ANALYTICAL METHODS

X-ray fluorescence (XRF) analysis

Whole rock XRF analysis was performed with a Spectro XEPOS spectrometer (Spectro Analytical Instruments GmbH) at the GeoZentrum Nordbayern (FAU Erlangen-Nürnberg, Germany). Accuracy and precision were controlled by internal and external standards (Basalt: BE-N; BR).

Electron probe microanalysis (EPMA)

EPMA was performed on carbon coated (ca. 20 nm) thin sections using the JEOL JXA-8200 Superprobe electron microprobe at the GeoZentrum Nordbayern (FAU Erlangen-Nürnberg, Germany). Synthetic and mineral standards were used. PETJ was used as analyzing crystal for Mn, Ti, Cr, Ca and P; TAP for Na, Mg, Si and Al; LIFH for Fe, Ni and V. The EPMA operated with an accelerating voltage of 15 kV, beam current of 15 nA and a diameter of 3 μm. Counting times for peaks were set to 10, 20, 30 and 40 seconds and background times were set between 10-15 seconds. Detection limits of the measured elements were <200 ppm for Mg, Si and Al; 200-400 ppm for Na, Ti, Ca, V and P; 400-600 ppm for Fe, Mn, Cr and Ni. Standard deviations range from 0.01 to 0.57.

Laser-ablation inductively coupled plasma–mass spectrometry (LA-ICP-MS)

To determine trace elements and REE concentrations in olivine of the drill core V6 LA-ICP-MS analysis was carried out on thin sections with an UP193FX New Wave research excimer laser coupled to an Agilent 7500 i quadrupole ICP-MS instrument at the GeoZentrum Nordbayern (FAU Erlangen-Nürnberg, Germany). Crater size was set to 15, 20, 35 and 50 μm. Helium (0.5 L/min) and Argon (1.15 L/min) were used as carrier gas. NIST SRM 612 was used as external reference material and Si by EMP analysis was used as internal standard. Measured Elements are ^{7}Li, ^{27}Al, ^{23}Na, ^{29}Si, ^{31}P, ^{39}K, ^{42}Ca, ^{47}Ti, ^{57}Fe ,^{42}Ca, ^{45}Sc, ^{51}V, ^{53}Cr, ^{55}Mn, ^{59}Co, ^{60}Ni, ^{63}Cu, ^{66}Zn, ^{73}Ge, ^{85}Rb, ^{88}Sr, ^{89}Y, ^{91}Zr, ^{93}Nb, ^{95}Mo, ^{118}Sn, ^{139}La, ^{140}Ce, ^{141}Pr,

^{146}Nd, ^{147}Sm, ^{153}Eu, ^{157}Gd, ^{159}Tb, ^{163}Dy, ^{165}Ho, ^{166}Er, ^{169}Tm, ^{172}Yb, ^{175}Lu, ^{178}Hf, ^{181}Ta, ^{182}W, ^{232}Th, and ^{238}U. Detection limits for REE vary from 0.01 to 0.42 ppm (La: 0.01-0.09 ppm; Ce: 0.01-0.11; Pr: 0.01-0.09 ppm; Nd: 0.03-0.49 ppm; Sm: 0.05-0.72 ppm; Eu: 0.01-0.17 ppm; Gd: 0.05-0.42 ppm; Tb: 0.01-0.10 ppm; Dy: 0.03-0.33 ppm; Ho: 0.01-0.12 ppm; Er: 0.02-0.20 ppm; Tm: 0.01-0.06 ppm; Yb: 0.03-0.29 ppm; Lu: 0.01-0.09 ppm). Their analytical errors are highly dependent on the concentrations and vary from <0.00 to 8.98 ppm (La: 0.01-0.15 ppm; Ce: 0.01-0.20 ppm; Pr: <0.00-0.06 ppm; Nd: 0.01-0.33 ppm; Sm: 0.03-0.30 ppm; Eu: 0.01-0.08 ppm; Gd: 0.06-0.32 ppm; Tb: 0.02-0.06 ppm; Dy: 0.21-0.53 ppm; Ho: 0.08-0.20 ppm; Er: 0.41-1.24 ppm; Tm: 0.16-0.52 ppm; Yb: 2.49-8.98 ppm; Lu: 0.51-2.03 ppm).

