# Grain Boundary Energy and Surface Energy of $\alpha$-U

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## INTRODUCTION

Metallic alloys of U-Zr and U-Pu-Zr are of interest for the Versatile Test Reactor and several microreactor designs. The $\alpha$-U phase (stable up to 935 K [1]) exists in metallic fuel and has a significant contribution in fuel performance. However, many fundamental material properties are still unknown, such as the behaviour of grain boundary (GB). GBs have an effect on plasticity [2], swelling [3], etc. of polycrystalline $\alpha$-U. Symmetric tilt grain boundary (STGB) is a common type of GB, which has a mirror symmetry across the tilt plane (plane contains tilt axis). The energy cost to make a cavity along a STGB (such as in alpha tearing) can be expressed as the energy difference of a STGB versus two surfaces of the same orientation [4]. Thus, the surface energy is also required to evaluate the mechanical performance of $\alpha$-U. To analyze the interfacial energy (GB energy, surface energy, etc.), molecular dynamics, along with a suitable interatomic potential, is a powerful tool [5]. In view of this, the current work has been conducted to evaluate the GB and surface energy of $\alpha$-U at four different temperatures (300 K, 400 K, 500 K and 600 K) employing molecular dynamics to determine the variation of inter-facial energy as a function of STGB orientation and temperature.

## METHODOLOGY

LAMMPS [6] along with the UMo ADP potential [7] is utilized to perform molecular dynamics simulation. To generate the super-cell, the simulation box is divided into two equal regions. Region one is tilted with respect to defined tilt axis and tilt angle (tilt angle is half of misorientation angle). Taking the refection of the former region across the tilt plane, region two is created. Hence a bicrystal of $\alpha$-U with two STGBs is formed. In order to maintain the periodic boundary condition in all directions, supercell size varies with tilt axis, misorientation angle and shear plane. While investigating the surface energy, only region one with $\alpha$-U atoms is created and region two is kept vacuum, thus two surfaces are created within a super-cell. An as-constructed STGB is shown in Figure 1.

A total of eighty STGBs are constructed, among them, twenty-four have tilt axis $\langle 1\ 0\ 0\rangle$ (shear plane (0 0 1)), twenty-four have tilt axis $\langle 0\ 0\ 1\rangle$ (shear plane (0 1 0)), and thirty-two have tilt axis $\langle 0\ 0\ 1\rangle$ (shear plane (1 0 0 )), such that entire scope of STGBs are studied with respect to three shear planes. These three categories are named as type A, type B, and type C, respectively.

A Nose-Hoover barostat and Langevin thermostat is considered to relax the system, and after reaching equilibrium, observable parameters are collected. Systems are equilibrated for 200 ps and energies are averaged over the final 50 ps. The

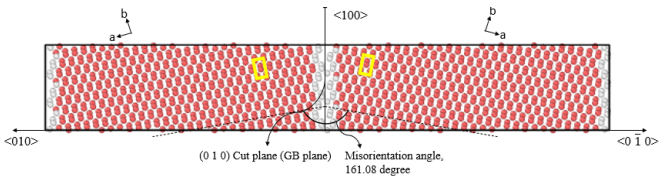


Fig. 1. The as-constructed $\alpha$-U symmetric-tilt GB (STGB) $(\bar{1}\ 12\ 0)$ with tilt axis $\langle 1\ 0\ 0\rangle$ and shear plane (0 0 1). Grain boundaries exist in the middle and on the edges of the super cell. Both red and white atoms are U, but with different coordination environments to illustrate grain boundaries. Yellow rectangles indicate the $\alpha$-U unit cell and a and b represent the original $\langle 1\ 0\ 0\rangle$ and $\langle 0\ 1\ 0\rangle$ crystal structure orientation respectively.

inter-facial energies are calculated as

$$E_{\text{inf}} = \frac{E^* - E_0}{A} \times N \tag{1}$$

where $E_{\text{inf}}$ is the interfacial (STGB or surface) energy per unit interface (STGB or surface) area, $E^*$ is the energy per atom of a system with an interface, $N$ is the number of atoms in the system with an interface, $E_0$ is the energy per atom of a defect-free system, and $A$ is the total area of the interface.

GB and surface energies are observed at 300 K, 400 K, 500 K, and 600 K where uranium is present in $\alpha$ phase.

