Supporting Information

Exploring nature-behaviour relationship of carbon black materials for potassium-ion battery electrodes

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Note S1: Bragg's law: $n\lambda = 2d \sin(\theta)$, in which *n* is an integer equal to 1, λ is the wavelength equal to 0.154187 nm, *d* is the vector of the displacement between reflection sites, and θ is the angle between the reflected ray and the plane formed by the material surface.

Note S2

According to the literature [1,2], the SEI layer thickness is supposed to be within ≈ 20 nm; so, if we suppose that a uniform SEI layer coverage was created after cycling, we can affirm that post-test analyses on the investigated electrodes are only related to SEI surface layer, while no information comes from underneath, since the nominal analysis depth for XPS technique is assumed to be ≈ 10 nm. Thus, there is no direct correlation in C1s peak components, before and after tests, for all the samples.



Figure S1. Voltage profiles of the first (left) and second (right) galvanostatic cycle at the current density of 0.05 A g^{-1} for the three carbon-based systems.



Figure S2. Differential capacity of rate performance cycling for Super P (a), C65 (b), and C45 (c) samples. The arrows are meant to point out the potential peak position shift.



Figure S3. Parameter present in the diffusion coefficient equation. Adapted and reproduced with permission from [3].



Figure S4. Potential profiles of Super P (a), C65 (b), and C45 (c) half-cells, taken at different current density values during the rate performance test. The percentage values of the capacity retained per each curve with respect to the 9^{th} cycle curve is shown in rectangular frames.



Figure S5 Nyquist plots obtained from the EIS measurements carried out on cells assembled with Super P (a), C65 (b), and C45 (c) before starting the charge/discharge testing as well as after cycles no. 1, 3, 10, and 20, at the constant specific current of 0.05 A g⁻¹. For all the three, the SEI formation through the cycle determine the decrease of the semicircle diameter, which represent the charge transfer resistance. In particular, C45 system after 20 cycles showed the lowest resistance to the ion transfer through the SEI layer.

Table S1. Relative atomic concentration values obtained for Super P, C65, and C45 samplesthrough XPS analysis.

| SAMPLES | Relative atomic concentration (at.%) | | | | |
|--------------------------|--------------------------------------|------|------|------|-----------------------|
| | C1s | O1s | F1s | K2s | Others |
| C-based electrodes (ref) | | | | | |
| 1- Super P | 90.8 | 0.5 | 8.7 | / | / |
| 2- C65 | 90.2 | 1.3 | 8.1 | / | Si2p: 0.4 |
| 3- C45 | 83.2 | 14.1 | / | / | Na1s: 2.7 |
| Tested electrodes | | | | | |
| 1- Super P in DEC | 25.6 | 35.7 | 15.7 | 19.7 | P2p: 2.3 Na1s: 1.0 |
| 2- C65 in DEC | 29.7 | 31.9 | 11.5 | 24.5 | P2p: 2.0 Na1s: 0.3 |
| 3- C45 in DEC | 30.8 | 30.5 | 13.6 | 22.7 | P2p: 2.2 Na1s: 0.1 |

References

[1] Cheng XB, Zhang R, Zhao CZ, Wei F, Zhang JG, Zhang Q. A review of solid electrolyte interphases on lithium metal anode. *Adv. Sci.* 2015;3:1500213.

[2] An SJ, Li J, Daniel C, et al. The state of understanding of the lithium-ion-battery graphite solid electrolyte interphase (SEI) and its relationship to formation cycling. *Carbon* 2016;105:52-76

[3] Cui S, Wei Y, Liu T, et al. Optimized temperature effect of Li-ion diffusion with layer distance in Li(Ni_xMn_yCo_z)O₂ cathode materials for high performance Li-ion battery. *Adv. Energy Mater.* 2016;6:1501309.