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?

The reviewer is correct, and the text has been modified accordingly.

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?

The reviewer is correct, and the text has been modified accordingly.

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

We have added a legend indicating crystallographic directions. We have also explained the red/blue atoms in the caption.

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?

Yes, stoichiometric is defined as U/Si = 1.5. In systems with a surface with a lower U/Si ratio, the second surface present in the system will have a higher U/Si ratio, and thus a high surface energy. This leads to systems with non-stoichiometric surfaces exhibiting a higher average surface energy.

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?

It is possible that we are seeing the beginnings of surface melting, but not definitive. There is certainly surface roughening, but there still exists a defined surface. That this energetic increase is associated with increased surface roughening has been included.

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.

The averaged value is net positive; however, the repulsion is not statistically significant. With a greater number of simulations, this interaction could become slightly attractive, but the important point is that the interaction is weak. This has been emphasized.

-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.

(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.

(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 thank the reviewer for a detailed examination of our equations. This was an error caught by the reviewer. It is in fact the total energy of the system with two interfaces that we are comparing with the energy of a pure crystalline system. The equation and description have been modified.

We also 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.

(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.

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. The tables have thus been moved to an appendix, so that the manuscript itself is not cluttered but the data is available.