

# Machine-learned potential for fission gas diffusion in uranium oxide nuclear fuels

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We report a machine-learned interatomic potential for uranium dioxide with xenon gas, as well as a machine-learned potential for uranium dioxide. Training datasets were constructed by leveraging a combination of density functional theory calculations with a Hubbard  $U$  correction and molecular dynamics simulations. Query-by-committee active learning procedures further automated the augmentation of training datasets. The efficacy of employing an equivariant message-passing neural network for iterative potential fitting was demonstrated by reproducing DFT +  $U$ -level forces and energies, despite the training datasets being much smaller than those for recently reported uranium dioxide MLPs. We found that our machine-learned potential for  $\text{UO}_2$  achieves strong agreement with experimentally observed thermophysical and thermomechanical properties across a temperature range 300–3000 K. Our second machine-learned potential for uranium dioxide with xenon successfully replicates reference DFT +  $U$  incorporation energies of xenon into various lattice sites. We also employed this MLP to calculate migration barriers for xenon diffusion via tetravacancy defect cluster mechanisms. The superlative ability of these machine-learned potentials to capture the behavior of uranium dioxide with xenon inclusion across a range of temperatures and defect chemistries lays the foundation for larger-scale molecular dynamics simulations of fission gas transport through uranium oxide fuel matrices.

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## I. INTRODUCTION

Nuclear power is rapidly gaining momentum as a crucial component of future energy strategies. Commitments to drastically increase nuclear capacity have been announced worldwide, with over 20 countries signing a declaration to triple nuclear energy output by 2050 at COP28 and an additional six countries affirming this declaration at COP29 [1].

Efficient and accurate fuel performance modeling will be key to achieving this multiplication in nuclear capacity. Simulations modeling the in-reactor evolution of materials properties are central to safety analyses and for determining the lifetimes of fuel elements [2]. For this reason, high-fidelity fuel performance codes are crucial to everything from licensing to operation for any reactor technology.

Fuel performance codes are primarily dependent on empirical models fitted to data that are readily available for traditional reactor materials and conditions. For example, widely used empirical models for the thermophysical properties of  $\text{UO}_2$  fuel have been employed for decades [3]. Beyond

purely empirical approaches, a suite of semiempirical potentials (SEPs) have been developed for  $\text{UO}_2$  by fitting pair potentials with experimental data [4–7]. One of the latest and most robust semiempirical potentials was developed by Cooper *et al.*, who leveraged the embedded atom method [8] to fit a many-body potential to experimental lattice parameters, elastic constants, and thermal expansion data [9]. This potential is widely used to describe interatomic forces and energies in molecular dynamics simulations for actinide oxides, and it has even been used to derive a heat capacity law for inclusion in the fuel performance code GERMAL V2 [10].

However, next-generation reactor technologies intend to reach higher burnups, extend into more extreme temperature regimes, and employ novel fuel forms and geometries [11]. Both by design and as a result of longer in-reactor usage, the microstructure of nuclear fuels will evolve differently than traditional  $\text{UO}_2$ . Commonly employed empirical models become inaccurate in light of these differences, and the same wealth of data that enables robust model development for traditional reactor materials is unavailable for advanced technologies [12].

For this reason, new approaches must be developed to ensure the continued accuracy of fuel performance models. A key improvement would be to derive fuel behavior and properties from first principles calculations at the atomic scale. Shifting from empirical models to such atomistic methods would provide physics-based mechanistic insight into complex phenomena that limit fuel element lifetimes.

One process of great interest is the diffusion of fission gas through the fuel matrix. Approximately 13% by weight

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of the products generated by the fission of a uranium-235 nucleus are Xe and Kr gas [13–16]. The retention of these fission gases in  $\text{UO}_2$  is kinetically unfavorable, leading to a multistage diffusion process involving migration of fission gas atoms through the bulk lattice, intragranular bubble formation within fuel grains, coarsening of intergranular bubbles along grain faces, and transport of bubbles along grain boundaries [14]. The interplay of these processes can ultimately result in the release of fission gases into the fuel-to-cladding gap. Such fission gas release is detrimental to the overall thermal efficiency of the reactor, pressurizes fuel pins, and expedites other fatal phenomena like pellet cracking and the onset of pellet-cladding interaction [17]. Despite the fact that fission gas diffusion is one of the primary sources of fuel element failure, its complex multiscale nature renders it challenging to simulate [18]. Even with much improvement in fission gas diffusion models [19], compounded errors resulting from assumptions made at every length scale leave unanswered questions about the behavior of Xe and Kr in fuel materials. Highly accurate models rooted in atomic-scale insights would greatly improve predictions made by fuel performance codes for fission gas diffusion.

Currently, density functional theory (DFT) is widely employed to calculate properties of nuclear fuels at the atomic scale. Early work studying point defect energetics in  $\text{UO}_2$  with local and semilocal DFT [20–22] has been refined through the implementation of a Hubbard parameter to address strong  $f$ -electron correlations [23,24]. Further advances continue to improve DFT +  $U$  approaches for strongly correlated materials like  $\text{UO}_2$ , such as the development of the  $U$ -ramping [25] and occupation matrix control (OMC) [26] methods to prevent convergence to metastable states. These developments have enabled complex studies of nuclear fuel material properties based on their electronic structure, extending beyond the energetics of point defects to provide insights into larger-scale phenomena like irradiation-induced creep [27].

Even so, atomistic modeling for nuclear fuels remains challenging because of significant computational limitations in reaching the required length and time scales and lingering questions in the theoretical treatment of  $f$  electrons. Though DFT +  $U$  calculations are considered highly accurate, their computational cost imposes a system size limit that is particularly restrictive for modeling defects. While DFT calculations are tractable for systems containing thousands of atoms, microstructural defects in nuclear fuels such as dislocations or cracks are on the order of millions or billions of atoms [28]. Additionally, the periodic boundary conditions typically used in DFT calculations for solids result in an artificial inflation of defect concentrations. It is therefore clear that an alternative tool is necessary to incorporate atomistic modeling into robust fuel performance codes without accruing unreasonable computational cost.

Machine-learned potentials (MLPs) have emerged as promising tools to extend DFT-level accuracy across longer length and time scales. MLPs are mathematical representations of the total energy and interatomic forces for a given atomic configuration generated by fitting a machine-learned model to a reference dataset [29]. When trained on an appropriate collection of quality reference data points, MLPs reproduce high-fidelity energies and forces for chemical sys-

tems at significantly reduced computational costs. MLPs have been employed for a wide range of materials classes and chemical systems up to the device scale, from amorphous solids to metal-organic frameworks [30–34].

Several MLPs have also been trained for  $f$ -block materials, including thorium dioxide [35], pure uranium [36–39], uranium mononitride [40], uranium carbide [41], and uranium dioxide [42–46]. Because  $\text{UO}_2$  is the most common nuclear fuel type and a wealth of experimental correlations and semiempirical potentials are available to benchmark novel machine-learned models, it will be the primary focus of this work. Differences in performance across reported  $\text{UO}_2$  MLPs can be attributed to the reference datasets used for training and the machine-learning architecture used for fitting and evaluation. Stippell *et al.* [43] employ a transfer learning strategy to train two ANAKIN-ME (ANI) [30] potentials on DFT and DFT +  $U$  training data, respectively. Gumber *et al.* [46] train a HIP-NN MLP with the DFT +  $U$  dataset generated by Stippell *et al.* and further refine the MLP using experimental data. Dubois *et al.* [44] compare the performance of an MLP trained using the Behler-Parinello high-dimensional neural network [47] (HDNNP) with a second MLP generated using spectral neighbor analysis approaches (SNAP). Both the HDNNP and SNAP potentials were trained using DFT +  $U$  data. Konashi *et al.* [45] use DeePMD-kit [48] to train an MLP using approximately 20 000 snapshots of  $\text{UO}_2$  from *ab initio* molecular dynamics (AIMD) simulations. Most recently, Zhong *et al.* [49] highlight their use of a neuroevolution potential to calculate the thermal conductivity of  $\text{UO}_2$ . While Alzate-Vargas *et al.* employ a hybrid MLP and Buckingham potential approach to simulate the incorporation of xenon into the uranium mononitride fuel lattice [40], we are unaware of any reported  $\text{UO}_2$ -based MLPs including fission gas atoms in their reference training structures.

In this work, we employ an active learning methodology to construct and benchmark two MLPs using the MACE architecture [50,51]. After testing the performance of an MLP trained exclusively for  $\text{UO}_2$  fuel against a reference DFT +  $U$  dataset, a second MLP is trained for  $\text{UO}_2$  with xenon inclusion. In doing so, we demonstrate the promise of MLP-driven molecular dynamics (MLP-MD) simulations for modeling fission gas diffusion in nuclear fuel materials. The MACE MLP trained for  $\text{UO}_2$  reproduces thermophysical and thermomechanical properties including the lattice expansion and heat capacity remarkably well across a wide range of temperatures. The MACE MLP for  $\text{UO}_2$  with xenon inclusion reproduces the energetics of both (i) xenon incorporation into a variety of trap sites and (ii) the migration of xenon through the  $\text{UO}_2$  fuel matrix via several diffusion pathways. This demonstrates the promise of using MLP methods to extend the accuracy of DFT +  $U$ -level results to the multiscale challenge of fission gas diffusion in nuclear fuels. Beyond making significant progress in calculating crucial nuclear fuel performance metrics, this work serves as a demonstration of repurposing available reference data to train new MLPs with extended capabilities.

First, we discuss the construction of DFT +  $U$  reference datasets for both MACE MLPs through active learning procedures. We also highlight the MACE architecture and its strengths as an equivariant message-passing neural network.

Following the training and testing of our MACE MLPs, we compare their performance to existing experimental data and potential models for the thermophysical and thermomechanical properties of  $\text{UO}_2$  as well as the energetics of xenon migration in the  $\text{UO}_2$  lattice. The MLPs presented in this work display robust performance across a wide temperature range, demonstrating strong agreement with existing experimental data and improving upon advances made by existing  $\text{UO}_2$  MLPs. The drastic computational speedup achieved by employing these models in place of expensive first-principles methods points to future work employing the MACE MLPs to investigate extended defect and fission gas mobility in uranium oxide nuclear fuels.

## II. METHODOLOGY

### A. Construction of the DFT + $U$ reference dataset

Fidelity of any MLP is entirely dependent on the quality of its reference training data. In this work, each training data point is defined as a pairing between (i) a unique set of atomic nuclei and their positions in space (the *configuration*), and (ii) its corresponding system energy and the forces acting on each individual atom in the  $x$ ,  $y$ , and  $z$  directions (the *label*). Therefore, construction of the reference dataset involves first generating a representative set of structures spanning the configurational space of interest and then labeling those structures at an appropriate level of theory.

#### 1. Sampling the configuration space for uranium dioxide with molecular dynamics

Exploration of the configurational space for  $\text{UO}_2$  to generate appropriate MLP training data is accomplished in two phases. First, we extract atomic configurations from MD simulations run using the force field potential developed by Cooper *et al.* for actinide oxides [9] (denoted as CRG) in LAMMPS [52]. Second, we employ an active learning procedure in the Atomic Simulation Environment (ASE) to select meaningful further additions to the training dataset.

For configuration generation using the CRG potential, simulation temperatures range from 300 K to 3000 K. These temperatures are chosen to be reflective of both typical in-reactor conditions and potential accident scenarios. While fuel centerline temperatures are not expected to exceed 2200 K under standard reactor operation, the extension of the training dataset to temperatures nearing the melting point of uranium dioxide (3138 K) accounts for scenarios such as a core melt accident [53,54].

