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Among the countries possessing natural vein graphite resources, Sri Lanka is renowned for its highly crystalline vein graphite with high natural purity and extensive mineralization. Sri Lankan Vein Graphite (SLVG) with its distinctive properties, should be able to play an important role in high-tech industrial applications. Value addition to Sri Lankan graphite has to be done in order to increase the revenue to the country. Graphite has already been introduced as a viable electrode material for the extensively used Li-ion batteries (LIBs). However, the application is limited by the fact that Li is a rare element. As an alternative, Na-ion batteries (NIB) have recently drawn significant attention but the ionic radii of Na⁺ being rather larger than that of Li, expansion of interlayer spacing of graphite is essential in order to use it for the anode of NIBs. This study aims to expand the interlayer spacing of vein graphite via thermal, microwave and chemical reduction of Graphite Oxide (GO) synthesized from natural vein graphite. It is because the reductions via various channels is expected to result in removal of selective functional groups and therefore end-up as expanded graphite. For that, Sri Lankan vein graphite from underground mines was collected and visually inspected to identify the different morphological varieties. Small chips of the selected variety were crushed and particle size fraction below 53 μm was separated by mechanical sieving. The powder sample was subjected to purification via acid leaching. GO was prepared by Tour Method using the purified Needle Platy Graphite (NPG) variety, which possesses the highest carbon content among the different morphological varieties of Sri Lankan vein graphite. The synthesized GO was reduced via chemical (CrGO) reduction with sodium hydroxide, microwave (MWGO) and thermal (TrGO) reductions and the resultant materials were characterized. The X-ray diffractograms obtained for NPG, GO, MWGO, CrGO have their major peaks corresponding to a d -spacing of 3.35, 8.37, 3.65, 3.73 Å. The thermally reduced Graphite Oxide (TrGO) showed its major peak at 13.78⁰ with a more favourable d -spacing of 6.42 Å, indicating a potentiality for the anode application of the NIB. Further, Fourier Transform Infrared spectroscopic analysis confirms that only few oxygen functional groups are reduced by the thermal

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treatment while most of the functional groups could be reduced by the other two methods. Scanning electron micrographs evidence for the existence of a honeycomb-like structure in the MWrGO and a puffed structure in the CrGO but a compact structure for TrGO. Altogether, this study shows that the thermal annealing of graphite oxide is a suitable method, among the investigated methods, to obtain interlayer spacing suitable for NIB anode from Sri Lankan vein graphite.

Keywords: *vein graphite, graphite oxide, expanded graphite, electrode materials, batteries*