## RESULTS

GB energy variation with misorientation angles from 0° to 360° at 500 K temperature for type A, B, and C STGBs are presented in Figure 2. Data points are connected via straight line to guide the eye. Because of the mirror symmetry of $\alpha$-U along (1 0 0) and (0 0 1) planes [8], no GB is formed at the 180° misorientation angle of type A, B, and C STGBs, or at the 0° misorientation angle of type B STGBs.

The number of cusps over the GB energy landscape are dependent upon the type of STGBs. For instance, type A STGBs ($\langle 1\ 0\ 0\rangle$ tilt axis) have three deep cusps at the $(0\ 1\ \bar{0})$, $(\bar{3}\ \bar{12}\ 0)$, and $(0\ \bar{1}\ 0)$ plane and also a minor cusp at plane $(\bar{3}\ 6\ 0)$. The GB energy profile of type B STGBs ($\langle 0\ 0\ 1\rangle$ tilt axis) only has a minor cusp at the $(\bar{2}\ 0\ \bar{1})$ plane. However. type C STGBs ($\langle 0\ 0\ 1\rangle$ tilt axis) have the highest number of cusps (eight), which are oriented at a mirror symmetry orientation with respect to the $\langle 0\ 0\ 1\rangle$ tilt axis, see Figure 2. Major cusps of type C STGBs are also mirror oriented oriented with respect to (0 0 1) STGB plane. Between these two cusps $(0\ 18\ \overline{25})$ STGB plane has the lowest GB energy (0.36 J/m$^2$) and $(0\ \overline{18}\ \overline{25})$ plane possesses the highest GB energy (1.39 J/m$^2$) among type C STGBs.

This study indicates that type C STGBs (0.94 J/m$^2$) have the highest average GB energy, followed by type A STGBs (0.78 J/m$^2$), with type B STGBs (0.75 J/m$^2$) exhibiting the lowest GB energies. Among the entire dataset of eighty STGBs, the lowest GB energy is observed for the type A STGB at the $(\overline{3}\ \overline{12}\ 0)$ STGB plane, where $\langle 1\ 0\ 0\rangle$ is the tilt axis, (0 0 1) is the shear plane, and the GB energy is 0.30 J/m$^2$. Low energy STGBs would be a likely candidate to form twins. The highest GB energy is found for a type C STGB at the $(0\ \overline{18}\ \overline{25})$ STGB plane, where $\langle 0\ 0\ 1\rangle$ is the tilt axis, (1 0 0) is the shear plane, and the GB energy is 1.39 J/m$^2$.

The effect of temperature on GB energy of $\alpha$-U STGBs is studied considering twenty-two STGBs (eight from type A, seven from type B, and seven from type C) and presented in Figure 3. All three types (A, B, and C) of STGBs experience an increase in GB energy with temperature (especially from 400 K to 600 K). However, the average change in GB energy within the studied temperature range (300 K to 600 K) with respect to GB energy attained at 300 K varies with STGB type; for example, type A by 15.8%, type B by 7.8%, and type C by 82%.

In the current work, surfaces which are only related to selected STGBs of $\alpha$-U are studied. Including the three primary surfaces, the surface energy of total eighty-three surfaces are calculated. Surfaces are classified as per the classification of STGBs. From Figure 4, the trend of surface energy of type A, B, and C surfaces with tilt angle is observed at 500 K.

No discernible trend is found for type A and C surfaces, whereas type B surface have a mirror symmetry with respect to the (1 0 0) plane. A similar pattern in the GB energy plot is observed if minor cusp is omitted, see Figure 2. Type A, B and C surfaces have an average surface energy of 1.25 J/m$^2$, 1.21 J/m$^2$, and 1.26 J/m$^2$, respectively. Among the studied surfaces, the maximum surface energy has been observed at the type C $(0\ \overline{18}\ \overline{25})$ plane with a surface energy of 1.52 J/m$^2$, and lowest energy is observed for the type A $(\overline{3}\ \overline{12}\ 0)$ plane with a surface energy of 1.0 J/m$^2$. At 500 K, the surface energy of three primary surfaces ((1 0 0), (0 1 0), and (0 0 1)) are calculated as 1.13 J/m$^2$, 1.39 J/m$^2$, and 1.11 J/m$^2$, respectively.