At these temperatures, we perform MD equilibrations for defect-free 12-atom fluorite  $\text{UO}_2$  unit cells, 24-atom supercells including a uranium Frenkel defect, 24-atom supercells including an oxygen Frenkel defect, and 24-atom supercells including a bound Schottky defect. For Schottky defects, oxygen vacancies occupy first-nearest-neighbor sites with respect to one another. Because atoms constituting persistent Frenkel defects are unlikely to occupy first-nearest-neighbor sites to their respective vacancy sites because of spontaneous recombination, second-nearest-neighbor sites are used. For 24-atom supercells, the 12-atom fluorite unit cell is doubled with respect to the  $a$ ,  $b$ , and  $c$  lattice vectors ( $2 \times 1 \times 1$ ,  $1 \times$

$2 \times 1$ , and  $1 \times 1 \times 2$  supercells, respectively). These doubled systems are intended to give a more complete picture of wavefunction interactions and bulk properties, particularly with respect to reducing the interaction of defects with one another upon implementation of periodic boundary conditions. Illustrative configurations are shown in Fig. 1.

Each type of supercell is equilibrated in the isobaric-isothermal (NPT) ensemble. A Nosé–Hoover thermostat and an anisotropic Nosé–Hoover barostat are used to maintain the temperature and pressure of the system, with damping parameters of 0.1 ps and 0.5 ps, respectively. Initial velocities are assigned to all atoms such that they obey a Maxwell–Boltzmann distribution centered around the designated temperature. A minimization step is first performed to allow the simulation box dimensions to relax. Following this minimization step, the supercell is equilibrated for 50 ps with a timestep of 2 fs. A total of 250 configurations are extracted from the resultant trajectory at intervals of 0.2 ps. Simulations are run for all configuration types at zero pressure.

In addition to these zero-pressure runs, simulations for the defect-free cells are also performed in the canonical (NVT) ensemble with expanded box volumes ranging from +0.08% to +13.51% to allow for prediction of thermal expansion by the MLP. These scaling factors align with the expected thermal expansion for  $\text{UO}_2$  in the temperature range under consideration [3]. Additionally, expanding the simulation box enforces exploration of more diverse regions of configurational space. A Nosé–Hoover thermostat with a damping parameter of 0.1 ps is used to maintain the temperature. The supercell is equilibrated for a total of 50 ps with a timestep of 2 fs. Configurations are extracted at intervals of 0.2 ps.

#### 2. Extending the uranium dioxide training dataset with active learning

While these MD equilibrations were carefully performed with the intent of spanning the regions of configurational space that  $\text{UO}_2$  may occupy under reactor conditions, further sampling is required to expand the training dataset and ensure reliable predictions by the MLP. In the event that the MLP must make predictions for a system that falls outside of its training data domain, errors are high and MLP-MD simulations quickly become unstable. However, manual determination of blind spots in MLP performance is tedious and often results in the selection of redundant configurations.

Active learning procedures are commonly employed to combat this issue [55–60]. Active learning circumvents exhaustive manual sampling techniques by instead employing the most current ML model to inform choices about which data points would be the most valuable to include in future model training iterations. By maximizing the value of each additional training data point, active learning reduces the number of expensive labeling computations that must ultimately be performed before a robust model is achieved. One such active learning approach is the query-by-committee method [61,62], in which a committee of MLPs is first trained using a preliminary set of manually selected data points and then MLP-MD simulations are run using this rudimentary set of models to explore areas of the potential energy surface for which prediction errors are high between members of

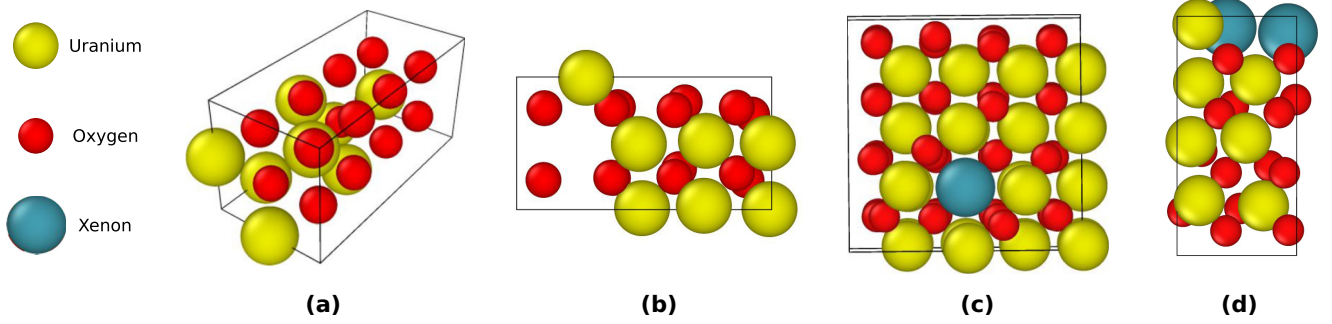


FIG. 1. Sample configurations included in the training datasets for the MACE MLPs. Configurations shown include (a) a  $1 \times 2 \times 1$   $\text{UO}_2$  supercell, (b) a  $2 \times 1 \times 1$   $\text{UO}_2$  supercell with a Schottky defect extracted from an MD equilibration run at 900 K, (c) a configuration with one xenon atom incorporated to a Schottky defect site, and (d) a configuration with two xenon atoms extracted from a higher-temperature MD equilibration run at 1500 K.

the committee. Configurations resulting in a high degree of “disagreement” between committee members are queued for labeling and addition to the training dataset. In this way, active learning procedures automate the selection of new training data points that significantly improve the versatility of the model.

In this work, a preliminary committee of eight models is trained using configurations selected via the MD equilibrations described above. This committee of models is then used as the engine for MLP-MD equilibrations again performed for temperatures ranging from 300 K to 3000 K. A series of volume scaling factors up to +4.31% are also initially applied to simulation boxes to extend the exploration of configurational space. For each possible combination of temperature and initial box volume, a configuration is first selected at random from the existing training dataset. The selected configuration is then evolved under the isothermal-isobaric (NPT) ensemble for up to 10 ps. A Nosé–Hoover thermostat and anisotropic Nosé–Hoover barostat are used to maintain temperature and pressure, with respective damping parameters of 0.1 ps and 0.5 ps. Every 10 fs, the prediction uncertainty is quantified by calculating the standard deviation across the eight committee members for both the energy and forces. If either of these uncertainty metrics exceeds the given threshold for a configuration in the equilibration trajectory, the equilibration is halted and that configuration is saved as a potential candidate for labeling and addition to the training dataset. Uncertainty thresholds are generally set to the mean absolute error (MAE) values found for energy and forces by evaluating the most recent generation of MLPs on a test set not included in potential training.

Following the completion of each active learning sweep, an uncertainty criterion  $u(X)$  is employed to select the candidate configurations having the highest prediction errors among committee models. This uncertainty criterion is defined by Dubois *et al.* [44] as

$$u(X) = \frac{\sigma_E(X)}{\sigma(\sigma_E)} + \frac{\langle \sigma_F(X) \rangle_{i,\zeta}}{\sigma(\langle \sigma_F(X) \rangle_{i,\zeta})}, \quad (1)$$

where  $X$  is the configuration from the pool of candidates,  $\sigma_E(X)$  is the standard deviation in energy predictions made by the committee members for configuration  $X$ ,  $\sigma(\sigma_E)$  is the standard deviation across all  $\sigma_E(X)$  values,  $\langle \sigma_F(X) \rangle_{i,\zeta}$  is the

average of the standard deviations in force predictions over all atoms  $i$  and directions  $\zeta$  for configuration  $X$ , and  $\sigma(\langle \sigma_F(X) \rangle_{i,\zeta})$  is the standard deviation across all  $\langle \sigma_F(X) \rangle_{i,\zeta}$  values. Configurations having a high uncertainty criterion score reflect regions of the configurational space that are not thoroughly sampled by the training dataset. In other words, high values for the uncertainty criterion reveal “blind spots” in the current iteration of the MLP. Therefore, configurations are sorted in order of their uncertainty criterion scores from highest to lowest, and are then queued for labeling and addition to the training dataset in this order. To maximize the benefit of additional reference calculations while managing the computational expense accrued for each iteration of the MLP, we assume that the configurations with the lowest uncertainty criterion scores will result in less significant improvements to MLP performance and label only the configurations having the highest uncertainty criterion scores with DFT +  $U$  for addition to the training dataset. A new committee of models is then trained on the augmented dataset, serving as the basis for the next active learning sweep. This active learning procedure is repeated until sufficiently few configurations are selected at a threshold of 14.5 meV/atom for energies and 98.6 meV/Å for forces. Because of the heightened disorder of structures drawn from high-temperature equilibrations and the corresponding higher uncertainties across MLP committee member predictions, later-stage active learning sweeps were performed across a narrower temperature range of 2000 K to 3000 K. For the final iteration of the MLP, MLP-MD simulations in LAMMPS ranging in temperature from 300 K to 3000 K remain stable when equilibrated under an NPT ensemble for 50 ps. More detailed information regarding the configurations added for each active learning sweep can be found in Sec. S2 of the Supplemental Material [63].

The stability of MLP simulations is also improved by the addition of larger supercell configurations to the training dataset. In running benchmarking MLP-MD simulations with each iteration of the potential, it became apparent that simulation stability decreased for supercells larger than those included in the training dataset (e.g.,  $4 \times 4 \times 4$   $\text{UO}_2$  supercells with 768 atoms). However, the rapidly increasing computational cost of performing quantum mechanical labeling calculations for these larger supercells is prohibitive. In order to increase the size of configurations in the training

dataset without necessitating further expensive labeling, the active learning procedure described above is modified to carefully select large supercells for which MLP prediction errors are sufficiently low to be considered as accurate as expensive quantum mechanical calculations. In the standard active learning procedure, configurations with the *highest* uncertainty criterion scores are queued for labeling and subsequently added to the training dataset. In the modified procedure, configurations with the *lowest* uncertainty criterion scores are added to the training dataset with the corresponding mean MACE energy and force labels across the committee of MLPs. This procedure is similar to knowledge distillation techniques that have been employed with the goal of student models outperforming teacher models while having the same complexity and number of trainable weights, as opposed to more traditional knowledge distillation in which the student models are designed to be more compact than teacher models [64,65]. Ensemble knowledge distillation methods, in which averaged metrics from an ensemble of teacher models are used to train student models, have resulted in enhanced MD speed and stability [66,67]. In this work, we combine such ensemble knowledge distillation methods in tandem with the calculation of the uncertainty metric given in Eq. (1) to train student models with ensemble predictions only for high-confidence configurations.

On average, the standard deviation in energy predictions across committee members for a given configuration selected via this method is 1.37 meV/atom. This procedure especially assists in selecting configurations for which uncertainty in force predictions is low. For the initial active learning sweeps, the standard deviation in force predictions averaged over all configurations is 132.26 meV/Å. Force errors for configurations acquired by the selection procedure for larger supercells are much lower, at 22.19 meV/Å. Beyond monitoring the statistical energy and force prediction errors, this method is validated through the MLP-MD simulations that are subsequently performed to benchmark the performance of the potential. By ensuring that the thermal expansion, heat capacity, and elastic stiffness tensor for uranium dioxide obtained by the MLP align with experimental results, the active learning and supercell selection procedures are shown to result in robust and physical improvements to the potential.

