

Supplemental Material

The hydrothermal alkaline alteration of potassium feldspar: a nanometer-scale investigation of the orthoclase interface

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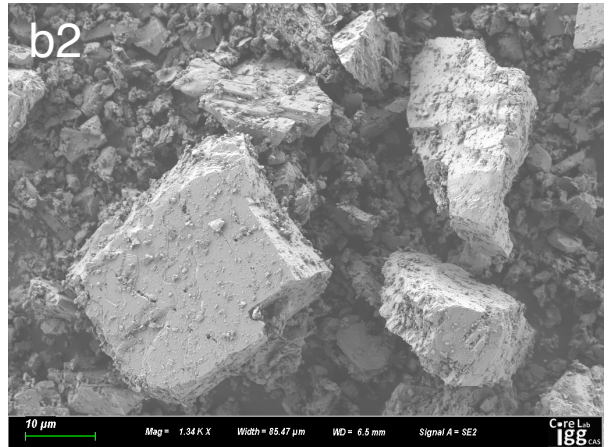
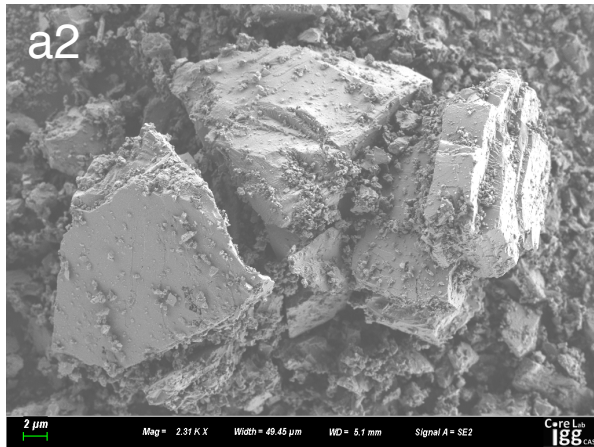
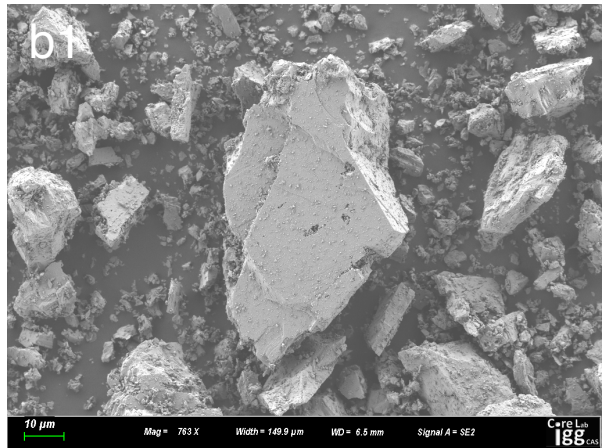
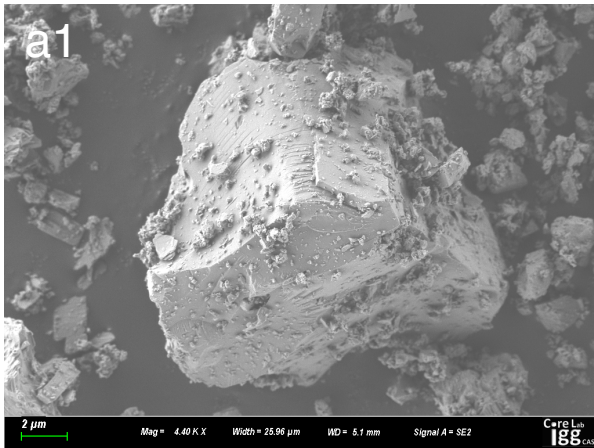
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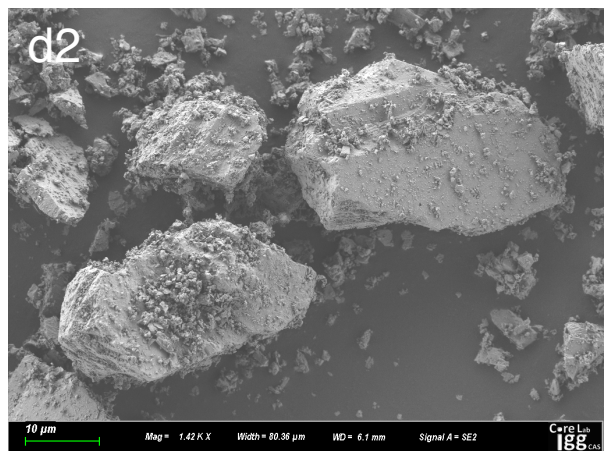
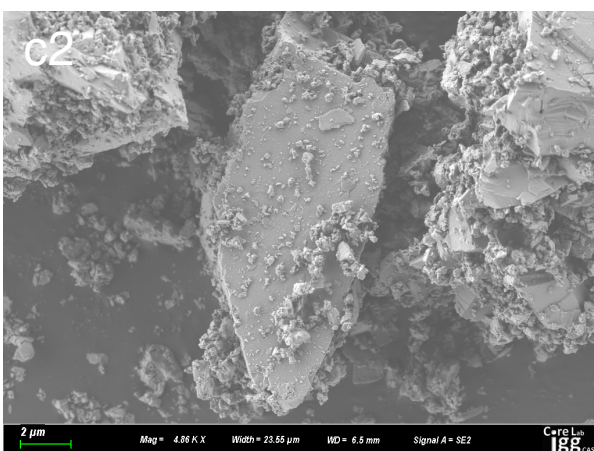
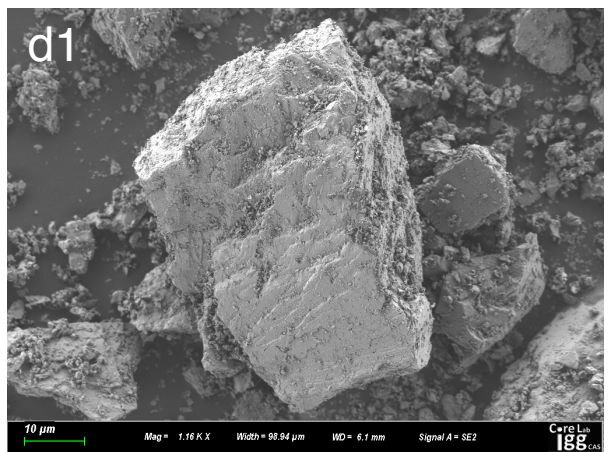
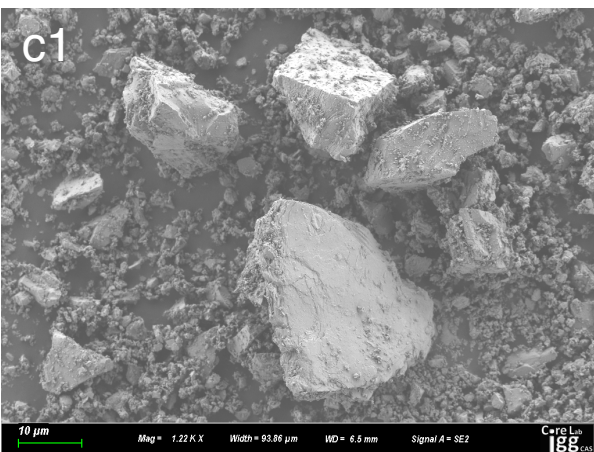
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Figures S1a-d



