International Atomic and Molecular Code Centre Network on Simulation of Plasma-Material Interaction Experiments

Summary Report of the 4th Biennial Technical Meeting

IAEA Headquarters, Vienna, Austria

29–31 July 2015

Prepared by

H.-K. Chung, Y. Ferro, G. Csányi, P. Krstic, B. D. Wirth, U. von Toussaint and B. J. Braams

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Abstract

The International Code Centre Network (CCN) is a group of experts developing codes and models for atomic, molecular and plasma-surface interaction data relevant to fusion applications. Variable subsets of the group are brought together by the IAEA Atomic and Molecular Data Unit in order to discuss computational and scientific issues associated with code developments. The 4th technical meeting was held 29–31 July 2015 and nine experts in the field of simulations of plasma-material interaction (PMI) experiments were invited to discuss the current status and future directions of the uncertainty quantification (UQ) activities for theoretical PMI data with special interest in problems of hydrogen retention in wall materials. Uncertainties associated with interatomic potentials, density-functional theory (DFT) simulations, molecular dynamics (MD) simulations, kinetic Monte Carlo (KMC) simulations and rate equation simulations for hydrogen retention and migration in fusion material were broadly discussed.

March 2016

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1. Introduction

Computational modeling of plasma behavior in fusion devices plays a critical role in developing a fusion reactor for energy use. To represent physical reality this modeling requires comprehensive data for very many atomic and molecular (A+M) and particle-surface interaction (PSI) or plasma-material interaction (PMI) processes. Only a limited amount of data is accessible to experiment or is found in the literature or in prepared databases, and computations are essential to generate adequate data for plasma simulations. The international Code Centres Network (CCN) is coordinated by the IAEA A+M Data Unit in order to address this issue, gather and provide access to computational codes and help to provide calculated A+M+PSI/PMI data relevant for modelers in fusion and plasma sciences. Starting with a consultants' meeting on "Establishment of Atomic and Molecular Computer Code Network" in 2005, the Unit has organized a series of meetings of code developers in the CCN to address code related issues including the accessibility of physics codes.

Recently, the fidelity of calculated data has been an issue of importance to both the data user community and the data producing community. Experimental data are almost always reported together with estimated uncertainties of measurement while theoretical data are seldom provided with such estimates. The use of data without the accuracy information brings in uncertainties to an overall result of global modeling and reduces the fidelity of the result toward applications.

In order to discuss the issue, the Unit organized the 3rd technical meeting on the international Code Centres Network (CCN3) held at IAEA headquarters in May 6-8 in 2013 where experts of theoretical A+M physics codes discussed general strategies on how to assess the accuracy of calculated data. Upon recommendation of the CCN3 meeting, the Unit subsequently organized a joint workshop at ITAMP (Institute of Theoretical Atomic, Molecular and Optical Physics, Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, USA) to address issues of uncertainty assessment for theoretical A+M scattering data and develop guidelines on how to estimate uncertainty of theoretical A+M Data (see https://www-amdis.iaea.org/DCN/Evaluation/.)

These activities are in accordance with an increasing effort in the engineering and physical sciences communities to establish mathematical and statistical foundations of Verification, Validation and Uncertainty Quantification (VVUQ) of complex models and simulations as reported by National Research Council (USA) (http://www.nap.edu/catalog/13395/assessing-the-reliability-of-complex-models-mathematical-and-statistical-foundations). The Unit participated in the BIPM (Bureau International des Poids et Mesures) Workshop on Measurement Uncertainty in order to address the UQ activities in the theoretical A+M sciences.

Following these efforts on A+M data the Unit wants to expand its UQ activities to theoretical PMI data. The issue has been raised at the Joint ICTP-IAEA Conference on Models and Data for Plasma-Material Interaction in Fusion Devices in November 2014, and also at the 21st International Conference on Plasma Surface Interaction in Controlled Fusion Devices (the PSI meeting) in Kanazawa, Japan in May 2014. With positive responses from the PMI code community, the Unit invited 9 experts to the 4th TM of the International Code Centre Network (CCN) on Simulations of Plasma-Material Interaction Experiments to discuss the current status and future directions of UQ activities for theoretical PMI data. To make this meeting relevant to fusion, discussion topics were focused on PMI related to hydrogen retention. Invited experts discussed uncertainties with interatomic potentials, density-functional theory (DFT) simulations, molecular dynamics (MD) simulations, kinetic Monte Carlo (KMC) simulations and rate equation simulations for hydrogen retention and migration in fusion materials.

