RESEARCH ARTICLE

Chemical intercalation of graphite using chromium trioxide for the anode application in rechargeable sodium-ion batteries

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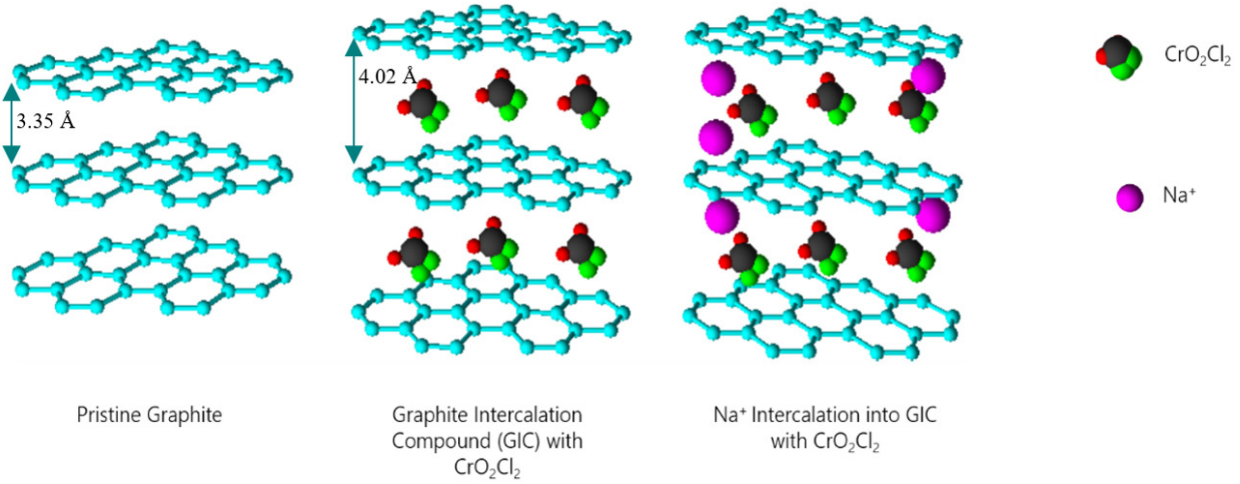

Highlights

- GIC prepared with CrO_3 using vein graphite for Na^+ rechargeable battery applications has never been reported before.
- Practically, retarded by the small interlayer spacing, pristine graphite has a negligible sodium storage capacity.
- The capacity of the battery assembled with CrO_3 treated GIC was higher than that of raw graphite.

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Chemical intercalation of graphite using chromium trioxide for the anode application in rechargeable sodium-ion batteries

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Abstract: Among naturally occurring graphite, Sri Lankan vein graphite (SLVG) is well known for its high initial purity in the range of 95 - 99% carbon, high crystallinity, extensive mineralization, and mode of occurrences. Due to its distinctive properties, Sri Lankan vein graphite has been proved to be a promising anode material for rechargeable Lithium-ion battery (LIB) applications. With the increasing demand for rechargeable energy sources, the supply of Lithium-ion batteries (LIB) could be hindered by the limited lithium reserves. Therefore, alternatives for Lithium in LIBs are being explored. In this context Sodium-ion batteries (SIB) have attracted much attention during the recent past. This study focused on studying the intercalation and deintercalation of Na^+ ions into the domain of Graphite Intercalation Compound (GIC), fabricated from natural vein graphite, for the anode application of rechargeable SIBs. Graphite Intercalation Compounds with CrO_3 (GIC-CO) were successfully synthesized using SLVG as the starting material. The synthesized GIC-CO exhibited an interlayer spacing of 4.02 Å which is regarded to be suitable for Na^+ ion intercalation. The anode material fabricated from GIC-CO showed a capacity of 26.9 mA h g^{-1} which is higher than that of pristine graphite (13 mA h g^{-1}). This shows the potential of utilizing this GIC for anode applications in SIBs. The anode could be improved by optimizing the molar ratios of CrO_3 and the reaction time to obtain expanded graphite making it appropriate for high-capacity SIB application.

Keywords: Sodium-ion battery; graphite intercalation compound, interlayer expansion, graphite, chromium trioxide.

