Lattice Expansion by Intrinsic Defects in Uranium by Molecular Dynamics Simulation

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Abstract:

A re-formulated and re-parameterized interatomic potential for uranium metal in the Charge-Optimized Many-Body (COMB) formalism is presented. Most physical properties of the orthorhombic α and bcc γ phases are accurately reproduced. In particular, this potential can reproduce the negative thermal expansion of the b axis in α -U while keeping this phase as the most stable phase at low temperatures, in accord with experiment. Most of the volume expansion in α -U by intrinsic defects is shown to come from the b axis, due to the formation of prismatic

loops normal to this direction. Glide dislocation loops forming stacking faults are also observed. Structures of both loop types are analyzed. An expansion simulation is conducted and the results are verified by using the Norgett-Robinson-Torrens model. Rather than forming extended defect structures as in α -U, the γ phase forms only isolated defects and thus results in a much smaller, isotropic, expansion.

1. Introduction

As of 2015, nuclear energy accounted for 19% of the US electricity production [1]. All of this energy is produced in pressurized water reactors (PWRs) and boiling water reactors (BWRs), which use uranium dioxide (UO_2) as the fuel. UO_2 exhibits good structural stability under irradiation, but its low thermal conductivity ($<5 \text{ W/mK}$) results in a high fuel centerline temperature and large radial temperature gradient across the fuel pellet, which ultimately limits the operating temperature of the reactor and the total power throughput of the nuclear plant.

Metallic fuels in fast reactors have smaller thermal gradients due to the fuel's much higher thermal conductivity ($\sim 30 \text{ W/mK}$). Because of this, despite lower melting points, metallic fuels actually have larger operating margin than oxides [2]. In addition, metallic fuels of U alloys (e.g. U-Zr or U-Pu-Zr alloy) have a higher density of fissile atoms and are easier to fabricate and reprocess than oxide fuels. Due to the absence of lightweight atoms (such as oxygen), they also thermalize fast neutrons less, keeping the neutron energy spectrum hard so that more fissile atoms can be bred. Examples of reactors that have employed metallic fuels include the fast spectrum Experimental Breeder Reactors I and II (EBR-I and EBR-II) in the US, the Dounreay Fast Reactor and the thermal Magnox reactors in the UK.

Because ^{235}U is a non-renewable and decreasing resource, ^{239}Pu must be utilized for sustained energy production by irradiating (breeding) ^{238}U . The U-Pu-Zr metallic alloy is a good fuel candidate for this purpose since the addition of Zr can increase the melting temperature of U-Pu alloy while suppressing the diffusion of cladding materials (Fe, Ni) into the fuel [3]. Nevertheless, uranium is still the main component of this alloy and many physical properties of pure and alloyed uranium are similar. Thus, a detailed understanding of pure uranium is critical to the investigation on the U-Pu-Zr properties and to the development of a metallic fuel system.

At one atmosphere, uranium exists in the orthorhombic α form at up to $\sim 1000 \text{ K}$ [4], at which point it transforms to tetragonal β and eventually to bcc γ phase, which melts at 1405 K [5]. At higher pressures ($>3 \text{ GPa}$), only the α and γ phase exist with a pressure-dependent phase

transformation temperature [6]. All three allotropic forms of uranium may be present under reactor operating conditions. The unit cell of α -U consists of four atoms as shown in Fig. 1. When the four nearest neighbors of each atom are bonded, the structure can be visualized as corrugated sheets extending in the *a* and *c* directions. The parameter *y* in atomic coordination controls the extent of corrugation.

Fig. 1. The structure of orthorhombic α -U in $3 \times 2 \times 2$ supercell. One unit cell has four atoms and they are represented by four colors (pink, brown, green and purple). All atoms/colors are equivalent (in the same Wyckoff position).

Uranium fuel shows substantial swelling, which can be classified into four types [7]: swelling by solid fission products, swelling by fission gases, breakaway swelling, and swelling from cracks and tears along grain boundaries to form microstructure of irregular cavitation. Each type of swelling has a characteristic degree of volume increase [8]. While all the swelling phenomena can be well measured and characterized experimentally, the detailed defect evolution processes and mechanisms leading to these types of swelling are not all well understood. The main swelling phenomenon in α -U is tearing and cavitation which, it has been suggested, results from anisotropic growth of individual grains with preferred orientation texture [3]. This is caused by the accumulation of interstitial loops in the (010) plane and vacancy loops in the (110) plane, giving rise to incongruent strain between grains whose stress is then relieved through plastic

deformation. By contrast, swelling in γ -U is mainly caused by agglomeration and expansion of gas bubbles in the relatively soft (compared to α -U) matrix [3].

Molecular dynamics (MD) simulation is a powerful tool to study dynamical phenomena since it is able to determine, track, and record the position of every atom in the system. However, this method has only recently become available for uranium as suitable interatomic potentials have been developed. Now, with several uranium potentials published in a short period of time, we use two such potentials for large-scale simulations – the Charge Optimized Many Body (COMB, refined in this work) potential and the Embedded Atom Method (EAM) [9] potential – to make a first attempt to investigate the swelling of uranium by intrinsic defects. The simulations are performed with the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) ^[10]. Since the extent of this swelling type is small ($< 1\%$ in our simulations), it would also be appropriate to refer to it as “expansion” rather than “swelling”, while the latter term is usually assumed to be mainly attributable to insoluble, extrinsic gaseous bubbles.

In the next section, all available uranium potentials of various formats will be compared to evaluate the quality of our COMB potential. Section 3 presents the simulation results on α -U by COMB; the defect structures formed during simulation will be analyzed and the expansion will be compared to experimental work using the Norgett-Robinson-Torrens (NRT) model. Some results for γ -U are also presented. Section 4 contains our conclusions.

2. Interatomic potential for U with improved performance

Despite the development and application of atomic-scale simulations using classical empirical interatomic potentials for a wide range of materials in the past thirty years, potentials for metallic uranium have been conspicuously absent. This absence can be attributed to the difficulty in modeling a material with such a low symmetry ground state structure (Fig. 1). The tetragonal β phase is usually not considered in MD potential development and has not been included in any existing potentials so far due to its limited stability range (temperature range less

than 110 K, pressure range less than 3 GPa [6]) and its structural complexity [11]. However, the bcc γ phase, which has a wide stability range [6] and can be stabilized at low temperature by alloying, should be included in addition to α in order to maximize the range of applicability of the potential since high-temperature properties of uranium are particularly important for reactor-related research. Thus, the combined effect of a low-symmetry ground state, a polymorphic system, and the need for an acceptable level of accuracy resulted in the long absence of potentials for U, even after many potentials of its oxide (UO_2) had been published [12-14].