This report contains an introduction in Section 1, proceedings of the meeting in Section 2, and discussions and recommendations in Section 3. The list of participants is provided in Appendix 1 and the meeting agenda in Appendix 2. The abstracts of presentations are provided in Appendix 3.

2. Proceedings

The CCN meeting was opened by the new head of Nuclear Data Section, A. Koning welcoming all participants. He emphasized the importance of the uncertainty estimates in data evaluation and suggested future collaboration between atomic and nuclear communities on various problems. B. J. Braams (Head, A+M Data Unit) and the scientific secretary H.-K. Chung (Atomic physicist, A+M Data Unit) welcomed participants and introductions of participants followed. Five rapporteurs (Y. Ferro, G. Csányi, P. Krstic, B. Wirth, U. von Toussaint) were designated to provide a summary of discussion sessions. The agenda was adopted (Appendix 2). The abstract of each presentation is attached in the Appendix 3 and presentations are available at Unit's home page https://www-amdis.iaea.org/CCN/Meetings4/.

Roberto Capote of Nuclear Data Section introduced the activities of uncertainty quantification applied to nuclear data and described the Unified Monte Carlo method, a method combining experimental and modelling uncertainties for data evaluation.

Hyun-Kyung Chung of Nuclear Data Section gave an overview of activities on uncertainty quantification of atomic and molecular data that the Unit has coordinated in the last 4 years.

Udo von Toussaint of Max-Planck-Institut für Plasmaphysik, Garching, Germany, described uncertainty quantification for plasma and plasma-wall interaction models in the community, the approaches, sensitivity analysis and emulators as well as examples.

Naohiko Otsuka of Nuclear Data Section introduced the concept of uncertainties and their quantification applied to experimental nuclear data.

Daiji Kato of National Institute for Fusion Science, Japan, presented Density Functional Theory calculations and their uncertainties of vacancy formation energies and interaction energies with hydrogen atoms in tungsten.

Yves Ferro, Aix-Marseille Université, France, presented the solution of the electronic structure of an atomic system by Density Functional Theory calculations and classified the sources of uncertainty from physical approximations or technical (or numerical) parameters or the lack of understanding of physics.

Bas Braams of Nuclear Data Section described the Bayesian method as a potential validation and uncertainty quantification method applied to atomic and molecular data.

Gábor Csányi of University of Cambridge, United Kingdom, described the function fitting method using basis functions in order to build a good interatomic potential with uncertainties and presented the method applied for tungsten of interest to fusion applications.

Takuji Oda of Seoul National University, Republic of Korea, discussed uncertainties in modeling of diffusion of hydrogen interacting with traps in bcc metals with emphasis on how to quantify the uncertainties of Density Functional Theory simulations and uncertainty propagation to the Kinetic Monte Carlo or Rate equation simulations.

Kazuhito Ohsawa of Kyushu University, Japan, reviewed simulation methods and results, their uncertainties of stable hydrogen configuration trapped in a tungsten vacancy.

Predrag Krstic of Institute for Advanced Computational Science, Stony Brook University, USA, discussed physical and computational aspects of quantum or classical molecular approaches, how to make choices, how to accelerate the calculation, what answers one should expect from each approach.

Duc Nguyen-Manh of Culham Centre for Fusion Energy, United Kingdom presented multi-scale approach based on first-principles calculations and reported on a systematic assessment of uncertainty quantification at different multi-scale levels of energetic and kinetic properties of nano-vacancy in tungsten and tungsten alloys.

Brian D. Wirth of University of Tennessee, Knoxville, USA gave an overview of multiscale materials modeling approach and a summary of observations from large-scale MD simulations on helium behaviour in tungsten. Also presented was an approach using Bayesian analysis for a global sensitivity analysis.

