**Supplementary Material**

1. **Analytical Methods**
2. **Whole-rock analysis**

Thirty-one representative samples (23 of Homrit Akarem granites, 6 of granodiorite from the country rock outcrop and 2 of granodiorite xenoliths within the HAGI) were analyzed for major, trace and rare earth elements at Activation Laboratories Ltd. (Actlabs, Canada). The major oxides were measured by lithium metaborate/tetraborate fusion ICP-AES. Trace and rare earth elements were measured by ICP-MS following lithium borate fusion and acid digestion. Loss on ignition (LOI) is determined by weight difference after ignition at 1000 °C. Precision and accuracy were controlled by analysis of international reference materials and replicate analyses and are 1% for major elements and 2% to 5% for trace elements. Detection limits are 0.01% for all major oxides except 0.001% for MnO and TiO2. Detection limits for each trace element are given in a column of Tables 3 and 4. Full details are on the laboratory website (actlabs.com, method 4 Lithoresearch).

1. **Mineral chemistry**

Mineral chemical analyses were performed at the Division of Geological and Planetary Sciences, California Institute of Technology, USA (Caltech) and at the Department of Geosicences, University of Oslo, Norway, on carbon-coated polished thin sections. Caltech operates a five-spectrometer JEOL JXA-8200 electron microprobe. Operating conditions were 15 kV accelerating voltage, 25 nA beam current, a focused beam (1 μm), 20 s on-peak counting times, the mean atomic number background subtraction method, a mix of natural and synthetic mineral standards, and the CITZAF matrix correction routine. This instrument was used for samples HAK 9 and HAK 9A. The University of Oslo operates a CAMECA SX100 electron microprobe. Operating conditions were 15 kV, 15 nA, 2 μm diameter beam, counting time of 10 s on-peak and 5 seconds each at low and high background positions, natural and synthetic mineral standards, and a ZAF matrix correction routine. This instrument was used for samples HA5, HA10, and HA16.

Backscattered electron images and energy dispersive X-ray spectra (EDS) were obtained with the Zeiss 1550VP field-emission scanning electron microscope (SEM) in the Division of Geological and Planetary Sciences at Caltech. To detect heavy elements, a 25 kV primary beam was chosen and spectra were collected in the 0-20 kV range. Deadtime was maintained below 60% and spectra were integrated for 60 s of live time. The EDS detector is a Si-drift type Oxford Max, and data were processed with Oxford’s AZTec software, using factory standards for quantification.

Li in zinnwaldite was analyzed by laser-ablation inductively-coupled plasma mass spectrometer (LA-ICP-MS), using a New Wave Research UP 193 Solid State Laser System coupled to an Agilent 8800 triple-quadrupole ICP-MS. The laser was operated at 10 Hz with a 75 m spot size. After 60 s washout time between samples and 15 s pre-sputtering, all analytes were counted for 45 s. The masses counted were 7 (Li), 23 (Na), 24 (Mg), 27 (Al), 28 (Si), 39 (K), 47 (Ti), 55 (Mn), 56 (Fe), 66 (Zn), and 89 (Y). NIST612 glass was the primary standard; it was analyzed before and after each unknown crystal for quality and drift control. and analyses were normalized to the mean of Si analysis of zinnwaldite by EPMA.

1. **Raman spectroscopy**

Selected mineral identifications were further verified by Raman spectroscopy. A Renishaw InVia micro-Raman instrument at Caltech with a 514 nm laser was used to collect spectra from Raman shifts of 100 to 4000 cm–1. An 1800 lines/mm–1 diffraction grating was used; scans over the wavelength range were 10 seconds each and three scans were averaged for each point. Laser power was kept at 10% in order to reduce sample heating and damage while providing adequate count rates. Cosmic ray-induced spikes and backgrounds were removed and these processed spectra were compared to the RRUFF.info library for mineral identification.

1. **Mössbauer spectroscopy**

About 100 mg of mica were separated under a binocular microscope from a crushed split of sample HAK9. It was impractical to remove fine quartz dust from the mineral separate, but the absence of Fe in quartz makes this a simple absorber rather than a major contaminant. The powder was gently ground in an agate mortar, then pressed between two pieces of scotch tape in a 1 cm hole in a Pb sample holder. Spectra were collected at 300 K in transmission geometry using a constant acceleration spectrometer with a 7 mCi source of 57Co in Rh. Velocity and isomer shift calibrations were performed by reference to bcc α-iron. Spectral integration was carried out until the signal-to-noise ratio reached an acceptable level, about 48 hours. No ferromagnetic component was apparent, so the spectra were fit to a pair of quadrupole doublets for Fe2+ and Fe3+ valences, and the areas of the doublets are used as approximate measures of molar fractions.