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Abstract

Aluminum Nitride is an active element of sensors that monitor the performance and well-being of the nuclear reactors due to its piezoelectric properties. Yet, the variations of its properties under irradiation are largely unexplored. Here, we report the results of the molecular dynamics simulations of the structural changes in AlN under irradiation via the knock-on atom technique. By creating and evolving the irradiation cascades due to energetic particle interaction with the atom of the crystalline lattice we determine the rate of the defect production as a function of the deposited energy. Further, we determine a displacement energy, a key characteristic that describes how efficient the defect production in the given material is. Comparison with the isostructural GaN is provided.

1 Introduction

In nuclear reactors, a way to measure physical paramaters is through ultrasonic sensors. In ultrasonic testing, a sound wave can be sent through a material, and used to probe for defects in it. This functions based one how sound waves will travel through the material and, principally, will reflect off of defects in that material. This ultrasonic testing can also be applied to measure properties of the material, such as temperature, pressure, and vibration. Of the materials chosen for these sensors, AlN has shown itself to be a very attractive candidate for use, due to it's stability and resistance to irradiation.

This project set out to investigate how the behavioral properties of AlN will change under irradiation. This is of particular interest due to it's use in Nuclear Reactors as a sensor, where it's piezoelectric effects can be used as a sensor to monitor the reactor's status[5]. Thus far, AlN has been seen to have stronger resistance to irradiation effect than GaN, and so motivates further development.

Studies of irradiation using molecular dynamics have been carried out many times before. As already mentioned, they've been used with respect to GaN to investigate how defect formation due to irradiation may behave under different circumstances, such as temperature, or differing irradiating energies[2][3]. This precedent work has already established some properties relating defect production and external factors, such at He et al. reporting a dependence on temperature, and Nord et al. investigating threshold displacement energies based on how the irradiated atom strikes the crystal.

Thus, we aim to apply much of the methodology of these simulations, and apply them to the case of AlN. In particular, the relationship between the formation of defects and the energy of the irradiated particle is the goal. This is investigated in two ways. Firstly, the question of how defects form if a particle of some energy strikes the lattice. And secondly, investigating what energy is required to form defects.

2 Methodology

In order to simulate radiation damage of AlN we employed Molecular Dynamics simulations with the LAMMPS package[4]. In these simulations, an irradiated particle known as the knock-on atom strikes the face of the crystal lattice and transfers a significant portion of its energy to the structure as a whole. Following this, the simulation is allowed to run until the entire system has once again returned to an equilibrium. Over the course of this simulation, some atoms may be knocked out of their place in an ideal structure and will instead settle in a different energy minima forming a defects. The hole this atom leaves behind is known as a vacancy. In this work, we only study low irradiation flux, that is, only one knock-on atom will strike the lattice.

The structure of AlN is wurtzite. We began with an orthorhombic primitive cell with 4 atoms and then replicated it to create a supercell containting 640,000 atoms. This size was found to be large enough such that the largest radiation cascade wouldn't interact with itself from periodic boundary conditions, and would also have the supercell present as a roughly cubic form.

In order to run the knock-on simulation, it was split into two parts. Firstly, the system was allowed to run with no knock-on atom to ensure we had reached a properly ideal crystal. This was accomplished by setting an NPT thermostat over the system at 300K, which would give us an appropriate volume for our intended temperature. Once the system had finished equilibrating, the primary damage simulations began. A knock-on atom on the face of the supercell was then given an energy ranging between 1k and 10k eV directed into the system, and was allowed to run under and NVT thermostat.

In addition, we also explored the displacement energy of AlN. In this analysis, the AlN crystal was started from a perfect crystal structure, and the knock on atom would be given a specific energy, where the exact energy is determined by a binary search algorithm. The direction this atom would be displaced towards would be in a random direction as well. In this approach, we search for a threshold displacement energy between by setting an upper and lower at 0 and 200 eV respectively. Then, the midpoint of our bounds were taken as a test energy, and then the simulation would be run from there. After the simulation finished, we can look at the energies at the end and, by comparing them with the energies of the perfect crystal lattice, decide if a defect was formed. Then, we can update the bounds to be higher or lower depending on if there was a defect present, and then repeat. This was repeated until a desired accuracy was obtained, and then that could be reported as a defect energy. It is worth noting, however, that the random direction chosen for the atom does play an effect on the defect energy. As such, this algorithm was run multiple times, and then an *average* defect energy was found.

