GDR IAMAT 2022

Machine-learning aided calculation of atomic-scale properties in chemically disordered (U, Pu)O_2 fuels

DE LA RECHERCHE À L'INDUSTRIE

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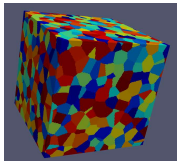
31/05/2022

Institut de recherche sur les systèmes nucléaires pour la production d'énergie bas carbone

LM2C & LI3A COLLABORATION

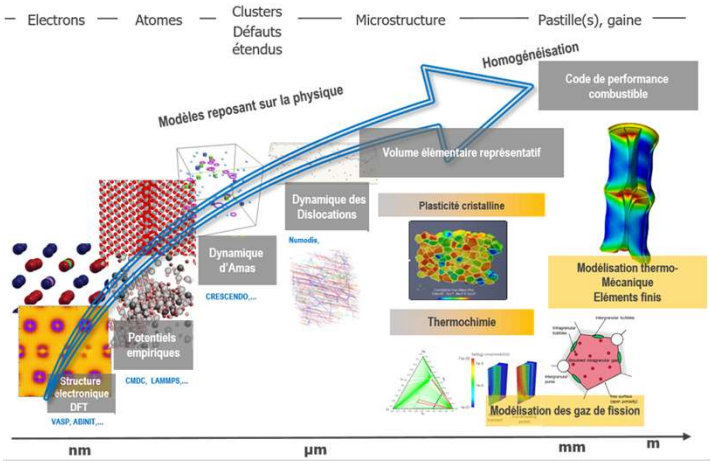
LM2C - Laboratoire de Modélisation Multi-échelle des Combustibles

- Study and modeling of physical phenomena to determine their physico-chemical properties, with the aid of models and scientific computing tools of the **PLEIADES [1]** platform.
- **Multiscale approach** - use of a variety of models at different levels of spatial (from nanometers to meters) and time scales (from picoseconds to years) to study a given system **[2]**.



Applications:

- PWR/RNT fuels at all scales
- All stages of fuel life (manufacturing, in-reactor operation, accidents, storage, reprocessing)
- Battery materials, corium, molten salts



LI3A - Laboratoire Intelligence Artificielle et Apprentissage Automatique

ADVANCED ARTIFICIAL INTELLIGENCE

- Frugal AI (Transfer Learning, Few Shot learning, generative models, simulations)
- Distributed AI
- Adaptive AI (Online L., Incremental L., …) [Streamer]
- Knowledge-based AI and reasoning [ExpressIF]
- Hybrid AI (rules learning, rules and data fusion) [PACT]

SIGNAL PROCESSING

- Digital signals intelligence
  - Audio analysis
  - Vibration & movement analysis
  - Spectroscopy and spectroimaging
- Point processes analysis

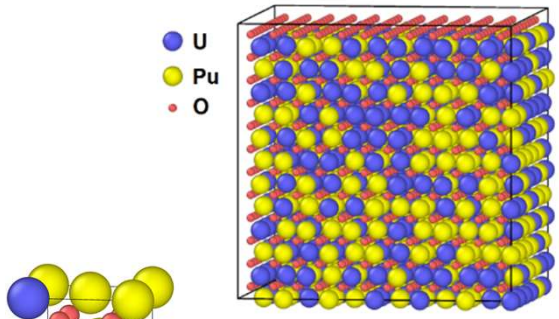
CYBER-PHYSICAL SECURITY

- AI for critical systems monitoring: resilience and security
  - Intrusion detection systems
  - Forensics
- Trustworthy AI
  - Adversarial robustness
  - Data and model privacy for AI

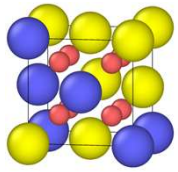
1. URANIUM-PLUTONIUM MIXED OXIDES

$(U, Pu)O_2$, MOX fuel

- Used in French pressurized water reactors (PWRs) and planned to be used in generation IV fast neutron reactors **[3]**.
- One of the objectives is the recycling of plutonium produced during the exploitation of UO_2 fuel in the reactor.
- This presents both economical and environmental benefits, which is why the study of this type of fuel is of much importance **[4]**, **[5]**.
- However, it is crucial to **predict the behavior of nuclear fuels** during their in-reactor operation - this allows to use their energy potential as efficiently as possible, within predefined **safety margins**.
- This task can be accomplished with **multi-scale approach**: the following work concentrates on the study of the properties of MOX fuel calculated at the **atomic scale**.



