Supporting Information for

Artificial Solid Electrolyte Interphase Formation on Si Nanoparticles through Radiolysis: Importance of the Presence of an Additive

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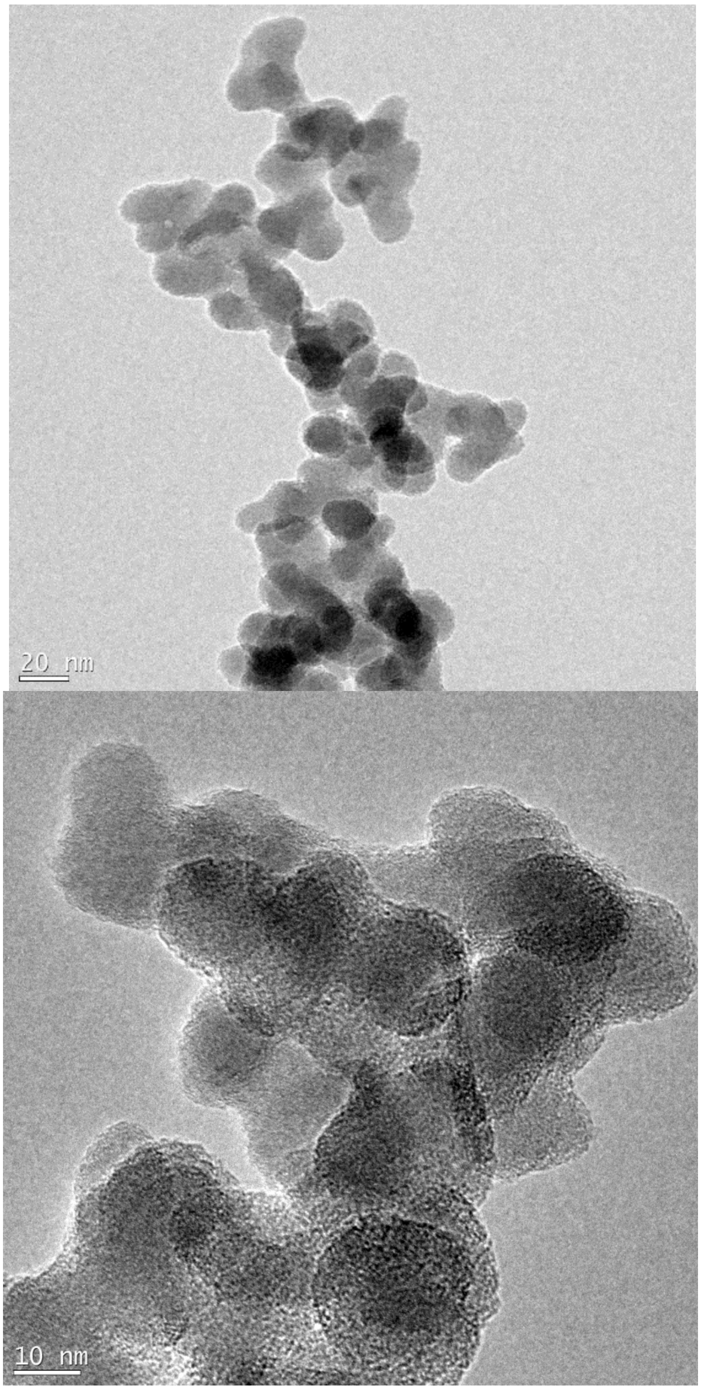

Figure S1. TEM images of the silicon nanoparticles after the thermal treatment and before irradiation.

Ion beam analysis

When a proton interacts with a target material, several physical phenomena occur: protons can be either backscattered or induce X-rays or γ-rays emission. The detection of the backscattered protons leads to the Rutherford Backscattering Spectrometry. RBS measurements can quantify precisely isolated elements or in high quantities. Nevertheless, it is limited in energy resolution for similarly concentrated neighbour elements (for instance, P and Si, for which ΔE = 30 keV). For these reasons, we do not have access to a quantitative composition of the samples. However, an alternative semi-absolute method can be used to compare the evolution of the content of the light element between the non-irradiated and irradiated samples.

