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KANDY, SRI LANKA

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810

Pro
"

Dye Sensitized Solar Cells with Poly(vinyl alcohol) Based Gel Polymer Electrolytes Containing Lithium Bis(oxalato)borate Salt	172
Z. Osman, I.M. Noor, M.A. Careem and A.K. Arof	
Poly(methyl methacrylate) Based Gel Polymer Electrolyte Containing Iodide/Triiodide Redox Mediator with Application in Perovskite Solar Cells	174
I.M. Noor, M.Z. Kufian and A.K. Arof	
Perspective of Excellent So-Called Perovskite Solar Cells: Panchromatic and Semiconducting $[\text{PbI}_2(\text{MENH}_3^+)_4]$-Aligned Molecular Structured Solar Cells	175
S. Yanagida	
Evaporation Method for High Quality Perovskite Thin Film on Different Textured Substrates for Tandem Solar Cells Application	177
L. Cojocaru, K. Wienands, J.C. Goldschmidt and S.W. Glunz	
Plasmon Enhanced Dye Sensitized Solar Cell Performance using TiO_2/Ag NW Nanocomposite Photoanode	179
M.G.C.M. Kumari, C.S. Perera, B.S. Dassanayake, M.A.K.L. Dissanayake and G.K.R. Senadeera	
Novel PV Power Analyzing System with Maximum Power Point Tracking Technique: Towards the Solution for Evaluation Challenges of Perovskite Solar Cells due to Hysteresis in I-V Measurements	186
P.V.V. Jayaweera, S. Kaneko, L. Cojocaru, S. Uchida and H. Segawa	
Preparation Methods and Characterization of Key Components in Solar Cells	187
B.O. Agyeman	
Broadband Absorption and Photo-thermal Conversion Properties of $\text{ATO}/\text{C}/\text{DEG}$ Nanofluids	188
N. Chen, H. Ma, C. Zhang, D. Wu, Z. Meng and H. Zhu	
Multi-Objective Optimization of Air-Based Hybrid PV/T Solar Panels	196
L. Ouhssaine, M. Siroux, M.E. Ganaoui and A. Mimet	
The Synthesis and Enhanced Photothermal Conversion Effect of Ag_2S Hollow Nanostructures	207
Y. Sun, W. Zeng, Q. Jiang, C. Zhang, H. Zhu and D. Wu	
Study of the Improvements of Solar Cell Performances by Concentration	213
K.M. Liyanage, S.S. Abeywickrama, G.D.K. Mahanama	
Enhanced Photocatalysis of H_2O_2 in the Presence of a Cobalt-Trimasic Coordination Polymer	217
D.U.G.S.N. Abeysingha, C.V. Hettiarachchi, and I.R. Perera	
Lithium-Ion Battery Systems for Solar Energy Storage	224
B.-E. Mellander, F. Larsson	
Performance of Li-ion Rechargeable Batteries Assembled with Electrodes Prepared from Developed Sri Lankan Vein Graphite and Nanostructured $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-x}\text{Na}_x)\text{O}_2$	229
T.H.N.G. Amaraweera, H.W.M.A.C. Wijayasinghe, N.W.B. Balasooriya, A.N.B. Attanayake, B.-E. Mellander and M.A.K.L. Dissanayake	

Electrical and Thermal
Secondary Batteries
J.H.T.B. Jayamaha,
M.A.K.L. Dissanayake

Performance of Developed
the Li-ion Rechargeable
B.T.S. Hewatillak
Prasanna

FTIR Analysis of Ethylene
W. Faruqi, T.M.W.J.
B.-E. Mellander

Evaluating Carrier Re-
Spectroscopy
S.S. Dissanayake and

Rechargeable Cell with
as an Energy Storage De-
W.A.D.S.S. Wernsing

Recycling of Si PV Modu-
N.N. Pradhan, G.R. B.

Investigation Properties of
K.A.C. Nilmini, J.V. V.

Effect of Temperature on
at Bimolecular Solar Pow-
A.M.K.L. Abeykoon, C.

A Study on Solar PV Po-
Faculty of Engineering, U-
A. Kanagasabaram, R.

Improved Electrochemical
Material for Sodium-Ion B-
R.I.C.N. Karunaratne,

Effect of Different Carre-
Polymer Electrolyte
H.N.M. Sarangaika, M.J.
Penna

Investigation of an all Poly-
Charge Discharge Test
K.W. Prasadini, W.A.D.