Four years ago, almost at the same time four different U MD potentials were published in the frameworks of COMB [15], EAM [16, 17], and a customized type which the creator called long-range (LR) [18]. The COMB potential mainly considered some static and dynamic properties of α -U, and was later expanded to include gaseous O_2 and fluorite-structured UO_2 , thereby becoming the first unified potential for these three systems [19]. The EAM potential developed by Smirnova [16] considered both α and γ phases, as well as their dynamic transformation process. It was later expanded to include Mo, U_2Mo , U-Mo bcc alloys and gaseous Xe to become a unified potential of the U-Mo-Xe system [9]. The EAM potential developed by Pascuet [17] considered properties of α -Al, α -U, γ -U and three U-Al intermetallic compounds, and is used to describe the diffusion properties of U and Al atoms in the uranium matrix. The LR potential developed by Dai included U, Nb and polymorphs of U-Nb alloys. Recently, a Modified EAM (MEAM) potential [20] was published with the emphasis on defect migration energies. This potential predicted a new $\alpha \leftrightarrow \gamma$ transformation path and demonstrated that the diffusion mechanism in α -U is vacancy-controlled. All potentials model the orthorhombic phase as the primary ground state but also take the high-temperature γ phase into consideration to varying extents.

Generally, the quality of an MD potential is evaluated by four criteria: flexibility (the ability to simulate multiple phases), accuracy (the fidelity of the calculated properties),

transferability (the ability to predict properties that are not considered in parameterization), and efficiency (the calculation speed in a computer) [21]. The following sub-sections will compare COMB with other existing potentials on the first three criterion.

2.1. Comparison of the COMB and EAM potential

Building on previous experience, we have developed a new COMB potential which provides a high-fidelity description of the cohesive energy, lattice dimensions, bulk moduli, defect formation energies, coefficient of thermal expansion (CTE) and transformation temperatures in α and γ , albeit by sacrificing some fidelity in the description of the α -U elastic constants (c_{ij}). The database for MD potential parameterization is obtained from experimental values and/or ab-initio calculations using Density Functional Theory (DFT) with the Generalized Gradient Approximation (GGA). Comparison between experimental measurements and DFT calculations provides verification of the accuracy of the database.

The basic formalism of the COMB potential used here has been presented previously [15]. This version of the uranium potential has a slightly different formalism, with the addition of two more attraction terms to the energy equation (Eqn. 1 in reference [15]):

$$E_i = \frac{1}{2} \sum_{j \neq i} f_s(r_{ij}) \left[A e^{-\lambda r_{ij}} - b_{ij} \left(B_1 \exp\{-\alpha_1 r_{ij}\} + B_2 \exp\{-\alpha_2 r_{ij}\} + B_3 \exp\{-\alpha_3 r_{ij}\} \right) \right] \quad (1)$$

B_2 , α_2 , B_3 and α_3 are the four new fitting parameters. The original bond-order term (Eqn. 3 in [8]) is now replaced by:

$$b_{ij} = \left\{ 1 + \left[\sum_{k \neq i, j}^N \left[F_C(r_{ik}) \exp(\beta_{ij}(r_{ij} - r_{ik})) g_{ij}(\cos(\theta_{jik})) \right] \right]^\eta + P_{ij}(\Omega_i) \right\}^{-1/2\eta} \quad (2)$$

The coordination term P_{ij} has been moved outside of the η -power bracket which describes the effect of many-body interaction. The new form eliminates a mathematical breakdown for some of the non-metals parameters since their P_{ij} are negative and cannot be raised to a power of a

fraction (η can be a fractional number). The old [22] and new formalisms of COMB are expected to have the same degree of flexibility to model a particular system. Unfortunately, even such a modest change in formalism usually means a total refit for systems with $\eta \neq 1$, which is the case for uranium [15]. The refitted U parameters for the new formalism are shown in Table 1. Almost all parameters in the new formalism have been changed completely.

Table 1: The parameters in the new COMB formalism for U. Most variables are defined in the previous version [15].

A (eV)	1208.055298
λ ($\text{\AA}^{-1}$)	2.385540
B ₁ (eV)	227.336212
α_1 ($\text{\AA}^{-1}$)	1.167905
B ₂ (eV)	203.804199
α_2 ($\text{\AA}^{-1}$)	1.061615
B ₃ (eV)	-77.431190
α_3 ($\text{\AA}^{-1}$)	0.945080
η	0.201069
β	2.799958
b ₆	-0.024662
b ₅	0.091080
b ₄	0.251867
b ₃	0.161477
b ₂	0.100591
b ₁	0.048940
b ₀	0.015389
R ^{min} ($\text{\AA}$)	3.7
R ^{max} ($\text{\AA}$)	3.85
c ₀	0.009953
c ₁	0.263795
c ₂	-0.244010
c ₃	0.038277

2.2. Refitted properties of α -U

Table 2 shows some key structural and mechanical properties of α -U for the refitted COMB potential, other MD potentials, and DFT calculations. For better comparison, those DFT works [23, 24] that did not perform elastic calculations are not listed. All DFT calculations are consistent in reproducing the lattice parameters when comparing to experiment, but over-estimating the elastic constants. The performance is not much improved even when all electrons are taken into calculations [25]. There is only one measurement of the elastic constants [26]. Extrapolation of the elastic constants at room temperature to 0 K tends to increase c_{ii} [25, 27], closing the gap between experiment and DFT calculations.

All MD potentials predict the cohesive energy and lattice parameters close to the experimental values. In order for COMB to obtain better dynamic properties and good point defect energies for both the α and γ phases, in this version some of the elastic constants are allowed to deviate from experimental values by more than 50%, especially the shear constants c_{ij} ($i = 4 - 6$). This is justifiable since elastic constants in α -U are known to be difficult to predict even with the high-fidelity DFT methods; our parameterization experience also shows that placing less tight control on shear constants can lead to significant improvement in the overall quality of the potential. All other MD potentials have similar issues as COMB in that their elastic constants also have large deviations from experimental or DFT values. Nevertheless, c_{ij} in COMB have the highest accuracy among all MD potentials.