In Molecular Dynamics, the choice of interatomic potentials to define the system is critical, since it will define how the atoms in the system interact. However, there are very few classical potentials developed for AlN. For this simulation, we choose the Buckingham Style potential with Coulomibc interactions[1] that reproduces reproduce basic behaviors of AlN well. The Ziegler-Biersack-Littmark (ZBL)[8] potential was also employed for very short range interactions.

The Buckingham-Coulomb potential is given by $E_{ij} = E_{Buck} + E_{Coul} = (A * exp(-\frac{r_{ij}}{\rho}) - \frac{C}{r_{ij}^6}) + (\frac{Z_i Z_j}{4\pi\epsilon_0 r_{ij}})$, where the first term in parentheses is the Buckingham potential, and the term in the second set of parentheses is the familiar Coulombic potential. A,B, and C are all constants, while Z_i and Z_j denote the charge on the two atoms, and r_{ij} is the distance separating the two. In the Buckingham potential, the term $Aexp(-Br)$ constitutes a repulsive force, and $-\frac{C}{r_{ij}^6}$ denotes an attractive force. Note that as the two atoms approach each other, the $\frac{1}{r_{ij}^6}$ term will begin to dominate, and will result in a very unphysical attractive force that will strongly bind the two atoms together, and in fact would happen very commonly during the molecular dynamics simulations. This necessitates more consideration onto the exact potentials we should use.

The chosen solution was to employ the ZBL potential at very short ranges.The ZBL potential is a potential that simulates two particles at very small separations, where the interactions between the 2 atoms can be seen as being dominated purely by the coulombic interactions of the positively charged. The ZBL potential is given by $E_{ij} = \frac{1}{4\pi\epsilon_0}\frac{Z_i Z_j e^2}{r_{ij}}\phi(r_{ij}/a) + S(r_{ij})$. a is a constant dependent on the charges of the 2 atoms, given by $a = \frac{0.46850}{Z_i^{0.23} + Z_j^(0.23)}$. $\phi(x)$ is defined to be $\phi(x) = 0.18157e^{-3.19980x} + 0.50986e^{-0.94229x} + 0.28022e^{0.40290x} + 0.02817e^{-0.20162x}$. Important to note is that as the distance goes to 0, $\phi(\frac{r_{ij}}{a})$ goes to 1, where the potential is purely Coulombic. $S(r_{ij})$is a switching force included to ensure that the force, energy, and curvature will smoothly ramp down to 0 between cutoffs.

In order to ensure that the overall potential we employed would smoothly transition between the two, the ATSIM[6] program was used to create a smooth spline between the short range interactions, dominated by the ZBL potential, and long range interactions, which are dominated by the Buckingham potentials.

In order to gather data once the simulations had been finished, the OVITO visualization program[7] was used. OVITO can read output from LAMMPS and use it to create visualizations of how the strucutre of the cell evolved over time. OVITO also comes with a Wigner-Seitz Analysis program, which was used to identify vacancies, interstitials, and antisites. This was the main method of analyzing the structure once it

had re-equilibrated after the knock-on simulation.

The Wigner-Seitz analysis program defines vacancies and interstitals by counting how many atoms reside in a Voronoi Cell. The Voronoi Cells are defined with respect to some reference structure (in this case, the equilibrated structure), and are structured such that each cell contains only one atom. At the end of the simulation, Wigner Seitz analysis will calculate the how many atoms reside in each Voronoi Cell. If there are no atoms in the Voronoi Cell, it's defined as a vacancy. If there are 2 or more atoms in a Voronoi Cell, it's defined as an interstitial. Additionally, if at the end of the simulation, the *type* of atom occupying a cell is different than the one inhabiting it originally, it is defined as an antisite.

