Supplementary information: Steering interzeolite conversion with alkali metal cations: lithium maximizes Al proximity in SSZ-13 zeolite genesis

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S1 Methodology on the calculation of the OSDA/Al ratio.

TGA was used to determine the mass decrease between 300°C and 800°C, the percentage obtained this way was corrected with the weight loss below 300°C, which is mainly due to water. Using the following formula, weight percentage of OSDA was determined.

$$\frac{\Delta weight(300°C-800°C)}{1-\Delta weight(50°C-300°C)} = weight\%(OSDA) \qquad (1)$$

Next, the $\frac{OSDA}{Al}$ ratio was determined using equation 2. The lithium was not taken into acount in this calculation due to its low prevalence and molecular weight, thus only giving a minor contribution to the final value. Also the average molar weight of a TO_2 atom was chosen to be 60 g/mol as in the final product either SiO_2 or AlO_2H is present. The $\frac{Si}{Al}$ was determined using ICP-AES.

$$\frac{\frac{weight\%(OSDA)}{194g/mol} / \frac{100-weight\%(OSDA)}{60g/mol}}{(\frac{Si}{Al}+1)^{-1}} = \frac{OSDA}{Al} \qquad (2)$$

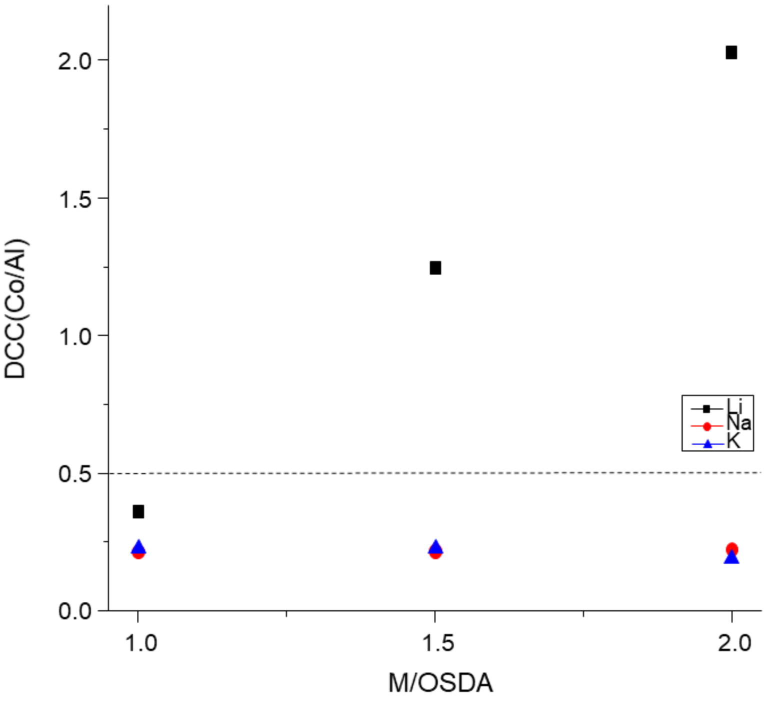

Figure S1: Zoom out of the DCC plot shown in figure 1C between M/OSDA 1 and 2. The dashed line represents the theoretical maximum for the DCC if only paired Al is responsible for Co^{2+} uptake.