3. Discussions

Discussion sessions in this meeting were primarily concerned with applications to fusion problems such as hydrogen and helium retention and transport in fusion materials such as tungsten. Discussion sessions on uncertainty assessment were led by Prof Ferro for electronic structure calculations, Prof Csányi for interatomic potentials, Prof Krstic for molecular dynamics simulations and Prof Wirth for multiscale materials modeling.

Uncertainty Quantification of Electronic Structures Calculations

The first discussion session was devoted to the topic of electronic structures calculations, mainly density functional theory (DFT) calculations and possibly more elaborate methods than DFT. Uncertainty assessment for DFT calculations for solids immediately runs into problems of model uncertainty. The "theory" in DFT is not constructive — it might have been called a heuristic — and it does not provide a path towards a converged solution of the many-electron problem. For small molecules the results of DFT calculations, for specific functionals, can be compared with presumably more accurate coupled-cluster calculations, but for solids that comparison does not appear to be feasible in general.

To a large extent, therefore, uncertainty assessment for DFT calculations involves the comparison of calculations using different functionals. It was proposed to establish a set of reference data to evaluate uncertainties of DFT calculations. The idea of having a set of different results including single-, di-, vacancies, interstitial and so forth is proposed in order to have an idea of the uncertainty. Prof Oda demonstrated the UQ approach by performing calculations using different pseudo-potentials, LDA, GGA, hybrid-GGA and meta-GGA functionals. A consistent set of data should be carefully produced with the same numerical criterion and functional.

For many fusion applications a molecular dynamics (MD) model employing an accurate potential energy surface (PES) contains essentially all the physics. (Quantum effects on the nuclear motion appear to be unimportant above about 200 K even for hydrogen, and effects of electronic excited states are only of interest for the initial stage of a high-energy particle impact.) However, the uncertainty assessment of an MD simulations must involve then an uncertainty assesment of the PES, which is a step beyond the uncertainty assessment of the underlying electronic structure (generally DFT) calculations. Based on other methods (continuum-like MRE, KMC), it is possible to identify the missing mechanisms that can be computed by DFT and refine against experimental results. These new results can in the end be used to improve MD potential.

Currently there are enough DFT calculations to build realistic potentials, particularly for tungsten and hydrogen in solid state. A large set of results including surface properties has recently been calculated. These data have now to be processed to establish realistic potentials used by larger scale simulations. However, it is not clear how one can relate uncertainties from DFT results in the range of 0.1 to 0.2 eV to the development of MD potentials.

More elaborate approaches beyond DFT do exist, however, they are not ready to be used with the systems of interest for fusion applications. Such approaches include MP2 scheme developed in the CRYSCOR code with Gaussian Type Function (local basis set) and MP2 and CCSD(T) in VASP (PW basis sets). It was concluded that it is currently not practical to go beyond DFT for realistic solids.

Uncertainty Quantification of Interatomic Potentials

From electronic structure point of view, the use of current bcc-W empirical potentials (such as Finnis-Sinclair or Tersoff-like bond-order potentials) for large scale MD and KMC simulations contains many important uncertainties due to the fact that tungsten is a transition metal located in the middle of periodic table of elements (as well as Cr and Mo). The shape of its electronic density of states (DOS) is characterized by a deep minimum at the Fermi energy and this feature cannot be reproduced by second-moment approximation from which Finnis-Sinclair potentials were constructed. The origin of uncertainty comes from angular dependence of d-orbitals in transition metals that require higher moments of DOS at least to the fourth moment approximation in order to reproduce the minimum within the DOS. The Tersoff-like bond-order potentials did include angular character of bonding but its functional form is valid only for sp-valent systems such as Si, C or SiC. The angular dependence for interatomic bonding between the d-electron orbitals had a completely different functional form this leads to uncertainty in using this potential for the tungsten case.

A classic way to get information on the reliability of empirical interatomic potentials is to construct a family of potentials by varying some of the parameters, and carrying out a large number of independent simulations to target key observables, and thus get information on the P (observable parameters) function. The idea is that observables that are robust in a wider region of parameter space are deemed more reliable. In some sense this is a test not of the potential, but of dependence of the observable on the detail of the potential, so it is a UQ of the modelling approach, rather than the potential. An example to this approach can be found in a paper by Frederiksen et al. (PRL, Vol. 93, No. 16, 2004, p. 165501.) Once the sensitivities of various observables are determined, these could be taken forward into higher level of UQ via noise parameters.