CSY-1

CSY-2



CSY-3

CHG-2

Figure S1e

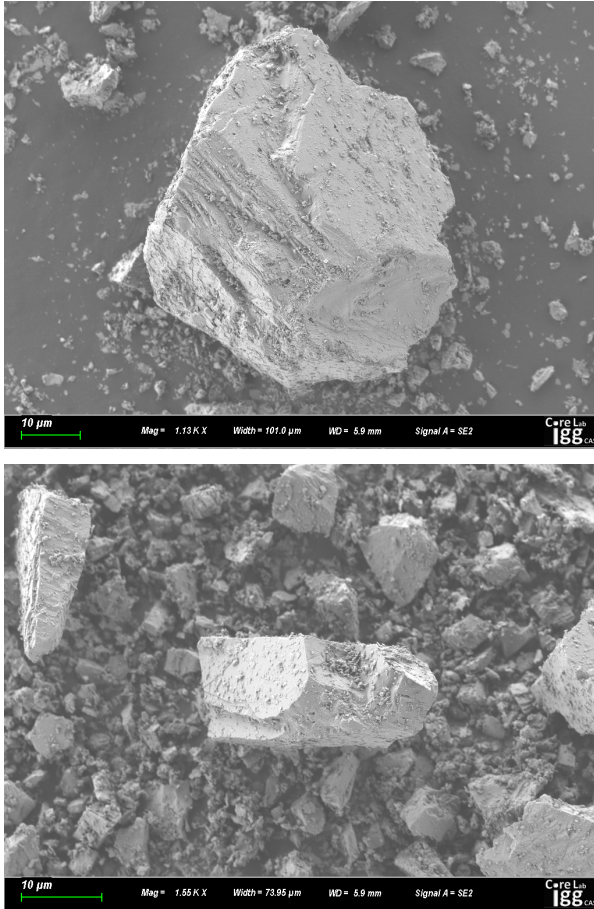


Fig. S1a-e. Orthoclase grains before alteration- two representative secondary electron SEM images of each sample are shown. Micrographs obtained with an SEM Merlin instrument operating at 1.2 kV and beam current of 500 pA.