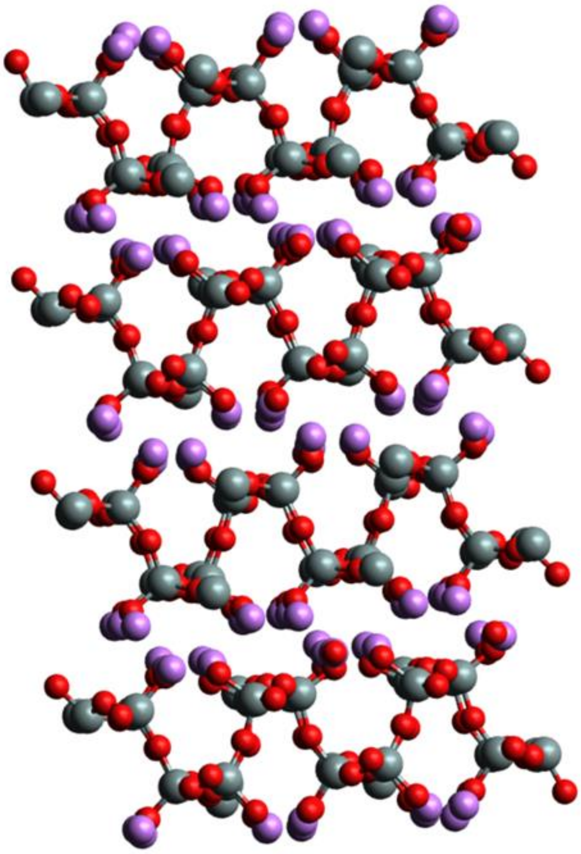

Figure S2: The structure of $Li_2Si_2O_5$, a layered phyllosilicate (silicon=grey, oxygen=red and lithium= pink) with Li^+ cations located in the intrplanar void balancing the negative charge of siloxane bonds.[1]

