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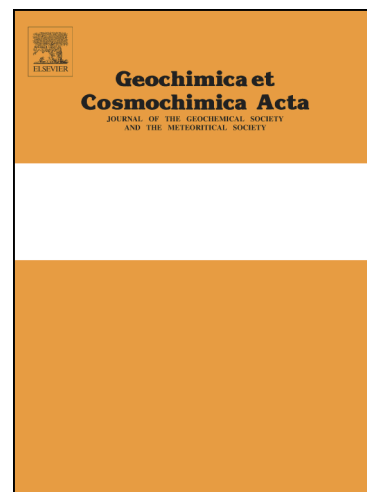
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1 Apparent energy of hydrated biomineral surface and apparent solubility 2 constant: an investigation of hydrozincite.

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8 9 Abstract

10 The apparent solubility (K_s^{app}) of hydrozincite $[Zn_5(CO_3)_2(OH)_6]$ was measured in samples of different
11 nature, including natural abiotic (“geologic”), synthetic (abiotic), and natural biominerals. A systematic
12 variation is recorded from $\log K_s^{app} = 6.2 \pm 0.1$ in geologic sample, $\log K_s^{app}$ between 7.0 ± 0.2 and 7.5 ± 0.2
13 in synthetic analogues, and $\log K_s^{app}$ between 8.8 ± 0.2 and 9.1 ± 0.2 in biomineral samples. Samples were
14 thoroughly characterized by using SEM, TEM, synchrotron radiation X-rays powder diffraction (SR-XRPD),
15 and Zn K-edge X-rays absorption spectroscopy (EXAFS). Refining SR-XRPD data, it was found a
16 significant increase (up to 10%) in the cell volume of synthetic and biologic hydrozincites with respect to
17 geologic samples. EXAFS analysis indicates small, but significant differences in the interatomic distances
18 between samples of different nature. Previous studies had shown that crystal size is in the nanometer range
19 for all samples, but decreases going from geologic to synthetic to biomineral samples. Combining these data
20 with structural data obtained in this study, the effects on solubility of particle size and of cell volume
21 increase were calculated by classical thermodynamic equations. The surface energy of hydrated hydrozincite
22 increases by at least one order of magnitude from geologic to biologic sample. The effect of cell volume
23 variation on apparent solubility is deemed negligible, being of the same order of magnitude of the error in
24 solubility measurements.

25 Thus, the different solubility of investigated samples is most likely ascribed to crystal size and surface
26 energy. The measured apparent solubility constants were used to build predominance diagrams; specifically
27 for biominerals, only the use of apparent K_s^{app} derived in this study predicts fairly well the seasonal variation
28 of hydrozincite biomineralization at Naracauli, Sardinia.

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