An investigation of surface energy variation with temperature is performed for a subset of STGBs, similar to the investigation of GB energy with temperature and plotted in Figure 5. Like GB energy, a general positive correlation between surface energy and temperature (specifically from 400 K to 600 K) is observed. Comparing among three types of surfaces, type C is significantly more sensitive to temperature, as the average change in surface energy with respect to 300 K for the Type A, B, and C surfaces is 4.4%, 2.5%, and 9.7%, respectively.

From Figures 2 and 4, it is also observed that the GB energy of $\alpha$-U is not only dependent on misorientation (or tilt) angle, but also on tilt axis and shear plane because of the unique crystallography of $\alpha$-U. Comparing the average interfacial energy of STGBs of $\alpha$-U with other U-based systems at 600 K (for the misorientation domain 0° to 90°), $\gamma$-U has the lowest GB energy, followed by $\alpha$-U and $U_3Si_2$ [9, 10]. An analogous result is found for the surface energy.

## CONCLUSIONS

Elucidating the dependence of crystallographic orientation and temperature on GB and surface energy, the current work is the most comprehensive study on inter-facial energy of $\alpha$-U to date. At 500 K, the global arithmetic average of the surface energy (1.24 J/m$^2$) is approximately 1.5 times the GB energy (0.82 J/m$^2$). With the increase of temperature, GB energy in general increases more rapidly than the surface energy. The information from this work will be utilized to study GB evolution in polycrystalline $\alpha$-U and deformation under irradiation, leading to the development of improved fuel performance models.

## ACKNOWLEDGMENTS

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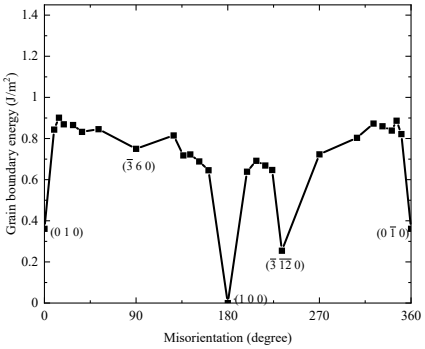
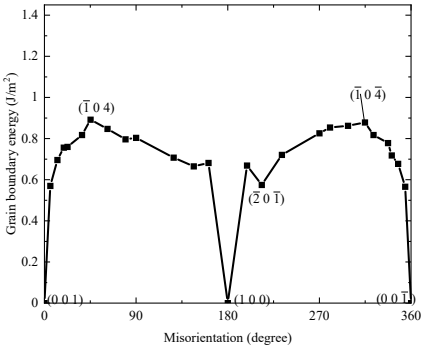


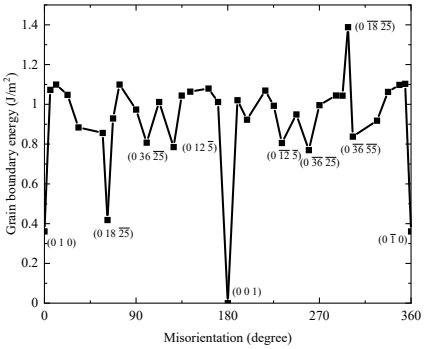
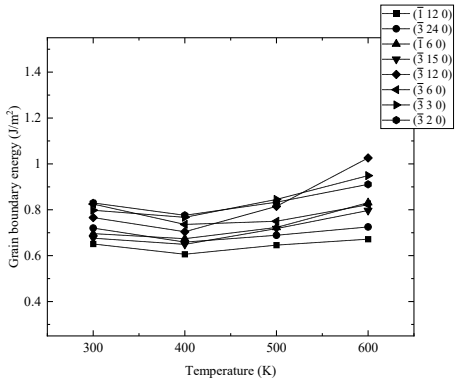
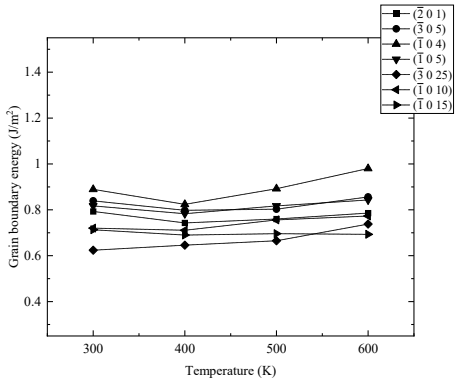




Fig. 2. GB energy of STGB (symmetric tilt grain boundary) of (a) type A, (b) type B, and (c) type C as a function of misorientation angles at 500 K. Key features are labeled with their STGB plane orientation.