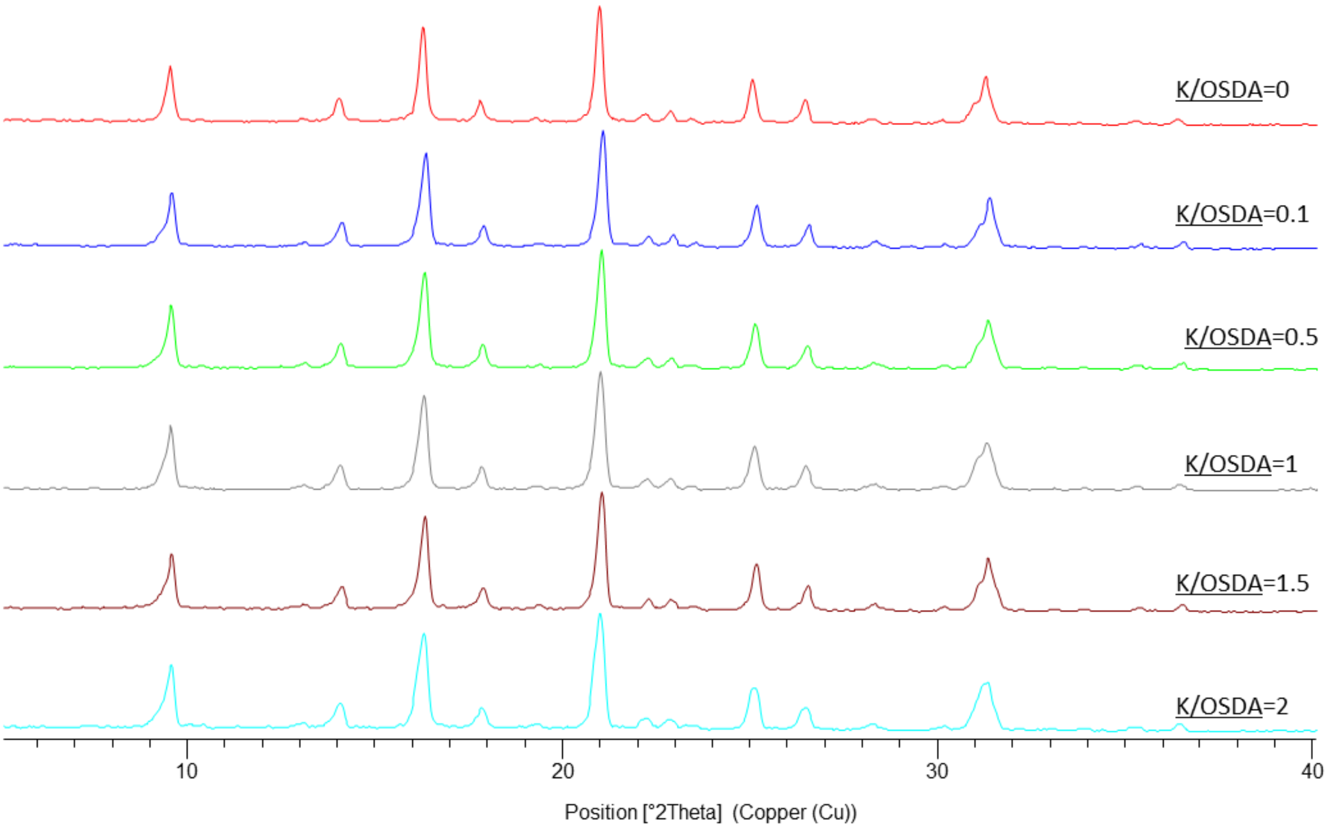

Figure S3: XRD spectra of SSZ-13 zeolites synthesised with different concentrations of K^+present in the mother liquor. All of them show pure and highly crystalline CHA zeolite.

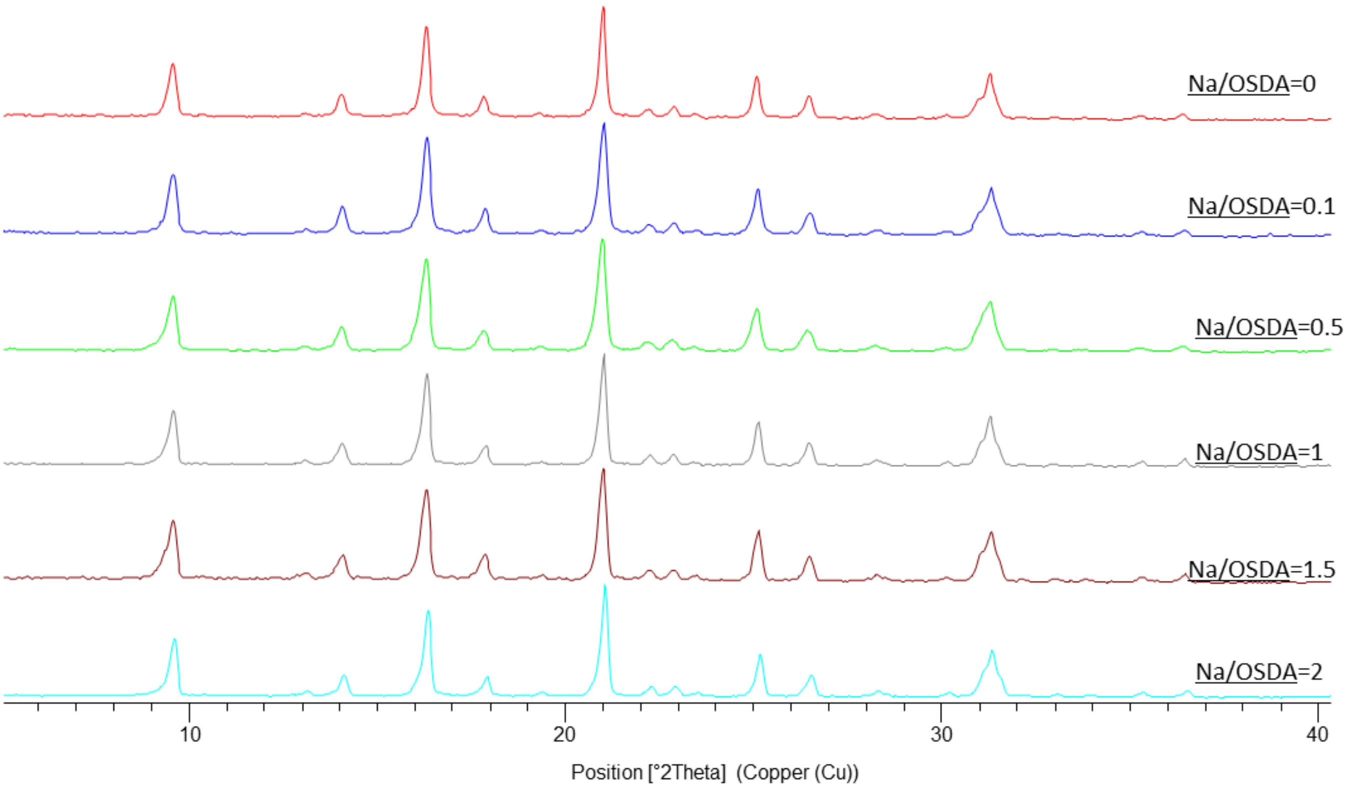

Figure S4: XRD spectra of SSZ-13 zeolites synthesised with different concentrations of Na+present in the mother liquor. All of them show pure and highly crystalline CHA zeolite.

