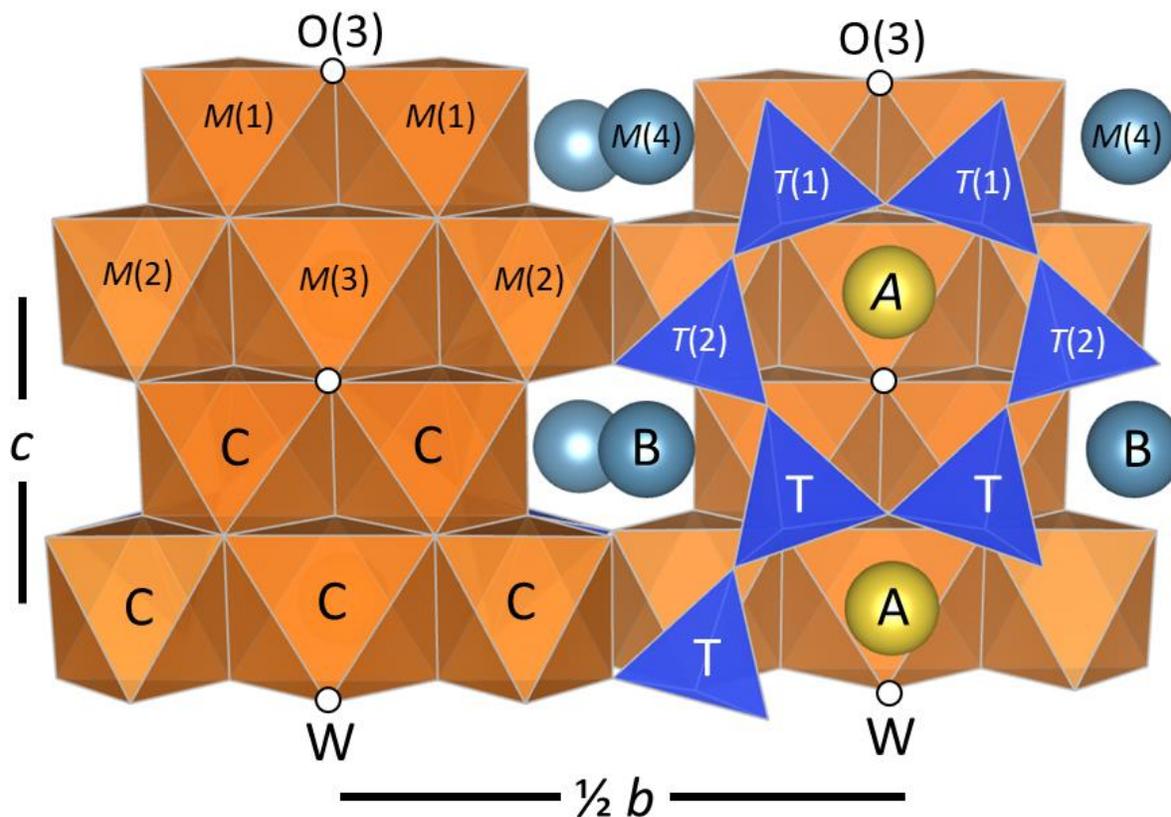


## Appendix A – Mineral formula calculation

The general formula for an amphibole is  $A_{0-1}B_2C_5T_8O_{22}W_2$ , where in this study A can be occupied by Na, K, Ca, and  $\square$  (a vacancy), B occupied by Ca, Na, and  $Fe^{2+}$ , C occupied by  $Fe^{2+}$ ,  $Fe^{3+}$ , and Al, T by Si, Al, and  $Fe^{3+}$ , and W by Cl, OH, and O. Supplemental Figure 1 shows the amphibole structure with the general formula as well as crystallographic site names indicated. Cations were calculated as follows. Ferric-iron fractions were set to the measured values for the samples for which Mössbauer analyses were available or estimated from the Cl content of the amphibole as described in the Results section. Total cations were calculated on the basis of  $O + OH + Cl = 24$ , where OH was calculated by adding sufficient  $H_2O$  that  ${}^A Ca$  is minimized ( ${}^A Ca < 0.05$ ), the sum of  $OH + Cl \leq 2$ , and the B and C sites contain 2 and 5 atoms, respectively. Cations were grouped as follows: First, all Si is added to the T cations followed by Al; if  $Si + Al < 8$ ,  $Fe^{3+}$  is added so that  $\Sigma T = 8$ . Second, the remaining Al and  $Fe^{3+}$  are assigned to C, to which  $Fe^{2+}$  is then added so that  $\Sigma C = 5$ . Third, the remaining  $Fe^{2+}$  is added to B, to which Ca is then added so that  $\Sigma B = 2$ . Finally, all K and Na along with any remaining Ca (which should be insignificant) are added to the A site. Adding large amounts of OH has the effect of diluting and lowering the total cations so that OH can be balanced with the total A-, B-, and C-group cations. There is a “sweet spot” where, by adding OH,  ${}^A Ca$  approaches 0 immediately before B- or C-group cations begin to fall below their required thresholds, which was the solution used when determining amphibole stoichiometry. For most of the synthesized amphiboles, there was insufficient Cl and OH to fill the  $O(3)$  site, so the remainder of the sites were assumed to be occupied by oxygen (Figure 3).

Supplemental Figure 1.



**Supplemental Figure 1.** A portion of the monoclinic ( $C2/m$ ) amphibole structure projected onto the (100) labeled with the sites in the general mineral formula  $A_{0-1}B_2C_5T_8O_{22}W_2$  in the lower half of the diagram, and the specific crystallographic sites, excluding most of the oxygens, labeled in the upper portion. Cl, OH, and O reside at the O(3) site.