Contents lists available at ScienceDirect

## Electrochimica Acta

journal homepage: www.elsevier.com/locate/electacta

# Effect of binder properties on electrochemical performance for silicon-graphite anode: Method and application of binder screening

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#### ARTICLE INFO

Article history: Received 3 March 2014 Received in revised form 15 April 2014 Accepted 12 May 2014 Available online 27 May 2014

Keywords: Lithium ion battery Binder Silicon anode Poly(acrylic acid) Poly(amide imide)

#### ABSTRACT

With increasing demand for lithium-ion batteries (LIBs) with high energy density, silicon-based negative electrode material has attracted much interest because of its high specific capacity. Practical utilization of Si remains unattainable, however, owing to severe volume expansion in the electrode, resulting in a loss of the electrical Si network, which is directly connected to drastic capacity fading of the cell. Therefore, there have been systematic studies on the characterization of fundamental binder properties to estimate the suitability of various binder materials. The binder properties are subdivided into mechanical and adhesion characteristics, electrode properties (rigidity and recovery), and phase separation behavior of slurry to correlate with the electrochemical performance and practical acceptance of candidate materials. Systematic screening showed that hybridization of poly(acrylic acid) (PAA) and poly(amide imide) (PAI) could complement each other's properties and the hybridized PAA–PAI was synthesized by a one-step, acid-catalyzed reaction. The PAA–PAI hybrid showed enhancement in overall properties as a result of co-polymerization and exhibited remarkable cycling performance after 300 cycles. Based on these results, it can be concluded that an understanding of binder characteristics provides useful insight into the search for a more efficient binder material, and fine tuning of fundamental binder properties through screening will be advantageous to the construction of more efficient LIB systems

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## 1. Introduction

Lithium-ion batteries (LIBs) are the most attractive energystorage systems based on their moderate rate capability, long cycle life, and high gravimetric and volumetric energy density [1–3]. As the demand for large-scale LIBs increases, they have emerged as promising power sources for hybrid-electric vehicles (HEVs), electric vehicles (EVs), and energy-storage systems (ESS) [4–8]. The relatively low energy density of currently available LIBs, however, presents a significant obstacle to their more widespread adoption, and much higher energy densities are needed for many practical applications. In this regard, silicon is one of the prospective anode materials owing to its excellent specific capacity (4200 mAh  $g^{-1}$ ), which exceeds that of a conventional graphite anode (360 mAh  $g^{-1}$ ) by an order of magnitude [9–11]. However, practical utilization

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http://dx.doi.org/10.1016/j.electacta.2014.05.062 0013-4686/© 2014 Elsevier Ltd. All rights reserved. of Si remains a challenge as the continuous irreversible electrochemical reaction of Si readily leads to poor cycling performance. This drawback mainly results from the intrinsic electrochemical nature of Si, where the electrochemical conversion of Si to Li<sub>x</sub>Si<sub>y</sub> is always accompanied by severe volume expansion in the electrode, causing it to pulverize and leading to isolation of the active material [12–14]. With the loss of an electrical network, the electrode is unable to process further electrochemical reaction, resulting in drastic capacity fading in cycling performance. Therefore, effective control of volume changes is the most indispensable requirement for utilization of Si. In this respect, an understanding of binder properties is essential as the primary role of the binder is to enhance the binding affinity between solid components (active material, carbon conductor) and the current collector in order to maintain an electronic network with a certain level of electrode uniformity to facilitate electrochemical reaction. Hence, many new candidate binder materials have been intensively investigated to promote binding interaction with the surfaces of Si particles or to facilitate Li<sup>+</sup> ion migration [15–20]; however, the operating principle







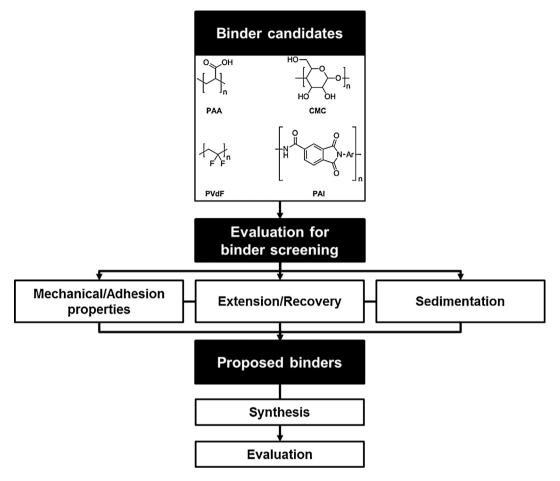


Fig. 1. Chemical structure of binder candidates and systematic strategy for binder screening method based on evaluation their properties.

of binders, namely, the main factor that enhances cycling performance, remains unclear. It has been observed that a binder with enhanced mechanical properties can help suppress volume expansion; however, its successful application is not assured because other factors including adhesion and slurry and electrode properties obviously affect the electrochemical performance as well. Therefore, a comprehensive assessment of the combination of binder properties with electrode properties is a useful and necessary probing tool to evaluate binder candidates. In this work, a systematic procedure for characterization of binder properties was established in terms of binding affinity, and the degree of extension and recovery in the electrode; it also included an analysis of phase separation behavior of slurries depending on the binder material (Fig. 1). The binding affinity is categorized according to the binding mechanism; i) mechanical properties of binders; ii) adhesion between the binder and Cu current collector. Insight into the effect of binding affinity on electrode performance will be useful in identifying the dominant factor of binder performance to maintain a desirable electrode network. As the factor of phase separation properties significantly affects electrode uniformity, which determines cell performance, an understanding of the phase separation behavior of various binders will be helpful to determine the processability of the material in industrial settings. Based on these assessments, promising binder materials were screened from candidates including (poly(acrylic acid) (PAA), carboxymethyl cellulose (CMC), poly(vinylidine difluoride) (PVdF), and poly(amide imide)(PAI)). A new hybridized binder was then synthesized, through which the complementary properties of individual materials were combined to form improved properties.

#### 2. Experimental

### 2.1. Measurement of mechanical and adhesion properties

To measure the mechanical properties of the binders, 5 g of each binder was dissolved in 10 mL of solvent (water for PAA (Aldrich, average MW: 100,000) and CMC (Aldrich, average MW: 90,000); *N*-methyl-2-pyrrolidone (NMP) for PVdF (Kureha, KF1100, average MW: 280,000) and PAI (Solvay, Torlon 4000T) to prepare the binder solution. After completion of binder dissolution, they were casted on Teflon (in case of water-based solution) or a glass mold (in case of NMP-based solution), respectively, and then dried in a vacuum oven overnight. The specimens were cut to pieces with the same dimensions (10 cm (width) × 30 cm (length) × 100 µm (thickness)) and their mechanical properties were measured by a tensile strength tester (JSV-H1000, Coretech).

To evaluate adhesion properties of binder, a mixture of the active material (silicon:graphite=3:7), binder, and carbon conductor (Super P) (in a weight ratio of 8:1:1) was dispersed in the solvent (water or NMP). The slurry formed from the dispersion was coated on a piece of copper foil, and the resulting electrode was dried in a vacuum oven at  $120 \,^{\circ}$ C for 12 h. After drying was completed, the electrode was completely attached onto slide glass with the identical dimensions ( $10 \,\mathrm{cm}$  (width)  $\times$  30 cm (length)  $\times$  100  $\mu$ m (thickness)) using a double-sided adhesive tape. Their adhesion properties were measured by a tensile strength tester (JSV-H1000, Coretech). Because delamination is only arisen between coating layer and Cu-current collector, recording stress measured in a tensile strength tester is considered as adhesive force.

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