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| TABLE S2. OBSERVED URANIUM POLYHEDRON BOND LENGTHS STRUCTURES WITH *α*- AND *β*-SHEET TOPOLOGIES  |
|  | U2Φ6-7 |  | U1Φ7 |  |
|  | <*U2*-*Φa*> | <*U2*-*Φyl*> | <*U2*-*Φeq*> | <*U2*-*Φ1*> | <*U2*-*Φ2*> |  | <U*1*-Φ> | <U*1*-*Φyl*> | <U*1*-*Φeq*> | Ref. |
| *α*-sheet topology |  |  |  |  |  |  |  |  |  |  |
| *α*-U3O8 | 2.217 | 2.075 | 2.274 | 2.544 | 4.172 |  | 2.219 | 2.074 | 2.277 | (1) |
| Protasite | 2.226 | 1.823 | 2.387 | 2.719 | 4.290 |  | 2.223 | 1.854 | 2.370 | (2) |
| Billietite | 2.200 | 1.753 | 2.379 | 2.812 | 4.388 |  | 2.233 | 1.804 | 2.404 | (2) |
| Becquerelite | 2.216 | 1.792 | 2.385 | 2.602 | 4.473 |  | 2.225 | 1.795 | 2.396 | (3) |
| Sr1.27[(UO2)O3.54(OH)1.46](H2O)3 | 2.202 | 1.807 | 2.360 | 2.631 | 4.424 |  | 2.202 | 1.807 | 2.360 | (3) |
| Cs3[(UO2)12O7(OH)13](H2O)3 | 2.258 | 1.765 | 2.455 | 2.623 | 4.427 |  | 2.241 | 1.772 | 2.429 | (4) |
| Na2[(UO2)3O3(OH)2] | 2.237 | 1.833 | 2.398 | 2.924 | 4.189 |  | 2.299 | 2.063 | 2.394 | (5) |
| Richetite | 2.202 | 1.799 | 2.364 | 2.704 | 4.417 |  | 2.247 | 1.878 | 2.395 | (6) |
| Agrinierite | 2.210 | 1.840 | 2.358 | 2.378 | 4.688 |  | 2.216 | 1.811 | 2.378 | (7) |
| Masuyite | 2.248 | 1.834 | 2.413 | 2.684 | 4.327 |  | 2.195 | 1.833 | 2.340 | (8) |
| Compreignacite | 2.243 | 1.811 | 2.416 | 2.857 | 4.361 |  | 2.224 | 1.800 | 2.394 | (9) |
| Cs3[(UO2)3O2(OH)3]2Cl(H2O)3 | 2.203 | 1.797 | 2.365 | 2.680 | 4.600 |  | 2.232 | 1.772 | 2.417 | (10) |
|  | 2.22(2) | 1.83(8) | 2.38(4) | 2.7(1) | 4.4(1) |  | 2.23(3) | 1.9(1) | 2.38(4) |  |
|  |  |  |  |  |  |  |  |  |  |  |
| *β*-sheet topology |  |  |  |  |  |  |  |  |  |  |
| *β*-U3O8 | 2.152 | 2.087 | 2.185 | 3.535 | 3.535 |  | 2.192 | 1.966 | 2.282 | (11) |
| Ianthinite | 2.211 | 2.055 | 2.289 | 3.635 | 3.678 |  | 2.265 | 1.799 | 2.451 | (12) |
| Spriggite | 2.076 | 1.839 | 2.194 | 3.498 | 3.538 |  | 2.232 | 1.937 | 2.349 | (13) |
| [U5+(H2O)2(UO2)2O4(OH)](H2O)4 | 2.180 | 2.424 | 2.059 | 3.644 | 3.644 |  | 2.200 | 1.786 | 2.365 | (14) |
| Wyaritite | -- | -- | -- | 3.535 | 3.623 |  | 2.185 | 1.812 | 2.334 | (15) |
| Wyartite-II | -- | -- | -- | 3.543 | 3.594 |  | 2.192 | 1.802 | 2.348 | (16) |
| K[(UO2)2(UO3)(OH)(NO3)2](H2O) | -- | -- | -- | 3.727 | 3.728 |  | 2.218 | 1.771 | 2.398 | (17) |
| Ba[(UO2)2(UO3)(OH)(NO3)2](H2O) | -- | -- | -- | 3.711 | 3.732 |  | 2.218 | 1.772 | 2.397 | (17) |
|  | 2.16(7) | 2.1(3) | 2.2(1) | 3.60(9) | 3.63(8) |  | 2.21(3) | 1.82(6) | 2.37(5) |  |
| ***a*** where *Φ* = O2-, (OH)-, and (H2O).References : (1) Loopstra (1977); (2) Pagoaga *et al*. (1987); (2) Pagoaga *et al*. (1987); (3) Burns & Li (2002); (3) Burns & Li (2002); (4) Hill & Burns (1999); (5) Li & Burns (2001) (6) Burns (1998a); (7) Cahill & Burns (2000); (8) Burns & Hanchar (1999); (9) Burns (1998b); (10) Li *et al*. (2001); (11) Loopstra (1970); (12) Burns *et al*. (1997); (13) Brugger *et al*. (2004); (14) Belai *et al*. (2008); (15) Burns & Finch (1999); (16) Hawthorne *et al*. (2006); (17) Unruh *et al*. (2010). |