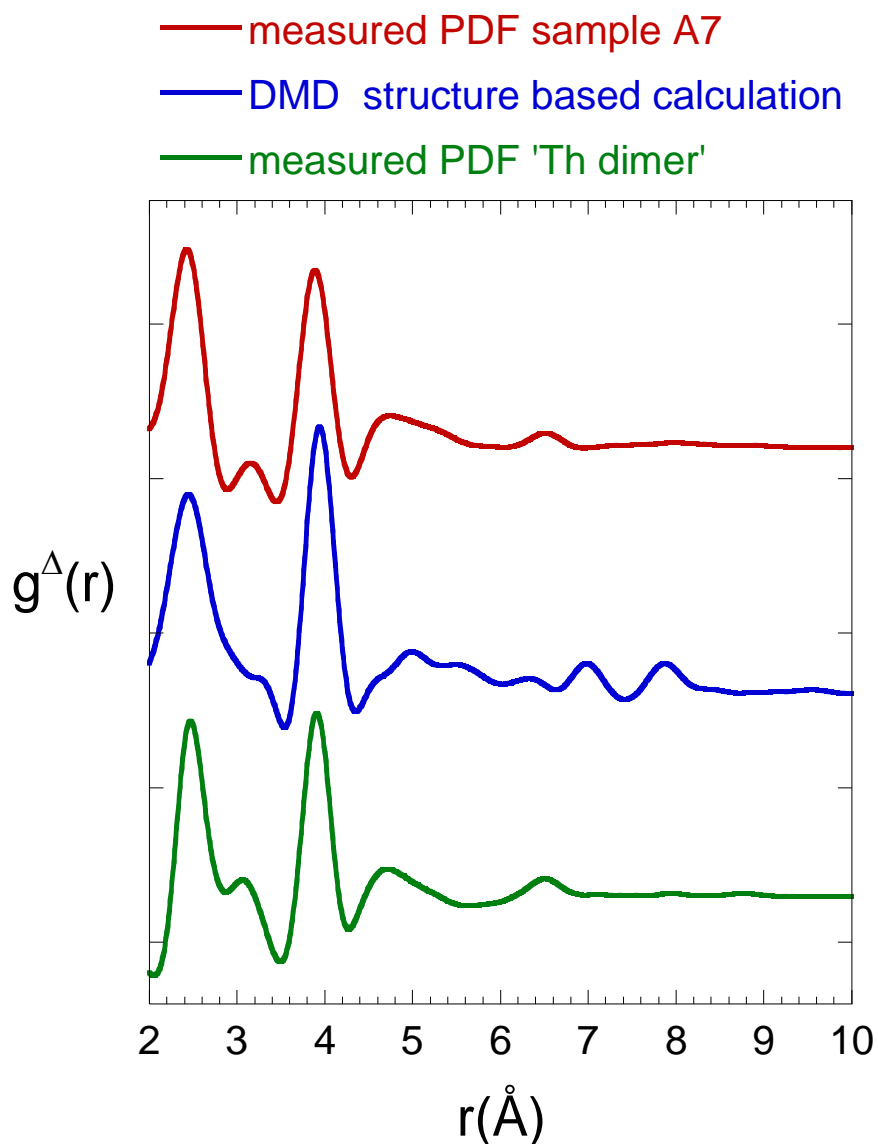


Supplementary Information

A.

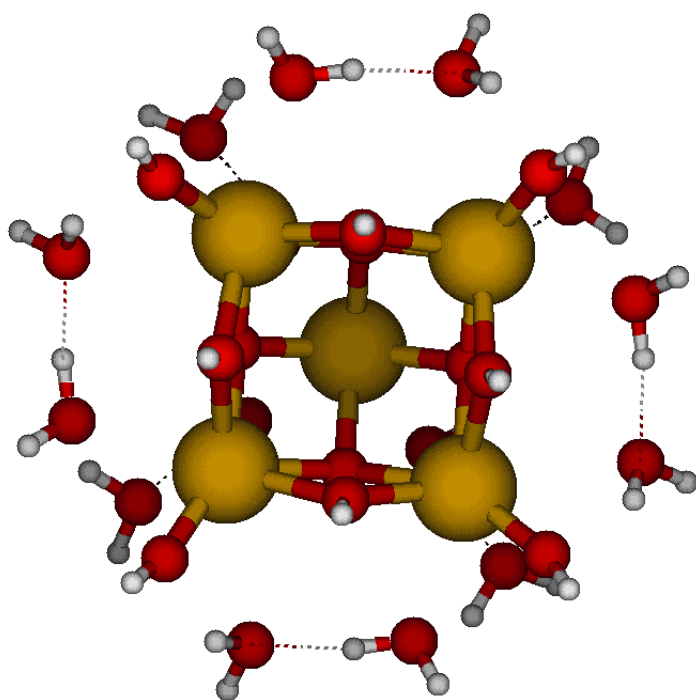
HEXS derived PDFs (top: sample 7, bottom: solution containing Th dimers, cf. Ref. ³); middle: theoretical PDF based on the favoured DMD structure.



