. To meet this purpose, BAC calculation at different discharge currents using an ANN has been proposed. This calculation can more accurately determine the BAC of other battery types, as well.

ANNs are mathematical systems that simulate biological neural networks. They consist of processing elements (neurons) organized in layers. The neural network used in this work is a back-propagation neural network (BPNN) [33–38]. A typical feed-forward neural network with back-propagation has three layers: the input, the hidden, and the output layers. The activation of a neuron is defined as the sum of the weighted input signals to that neuron:

$$\mathrm{Net}_j = \sum_i W_{ij} X_i + \mathrm{bias}_j \tag{2}$$

where W_{ij} is the weight-connection to neuron j in the current layer from neuron i in the previous layer, X_i is the input signal and bias j is the bias of neuron j. The Net_j of the weighted inputs is transformed with a transfer function, which is used to get to the output level. Several functions can be used for this purpose, but the “sigmoid function” is applied most often. This function is as follows:

$$y_j = \frac{1}{1 + \mathrm{e}^{-c\mathrm{Net}_j}} \tag{3}$$

where y_j is output of neuron j and c is a constant ($c \neq 0$). The network learns by adjusting its weights according to the error E [Eq. (4)]. The goal of training a network is to change the weights between the layers in a direction that minimizes the error E.

$$E = \frac{1}{2} \sum_p \sum_k (y_{pk} - t_{pk})^2 \tag{4}$$

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