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Structure of nanocrystalline calcium silicate hydrates

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Nanocrystalline calcium silicate hydrates (C-S-H) are the main hydration products and the main binding phases in many types of cement, including ordinary Portland cement. As a result of its ubiquity in these engineered systems, C-S-H controls main cement physical and chemical properties. These properties depend on C-S-H calcium to silicon atomic ratio (Ca/Si), which is commonly described as ranging between 0.6 and 2.3. This means that C-S-H structure dictates cement macroscopic properties. As a consequence, its crystal-chemistry must be understood to be able to understand (and thus predict) cement properties. However, despite decades of study, its crystal structure is still a matter of debate. Depending on the authors and on the calcium to silicon ratio, the structure is described similar to one or more of the following minerals: tobermorite, jennite and possibly portlandite. Such inaccuracy largely results from its X-ray diffraction patterns, which exhibits only a few weak and mostly asymmetrical diffraction maxima and thus cannot be refined using classical methods. By using a specific formalism for the analysis of X-ray diffraction patterns, previously applied to phyllosilicates and phyllo-manganates, it is here proposed that C-S-H can be described as a lamellar structure similar to nanocrystalline and turbostratic tobermorite, turbostraticism meaning that there is the systematic presence, between adjacent layers, of a random rotation about the normal to the layers and/or a random translation in the layer plane. This model was validated using complementary methods (including transmission electron microscopy, synchrotron X-ray absorption spectroscopy and synchrotron high-energy X-ray scattering). From analysis of literature data, it is proposed that the evolution of C-S-H structure as a function of Ca/Si can be described as interstratification of two different types of layers having calcium to silicon ratios of 0.6 and 1.25, plus discrete $\text{Ca}(\text{OH})_2$ at higher ratios.