Another problem with the above approach is that the empirical potentials even with variable parameters only cover a very small manifold of possible interatomic potentials, and their true error is due to the fact that the real potential is not on that manifold. For example this is the case when the parameters of a Finnis-Sinclair type potential are varied, which altogether miss important effects as discussed above.

A couple of interesting directions are offered by having fully flexible many-body potentials (machine learned potentials).

1) The Gaussian process (GP) based potentials offer an intrinsic error bar on the potential energy and derived quantities (forces, stresses), but it is difficult to convert this into an uncertainty for a complex observable that is the result of a long simulation - it's more of a diagnostic tool that can be used to invalidate a simulation if it encounters configurations that are far from the training data.

2) A GP framework offers the possibility of a systematically improvable potential, with more training data (and higher evaluation cost) leading to higher accuracy. This is mathematically true in principle, in the scaling regime, but completely untested in reality. (But have seen it in rudimentary examples e.g. temperature-dependent phonon spectra of graphene as a function of potential cost)

3) The ability to generate a general potential that is nevertheless smooth gives rise to the following possibility. One could take an empirical potential that is to be used for a simulation and *add* to it a small perturbation, i.e. a Gaussian process with random coefficients, and do the long complex simulation with the sum. Sampling the GP repeatedly gives potentials that are not on a small dimensional manifold, and therefore go a long way to rectify the problems of classic approach. The big advantage is that this can be done without any accurate training data whatsoever, it is purely a test of the empirical potential manifold. However, the results will depend on the structure of the noise that is added. This is not a systematic approach yet.

Uncertainty Quantification within Molecular Dynamics Simulations

Classical molecular dynamics (CMD) is only as good as the interatomic multibody potential model used. Creation, calibration and validation of these multibody, semi-empirical potentials is an important computational/experimental challenge which was extensively discussed at the meeting comparing Finnis-Sinclair and Tersoff-like potentials for W. For simulations of metallic systems, the Embedded Atom Method is a relatively accurate technique that is related to the second moment of tight binding theory. Validated EAM potentials can be very accurate for a variety of mechanical properties of metallic systems, but tend to be complex and not easily mixed as it is generally necessary to specifically parameterize a given alloy. Simulating effects of chemical reactions with contaminants EAM potentials are not satisfactory, requiring development of Reactive Bond Order Potentials (RBOP, which exist for a number of mixed materials, including metals, and are being further developed, using DFT parameters inputs). Additional complications arise due to the polarization features and long-range bonding interactions where classical molecular dynamics might not provide physically believable answers because of its intrinsic inability to treat the coordinate-dependent charging/polarizations. More complex interaction potentials that treat many-body interactions lose their accuracy with the increase of a system mixing order and the added complexity of these methods can increase computation time by two orders of magnitude.

A more accurate solution, with comparable computational feasibility is in approximate quantum mechanics in combination with classical dynamics. Namely, the specifics of the coordinate-dependent charging are polarizations defined by electron dynamics. The *Quantum-Classical Molecular Dynamics (QCMD)* based on Density-Functional Theory or its approximations could make this approach orders of magnitude more computationally intensive then CMD and yet does not rely on predefined potentials. In principle it can handle important electronic transitions in the dynamics process of the polarized materials (charge transfer) and yet present the largest computational challenges.

Alternative approaches like self-consistent-charge tight-binding (SCC-DFTB) or localized basis set approaches are the practical starting points and there has been already an extensive experience developed over a decade in using these methods in the mixed material dynamics. However, these approximate approaches contain their own challenges, specifically in the development and application of the pair-potentials for mixed materials used to accelerate the solution, which requires a further development. Although about three-orders of magnitude slower than classical MD, these are still about three orders of magnitude faster than QCMD based on DFT, thus tractable with current

supercomputing capabilities. However, the uncertainty assessment of the underlying DFTB potentials needs to be addressed still.