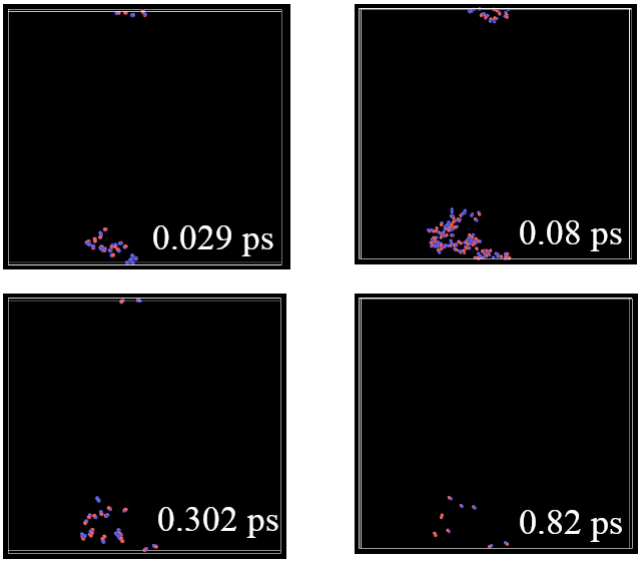
3 Results

3.1 Production of Defects

As the knock-on atom first collides with the crystal lattice, it will immediately dump much of its kinetic energy into the surrounding atoms. This sudden spike of energy will continue as those atoms will also begin to displace more and more atoms surrounding it, creating the radiation cascade. Due to this high energy displacement at the very beginning of the irradiation, defects are almost surely formed. During this initial knock-on is when the maximum number of defects will be formed. As the simulation continues however, some interstitials and vacancies will begin to anneal as the system beings to return to equilibrium. This results in the total number of defects declining as the run continues, until eventually the system comes to rest at another equilibrium which will contain some defects inherent in it's structure.

By visualization and Wigner-Seitz analysis, this process of an increasing number of defects and then them dropping down can be clearly seen. In the following 4 snapshots, this behavior can be seen very clearly.

Figure 1: 4 Snapshots of all defects due to a 4 keV knock on atom at different points in time. Size of atoms exaggerated for clarity.



These are taken in chronological order and show the total defects at different points in time. As is to be expected, at the very beginning defects very quickly form as the knock-on atom deposits almost all of it's kinetic energy at once. This causes the radiation cascade to begin forming, and ballooning outward from where the knock-on atom first struck the lattice. Of note is that as the knock-on atom recoils from it's first

collision, it will begin to travel backwards from where it came, and eventually cross the periodic boundary and being to interact with atoms at the top of the structure. However, at this point the atom has already deposited much of it's kinetic energy and so it's impact will be much less extreme compared to the initial knock-on.

By the time the system has settled into another equilibrated state, many of the interstitials and vacancies have disappeared, leaving behind a equilibrated structure with much fewer defects inherent in it's formation. This comes about because defects that are close enough together can spontaneously recombine with each other, thus dropping the total number of defects. However, in a more realistic scenario where the AlN will be continually bombarded with more irradiated particles, this irradiation will continuously produce more defects.

Energy Step	Maximum Interstitials	Maximum Time	End Interstitials	End time
1 KeV	32	.07 ps	1	0.55 ps
2 KeV	41	.0693 ps	4	0.6 ps
3 KeV	46	.0618 ps	6	0.7 ps
4 KeV	117	.08 ps	8	0.82 ps
5 KeV	137	0.12 ps	8	0.7 ps
6 KeV	178	0.1582	12	2.78 ps
7 KeV	207	0.125 ps	18	0.9 ps
8 KeV	216	0.1 ps	14	1.322 ps
9 KeV	273	0.122 ps	14	1.287 ps
10 KeV	297	.1105 ps	17	1.1005 ps

