Comments from the editors and reviewers:

-Reviewer 1

- The author systematically investigates the various properties relevant to the grain-boundary of U3Si2 using a molecular dynamics technique. However, there are still unclear points for understanding in the present paper.

1. P7, line2: The author writes “…maxima are observed at the (730) and (530) planes…” Is it correct? The GB energy at (910) plane is larger than one at the (730) plane, isn’t it?

Verifying that I wasn’t stupid or that I was stupid for a specific reason.

*It seems the reviewer is right for the post-translation results. We may need to just re-phrase it.*

1. P8, line3-4: The author writes “…the most prominent maximum remains at the (730) plane”. Is it correct? The GB energy is highest at the (530) plane at the temperature range investigated, isn’t it?

Verifying that I wasn’t stupid or that I was stupid for a specific reason.

*It seems the reviewer is right.*

1. P12, Fig.6: For what do the blue and red spheres stand? The lattice direction is unclear.

Will label the figure and add in xyz spatial figure thing.

1. P13, line10-12: According to Fig. 7, the surface energy is the lowest at the 1.4-1.45 of U/Si. This is different from the description by the author. Does the author use the “stoichiometric” as U/Si = 1.5?

Verifying this.

1. P14, line2-4: What is the reason why the surface energy increases suddenly from 1200 K to 1600 K? Is the surface melting relevant to that increase?

I don’t have a good answer to this. Possibly? The surfaces are certainly getting rougher, but melting might be too strong of a word.

*We are dealing with only the potential energy here. The increase in temperature causes an increase in the lattice parameter, along with an increase in the potential energy. We can probably test this by doing 0K calculations with strains to see if this is caused by the increase in lattice parameter. The surface disorder can certainly be a possibility. Another possibility is cutoff used in the potential. From 1200-1600 K some atoms are moving out of the cutoff.*

P16, line18-20: According to Fig. 10, the segregation energy of U interstitial at the (210) plane is not attractive, unlike other cases. If possible, the author had better give the reason here.

No idea! Not sure what I will say here.

-Reviewer 2

- The authors used molecular dynamics method to investigate several properties of U-Si material, including gain boundary energy, surface energy, void surface energy, and segregation energy of point defects to grain boundary. Different GB and surface structures were studied. The topic itself is interesting, but there are some important concerns about the technical points.

(1) For the construction of grain boundary. The authors applied periodic boundary condition in only two directions (x- and z- directions), which means, in Fig.1, there is only one GB in the middle and there are two free surfaces on both end of the model (y-edges). This is inconsistent with the description “where grain boundaries exist in the center of the supercell (denoted by a dashed black line) and on the y-edges of the supercell.”

There are indeed periodic boundaries on the y-edges. We apologize for the confusion and the text has been modified to more clearly illustrate the nature of the supercell. TODO- modify text

(2) The simulation sample is extremely small. To my understanding, such kind of simulation can be easily performed on PC rather than running on a High Performance Computer as acknowledged in the final part. Although the authors mentioned that the current simulation system is large enough to obtain accurate energy values, the calculated result of energy using the introduced methods in the manuscript can be more trustable by increasing the size of the sample (the GB and surface area). The increase of model size along x- and z- directions is necessary.

It is possible that we did not adequately convey the sample sizes utilized to generate data. For grain boundaries energies 500 simulations were performed. For surface energies, 200 simulations were performed. For grain boundary defect segregation energies, 5760 simulations were performed. The sample size for grain boundary energies has been doubled and for the surface energies has been quadrupled, per the reviewer’s suggestions, and the scope of the sample has been better explained. TODO-simulations, shouldn’t take too long

(3) The methods used to calculate the interface energy and segregation energy are debatable. For example, ‘E\* is the energy per atom’ in equation (1), do you count the energy of atoms one by one? Also, the energy of atoms at the interface area can be quite different due to their positions, the energy difference (E\*-E) cannot be multiply the number of atoms (N) directly. In equation (2), according to the authors description, the result of E(def)-E(bulk) is the defect formation energy in the bulk rather than the defect segregation energy to GB. E(seg) should be calculated by considering the difference of GB energy before and after the segregation.

We ensured that we were obtaining the correct grain boundary energies by indeed looking at individual atomic energies, comparing the energy to their energy in the perfect equilibrium system, and calculating energy differences and thus a grain boundary energy. This yields identical results to comparing the energy per atom of the total system to the energy per atom of the perfect equilibrium system.

The formation energy of a defect near the grain boundary is compared to the formation energy of a defect far away from the grain boundary. Thus, the only difference in the two systems is the location of the defect, and this energetic difference is the segregation energy. We apologize for the confusion again and the text of the manuscript has been modified to more clearly illustrate our methods. TODO-modify text

*The reviewer has a point that the per atom energy we used is probably an averaged per atom energy, not the exact energy of each individual atom (please verify). Why don’t we just use the total energy in the equation? It is sufficient to derive the GB energy and segregation energy. Also, it removes the number N from the equation, which sometimes implies size dependence. N is not important and it even does not need to be known.*

(4) Some of the results are unnecessarily duplicative. Fig.2 and Table.1 show the same result, why are they presented in two different ways? Similarly, Fig.5 and Table.2, Fig.8 and Table.3, Fig.9 and Table.4.

This has been condensed. The authors wanted to be inclusive (and perhaps were so to excess). We felt that visual trends were important, but also that generally it is the numbers that are the usable pieces of data, so we included both originally. Selectively a graph or table has been deleted so as to not excessively duplicate data. TODO-modify graphs/tables

*I don’t know if we should condense it. The plots are good to show the trends, but the numbers are sometimes useful too. For instance, some reader may want to compare the exact values for benchmarking, or some upper-scale modelers want a number to use. It’s sometimes painful to digitize a picture to get a number. An option is to put the tables to appendix or somewhere so the readers can still access.*