**General comments from the authors:**

Given the ability to revise the manuscript and the comments from the reviewers, the authors decided to spend the full allotment of time to continue this work and present a more complete picture of results. Now, an updated U-Si MEAM interatomic potential is presented within the paper. The methodology for fitting the potential has been slightly modified, adding in a step to modify pure Si MEAM parameters in order to fit U3Si2 properties. We believe this more fully illustrates the possible utilization of the MEAM formalism in U-Si and the inherent difficulty associated with the accurate description of this complex system.

The result is that the new potential has various advantages over the previously presented potential. These include a dramatically higher melting point, destabilization of a metastable phase discovered in the revision process, and improvement of point defect properties. The one drawback has been a slight degradation of elastic properties.

The authors continue to believe that once there is further information provided on this system, these potentials can (and should) be refined in the future.

Comments addressing reviewer concerns are below in blue.

**Comments from the editors and reviewers:**  
-**Reviewer 1**  
  
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This paper reports on a new MEAM potential for the U-Si system. The model may be used to support some grand and (notably) as-yet unwritten multiscale fuel performance code of new, accident tolerant fuels. This is an area of some current interest to the nuclear industry although the fuels themselves are still many years from standard use.

The paper is a record of the potential derivation with no new physical insight into the system and the authors have followed a well-trodden path in fitting their potential. I don’t have a problem with this, a good potential model is very valuable and would be highly cited, but I’m concerned that this comes across as a work in progress rather than a finished, validated potential. The authors even report this in their discussion, ‘...*future refinement of the potential should be undertaken once more atomistic and fundamental property information has been gathered*’. In particular, I was surprised and a bit disappointed at the poor replication of the DFT point defect values in spite of a nominally robust fitting process: this undermines the use of the potential in cascade simulations and also prevents the accurate prediction of fission product migration because the defect chemistry will be all wrong. A key, unanswered question for me was whether this is the best we can expect from a parameterised potential or whether future work should focus on fitting using a different (non-EAM) formulation, or by refitting the original Moore parameters or just by including more/different fitting data?

This has been (continues to be) explored along a parallel line via fitting a Tersoff U-Si potential as well as refinement of the Moore UZr potential. Both attempts have proven less successful in describing the U-Si system than the approach presented in the manuscript. It is theoretically possible that improvements can be achieved in the description of U3Si2, but at the cost of losing accuracy in the pure systems. Given that a variety of potential formalisms have been employed to describe pure U and that none are without flaws, generating a near-perfect potential for U3Si2 is currently exceedingly challenging. The authors feel that given the available potentials, the fitting methodologies and the fitting targets, this process has been a success.

Nevertheless, this merits publication. It’s a new contribution to an interesting system, the authors have followed an entirely sensible approach, and a lot of the validation (or not) of the potential will follow once it is used in anger to predict real fuel properties. I’ve condensed my concerns down into three main points, which require some thought, and then a bunch of minor stuff that should be revised before re-submission.

**Major Points**

1. **Applicability of Potential:** The discussion section should have a couple of paragraph outlining what this potential could, and more importantly, should not be used for. E.g. surely the poor replication of the U and Si intrinsic defect energies mean that the cascade simulations don’t represent reality? Whereas the reasonable elastic properties and thermal expansivity mean there’s a host of simulations of grain boundaries, dislocations, bubbles etc which would be okay? The use of these fuels will rely on accurately predicting the loss of thermal conductivity due to the accumulation of fission products and radiation defects. So does / will the potential predict the thermal conductivity of the U-Si phases correctly?

The discussion section has been expanded to include details of what the potential is suitable for and what the potential is not suitable for. As for thermal conductivity explicitly, the major component of the thermal conductivity will be electronic, not phonon-based, and thus molecular dynamics is not the most suitable tool to explore such phenomena. However, given reasonable elastic constants, phonon modes should also be reasonable, and the phonon contribution to thermal conductivity can be calculated.

1. **Cascade simulations:** Is the short range repulsion in the potential physically sensible given that it’s fitted to largely equilibrium bulk properties which will have energies of fractions of an eV? I.e. in a ~1KeV U-Si collision (as will occur in the cascade simulations) are the forces generated from the potential accurate? In an oxide this would be handled by a spline curve to a dedicated short range potential. What is done here to ensure these are correct?

The potential is splined to a ZBL potential to ensure proper short-range behavior at high energies. This detail has been included in the manuscript.

1. **Surface simulations:** I wasn’t sure this added a lot to the paper, the numerical stability of the potential more or less follows from the sensible elastic constants. If included, it needs a lot more details: what is the supercell size, what is the vacuum gap, how is the cell relaxed (are x- and y- allowed to vary independently)? Please put (three) crystallographic axes on diagrams. Where are the DFT results from (Ref. 49 reports their use in a rate theory model but not the original work)? The comparison with the DFT results is poor, but this is not unexpected as they will presumably be at 0K and with a much smaller supercell. What happens if a small cell is relaxed at 0K using the new potential? If there’s a genuine 30% drop in the {100} surface energy due to relaxation, which doesn’t occur with the {001} surface, then I think this would dramatically change the expression of the different surfaces (for example in bubble facets) and is an interesting result that needs reporting.