One particularly difficult task in uranium potential development is to capture the anisotropic lattice dimension change in α -U. As temperature increases, uranium shows expansion in the a and c axes but contraction in the b axis; the total volume increases, as does the γ parameter. This gives positive and negative coefficients of thermal expansion (CTE) in different directions. Fig. 2 shows the variation of lattice dimension and volume calculated by the COMB (this work) and EAM potential [9] as a function of temperature. Experimental data [28] are shown for comparison. All curves are normalized by their values at 0 K. Both the COMB and

Table 2: Key structural properties of orthorhombic α -U calculated by MD potentials (COMB, MEAM, EAM and LR), DFT (GGA and full potential), as compared with experiment. E_c : cohesive energy; y: coordinate parameter (see texts explaining Fig. 1).

	MD					DFT					Experiment
	COMB	MEAM ^a	EAM ^b	EAM ^c	LR ^d	GGA	FP ^e	GGA ^f	GGA ^g	GGA ^h	
	(this work)	(Fernandez)	(Smirnova)	(Pascuet)	(Dai)	(this work)	(Soderlind)	(Taylor)	(Li)	(Beeler)	
E_c (eV)	-5.302	-5.547	-4.185		-5.55	-6.73					-5.1 ⁱ
a (Å)	2.830	2.721	2.842	2.775	2.850	2.800	2.845	2.797	2.811	2.793	2.854 ^j
b (Å)	5.841	6.381	5.714	6.072	5.912	5.863	5.818	5.867	5.913	5.849	5.870 ^j
c (Å)	4.864	4.858	4.891	4.936	4.897	4.899	4.996	4.893	4.894	4.894	4.955 ^j
y	0.108	0.093	0.0988	0.1044	0.121	0.0975	0.1025	0.098	0.098	0.098	0.1024 ^j
V (Å ³ /atom)	20.11	21.09	19.86	20.79	20.63	20.11	20.67	20.07	20.34	19.99	20.75
c_{11} (GPa)	249	436	134		244	306	300	293	255	299	215 ^k
c_{12} (GPa)	61	76	84		63	60	50	60	53	59	46 ^k
c_{13} (GPa)	43	19	103		59	23	5	30	45	30	22 ^k
c_{22} (GPa)	139	211	253		117	228	220	227	201	231	199 ^k
c_{23} (GPa)	123	84	82		94	144	110	147	136	144	108 ^k
c_{33} (GPa)	214	282	320		128	375	320	331	345	364	267 ^k
c_{44} (GPa)	41	89			51	158	150	149	127	100	124 ^k
c_{55} (GPa)	18	42			14	131	93	117	115	150	73 ^k
c_{66} (GPa)	26	59			19	99	120	95	88	132	74 ^k
B (GPa)	116	143	109		102	149	130	147	141	151	115 ^k , 125 ^l
G (GPa)	35	88			29	115	118	113	104	111	88

Reference: a[20]; b[9]; c[17]; d[18]; e[25]; f[29]; g[30]; h[31]; i[32]; j[33]; k[26]; l[4].

Fig. 2. Comparison of the change of structural parameters as a function of temperature for α -U simulated by the COMB (dashed line, this work) and EAM (dotted line, [9])

potentials with experiment (solid line, [28]) for the (a) a axis, (b) b axis, (c) c axis, (d) y parameter, and (e) atomic volume. All curves are normalized by their values at 0 K; the experimental 0 K values are extrapolated from the high-temperature profile in order to avoid the cryogenic abnormal lattice change in α -U.

Fig. 3. Comparison of α -U thermal expansion coefficients (CTE) of the a, b, c axes and volume between COMB and experiment [28].

the EAM potentials show good agreement with experiment in the a and c axes and volume. The trend of decreasing b axis with increasing temperature is successfully captured by the COMB potential but not by EAM. As we have found in previous work [15], the y parameter and b tend to change in different directions (as one increases, the other decreases) in order to keep neighbor shell order unchanged; here both potentials reproduce this trend. For the EAM potential, b increases in the whole temperature range while y decreases up to 700 K; at 800 K, the abrupt increase of y indicates the $\alpha \rightarrow \gamma$ transformation is about to occur. In α -U symmetry, the γ phase corresponds to $y = 0.25$. Pascuet [17] and Fernandez [20] have shown that their potential also reproduces the signs of lattice expansions (in which they refer as “linear expansion”). To further

evaluate the quality (transferability) of COMB potential, the calculated CTE is compared with experiment and shown in Fig. 3. The CTE curves correspond to the temperature derivative of the lattice curves (Fig. 2); the reproduction of the CTE profiles in a full MD simulation is an even stricter test of the potential quality than the comparison of static values shown in Table 2. Despite some quantitative differences, COMB correctly reproduces the order and sign of all CTEs as $\text{CTE(V)} > \text{CTE(a)} > \text{CTE(c)} > 0 > \text{CTE(b)}$ at high temperatures, where the parenthesis indicates the physical property which the CTE is referring to.

The defect formation energies (DFE) calculated by COMB, EAM and DFT are shown in Table 3. All defect structures have been relaxed; this is the forces on all atoms have been reduced to very low values. “I4” and “I5” denote the two different interstitial sites at which the defect atom is respectively bonded with four and five nearest equal-distance neighbors before atomic relaxation [15]. Vacancy energies from all computational methods are in good agreement with each other, although they are slightly higher than experimental data. Since vacancy energies are usually 1/3 to 1/4 of cohesive energies for metals, the good reproduction of E_c and vacancy DFE in COMB indicates that this potential is well-parameterized. For interstitials, after atomic relaxation the local structure is highly distorted and the configuration difference between the I4 and I5 sites becomes less distinct; thus, it is not surprising that these two sites have similar relaxed defect energies. However, the structural distortions are not identical among COMB, EAM and DFT, and it is expected that their interstitial energies will have some differences. While the COMB values for the interstitial formation energies are lower than experiment, they are not anomalously small for metal interstitials. As a result, we believe that the performance of this potential in the expansion simulation will not be substantially impaired. Finally, the (001) surface energy is calculated and shown to match with experiment, but it is substantially smaller than other MD results and the two independent GGA calculations.

