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VACANCY ORDERING IN MULLITE – A SUPERSPACE MODEL

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Single crystal X-ray diffraction was applied to analyze the reciprocal space of a commercial mullite sample with composition $Al_{4.84}Si_{1.16}O_{9.38}$ (Z=1) to analyze the vacancy ordering, that was generally believed to be short-range ordered. Sharp satellite reflections were visible in the diffraction patterns. The development of a superspace model required the use of different scaling factors for main and satellite reflections, respectively, indicating that only a fraction of the sample is ordered and a larger fraction is disordered. The main feature of the superspace model is the presence of oxygen vacancies in the expected environment with tetrahedra triclusters. The vacancies group together forming vacancy-dense sections that are separated by vacancy-free sections. These sections alternate along c and along a, which explains the doubling of the c parameter and also the incommensurate modulation along a. The amount of vacancies per vacancy-dense section, i.e. the length of the vacancy-dense sections, varies to match the composition given by the refined vacancy concentration $x = 0.426(5)$. A commensurate approximation with a $17 \times 1 \times 2$ supercell contains 14 vacancies corresponding to a vacancy concentration $x = 14/34 \approx 0.412$. We get to the conclusion that long-range order and short-range order coexist in mullite because satellite reflections were not detected in many measurements. Accordingly mullite is mostly disordered and a small, variable amount of domains with a long-range ordered vacancy distribution is present. This explains why satellite reflections in X-ray diffraction patterns are weaker than expected before and also why higher orders are visible in several electron diffraction patterns.