### 3. Adaptations to the sampling procedure for xenon inclusion in the training dataset

Constructing the training dataset for a MACE MLP for  $\text{UO}_2$  with xenon gas inclusion follows the same sequence of initial sampling and active learning phases. Because instabilities in the molecular dynamics simulations were encountered when sampling with the CRG potential for actinide oxides with fission gas [68], xenon-bearing systems are evolved in ASE using the MACE-MP-0 MLP [69] as a preliminary scan of the configurational space. MACE-MP-0 is an interatomic potential trained using the MACE architecture and the Materials Project Trajectory (MPtrj) dataset [70], which spans the periodic table. All configurations used in these rudimentary equilibrations are 24-atom  $\text{UO}_2$  supercells with either (i) one xenon atom incorporated into an interstitial site, (ii) one xenon atom incorporated into a Schottky defect site, or (iii) two

total xenon atoms, one in an interstitial site and the other in a Schottky defect site. By considering Xe-Xe interactions, this third configuration type lays the foundation for larger-scale simulations investigating the experimentally observed aggregation of xenon into nanometer-scale bubbles in the rim region of  $\text{UO}_2$  fuel pellets [71]. These initial configurations are equilibrated in the canonical (NVT) ensemble for 50 ps with a timestep of 2 fs. Langevin dynamics are implemented with a friction coefficient of 1 ps<sup>-1</sup>. Again, 250 configurations are extracted from the simulation trajectory at intervals of 0.2 ps.

We again employ the query-by-committee active learning procedure to improve the performance and stability of the  $\text{UO}_2$  MACE MLP with xenon. Using a committee of seven MACE models and an identical procedure to that described in Sec. II A2, new data points are selected to augment the training dataset. After ten iterations of this active learning procedure have been completed, we reintroduce the  $\text{UO}_2$ -only configurations gathered for the original  $\text{UO}_2$  potential.

While the initial MD equilibrations and subsequent active learning iterations allow for a preliminary scan of xenon-bearing configurations, the concentration of xenon in these small supercells is much greater than that observed in used nuclear fuel. The number of xenon atoms produced in a fuel element at a given burnup can be estimated by

$$N_{\text{Xe}} = Y_{\text{Xe}} \times N_{\text{fission}}, \quad (2)$$

where  $Y_{\text{Xe}}$  is the fission yield of xenon from uranium-235 fission events and  $N_{\text{fission}}$  is the expected number of fission events.  $N_{\text{fission}}$  is directly related to fuel burnup, with each uranium-235 fission event accounting for approximately 200 MeV of the total energy generated per unit fuel mass loaded [72]. Because of this, the total number of xenon atoms produced can be estimated across a range of burnup values. While 62 MWd/kg U is the average burnup of fuel discharged from the current fleet of light and boiling water reactors [73], advanced reactor designs promise to reach burnups higher than 150 MWd/kg U prior to discharge [74]. Therefore, for the current fleet, the estimated percentage xenon produced by weight is 0.441%. For advanced reactor designs achieving higher burnups, this percentage could approach values as high as 1.48 wt%.

To accurately reflect these expected percentages by weight of xenon, supercell sizes must be larger than desirable for expensive quantum mechanical labeling computations. For extremely high burnups above 150 MWd/kg U, a 96-atom  $2 \times 2 \times 2$  supercell with one xenon atom equates to the predicted weight percentage of xenon. While such calculations are feasible, this high burnup is far from characteristic. For a fuel element with a local burnup of 50 MWd/kg U, a realistic value for uranium oxide fuel used in a current power reactor, a 324-atom  $3 \times 3 \times 3$  unit cell with one xenon atom would be needed to equate to the total amount of xenon expected to have evolved over the entire operational lifetime of the fuel. Because of the diffusion of fission gas atoms in oxide fuels, the experimentally observed local xenon concentration necessarily deviates from these estimates [71,75,76]. Even so, fuel regions with high fission gas retention still reach approximately 0.8 wt% xenon, corresponding to a 216-atom supercell with a single xenon atom.

Therefore, in order to include xenon-bearing supercells of sufficient size in the training dataset without incurring exorbitant computational cost, the supercell selection process described in Sec. II A2 for uranium dioxide is modified and employed to acquire xenon-bearing supercells of sufficient size. After the ten active learning iterations and reintroduction of the  $\text{UO}_2$ -only configurations, the committee of MACE MLPs is used to make energy and force predictions for larger supercells with appropriate xenon concentrations. Configurations for which committee agreement is high are carefully selected for addition to the training dataset. Just as with the  $\text{UO}_2$  MACE MLP, xenon incorporation energies and migration energetics are monitored before and after the addition of larger supercells to the dataset to ensure that the fidelity of the model is maintained. Representative xenon-bearing configurations are shown in Fig. 1.

#### 4. Density functional theory for uranium oxides

Configuration energy and force labels are calculated using density functional theory. DFT calculations are performed using the Vienna *ab initio* simulation package (VASP) [77–79]. Projected augmented wave pseudopotentials are used to construct descriptions of the ions, for which uranium has 14 valence electrons ( $5f^2 6s^2 6p^6 6d^2 7s^2$ ), oxygen has six valence electrons ( $2s^2 2p^4$ ), and xenon has eight valence electrons ( $5s^2 5p^6$ ). A consistent plane wave cutoff of 650 eV is applied to all calculations [80,81]. The exchange-correlation energy is modeled using the Perdew, Burke, and Erzerhof parametrization of the generalized gradient approximation, revised for solids (PBEsol) [82,83]. A  $\mathbf{k}$ -mesh spacing of  $0.25 \text{ \AA}^{-1}$  is found to be sufficient to achieve a convergence tolerance of  $\pm 0.5 \text{ meV/atom}$ . The tetrahedron method with Blöchl corrections is used to integrate over the Brillouin zone [84].

It is well known that the self-interaction error of standard density functional theory renders the treatment of  $d$ - and  $f$ -electrons inaccurate [85]. For example, standard DFT predicts that  $\text{UO}_2$  is a metal, when it is in fact an insulator. A common solution used to restore electron localization in such strongly correlated materials is to add the Hubbard  $U$  parameter correction to the exchange-correlation functional [86]. We introduce this energetic penalty to delocalization according to the rotationally invariant Dudarev approach with  $U = 4.50 \text{ eV}$  and  $J = 0.50 \text{ eV}$  [87]. The resultant effective  $U$  parameter of  $4.00 \text{ eV}$  is reflective of values typical for DFT +  $U$  calculations of  $\text{UO}_2$  [23,87–89], as it has been shown to align with both core photoemission spectra for actinide dioxides [90] and the range of effective  $U$  values found for  $\text{UO}_2$  using *ab initio* methods [91–93].

However, because the introduction of this  $U$  parameter energetically penalizes partial orbital occupation, calculations may converge to local minima in the potential energy surface. Several methods have been introduced to avoid convergence into these metastable states, including occupation matrix control (OMC) [24] and  $U$ -ramping [25]. Even so, these methods do not guarantee convergence to the ground state. Additionally, OMC methods are rooted in the crystal field splitting of  $f$ -orbitals in a cubic field [24]. At high temperatures, disorder in the crystal lattice results in deviations from such a cubic geometry, rendering the implementation of OMC inconsistent

across all training configuration geometries. Because of the computational expense of implementing such methods without guarantee of their fidelity, especially for the number of calculations necessary to generate a sufficiently large training dataset, neither were employed for the labeling of atomic configurations for potential fitting. In Sec. S5 of the Supplemental Material, however, we demonstrate that the MACE MLPs trained in this work still robustly capture ground-state energies of configurations that otherwise result in metastable solutions when a  $U$ -ramping or OMC procedure is not employed [63].

Experimentally,  $\text{UO}_2$  has been found to exhibit non-collinear  $3\mathbf{k}$  antiferromagnetic ordering below its Néel transition temperature,  $T_n = 30.8 \text{ K}$  [94–96]. While recent work has considered this noncollinear magnetism through the inclusion of spin-orbit coupling [97,98], the computational expense can be prohibitive when many calculations are required. For this reason, first-principles studies of  $\text{UO}_2$  generally neglect spin-orbit coupling and instead approximate the ground-state magnetic state to have collinear  $1\mathbf{k}$  antiferromagnetic ordering [23]. This compromise has been justified by the argument that spin-orbit coupling is more than an order of magnitude weaker than Coulombic interactions in  $5f$  systems and that energetic discrepancies are marginal [88]. The inclusion of relativistic effects is also expected to increase the number of metastable states [99]. For these reasons, in addition to the extravagant computational expense that would be accrued by implementing spin-orbit coupling for thousands of DFT calculations, a  $1\mathbf{k}$  antiferromagnetic configuration is assumed in which the spins of uranium atoms alternate along the  $[001]$  direction.

However, while this  $1\mathbf{k}$  ordering is the current standard for DFT +  $U$  calculations of  $\text{UO}_2$ ,  $\text{UO}_2$  exhibits such long-range order only at low temperatures and is paramagnetic at all temperatures relevant for in-reactor use. The  $1\mathbf{k}$  ordering is nonetheless employed here in line with our aim to reproduce DFT +  $U$ -level accuracy using MLPs, though we acknowledge improved magnetic treatments as an important avenue for future work.

#### B. MACE architecture and training

Machine-learned interatomic potentials are trained using the MACE architecture. MACE is a message-passing neural network that uses a computationally efficient tensor product structure to parametrize the mapping of a given atomic configuration to its corresponding potential energy surface [50,51]. Crucially, MACE employs the generalized Clebsch-Gordan coefficients to symmetrize higher-order, equivariant tensor features. Because many-body messages are constructed as linear combinations of these equivariant features, MACE MLPs are better able to capture physical properties that transform in particular ways under a given symmetry operation [50]. A more detailed description of the MACE architecture, summarized from Batatia *et al.* [51] and Kovács *et al.* [100], can be found in Sec. S1 of the Supplemental Material [63].

In this work, two message-passing layers are used with 128 equivariant feature channels. All radial features are generated using eight Bessel basis functions. A fifth-degree polynomial envelope is used to ensure that these functions decay to zero as the interatomic distance approaches a cutoff

radius of 5.0 Å, which is extended to 10.0 Å by the usage of two-layered models. Angular features are described with spherical harmonics up to  $l = 3$ . The maximum body order employed for construction of higher-order features is  $\nu = 3$ . These higher-order features are also symmetrized using generalized Clebsch-Gordan coefficients to ensure  $L$ -equivariance, where  $L = 1$  for the models presented in this work.

MACE MLPs are trained using a single NVIDIA V100 GPU. For all training iterations, a random 10% of the dataset is withheld as a test set for model performance evaluation. The training dataset is composed of the remaining 90%. Validation is performed on 10% of the training dataset. Training data is reshuffled after each epoch of training. A stochastic weight-averaging procedure is used in the final 20% of training epochs in which the energy weighting factor in the loss function is increased by a factor of 1000 to decrease errors in energy predictions.

For the MACE MLP for  $\text{UO}_2$ , new models are trained for each iteration of the potential using sequentially augmented datasets from the active learning procedure described above. For the MACE MLP for  $\text{UO}_2$  with xenon inclusion, the most recent iteration of the MLP is used as a “foundation model,” with the fitted parameters being fine-tuned with the augmented dataset from active learning.

### C. MD-MLP benchmarking for thermomechanical properties

The MACE potential for  $\text{UO}_2$  is benchmarked against experimental data, results from semiempirical potentials, and results from recently reported machine-learned potentials. All benchmarking studies are performed by running MD-MLP simulations in LAMMPS with the  $\text{UO}_2$  MACE MLP.