Table 3: Comparison of relaxed defect formation energies (DFE) of α -U among MD (COMB and EAM), DFT results and experiment. “I4” and “I5” denote two different interstitial

sites; “FP” denotes Frenkel pair and uses the lower of I4 and I5 values for calculation. The (001) surface energy is also considered.

	MD				DFT		Experiment
	COMB (this work)	MEAM ^a (Fernandez)	EAM ^b (Smirnova)	LR ^c (Dai)	GGA (this work)	GGA ^d (Taylor)	
I4 (eV)	2.6	3.96	3.6		4.9		
I5 (eV)	2.6		3.4		4.3		
V (eV)	1.9		2.0		1.7	1.95	1.5 ^e
FP (eV)	4.5		5.4		6.0		
(001) (J/m ²)	1.05	2.02		1.53	1.8	1.4	1.0 ± 0.5 ^f

Reference: a[20]; b[9]; c[18]; d[29]; e[34]; f[35].

2.3. Predicted properties of β -U

The β phase is actually not considered in the parameterization process (Table 1). The only requirement on this phase is that it must have an energy of 0.1 eV higher than α . Calculation of some basic static properties of β and the comparison with DFT results can further test the flexibility of the COMB formalism and the quality of this parameter set. The comparison is (Table 4) surprisingly reasonable, providing further evidence supporting the reliability of this COMB potential. If data on the β phase had been included in the training set, the agreement could have been improved further. Nevertheless, COMB is not designed to simulate β -U since the phase will transform to the more stable γ under small thermal agitation.

Table 4: Selected structural properties of tetragonal β -U calculated by COMB, DFT (GGA), as compared with experiment. $E(\beta-\alpha)$: the amount of energy by which β is higher than α .

	COMB (this work)	GGA (this work)	GGA ^a (Li)	Experiment ^b
$E(\beta-\alpha)$ (eV):	0.12	0.099		> 0
a (Å)	10.98	10.46	10.52	10.76
c (Å)	5.53	5.66	5.71	5.65
V (Å ³ /atom)	22.2	20.6	21.1	21.8
c_{11} (GPa)	188	298	218	

c_{12} (GPa)	100	51	99
c_{13} (GPa)	74	63	80
c_{33} (GPa)	215	238	214
c_{44} (GPa)	7.8	46	60
c_{66} (GPa)	27	74	61
B (GPa)	120	132	130

Reference: a[30]; b[11].

2.4. Refitted properties of γ -U

The modeling of a bcc material itself is generally straightforward for the interatomic formalism used in most MD potentials. The challenge in modeling bcc γ -U is two-fold: (1) to ensure that it is not more stable at $T = 0$ K than the α phase and, at the same time, (2) to maintain some level of accuracy in structural properties (the criteria of flexibility and accuracy). This quickly becomes a difficult task since classical interatomic potentials tend to stabilize the most symmetric structures — in this case, the γ phase itself. The properties of γ -U reproduced by different potentials are shown in Table 5, including a new uranium MEAM potential [36] that solely considers the γ phase. Only the lattice parameter, and to a lesser extent the bulk modulus, are accurately reproduced by all potentials. COMB does a good job in satisfying the two above requirements. Although the energy difference, $E(\gamma-\alpha)$, is very small in COMB, we have still managed to transform the γ phase into α slowly by subjecting the γ structure to thermal cycling at temperatures below the phase transformation temperature $T_r(\alpha \rightarrow \gamma)$, showing that this small and positive value of $E(\gamma-\alpha)$ is physically meaningful. This transformation is not observed for the EAM potential [9] due to the fact that its γ -phase is more stable.

We note that the relative magnitude of c_{11} and c_{12} is reversed in COMB compared to DFT, which shows an elastic instability by giving $c_{11} < c_{12}$ at $T = 0$ K. However, γ -U is known to be stable only at $T > 1045$ K [5], above which temperature it is required that $c_{11} > c_{12}$. This indicates that these two elastic constants in γ -U will reverse in magnitude as temperature changes. Such a reversal is very difficult to reproduce in MD and we do not force COMB to

Table 5: Key structural properties of bcc γ -U calculated by MD potentials (COMB, MEAM, EAM and LR) and DFT (GGA and LDA), as compared with experiment. $E(\gamma-\alpha)$: the amount of energy by which γ is higher than α ; DFE: defect formation energy; CTE: coefficient of thermal expansion; T_r : phase transformation temperature. T_m : melting temperature.

	MD					DFT						Experiment
	COMB	MEAM ^a	EAM ^b	MEAM ^c	LR ^d	GGA	LDA ^e	GGA ^f	GGA ^g	GGA ^h	GGA ⁱ	
	(this work)	(Fernandez)	(Smirnova)	(Beeler)	(Dai)	(this work)	(Crocombette)	(Taylor)	(Shang)	(Li)	(Beeler)	
$E(\gamma-\alpha)$ (eV)	0.008	0.15	-0.009		0.018	0.28	0.156	0.24		0.278		> 0
a_0 (Å)	3.417	3.463	3.547	3.503	3.474	3.433	3.37	3.43		3.455	3.427	3.48 ^j
V (Å ³ /atom)	20.0		22.3	21.5	21.0	20.2	19.15	20.18		20.62	20.12	21.1 ^j
c_{11} (GPa)	167			111	74	95		161	84	103	86	
c_{12} (GPa)	104			117	56	156		184	157	142	155	
c_{44} (GPa)	50				12	35		56	38	46	37	
B (GPa)	125		92	115	62	136	170	176	132	129	132	113 ^k
DFE I (eV)	2.1		1.8			5.2						
DFE V (eV)	1.4		6.3	1.6		1.2						> 1.0 ^l
DFE FP (eV)	3.5		8.1			6.4						
$T_r(\alpha \rightarrow \gamma)$ (K)	900 $\pm$ 50	972										935 ^m
$T_m(\gamma \rightarrow L)$ (K)	1400 $\pm$ 50	1505	1800 $\pm$ 50	1410 $\pm$ 10								1405 ⁿ
CTE $\times 10^6$	21		8	19								24.6 ^k
												22 ^l