CHG-12

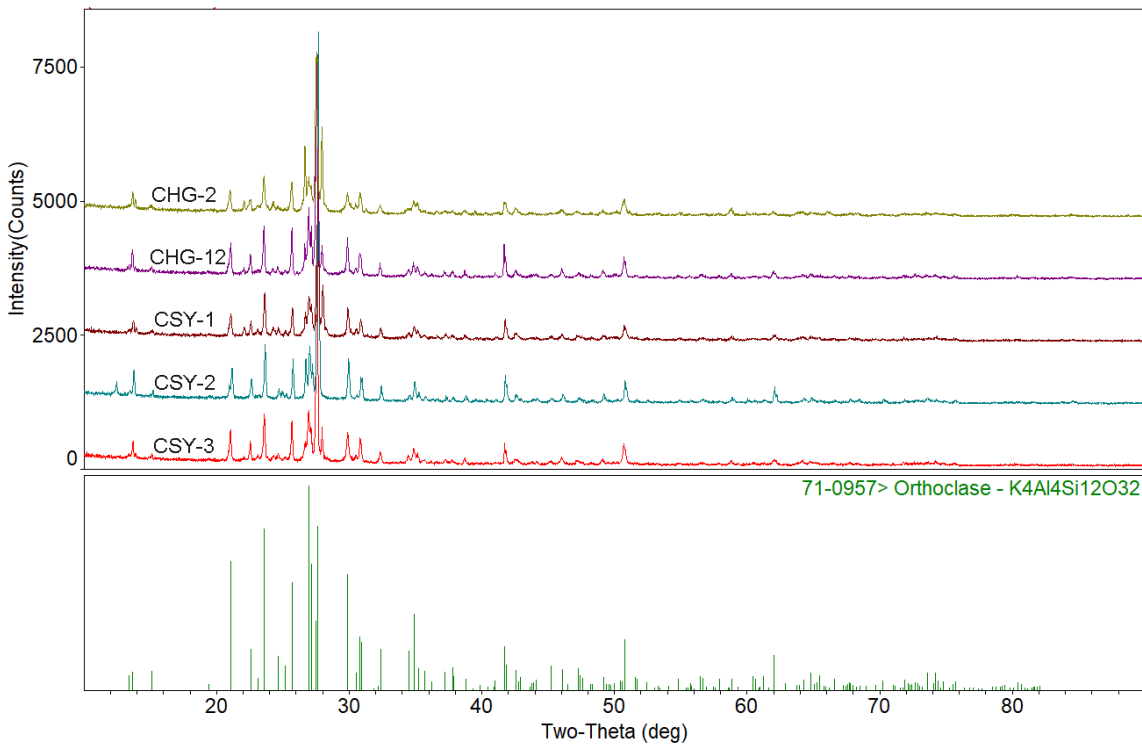


Fig. S2. Enlarged view of powder X-ray diffraction (XRD) patterns corresponding to the 5 unaltered orthoclase samples. The bottom figure is a standard reference XRD pattern file for orthoclase.

CHG-2

Hist 1

Lambda 1.5406 A, L-S cycle 222

Obsd. and Diff. Profiles

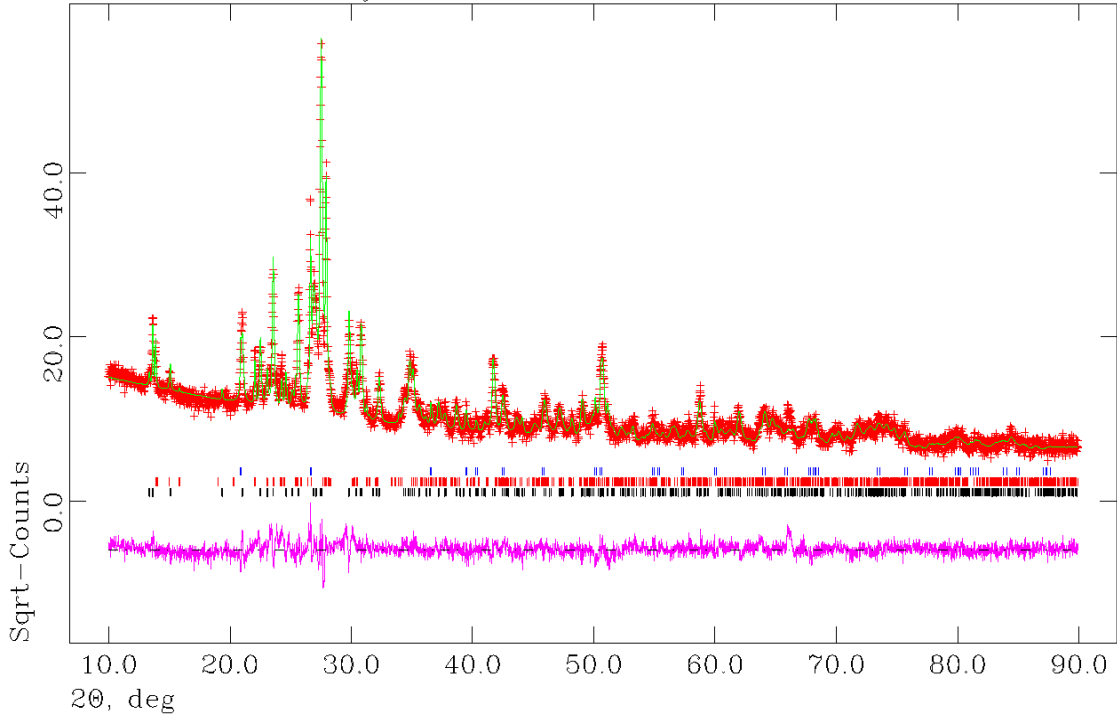
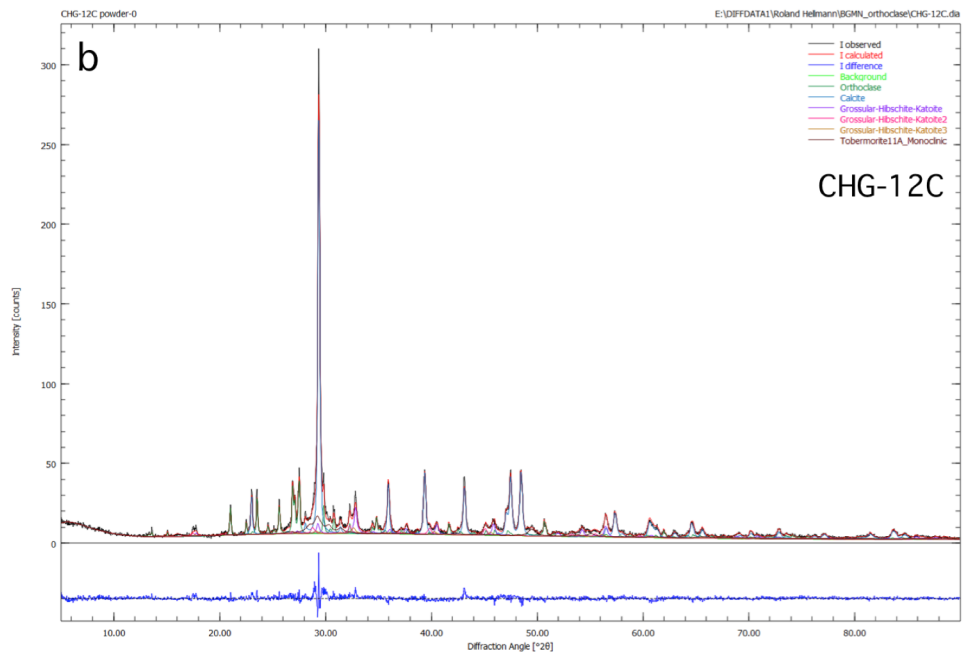
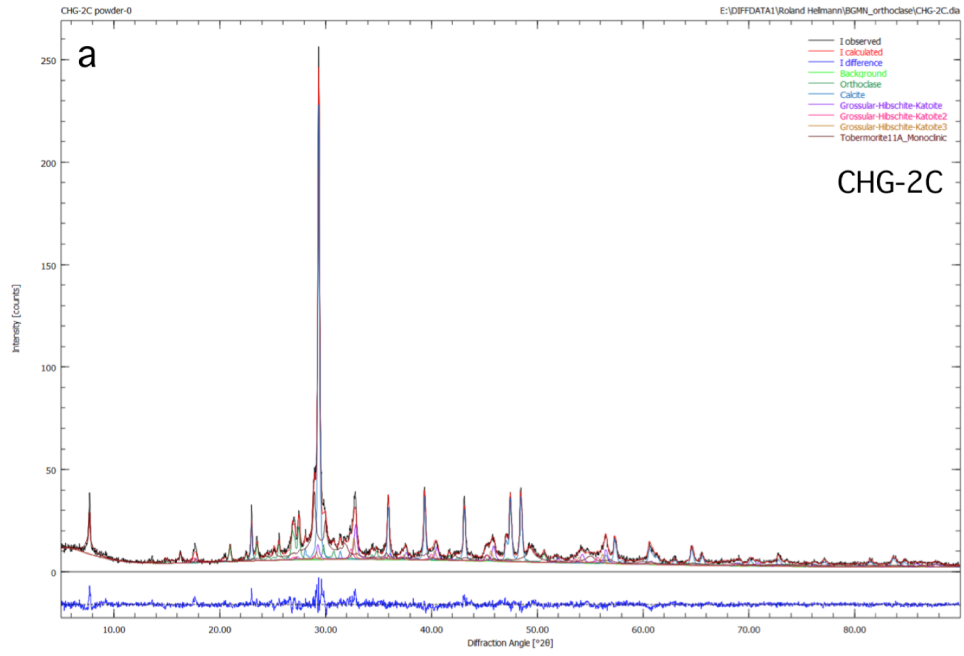


