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Evaluation of Americium’s Molecular Dynamics Pair Potential for use in Alphavoltaic Modeling

There are two primary means to design a device of capturing the radioactive decay. The first method used in deep space probes and remote hardwares, called a radioisotope generator, utilizes the heat from the decay of a large number of radioactive materials, which is expensive and heavy; and the other method is by the direct decay capture in a semiconductor device that is called an alphavoltaic or betavoltaic power supply. Most of these devices exist as prototypes and suffer from a low efficiency and short lifetime.\(^1\) The focus of this research project is on the second kind.

An alpha emitter was selected based on its power and well-defined stopping distance, or penetration depth of the alpha particle into the semiconductor, which are more applicable than a beta emitter. The power generation has two types, i.e., direct or indirect. In an indirect type, radiation is absorbed by a secondary material and then is reemitted at a different wavelength. Since the emitter size is a primary concern, the direct method will be used to limit the amount of materials exposed to radiation. The problem then is that the heavier alpha particles cause more material damage than the beta ray. So testing more resilient materials becomes critical for alpha emitters.

Before building of a physical device, to save on cost and potential contamination hazards, molecular dynamics (MD) simulations are performed. To start an MD simulation, the force between any two molecules must be well defined. There are many types of force calculations with
varying levels of complexity. In this project, because of the metallic nature of the isotope, it is desired to use an embedded atom method (EAM) potential. The EAM potentials do very well at describing the long-range order presented in metallic systems. Upon investigating the literature, no such potential exists. An alternative one is found, i.e., the Lennard-Jones (LJ) potential that only requires a few constants to be specified. The available americium LJ potential was of the 12–6 form:

\[ E = 4\varepsilon \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^{6} \right] \]

where \( \varepsilon \) is an energy constant, \( \sigma \) is a distance constant, and \( r \) is the distance between two atoms.

The problem is that while the required constants exist, they have not yet been validated. The primary purpose of this project is to validate the LJ potential to allow further development of an alphavoltaic power supply.

**Simulation Setup**

Very little information of americium is available to the public. To validate the LJ potential, three metals have been chosen, i.e., titanium, uranium, and americium. Titanium is selected to take advantage of my previous work, as all material properties are well known. Uranium is selected because it is the nearest element to americium that has sufficiently available data. The behavior of americium follows the same trend as uranium over a temperature range. Even though no EAM potential exists for americium, an EAM calculation was made for the other elements so a secondary evaluation could be made to ensure the correctness. And due to the wide range of material properties to be tracked, two simulations have been run for each metal at each selected temperature. One simulation is made to calculate thermal conductivity and diffusivity. The other is for the calculation of density, potential energy, volume to find the melting point and thermal expansion coefficient. The reason for the split simulations has to do with the MD control scheme used to regulate the energy inside the system changes depending on critical values that need to be conserved. For the melting simulation, an NPH (that is constant pressure, constant enthalpy) scheme was used to prevent the thermostat in the simulation from artificially altering the energy; while an NVE (that is constant energy, constant volume) scheme is used to maintain the system’s temperature and volume.
Results

Titanium data show that the LJ and EAM potentials behave similarly until the temperature approaches the melting point. Typically, at the melting point, metal atoms’ self-diffusion rises in a non-linear fashion, because the atoms instead of being trapped in the lattice are now able to freely move around each other. While not as the primary purpose of the diffusion calculation, it did provide a secondary means to check for melting. The LJ potential does not make the melting transition of phase change for titanium as seen in Fig. 1. However, the EAM potential does make the transition but not as quickly as the reference data due to the instability of the EAM potential at higher temperatures. The melting and thermal expansion simulations are still running.

The uranium simulations were all completed and much like titanium simulations, the diffusion data from the LJ potential did not trend as expected at higher temperatures but despite this its low temperature data gave closer to the reference. The EAM diffusion as seen in Fig. 2 did however give proper response, and, again like the titanium curve, was separated from the reference data early and did not rise as quickly as the reference data. The EAM uranium melting simulation was constructed as infinite crystal by using the wrapped boundaries that gave a density of 18.5 g/cm$^3$ and a 3% error compared to the actual value of 19.1 g/cm$^3$. The linear thermal expansion was 0.00000224 K$^{-1}$ compared to 0.0000139 K$^{-1}$ resulting in a large error can be attributed to the mismatch in the lattice type between the simulated uranium and natural uranium. Natural uranium has an orthorhombic structure; but to simplify the mathematics, it is needed to generate the potential constants in a body centered cubic structure as the lattice for the simulated uranium. At

![Figure 1: a) Calculated diffusion coefficients of titanium with reference data, b) Magnified view to better show EAM melting behavior.](image-url)
the melting point, the infinite crystal did not melt due the restrained nature of the crystal, which was verified by running the simulation again but this time with an exposed face. The potential energy plot made a sharp turn-up indicating the released energy associated with melting. After evaluating the uranium and titanium simulations, the LJ potential is only suitable for low temperature applications and will need significant tuning to generate correct properties.

The americium simulations were carried out using the LJ potential despite the apparent unsuitability for us. While no information is available for diffusion comparing the low temperature response of titanium and uranium to that of americium, our results still indicate that LJ does not perform well. The plotted diffusion immediately goes non-linear when it should have experienced a slow rise until approaching near the melting point. While attempting the melting simulation, the potential failed to produce a stable temperature control needed to perform a calculation of thermal expansion.
All the simulations have been run on a purpose-built Raspberry Pi cluster made of 15 cards functioning as individual nodes of a single computer. The diffusivity simulations took a shorter amount of time compared to the melting simulations. Total run time is approximately 650 hours (that is 27 days) for the simulations with data outputs, and none of the equation and temperature control setup has been recorded to minimize the usage of the hard drive space. In general, the LJ simulations take longer than the EAM simulations of the same type with the total amount of data generated by both is about 34 GB. And currently many simulations are still being run to finalize the data set for titanium.

**Conclusion/Future Considerations**

To continue working with americium, a different potential will have to be used and more modern information is required. Current efforts are hindered by the only available data being the piecemeal declassified papers from the 1950s and 1960s and even so, there were large discrepancies in these papers such as the reference diffusion values used in Figure 2. An LJ style potential may still be capable for americium calculations. When the original LJ potential was generated, it was designed for the inert (noble) gases, however, the attractive and repulsive elements can be altered by changing the exponents from 12 and 6 to a value that better reflects the metallic behavior of the element. 

Without much reliable data on americium, another means of testing for material properties is desired. An initial investigation has been made to perform a first principles calculation. However, much like no validated potential exists for conventional MD simulation, the same was also true for the first principles calculation in that no validated pseudopotential exists. If a pseudopotential can be made, then an EAM potential can be constructed, based on which a conventional macroscale continuum simulation can be made as well.

**Bibliography**


4) Self-Diffusion in Gamma Uranium. July 1959


