

## Support Information

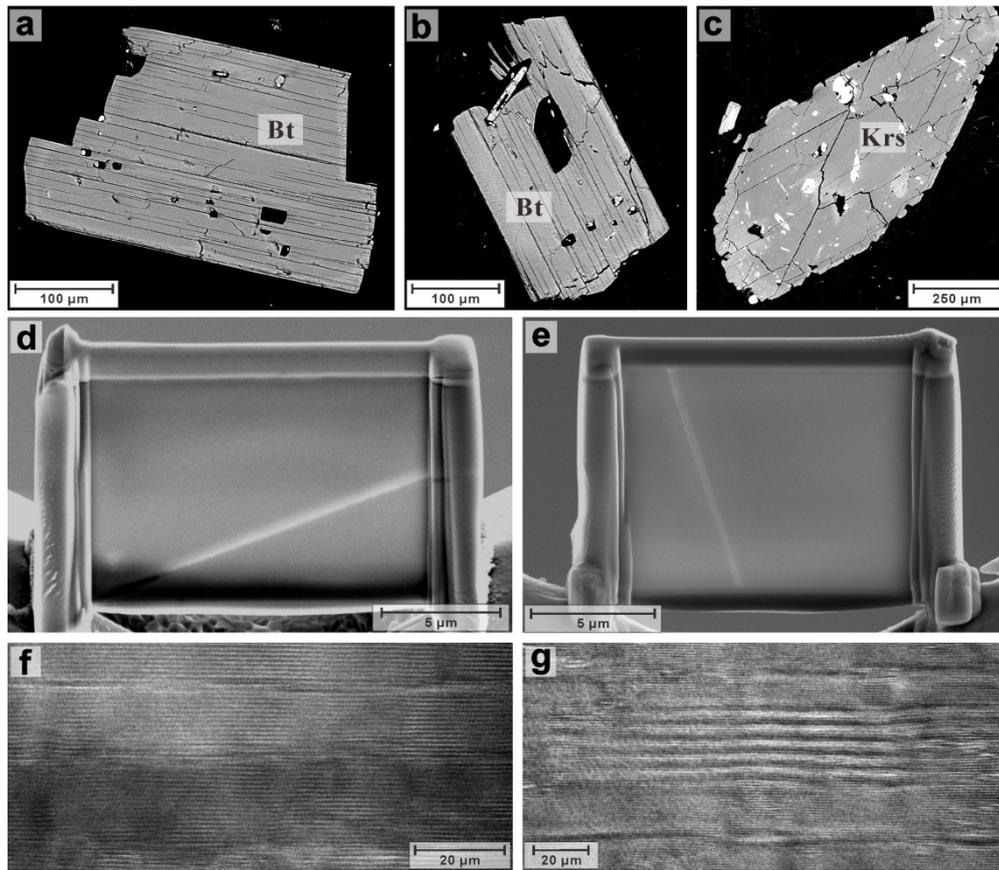
**Table S1. Calculated integral results at the Fe  $L_{3,2}$ -edges of EELS and corresponding  $\text{Fe}^{3+}/\sum\text{Fe}$  ratios.**

area	biotite				alteration product			
① No.	$L_3$	$L_2$	$L_3/L_2$	$\text{Fe}^{3+}/\sum\text{Fe}^{\text{②}}$	$L_3$	$L_2$	$L_3/L_2$	$\text{Fe}^{3+}/\sum\text{Fe}$
1	5524518.33	1806274.32	3.06	0.30	6710645.49	1889302.77	3.55	0.39
2	40406318.43	13186318.53	3.06	0.30	48896318.76	13816164.49	3.54	0.39
3	6384352.35	2091164.07	3.05	0.30	7746546.98	2180871.23	3.55	0.39
4	8956029.10	2939311.91	3.05	0.30	10916681.54	3078520.68	3.55	0.39
5	28793744.25	9410645.11	3.06	0.30	35132265.25	9895822.46	3.55	0.39
6	80545214.50	26408267.05	3.05	0.30	98106576.75	27635655.42	3.55	0.39
avg.	--	--	3.06	0.30	--	--	3.55	0.39