Fig. S3. Rietveld fit of the sample CHG-2. The red crosses are the measured data, and the solid light green line is the calculated diffraction pattern. Below this, the vertical tics (from top to bottom, i.e., blue, red, black) indicate the positions of allowed $K\alpha_1$ and $K\alpha_2$ reflections of orthoclase, albite, and quartz, respectively (the latter two phases are considered as impurities). The rose-colored pattern at the bottom represents the residuals (difference between measured and calculated diffraction pattern).

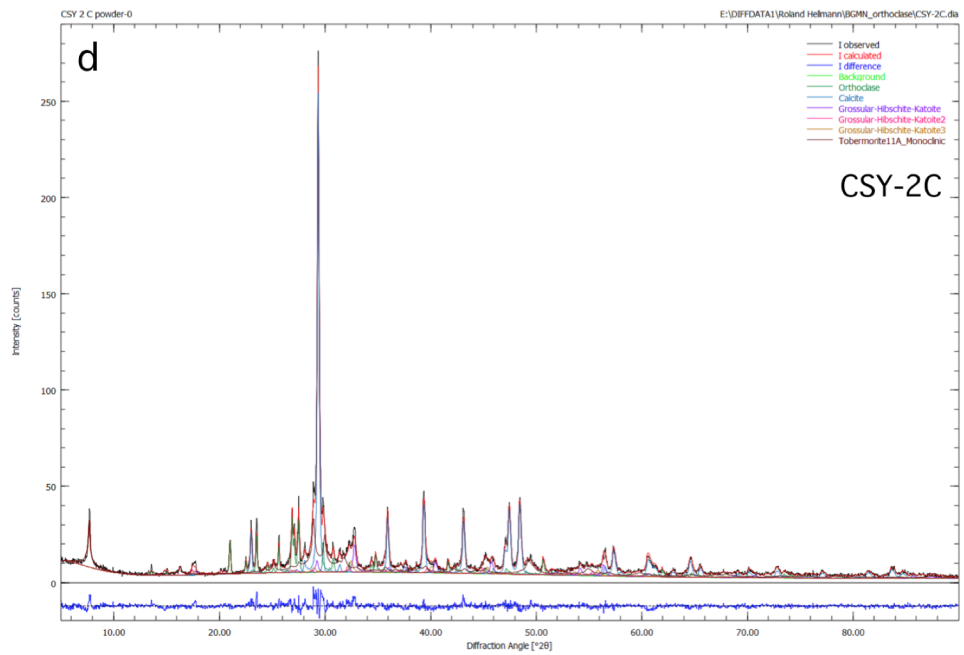
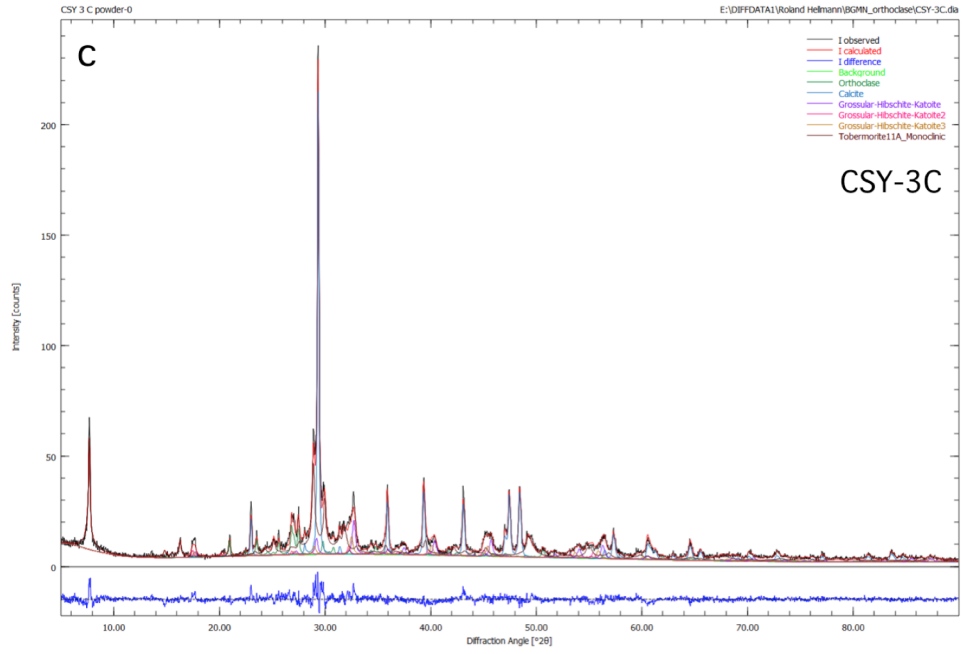
Note S1.