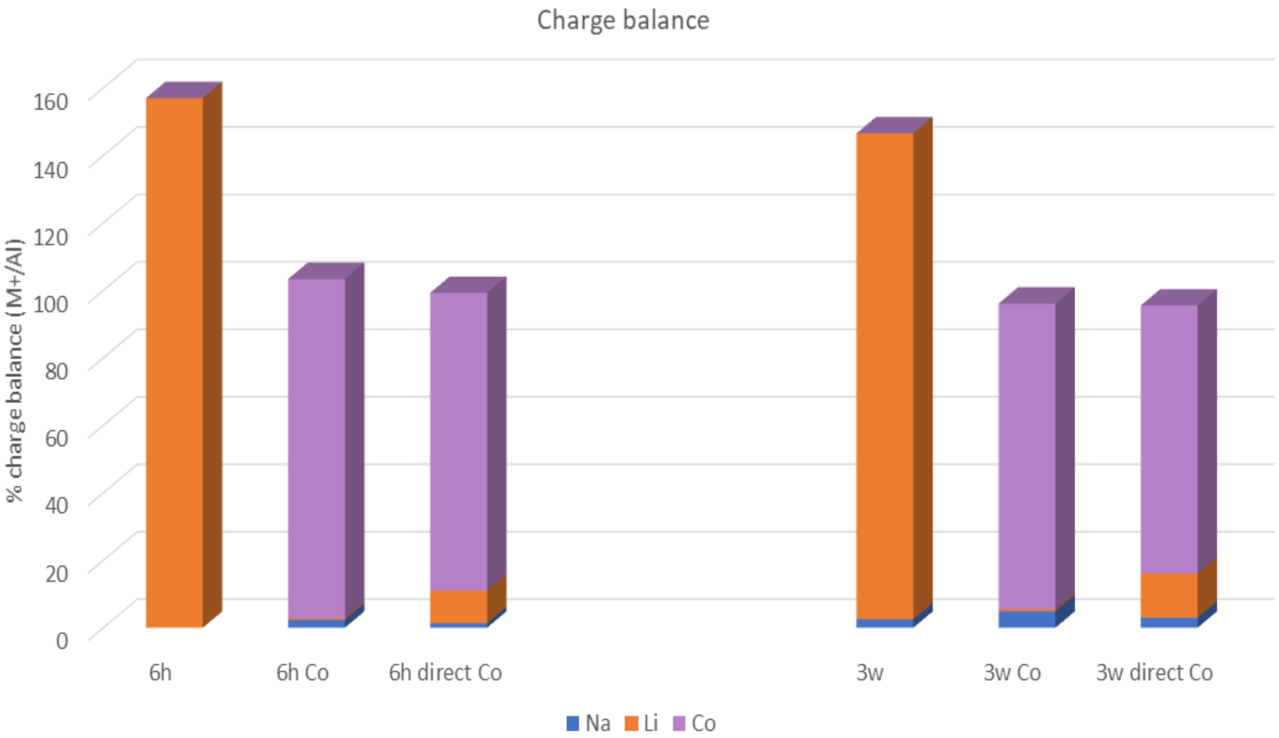

Figure S5: Charge balance of a calcined zeolite synthesised in a Li/OSDA=0.5 after 6 hours (6h) and 3 weeks (3w) of hydrothermal treatment. The charge balance, as represented by a cation/Al ratio was taken right after calcination (6h-3w), after a normal Co-exchange procedure (6h Co-3w Co) and finally after a Cobalt exchange procedure which skips the 3x NaCl exchange (6h direct Co-3w direct Co) (See Methods for more info). From this it was concluded that the Li^+ cations themselves are not interacting with Co^{2+} cations resulting in high DCC. The total charge balance being close to 100% in most cases also rules out the role of defects. Small amounts of Na are present in all samples due to the analysis procedure.

