

We would like to thank the referees of Physical Review B for considering our paper for submission and giving us valuable suggestions. We have complied with the suggestions, and here is the summary of the improvements we have made to the paper and our response to the referees (in red colour). Wherever changes have been made in the manuscript they are highlighted in red.

Report of the First Referee -- BT13108/Talapatra

This paper presents a rigorous high-throughput exploration of the phase stability of novel classes of candidate MAX phases. This is a fantastic paper which really demonstrates the power of such an approach. For once, high-throughput methods are used without ignoring the effect of temperature and by considering the possibility solid solutions. The paper also includes some attempts at identifying a physical basis for the trends in stability that were uncovered.

We thank Referee # 1 for his appreciation of the work.

I only have a few minor comments:

1) Perhaps better explain how the transition temperatures were approximated. In particular, in Equation (3), what is ΔH_f ? Is it the formation energy of the ordered phase, of the disordered phase or of their difference? Perhaps just write down the free energy of each competing phase and state which equality between them is used to define each transition temperature.

The critical temperatures are calculated for the particular compositions (50%). Here the ΔH_f refers to the formation energy of the particular composition w.r.t the end-members and is used to obtain the corresponding solid solution temperature. We intend it to indicate a crude approximation of the solid solution temperature for the individual composition and as such it does not include the effect of the competing phases. The manuscript has been modified to explain this more clearly.(Section IV.A)

2) It would be useful, for the experimental audience, to mention some of the limitations of the calculations, namely, the use of a perfectly random solution approximation and the omission of the effect of lattice vibrations. These limitations are understandable given the number of systems considered, of course, but they should be mentioned.

The manuscript has been modified and we have pointed out the fact that the calculations are OK calculations and do not consider temperature effects or vibrational effects (Section IV)

3) Suggestion: For the most promising system, it may be worth calculating a phase diagram. Within a random solution approximation, it should be easy. A simple Monte Carlo run could also be straightforward.

Calculation of a phase diagram is the next logical step in this work. We have just completed a work (by the second author of this paper) in which we have carried out a complete finite temperature phase diagram calculation (taking into account all experimental and theoretical competitive phases) by building on the current work. It is a comprehensive work and in our opinion merits a full paper and hence we do not include it in this work.

4) Minor issues:

a) On pp. 26–27, it is not completely clear if $\text{Ti}_{1/3}\text{Zr}_{2/3}$ decomposes. On the one hand, a "decomposition reaction" is given in (7), on the other hand it is stated that it "will not decompose". Also, the term "relatively stable" is a bit ambiguous.

In Equation (7) we present a *possible* decomposition reaction based on the OQMD database. However, we show that it is not energetically favorable, since the energy of $\text{Ti}_{1/3}\text{Zr}_{2/3}$ is more negative than that of the products. Hence we say that it will not decompose. We use the term 'relatively stable' because we are aware that this is only one possible decomposition reaction and others may be possible. Additionally, there may be other unknown competitive phases which we cannot take into account. Hence, in order to be completely correct and not make a false claim, we use the terminology 'relatively stable'. In the manuscript, we have rephrased the paragraph to make our meaning clearer.

b) Equation (1) looks a bit strange. Why the "[=]" or "~"?

We thank the reviewer for pointing out this typographical error which unfortunately escaped our notice. It has been corrected. The "[=]" should be '=' and the "~" is '≈' (approximately equal).

This paper reports a very comprehensive study on the alloy thermodynamic properties of 42 selected MAX pseudo-binary alloy system on the M sublattice site. As mentioned by the authors, MAX is a broad class of material systems with remarkable mechanical properties, oxidation resistance and thermal stability. Through alloy designing, new stable phases with interesting properties could be discovered to meet the demands of different applications. However, it is not an easy task for experiments and computations. This work made valuable contributions from this aspect. Overall, this referee believes it is a publishable work. There are several comments.

We thank Referee # 2 for his encouraging words.

(1) It is better to provide details of their CE results in the supporting information. For example, how much is the CV score, how many pair clusters were used and how many body clusters were used?

The CV values have been tabulated in the supplementary data document. CE calculation results for all systems including cluster information, eci data, ground states etc. have been included as a compressed data file (CE_results.tar). We thank the reviewer for this comment. It definitely adds necessary detail to the work.

(2) The value of the DOS and pDOS results is not clear for this referee. What physical insights can be obtained?

The intent of the authors was to present a systematic analysis of the difference in the DOS and pDOS of the three regimes. The DOS reflect the stability of the phases. Regime I (weak ordering) shows a shallow trough at the fermi level, corresponding to the low formation energy. In Regime II (phase separation), we see a peak in the vicinity of the fermi level corresponding to the positive formation energy (w.r.t the end members). Finally, for Regime III (strong ordering) we see a deep trough corresponding to the highly negative formation energy. This explanation has been added to the to make the distinctions clearer.

(3) The authors did not define the variables used in Eq. (5) and (6) on page 24. Also they used $r_{\{\pi\}}$ in equation but r_{π} in main text. It is better to be consistent.

This error has been corrected. Thank you for pointing out the same.

The authors thank the referees for their valuable suggestions. We believe they have added necessary detail and clarity to this work.