Supercell built from 6x6x6 primitive cells – 2592 atoms in total.



Single primitive cell of fluorite type.

1. URANIUM-PLUTONIUM MIXED OXIDES

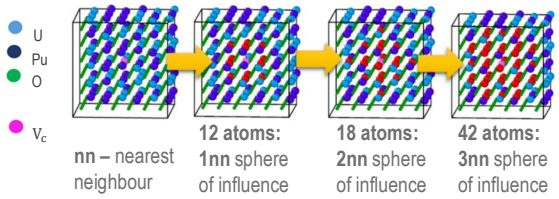
Chemical disorder of MOX fuel

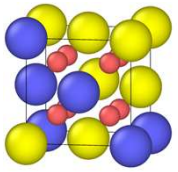
- MOX fuel has a fluorite-type crystallographic structure. It is a face-centered cubic (fcc) lattice, combined with a simple cubic sublattice occupied by oxygen atoms.
- $(\mathrm{U,Pu})\mathrm{O}_2$ is a **chemically disordered** compound, where plutonium and uranium are distributed on the same crystal lattice.

As a consequence, there are **many possible configurations** influencing the properties that depend on the size of the configuration space … and **high computational cost!** **[6]**, **[7]**, **[8]**.

Example - calculation of the formation energy of defects E^f_{BSD} with interatomic potential:

- Ensemble of **12** atoms (0.384 nm) - $\mathbf{14}\ \boldsymbol{d}$
- Ensemble of **18** atoms (0.543 nm) - $\mathbf{910}\ \boldsymbol{d}$
- Ensemble of **42** atoms (0.768 nm) - $\mathbf{42 * 10^6}\ \boldsymbol{y}$
- …
- …supercell made of **2592 atoms.**





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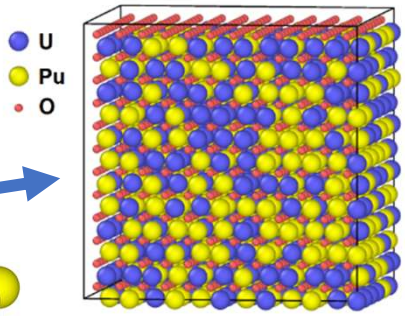

Supercell built from 6x6x6 primitive cells – 2592 atoms in total.

2. Objective of the work

2. OBJECTIVE OF THE WORK

Generative Machine Learning methods in atomic-scale modeling

The objective of this work is to design a new generative ML tool, coupled with atomic-scale modeling methods, that is able to go beyond the limitations of classical approaches – to be able to study the **properties of chemically disordered** compounds like uranium-plutonium mixed-oxide nuclear fuels.

One of such properties is the **concentration of thermal defects.**

2. OBJECTIVE OF THE WORK

Formalization of the objective

Calculation of the concentration of thermal defects - knowing such property is important to model the microstructure evolution of the MOX fuel. Just say that it is an ensemble average and we need to calculate all energies for that

$$C_{\mathrm{d}}(T) = \left\langle \exp\left(-\frac{E_{\mathrm{d}}^{\mathrm{form}}(\chi_i)}{k_{\mathrm{B}}T}\right)\right\rangle = \mathbf{E}_{E_{\mathrm{d}}^{\mathrm{form}}\sim p(E_{\mathrm{d}}^{\mathrm{form}})}\left[\exp\left(-\frac{E_{\mathrm{d}}^{\mathrm{form}}(\chi_i)}{k_{\mathrm{B}}T}\right)\right]$$

- C_{d} - concentration of defect d
- χ_i - atomic configuration
- $E_{\mathrm{d}}^{\mathrm{form}}(\chi_i)$ - formation energy