A Coordination Polymer of
Wastewater
W.M.Y.H. Wijesundara,

Electrical and Thermal Properties of Nano-Composite Gel-polymer Electrolytes for Sodium-ion Secondary Batteries	235
J.H.T.B. Jayamaha, L. Wewagama, K. Vignarooban, K.S. Perera, K.P. Vidanapathirana, M.A.K.L. Dissanayake, S. Senthuran, P. Ravirajan, M.A. Careem and A.K. Arof	
Performance of Developed Shiny-Slippery-Fibrous Type Sri Lankan Vein Graphite Anode in the Li-Ion Rechargeable Battery	241
H.P.T.S. Hewathilakea, N.W.B. Balasooriya, H.W.M.A.C. Wijayasinghe and H.M.T.G.A. Pitawala	
FTIR Analysis of Ethylene Carbonate Electrolytes for Batteries and Dye Sensitized Solar Cells	246
M. Furlani, T.M.W.J. Bandara, H.D.N.S. Fernando, S. Bertilsson, M. Friesel, I. Albinsson and B.-E. Mellander	
Evaluating Carrier Recombination Dynamics of Photovoltaic Materials Using Terahertz Spectroscopy	251
S.S. Dissanayake and M.-J. Sher	
Rechargeable Cell with Poly-n-methylpyrrole Cathode and Gel Polymer Electrolyte to be Used as an Energy Storage Device	252
W.A.D.S.S. Weerasinghe, K.W. Prasadini, K.P. Vidanapathirana and K.S. Perera	
Recycling of Si PV Modules	258
N.N. Pradhan, G.R. Bader and A.R. Weerasinghe	
Investigation Properties of Type II Solar Radio Burst with CME Using Math Lab	263
K.A.C. Nilmini, J.V. Wijesekara and E.M.B.V Ekanayake	
Effect of Temperature on the Photovoltaic Characteristics of Polycrystalline Silicon Solar cells at Hambantota Solar Power Plant	270
A.M.K.L. Abeykoon, G.M.L.P. Aponsu, H.M.B.I. Gunathilaka and H.A.V. Nadeera	
A Study on Solar PV Power Generation Influencing Parameters Using Captured Data from Faculty of Engineering, University of Jaffna Solar Measuring Station.	276
A. Kanagasundaram, R. Valluvan and A. Atputharajah	
Improved Electrochemical Performance of Li-Substituted $\text{NaNi}_{0.4}\text{Mn}_{0.4}\text{Co}_{0.2}\text{O}_2$ Cathode Material for Sodium-Ion Batteries	284
R.I.C.N. Karunarathne, H.W.M.A.C. Wijayasinghe and M.A.K.L. Dissanayake	
Effect of Different Current Collectors in Mg Battery with Conventional PEO Based Gel Polymer Electrolyte	289
H.N.M. Sarangika, M.A.K.L. Dissanayake, G.K.R. Senadeera, J.A.K. Sampath and T.T.M. Perera	
Investigation of an all Polymer Redox Capacitor using Cyclic Voltammetry and Galvanostatic Charge Discharge Test	295
K.W. Prasadini, W.A.D.S.S. Weerasinghe, K.P. Vidanapathirana, K.S. Perera	
A Coordination Polymer of Titanium for Efficient Removal of Cationic and Anionic Dyes from Wastewater	300
W.M.Y.H. Wijesundara, H.A.I.R. Perera and C.V. Hettiarachchi	

PERFORMANCE OF LI-ION RECHARGEABLE BATTERIES ASSEMBLED WITH ELECTRODES PREPARED FROM DEVELOPED SRI LANKAN VEIN GRAPHITE AND NANOSTRUCTURED $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-x}\text{Na}_x)\text{O}_2$

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ABSTRACT

Rechargeable batteries, such as Lithium Ion Batteries (LIBs,) offer viable solutions for storing electrical energy harnessed through solar cells and other renewable energy sources such as hydro, wind and biomass. Investigation of low cost and performance enhanced electrode materials for LIBs is an important task for the advancement of the next generation of LIBs. In this context, Sri Lankan natural vein graphite has been identified as a promising anode material for the LIBs. Moreover, the Na doped $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-x}\text{Na}_x)\text{O}_2$ cathode material shows further enhanced electrochemical performance. However, the performance of full LIB cell comprising a vein graphite anode has not yet been reported. Hence this abstract presents the outcome of our study conducted on a full LIB cells assembled with electrodes fabricated from developed Sri Lankan vein graphite and locally synthesized nanostructured $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-x}\text{Na}_x)\text{O}_2$.

For the anode electrode preparation, vein graphite (99.99 % carbon) purified through acid leaching was further modified by mild chemical oxidation. For the cathode, phase pure nano-structured $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-x}\text{Na}_x)\text{O}_2$ was locally synthesized by employing the glycine nitrate combustion technique. The electrodes were fabricated by tape casting the developed electrode materials onto respective current collectors by employing the doctor blade method. Finally, CR2032 type LIB coin cells were assembled with these electrodes together with the non-aqueous liquid electrolyte of LiPF₆.

