

## **Additional Reflections and Polytypic Sequences in Polygonal Serpentine**

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The existence of polygonal serpentine was first noticed through additional reflections in XRPD patterns. Later, HRTEM revealed micrometric cross-sections, consisting of polygonally arranged lizardite sectors. They occur rotated by 12° or 24°, originating the “magic” numbers of 30 and 15 sectors per fiber (PS-30 and PS-15).

Adjacent sectors are based on different, regularly repeated polytypic stacking sequences. The stacking vector between adjacent layers changes by  $\pm b/3$  (equivalent to  $\mp b/6$ ) from one sector to the next. Moving counterclockwise, couple of layers are stacked by (*o*) orthogonal, (*r*) right or (*l*) left pointing vectors in PS-30, and by (*o*), (*l*) and (*r*) pointing vectors in PS-15. The two sequences arise because adjacent sectors have (001) rotated by 24° and 12° in PS-15 and PS-30, respectively; therefore, the PS-15 *olr* sequence arises eliminating bracketed sectors in the *o(r)l(o)r(l)o(r)l.* sequence of PS-30. Two-layer and multilayer polygonal polytypic arrangements are common, still matching the same rules as one-layer arrangements.

HRTEM observations indicate continuous 1:1 layer, with no tetrahedral inversion between adjacent sectors. The complex [100] SAED patterns with five-fold symmetry are reproduced by properly overlapped *hk0* reciprocal lattice planes.

The contemporaneous presence of different unit-cells removes degeneracy in  $d_{hkl}$  values of adjacent sectors, thus leading to clusters of additional reflections (*e.g.*, 020 and 020<sub>*r,l*</sub>; 021<sub>*r*</sub>, 021<sub>*o*</sub> and 021<sub>*l*</sub>; 022<sub>*r*</sub>, 022<sub>*o*</sub> and 022<sub>*l*</sub> in PS-30) in the XRPD patterns.

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