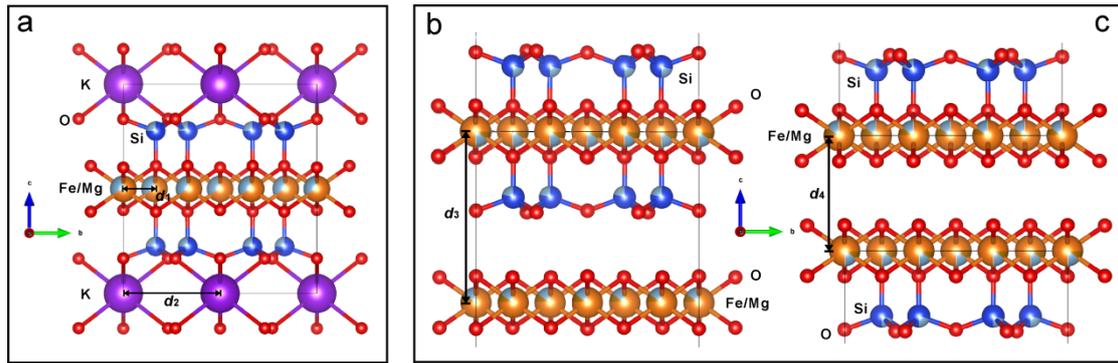
①  $L_2$  and  $L_3$  represent the signal integral of 2.1eV-width windows at the Fe  $L_3$ - (708.85-710.95 eV) and Fe  $L_2$ -edges (719.65-721.75 eV) in the background-subtracted EELS spectra, respectively.

② The ratios of  $\text{Fe}^{3+}/\sum\text{Fe}$  are acquired on the basis of the ratio of  $L_3/L_2$  by plot the latter in the diagram matching  $\text{Fe}^{3+}/\sum\text{Fe}$  and the calculated ratio of Fe  $L_{3,2}$ -edges integral results (Aken et al. 1998; Aken and Liebscher, 2002).

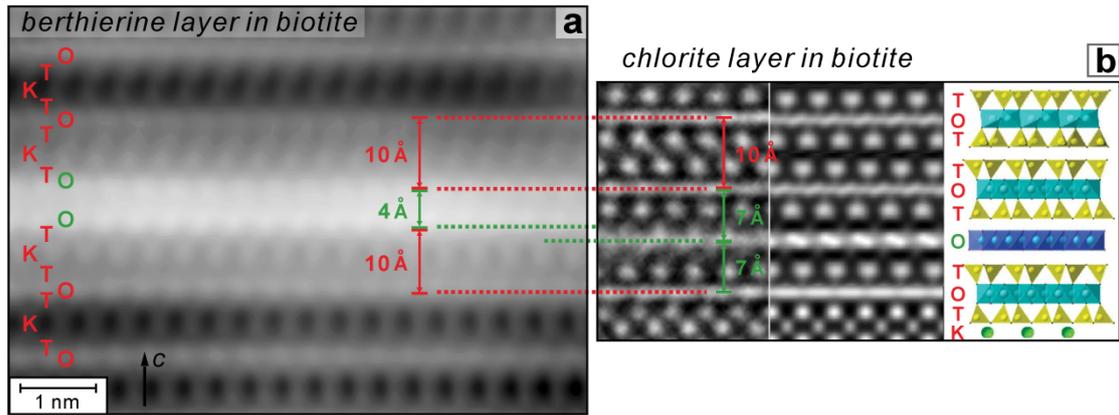
**Standard deviation values:** According to studies of Aken et al. (1998; 2002), the integral intensity ratio of  $I(L_3)/I(L_2)$  computed from two 2.1eV-width windows (i.e., 708.85–710.95 eV and 719.65–721.75 eV) is dependent on the ferric iron percentage, i.e.,  $\text{Fe}^{3+}/\text{Fe}$ . The result of the connection was a universal curve that may be used to evaluate  $\text{Fe}^{3+}/\text{Fe}$  ratios in the Fe-species. The statistical standard deviation of the  $I(L_3)/I(L_2)$ -values is less than 5%, and the absolute systematic errors for quantitative determination are roughly 0.05 for  $\text{Fe}^{3+}/\text{Fe} < 0.4$  and 0.03 for  $\text{Fe}^{3+}/\text{Fe} > 0.6$ . The majority of experimental errors can be attributed to random error during spectrum capture. Here, the averages of six sets of EELS data were calculated, and the results indicated an  $L_3/L_2$  value inaccuracy of 0.3%.



**Figure S1.** The SEM images of **(a-b)** biotite and **(c)** kaersutite phenocrysts, **(d-e)** the FIB sections cut from the edge of the biotite, and **(f-g)** the corresponding TEM images exhibiting defective regions in the host biotite.



**Figure S2.** Diagrams of a normal biotite with TOT-K unit **(a)**, a chlorite unit with TOT-O unit **(b)**, and the assumed structure of the defective region in this study with TO-OT stacking sequence **(c)**.



**Figure S3.** Comparison between (a) berthierine twin structure with a stacking sequence of TO-OT, and (b) one chlorite layer with TOT-O structure in the host biotite. Adjacent biotite units (octahedral sheets marked by the red lines) besides the defective structures (octahedral sheets marked by the green lines) in these two images have been matched.