INTRODUCTION

The booming introduction of numerous novel electrical equipment, involving all kinds of consumer electronics, electric tools, and electric vehicles, leads to a heavy demand for rechargeable portable energy sources. Lithium-ion batteries (LIB), characterized by high specific energy, high efficiency, and long life, have been the common power source of choice for the consumer electronics market with a production of the order of billions of units per year (Scrosati & Garche, 2010). However, this large-scale production of LIBs may be hampered by the limited lithium resource reserves (Hou *et al*., 2017; Li and Bjerrum, 2002; Wang *et al*., 2017). Cheaper and more abundant elements such as sodium, magnesium, and aluminum-based battery systems have been recently investigated as an alternative to LIBs (Gofer *et al*., 2009; Li and Bjerrum, 2002; Slater *et al*., 2013). Among these elements, sodium has attracted much attention as it is the second-lightest and the smallest alkali metal next to lithium in the periodic table. However, the charge storage capacity of graphite is related to the number of ionic intercalants. That is directly limited by the size of the intercalating ion and is also dependent on the structure and morphology of the graphite materials used (Wang *et al*., 2017). Na^+ has a larger ionic radius (1.06 Å) than Li^+ (0.76 Å), which has a direct impact on mass transport and storage in the electrochemical process. It cannot be easily intercalated into graphite interlayers, as Li^+ ions can in LIBs (Slater *et al*., 2013).

Figure 1 depicts the schematic diagram showing the crystalline structure of graphite. Pristine graphite has an interlayer spacing of 3.354 Å (Hewathilake *et al*., 2018). Under ambient conditions, Lithium ions are gradually inserted between single graphite layers, and a maximum of one lithium per six carbons (LiC_6) is expected to be intercalated, in the Van der Waals gaps above and below a carbon hexagon (Figure 1) (Deng, 2015). Therefore, it has a theoretical capacity of 372 mA h g^{-1} Vs Li/Li^+ in LIBs (Deng, 2015). It was found that sodium-graphite intercalation compounds (Na-GICs), like NaC_6 and NaC_8 caused by the high redox potential of Na/Na^+, are quite unstable due to the stress caused by the stretching of C-C bond lengths to the unstable Na-GICs (Nobuhara *et al*., 2013; Zhang *et al*., 2019; Perveen *et al*., 2020). However, according to previous studies, based on theoretical calculations, a minimum interlayer distance of 3.7 Å is essential for Na^+ insertion into carbon materials in conventional ester-based electrolytes (Cao *et al*., 2012; Zhang *et al*., 2019). They also stated that alterations of the graphite anode such as increasing the interlayer spacing, edge functionalization, and the formation of defects can be used to adjust the energy of sodiation (Zhang *et al*., 2019). Different types of structurally modified graphite with large interlayer distances in the range of 3.7 - 4.0 Å have been studied as the anodes for SIB. This mostly includes reduced

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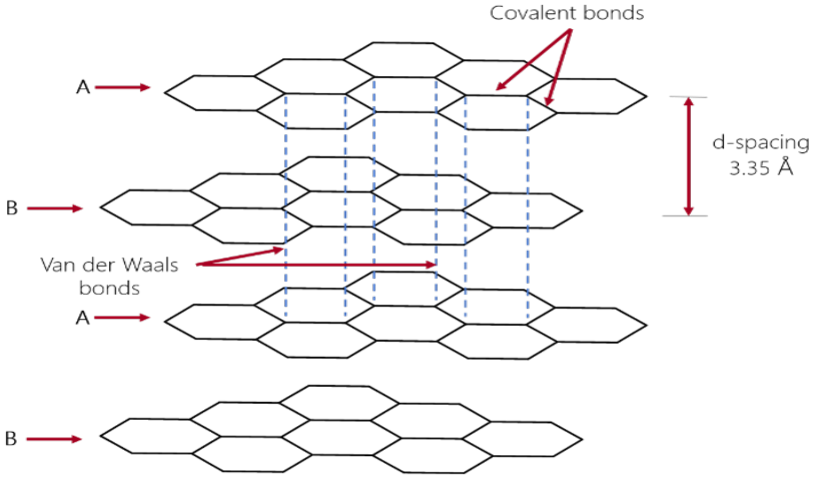

Figure 1: Schematic diagram showing the crystalline structure of graphite.

graphene oxide (rGO) as the anode material (Zhang *et al*., 2019). Apart from rGO, interlayer expansion could also be achieved through the preparation of graphite intercalation compounds (GIC) (Mittal *et al*., 1998).

