## **Electronic Supplementary Information**

## Nanoscale, surface-confined phase separation by electron beam induced oxidation

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**Figure S1:** HAADF image of a TEM lamella of EBC cured Co<sub>2</sub>SiCO FEBID grown at 6.3 nA and 5 kV. The EBC is carried out with an electron dose of 1330 nC· $\mu$ m<sup>-2</sup> using the same beam parameters. The EDX Pt<sub>L</sub>, Nb<sub>L</sub>, Co<sub>K</sub>, Si<sub>K</sub> and O<sub>K</sub> maps of the same area.



**Figure S2:** EDX Co<sub>K</sub>, Si<sub>K</sub> and O<sub>K</sub> maps as well as a HAADF image of a TEM lamella showing an oxidative EBC treated Co<sub>2</sub>SiCO FEBID deposited at 6.3 nA and 5 kV. EBC was carried out with an electron dose of 1330 nC· $\mu$ m<sup>-2</sup>. The larger lower resolution HAADF image shows the homogeneous phase separation at the surface. The graphical representation shows a line scan EDX from the area shown above. The substrate in this case is copper.



**Figure S3:** EDX Co<sub>K</sub>, Si<sub>K</sub> and O<sub>K</sub> maps as well as a HAADF image of a TEM lamella showing an oxidative EBC treated Co<sub>2</sub>SiCO FEBID deposited at 6.3 nA and 5 kV on a micromembrane heater. EBC was carried out with an electron dose of 1330 nC· $\mu$ m<sup>-2</sup>. The graphical representation of an EDX line scan accumulating the signal intensity of the whole area shown above and the atomic fractions neglecting the carbon content in the deposit (section of the FEBID deposit up to the maximum signal intensity of phase separated Co).



**Figure S4:** Illustration of the crystallinity of forming CoO nanoparticles at the surface in the oxidative EBC without additional Nb-based coating (upper images) and with 2 nm Nb-based coating (lower image).



**Figure S5.** SEM image showing Co<sub>2</sub>SiCO NWs and HAADF image illustrating the bending after the EBC curing using a dose of ~4000 nC· $\mu$ m<sup>-2</sup> and ~6000 nC· $\mu$ m<sup>-2</sup>.



**Figure S6.** The graphical representation of an EDX line scan of an area illustrated in the overlay of EDX Si and Co maps.



PtCx

**Figure S7:** 4-point contacts to an 80 nm thick, oxidative EBC-treated Co<sub>2</sub>SiCO pad. Prior to FEBID of the  $PtC_x$  leads, protective, electrically insulating  $NbN_xC_y$  FEBID layers were deposited on the edges to ensure only top contact.  $PtC_x$  was also treated with EBC after deposition to increase the conductivity. A scheme of the different materials completes the image. Electron transport could occur via the Co-based top layer or via the Co<sub>2</sub>SiCO bulk, but the CoO/SiO<sub>2</sub> appears to be electronically insulating.