

Here, we present details of how we determined that most of our zirconolite analyses vertically sample multiple phases. Appendix Figure 1 shows that concentrations of other elements correlate inversely with the SiO_2 concentration, implying that SiO_2 from other minerals effectively ‘dilutes’ the other oxides as opposed to Si replacing cations in the zirconolite crystal. If Si were replacing one or a few cations, we would expect the concentrations of the substituted cations to decrease as Si increases while the concentrations of the cations Si does not replace should remain constant as Si increases. In the largest zirconolite grain in the probe mount, an irregularly-shaped occurrence (Fig. 4b,5), initial analyses yielded as much as 22.2 wt% SiO_2 . We addressed this issue as described in the analytical section; here we further discuss the mineral chemical trends because it is important to establish the chemical composition of this occurrence of zirconolite. Suspecting the high SiO_2 might be a sampling issue, we imaged the grain with back-scattered electrons at different accelerating voltages (BSE tomography) and then modeled the thickness of the overlying phase (using the Casino program version 2.42; Hovington et al. 1997) to be only 70-250 nm. Other portions of the irregular occurrence are overlain by K-feldspar (Fig. 5) modeled to be only 120-220 nm thick. All analyses on the thin or overlain portions of this zirconolite grain also contain a compositional component from the overlying and underlying phases. We conclude that the SiO_2 , Al_2O_3 , and Na_2O components of our zirconolite analyses were contributed by electron beam interaction with nearby (vertically distributed) phases, specifically quartz, plagioclase, and K-feldspar.

Appendix Figure 1. Variations in concentrations of ZrO_2 and other elements with SiO_2 in zirconolite of 12032,366-19 reveals that, in general, the more SiO_2 contained in an analysis, the lower the concentration of everything else. ZrO_2 , however, increases as the concentrations of most other oxides increase (except for SiO_2) indicating that the actual zirconolite composition contains little or no SiO_2 .