Graphite intercalation compounds (GICs) are compounds with intercalated foreign species into the interplanar interstitial sites of the graphite crystal such that the layer structure of the graphite lattice is retained. These GICs exhibit excellent physical and chemical properties comparable to those of pristine graphite (Xu *et al*., 2017). In graphite, the carbon atoms within a layer are strongly bound while the carbon atoms in adjacent layers are weakly bound. As a result, the intercalating substance occupies and thereby expands the interplanar spacing of the graphite crystal without disrupting the carbon layers(Chung, 2016). These compounds exist in concentration stages marked by differences in the periodic stacking sequence of the carbon layers and the intercalate layers. The number of carbon layers between two intercalated layers defines the 'stage' of the compound. As the concentration of the intercalate increases, the stage decreases (Rudorff, 1988).

In a study by Mittal *et al*., 1998, they proved that by using CrO_3 and graphite in HCl at room temperature under normal atmospheric conditions, GICs could successfully be produced. They also stated that the stage structure of the resultant GICs could be controlled by the amount and strength of the HC1, amount of $CrO_{3,}$ and the reaction time (Mittal *et al*., 1998). Hence, intercalation into graphite interlayers using CrO_3 could result in an interlayer spacing that could facilitate the intercalation and de-intercalation of Na^+ ions into the domain of GIC. The synthesis of GIC by intercalating graphite with CrO_3 prepared using vein graphite has never been studied before. Therefore, the main objective of this study is to synthesize GIC using natural vein graphite and CrO_3 using HCl followed by an investigation of the intercalation and de-intercalation of Na^+ ions into the domain of GIC, with the ultimate aim of using it in SIBs applications.

MATERIALS AND METHODS

For the present study, Needle Platy Graphite (NPG), which usually possesses the highest purity among the four structural varieties of Sri Lankan vein graphite (Hewathilake et al., 2018) was collected from a deep underground mine. Figure 2 shows a sample of the Needle Platy Graphite variety of Sri Lankan Vein Graphite. It was then crushed using a laboratory disk mill and particle size fractions < 53 μm were separated by mechanical sieving. The collected sample was purified by the acid leaching method (Amaraweera et al., 2013, 2018).

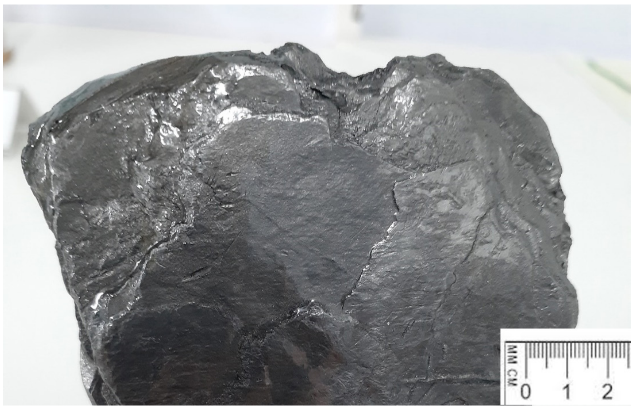

Figure 2: Needle Platy Graphite variety of Sri Lankan Vein Graphite.

One gram of purified NPG was taken into a beaker, and 8.5 g of CrO_3 and 35% HCl (7 mL) (Sigma Aldrich) were added to it, and the resultant mixture was stirred at room temperature for two hours. The product was washed with deionized water several times to remove unreacted CrO_3 and finally washed with acetone.

The washed product was dried at 60 ℃ in a vacuum oven overnight to obtain the graphite intercalation compound prepared with chromium trioxide (GIC-CO).

In order to analyze the crystal phase present, the resultant material was characterized by powder X-ray

Diffractometry (PXRD) with 'Rigaku-Ultima IV' Powder X-ray diffractometer, using Cu- Kα_1 radiation ($\lambda = 1.54$ Å). Fourier Transformer Infrared (FTIR) spectroscopy (Nicolet 6700 spectrometer) was used to identify functional groups present in GIC, within the 500 - 4000 cm^{-1} region. Scanning Electron Microscopy (SEM) (EEVO/LS 15 ZEISS) was used for morphological observation of the GIC.