In the PIXE and PIGE techniques, the same equation gives the concentration *Cz* of one element (*Z*) using external standard:

$$= [C_Z]_{std} \left[\frac{Q}{S\,A(Z)}\right]_{std} \left[\frac{S\,A(Z)}{Q}\right]_{unknown}$$

where *Q* is the integrated bombarding charge in coulombs, *S* the stopping power of the sample and *A(Z)* the integral of the peak area associated to X-ray or γ-ray transitions observed for an element *Z*.

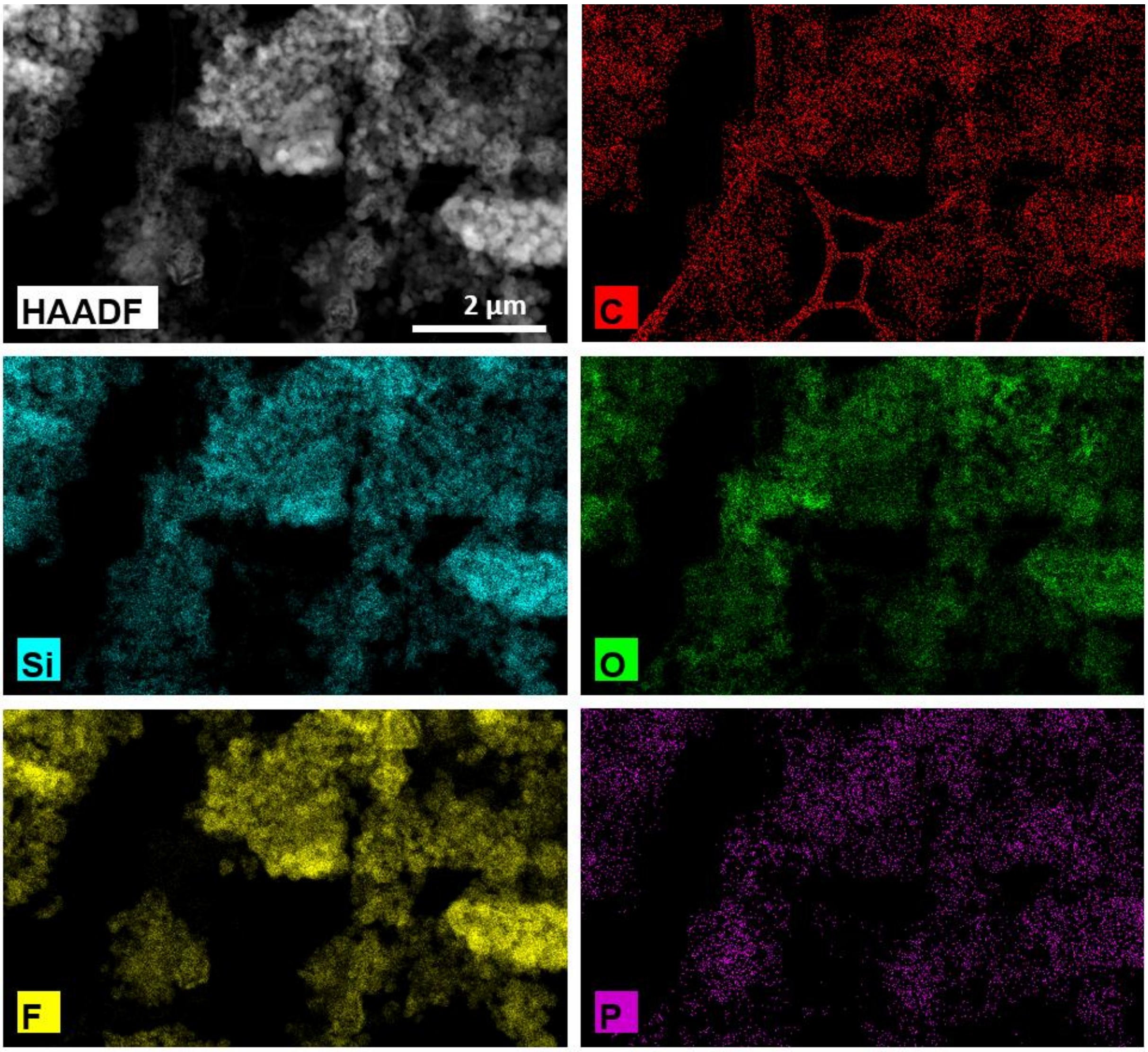

Figure S2. EDX images of the silicon nanoparticles irradiated at 200 kGy in EC/DEC/FEC/$LiPF_6$. Carbon is in red; silicon in pale blue; oxygen in green; fluorine in yellow and phosphorus in purple.

