## Non-isodiametric growth and confinement effect in the mineralisation of witherite

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Fig. S1. SEM images of mineralized products when  $[Ba^{2+}]/[Ca^{2+}]$  is (a) 1:1, (b) 2:1 and (c) 4:1.



**Fig. S2**. XRD patterns of mineralized products prepared with  $[Ba^{2+}]/[Ca^{2+}]$  ratios of 1:1, 2:1and 4:1, respectively.



Fig. S3. SEM images of samples #1-#4 before secondary dispersion.



Fig. S4. XPS high resolution spectra of sample #1: (a) C1s, (b) O1s, and (c) Ba3d peaks.



**Fig. S5**. XPS high resolution spectra of sample #2: (a) C1s, (b) O1s and (c) Ba3d, and (d) electronic energy spectrum distribution image of element N, with inset.



**Fig. S6**. XPS high resolution spectra of sample #3: (a) C1s, (b) O1s and (c) Ba3d, and (d) electronic energy spectrum distribution image of element Ca, with inset.



**Fig. S7**. XPS high resolution spectra of sample #4: (a) C1s, (b) O1s and (c) Ba3d, and electronic energy spectrum distribution images of elements (d) Ca and (e) N, with insets.



Fig. S8. (a) DSC and (b) TG curves of pure egg white.



**Fig. S9**. (a) Selected regions in sample #4, based on a TEM image; (b) and (c) mapping images of the  $Ca^{2+}$  and the  $Ba^{2+}$  electron energy spectra, respectively; (d)  $Ca^{2+}$  content of different regions in (a).

	Atomic %	С	0	Ba	Ca	Ν
Sample numb	ber					
#1	-	45.63	38.79	15.53	0.02	0.03
#2	2	45.61	35.62	13.39	0.02	5.35
#3	3	48.85	36.31	13.53	1.26	0.05
#4	Ļ	49.29	34.43	11.85	0.89	3.53

Table S1. The atomic percentage of each element in samples #1-#4 as tested by XPS.

 Table S2. Relative percentages of Ca and Ba in selected areas of sample #3 as electronically measured.

Са	Ba	Total
0.56	99.44	100.00
0.87	99.13	100.00
1.31	98.69	100.00
1.28	98.72	100.00
1.22	98.78	100.00
0.62	99.38	100.00
	Ca 0.56 0.87 1.31 1.28 1.22 0.62	Ca       Ba         0.56       99.44         0.87       99.13         1.31       98.69         1.28       98.72         1.22       98.78         0.62       99.38

Table S3. Relative percentages of Ca and Ba in sample #4 as electronically measured.

Spectral	Ca	Ba	Total
Zone 1	1.77	98.23	100.00
Zone 2	1.78	98.22	100.00
Zone 3	3.29	96.71	100.00
Zone 4	3.91	96.09	100.00
Zone 5	4.11	95.89	100.00

Faces KJ/mol substitution rate	(111)	(002)
0%	1.025	0.9288
3%	0.9916	0.9288
4%	0.9510	0.9288
5%	0.9188	0.9288

**Table S4.** Surface Energy Calculation of Different Replacement Ratios of faces (111) and (002)surface Ca2+ of BaCO3 Crystals.

**Table S5.** The FWHM values of the two plans of all samples.

FWHM values Sample number	(111)	(002)
Sample #1	0.306	0.171
Sample #2	0.341	0.271
Sample #3	0.355	0.230
Sample #4	0.358	0.231