Electrochemical performance investigations were conducted by assembling Na-ion half-cells with electrodes fabricated with the GIC-CO. For electrode fabrication, 70% of GIC-CO powder was mixed with 15% of conductive carbon (Carbon Black), 15% PVDF (Sigma Aldrich) in NMP (Sigma Aldrich) using a mortar, and mixed using a pestle for 30 min. The resulting slurry was then casted on a copper foil. After drying it at room temperature for 12 h, the sample was then transferred into a standard vacuum oven and kept at 100 °C overnight. In an argon-filled glove box, a coin cell (CR2032) was assembled from a fabricated electrode, a sodium metal counter electrode, a blown glass fiber separator, and a liquid electrolyte. The electrolyte was prepared by mixing 1.0 mol dm^{-3} $NaPF_6$ (sodium hexafluorophosphate) dissolved in a 1:1 volume ratio solution of ethylene carbonate (EC) and dimethyl carbonate (DMC) (Sigma Aldrich) solvent. Galvanostatic charge/discharge cycles were carried out using a Landt CT2001A battery testing station. The half cells were cycled between 0.00 and 2.5 V (versus Na/Na$^+$) at a 0.2 C rate.

RESULTS AND DISCUSSION

Crystallographic characteristics

Figure 3 shows Powder X-ray Diffractograms of graphite (a), graphite intercalation compound prepared by chromium trioxide (b). The number in front of *hkl* stands for the stage number of the GIC and G stands for unreacted graphite. PXRD [Figure 3(a)] shows that graphite has a major peak corresponding to an interlayer spacing of 3.35 Å with a relatively high diffraction intensity due to good order along the c-axis direction (Touzain *et al*., 2010). However, after being treated with CrO_3 and HCl, the major PXRD peak shifts to 22° with a corresponding interlayer spacing of 4.02 Å [See Figure 3(b)]. The peak structure corresponds to the formation of the stage 3 graphite intercalation compound which is reflected in four adjacent characteristic diffraction peaks (Lin et al., 2016; Mittal et al., 1998; Skowroński, 1988). As shown in Figure 3 (b), four adjacent diffraction peaks (13.02°, 22°, 24.7°, and 28.75°) are visible, from which a stage-3 intercalation structure is able to be determined definitely. Here, an intensity-reduced graphite peak at ~ 24° denotes that part of the graphite structure is retained with incomplete intercalation (Lin *et al*., 2016).

The intercalation could have occurred from the following reaction given in Equation 1 (Mittal *et al*., 1998). In the presence of concentrated HCl, the intercalation of CrO_3 to graphite is primarily achieved by CrO_2Cl_2 (Mittal *et al*., 1998).

$$CrO_3 + 2HCl \rightarrow CrO_2Cl_2 + H_2O \qquad (1)$$

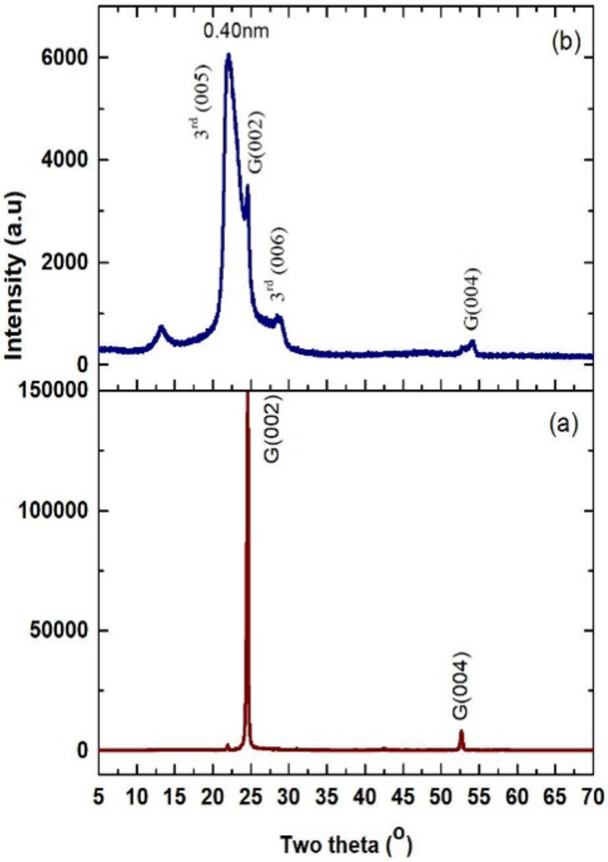

Figure 3: Powder X-ray Diffractograms of graphite (a) Graphite intercalation compound prepared by chromium trioxide (b) The number in front of *hkl* stands for the stage number of the GIC and G stands for unreacted graphite.