Reference: a[20]; b[9]; c[36]; d[18]; e[23]; f[29]; g[37]; h[30]; i[31]; j[38]; k[6]; l[39]; m[4]; n[5].

capture this instability condition of $c_{11} < c_{12}$ at 0 K while simultaneously requiring it to show $c_{11} > c_{12}$ at high temperature. By contrast, the MEAM potential [36] does display this instability condition at 0 K and demonstrates a good quality in simulation of γ -U, but the ground state α phase is not considered at all in that work. The EAM potential [9] can stabilize γ -U during energy minimization, but after thermal annealing at 500 K the structure can reach an energy state 0.005 eV/atom lower and the structure becomes “body-centered pseudo-tetragonal” (e.g. $a \approx b > c$), which means this EAM potential also exhibits an instability at 0 K (the un-annealed γ is already 0.004 eV lower than α , so the annealed γ is 0.009 eV lower as shown in Table 5). The LR potential does predict γ to be less stable than α and does not show the instability; however, the elastic constants (especially B) are substantially underestimated. The other MEAM potential [20] provides little information on this phase, so its quality is not assessed here.

Reproducing the $\alpha \rightarrow \gamma$ transformation temperatures T_r and the melting temperatures T_m of γ are strict tests of the potential quality (transferability) since they are difficult to fit in potential development. A polycrystalline structure is created for the evaluation of T_r and T_m . The structure contains four grains of random orientations, while each grain has the shape of an fcc Wigner-Seitz cell and all grains are combined in such a way so that their grain centers form a fcc lattice. Including grain boundaries in the system enables more accurate evaluation of the transformation/melting temperatures. Experimentally, melting is generally nucleated at defect site (thermodynamic melting) [40]; if no defect site is provided in MD simulation, the system will super-heat until the solid is mechanically unstable (mechanical melting) which always occur at a higher temperature than thermodynamic melting. With grain boundaries, COMB predicts both T_r and T_m in very good agreement with experiment ($T_r = 900 \pm 50$ K in COMB vs. 935 K in experiment; $T_m = 1400 \pm 50$ K vs. 1405 K). Test on EAM shows that it overestimates the stability range of the γ phase; the energy of γ is always 0.003 – 0.009 eV below α at all temperatures. Given the fact that bcc γ is more symmetric and thus has a larger entropy, it is clear that the real ground state for this EAM potential is γ .

With many of the properties of different uranium phases well-reproduced in the COMB potential, the simulations of defected uranium structures are expected to be quantitatively correct. Results of these simulations are presented in the next section.

3. Simulation of uranium expansion

Before the simulation results are analyzed, some terms characterizing the extent of expansion or defect state must be specified. The "defect energy ratio" (DER), "volume expansion ratio" (VER) and "axial expansion ratio" (AER) are defined in order to reveal the physics of uranium expansion under the effect of intrinsic defects. In this work, the DER of a structure with an initial concentration of $x\%$ Frenkel pairs (FPs) is defined as:

$$DER = 100 \left[\frac{E(x, T) - E_0(T)}{E_{FP}} \right] \quad (3)$$

where $E(x, T)$ and $E_0(T)$ are respectively the energies per atom of the defected and perfect structures with defect concentration $x\%$ at temperature T , and E_{FP} is the defect energy of one FP at 0 K in COMB (4.5 eV). If the FPs do not interact with each other or their concentration is small, the DER gives the FP concentration in the system ($DER = x$). For α -U structures containing 4% and 7% FP, the initial DERs are respectively 3.8% and 6.0%, indicating that the assumption of non-interacting FP is approximately true at these concentrations. DER can be used as an approximation of the FP concentration during annealing, since direct counting of the number of defects is almost impossible for two reasons: first, a single isolated defect can cause substantial displacement on nearby atoms, forming dumbbells or dumbbell chains, making accurate determination of the actual defect number difficult; second, defect morphologies other than point defects – dislocation loop and stacking fault – can form after annealing, such that it is not appropriate to describe the system by a single FP concentration alone. But in a dense, unary system where the number of atoms is constant, all defects (point defects, defect clusters, dislocations or grain boundaries) formed can be regarded as FP so the use of DER to measure the

“closeness” of the system to its actual ground state still makes sense: the smaller the DER, the fewer defects a system has and the closer the system is to its pristine ground state.

The VER measures the extent of expansion from changes in dimensions and calculates the percentage of volume increase normalized by the volume in the perfect structure. In this work it is defined as:

$$VER(x, T) = 100 \left[\frac{V(x, T)}{V_0(T)} - 1 \right] \quad (4)$$

where $V(x, T)$ and $V_0(T)$ are respectively the volume per atom of the defect and perfect structures with defect concentration $x\%$ at temperature T . Similarly, the AER A_i is defined as:

$$A_i(x, T) = 100 \left[\frac{L_i(x, T)}{L_{i0}(T)} - 1 \right] \quad (5)$$

where $L_i(x, T)$ and $L_{i0}(T)$ are respectively the axis lengths in the i direction of the defect and perfect structures at temperature T and concentration $x\%$, and i can be either a , b or c . If A_i is not too large, $VER \approx A_a + A_b + A_c$.

3.1. Anisotropic expansion in α -U single crystal

In order to better model the effects of irradiation, we performed a multi-stage simulation with a structure containing $36 \times 18 \times 21$ unit cells ($102 \times 105 \times 102 \text{ \AA}^3$) with 54432 atoms. A structure of this size may not be considered “large” in MD simulations using conventional potentials such as EAM, but given the fact that COMB is at least 100 times slower than EAM [41], this size is a good balance between computational cost and microstructure sampling. The whole simulation is divided into several stages; in each stage a certain number of defects are manually introduced into the system in order to mimic the structural damage by radiation. Traditionally, MD simulation of radiation damage can be divided into two phases: the ballistic phase, in which an atom of very high energy causes severe regional structural disturbance, and

the kinetic phase, in which the structure equilibrates by annealing defects created in the ballistic phase. In this work, we follow the procedure implemented by previous simulations in which the ballistic phase is by-passed by manual creation of randomly distributed FPs [42, 43]. Although in real cascade simulation, the ballistic phase creates an inhomogeneous distribution of vacancies and interstitials (vacancies concentrated at the center and interstitials at the periphery of the radiation damage region) that will very likely affect the formation of defect clusters, our approach is justified since the primary goal of this multi-stage simulation is to test the performance and robustness of this new COMB potential under a dynamic and defected environment, given that the uranium static properties in this potential is already superior (Table 2 and Table 5). The ballistic phase could be realized in future use of COMB with the addition of the ZBL [44] potential to account for small-distance interactions.