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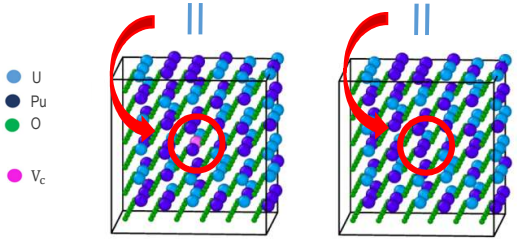
- C_{d} - concentration of defect d
- χ_i - atomic configuration
- $E_{\mathrm{d}}^{\mathrm{form}}(\chi_i)$ - formation energy

Formation energy:

$$E_{\mathrm{d}}^{\mathrm{form}}(\chi_i) \overset{*}{\approx} E_{BSD}(x_c) - E(x_c)$$



*simplified formula

$$X = \left(E_{\mathrm{d}}^{\mathrm{form},1}, E_{\mathrm{d}}^{\mathrm{form},2}, E_{\mathrm{d}}^{\mathrm{form},3}, \dots, E_{\mathrm{d}}^{\mathrm{form},N} \right)$$

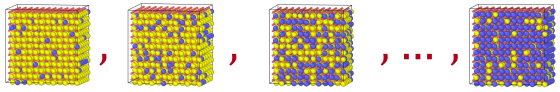
2. OBJECTIVE OF THE WORK

Objective: Calculation of the concentration of defects

$$C_{\mathrm{d}}(T) = \left\langle \exp\left(-\frac{E_{\mathrm{d}}^{\mathrm{form}}(\chi_i)}{k_{\mathrm{B}}T} \right) \right\rangle$$

Challenge: the size of configuration space X

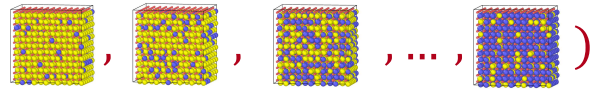
$$X = \left(E_{\mathrm{d}}^{\mathrm{form},1}, E_{\mathrm{d}}^{\mathrm{form},2}, E_{\mathrm{d}}^{\mathrm{form},3}, \dots, E_{\mathrm{d}}^{\mathrm{form},N} \right)$$



2. OBJECTIVE OF THE WORK

Challenge: how to obtain a database of $E_{\mathrm{d}}^{\mathrm{form}}$ with lower cost?

$$X = \left(E_{\mathrm{d}}^{\mathrm{form},1}, E_{\mathrm{d}}^{\mathrm{for},2}, E_{\mathrm{d}}^{\mathrm{fo},3}, \ldots, E_{\mathrm{d}}^{\mathrm{for},N} \right)$$