As an added comment for the interested reader, a new nomenclature by the CNMNC of the IMA redefines the garnet supergroup (Grew et al. 2013), such that all have a common formula: $\{X_3\}[Y_2](Z_3)\varphi_{12}$, where X , Y , and Z refer to dodecahedral, octahedral, and tetrahedral sites, respectively, and φ denotes O, OH or F. Under this new classification scheme, hibschite is now referred to as grossular, and the former grossular becomes anhydrous grossular. However, to facilitate comparisons with previous studies, we prefer to use the former, traditional terminology.

Figures S4a, b



Figures S4c, d



Figures S4e,f

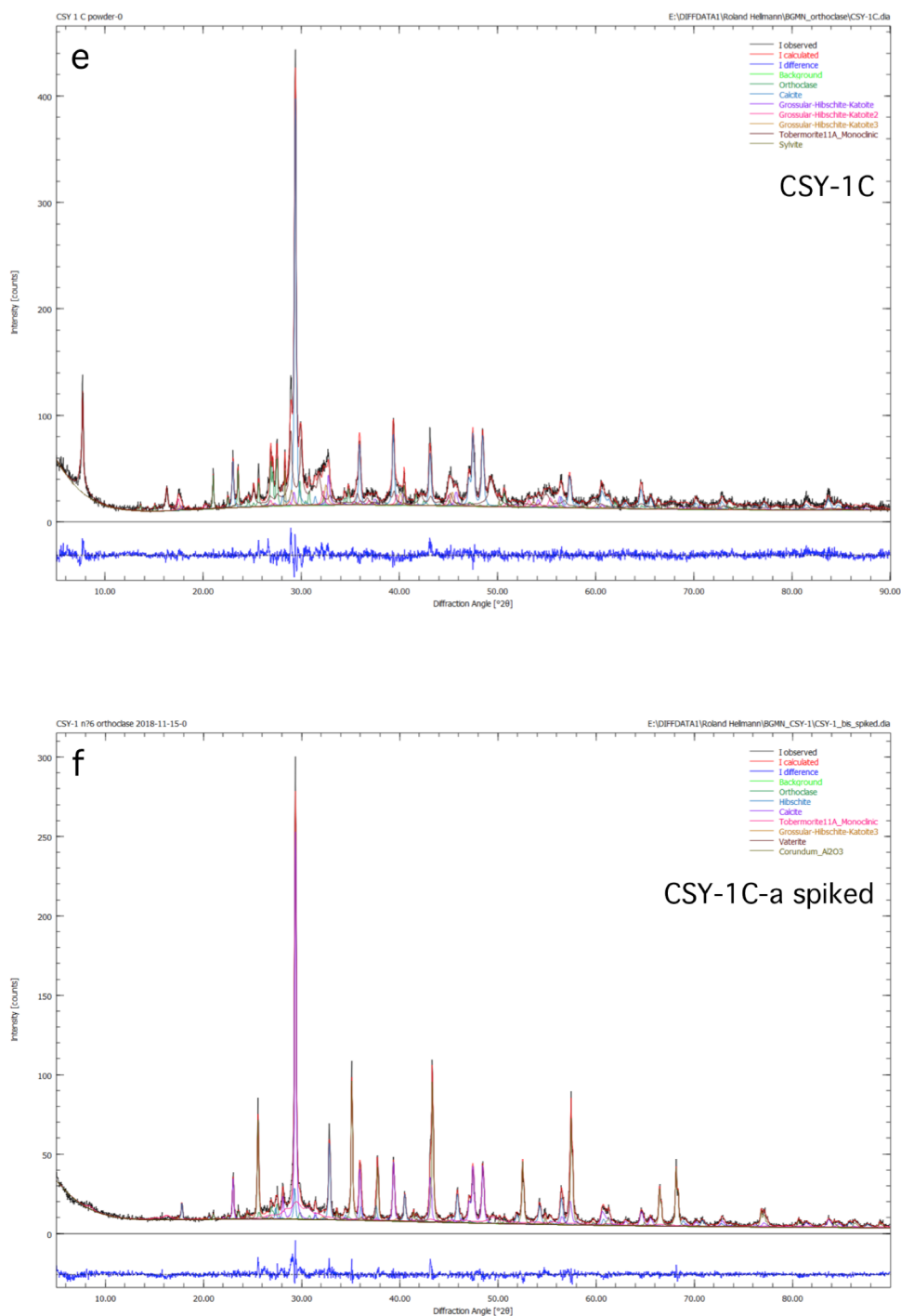
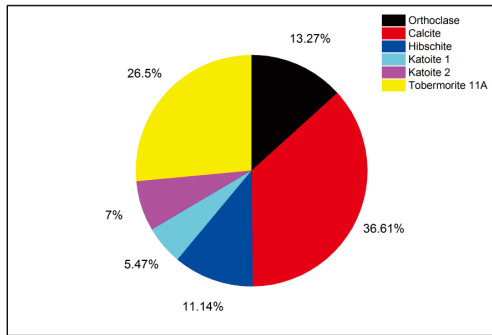
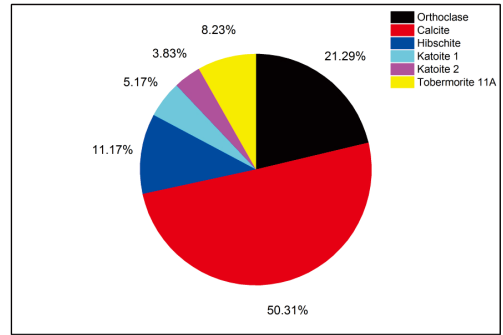


