The formation of Mg-orthocarbonate through the reaction $MgCO_3 + MgO = Mg_2CO_4$ at Earth's lower mantle P–T conditions
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Response to the reviewers

We would like to gratefully thank the reviewers for the careful read and precious suggestions, which make our manuscript more clear. Changes have been done in accordance with all the suggestions. In the following we address the comments point by point.

Reviewer 1

General considerations

Reviewer Point P 1.1 — All of the crystal structures models discussed in Figure 1 should be presented in parallel to Figure 2, and there cifs should be provided in SI. The quality of Fig 2 should be improved. In the current version we can not understand the connections of the polyhedrals directly.

Reply: The figure of olivine-like Mg_2CO_4-Pnma was added to the Figure 2. As the other structures do not have stability fields we do not present their models, not to overload the figure. The references on the works, based on which we have constructed structural models have been added. The quality of the Fig2 have been improved and the figure illustrating connections between polyhedrons have been added. As is is problematic to illustrate connections of polyhedrons for the whole structure, the interconnections of only separate polyhedrons have been shown.

Reviewer Point P 1.2 — The previous exploration of Mg_2CO_4 should be introduced for a little bit more, like the paper Stability of Magnesite under the Lower Mantle Conditions, by Tomoo KATSURA, et al, published on Proc. Japan Acad., 67, Ser. B (1991) and its ref (14).

Reply: The suggested reference and discussion have been added.

Reviewer 2

Reviewer Point P 2.3 — The authors predicted that Mg_2CO_4-Pnma is isostructural to Mg_2SiO_4 (forsterite), not Ca_2CO_4-Pnma, please explain the origin from the structural packing perspective.

Reply: The discussion of structural trends in the systems Mg_2SiO_4, Ca_2SiO_4 and Ca_2CO_4 have been added. Mg_2CO_4-$P2_1/c$ is the deformed analogue of Ca_2CO_4-Pnma. Mg_2CO_4-$Pnma$ is the lower pressure form, which is observed only for orthocarbonates with small cation like Mg and it is not observed for orthocarbonates of Ca, Sr, and Ba.

Reviewer Point P 2.4 — Electronic structure and chemical bonding is critical for understanding the phases and its connection with others, e.g. Mg_2SiO_4, Ca_2CO_4, which is missing in the manuscript.

Reply: The analysis of electronic denisty distribution (Mulliken charges, Baader analysis) of Mg_2CO_4-Pnma and isostructural to it Mg_2SiO_4-Pnma has been added.

Reviewer Point P 2.5 — The Raman spectra of predicted phases may be added for the later identification in experiments.

Reply: The Raman spectra of the predicted Mg_2CO_4-Pnma has been added.

Reviewer 3

Reviewer Point P 3.6 — This is an interesting theoretical paper on Mg orthocarbonate polymorphs. The only point that attracts my attention is why Mg_2CO_4 follows the olivine-larnite phase transition at high pressure (similar to Ca_2SiO_4) and not the Mg_2SiO_4 phase transitions that involve formation of such phases as poirierite, wadsleyite (probably also wadsleyite II) and, finally, ringwoodite (i.e. the spinel-structured phase). Were these phases among the candidates for the high-pressure theoretical phases? From Figure 1, it seems that the answer is 'yes'. But then why the authors refer in Figure 1 to Zn_2SiO_4 polymorphs and not to Mg_2SiO_4 polymorphs (ringwoodite, wadsleyite)? If they want to stress geological importance of their findings, it would be reasonable to indicate clearly parallels with the Mg_2SiO_4 polymophism, which is highly important for natural systems.

Reply: We thank the reviewer for this comment. The strucutures of ringwodite and wadsleyite are the same as some of the structures of Zn_2SiO_4 presented on Figure1. The results of the optimisation does not depend on what model are used for the preparation of Mg_2CO_4 structures, Zn_2SiO_4 or Mg_2SiO_4. In the revised manuscript we have changed Zn_2SiO_4 on Mg_2SiO_4 to show comparison of Mg_2CO_4 and Mg_2SiO_4 systems. The enthalpies of Mg_2CO_4 in the form of poriereite and wadsleyite have been calculated and corresponding discussion has been added.