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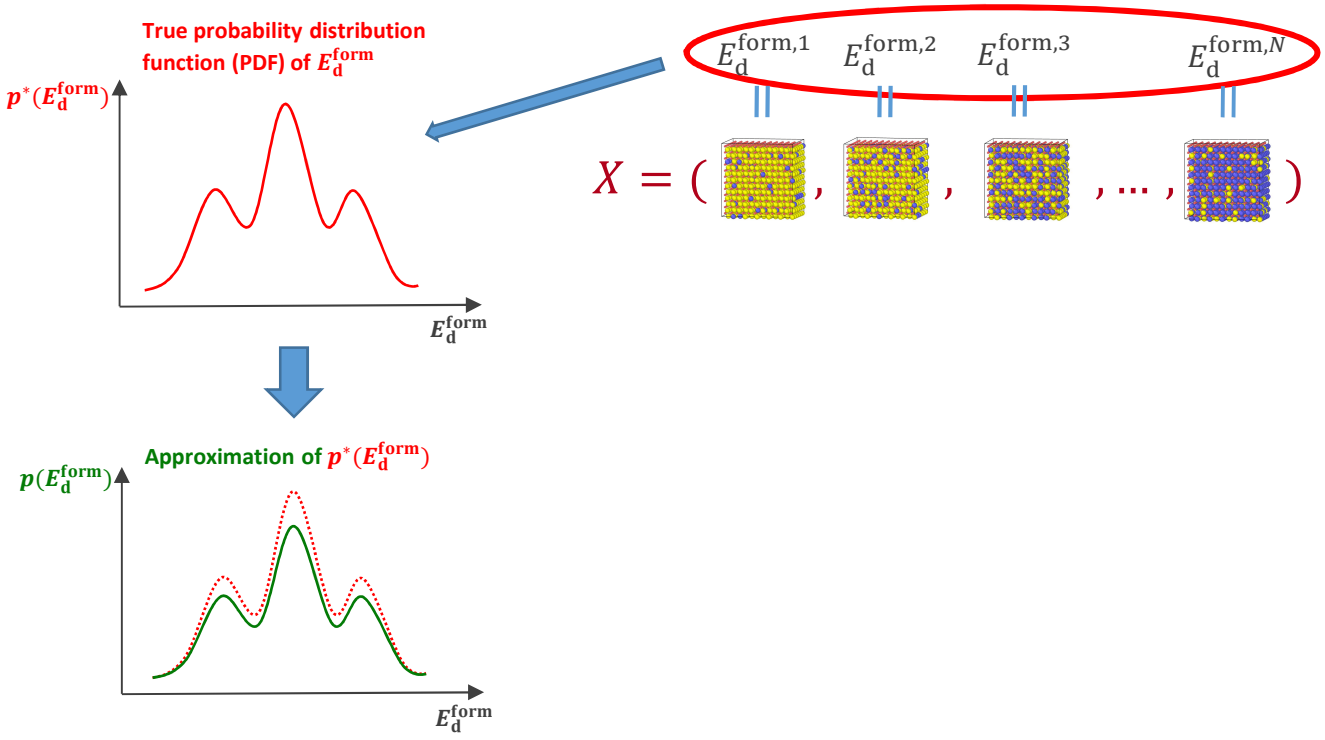

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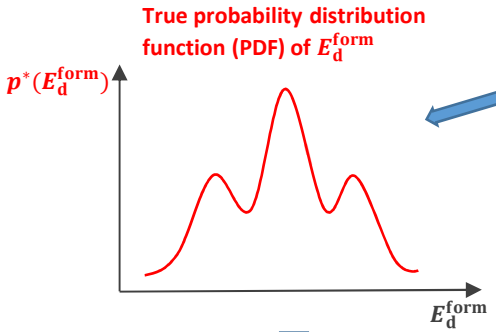
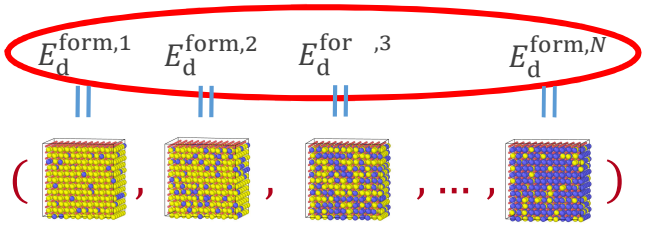
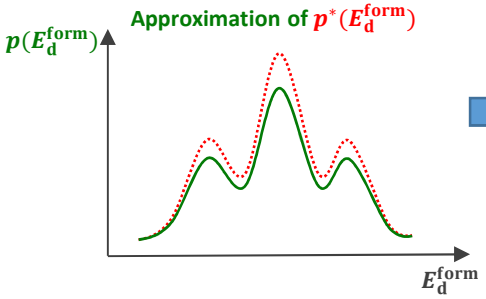




$p^*(E_{\mathrm{d}}^{\mathrm{form}})$ – sampling is difficult and computationally expensive

$p(E_{\mathrm{d}}^{\mathrm{form}})$ – defined with a tractable probability distribution that is simple and quick to sample



Monte Carlo (MC) approximation of $C_d(T)$

$$C_{\mathrm{d}}(T) = \mathbf{E}_{E_{\mathrm{d}}^{\mathrm{form}} \sim p(E_{\mathrm{d}}^{\mathrm{form}})}\left[\exp\left(-\frac{E_{\mathrm{d}}^{\mathrm{form}}}{k_{\mathrm{B}}T}\right)\right]$$

The objective is to obtain an **approximation of $p^*(E_{\mathrm{d}}^{\mathrm{form}})$** with a **minimal amount** of configurations.