Surface Characteristics

The FTIR spectrum of graphite and graphite intercalation compounds prepared by chromium trioxide (GIC-CO) is shown in Figure 4. It evidences the presence of some functional groups as given below. Strong broad vibrational bands from the 3200 - 3600 cm^{-1} region are responsible for stretching vibrations of the hydroxyl group, where the hydroxyl groups may be from absorbed water molecules or phenolic OH or OH from carboxylic groups. The stretching vibration of the OH group is prominently higher in GIC-CO than that of graphite. Further, carbonyl groups C=O stretching at 1710 - 1700 cm^{-1} have increased when the graphite reacts with CrO_3 in the presence of HCl. Alcoholic groups O-H stretching at 1390 cm^{-1} were also observed, with high absorption values (Yu *et al*., 2016). The reason behind the functionalization of the treated graphite could be explained by the formation of CrO_2Cl_2, a highly reactive compound, and powerful oxidizing agent which could have oxidized the graphite (Mittal *et al*., 1998).

Surface morphology

Figure 5 shows the SEM images of graphite (a) and the graphite intercalation compound prepared by chromium trioxide (GIC-CO) (b). As evidenced from the SEM images, graphite shows a compact layered structure [Figure 5(a)] while on the contrary, GIC-CO evidenced

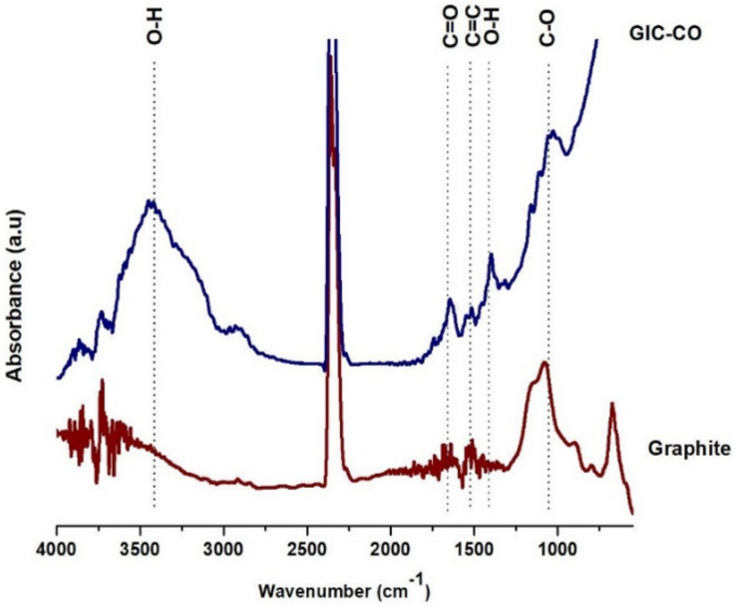

Figure 4: FTIR spectra of graphite (bottom graph) and Graphite Intercalation Compound prepared by Chromium trioxide (GIC-CO) (top graph).