#### 1. Lattice expansion and heat capacity

To calculate the thermal expansion and heat capacities of  $\text{UO}_2$  from 300 K to 3000 K, a  $3 \times 2 \times 2$  supercell of the 12-atom fluorite unit cell is equilibrated at zero pressure under an NPT ensemble for 60 ps with a timestep of 1 fs. Simulation temperatures are set by initializing atom velocities according to the corresponding Maxwell–Boltzmann distribution. A Nosé–Hoover thermostat and an anisotropic Nosé–Hoover barostat are used to maintain the temperature and pressure of the system; damping parameters are 0.1 ps and 0.5 ps, respectively.

The lattice parameters and enthalpies are then averaged over the final 30 ps of the simulation. Lattice parameters are directly extracted, reflecting the thermal expansion of  $\text{UO}_2$ . The specific heat capacities,  $c_p$ , are found by differentiating the enthalpy increment with respect to temperature,

$$c_p = \frac{1}{n} \left( \frac{\partial H}{\partial T} \right)_p, \quad (3)$$

where  $n$  is the number of moles and  $H$  is given by a polynomial fit to the enthalpy increment.

#### 2. Elastic properties

The temperature-dependent elastic stiffness tensor was obtained using a generalized form of Hooke’s law. For small deformations, the elastic stiffness tensor  $C_{ij}$  is defined as the

proportionality constant between the strain and stress tensors [101]. This is expressed in Voigt notation as

$$\sigma_i = C_{ij} \epsilon_j, \quad (4)$$

where  $\sigma$  is the stress tensor,  $\epsilon$  is the strain tensor, and  $i$  and  $j$  are indices representing pairs of the Cartesian coordinates. With Voigt notation, where  $xx = 1$ ,  $yy = 2$ ,  $zz = 3$ ,  $yz = 4$ ,  $xz = 5$ , and  $xy = 6$ , the elastic stiffness tensor can be expressed as a  $6 \times 6$  matrix. Because of symmetry,  $\epsilon_4 = 2\epsilon_{yz}$ ,  $\epsilon_5 = 2\epsilon_{xz}$ , and  $\epsilon_6 = 2\epsilon_{xy}$ .

Simulations follow the methodology employed by Clavier and Thompson [102]. At each temperature of interest, a  $3 \times 3 \times 3$  supercell of the 12-atom  $\text{UO}_2$  unit cell is equilibrated in an NPT ensemble for 30 ps with a timestep of 1 fs. A Nosé–Hoover thermostat with a damping parameter of 0.1 ps is combined with a Nosé–Hoover anisotropic barostat having a damping parameter of 0.5 ps to maintain simulation parameters. The volume of the simulation box is averaged over the final 5 ps of the equilibration to obtain a nominal volume at the temperature of interest.

A series of elementary deformations are then applied to a simulation box having this nominal volume. Scaling factors of  $\pm 0.75\%$  and  $\pm 1.5\%$  are applied to the simulation box for every  $j$  from 1 to 6. Scaled systems are then equilibrated in an NVT ensemble for 60 ps with a timestep of 1 fs and a Nosé–Hoover thermostat damping parameter of 0.1 ps. For each of these applied strains ( $\epsilon_j$ ), the stress tensor ( $\sigma_i$ ) is taken to be the inverse of the pressure tensor and averaged for  $i = 1 - 6$  over the final 30 ps of the equilibration [103].

After this series of deformations has been performed at a given temperature, a linear fit is applied to the resulting stress-strain curve to obtain element  $C_{ij}$  of the elastic stiffness tensor.

The bulk modulus is then calculated using the standard relationship between  $C_{11}$  and  $C_{12}$ ,

$$K = \frac{C_{11} + 2C_{12}}{3}. \quad (5)$$

Zener ratios are also calculated as a measure of crystal structure anisotropy [104] using

$$\alpha_r = \frac{2C_{44}}{C_{11} - C_{12}}. \quad (6)$$

Elastic isotropy is defined as  $\alpha_r = 1$ , with deviations from  $\alpha_r = 1$  being indicative of an elastically anisotropic material for which stress responses are not equivalent in all directions.

### D. Xenon incorporation energy formalism

Energies are calculated for xenon incorporation into an interstitial site, an oxygen vacancy, a uranium vacancy, or a bound Schottky defect. These reactions are given by the following expressions:

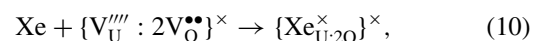
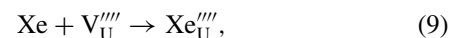
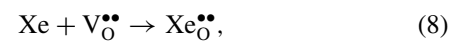


TABLE I. Composition of the training dataset for the  $\text{UO}_2$  MACE MLP.

| Configuration Type         | Number of Configurations |
|----------------------------|--------------------------|
| Defect-Free, Zero-Pressure | 3718                     |
| Defect-Free, Expanded      | 807                      |
| Frenkel Defect (O)         | 1182                     |
| Frenkel Defect (U)         | 521                      |
| Schottky Defect            | 692                      |
| Total Configurations       | 6920                     |
| Training Configurations    | 6228                     |
| Test Configurations        | 692                      |

where defects are denoted in Kröger–Vink notation [105]. For xenon incorporation into Schottky defect sites, energies are reported for three distinct defect configurations in which the oxygen vacancies occupy either first-, second-, or third-nearest-neighbor sites with respect to one another.

### E. Energetics of xenon diffusion

Migration barriers for the diffusion of defect clusters within the  $\text{UO}_2$  fuel lattice are calculated using the nudged elastic band (NEB) method in ASE [106–109]. Initial and final states are first relaxed with the FIRE optimizer [110] until the force on all individual atoms is less than  $0.01 \text{ eV}/\text{\AA}$ . Ten beads are used for all atomic motions. Intermediate images are generated using the image-dependent pair potential (IDPP) approach [111]. All images are again relaxed with the FIRE optimizer until forces are converged to within  $0.05 \text{ eV}/\text{\AA}$  to obtain the minimum-energy pathway (MEP). This methodology is implemented for each committee member of the final MACE MLP, generating a distinct pathway with each potential.

## III. RESULTS AND DISCUSSION

### A. Construction of training datasets

The composition of the training dataset for the  $\text{UO}_2$  MACE MLP is shown in Table I. The dataset is comprised of 6920 distinct labeled configurations. Of these, 692 configurations were held out as a test dataset for testing of the MLP.

The composition of the training dataset for the  $\text{UO}_2$  MACE MLP with xenon inclusion is shown in Table II. The dataset is comprised of 11 362 labeled configurations, with 1136 of these being held out as a test dataset. This dataset includes all 6290 labeled configurations used to train the  $\text{UO}_2$  MACE MLP, demonstrating the utility of leveraging preexisting datasets to extend the application space for a given model.

The relatively small size of these training datasets can be attributed to the strengths of the MACE architecture, the active learning methodology employed, and the intended use cases for the MLP. Because of the higher-order equivariant features employed by MACE models, it is possible to achieve exceptional performance for a broad range of temperatures using 6920 and 11 362 configurations to train the  $\text{UO}_2$  and  $\text{UO}_2$  with Xe MLPs, respectively. In view of differences in model architecture and training choices, the dataset size used to construct other recently reported  $\text{UO}_2$  MLPs varies greatly.

TABLE II. Composition of the training dataset for the  $\text{UO}_2$  MACE MLP with xenon.

| Configuration Type                             | Number of Configurations |
|------------------------------------------------|--------------------------|
| $\text{Xe}_i$                                  | 2,926                    |
| $\text{Xe}_{\text{U:20}}^\times$               | 761                      |
| $\text{Xe}_i + \text{Xe}_{\text{U:20}}^\times$ | 280                      |
| Isolated Xe                                    | 1                        |
| Increased [Xe]                                 | 474                      |
| Defect-Free, Zero-Pressure                     | 3718                     |
| Defect-Free, Expanded                          | 807                      |
| Frenkel Defect (O)                             | 1182                     |
| Frenkel Defect (U)                             | 521                      |
| Schottky Defect                                | 692                      |
| Total Configurations                           | 11 362                   |
| Training Configurations                        | 10 226                   |
| Test Configurations                            | 1136                     |

Between their two MLPs, Stippell *et al.* utilize 25 000 configurations for their DFT-based MLP but then use a refined dataset of 2000 DFT +  $U$  solutions to retrain the DFT-trained model [43]. Similarly, Konashi *et al.* collect 20 000 configurations [45]. However, though the ANI and DeePMD-kit architectures used to train these potentials require larger training datasets, their other features such as evaluation time may render them advantageous for certain use cases.

For any of these MLPs, the breadth of the configurational space sampled also affects the necessary dataset size. Because the use cases for these MLPs were explicitly defined prior to their construction, the active learning methodology employed to sample the configuration space was focused. This results in smaller dataset sizes than would be necessary for a more generalizable MLP. Even so, we find that the MACE MLPs capture a broad region of the potential energy surface with a relatively small dataset. Importantly, this reduction in the number of training data points reduces the cost of labeling reference configurations with DFT +  $U$ .

Further detail regarding the composition of these datasets, as well as their evolution through active learning iterations and the dataset augmentation workflow, is provided in Sec. S2 of the Supplemental Material [63].

### B. MACE potential performance

Parity plots evaluating MLP fitting to reference DFT +  $U$  energies and forces and giving the  $R^2$ , RMSE, and MAE values for MLP performance on the test dataset are shown in Fig. 2. RMSE and MAE values are additionally given in Table III. Performance statistics for both MACE MLPs are compared to those reported for other  $\text{UO}_2$  MLPs in Table S3 within the Supplemental Material [63]. The RMSE, MAE, and  $R^2$  values for both energies and forces align well with reported values for MACE MLPs trained for other systems [69]. These statistics are also similar to those reported for other  $\text{UO}_2$  MLPs [43–45]. For both MACE MLPs, force errors are notably lower than those found for other  $\text{UO}_2$  MLPs. While these metrics are obtained through testing on different datasets across referenced MLPs and therefore cannot be directly compared, it remains worthwhile to note that force errors for our