2. OBJECTIVE OF THE WORK

Chosing a method for approximation of $p^*\left(E_{\mathrm{d}}^{\mathrm{form}}\right)$: Data vs energy evaluation

1. **When data is available**
   - Simple approach with a NN
2. **When energy evaluation is quick**
   - Markov Chain Monte Carlo (MCMC)
   - Active Learning

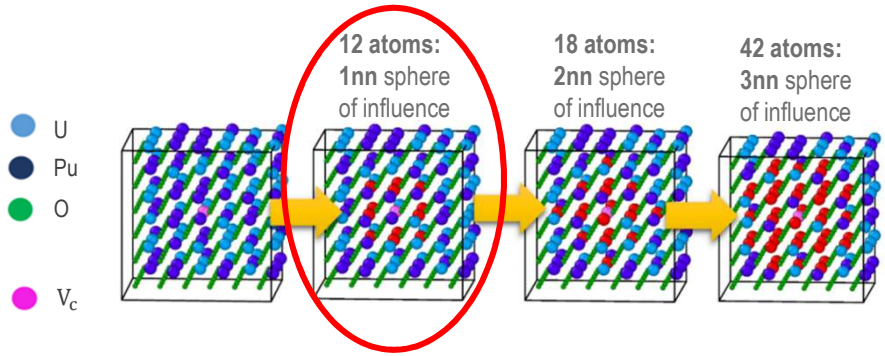
Case of MOX fuel

1. **Very little data is available**
2. **Each energy evaluation is computationaly expensive**

2. OBJECTIVE OF THE WORK

Semi-supervised approach to chemical disorder

Basing on the observations from previous studies, and on the first results of this work, we could recognize **the most influencial factor** for E_d^{form} calculation **[8]**, **[9]**.

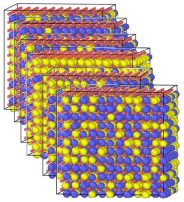


- Since there is very few data available and generating new data is expensive, the idea was to describe the data in a way, that will **accelerate the learning process.**
- The goal was to be able to build such training dataset that is **small**, but contain **enough information** to obtain **accurate approximation** of $p^*\left(E_d^{form}\right)$

2. OBJECTIVE OF THE WORK

Semi-supervised approach to chemical disorder

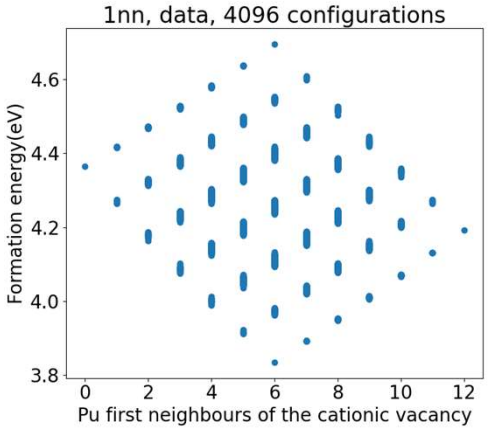
Atomic configurations



Description of configurations, based on the 1nn atomic composition

$$\begin{bmatrix} N_{Pu}^{c} \\ E_{\mathrm{d}}^{\mathrm{form}} \end{bmatrix}$$

- No. Pu near cationic vacancy
- Formation energy

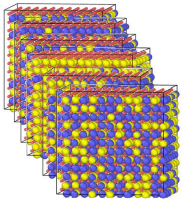


We described atomic configurations with a simplistic descriptor that consist of the number of Pu atoms near cationic vacancy N_{Pu}^{c} and formation energy $E_{\mathrm{d}}^{\mathrm{form}}$.

