

Influence of flake/potato graphite particle ratio on SEI formation and negative electrode performance in lithium-ion batteries

S. Chhor,<sup>1,2</sup> S. Mailley,<sup>2</sup> E. DeVito,<sup>2</sup> P. Mailley,<sup>3</sup> et G. Gébel<sup>1</sup>

<sup>1</sup> CEA-INAC, 17 rue des Martyrs - 38054 GRENOBLE Cedex 9, France

<sup>2</sup> CEA-LITEN, 17 rue des Martyrs - 38054 GRENOBLE Cedex 9, France

<sup>3</sup> CEA-INES, 50 Avenue Du Lac Léman, 73377 Le Bourget Du Lac, France

Email : sarine.chhor@cea.fr

Graphitic carbons are recognized in lithium-ion batteries to play a major role in irreversible charge loss (due to the formation of the solid electrolyte interphase SEI) and in overall cell performance.

The behavior of graphite in lithium-ion batteries depends on the morphology and chemistry of its surface. [1] Graphite exhibits two kinds of surface with different specific reactivities: the basal plane surface mainly contains sp<sub>2</sub> carbon atoms and the prismatic (edge) surface contains both carbon and oxygen atoms. [2] Lithium ions insertion during charge/discharge takes place via the prismatic surfaces. During the first intercalation of lithium ions into graphite, the solvent and the lithium salt are reduced to form a surface film (SEI) on graphite electrode. This film has to be protective and effective, i.e. assure the protection of the graphite electrode from further electrolyte decomposition and electrode exfoliation while permitting the transport of lithium ion.

The active material in graphite electrode is ideally a round-shape graphite like mesocarbon microbeads. In this work, a combination of two types of graphite was used as intercalation materials: flake-shape graphite (7.5 m<sup>2</sup>/g, d=6.5µm) and potato-shape graphite (17 m<sup>2</sup>/g, d=32µm). The flake-shape graphite can improve the electrical percolation between the potato-shape graphite. Different electrode formulations containing varying ratios of both particles were assayed to study their influence on the irreversible charge loss and improve the cyclability of the graphite electrode. We aim to optimize the formulation, understand the role of each particle in the different formulations and thus better control the SEI formation.

The influence of flake/potatoe graphite ratio is evaluated by following the anode performance in lithium/graphite half-cell using charge/discharge tests and cyclic voltammograms. In the first cycle, SEI formation starts at higher potentials with flakes graphite (~0.7V) than potatoes graphite (~0.5V) at room temperature and at 40°C. (Fig.1)

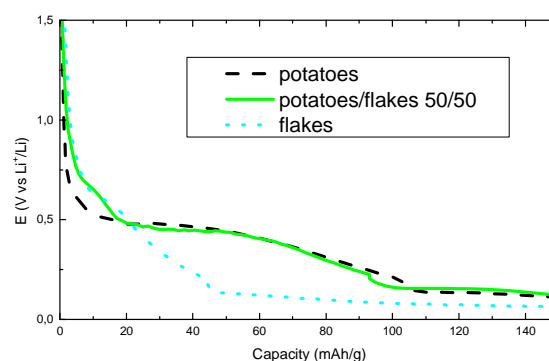


Figure 1. Discharge curve in the first cycle (between 1.5V and 0.1V)

The irreversible charge loss is linearly dependant on the ratio at 40°C. A contrasted evolution was recorded at room temperature, which could be ascribed to the electrode surface accessibility to the electrolyte. (Table 1)

Graphite composition	Irreversible capacity at RT (mAh/g)	Irreversible capacity at 40°C (mAh/g)
Potatoes	134 ± 9	41 ± 2
Potatoes/Flakes 50/50	128 ± 6	56 ± 2
Flakes	57 ± 1	72 ± 2

Table 1. Irreversible capacity at RT and 40°C for different ratios of potatoes/flakes graphite

The cyclability of the cell is optimum for a 50/50 mix. (Fig. 2)

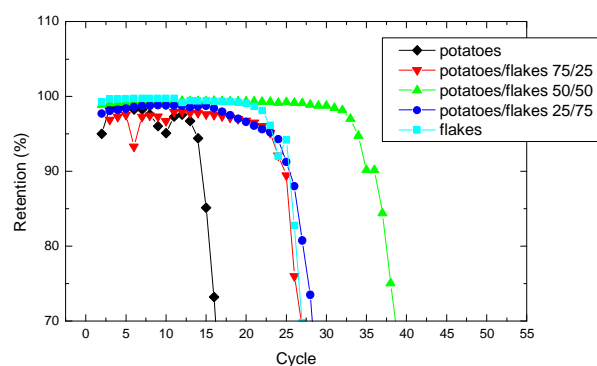


Figure 2. Capacity retention for different ratios of potatoes/flakes graphite

To support this finding, SEM and XPS analysis were also done to follow the evolutions in the morphologies and the chemistries of the SEI and the graphite particles.

[1] Winter, M., P. Novak, et al. (1998). "Graphites for lithium-ion cells: The correlation of the first-cycle charge loss with the Brunauer-Emmett-Teller surface area." *Journal of the Electrochemical Society* **145**(2): 428-436.

[2] Placke, T., V. Siozios, et al. (2012). "Influence of graphite surface modifications on the ratio of basal plane to "non-basal plane" surface area and on the anode performance in lithium ion batteries." *Journal of Power Sources* **200**: 83-91.