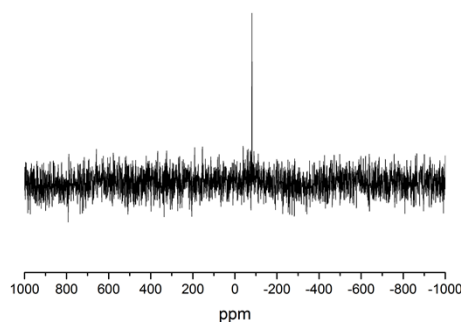
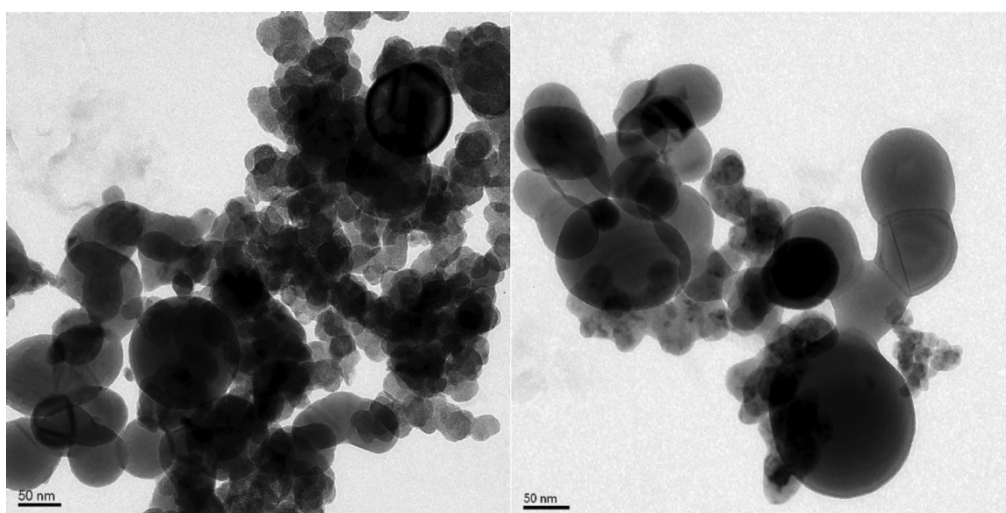


## Electronic Supplementary Information (ESI)



**Fig. S1**  $^1\text{H}$  decoupled  $^{29}\text{Si}$  MAS spectrum of nSi/CA/gellan gum composite at 39.76 MHz and 25 kHz MAS, acquired at a relaxation delay 60 s and 5k transients.

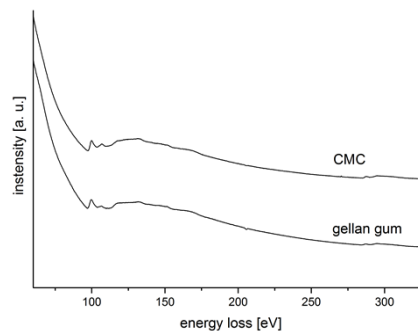


**Fig. S2** TEM measurements of nano silicon particles with Super C65 in CMC binder matrix (left) and gellan binder matrix (right).

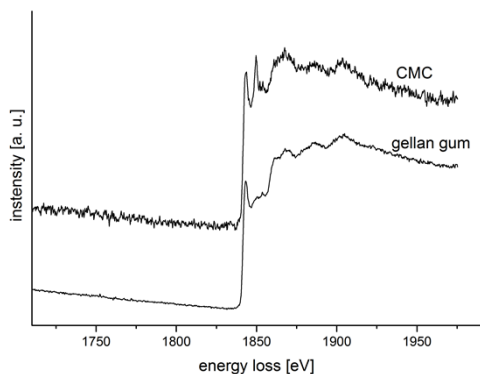
### EELS measurements

The fact that there are differences in the bonding character of the individual binders is strengthened by the EELS measurements where the silicon signals exhibit a difference for each binder system.

Fig. S2 and S3 show the electron loss spectra of the samples with two different binders. The characteristic energy region for silicon shows a difference in signals and intensities, which underline the results of the IR and electrochemical measurements. The spectra elucidate a difference in the way silicon is bonded in the matrix. From this point of view, we can assume that the EELS spectra show a significant difference in the bonding character and / or the ratios of specific bondings, particularly hydrogen bonding and covalently joined, depending of the use of binder.



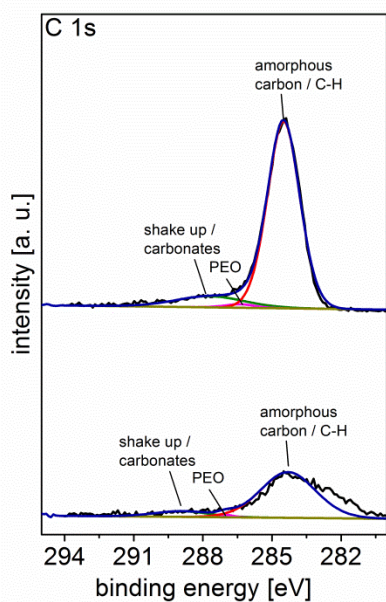
**Fig. S3** EELS measurement of nano silicon particles with Super C65 in a CMC binder matrix (top) and gellan binder matrix (bottom) in an energy region of 60 to 330 eV.