The surface simulation section has been expanded such that interfacial energies have been included, as well as the void surface energy reported. More details on the simulations have also been included. This result can directly be used in phase field simulations or rate theory simulations as an input parameter for surface energy. The DFT results that were included for comparison have been published as part of a rate theory study. The authors of that study have communicated that a full paper that includes this surface energy work will be published at a later date. We felt it was important to include a comparison with any data available, so this citation and the comparison have been included.

**Minor Points**

1. **Page 2, Paragraph 2** - you’ve confused ‘molecular dynamics’ with ‘molecular dynamics using pair potentials’. You can do MD with DFT, it takes longer but the maths is the same.

Paragraph has been lightly edited to include the concept of MD-DFT, distinct from MD with pair potentials.

1. **Page 6**, I didn’t like (or I didn’t understand) the statement ‘*A variety of other simulations were performed to fine-tune the potential, the entirety of which is beyond the scope of this paper’*. The use of LAMMPS to simulate properties which are then fed into some sort of least squares fit is a good one, but this statement is just clouding the issue. What properties from which phases were used to fit the potential?

Not all simulations that were performed to fine tune the potential were included as fitting targets. Many simulations were performed periodically as sanity checks and served to inform the targets. It was our belief that only the fitting targets should be explicitly included within the paper. This paragraph has been refined to more clearly represent this information and the targets more clearly stated. Examples of such sanity checks have been included.

1. **Tables 1 & 3:** Units and please make the variable names consistent with your EAM/MEAM derivation in Section 2 (e.g. what is esub, asub etc?). *C*min/max values need clarification as to which is species ‘1’ and ‘2’.

Provided units where applicable. Defined 1 and 2 species. Modified labels to more closely match equations.

1. **Table 4:** Either units or I guess alternatively specify what internal units LAMMPS is using for these values to be correct (units metal / atom\_style atomic?).

These are unitless. Mostly on/off switches. Except for delr, where units are included. The information as now presented is complete for any users wishing to reproduce our results.

1. **Figures 2 & 3:** I didn’t really see the point of these and Figure 4 seemed to convey the same information much more succinctly. JNM readers are hardly going to get their rulers and protractors out to measure your distances / angles. If you want to put a proper crystal structure diagram of U3Si2 in then please do so and talk about the distortion in the text.

In place of the two figures, a single figure displaying the experimental structure is now shown, with discussion of the deviations between the experimental and predicted structures in the text.

1. **Table 5 & 6:** Units.

Units input in captions.

1. **Figure 7:** Values should be normalised to the system size, so per formula unit or per atom. Is the zero of energy the atoms isolated at infinity?

The figure has been regenerated with energy per atom and volume per atom instead of totals. The energy of a system is zero when all atoms are beyond the cutoff radius with respect to one another, which is 6 angstroms, as defined in table 3.

-**Reviewer 2**  
  
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The authors report the derivation of a set of interatomic potential parameters for modelling U-Si compounds. The topic is well in line with the Journal's theme and addresses novel issues with state-of-the-art techniques.  In view of the quality of the study and the promising capabilities of the potential, I recommend to accept the manuscript for publication into the Journal of Nuclear Materials.

Herewith some minor comments / typo corrections which could be addressed by the authors:

\* p3, under eq.2: trailing "{" sign before E^u(R)

Brace is meant to be there, but it is unclear in the formula, the size of the brace has been increased to more clearly illustrate the equation.

\* p4, eq.7: ln is a function => not in italic (use "\ln" instead of "ln")

This has been corrected.

\* p6: "A variety of other simulations were performed to fine-tune the potential, the entirety of which is beyond the scope of this paper." => Is it described somewhere? Some more details might interest readers who would like to apply a similar

procedure.

Not all simulations that were performed to fine tune the potential were included as fitting targets. Many simulations were performed periodically as sanity checks and served to inform the targets. It was our belief that only the fitting targets should be explicitly included within the paper. This paragraph has been refined to more clearly represent this information and the targets more clearly stated. Examples of such sanity checks have been included.

\* p 10: the distortion is not clearly visible on the pictures. Could it be indicated more clearly? One speaks of distances between atoms, but the types of atoms is not indicated. Is it U-U, U-Si or Si-Si distances?

In place of the two figures, a single figure displaying the experimental structure is now shown, with discussion of the deviations between the experimental and predicted structures in the text. It has been added that the first nearest neighbor peak is a Si-Si pair.

\* Figures 2 and 3: the caption could provide more details on what to observe

More discussion has been included in the text regarding the consolidated figures.

\* p 15: no phase change observed: is the system large enough to observe a phase change? Phase changes typically initiate in a small region and propagates; the small system size may inhibit such events.

This is meant to imply a crystal structure change. Thus, the word ‘phase’ has been changed to ‘crystal structure’. Such systems are large enough to observe a change in the crystal structure with applied temperature.

\* p17: damage after a cascade is reported, as a mean to 'validate' the potential. Although limited experimental validation exists, one might compare the results to theoretical expectations (NRT law?)

The difficulty in this is the lack of threshold displacement energy. Thus, a value cannot be generated for displacements via NRT, unless an estimate of the displacement energy is made in some way. This is largely to illustrate that the system behaves ‘typically’ under irradiation.