Table S1: Micropore volume of some selected samples of the concentration series of Li^+.

Li/OSDA	**micropore volume (cm^3/g)**
0	0.32
0.1	0.31
0.2	0.32
0.5	0.32
1	0.30
1.5	0.23
2	0.20

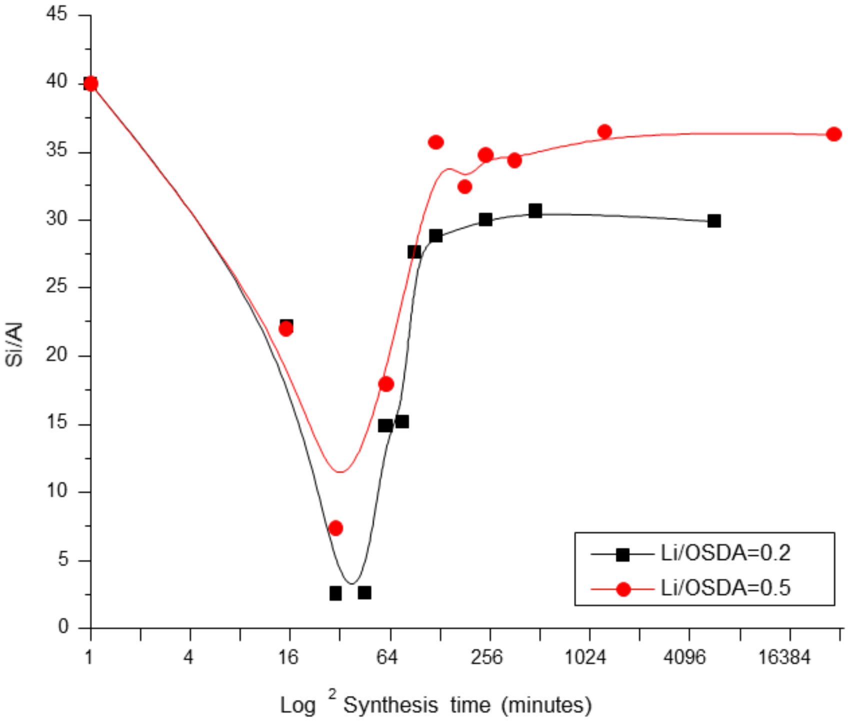

Figure S6: Si/Al ratio of a Li/OSDA=0.2 and 0.5 synthesis throughout time. Notice the similar course of actions taken by both. Lines are added as a guide to the eye.