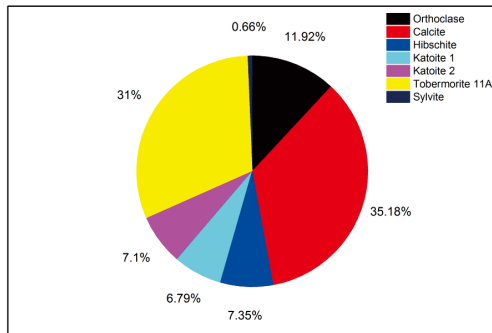
Fig. S4a-f. Fitted X-ray powder diffraction-Rietveld refinement patterns, including residuals, for the altered orthoclase samples. The diffraction patterns include the 5 original orthoclase samples, as well as sample CSY-1C-a. The latter sample is a replicate, and was produced by alteration of raw sample CSY-1, but without the 105 °C heat treatment step- see main text. The reaction product was then spiked with the addition of a precise amount of Al_2O_3 (corundum) as an internal crystalline standard. The diffraction pattern of CSY-1C-a is virtually identical to that of CSY-1C, except that the former doesn't have a well-defined and sharp tobermorite 11Å peak ($8^\circ 2\theta$). We interpret that the small, rounded peak in CSY-1C-a is evidence for nanocrystalline or poorly crystalline tobermorite.



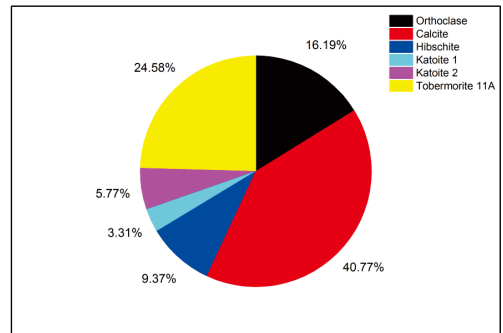
CHG-2C



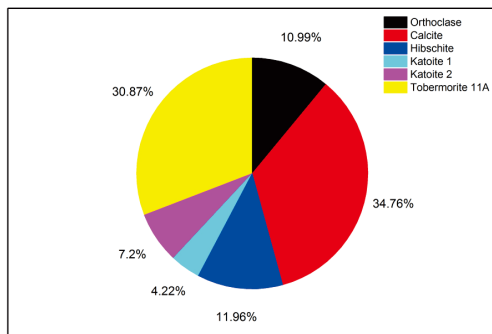
CHG-12C



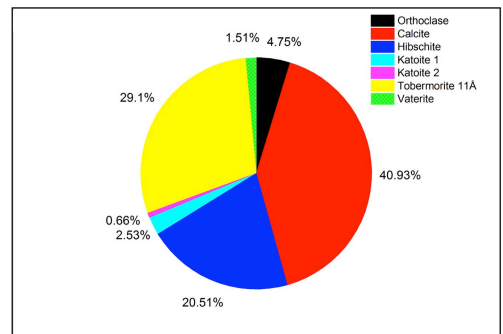
CSY-1C



CSY-2C



CSY-3C



CSY-1C-a

Fig. S5. Partitioning of crystalline phases comprising the hydrothermal product of all altered orthoclase samples, as quantified by X-ray diffraction-Rietveld refinement (mass %). Sample CSY-1C-a is the replicate sample- the phase partitioning excludes the internal standard (see main text). Katoite 1, 2 refers to different degrees of hydration.

