



An eco-friendly approach of designing arecanut husk ash into high performance Li ion battery anodes

Deepthi Panoth^a, K. Brijesh^b, K.V. Baiju^a, HS Nagaraja^b, N Ponpandian^c, Anjali Paravannoor^{a,*}

^a School of Chemical Sciences, SAT Campus, Kannur University, Kerala, India

^b Department of Physics, NIT Suratkal, Karnataka, India

^c Department of Nanoscience and Technology, Bharathiar University, Coimbatore, India

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ABSTRACT

Designing a facile synthesis approach for large scale production of Si based anode materials that would resist the huge volume expansion and subsequent pulverization during cycling is a major challenge that hinders their commercialization. The present paper uses a green approach for the synthesis of a Si based composite electrode material sourced from arecanut husk ash. In addition, the conventional organic electrolytes are replaced with Room Temperature Ionic liquid based solvents to improve the safety as well as stability. It was found that the anode materials exhibit an exceptional stability of 85.3 % at the end of 100 cycles when used in half cells.

1. Introduction

The drastic volume change of Si anodes in Li ion batteries is a major challenge despite their huge theoretical capacity and most of the pioneering works consider nanostructuring, synthesis of yolk shell/core shell structures and the incorporation of active or inactive matrix materials to address the same [1–3]. However, fabrication methods of these structures are all limited to High temperature sophisticated procedures affecting scalability. In yet another perspective, irrespective of the process, the Si is ultimately sourced from sand or aquatic sediments posing a major environmental challenge. Using agricultural waste would be a sustainable ‘Go green’ approach and Rice husk and Sugar cane bagasse are already being explored for synthesis of Si nanostructures despite the low yield (2–5%) and lack of control in terms of size and morphology. [1,4].

Hence, a facile and cost-effective approach is proposed to fabricate porous Si spheres embedded in a silicate matrix, (CaSiO₃, CSM). An in-situ approach is adopted with Arecanut husk as the Si source, following magnesiothermic reduction. The environment friendly, low cost and sustainable approach can be a better solution for waste disposal from arecanut industry into value added products.

2. Materials and methods

Materials: Starting materials used for the synthesis were Areca nut husk, Magnesium powder: (~325 mesh, 99.8% - Alfa Aesar) Sulfuric

acid (H₂SO₄; 97%) and Hydrochloric acid, (HCl – 35%). Areca nut husk was burned in open air to form areca nut husk ash followed by acid leaching and calcination at 200, 600 and 800 °C in air. The as obtained ash was made to undergo magnesiothermic reduction at 850 °C for 4 h under vacuum followed by washing in ethanol, acetone and HF. A detailed synthesis procedure along with the methods for material and electrochemical Characterizations are given in the Electronic [Supplementary Information \(SI-I\)](#).

3. Results and discussion

The formation mechanism of the Si nanospheres embedded in Calcium silicate matrix is described in the Electronic [Supplementary Information \(SI-II\)](#) wherein preserving the Ca and Si content is the key factor. [Fig. 1 \(a\)](#) shows the XRD spectra of the pristine Si/CSM composite. The signals of Si/CSM samples correspond to the Si and β-CaSiO₃ phases consistent with reported data (JCPDS No. 76–0186 β-CaSiO₃ and JCPDS No. 27–1402Si) [5,6]. The XRD spectra of the AHA samples treated at various temperatures are given in the Electronic [Supplementary Information \(SI-III\)](#) indicating the presence of amorphous phases in the 200 °C, and more crystalline phases with signals from both Silica and CaSiO₃ at higher temperatures of 600 and 800 °C. [Fig. 1\(b\)](#) exhibits the morphological features of the Si/CSM samples as unveiled by SEM analysis. The sample demonstrates spherical particles embedded in a sheet like matrix, with a uniform size distribution (~50 nm). In order to further analyse the morphological characteristics,

* Corresponding author.

E-mail address: anjali.nano@gmail.com (A. Paravannoor).

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