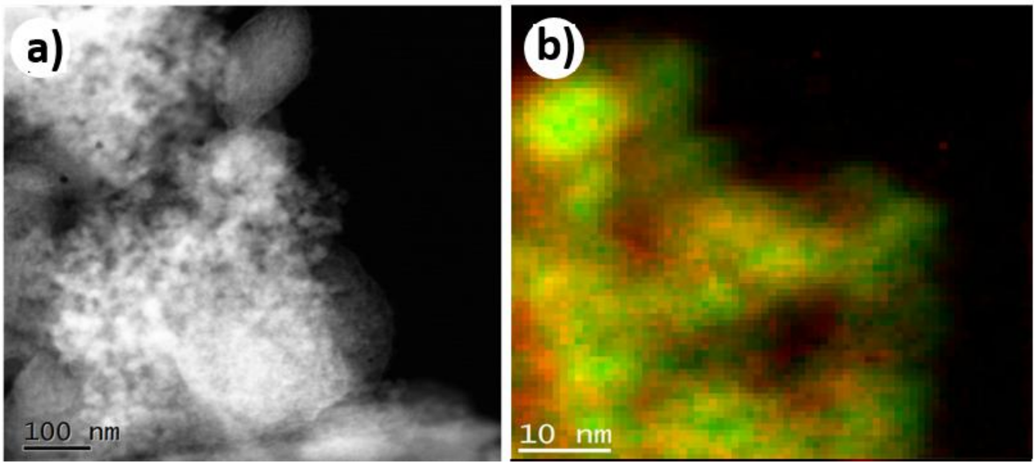

Figure S3. STEM images and EELS mapping of Si nanoparticles after irradiation at 110 kGy in an EC/DEC/FEC/$LiPF_6$ electrolyte. (a) is the same as (b) but with a larger scale to illustrate the morphology of the SEI. The green color stands for silicon and the red one for carbon. The yellow color arises from regions with less carbon amount than the red ones.

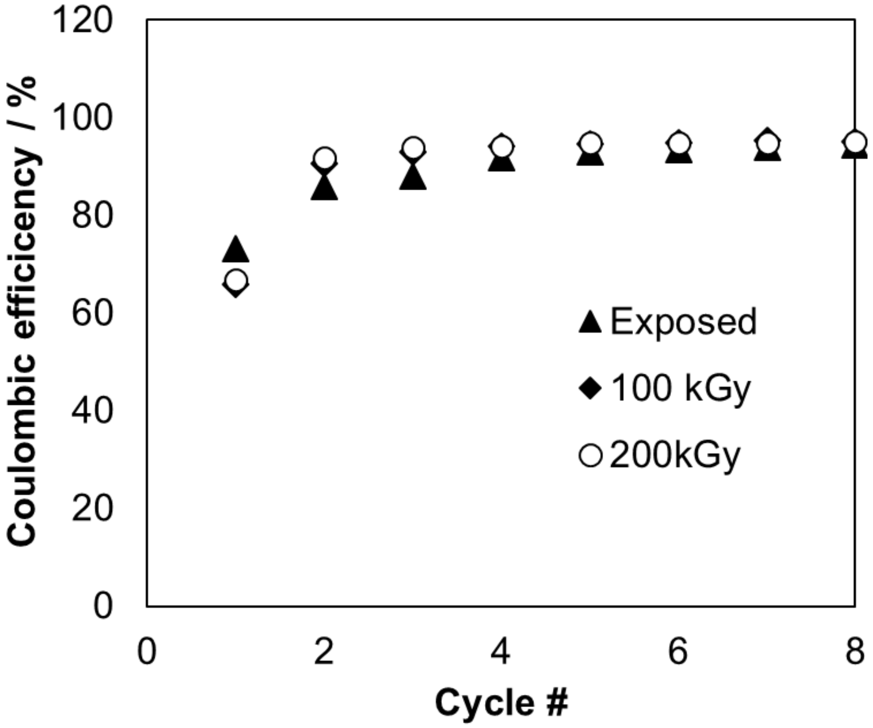

Figure S4. Coulombic efficiency during the first 8 cycles of the exposed and irradiated samples at C/20.