Fayalites of the drill core KL24 samples and the fayalite cumulate whole rocks (fused glasses) were analyzed with an Analyte Excite 193 nm Laser, which was coupled to an Agilent 7500 quadrupole ICP-MS instrument at the GeoZentrum Nordbayern, Erlangen. Crater size was set to 35 μm. Helium (0.9 L/min) and Argon (0.9 L/min) were used as carrier gas. NIST SRM 612 was used as external reference material and Si, Ca, Fe by EMP analysis was used as internal standard. Measured elements are the same as above. Detection limits for REE vary from <0.00-0.04 ppm (La: 0.01-0.01 ppm; Ce: <0.00-0.01; Pr: <0.00-0.01 ppm; Nd: 0.02-0.04 ppm; Sm: 0.02-0.04 ppm; Eu: 0.01-0.01 ppm; Gd: 0.02-0.03 ppm; Tb: <0.00 ppm; Dy: 0.02-0.03 ppm; Ho: <0.00-0.01 ppm; Er: 0.01-0.02 ppm; Tm: <0.00-0.01 ppm; Yb: 0.03-0.03 ppm; Lu: <0.00 ppm). Their analytical errors are highly dependent on the concentrations and vary from <0.00-60.58 ppm (La: <0.00 ppm; Ce: 0.01-0.02 ppm; Pr: <0.00-0.01 ppm; Nd: 0.02-0.06 ppm; Sm: 0.03-0.07 ppm; Eu: <0.00-0.01 ppm; Gd: 0.08-0.25 ppm; Tb: 0.03-0.10 ppm; Dy: 0.45-2.60 ppm; Ho: 0.17-0.88 ppm; Er: 1.91-10.95 ppm; Tm: 0.49-2.88 ppm; Yb: 10.46-60.58 ppm; Lu: 2.6-11.21 ppm).

Analytical details such as the analytical error and detection limits for each LA-ICP-MS spot can be found in the supplementary data (Table DR4). Every fayalite grain was

analyzed in line sections to exclude inclusions and represent geochemical differences in each grain.

Atom probe tomography (APT)

Background: APT is a technique whereby single atomic or small molecular ions are field evaporated and projected onto a 2D detector. This 2D distribution is extended into 3D by taking the arrival sequence at the detector into account (Bas et al., 1995). Triggering the field evaporation of the ions with a pulsed laser (Gault et al., 2006), the departure time of the ions and their arrival time at the detector can be used to determine their individual mass to charge ratio und thus chemical and isotopic identity. Since the sample is on the order of a few tens of nm and the detector some tens of mm, also a very high spatial magnification is achieved. This way, chemical inhomogeneities in crystals can be effectively investigated at the atomic scale.

Sample preparation: Based on the LA-ICP-MS data HREE-rich core domains of sample KL24 203m were selected for APT studies and extricated from the thin section using focused ion beam SEM. The sample to be investigated by APT should have the shape of a sharp needle with an apex radius of typically less than 100 nm, which is directly taken from the thin section using a focused ion beam system. The location of the sample is marked in Fig. 4A. The 'lift-out' preparation was conducted after the method propagated by Thompson et al. (2007). All coarse milling operations were carried out using 30keV Ga^+ ions, with the ion energy reduced to 10keV for the final milling operations. As a result, Ga and associated damage are absent in the analyzed volume.

Atom probe experiment: The atom probe experiment was conducted on a CAMECA LEAP 4000X HR atom probe (CAMECA Inc. Madison, WI) with a picosecond-pulse ultraviolet laser at the Department of Materials Science and Engineering (FAU Erlangen-Nürnberg, Germany). The measurement was carried out at 40K base temperature, with a pulsed laser with a wavelength of 355 nm triggering field evaporation with a pulse energy of 90 pJ in a spot of ~ 1.5 μm diameter. The pulse repetition rate was 100 kHz to ensure heavier ions are

captured within one pulse period. The DC bias voltage was regulated to a detection rate of 1kHz. The used instrument is equipped with an electrostatic mirror (reflectron), resulting in a flight path length of the ions of ca. 350 mm. This yields a mass resolution $\Delta M/M$ of ca. 1/500 at full width 10th of maximum for $^{56}Fe^{++}$, the most common ion. The resulting dataset size is around 73M ions.

