**Supplementary Table 1.** Average crystallite size estimated for the pathological deposits of the human valve tissues **by the Sherrer equation.**

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Sample** | **D002**  (nm) | **D120**  (nm) | **D121** (nm) | **D300** (nm) | **D212** (nm) | **D310**  (nm) | **D113**  (nm) | **D222** (nm) | **D213** (nm) | **D004** (nm) | **Average crystallite size** (nm) |
| **2θ**  25.81 | **2θ**  28.95 | **2θ**  31.8 | **2θ**  32.9 | **2θ**  39.23 | **2θ**  39.86 | **2θ**  43.80 | **2θ**  46.72 | **2θ**  49.45 | **2θ**  53.27 |
| **Tv2a**  FWHM (°) | 37 0.22 | 20 0.40 | 15 0.55 | 16 0.52 | 21 0.40 | 16 0.51 | 28 0.30 | 20 0.43 | 23 0.38 | 27 0.33 | 22 (7) |
| **Tv3m**  FWHM (°) | 26 0.26 | 26 0.31 | 10 0.85 | 16 0.51 | 21 0.41 | 15 0.55 | 23 0.37 | 18 0.47 | 21 0.42 | 26 0.34 | 20 (5) |
| **Tv6a**  FWHM (°) | 37 0.22 | 23 0.34 | 16 0.51 | 17 0.49 | 22 0.38 | 17 0.50 | 28 0.31 | 21 0.41 | 24 0.37 | 29 0.31 | 23 (6) |
| **Tv9ab**  FWHM (°) | 37 0.22 | 23 0.35 | 17 0.48 | 17 0.48 | 21 0.39 | 19 0.45 | 30 0.28 | 22 0.39 | 24 0.36 | 28 0.31 | 24 (6) |
| **Tv12a**  FWHM (°) | 33 0.25 | 26 0.32 | 16 0.52 | 16 0.50 | 22 0.38 | 16 0.51 | 25 0.34 | 22 0.40 | 23 0.38 | 26 0.34 | 22 (5) |
| **Tv14a**  FWHM (°) | 34 0.24 | 25 0.32 | 14 0.59 | 15 0.55 | 22 0.39 | 16 0.52 | 23 0.37 | 21 0.41 | 23 0.38 | 27 0.33 | 22 (6) |
| **Tv15m**  FWHM (°) | 33 0.25 | 21 0.39 | 10 0.81 | 15 0.55 | 21 0.40 | 15 0.57 | 28 0.31 | 19 0.46 | 21 0.42 | 27 0.33 | 21 (7) |
| **Tv18ab**  FWHM (°) | 35 0.23 | 22 0.38 | 16 0.52 | 16 0.51 | 22 0.39 | 17 0.50 | 30 0.29 | 21 0.41 | 23 0.38 | 29 0.31 | 23 (6) |

\*a = aortic valve, †m = mitral valve, ‡ab = bicuspid aortic valve, **D = mean diameter of the coherent-scattering domains derived by the Sherrer equation, FWHM= the Full Width Half Maximum for the diffraction peak under consideration; 2θ= the Bragg angle.**

The unit cell parameters revealed little variations from geological hydroxylapatite and from bioapatite of mineralized tissues. The *a* parameter contraction and the *c* elongation detected can be associated to the presence of CO32- group in the apatite lattice as we have previously documented [S. Mangialardo, V. Cottignoli, E. Cavarretta, L. Salvador, P. Postorino, A. Maras, “Pathological biominerals: Raman and Infrared studies of bioapatite deposits in human heart valves,” *Applied Spectroscopy*, vol. 66, pp. 1121-1127, 2012].

**Supplementary Table 2.** Cell parameters calculated for pathologic bioapatite (Space Group: *P63/m*)

|  |  |  |  |
| --- | --- | --- | --- |
| **Sample** | **Sex, Age** | ***a*(Å)** | ***c*(Å)** |
| Tv2a\* | M, 75 | 9.4172(5) | 6.8954(4) |
| Tv3m† | F, 81 | 9.4161(8) | 6.8948(6) |
| Tv6a | M, 75 | 9.4153(8) | 6.8952(7) |
| Tv9ab‡ | M, 71 | 9.4165(7) | 6.8957(6) |
| Tv12a | M, 68 | 9.4165(8) | 6.8951(7) |
| Tv14a | M, 69 | 9.4161(9) | 6.8958(7) |
| Tv15m | M, 64 | 9.4161(9) | 6.8961(7) |
| Tv18ab | M, 41 | 9.4158(5) | 6.8958(4) |

\*a = aortic valve, †m = mitral valve, ‡ab = bicuspid aortic valve