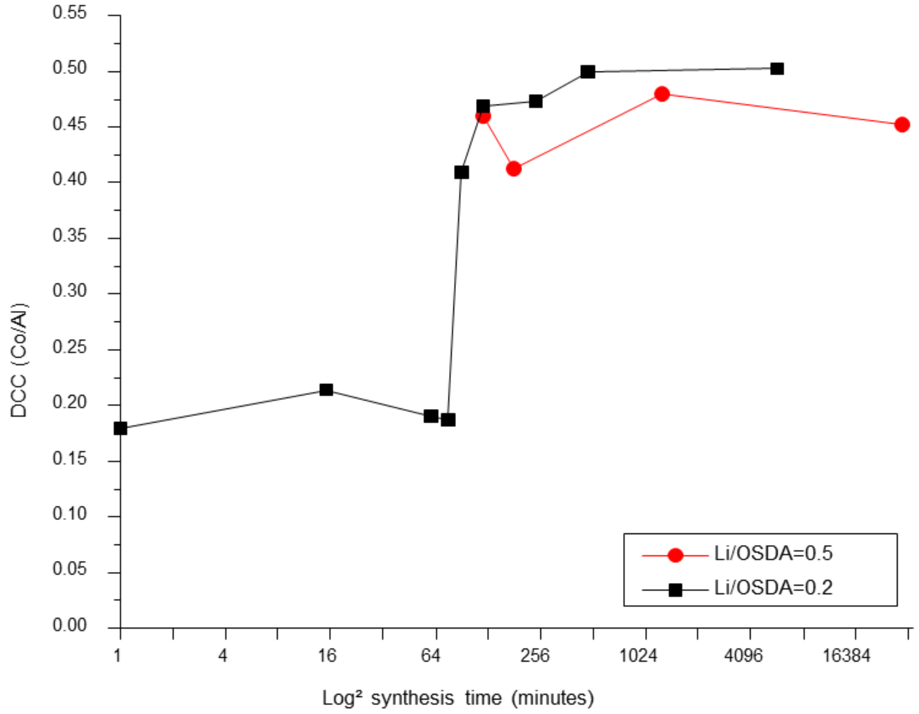

Figure S7: Dcc as a function of synthesis time for a synthesis with Li/OSDA=0.5 and Li/OSDA=0.2, showing no reduction in DCC over time as been observed earlier.

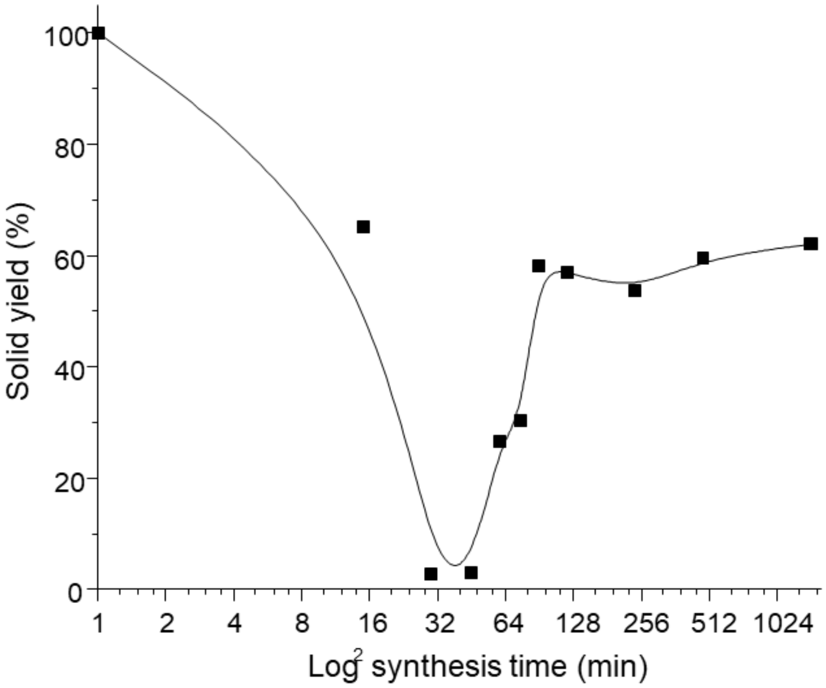

Figure S8: Solid yield as a function of time of the Li/OSDA=0.2 system. Lines are added as a guide to the eye.

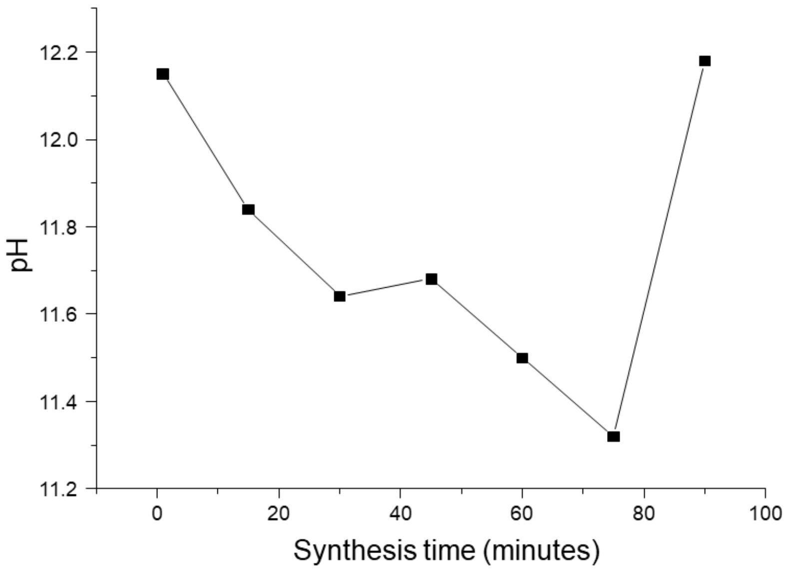

Figure S9: pH of a synthesis containing a Li/OSDA ratio of 0.2 in function of synthesis time.