Mass spectrum analysis: A mass spectrum was calculated by applying a correction for the longer flight paths of ions that are detected away from the measurement axis (bowl correction) and the shift in flight time due to the gradual bias voltage increase during the experiment. The relevant part of the resulting mass spectrum (histogram of mass-to-charge state) is displayed in Fig. 3A. Isotopic abundances are based on their average natural abundances. We further created ranges within which certain detected ions are identified as a given species based on the ion identifications. These are denoted above their respective ranges. For Li, only Li^+ was found to occur. All REE have a charge state of 2^+, only Y occurs as Y^+. The mass spectral regions pertaining to Y^{++} and 1^+ for the RE elements were not observed to contain significant spectral peaks. The isotopic pattern for three of the four Er isotopes and three of the five Yb isotopes are in accordance with their natural occurrences. This was not confirmed for the single isotope elements Ho and Tm. The peak at 89 Da is attributed to Y^+, while there is some overlap with $^{56}Fe^{16}O_2^+$, which was accounted for in the concentration calculation. The concentrations of the REE and Li were calculated either based on the uniquely identified isotopes, or on their natural abundances due to ion overlapping. The ion identifications allow us to calculate overall concentrations for ions, for which not all isotopic combinations were unambiguously identified. In this case, we based the concentration calculation on all ranges that contained less than 1 % (relative) of any other ion species. The concentrations of the remaining isotopic combinations were then calculated based on those, using the average natural abundance according to the Commission on Isotopic Abundances and Atomic Weights of the International Union of Pure and Applied Chemistry

(www.ciaaw.org). The concentration of trace elements with overlapping peaks of the majority ions like Dy and Lu was not determined. The APT results are shown in Fig. 3 for a dataset of the fayalite containing ca. 130M atoms or about 650 nm in its longest direction. The natural isotopic abundance of the multi-isotopic elements studied here was used as confirmation for the correct identification of the element and that an individual isotopic peak does not share its mass-to-charge with another ion. Therefore, based on the peak height of the most abundant isotope, the expected peak heights for each observed ion was marked with a circle. For quantification and visualization purposes, those ions that fall in the mass-to-charge range with the corresponding color were counted as the species denoted above it, with corrections for overlaps.

For all trace elements that were analyzed, including the REE and Y, additionally a background substraction was used. In this case, the number of ions in a peak was determined by establishing a locally linear background estimate and integrating the number of ions above this background estimate.

3D spatial analysis: To confirm the absence of nano-inclusions, we tested the observed 3D distribution of individual atomic species against a random distribution, using the method of Felfer et al. (2015). We partitioned the 3D space in Voronoi cells, based on the individual 3D atomic positions and calculated the distribution of their size. This size distribution was tested against a distribution from 'randomized data', where a number of atomic positions, equal to the number of test atoms was chosen at random from the whole dataset (random swapping). The Kolmogorov-Smirnov test with a confidence interval of 95% was used here. This approach accounts for any density variations that may be present in the 3D data due to reconstruction artefacts, most importantly local magnification (Miller & Hetherington, 1991). However, since in the present case a single-phase crystal was investigated, such artefacts were only present to a minor degree. No statistically significant deviation between the observed distribution and a randomized distribution was observed for Li, or any of the identified RE

elements (Yb, Er, Ho, Tm) or Y. Therefore, discrete micro- or nano-inclusions are absent in the investigated volume of the fayalite crystal.

References cited in the Data Repository:

Bas, P., Bostel, A., Deconihout, B., and Blavette, D., 1995, A general protocol for the reconstruction of 3D atom probe data: Applied Surface Science, 87-88, 298–304, doi:10.1016/0169-4332(94)00561-3.

Felfer, P., Ceguerra, A. V., Ringer, S. P., and Cairney, J. M., 2015, Detecting and extracting clusters in atom probe data: A simple, automated method using Voronoi cells: Ultramicroscopy, 150, 30–36, doi:10.1016/j.ultramic.2014.11.015.

Gault, B., Vurpillot, F., Vella, A., Gilbert, M., Menand, A., Blavette, D., and Deconihout, B., 2006, Design of a femtosecond laser assisted tomographic atom probe: Review of Scientific Instruments, 77(4), 043705, doi:10.1063/1.2194089.

Miller, M.K., and Hetherington, M.G., 1991, Local magnification effects in the atom probe. Surface Science, v. 246, p 442-449, doi.org/10.1016/0039-6028(91)90449-3.

Thompson, K., Lawrence, D., Larson, J.D., Olson, J.D., Kelly, T.F., Gorman, B., 2007, In situ site-specific specimen preparation for atom probe tomography, Ultramicroscopy, v. 107, p. 131–139, doi.org/10.1016/j.ultramic.2006.06.008.