2. OBJECTIVE OF THE WORK

Semi-supervised approach to chemical disorder

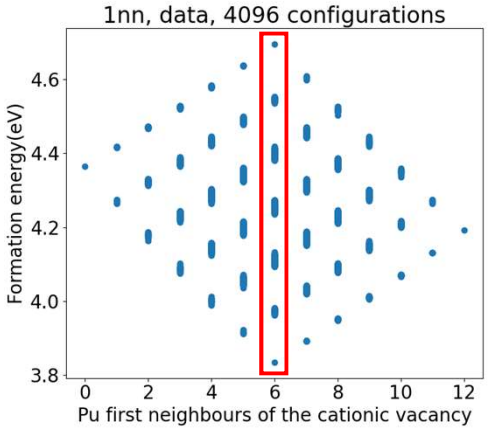
Atomic configurations



Description of configurations, based on the 1nn atomic composition

$\begin{bmatrix} N_{Pu}^{c} \\ E_{\mathrm{d}}^{\mathrm{form}} \end{bmatrix}$ - No. Pu near cationic vacancy

- Formation energy

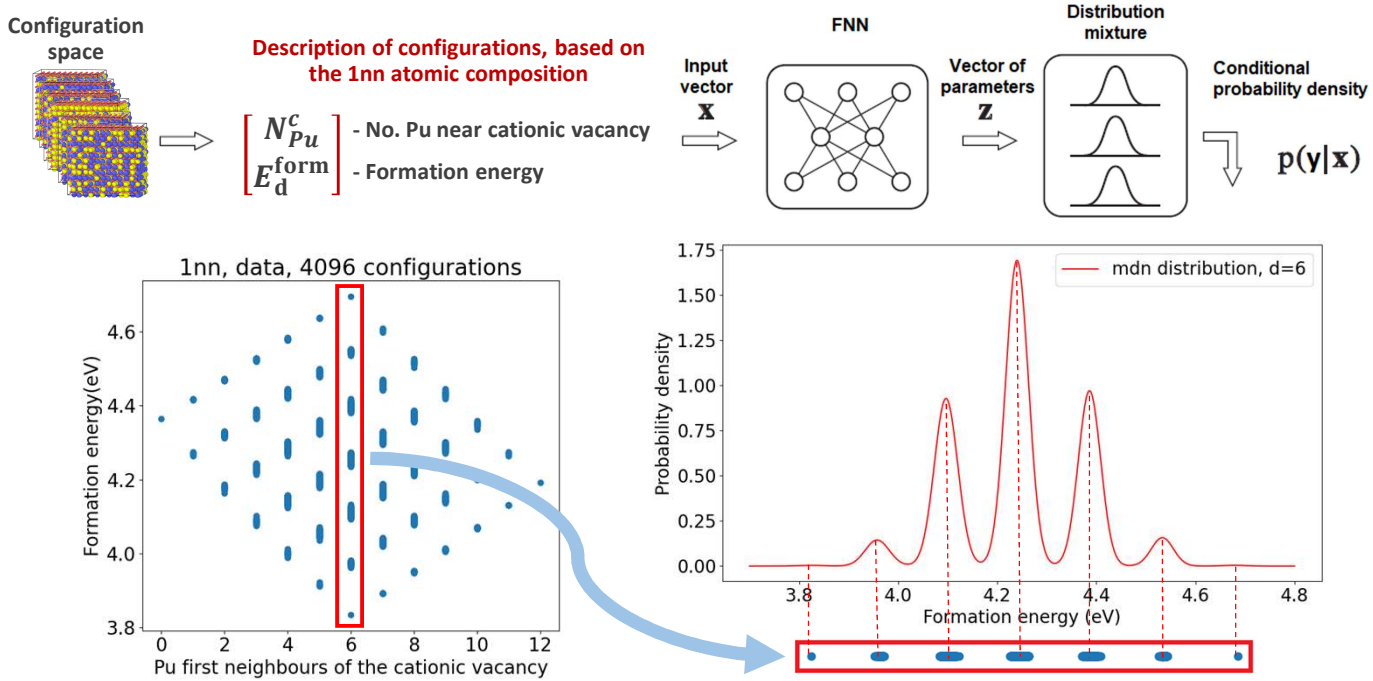


We described atomic configurations with a simplistic descriptor that consist of the number of Pu atoms near cationic vacancy N_{Pu}^{c} and formation energy $E_{\mathrm{d}}^{\mathrm{form}}$.

2. OBJECTIVE OF THE WORK

Mixture Density Network (MDN)

- **Simple composition** of the output distribution (Gaussian mixture)
- Possibly **small amount of data** required for training **[9]**, **[10]**.