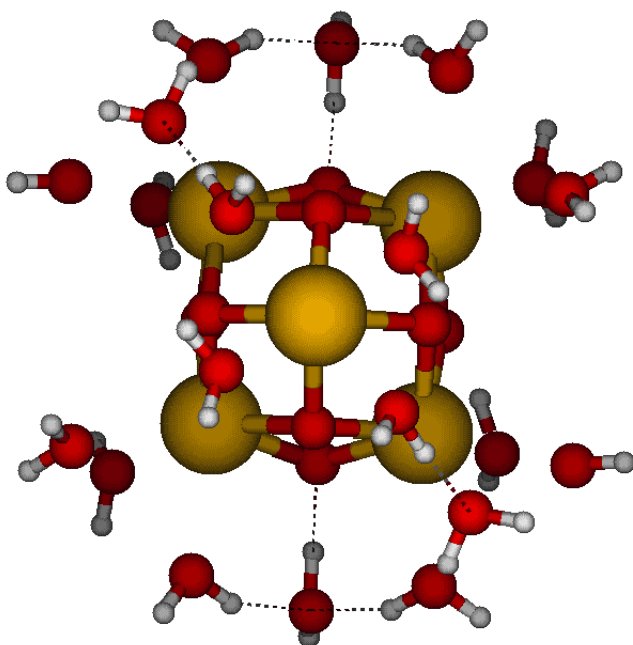
B.

Pentamer structures considered for quantum chemical calculations (cf. Table 3) in addition to the favoured DMD structure - including square-pyramides with different O/OH-bridges and a DMD derived structure, where the dimer bonds are broken and four monomers are binding to the central Th(IV) (5M):

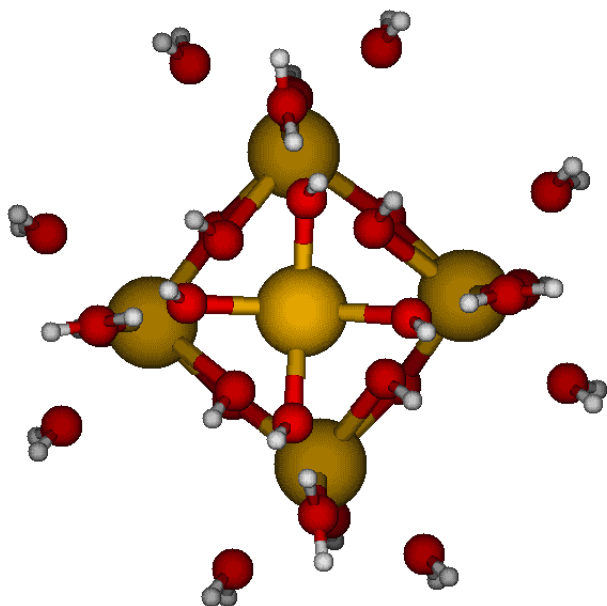
1. $\text{Th}_5\text{O}_4(\text{OH})_8(\text{H}_2\text{O})_{16}$ (pyramidal)



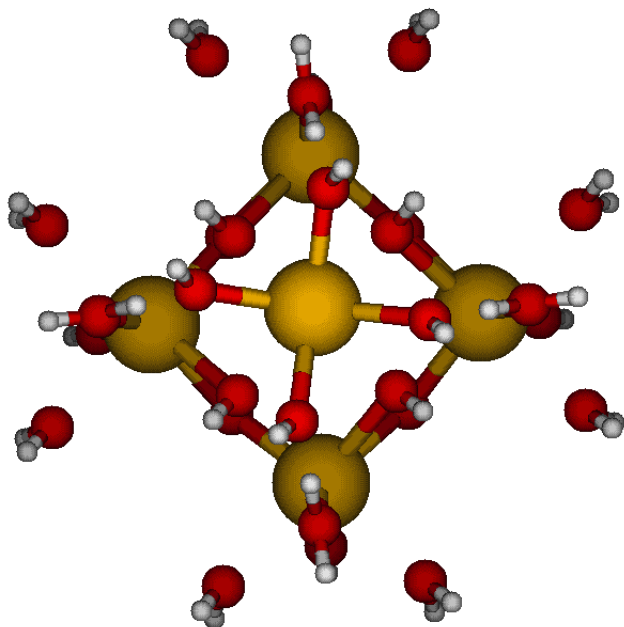
2. $\text{Th}_5\text{O}_8(\text{H}_2\text{O})_{20}$ (pyramidal)



3. $\text{Th}_5(\text{OH})_{16}(\text{H}_2\text{O})_{12}$ (pyramidal)



4. $\text{Th}_5\text{O}_5(\text{OH})_8(\text{H}_2\text{O})_{16}$



5. $\text{Th}_5(\text{OH})_{16}(\text{H}_2\text{O})_{16}$ (5M)