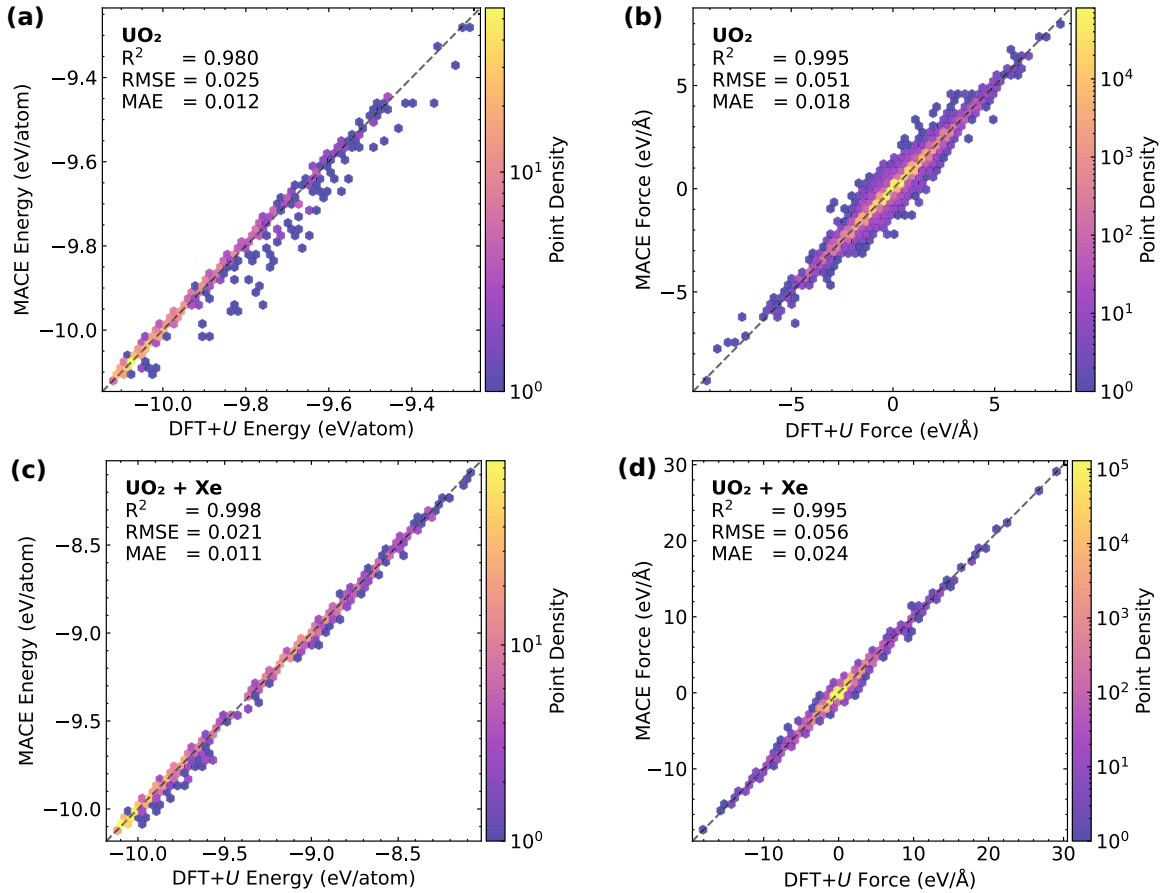


FIG. 2. Performance of the  $\text{UO}_2$  MACE MLP (a) and (b) and the  $\text{UO}_2$  with Xe MACE MLP (c) and (d) through comparison with the held-out test set of reference DFT +  $U$  data.

MACE MLPs are lower by an order of magnitude. This is attributable to the fully equivariant features employed in the construction of the MACE potentials, which capture a higher degree of accuracy when predicting forces. Additionally, the loss function employed by the MACE MLP training protocol weights the force prediction errors more heavily than energy prediction errors by a factor of 100, meaning that the forces are strongly prioritized in the final MACE MLP parameter weights.

It is worthwhile to note that a number of predictions for these test configurations fall below the parity line, especially for the  $\text{UO}_2$  potential. These off-diagonal configurations can be attributed to reference DFT +  $U$  calculations that converged to metastable states with energies above the ground state, as evidenced by the underprediction of DFT +  $U$  energies by our MACE potential. Observing similar trends, Stippell *et al.* also ascribe this phenomenon to the tendency

of DFT +  $U$  reference calculations to converge to metastable states and reject these configurations from their datasets. In Sec. S5 of the Supplemental Material [63], we show that retaining these configurations in our training dataset does not compromise the fidelity of the MACE MLP, as the underpredictions of DFT +  $U$  energies by our MACE potential are generally reflective of ground-state energies.

### C. Thermophysical and thermomechanical properties of $\text{UO}_2$

The MACE MLP for  $\text{UO}_2$  was then employed to calculate thermophysical and thermomechanical properties of  $\text{UO}_2$ . The thermal expansion, heat capacity, and elasticity matrix are all crucial parameters for larger-scale fuel performance models. The MACE MLP performance is benchmarked against widely used equations fitted to experimental data [3,112], semiempirical pair potentials [7,9], and other neural network

TABLE III. Statistics for energy and force predictions made by both the  $\text{UO}_2$  MACE MLP and the  $\text{UO}_2$  MACE MLP with xenon inclusion, relative to reference DFT +  $U$  calculations.

| MLP                                  | Energy (meV/atom) |       | Force (meV/Å) |       |
|--------------------------------------|-------------------|-------|---------------|-------|
|                                      | RMSE              | MAE   | RMSE          | MAE   |
| This work, $\text{UO}_2$             | 25.02             | 12.33 | 51.26         | 17.61 |
| This work, $\text{UO}_2 + \text{Xe}$ | 20.87             | 11.49 | 56.50         | 23.80 |

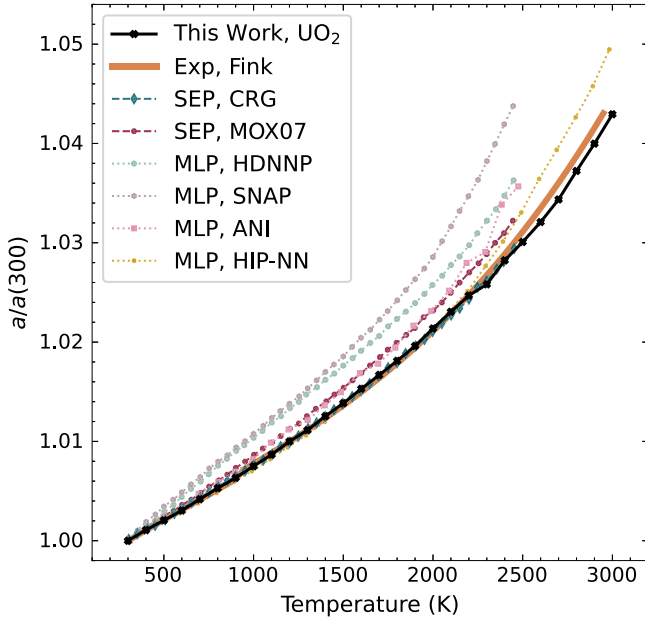


FIG. 3. Thermal lattice expansion for  $\text{UO}_2$  relative to lattice parameter at 300 K. Our MACE MLP for  $\text{UO}_2$  demonstrates excellent agreement with experiment. Because the CRG semiempirical potential was fit to experimentally observed lattice parameters and thermal expansion, its predictions also closely replicate experimental values.

potentials for  $\text{UO}_2$  [43–46]. While the MACE MLP for  $\text{UO}_2$  was used for these benchmarking studies, results for the  $\text{UO}_2$  MLP with Xe are also included in Sec. S6 of the Supplemental Material [63].

### 1. Lattice expansion

The thermal dependence of the lattice parameter from 300 K to 3000 K is shown in Fig. 3. The MACE MLP reproduces the experimental results summarized by Fink with high accuracy, deviating on average from this experimental correlation by only 0.042%. At temperatures above 2500 K, the MACE MLP slightly underestimates the lattice parameter by 0.139% on average. The MACE MLP improves upon the overestimation of the lattice parameter by the MOX-07, HDNNP, SNAP, and Stippell potentials. The CRG pair potential was explicitly fitted to the lattice expansion of  $\text{UO}_2$ , meaning it most closely reproduces the experimental values.

It is worthwhile to note that while the thermal expansion relative to the lattice parameter at 300 K is reproduced by the MACE MLP, the absolute results are inflated from experimental values by 0.81%, on average. For DFT +  $U$  calculations with the PBE functional, overpredictions in the lattice parameter are commonly documented [24,43,89]. Increasing the effective  $U$  value from 2.0 eV to 7.0 eV has been shown to result in an increase of the lattice parameter up to 5.53 Å, demonstrating the sensitivity of the lattice parameter to the Hubbard correction [89]. This inflation in the lattice parameter can be attributed to the localization of electrons with increasing  $U$  [24]. Chen and Kaltsoyannis also found high-energy solutions with large lattice parameters for solutions having particular occupation matrices [89]. Even though the MACE MLPs reproduce this expected overprediction in

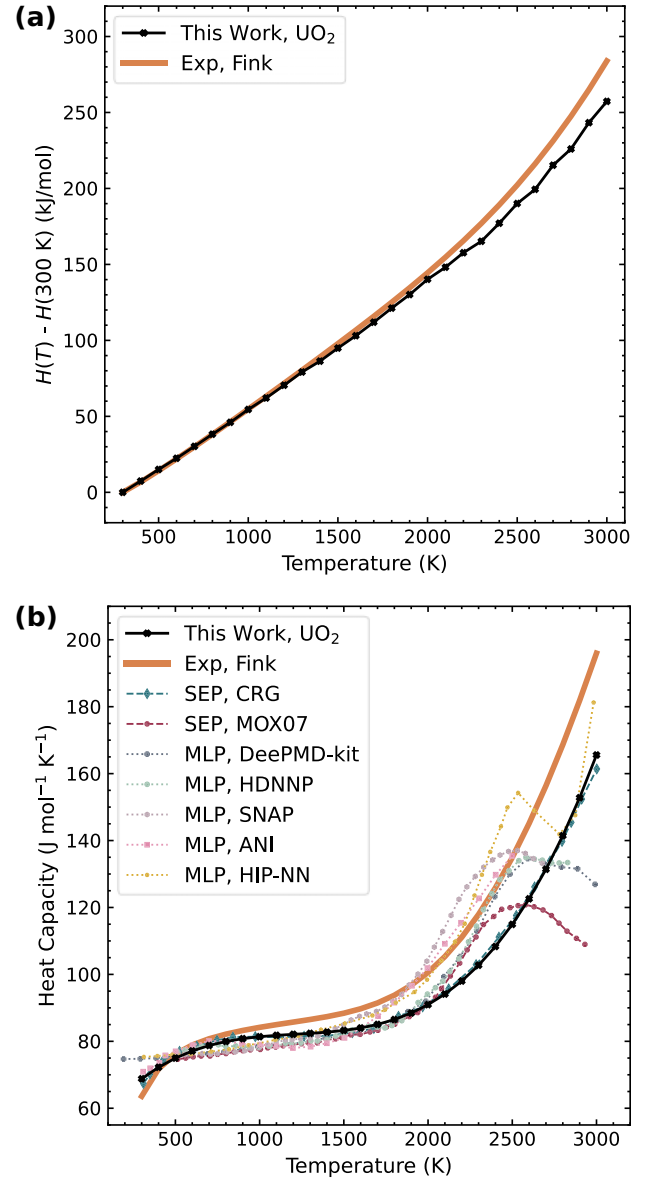


FIG. 4. Enthalpy increment for  $\text{UO}_2$  as calculated with the MACE MLP for  $\text{UO}_2$  (a) and the heat capacity (b), obtained by fitting the enthalpy increment with a fourth-degree polynomial.

lattice expansion, the trend itself for the  $\text{UO}_2$  lattice expansion using the MACE MLP trained on DFT +  $U$  reference data is remarkably accurate with respect to experiment. A comparison of the absolute temperature-dependent lattice parameters found using the MACE MLP to experiment is given in Fig. S6 within the Supplemental Material [63].

### 2. Heat capacity

The enthalpy increment extracted from constant-pressure MD simulations is shown in Fig. 4. Results are compared to a best fit of available enthalpy data given by Fink. At temperatures ranging from 300 K to 2300 K, MACE MLP results for the enthalpy increment are within 5% of the Fink correlation. At high temperatures above 2300 K, the underestimations made by the MACE MLP range from 6% to 10%.

Previous work has obtained the specific heat by fitting a polynomial function to this enthalpy increment and taking its first derivative [3,43–46]. We employ this technique, with heat capacity results given in Fig. 4.

We provide the raw enthalpy increment results in addition to heat capacities derived from polynomial fits of lower and higher orders because of disagreement in the literature regarding a premelting transition in  $\text{UO}_2$ . Fink makes note of a  $\lambda$ -phase transition in solid  $\text{UO}_2$  at 2670 K, supported by results from neutron scattering experiments by Hutchings *et al.* [112] and cooling curve analysis by Hiernaut *et al.* [113]. Ronchi and Hyland [114] provided a theoretical explanation for this observed  $\lambda$ -phase transition: they found that the increase in the heat capacity from room temperature to 1000 K is because of harmonic lattice vibrations, the increase from 1000 K to 1500 K is attributable to anharmonic lattice vibrations, the increase from 1500 K to 2670 K is governed by the formation of Frenkel defects, and the  $\lambda$ -phase transition marks a shift where the Frenkel defect concentration is saturated and Schottky defects become more crucial to changes in heat capacity.