3. Results of the MDN approach

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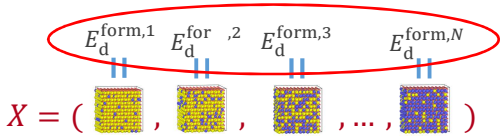
3. RESULTS OF THE MDN APPROACH

Treatment of chemical disorder in MOX fuel with MDN method

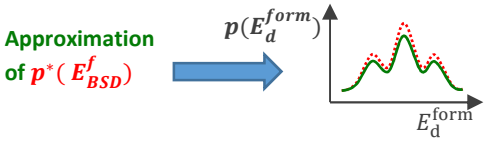
- **Objective:** Calculation of the concentration of defects $C_d(T)$ - $E_{\mathrm{d}}^{\mathrm{form}}$ database of configurations is required.

$$C_{\mathrm{d}}(T) = \left\langle \exp\left(-\frac{E_{\mathrm{d}}^{\mathrm{form}}(\chi_i)}{k_{\mathrm{B}}T}\right)\right\rangle$$

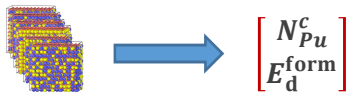
- **Challenge:** The size of the configuration space *X* - **high computational cost** of $E_{\mathrm{d}}^{\mathrm{form}}$ for every configuration.



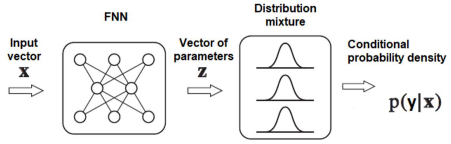
- **Approach:** Approximation of the true probability distribution $p^*(E_d^{form})$ – to obtain the database of $E_{\mathrm{d}}^{\mathrm{form}}$ with **lower cost**.



- **Description of data:** to build a dataset that accelerates the training process and limits the number of configurations needed.



- **ML model:** Mixture Density Network – **simple composition** of the output distribution, with possibly **small amount of data** required for training.