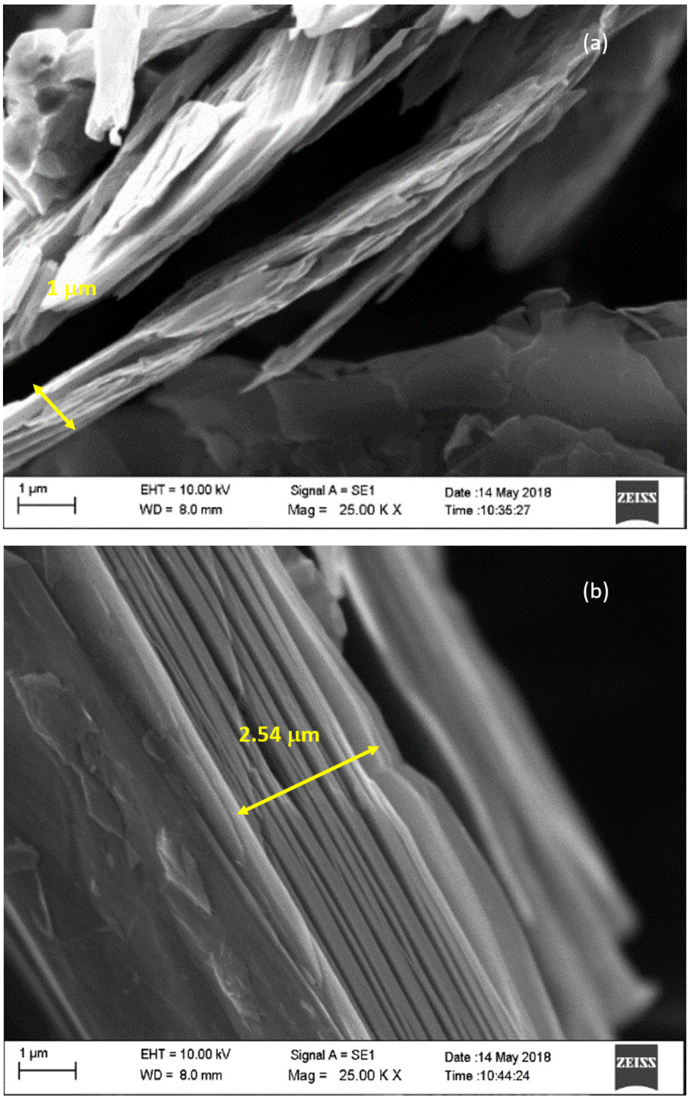

Figure 5: (a) SEM images of graphite (b) Graphite intercalation compound prepared by chromium trioxide (GIC-CO).

an expanded “accordion” like structure [Figure 5(b)]. As explained earlier, Intercalation retains the stacked structure of graphite, but the interlayer spacing is widened, often by several angstroms or more, which may result in the electronic separation of the individual layers (Rudorff, 1988).

Mechanism of the intercalation

According to the results obtained from the powder X-ray diffractometry, the Graphite Intercalation Compound prepared by chromium trioxide (GIC-CO) showed a higher interlayer spacing compared to the pristine graphite. CrO_2Cl_2 formed from the reaction between HCl and CrO_3 enters into the graphite domain when stirred at room temperature for 2 h. The intercalation retains the stacked structure of graphite, but the interlayer spacing is widened as evidenced by the powder X-ray diffractogram. Figure 6 illustrates a mechanism proposed for intercalation of CrO_2Cl_2 into the graphite domain and how Na^+ intercalation could occur. Hence, GIC-CO, which reported a broader interlayer space of 4.02 Å, was selected to fabricate electrodes for investigating its performance as the anode material in the rechargeable sodium-ion battery.

Electrochemical Performance of the synthesized GIC versus Na/Na^+

The electrochemical performance of this GIC-CO as the anode material of the Na-ion rechargeable battery was investigated by carrying out electrochemical charge-discharge cycle tests. Cells were cycled between 0.00 and 2.5 V (versus Na/Na^+) at a 0.2 C rate. The capacity was calculated based on the mass of the active material.

Figure 7 shows the first discharge and charge cycle curve of the cell assembled with GIC-CO versus Na/Na^+ at a 0.2 C current rate between 0.00 and 2.5 V. The battery assembled in the present study, with GIC-CO, showed a specific discharge capacity of 215.9 mA h g^{-1} with a charge capacity of 26.9 mA h g^{-1} for the first cycle. An irreversible capacity of 189 mA h g^{-1} can be observed for this assembled battery. During the first discharge process in the electrochemical test cells, some of the sodium ions might have reacted at the surface of the carbon electrode with electrolyte species to form an electronically insulating but ionically conductive surface layer known as the solid electrolyte interphase (SEI) (Stevens & Dahn, 2001). This SEI formation is one of the

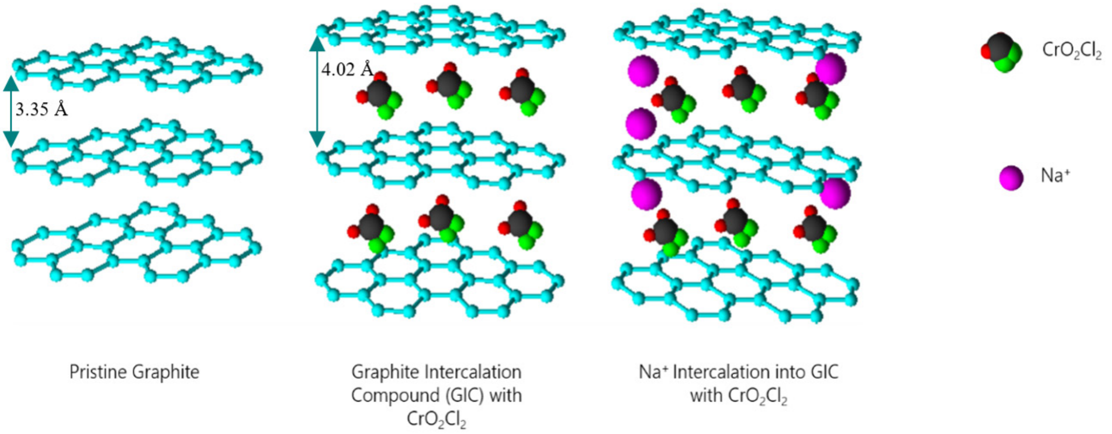

Figure 6: Graphite Structure and CrO_2Cl_2 and Na^+ intercalation into graphite domain.