This identification of a Bredig transition [115], which suggests that there is a peak in the heat capacity at  $T = 0.8T_m$ , where  $T_m$  is the melting temperature, was also reported by other experimental and theoretical works [10,116,117]. However, many have been unable to replicate this peak in the heat capacity [118,119]. Ronchi, who had previously provided a theoretical basis for the transition, later collected conflicting data for the heat capacity above 2700 K. This updated work argues instead that the effects of the  $\lambda$ -phase transition are negligible and an increase in  $C_p$  is still observed above 2670 K [118]. Rectifying these disparities is a nontrivial exercise because of challenges in experimental verification of the heat capacity at temperatures above 2300 K. For this reason, the Fink recommendation of a strictly increasing heat capacity remains widely implemented.

Recently reported MLPs for  $\text{UO}_2$  have highlighted their reproduction of this Bredig transition as a primary strength of their potentials [44]. Others have been more conservative in reporting the transition, noting that it is not experimentally confirmed and that the peak they observe in the heat capacity is gradual [45]. A more detailed analysis of the sensitivities of the heat capacity at both high and low temperatures to the order of the polynomial fit, including a demonstration that a Bredig transition peak can be derived from the same enthalpy increment data by increasing the order of the polynomial fit, appears in Sec. S6B of the Supplemental Material [63]. For this reason, the raw enthalpy increment data remains a crucial benchmark for  $\text{UO}_2$  MLP performance.

Regardless of the fit used, the MACE MLP predictions show strong agreement with the Fink recommendation and reported potentials. Heat capacity predictions are particularly improved at temperatures below 500 K in comparison to other MLPs. Though the MACE MLP underpredicts the specific heat at higher temperatures, the trend itself follows the Fink correlation. Further experimental confirmation is required to make a determination regarding the Bredig transition in the heat capacity, but the underlying enthalpy increment found using the MACE MLP are accurate with respect to reported experimental results.

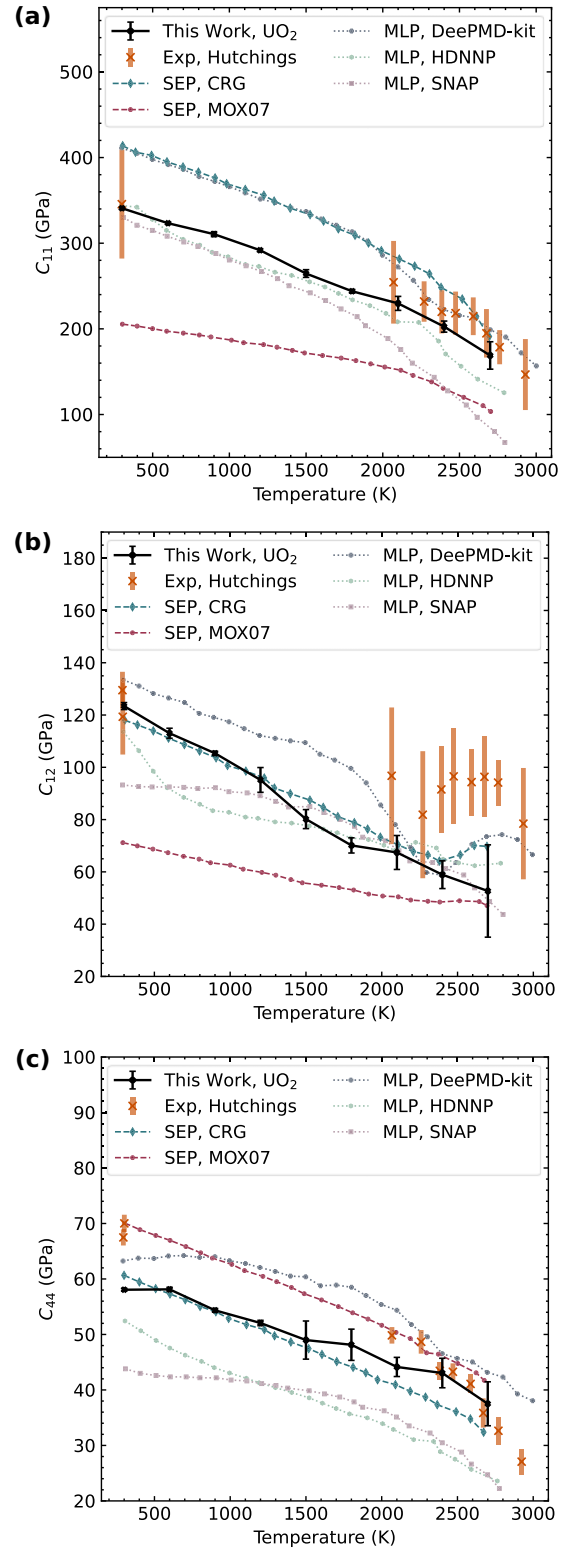


FIG. 5.  $C_{11}$  (a),  $C_{12}$  (b), and  $C_{44}$  (c) for  $\text{UO}_2$ , obtained by applying small elementary deformations to the supercell and averaging the stress tensor.

### 3. Elastic properties

Temperature-dependent  $C_{11}$ ,  $C_{12}$ , and  $C_{44}$  elastic constants are given in Fig. 5. Errors are given as the standard deviation across symmetry-equivalent entries of the

elasticity matrix, where  $C_{11} = C_{22} = C_{33}$ ,  $C_{12} = C_{21} = C_{23} = C_{32} = C_{13} = C_{31}$ , and  $C_{44} = C_{55} = C_{66}$ . All experimental data points presented for comparison are those made with the triple-axis spectrometer in the PLUTO reactor at Harwell, reported by Hutchings [112]. Zero-temperature elastic constants are also provided in Table S5 within the Supplemental Material [63].

Uncertainties are generally larger at higher temperatures because of the inherent disorder introduced into the system as the melting temperature is approached. At this limit, the assumption of cubic symmetry becomes less accurate and  $C_{11}$ ,  $C_{12}$ , and  $C_{44}$  are no longer completely descriptive of mechanical behavior.

Generally, material softening is observed as temperature increases. The MACE MLP predicts relatively linear decreases in both  $C_{11}$  and  $C_{44}$  from 300 K to 2000 K. An onset of a more rapid, nonlinear decrease in both  $C_{11}$  and  $C_{44}$  appears to occur above 2000 K. This trend is also observable in the Hutchings neutron scattering data and for the CRG, HDNNP, SNAP, and Konashi potentials. Dubois *et al.* attribute this nonlinear decrease to the aforementioned Bredig transition, with rapid softening resulting from increased mobility of the oxygen sublattice. As previously discussed, we note that this Bredig transition is not fully accepted for  $\text{UO}_2$ . Regardless, such softening is unsurprising at high temperatures approaching the melting point of  $\text{UO}_2$ . Values found using the MACE MLP for  $C_{11}$  fall within error of the Hutchings results despite slightly underestimating the absolute values reported. Similarly, MACE MLP results for  $C_{44}$  are generally aligned with or underestimate the Hutchings constants. However, the MACE MLP improves on underpredictions made by the HDNNP, SNAP, and MOX07 potentials for  $C_{11}$ . While the Konashi potential appears to more accurately reproduce experimental results for  $C_{11}$ , the MACE MLP improves upon its overprediction of  $C_{44}$ .

Uncertainties are especially large for the  $C_{12}$  constant. Because the  $C_{12}$  constant describes the coupling of strain in one direction and stress in a different direction, it is generally more sensitive to anisotropic effects introduced at high temperatures. Indeed, Zener ratios for all temperatures fall between  $\alpha_r = 0.530$  and  $\alpha_r = 0.645$ , indicating elastic anisotropy. This explains the high uncertainties reported experimentally for  $C_{12}$ , as well as the diversity in trends predicted by available potentials for  $C_{12}$  with increasing temperature. The MOX07 potential predicts a constant decrease in  $C_{12}$  with increasing temperature. The SNAP potential predicts a similar constant decline until 1600 K, but a subsequent nonlinear decrease with a more pronounced profile beyond that temperature threshold. The HDNNP potential, on the other hand, indicates significant softening from 300 K to 800 K. Aligning with the supposed Bredig transition, the CRG and Konashi potentials predict an increase in  $C_{12}$  above 2300 K. The MACE MLP predicts a linear decrease from 300 K to 1200 K, at which point a more rapid decline of  $C_{12}$  is observed. Unlike the other reported potentials, we then observe a return to a more modest decrease in  $C_{12}$  above 2000 K. Refinement of experimental data and consideration of the anisotropic nature of  $\text{UO}_2$  would allow for further clarification of the temperature dependence of the  $C_{12}$  constant, though the MACE MLP demonstrates strong benchmarking against available data.

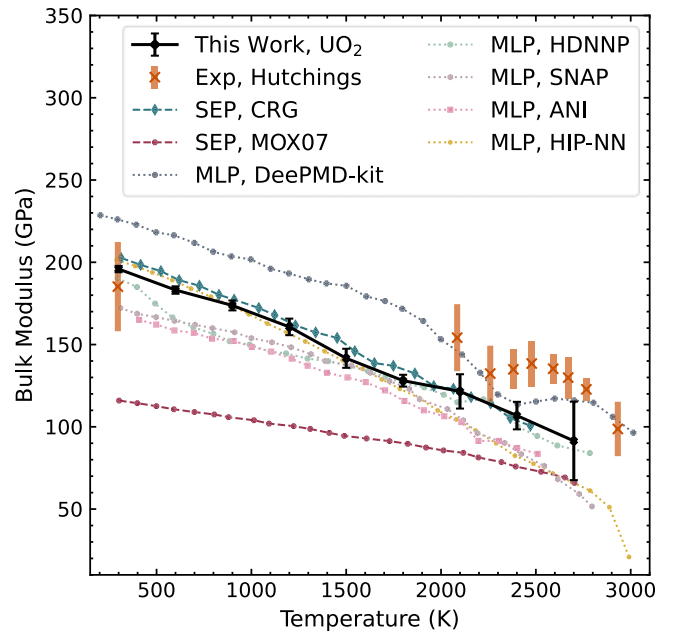


FIG. 6. Bulk modulus for  $\text{UO}_2$ , obtained with the MACE MLP for  $\text{UO}_2$  via relationships between components of the elastic tensor.

Results for the temperature-dependent bulk modulus are given in Fig. 6. The results found with the MACE MLP align reasonably well with the Hutchings dataset and the CRG potential. Additionally, predictions made with the MACE MLP improve upon the underpredictions by the HDNNP, SNAP, and MOX07 potentials. The Konashi potential appears to capture a peak that appears in the bulk modulus at temperatures above 2500 K, yet large errors reported for data points composing this feature in the Hutchings dataset cast doubt on its validity. Calculations for the bulk modulus also assume an elastically isotropic material, and therefore suffer the same uncertainties as those discussed above for the  $C_{12}$  constant.

#### D. Energetics of xenon in $\text{UO}_2$

To demonstrate the utility of the MACE MLP for simulating the diffusion of Xe gas in  $\text{UO}_2$ , we present the energetics of xenon incorporation and diffusion through the fuel matrix. We compare the performance of our models to both DFT +  $U$  reference calculations and results found using semiempirical potentials.