Figures S6a-c

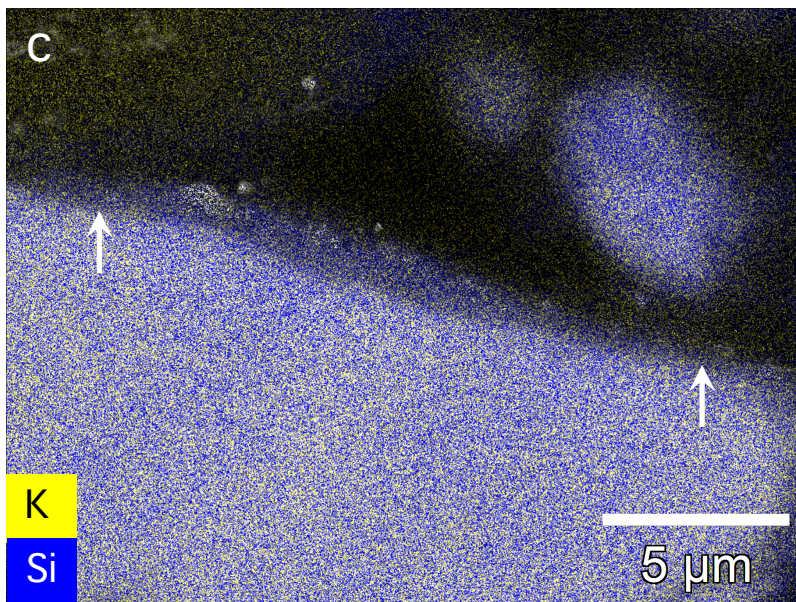
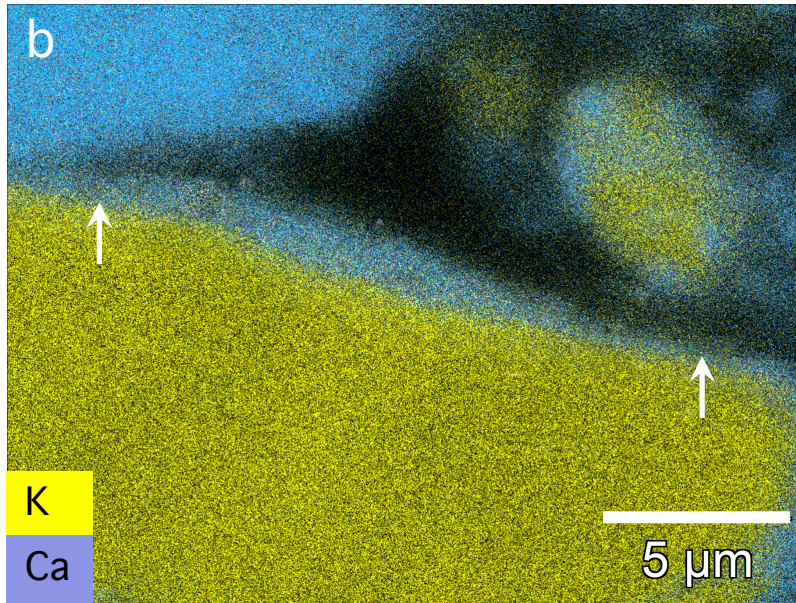
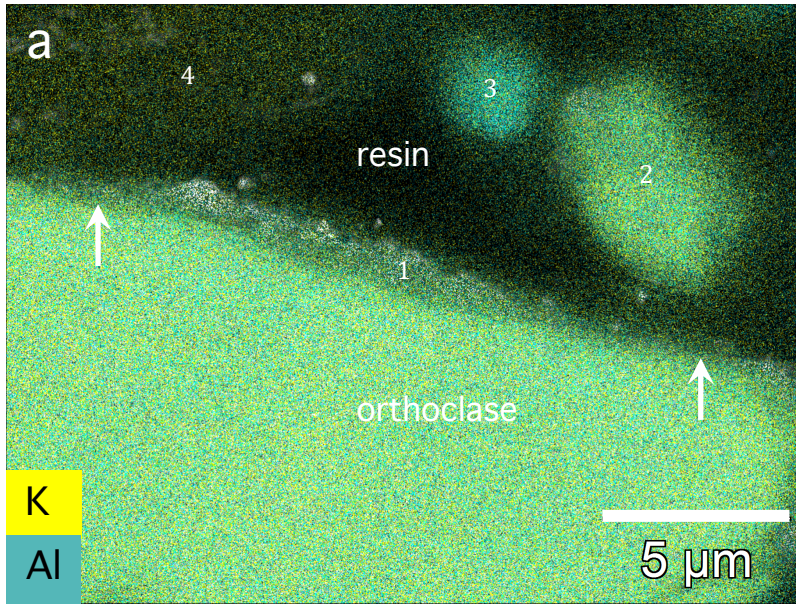


Fig. S6a-c. Binary elemental overlay maps of the same grains and areas as shown in Figs. 3a-f (main text) reinforce the conclusions drawn from the single elemental maps. Nonetheless, these binary maps more clearly illustrate certain qualitative relations between K, Al and Si. The K-Al map reveals that although well correlated in entity 2, entity 3 is relatively enriched in Al but not in K. In an opposite sense, entity 4 contains K but is Al-poor. The K-Ca map shows that these two elements are in general well-correlated. In parts of entity 2, however, this correlation breaks down, as there are distinct peripheral sub-regions that are rich in Ca, but not in K. In a similar manner, K and Si are correlated in the rind (1), whereas in entities 2, 3, and 4, certain sub-areas show enrichment in Si but not in K, or the converse, depletion in Si and higher levels of K. In all figures, the white arrows point to the orthoclase-precipitate interface. The numbers refer to specific entities of precipitate- see also main text and Figs. 2 and 3 therein.