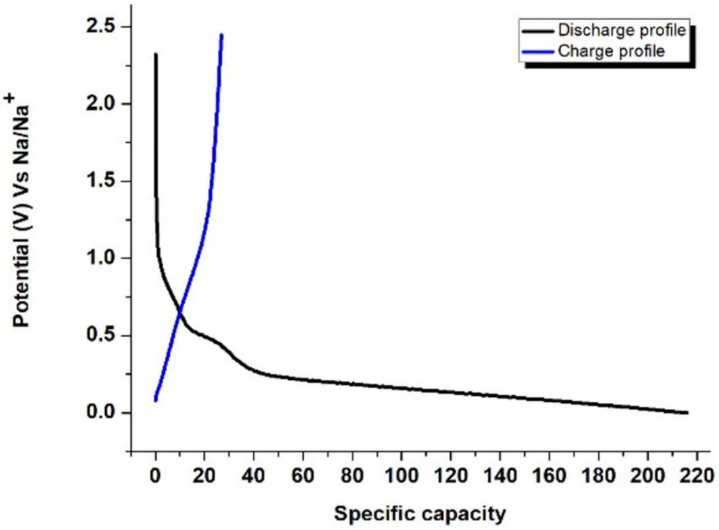

Figure 7: The first discharge and charge cycle curve of the assembled cell with GIC-CO versus Na/Na^+ at 0.2 C current rate between 0.00 and 2.5V.

primary contributors to irreversible capacity essentially the amount of charge lost between the first discharge and subsequent charge cycles for graphite electrodes (Stevens & Dahn, 2001). Carbonate-based electrolytes (EC/DMC) form relatively thick insulating SEI layers compared to ether-based electrolytes on the graphite surface, which block Na^+ solvent transport (Sun, 2017).

The theoretical capacity of Na^+ in graphite is 35 mA h g^{-1} (Ge & Fouletier, 1988). Practically, retarded by the small interlayer spacing, pristine graphite has a negligible sodium storage capacity of 13 mA h g^{-1}, which is widely known and has been reported previously (Wen *et al*., 2014). The present study using GIC-CO electrode versus Na/Na^+ at a 0.2 C current rate between 0.00 and 2.5 V resulted in a capacity of 26.9 mA h g^{-1}. Hence, when compared to pristine graphite, GIC 6 prepared with CrO_3 shows a capability of accommodating a higher number of sodium ions within its layers. This shows the potential of utilizing this GIC for anode applications in SIBs.

The resultant capacity could be the result of intercalation compounds within the graphene layers, which could be blocking the entrance and exit of Na^+ ions into its layers. The SEI layers could have also contributed to the blocking of the Na^+ entrance and exit into the graphite layers. On the other hand, according to another study, GIC prepared with CrO_3 in HCl could be exfoliated using H_2O_2 (Skowroński, 1988). Hence, the present study suggests that this could further be improved by moderating the molar ratios of CrO_3 and the reaction time, and with the addition of H_2O_2, it could result in better interlayer spacing suitable for the anode application of SIBs.

CONCLUSION

A graphite intercalation compound with CrO_3 (GIC-CO) was successfully prepared using Sri Lankan vein graphite as the starting material. The prepared GIC-CO exhibited an interlayer spacing of 4.02 Å. This spacing is considered suitable for Na^+ ion intercalation. GIC-CO was used to fabricate the anode material for Na^+ intercalation. The resultant capacity of the assembled cell with GIC-CO versus Na/Na^+ at a 0.2 C current rate between 0.00 and 2.5 V was 26.9 mA h g^{-1}. This capacity is higher than that of pristine graphite (13 mA h g^{-1}). This shows the potential of utilizing this GIC for anode applications in SIBs.

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DECLARATION OF CONFLICTS OF INTERESTS

The authors declare no competing interests

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