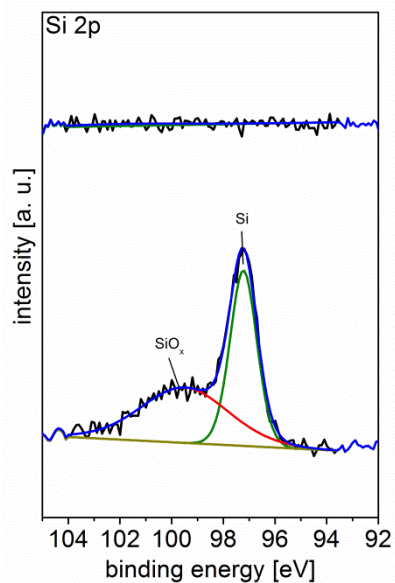
**Fig. S4** EELS measurement of nano silicon particles with Super C65 in a CMC binder matrix (top) and gellan binder matrix (bottom) in an energy region of 1700 to 2000 eV.

### XPS spectra

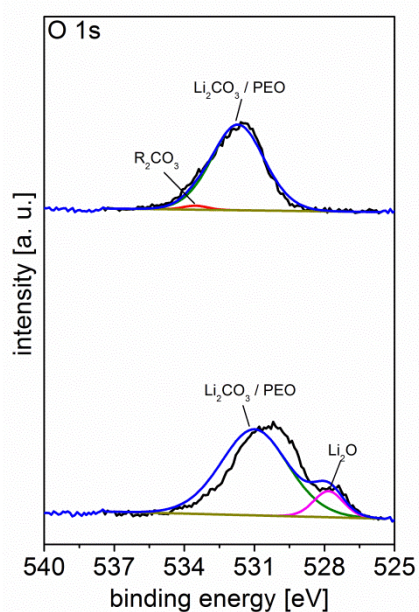
In the following the XPS spectra of a cycled graphite free nSi/CA/gellan gum electrode before and after the application of the sputter depth profile (SDP) are shown.



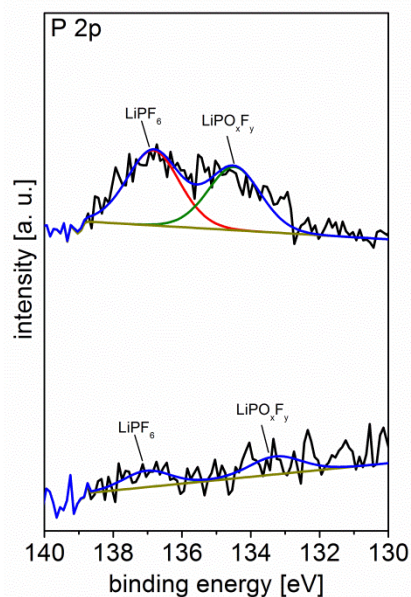
**Fig. S5** XPS C 1s spectrum of the cycled anode before SDP (top) and after SDP (bottom). The plots indicate the different components of the SEI layer. The corresponding component is labelled in the Figure.



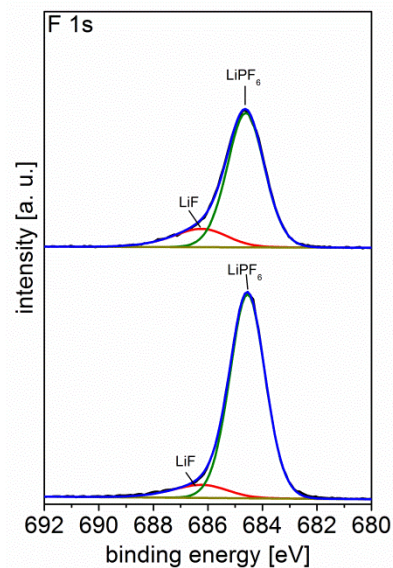
**Fig. S6** XPS Si 2p spectrum of the cycled anode before SDP (top) and after SDP (bottom). The plots indicate the different components of the SEI layer. The corresponding component is labelled in the Figure.



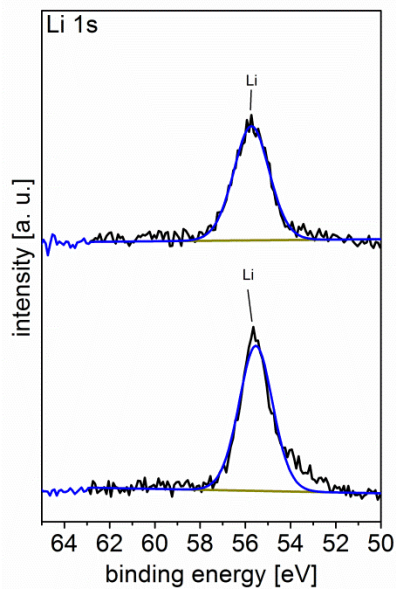
**Fig. S7** XPS O 1s spectrum of the cycled anode before SDP (top) and after SDP (bottom). The plots indicate the different components of the SEI layer. The corresponding component is labelled in the Figure.



**Fig. S8** XPS P 2s spectrum of the cycled anode before SDP (top) and after SDP (bottom). The plots indicate the different components of the SEI layer. The corresponding component is labelled in the Figure.



**Fig. S10** XPS F 1s spectrum of the cycled anode before SDP (top) and after SDP (bottom). The plots indicate the different components of the SEI layer. The corresponding component is labelled in the Figure.



**Fig. S9** XPS Li 1s spectrum of the cycled anode before SDP (top) and after SDP (bottom). The plots indicate the different components of the SEI layer. The corresponding component is labelled in the Figure.