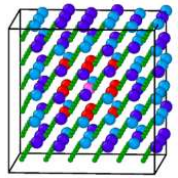


3. RESULTS OF THE MDN APPROACH

Objective: Calculation of the concentration of defects C_{BSD}

Results for the **1nn sphere of influence**

1nn sphere of influence



Calculation of $C_{BSD}(T)$ from data

No. configurations: **4096**

Interatomic potential: **5 min / conf.**

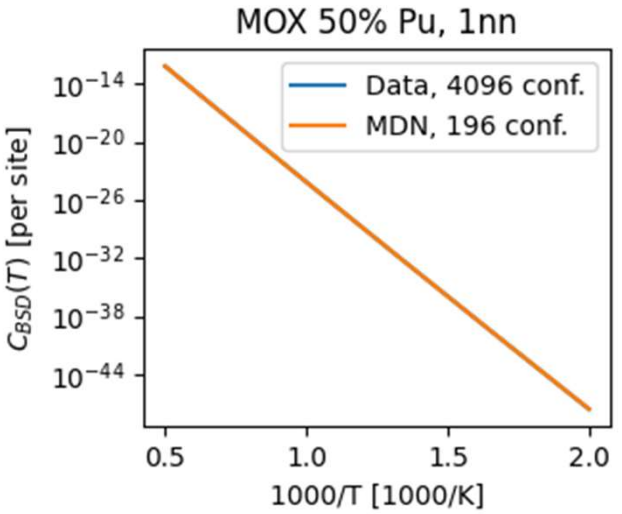
CPU time: **~341.3h**

Calculation of $C_{BSD}(T)$ with MDN

No. configurations: **~200**

Interatomic potential: **5 min / conf.**

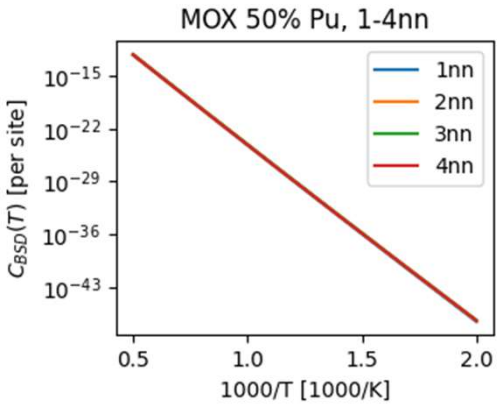
CPU time: **~16.3h**



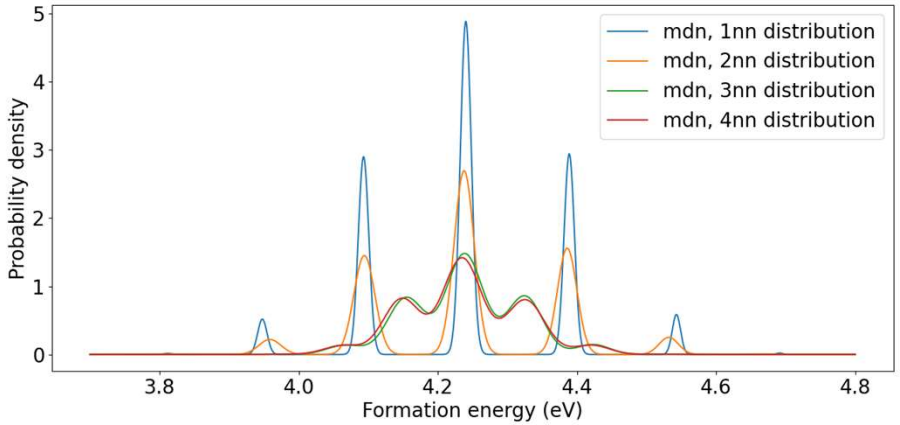
3. RESULTS OF THE MDN APPROACH

Effect of local composition on the C_{BSD} calculation

- Thanks to the fact that MDN requires very few data, it can be used for the verification of the range of influence of **local composition** on the C_{BSD} calculation.
- Here, 4 MDN models were trained on different datasets, for the different spheres of influence around the defect.



MDN models evaluated for the descriptor d=6 (6 Pu 1nn atoms)



- ✓ Spheres beyond **3nn** have **negligible effect** on E^f_{BSD} calculation.
- ✓ MDN requires the same amount of training data, **independently of the size** of configuration space.

3. RESULTS OF THE MDN APPROACH

Conclusions & Limitations

- ✓ MDN can serve as a robust **semi-supervised** method to evaluate **local-atomic dependend** properties.
- ✓ MDN requires a relatively **small amount of samples** for training (~200).
- ✓ MDN is **independent** of the size of the configuration space.
- ❑ Training database of the MDN model was builed based on the observed influence of the 1nn.
- ❑ Calculations were done with the use of CRG interatomic potential **[11]**.
- ❑ The influence of **local order** of atomic configurations was assumed to be negligible.

Future perspectives

- ➢ MDN can be used **readily** with other types of potentials.
- ➢ It can be used to investigate **different types of defects .**
- ➢ It is possible to train MDN directly on **DFT data.**

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MERCI DE VOTRE ATTENTION

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4. FUTURE PERSPECTIVES

MDN approach

- There are many possible uses of the MDN approach:
  - MDN can be used **readily** with other types of potentials
  - It can be used to investigate **other types of defects**
  - It is possible to train MDN directly on **DFT data**
- However, the MDN approach is limited by the assumptions taken.
- The biggest of them is the assumption about the **uniform distribution** of weights $w(x_c)$
- This is restricting the possible uses of MDN in other tasks where the influence of **local order** of atomic configurations is not negligible.

Chosing a method for approximation of $p^*\left(E_{\mathrm{d}}^{\mathrm{form}}\right)$: Data vs energy evaluation

1. **When data is available**
   - Simple approach with a NN
2. **When energy evaluation is quick**
   - Markov Chain Monte Carlo (MCMC)
   - Active Learning

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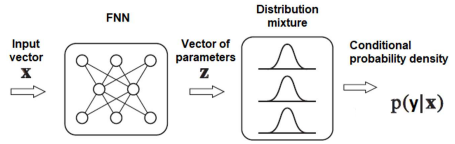
➢ Semi-supervised approach with MDN



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