At each stage of the current simulation setup, a 1% concentration of FPs is randomly created. The system is equilibrated at 0.01 K for 50 ps with its volume free to expand (which is essentially a 0 K relaxation process), followed by an annealing cycle in which the structure is heated from 0 K to just below melting and then cooled back to 0 K, both in increments/decrements of 100 K every 50 ps. During the annealing cycle, FPs annihilate or cluster with each other to form isolated or extensive defects, reducing the volume expansion that has occurred during the 0 K equilibration. In the next stage, another 1% concentration of FPs are created in the final structure from the previous stage, and the new structure is subjected to another energy equilibration and annealing cycle. This process is repeated eight times for α and eleven times for γ . Since at each stage the FP generation creates a displacement per atom (dpa) of 0.01, the total cumulative dpa is at least 0.08.

The final DER, VER and AERs are calculated after each stage and are reported in Fig. 4. After the first stage, all AERs have roughly the same value indicating isotropic expansion. Starting from stage 2, A_b quickly increase to become commensurate with VER, while A_a and A_c oscillate around zero. As the discussion below (section 3.2.1) shows, this is indicative of almost

all of the newly-added defects clustering to form prismatic dislocation loops that are normal to the b axis. i.e., in the (010) planes. The DER and VER have similar values, which is expected according to their definitions. They increase almost linearly with dpa, but with a much smaller slope which does not extrapolate to zero at zero dpa. This is most likely due to the fact that extended prismatic loops require a much smaller volume expansion for each additional atom than the average atomic volume required to initially create the loop.

Fig. 4. Evolution of DER, VER and individual AERs in α -U calculated by COMB potential after each simulation stage. The legend “AER b (initial)” shows the value of A_b before each simulation stage.

The AER of the b axis (A_b) after energy equilibration and before annealing cycle is presented in Fig. 4 as “AER b (initial)” for each stage. This value is generally negative and always smaller than the final A_b of the same stage. Physically this means that, as random defects are introduced in uranium, its b axis contracts; as defects annihilate and interact to form prismatic loops, the b axis re-expands. This process happens at each injection of new FPs. By contrast, the *initial* A_a , A_c and VER (not shown in Fig. 4) are always positive and much larger than their *final* counterparts, indicating conventional expansion. We conjecture that the initial contraction of b axis is due to the local structural relaxation and the re-adjustment of the highly

directional bonding between interstitial and lattice atoms. α -U is known for showing grain boundaries (GB) tearing due to anisotropic dimension change during thermal expansion [28] and radiation damage [45], but Fig. 4 shows that the defect annihilation process itself can also cause anisotropic dimension change in which the b axis first contracts and then expands, whereas the a, c axes and volume first expand and then contract. The final tearing phenomenon could be the result of a combination of these three effects (thermal expansion, irradiation damage and intrinsic contraction).

3.2. Defect structures in α -U

It is well-known that dislocation loops can exist in two different forms: the prismatic loop and the glide/slip loop [46]. The prismatic loop can be formed by condensing extra interstitials or vacancies into a loop, with pure edge component everywhere on the loop edge and a Burgers vector normal to the loop plane. The glide loop can be formed by applying a shear stress on the slip plane of the structure. The resulting loop has both edge and screw components depending on its orientation relative to the shear direction; its Burgers vector lies on the loop plane. Both types of loops are observed during defect annihilation of α -U and the corresponding analysis of both structures will be discussed in detail below.

3.2.1. Prismatic dislocation loop

The prismatic loop is the most commonly observed defect type at dpas of 0.02 and higher. Fig. 5(a) and (b) show a typical structure from an annealed system with a dpa of 0.07. The defect structure is viewed along the principle axes of b and c. Fig. 5(c) and (d) show the idealized models in the same directions without atomic relaxation. Because the perfect α -U structure can be visualized as the stacking of corrugated planes along the b direction, this prismatic defect structure is manifested as additional loop planes inserted into the normal structure stacking (Fig. 5d). These loops are inserted in such a way that, when viewed in the b direction, all loop atoms

are directly above or below their neighboring sheets and almost no interstitial can be detected (Fig. 5a and c). This causes almost no atomic displacement in the a and c directions, corresponding to the very low values of A_a and A_c observed in the multi-stage simulation (Fig. 4). When viewed in directions other than b (Fig. 5b), a buckling region with atomic displacement can be clearly seen, giving a large A_b that accounts for almost all expansion. The dominance of A_b over A_a and A_c in Fig. 4 confirms that prismatic loop formation is the dominant defect formation mechanism at dpa of 0.02 and higher.

Fig. 5. Snapshots of part of the final structure at stage 7 (cumulative dpa = 0.07) showing the formation of a prismatic dislocation loop perpendicular to the b axis. Different views are shown along (a) the b axis and (b) the c axis. All atoms are shown in order to demonstrate the location of defect atoms and their bonding relation with the perfect atoms. Idealized models are shown in (c) and (d) for the same viewing directions as (a) and (b) but with no local structural relaxation. This prismatic dislocation is essentially an extra layer of atoms (brown) inserted into the perfect α -U structure (purple) between two adjacent b planes. Atoms are displaced exclusively in the b direction but not in the a or c directions, explaining the different expansion behavior of these axis shown in Fig. 4.

3.2.2. *Glide extended dislocation loop*

Since each kind of glide dislocation loop associates with one slip system, it is reasonable to discuss the slip systems in α -U before analyzing the glide loop structure. In α -U, the (010)[100] system has been proposed to be the primary deformation slip system based on experimental observation [47]. This seems to be physically reasonable since the slip plane and slip direction correspond to the close-packed plane and direction – a general rule for other metals of fcc, bcc and hcp symmetry. Some authors [48] even conjectured that the bonding in α -U was partially covalent and each corrugated sheet in the (010) plane was a “giant molecule”. However, DFT calculations [25, 49] did not show any obvious gap in the electronic density of states in α -U; nor did measurements of diffusion constants [50] along the three principle directions show large anisotropy. These results indicate that bonding in α -U is of typical metallic type. However, because the nearest four neighbors lies within the corrugated layer, the intra-layer bonding is stronger than inter-layer bonding and the (010) corrugated plane is expected to be the slip plane.