##### 1. Xenon incorporation energies

The incorporation energies of a single xenon atom into an interstitial site, a uranium vacancy, an oxygen vacancy, and Schottky defect sites are given in Table IV. The MACE MLP was used to find xenon incorporation energies into three distinct Schottky defect configurations, for which the oxygen vacancies occupy first-, second-, or third-nearest-neighbor sites with respect to one another. As literature references do not generally specify which of these configurations their incorporation energies represent, those values are given in an additional column.

The MACE MLP for  $\text{UO}_2$  with Xe reproduces the ordering of incorporation energies found by all DFT reference

TABLE IV. Xenon incorporation energies given in eV for the MACE MLP, compared to DFT +  $U$  reference values. Errors are given as the standard deviation in predictions made by all committee members.

|                                                     | MACE MLP(eV)                    | DFT + $U$ (eV)            |                              |                          |                             |
|-----------------------------------------------------|---------------------------------|---------------------------|------------------------------|--------------------------|-----------------------------|
|                                                     | This Work, UO <sub>2</sub> + Xe | Cooper <i>et al.</i> [68] | Thompson and Wolverton [120] | Geng <i>et al.</i> [121] | Nerikar <i>et al.</i> [122] |
| Xe <sub>i</sub>                                     | 9.83 ± 0.21                     | 8.94                      | 9.73                         | 9.75                     | 11.11                       |
| Xe <sub>O</sub> <sup>••</sup>                       | 7.79 ± 0.10                     | 6.85                      |                              | 7.53                     | 9.50                        |
| Xe <sub>U</sub> <sup>'''</sup>                      | 3.03 ± 0.26                     | 3.70                      |                              | 3.87                     | 2.50                        |
| {Xe <sub>U:2O</sub> <sup>x</sup> } <sup>x</sup>     |                                 | 0.49                      | 1.06                         | 0.18                     | 1.38                        |
| {Xe <sub>U:2O,1NN</sub> <sup>x</sup> } <sup>x</sup> | 0.43 ± 0.03                     |                           |                              |                          |                             |
| {Xe <sub>U:2O,2NN</sub> <sup>x</sup> } <sup>x</sup> | 1.30 ± 0.05                     |                           |                              |                          |                             |
| {Xe <sub>U:2O,3NN</sub> <sup>x</sup> } <sup>x</sup> | 1.52 ± 0.11                     |                           |                              |                          |                             |

calculations. This ordering is easily supported with a steric argument. Incorporation of a xenon atom into an interstitial site is the least favorable, as it exerts the most strain on the lattice itself. The oxygen vacancy site is also relatively unfavorable for xenon incorporation, with an incorporation energy of 7.64 eV as found by the MACE MLP. Incorporation into a uranium vacancy site is more favorable than incorporation into an interstitial or oxygen site, as it is a larger site that is better able to accommodate the xenon atom. As expected, incorporating a xenon atom into a Schottky defect site has the lowest energy relative to other trapping sites. We find the incorporation energy to be lowest for a Schottky defect with its oxygen vacancies in first-nearest-neighbor sites with respect to one another.

Other authors have ascribed discrepancies between reference values for these incorporation energies to the tendency of DFT +  $U$  calculations to converge to metastable states [120]. It is likely more significant that, where referenced calculations employ methods to avoid such results, each use a different approach. Nerikar *et al.* [122] do not employ a scheme to mitigate convergence to metastable states, Geng *et al.* [121] develop a “quasiannealing” procedure, and both Cooper *et al.* [68] and Thompson and Wolverton [120] apply  $U$ -ramping to the UO<sub>2</sub> supercell before defect calculations. Therefore, some disagreement between incorporation energies found using varying methods is expected. Even so, differences between the MACE MLP results and reference DFT +  $U$  calculations are comparable to the disparities between DFT +  $U$  results found using different methods to converge to the ground state.

## 2. Schottky defect migration in uranium dioxide

First, we used the diffusion of a Schottky defect through a  $2 \times 2 \times 2$  UO<sub>2</sub> supercell without any xenon incorporation to benchmark the performance of both MACE MLPs in predicting diffusion behavior. This approach is in line with that taken by Thompson and Wolverton [123]. The Schottky defect diffusion pathway is shown in Fig. 7.

Predictions made by both MACE MLPs for the energetics of this Schottky defect diffusion pathway demonstrate reasonable agreement with reported DFT +  $U$  results [123]. DFT +  $U$  solutions are also generated in this work by performing calculations for the configurations generated through the MLP NEB pathways. For these calculations,  $U_{\text{eff}}$  is increased sequentially from 0.0 eV to 4.0 eV in increments of 0.5 eV. Results are shown in Table V.

Both reference DFT +  $U$  calculations and MACE MLP predictions find that the migration of a uranium atom is the rate-limiting step, though the MACE MLPs predict lower migration barriers for the uranium movement than reported DFT +  $U$  results (DFT +  $U$  = 5.88 eV; MACE MLP without Xenon =  $4.67 \pm 0.46$  eV; MACE MLP with Xenon =  $5.09 \pm 0.27$  eV). This discrepancy can be traced to differences in the relative energies of the three distinct Schottky defect configurations. Thompson and Wolverton find that a Schottky defect in which the oxygen vacancies occupy first-nearest-neighbor (1NN) sites with respect to one another has a formation energy of 0.76 eV relative to a Schottky defect in which the oxygen vacancies occupy second-nearest-neighbor (2NN) sites. The MACE MLP for UO<sub>2</sub> with xenon predicts a lower relative formation energy of 0.50 eV for the 1NN configuration, contributing to the reduced migration barrier by both MACE MLPs.

The errors in the migration barrier energies found using our MACE MLPs likely arise from several factors. First, as detailed in Sec. III B, the loss function used as part of the MLP training protocol heavily weights the force errors in comparison to energy errors. This results in decreased precision for metrics governed by system energies, including migration barriers. Additionally, the training datasets for the MACE MLPs do not include configurations drawn from NEB pathways. Therefore, because the MLPs are making predictions for configurations outside of their training domain, higher errors can be expected.

The benefit of fine-tuning the most recent iteration of the MACE MLP (as was done for the UO<sub>2</sub> MLP with xenon inclusion) rather than training a new MLP with each augmented dataset (as was done for the UO<sub>2</sub>-only MLP) is also apparent in the standard deviations between each MLP’s respective committee of members for the uranium migration energetics. While both MACE MLP predictions for the uranium migration barrier are within error of one another, committee members for the UO<sub>2</sub>-only MLP display greater disagreement. When training the MACE MLP for UO<sub>2</sub> with xenon, retaining the parameter weights from a previous iteration and fine-tuning on new data points led to a large reduction in disagreement across committee member predictions.

The two oxygen migrations have very similar energetic costs, with the transition of the Schottky defect from a 1NN to a 2NN configuration having a slightly higher migration barrier than the transition from a 2NN to a 3NN configuration. Thompson and Wolverton attribute this ordering of Schot-

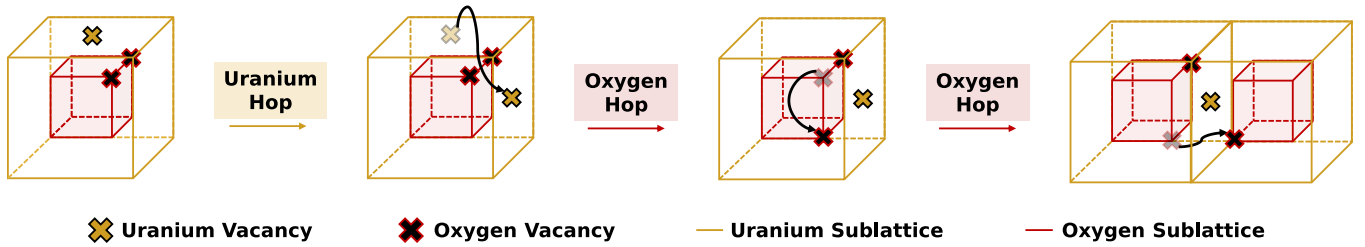


FIG. 7. Diffusion pathway for the migration of a Schottky defect in  $\text{UO}_2$ . Adapted from Thompson and Wolverton [123].

tky defect stability to the interplay between electrostatic and surface energy considerations [120]. Though the 3NN configuration minimizes Coulombic repulsion between the oxygen vacancies, the 1NN configuration minimizes the number of unsatisfied bonds in the lattice by having the most compact vacancy structure. Therefore, we similarly observe that the 2NN configuration is more stable than both the 1NN and 3NN configurations.

### 3. Xenon migration via tetravacancy defect clusters

Subsequently, two diffusion pathways of xenon within tetravacancy defect clusters ( $V_{\text{U}_2}\text{O}_2$ ) in larger  $4 \times 4 \times 4$  supercells were investigated. The so-called “direct diffusion” and “indirect diffusion” pathways are shown in Fig. 8. The primary distinction between the direct and indirect mechanisms is the migration path of the uranium vacancy. The direct pathway, originally proposed by Govers *et al.* [124], involves the migration of a uranium vacancy via first-nearest-neighbor sites with respect to the Xe atom. The indirect pathway, alternatively, was proposed by Thompson and Wolverton [123] and is characterized by the temporary dissociation of the uranium vacancy from the defect cluster to a second-nearest-neighbor site from the xenon. The uranium vacancy then rebinds to the defect cluster in a different first-nearest-neighbor site following the corresponding oxygen motions.

Configuration energy as a function of reaction coordinate is shown for the direct and indirect pathways in Fig. 8, as an average of predictions made by all committee members. Comparisons are made to semiempirical potential results reported by Thompson and Wolverton [123], who evaluated the performance of two pairings of semiempirical potentials. The first pairing is a U-O potential developed by Basak *et al.* [4] with Xe-O and Xe-U potentials formulated by Geng *et al.* [121]. The second pairing combines a U-O potential from Morelon *et al.* [5] with Xe-O and Xe-U pair interactions from Chartier *et al.* [125]. Uranium atom movements are shaded in yellow, while oxygen atom movements are shaded in red. For

comparison to the semiempirical potential results, xenon migrations are shaded in teal. Comparisons of migration barriers found using the MACE MLPs and by several reference studies are given in Table VI.

While these results arise using the same initial and final atomistic structures for each step in the NEB pathway, these structures as well as the intermediate images generated through the NEB protocol are relaxed by each individual potential. This results in more detailed qualitative information regarding each potential’s prediction for the nature of xenon migration in  $\text{UO}_2$ . For example, while Thompson and Wolverton note a small barrier for these xenon migrations at the outset and conclusion of the indirect migration pathway when employing the Basak and Geng potential pairing, MACE MLPs predict a nonexistent barrier because of the equilibration of xenon atoms to the center of the defect cluster.

Overall, uranium movements are the most energetically costly. For the direct pathway, this migration barrier is found to be 5.50 eV. For the indirect pathway, the barrier to uranium motion through the lattice is 0.42 eV lower, at 5.08 eV. While the temporary dissociation of the uranium vacancy from the tetravacancy cluster via the indirect pathway reduces the available size of the xenon trapping site, it also reduces the interactions of the migrating uranium with the xenon atom. Thompson and Wolverton achieve this same result using both semiempirical potential pairings, with the Basak and Geng potentials finding the indirect pathway to have a migration barrier 0.9 eV lower than that of the direct pathway. DFT +  $U$  comparison of the direct and indirect pathways also finds that the temporary dissociation of the uranium vacancy is more favorable by 0.48 eV [19].

Beyond the relative barrier heights of the direct and indirect pathways, the energy traces as a function of reaction coordinate display a handful of differences from reference semiempirical potential results.