The initial discharge capacity of the cell assembled using the developed graphite anode and $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2)$ cathode was 98 mA h g⁻¹ at C/5 rate between 3.0 and 4.2 V, at room temperature. More interestingly, the cell assembled with the Na doped $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-x}\text{Na}_x)\text{O}_2$; x= 0.04 cathode with the same graphite anode showed a higher discharged capacity of 135 mA h g⁻¹ under the same operational condition. Furthermore, this full cell showed a higher capacity retention over cycling. Altogether, it clearly shows the very promising cell performance of full LIB cell comprised of electrode fabricated from developed Sri Lankan vein graphite and locally synthesized nano-structured $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-x}\text{Na}_x)\text{O}_2$.

Keywords: vein graphite, $\text{Li}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3-x}\text{Na}_x)\text{O}_2$, lithium-ion batteries, cell performance.

INTRODUCTION

Invention of new techniques to store more energy is a crucial area of research to address the increasing challenge of energy needs for the present technology era. Rechargeable Lithium Ion battery (LIB) is a very promising rechargeable energy storage device. Current LIBs are based on expensive electrode materials such as synthetic graphite as the anode material and lithium metal oxides as the cathode material. However, the high unit cost and unsolved materials-related problems are keeping the LIBs from reaching the common masses as a cheaper and reliable portable power source [1]. Modified vein graphite in Sri Lanka has been identified as a cost effective alternative to the synthetic graphite [2]. However, effect of vein graphite anode on the performance of LIB has not yet been identified. Therefore, present study aims to investigate the electrochemical performance of lithium ion battery employing anode fabricated from purified vein

with inverse temperature showed
the electronic transference number
conducting NCGPE sample, confirming
thermal studies showed that the test
-14.9 °C.

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PERFORMANCE OF DEVELOPED SHINY-SLIPPERY-FIBROUS TYPE SRI LANKAN VEIN GRAPHITE ANODE IN THE LI-ION RECHARGEABLE BATTERY

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ABSTRACT

Rechargeable Lithium-Ion Batteries (LIBs) are at present the most demanded rechargeable battery system due to its higher capacity, energy density and excellent cycleability. However, many investigations are presently proceeded on the development of the electrode materials in order to achieve higher efficiency in a more cost effective manner. Recently, natural graphite was identified as a potential low cost anode material for LIBs. Sri Lanka has its own established name for the high quality vein type natural graphite containing 95-98% of initial purity. Sri Lankan vein graphite (SLVG) has been categorized into four structurally distinct morphological varieties namely, shiny-slippery-fibrous graphite (SSF), needle-platy graphite (NPG), coarse striated-flaky graphite (CSF) and coarse flakes of radial graphite (CFR). Anisotropy of the surface and impurities present in trace levels cause profound alteration of chemical and electrochemical reactivity in natural graphite. Our recently introduced acid digestion method could result over 99.9 % of purity for all the four morphologies of SLVG, while simultaneously undertaking the surface modification. Further, studies have proved that this treatment method could enhance the performance of the NPG type SLVG anode. Therefore, this study aims to evaluate and compare the electrochemical performance of SSF type SLVG anode that was developed through acid digestion treatment.

The treated SSF type SLVG powder (<53 µm) developed by acid digestion was used for this study. Electrochemical half-cell studies were carried out using CR 2032 coin cells with treated SSF type SLVG as active material and 1 M LiPF₆ (EC: DMC; vol. 1:1) as the electrolyte. All the assembling steps were performed in an argon filled glove box under the controlled oxygen and moisture levels. Galvanostic charge-discharge studies were conducted at 0.2 current rate with "Landat" battery testing instrument. Treated anode shows high reversible discharge capacity of 440 mA h g⁻¹, which is higher than the theoretically expected capacity for a graphite anode. Initially, the first cycle discharge capacity was recorded as 517 mA h g⁻¹. In contrast to the NPG type SLVG anode treated with same method, the SSF type SLVG anode gradually faded from the 1st cycle to 5th cycle and achieved its stable reversible capacity. However, after the 5th cycle, the treated SSF anode could be able to maintain a high reversible capacity of 440 mA h g⁻¹ up to the 50th cycle. Further, charge-discharge curves didn't give evidences for any strong passivation reaction and a continuous formation of SEI layer could be observed up to 4th cycle. However, it may be absent or declined to a very low level after the 5th cycle. Hence, lithiation and delithiation occurred very smoothly without any degradation reaction and it achieved a high reversible capacity for the first 50 cycles while maintaining over 99% of Columbic efficiency. Altogether, this study reveals the promising performance of our developed shiny-slippery-fibrous type Sri Lankan vein graphite anode in the Li-ion rechargeable battery.

Keywords - vein graphite, anode, lithium-ion rechargeable battery, cell performance.