3.2 Displacement Energy

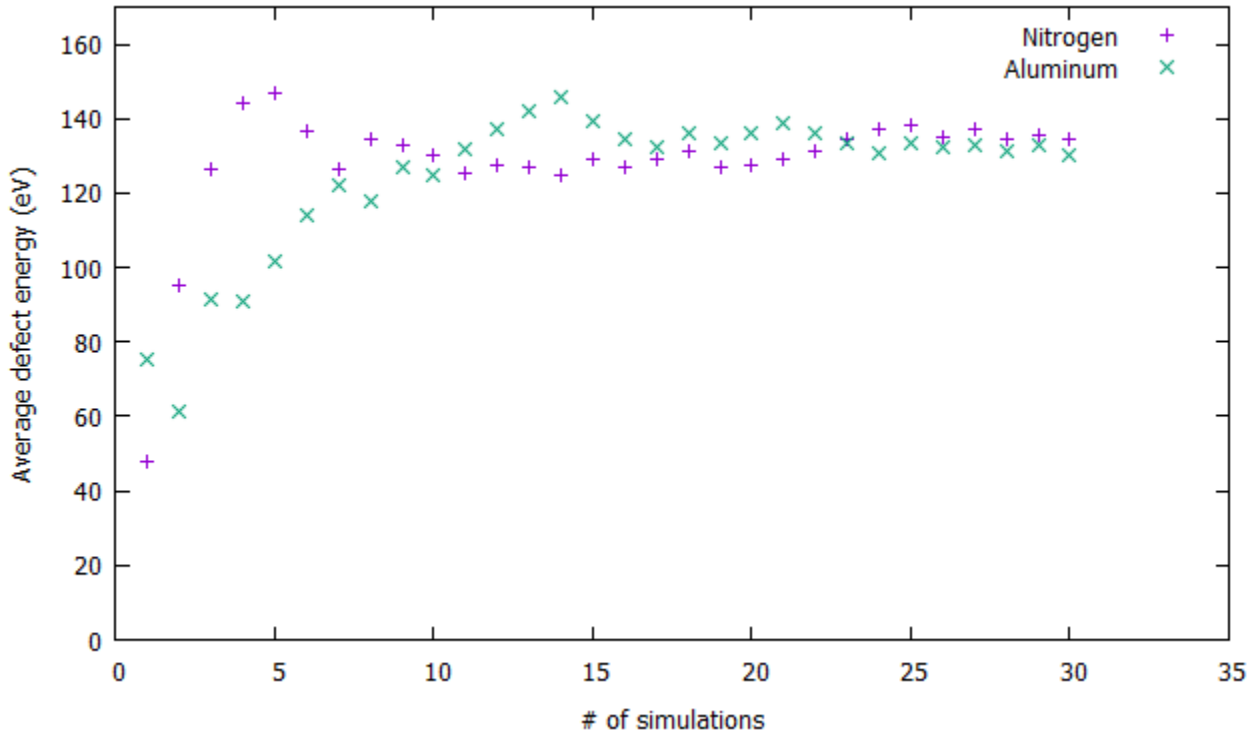
Defect energy is a statistical value important to the study of damage calculations. It is defined as the lowest energy required in order to create a permanent defect if the atom is sent in a certain direction. It is also, however, a statistical value by nature. So in order to obtain an accurate value, the binary search tree algorithm must be run multiple times over, and then averaged to obtain a reasonable result.

In the interest of efficiency, the system we tested our defect energies on also differs from our investigation into defect production. Since our knock-on atom is now travelling much slower than any of the previous simulations, the size of the simulation cell can be decreased in the interest of efficiency. This new cell has 17,280 atoms, and was once more chosen with the interest of ensuring that the radiation cascade never interacts with itself.

In addition, we also investigated whether type of knock-on atom would play a significant role in the average defect energy. And so, in addition to running the simulations as done previously, we also elected to have one set of binary search algorithms work when an aluminum atom is displaced.

The graph shows very clearly that there appears to be no dependence on particle type when it comes to displacement energy, suggesting that it's more dependent strictly on energy of the knock on atom. In addition, the defect energy exhibited here is much higher than the defect energy of GaN. While the work of Huen et al. reported an average displacement energy of about 50.1 eV, the displacement energy here shows to be on the scale of 130.

Figure 2: Average defect energy vs Number of simulations run



4 Conclusions

We used the Molecular Dynamics Package LAMMPS extensively in our analysis of AlN and how it's properties change when it is subject to irradiation, and the subsequent defect formations. From this, we found multiple dependencies on the energy of the knockon atom with regards to how extensively defects are formed, and what energies are required to form defects.

In regards to how defects are formed, we discovered that there is a strict dependence on the knock-on energy of the atom. This range included the total number of interstitials, at both maximum and minimum time, and the timeframe over how these occur. Specifically, higher energies correlate to higher numbers of defects at both the beginning, and end of the simulations. As well, the timeframe to reach those expands, meaning that as the energy of the knockon atom increases, the system will take more time to anneal out all of it's defects. This however, did not seem to be as completely linear as expected.

In addition, a limited number of runs were used to investigate the defect energy of AlN. In addition, this setup was run with both Aluminum and Nitrogen as knock-on atoms to investigate if there were any major changes. Based on these small number of runs, it appears that there is no significant dependence on atom type, Aluminum or Nitrogen, on the average energy defect. From this, it seems that the most important factor in average energy defects (without respect to direction) is strictly based on how energetic the incoming particle is.

Of interesting to note is the comparison to GaN that has been worked on previously. When compared to the work of He et al. on GaN, our AlN structure seemed to be much more resistant to overall damage. This manifestedin multiple ways, including a lower number of maximum and minimum defects, faster time to anneal, and a higher average defect energy. All of this combined point to AlN as being a material resistant to irradiation as a whole.

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