The glide loop occurs much more rarely than the prismatic loop in our simulation. Fig. 6 shows a typical glide loop structure from a system containing 3% initial FPs annealed at 600 K for 850 ps. Fig. 6(a) and (b) show the loop structure with all atoms visible. The head/tail and body of the loop, usually respectively referred as partial dislocation and stacking fault (SF), are identified by red arrows. Idealized models in the same viewing directions are shown in Fig. 6(c) and (d) for better illustration. The partials and the SF body are outlined by grey dotted boxes. The normal stacking order of atomic layers in α -U can be denoted as ABAB..., in which layer A can transform into B by displacing itself by half of the $\langle 100 \rangle$ unit cell length. Under defect interactions, part of the structure may have its stacking order change to ABAABA... where two planes of the same position are packed next to each other. In this case, atoms in these two planes are closer to each other and form interlayer bonds, showing a bonding pattern that is different from that in the perfect structure when viewed along the b direction. The regions that show changes to the bonding pattern are identified as partial dislocations; the region between the two partials that shows no change to the bonding pattern is identified as a stacking fault (Fig. 6b and d).

Fig. 6. Illustration of an extended dislocation loop with Burgers vector $b = [100]$. Views along the (a) c axis and (b) b axis with all atoms shown. The partial dislocations and stacking fault are identified; idealized models are shown in (c) and (d) for the same viewing directions. Green letters A and B indicate the stacking sequence; leading partial, stacking fault and trailing partial are indicated by grey dash boxes; interlayer bonding in the partial and stacking fault regions are indicated by red arrows.

3.3. Comparison to experiment

It has long been known that the anisotropic dimensional change of single crystal α -U under irradiation has a contraction in the a axis, an expansion in the b axis, and almost no change in the c axis [5]. Buckley [51] observed that the contraction/expansion is even more pronounced at 77 K, and suggested that aggregation of vacancies and interstitials as plates on different crystal planes might be the reason for this anisotropic shape change. By measuring the anisotropic change of sample length and electrical resistivity under neutron irradiation, Loomis and Gerber

[45] obtained supporting results that interstitials in α -U are preferentially condensed in (010) planes to cause expansion, and vacancies condensed in (110) to cause contraction. In the results discussed above, expansion by interstitials is verified but no contraction is observed. By assuming these two processes are independent, we now try to make a quantitative justification of the expansion exclusively by comparing the simulation results with the experimental measurement [45]. We first estimate the number of FPs in our simulation using a model developed by Norgett, Robinson and Torrens (NRT model) [52], and then back-calculate the energy required in actual experiment to cause the same number of defects observed in our simulation. This energy is converted to dose and expansion ratio according to Loomis' result. The expansion ratio calculated is compared to A_b (AER_b in Fig. 4) we obtained in our simulation.

The NRT model predicts the number of FPs produced for a given Primary Knock-on Atom (PKA) energy:

$$N_{NRT} = 0.8E / 2E_d \quad (6)$$

where N_{NRT} is the total FPs number predicted by the NRT model, E is the energy available in PKA for elastic collision, E_d is the threshold energy of displacement for uranium atoms and can be estimated by the Sudden Approximation method [53] as used in Ref [54]. COMB gives $E_d = 59$ eV in $\langle 010 \rangle$, 47 eV in $\langle 001 \rangle$ and 49 eV in pseudo $\langle 021 \rangle$ direction (its exact direction depends on the y parameter). For simplicity we choose $E_d = 50$ eV. We realize that this E_d is a little large, since a typical value of 25 eV is proposed for metals [55]; but this two-fold difference will not qualitatively affect our analysis and conclusion. The NRT model predicts N_{NRT} based on energetics considerations alone and does not take account of the subsequent recombination. To estimate the energy in the system, we need to use the initial FP concentration in our calculation. Using the 5% cumulative concentration as an example, Eqn. (6) gives $E = 5\% \times 54432 \times 2 E_d / 0.8 = 0.34$ MeV. Assuming each uranium fission produce 200 MeV of energy and only 10% of this energy is applied to ballistic interaction, the fraction (τ) of U atoms fissioned in our MD system is $\tau = 0.34/20/54432 = 3.1 \times 10^{-7}$. The growth coefficient G can be defined as [45]:

$$G_i = \frac{\ln L_i / L_{i0}}{\tau} \approx \frac{\Delta L_i / L_{i0}}{\tau} = \frac{A_i}{\tau} \quad (7)$$

where $\Delta L_i = L_i - L_{i0}$ is the change of axis length. The growth coefficient for the b axis at 50 K is $G_b = 21500$ [45]; this gives $A_b = 6.7 \times 10^{-3}$ according to Eqn. (7). The MD simulation gives $A_b = 1.81 \times 10^{-3}$ (Fig. 4). Results from other concentrations are also calculated and presented in Fig. 7.

Care must be taken when comparing cryogenic physical properties of uranium. Loomis and Gerber had also reported several G_b values that are larger than 21500 at lower temperature; however, we do not use these values to compare with our MD results at 0 K. At $T < 43$ K uranium supports a charge-density wave (CDW) in which distortions appear in various directions and divides the materials into small domains [5]. The elastic constants, specific heat and electrical resistance show abrupt changes within this temperature range [56]. In order to avoid this cryogenic effect which is not present in the MD simulations, we choose a value measured at the lowest possible temperature (50 K) without introducing the CDW effect.

Fig. 7. Calculation of the axial expansion ratio (A_a and A_b) by MD using COMB and EAM potentials, as comparing with the NRT models using two different displacement energy.