Figure S7

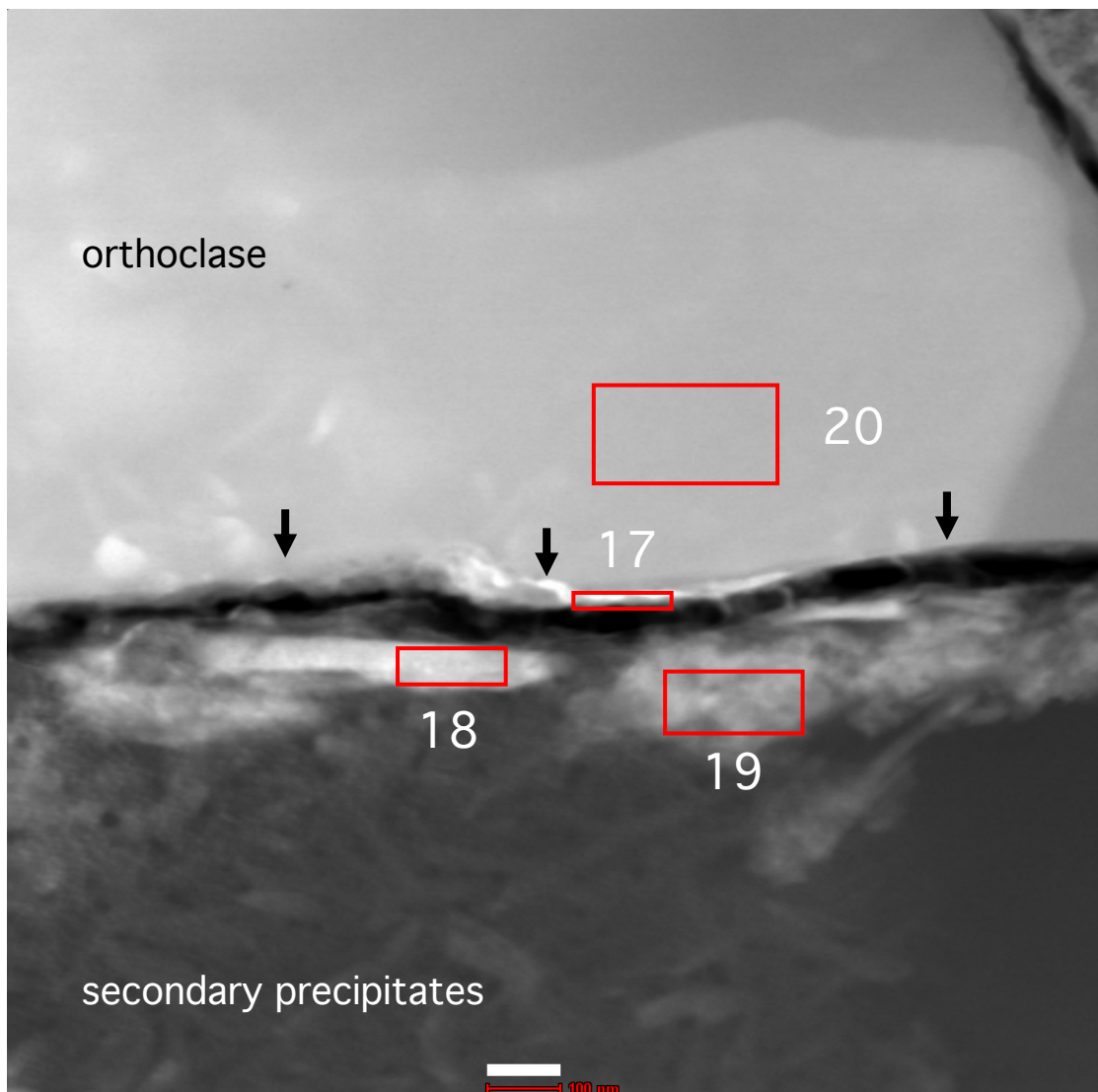


Fig. S7. HAADF-STEM image of FIB thin foil showing cross sectional view of orthoclase grain (CSY-3C) and secondary precipitates. This interface (black arrows) is close to the one shown in Fig. 4c (main text). The red boxes indicate where chemical analyses by EDXS were made; the Al/Si, K/Si and Ca/Si ratios are reported in Table 3 in the main text. Area 17, which shows an elevated K/Si ratio, may be within an amorphous phase. The reason for the large light grey region on the orthoclase grain is unknown, but is likely an artifact (redeposition) created either during FIB preparation or by the TEM electron beam during analyses (see main text, section 3.3.5). The white scale bar reads 100 nm.

References

Grew, E.S., Locock, A.J., Mills, S.J., Galuskina, I.O., Galuskin, E.V., Hålenius U., 2013. IMA Report Nomenclature of the garnet supergroup. *Amer. Miner.* 98, 785–811.