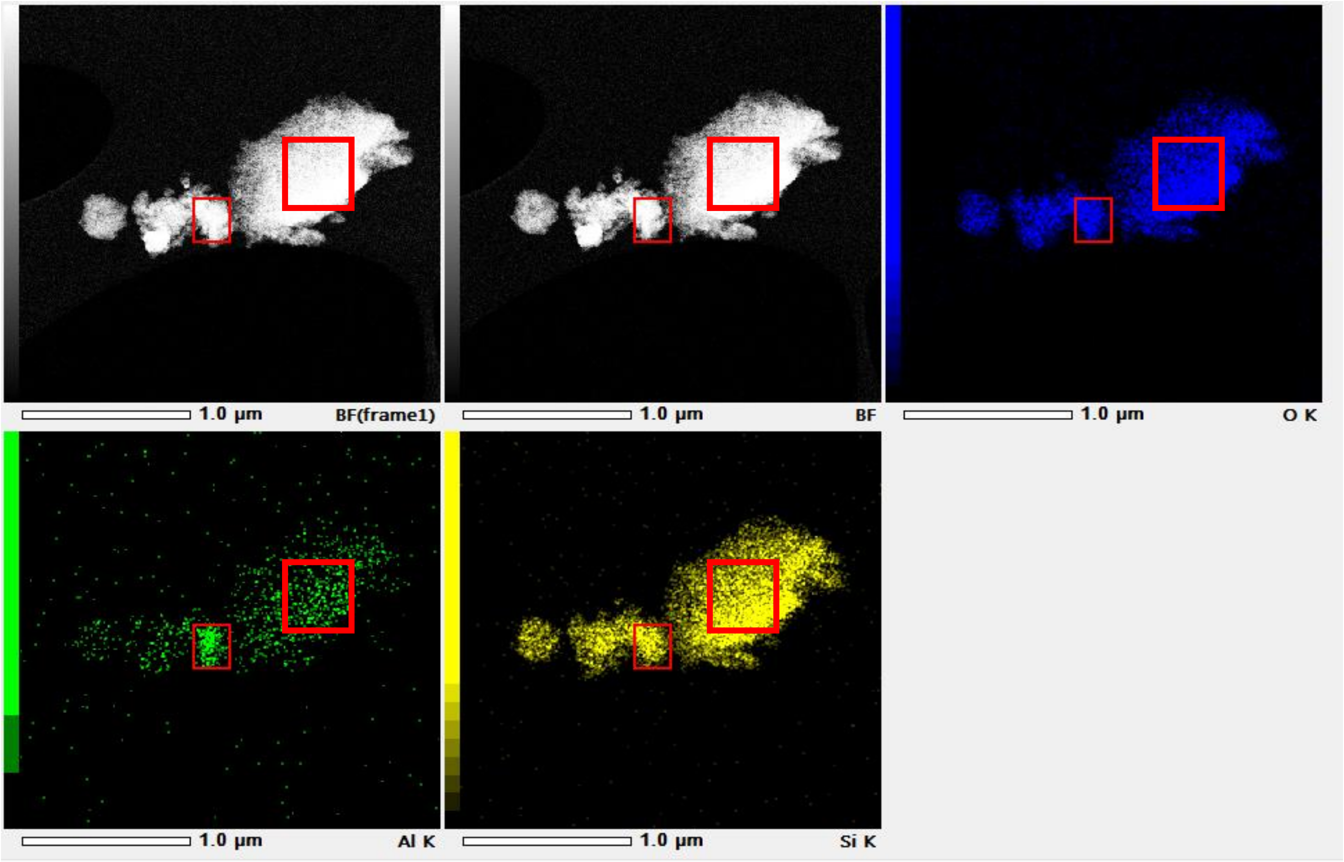

Figure S10: EDX analysis of the particles displayed in fig 4C-D after 75 minutes of synthesis. A more aluminous particle can be observed in the small red square. The two squares represent the zones used for elemental mapping and determination of the Si/Al ratios in figure 5C.