INTRODUCTION

Arrival with the novel technological aspects, the energy demand for the portable electronic devices and electrical vehicle increases rapidly. In order to cater this demand, several types of rechargeable batteries have been introduced. Due to their higher capacity, energy density and excellent cycleability, Rechargeable Lithium-Ion Batteries (LIBs) become the most prominent rechargeable battery type [1-2]. However, most of the current research studies are focusing on cost-effective electrode materials instead of synthetic electrode components, which are presently problematic with performance issues and high cost. Many of recent investigations have proved the



IMPROVED ELECTROCHEMICAL PERFORMANCE OF Li-SUBSTITUTED $\text{NaNi}_{0.4}\text{Mn}_{0.4}\text{Co}_{0.2}\text{O}_2$ CATHODE MATERIAL FOR SODIUM-ION BATTERIES

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ABSTRACT

Low cost rechargeable batteries, made using abundant materials, operating in safe voltage window are much attractive for large scale energy applications involving solar energy integration and electric grids. In this regard, Sodium-Ion Battery (SIB) has become a promising solution for future issues related to the high demand for energy storage devices, mainly due to the high natural abundance and low cost of sodium resources. As a result, extensive research efforts have been directed to search for suitable functional materials, especially for the anode and the cathode of SIBs. Though few successful studies on this subject have been reported in recent years, many challenges are still remained in developing reliable electrodes for SIBs.

$\text{NaNi}_{0.4}\text{Mn}_{0.4}\text{Co}_{0.2}\text{O}_2$ adopts a layered structure, where the transition metal atoms are located in octahedral sites between oxygen layers. In attempting to improve the electrochemical performance of $\text{NaNi}_{0.4}\text{Mn}_{0.4}\text{Co}_{0.2}\text{O}_2$, substitution of Li for Ni was investigated under the parent study. Using X-ray diffraction (XRD) and scanning electron microscopy (SEM), the structure and morphology of the cathode material were characterized. The electrochemical performance of the Li doped cathode was investigated in terms of charge-discharge curves and cycleability at different C rates.

The XRD measurements demonstrated that these prepared materials have pure hexagonal P- type layered structure with R3m space group comparable with the parent structure. SEM images exhibited the existence of globular-shaped sub-micron primary particles that agglomerated to form into micron size, softly bounded secondary particles consists of different micron size cavities. The charge-discharge test exhibits that the Li-doped materials has an initial specific discharge capacity of 153 mA h g^{-1} at C/125 rate, which gradually decreased to 125 mA h g^{-1} after six cycles. However, further investigations have to be performed in order to study the cycling stability of the material for the cathode application in the sodium ion battery.

Keywords: Sodium-ion batteries (SIBs); Cathode; Transition metal oxides.

1. INTRODUCTION

Na-intercalation batteries are appearing as an important alternative to Li intercalation systems, hence extensive research efforts have been directed to develop high capacity cathode materials for Sodium Ion Batteries (SIB) [1-5]. Among them, layered transition metal oxides have become most reliable promising candidates for the positive electrodes of SIBs due to their high specific capacities [1-3]. Although promising, they exhibit capacity fading due to the structural instability during charging/discharging process. Therefore, most of recent research investigations have been focused to enhance the electrochemical performance by partially substituting different elements to the host structure, while keeping the structural stability of the material. In addition, some foreign ions can be used to improve the electrical conductivity and suppress phase transformation, which leads to the enhanced electrochemical performance of the materials. Recently, it was found that structural doping of lithium in manganese to be effective in enhancing the structural stabilization of the cathode upon cycling. Nevertheless, these transition metal oxide cathodes are still much inferior to that of Li-based cathode analogs, limiting their practical energy densities [4].

In this paper, we report f
 $\text{NaNi}_{0.4}\text{Mn}_{0.4}\text{Co}_{0.2}\text{O}_2$ syn

2. EXPERIMENTAL

2.1 Syntheses of cathode

The nominal con
prepared by glycine ni
(Fisher Scientific), MnN
(99.2%, VWR) and LiN
components were mixed
Glycine:Nitrate ratio as
temperature. The result
temperature of resulting
decompose the nitrate ar
calcined at 900°C for 3 h

2.2 Material characteriz

Powder X-ray di
instrument with Cu (K_α)
of the synthesized mater
(ICP-MS). The morpho
microscope (SEM,ZES
spectroscopy (EDS).

2.2 Electrochemical cha

The electrodes us
active material, 10 wt%
thoroughly mixed in N-r
slurry and then cast on a
 $^\circ\text{C}$ for 12 hours in vac
electrochemical behavior
containing pure sodium
separator, assembled in
hexafluorophosphate) dis
dimethyl carbonate (DM
were performed in the v
battery cycling tester, re

3. RESULTS AND DIS

The powder XRD
diffraction peaks can be
except some extra peaks
the presence of a second
Two other peaks, which