It is important to recall that the NRT models are empirical equations that attempt to correlate the PKA energy with the observed number of defects during cascade simulations. So, when this model is used to interpret our simulation results and the experimental measurement [45], it would not be surprising to have substantial discrepancy in the prediction of the extent of expansion. However, given the simplicity of these models in interpreting cascade simulations, the great physical difference between our multi-stage simulation and a realistic irradiation experiment, and various assumptions made in interpreting our simulation results, we think an agreement within an order of magnitude between the MD prediction and the NRT model is reasonable. If $E_d = 25$ eV is used, as recommended by Kinchin [55], instead of 50 eV given by COMB, the resulted NRT curve is even closer to the MD calculation.

In experiment [45], both lattice contraction (in the a axis) and expansion (in the b axis) were observed but only expansion is accounted for in our simulations despite the fact that both vacancies and interstitials are created during setup. In separate simulations, structures having only vacancies (1% or 2%) were created and annealed; the a and b axis did contract but their AERs were ten times smaller compared to the equivalent dpa in Fig. 4. Thus, we conclude that the experimentally observed contraction of the a axis might not be exclusively caused by the condensation of vacancies. Some other effects such as interaction with dislocations, grain boundaries, or extrinsic noble gas bubbles may play a role in lattice contraction. This awaits further investigation.

To examine the effect of the choice of potential, the same multi-stage simulation has been conducted using the EAM [9] potential. The A_a and A_b are shown in Fig. 7. The VER (not shown) from EAM is ten times as large as that in COMB; half of this large expansion originates from the a axis. Despite different values of AERs showing anisotropic expansion, a closer look of the defect structure does not reveal any particular structural morphology, e.g. no formation of dislocations. Because α is slightly less stable than γ in this potential at all temperatures, the annealing cycle only heats the structure to 500 K to avoid the phase transformation occurring fast

enough within the time scale of our simulation. But a thin layer of γ region perpendicular to the a axis still forms and grows gradually during each stage, giving a large A_a (in this EAM potential, the atomic volume of γ is 12% larger than α , see Table 2 and 4). So even though the calculated A_b in EAM is in better agreement with the NRT model using the universal E_d value of 25 eV, the much larger A_a and the instability of the α phase disqualify this potential from use in swelling simulations.

Based on the analysis above, we conclude that the ability to prevent or retard the formation of extensive defects in α -U is a key factor for homogeneous expansion in this phase. The influence on defect formation and atomic diffusion in α -U structure at microscopic scales by the addition of Pu and Zr deserves more attention in order to gain deeper understanding on the performance of this promising metallic fuel in fast reactors.

3.4. Uranium expansion in γ -U structure

In this section, a corresponding simulation is performed for the expansion of the bcc γ -U phase. The structure is constructed with $30 \times 30 \times 30$ unit cells ($102.5^3 \text{ \AA}^3$) and 54000 atoms. The final DER, VER and AERs for eleven stages are plotted in Fig. 8. Compared to the counterpart α -U in Fig. 4, γ -U shows homogeneous expansion due to cubic symmetry and a much smaller VER ($< 0.05\%$). The residual defects are mostly isolated vacancies and $\langle 111 \rangle$ interstitial dumbbells. No extensive clusters (other than $\langle 111 \rangle$ dumbbell chain) or dislocations form. This gives a VER that is about ten times smaller than α -U under the same dpa condition, demonstrating the extraordinary ability of cubic material to annihilate defects and sustain continuous intrinsic defect creation. Moreover, the bcc phase has a considerable ability to self-heal from the amorphous state. For a one-time defect creation of 15% FPs (not shown here), the γ -U structure first became amorphous, but as the structure is annealed at 900 K, the bcc crystal structure nucleates from the amorphous phase with very few residual defects (DER $\sim 0.04\%$). In comparison, for α -U with 10% FPs, the structure generates too many stacking faults

that the original crystal lattice is no longer discernible, and it shows no sign of recovery during thermal annealing.

Fig. 8. Evolution of DER, VER and individual AERs in γ -U calculated by COMB potential after each simulation stage. The volume expansion is about ten times smaller than α -U (Fig. 4) under the same dpa dose.

The much smaller expansion of γ -U renders this phase preferable in fast reactors; indeed it can be stabilized by alloying with other bcc elements. At high temperature this phase softens and swells mainly by agglomeration of noble gas bubbles [3]. Lattice expansion by intrinsic (or soluble extrinsic) defects is no longer the major concern in γ -U as illustrated in this work.

4. Conclusions

In this work, the uranium COMB potential published previously [15] has been re-parameterized. Most properties of both the orthorhombic α and bcc γ phases, including the cohesive energies, elastic constants, bulk moduli, coefficients of thermal expansion and defect formation energies, are substantially improved. The potential demonstrate excellent flexibility (polymorphs of α and γ), accuracy (Table 2 and Table 5) and transferability (CTEs of α and γ , T_r and T_m). Although it is computationally more expensive than EAM, with the help of Moore's

law (computer speed doubles in every two years), this refitted COMB is nevertheless a strong competitor in uranium potential repository, especially for simulations in a defect environment.

Multi-stage simulations on α -U structure with random FP defects show anisotropic expansion almost exclusively occurs in the b direction. This is caused by the condensation of interstitials to form prismatic loops on the (010) plane. The structure of this prismatic loop is analyzed, demonstrating the mechanism of causing anisotropic expansion. A glide dislocation loop structure is also found to exist on the (010) plane. The edge component of this loop lies in the $\langle 100 \rangle$ direction and can split into partials in a similar manner as in close-packed materials. By using the NRT model to estimate the number of Frenkel pairs, the b-axis expansion is shown to lie within one order of magnitude of experiment. This result is satisfactory given the simplicity and the empirical nature of the model. Lattice contraction by vacancies has not been observed in this work; this phenomenon is believed to relate to other types of defects (such as dislocations) or elements (such as noble gas), which are missing in the setup of current work. Brief simulations of γ -U shows that its isotropic expansion has a magnitude that is at least 10 times smaller than that of the α phase, with defects being mostly isolated point defects or $\langle 111 \rangle$ dumbbell chains.

This work provides the first simulation results of how intrinsic defects in α -U agglomerate and cause anisotropic expansion, a first step towards the understanding of the expansion/swelling mechanism of α -U from a microscopic viewpoint. As the U-Pu-Zr alloy becomes a promising fuel in fast reactor, researches on the atomic migration, diffusion and interaction in this material will be very helpful in capturing the behavior of this fuel under conditions of high temperature, stress and radiation.

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