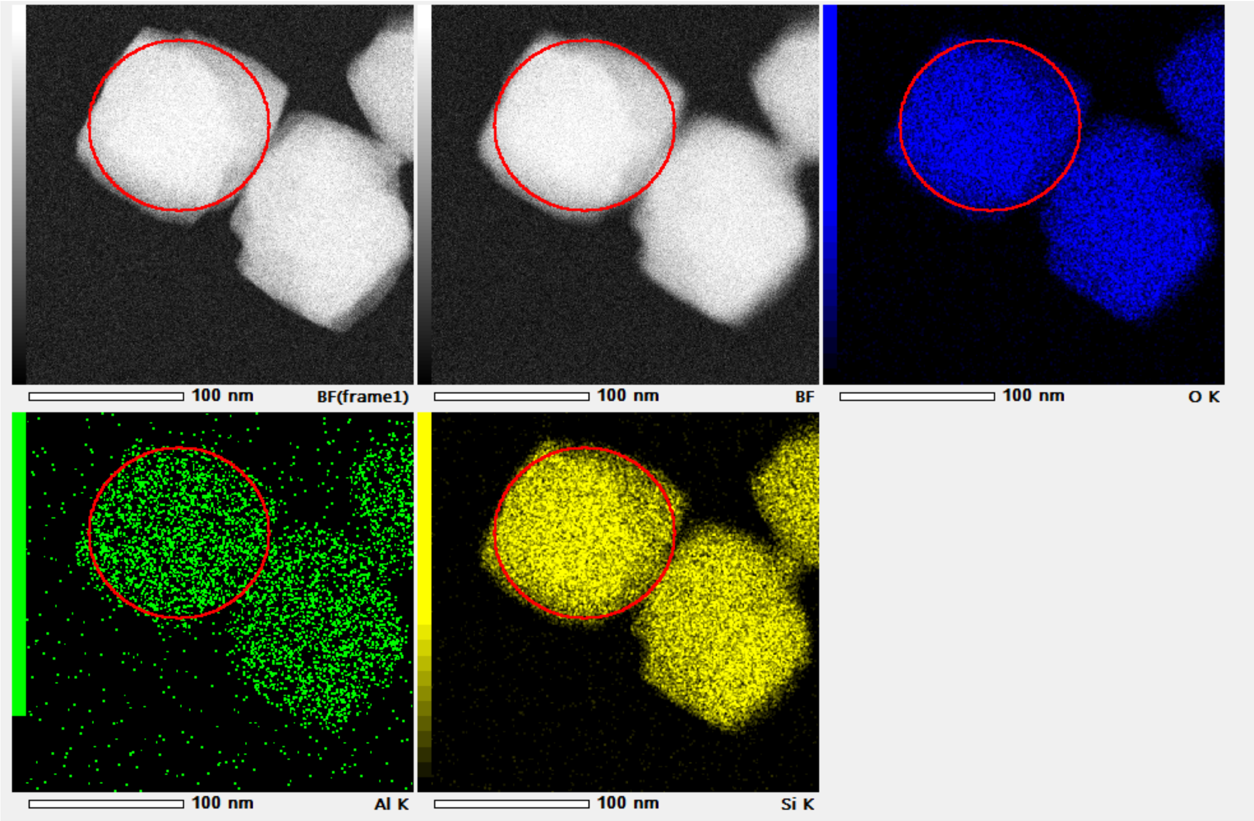

Figure S11: EDX analysis of a particle that underwent 120 min of synthesis. No sign of elemental zoning can be seen. The circle represents the zone used for elemental mapping and determination of the Si/Al ratio in figure 5E.

References

1. Lodesani F., Menziani M. C., Maeda K., Takato Y., Urata S., Pedone A., *Disclosing crystal nucleation mechanism in lithium disilicate glass through molecular dynamics simulations and free-energy calculations*, Sci Rep 10, 17867 (2020). https://doi.org/10.1038/s41598-020-74764-9