With respect to the direct pathway, the oxygen migrations exhibit complicated energetics that deviate from both the Basak/Geng and Morelon/Chartier results. We observe

TABLE V. Migration barriers for the migration of a bound Schottky defect in a  $2 \times 2 \times 2$   $\text{UO}_2$  supercell. Mechanistic steps correspond to those shown in Fig. 7. Errors are given as the standard deviation between committee member predictions.

| Mechanistic Step                 | This work              |                                    |                | Thompson and Wolverton [123] | Basak [4]                |
|----------------------------------|------------------------|------------------------------------|----------------|------------------------------|--------------------------|
|                                  | $\text{UO}_2$ MLP (eV) | $\text{UO}_2 + \text{Xe}$ MLP (eV) | DFT + $U$ (eV) | DFT + $U$ (eV)               | Empirical Potential (eV) |
| 1: U Hop                         | $4.67 \pm 0.46$        | $5.09 \pm 0.27$                    | 6.67           | 5.88                         | 6.72                     |
| 2: O Hop (1NN $\rightarrow$ 2NN) | $1.59 \pm 0.07$        | $1.71 \pm 0.11$                    | 2.15           | 1.77                         | 2.14                     |
| 3: O Hop (2NN $\rightarrow$ 3NN) | $1.32 \pm 0.05$        | $1.41 \pm 0.13$                    | 1.71           | 1.34                         | 1.96                     |

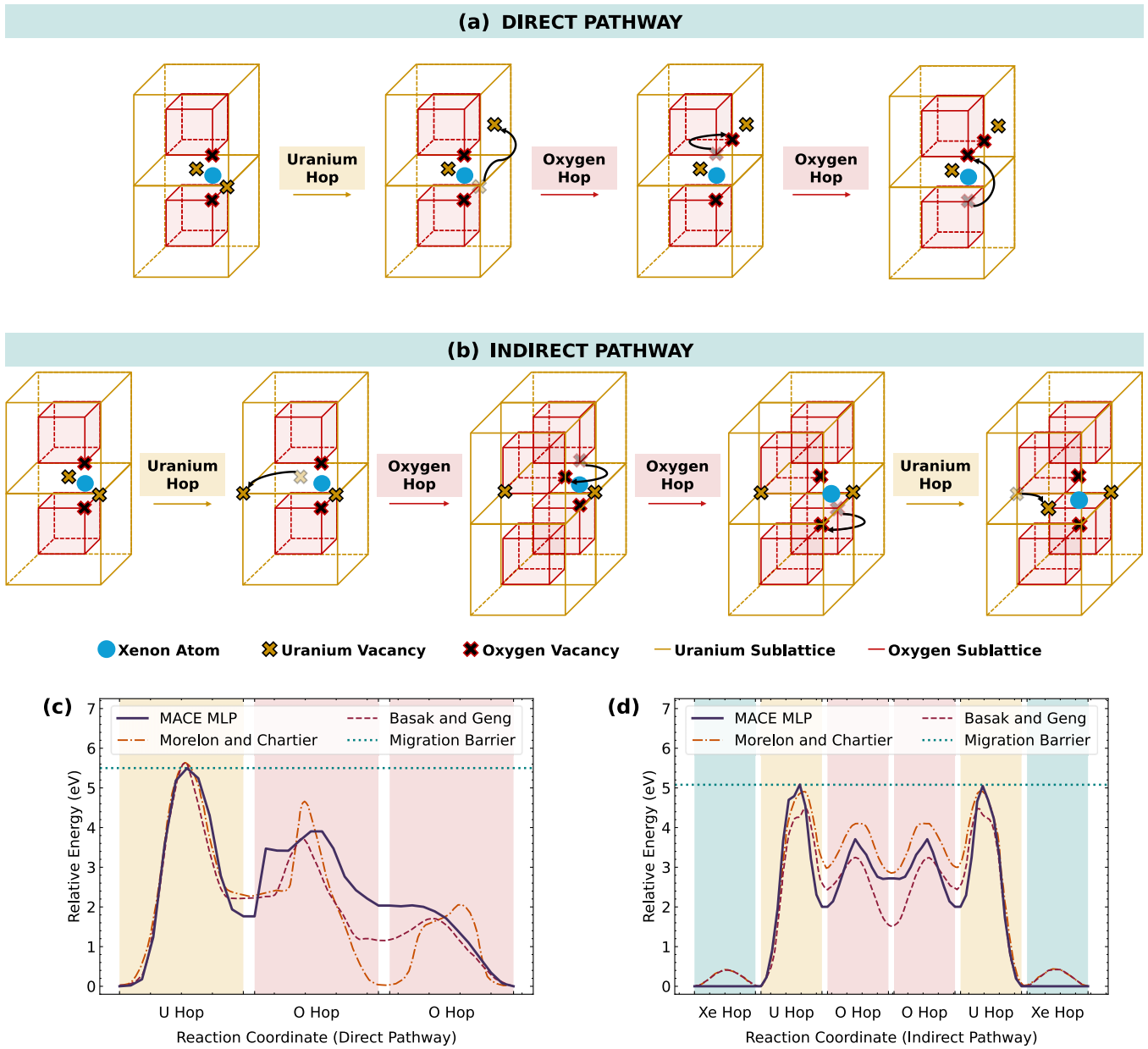


FIG. 8. Diffusion pathways for a xenon atom in the bulk  $\text{UO}_2$  lattice via the direct (a) and indirect mechanisms (b), differentiated by the temporary dissociation of the uranium vacancy in the indirect pathway. Corresponding energy profiles for these pathways (c) and (d) are shown relative to the lowest-energy configuration. Errors are given as the standard deviation between committee member predictions.

a local minimum in the minimum-energy pathway for the first oxygen migration. This corresponds to the migrating oxygen occupying an interstitial position furthest from neighboring oxygen atoms, suggesting that the pathway is strongly directed by the minimization of electrostatic repulsions.

Additionally, this migrating oxygen atom equilibrates to a nonlattice site in order to accommodate the xenon atom, as the initial uranium migration reduces the size of the trapping site. The xenon atom also equilibrates partially into that migrating oxygen's initial position, reducing its steric interaction with

TABLE VI. Migration barriers for a single xenon atom diffusing via a tetravacancy cluster mechanism. Values are reported for both the direct and indirect pathways.

|                  | MACE MLP (eV)<br>This Work, $\text{UO}_2 + \text{Xe}$ | DFT + $U$ (eV)<br>Andersson <i>et al.</i> [19] | Semiempirical Potentials (eV) [123]<br>Basak/Geng [4,121]      Morelon/Chartier [5,125] |     |
|------------------|-------------------------------------------------------|------------------------------------------------|-----------------------------------------------------------------------------------------|-----|
| Direct Pathway   | $5.50 \pm 0.07$                                       | 5.73                                           | 5.6                                                                                     | 5.6 |
| Indirect Pathway | $5.08 \pm 0.09$                                       | 5.25                                           | 4.5                                                                                     | 4.9 |

the lattice. The interstitial positioning of the oxygen atom accounts for the elevated energy of the tetravacancy defect cluster between the first and second oxygen migrations.

Similarly, with respect to the indirect pathway, the equilibration of the migrating oxygen to a non-lattice site after the first oxygen hop to accommodate the incorporated xenon leads the MACE MLP to predict a migration pathway intermediate with a higher energy than the Basak/Geng potential pair. According to simulations with the MACE MLP, the xenon does not occupy a uranium lattice site but rather equilibrates to the center of the tetravacancy defect cluster. However, Thompson and Wolverton report xenon occupation of those uranium vacancies. This offers an explanation for the differences in migration energetics observed between the empirical models and our MLP. Even so, we observe the same small local minimum in the energetic profiles of the uranium migrations as that found using the semiempirical potentials. This corresponds to the occupation of an interstitial site by the migrating uranium.

#### IV. CONCLUSIONS

The MACE equivariant message-passing neural network architecture was leveraged to train two MLPs for  $\text{UO}_2$  and  $\text{UO}_2$  with xenon inclusion. Training datasets were generated for this work using DFT +  $U$  calculations and query-by-committee active learning procedures, resulting in MLPs that reproduce DFT +  $U$ -level accuracy at a fraction of the computational cost. Robust energies and forces for systems containing hundreds of atoms can be obtained in seconds with MACE MLPs, whereas reference DFT +  $U$  calculations require intensive resources and sporadically successful techniques to coax systems into the ground state. Similar prediction errors for energies and forces were obtained for these MACE MLPs as have been reported for other MLPs, even though our potentials were trained with thousands fewer data points. This illustrates the efficacy of the equivariant features inherent to MACE model construction.

Our MACE MLP for  $\text{UO}_2$  outperforms existing semiempirical potentials and MLPs when calculating the thermal lattice expansion and temperature-dependent heat capacity of  $\text{UO}_2$ . While results for the elasticity tensor and bulk modulus showed increased uncertainty at temperatures above 2000 K, this uncertainty is also reflected in available datasets for benchmarking. Further experimental characterization of  $\text{UO}_2$  in this high-temperature regime will be necessary to definitively resolve disagreement among available models, particularly regarding the proposed premelting transition. Overall, the MACE MLP for  $\text{UO}_2$  is found to be a robust engine for MLP-MD simulations modeling the behavior of  $\text{UO}_2$  under in-reactor conditions.

We also report an MLP trained to model xenon inclusion in  $\text{UO}_2$  nuclear fuel. Xenon incorporation energies into a variety of trap sites found using the MACE MLP align with those reported in DFT +  $U$  reference datasets. This suggests that larger-scale MLP-MD models of xenon diffusion through the lattice will be governed by an accurate defect energy landscape. To demonstrate this, nudged elastic band simulations employing the MACE MLP were performed to investigate the energetics of xenon diffusion via a tetravacancy defect cluster. Migration barriers for both a direct and indirect

tetravacancy diffusion mechanism align with those reported in the literature, achieving accurate enthalpies for inclusion in diffusion models at modest computational cost. Beyond these quantitative results, qualitative characteristics of these pathways give further insight into xenon movement in bulk  $\text{UO}_2$ . These studies lay the groundwork for more extensive work modeling xenon bubble formation, equilibration, and diffusion through the  $\text{UO}_2$  lattice using MLP-MD methods. More complete modeling of xenon diffusion through the  $\text{UO}_2$  lattice will necessitate consideration of trap site charge states, the stoichiometry of  $\text{UO}_2$  in oxygen-deficient or oxygen-rich conditions, and entropy contributions [19,22,126–129].

Beyond the success of both MACE MLPs presented here in addressing pressing challenges in nuclear fuel performance modeling, this work demonstrates the advantage of capitalizing on existing training datasets to extend MLP usage beyond its originally imagined application space. By supplementing the original  $\text{UO}_2$  training dataset with additional xenon-bearing configurations, the scope of questions that can be answered with the resulting model widens greatly.

Future work will involve the use of MLPs to generate data for fuel performance codes. Extension of the xenon potential to include krypton will also facilitate more detailed fission gas diffusion modeling.

Beyond the employment of these MLPs to improve fission gas diffusion models with mechanistic insights, addressing shortcomings in the reference calculations themselves will maximize the benefits of the computational speedup achieved by employing machine-learned potentials. While MLP training greatly reduces the computational cost of extending DFT +  $U$ -level accuracy to larger length scales, it is even more desirable to do so with even higher-quality training data. By making improvements to the type of data used in MLP training, new frontiers in fuel performance modeling can be achieved by extending insights from detailed first-principles calculations to grain- and pellet-scale simulations.

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## DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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