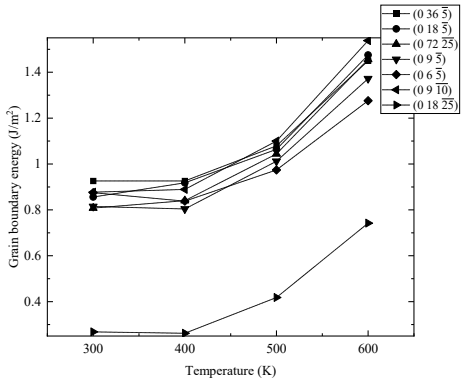
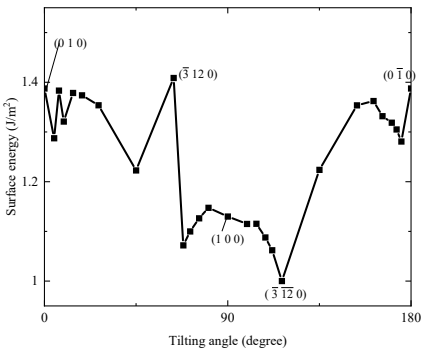
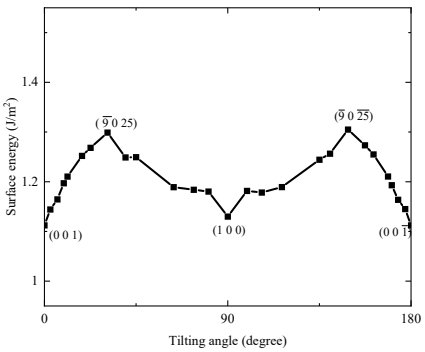




Fig. 3. GB energy of selected STGBs of (a) type A, (b) type B, and (c) type C as a function of temperature



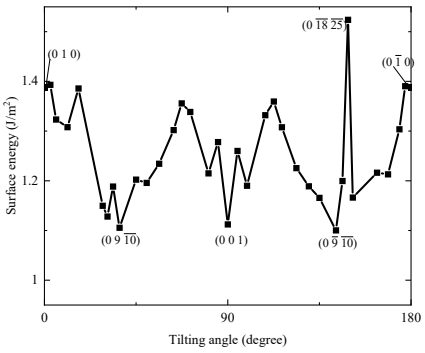
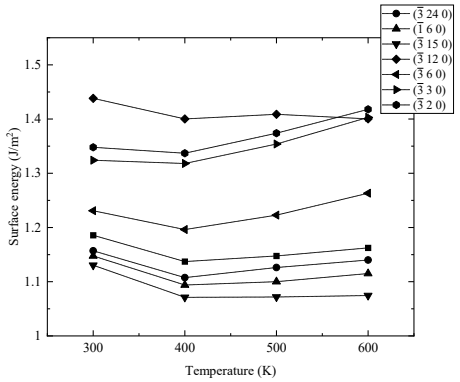
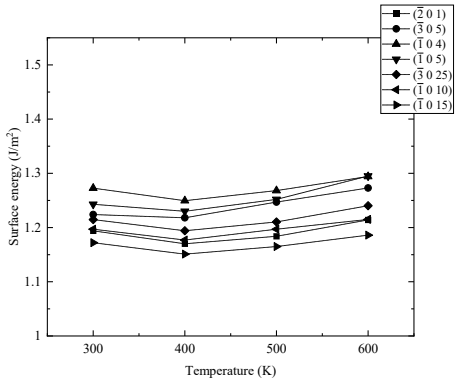




Fig. 4. Surface energy of surfaces of (a) type A, (b) type B, and (c) type C as a function of tilt angle at 500 K.



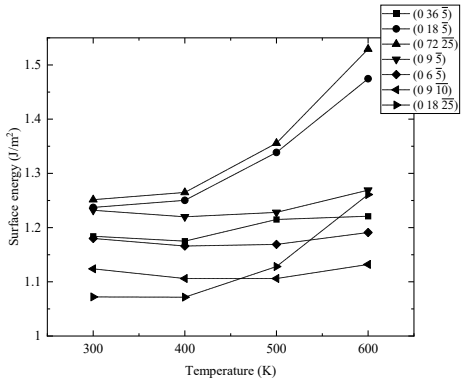




Fig. 5. Surface energy of selected surfaces of (a) type A, (b) type B, and (c) type C as a function of temperature.