



**Fig. 2.** Long-range XPS spectra for samples CGs and CGDs (low resolution, PE = 50 eV) in A. Short-range spectra for (B) Si2p, (C) Na1s, (D) Ca2p, and (E) O1s. Spectra are not normalized in intensity.

CGD1, CG2, and CG3. We can deduce that this is related to the formation of NaCl and CaCl<sub>2</sub> aggregates embedded into the silicate matrix of CGs and CGDs. C1s core level analysis is reported in *SI Appendix, Fig. S2A*. The energy region where the C1s peak is expected is superimposed on the complex Na Auger emission. However, comparing CGs and CGDs through BE alignment, we highlight some differences between these two surfaces. Two CGDs show a more pronounced lower binding energy shoulder where the C1s core level should be located (“expected C1s range” zone) and an increased level of carbon in these samples can be hypothesized. From this analysis, we conclude that the decanol is incorporated in the CGDs structures albeit in various ways and with different kinetics, varying its quantity in different CGD crystal regions.

In light of these observations, we chose to analyze the CG and CGD systems incubated also over a longer period (1 mo). We used a technique enabling a more in-depth surface analysis: scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM EDXS). Relevant results are discussed here and corresponding figures can be found in

(*SI Appendix, Fig. S3*). Inorganic mineral surfaces created after 1 d of incubation with CaCl<sub>2</sub> with or without decanol show an analogous overall composition, similar to the one observed through XPS. Inorganic mineral surfaces generated after 1 mo of incubation without or with decanol also show an overall similar composition with an increased amount of Cl and Ca. In the case of the sample incubated without decanol, a slight silicon presence is noted that is instead absent in the case of decanol treatment. We presume a higher substitution of chlorine ions to silicate and an increased thickness of the salt crystals present on the surface in the case of the CGD system. The carbon quantity after 1 mo is higher in the decanol-treated sample, and this could be related to decanol integration inside the chemical garden structures.

We then analyzed specific surface structures of each system after 1 d and 1 mo of growth. Images are reported in *SI Appendix, Fig. S3*. The surface of the CG system after a 1-d incubation time shows a smooth layer of Na<sub>2</sub>SiO<sub>3</sub> with NaCl crystals. The surface of the CGD system (also 1 d incubation time) shows instead a smooth layer of Na<sub>2</sub>SiO<sub>3</sub> with some Na<sub>2</sub>CO<sub>3</sub>, NaCl, and