Supplementary information

Large scale green production of ultra-high capacity anode consisting of graphene encapsulated silicon nanoparticles

Ali Reza Kamali^{a,b*}, Hyun-Kyung Kim^b, Kwang-Bum Kim^c, Vasant R. Kumar, D.J.Fray*^b

a) School of Metallurgy, Northeastern University, Shenyang 11018, China

b) Department of Materials Science and Metallurgy, University of Cambridge, CB3 0FS, UK

c) Department of Material Science and Engineering, Yonsei University, 50 Yonsei-Ro, Seodaemun-

Gu, Seoul 120-749, Republic of Korea

Emai: ali@smm.neu.edu.cn, alirezakam@yahoo.com (a.r.Kamali)

Fig. 2a exhibits an electron SEM image of the graphite electrode material used for the molten salt preparation of graphene-silicon nanocomposite. The micrograph exhibits the presence of graphite flakes with irregular grain sizes up to several micrometers and various particle shapes. The XRD pattern taken from the graphite material in the powdered form in the twotheta range from 20° to 30° is shown in Fig.1S and the data extracted from this pattern is summarized in Table 1S. The results obtained demonstrate the appearance of the sharp (002) reflection at 2theta=26.36° corresponding the d-spacing of 0.338 nm. An electrode made of this material was exfoliated in molten LiCl to produce graphene nanosheets. Fig.1S exhibits a SEM micrograph of the graphene nanosheets produced in molten salt [1]. Fig. 2S and Table 1S compare the XRD pattern of the graphene product with that of the graphite starting material on the same scale of the intensity axis. The low intensity of (002) peak in the XRD pattern of the graphene product indicates that its hexagonal crystallites are much less abundant in (002) planes in comparison with those in the graphite starting material. It shows that the graphite has largely been exfoliated during the molten salt process. Fig. 2b shows a bright field TEM micrograph of the Si nanoparticles used for the fabrication of the nanocomposite. It should be noticed that SEM study of silicon nanoparticles was difficult due to the extensive electron charging effect caused by the semi-insulating Si surfaces, as can be seen in Fig. 3S. The silicon nanoparticles in the graphene-Si nanocomposite, however, didn't show this charging effect (Fig.2) which can be attributed to the presence of a conductive network of 3D graphene nanosheets. Fig. 4S shows the powder X-ray diffraction pattern (blue line) and the Rietveld refinement profile (red line) as well as peak information for GW-Si fabricated in the molten salt. Fig. 5S shows the Coulombic efficiency of GW-Si containing 50wt% Si, shown in Fig. 5b, with high magnification.



Fig.1S. SEM micrograph of graphene nanosheets produced by the cathodic exfoliation of graphite in molten salt.



Fig. 2S. X-ray diffraction pattern of the graphite starting material, and graphene powders produced by the exfoliation of the graphite in molten salt.

Table 1S. XRD data for the hexagonal (002) peak of the graphite raw material and graphene product.

	Pos. [°2θ]	Height [cts]	FWHM Left [°20]	d-spacing [Å]
Graphene	26.5253	5000	0.3936	3.36044
Graphite	26.3556	41000	0.3375	3.38170



Fig.3S.SEM micrograph of silicon nanoparticles. It is clear that the nanoparticles tend to agglomerate into clusters. The electron charging effect caused by the semi-insulating Si surfaces make the harsh white-dark contrast.



Fig. 4S. Powder X-ray diffraction pattern (blue line) and Rietveld refinement profile (red line) as well as peak information for GW-Si fabricated in the molten salt.



Fig. 5S. Coulombic efficiency of GW-Si containing 50wt% Si, shown in Fig. 5b, with high magnification.