

Interactive comment on “Experimental diagenesis: Insights into aragonite to calcite transformation of *Arctica islandica* shells by hydrothermal treatment” by Laura A. Casella et al.

Anonymous Referee #4

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This study provides important new data and approaches to understand the transformation of biological aragonite to calcite, a problem that has significant impact on many aspects of the biogeosciences and still lacks a thorough understanding of the major underlying processes.

The paper is well written; figures are of excellent quality and support very well the presentation of data and discussion. The organization of the paper could be improved, and some recommendations to this end can be found in the detailed comments listed below, together with some editorial suggestions and question that could be addressed to improve the discussion of the extensive, well documented dataset.

First paragraph of ‘Abstract’ may be moved to ‘Introduction’.

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Line 52: I understand from the text that experimental conditions were at 100° and 175°C, and not ‘between’ these temperatures.

Line 58: ‘...in all experiments below 175°C...’ This implies that the described observation occurs at, e.g., 174°C. Be precise.

Line 72: Again, this statement implies that 175°C is a critical threshold temperature, while it is simply the temperature at which many of the experiments were run.

Line 82: ‘...limited vital effect...’ This is certainly an over-simplification, as ‘vital effects’ depend on the complexity of the biomineralisation process of the organism, the element or isotope system considered; and can be negligible or significant.

Line 91-106: Do we need to know that much about *A. islandica* in order to understand the significance of this paper, which is more concerned about general aspects of biological aragonite diagenesis? I suggest shortening this paragraph as it distracts from the main focus of the paper.

Line 124: ...natural or synthetic aragonite...

Lines 128-130: edit capitalized words

Lines 132-140: These statements should be at the beginning of the ‘Introduction’.

Lines 199-200: No isotope data are reported in table 1.

Line 200: Something wrong with this sentence; as no isotope data are reported or discussed, sentence could be omitted.

Line 205: According to table 2, samples were experimentally altered during 7, 28, and 84 days at temperatures of 100 or 175°C. Be precise.

Line 232: It may be advisable to refer to inner and outer shell layers, consistent with the terminology in Fig. A1. ‘Shell portion facing seawater’ is somewhat misleading, as only the surface of the shell may be in contact with seawater, and is additionally protected

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by the periostracum (see Fig. A1).

Lines 273-275: No explanation is offered for the high Sr-concentrations along growth lines throughout the text. Is Sr part of the organic matrix, or the mineral aragonite?

Line 288: I suggest reporting the EBSD results in a separate section with its own subheading.

Line 315: The observation of calcite formation is a very fundamental result that should be higher up in the 'Result' section, not at its end, hidden in the description of aragonite microfabrics.

Line 368: Is there evidence that supersaturation was reached in the 100°C experiments? The experimental fluids should be initially undersaturated, no aragonite dissolution is reported from the experiments, and the data in Table 1 are not sufficient to estimate saturation states.

Line 370: Fig. A3 shows alteration at 100°C, and not at 175°C.

Line 375: Only the first paragraph of this section is about the time lag of calcite replacement, while 4/5 of the section are about orientation of newly formed calcite crystals. The latter discussion is very important, and certainly deserves its own subheading.

Line 445: The pH of the solution and/or CO₂ partial pressure must be considered important, as they control the solubility of the original phase. Experiments of the current study were run with fluids with relatively high pH values and presumably very low pCO₂ (no data provided). This should be discussed when comparing the present result with older studies that observed more rapid reactions with low-pH solutions (lines 435-436).

Line 470-493: I disagree with many statements in this section. Aragonite dissolution is known from many near-surface environments and occurs essentially at surface temperatures, e.g. in the vadose environment. Factors other than temperature, e.g. pH of vadose fluids, anoxic diagenetic environments with sulfate reduction etc., have to be considered. Much has been written about a taphonomic bias related to shell min-

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eralogy as well as about carbonate diagenesis, and these complex issues cannot be adequately addressed in such a short paragraph. It is not strictly relevant to the main topic addressed in this paper and I suggest omitting it.

Line 499: While it is correct that the inner shell layer is closer to the soft body of the bivalve, this is probably quite irrelevant for the observed differences in shell structure and porosity. Inner and outer shell layers are precipitated from the extrapallial fluid, i.e. at about the same distance from the tissue of the mantle margin where the shell is formed.

Table 1: Please clarify that all data (except Mg) are referring to fluids before the experiment. Did changes of any of the tabulated items occur during the experiment, or was this not analyzed?

'hydrothermal': rephrase to 'solution', 'diagenetic fluid', or 'experimental' fluid. 'Hydrothermal' is not synonymous with 'hot water'.

What about the importance of the organic matrix, its decay, and its role of providing pathways for fluids and in changing the chemistry of fluids? The time lag observed before nucleation of calcite crystals could be related to the time required for disintegration of the organic matrix, formation of local acidic conditions due to the addition of CO₂ from the inorganic breakdown of organic molecules, and the permeability created in the process.

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