The development of the atomistic potentials has been historically based on the parameters developed by the DFT approached to the atomic structure. Although DFT has played an important role in the material structure computations in the previous decades, it is currently becoming possible through development of both hardware and software supercomputing capabilities to move at the next step and significantly improve quality of the classical MD potentials by using more advanced and more accurate methods of computational chemistry, with a significant positive impact to the UQ of the CMD. It is to be noted that there has been a lot of resistance and conservativism in the part of the material sciences community to this progress. This resistance is justified in case of the bulk-material research, but cannot be accepted for size of atomic clusters used for the development and parameterization of the CMD potentials.

Uncertainty Quantification within Multiscale Materials Modeling

Common framework for multiscale modeling, based on an information passing paradigm and a 'bottom-up' focus building from electronic structure calculations to atomistic simulations to kinetic Monte Carlo and/or continuum reaction-diffusion based reaction rate theory or cluster dynamics was identified. In particular, several key examples of multiscale simulations were presented by Nguyen for DFT-based calculations of binding energies to experiments of gas-defect concentrations; by B. Wirth showing a demonstration of parameterizing a cluster dynamics code based on atomistic data from available W-He interatomic potentials, and then benchmarking the continuum code against molecular dynamics databases; and by Oda that demonstrated the use of kinetic Monte Carlo simulations of H diffusion and permeation in tungsten containing defects based on DFT parameterization.

In terms of uncertainty quantification approach, von Toussaint presented a methodology for sampling the variables of interest in plasma material interaction models, while Wirth demonstrated the use of a polynomial chaos expansion to evaluate the impact of uncertainty in the knowledge of helium – vacancy cluster thermodynamics on the amount of retained helium in tungsten exposed to high flux helium plasma exposure. Overall, the discussion indicated a strong degree of agreement that these approaches to performing UQ on a multiscale materials modeling approach were appropriate and promising.

There was some discussion that global sensitivity analysis applied to KMC simulations could potentially take advantage of the parallel characteristics of the KMC method and this approach should be further utilized. The global sensitivity analysis approaches indicated that the propagation of uncertainties is quite well in hand, however, that the underlying uncertainties may not be fully well known or understood. Correspondingly, the approach with Bayesian-based sampling methodologies involving polynomial cluster expansion represents an approach to over-coming the lack of understanding, especially when coupled to the highly detailed and systematic DFT studies.

Discussions continued on the varying predictions of the Juslin et al., versus the Li et al., hydrogen surface behavior observed in MD simulations of H implantation into W. It was discussed whether a sensitivity/UQ analysis could be performed that interpolated between the two different potentials that might elucidate the underlying reason for such disparate behavior. However, it was noted that these two potentials involve differences in the base W bond-order potential, as well as the W-H potential, and this may complicate such an attempted analysis using an interpolation between the two different potential fits.

Finally, the discussion concluded by highlighting additional aspects of the plasma surface interaction physics which must be evaluated in future work. In particular, the importance of impurities and specifically the role of C, N and O impurities on the behavior of vacancies and gas atom complexes in tungsten was identified, in addition to additional emphasis on understanding the impact of tungsten surface orientation on the diffusion and permeation of H, and the He-H clustering behavior.

4. Summary and Recommendations

The objective of the meeting was to review simulation methods for PMI experiments with special interest in the sources of uncertainty of the simulations. Relevant methods included the development of interatomic potentials, DFT simulations, MD simulations, KMC simulations and Rate Equation simulations and the application of these methods for the simulation of hydrogen retention and migration in fusion materials and the calculation of particle reflection, sputtering, erosion and changes in the microstructure of materials.

The meeting was centered on discussions about the current status, challenges and open issues and the future directions of Uncertainty Quantification (UQ) activities for theoretical PMI data, especially in connection with simulations of experiments.

1. There was general agreement that the quantification of uncertainties is the key for any reasonable kind of predictive modeling activities. At the same time it was emphasized that the present state-of-the-art does not yet allow this quantification for many interesting areas of PMI. The main reasons for this are as follows:
   - The model descriptions for the systems of interest are generally incomplete, i.e., for a desired level of accuracy it is unknown which physical processes have to be included and which can be neglected or replaced by an 'effective' process. Here an intense interaction between experiment and modeling in identifying the relevant processes is mandatory. A stronger participation of experimentalists in future workshop on UQ appears desirable (e.g., how reliable are binding energies derived from thermal effusion spectroscopy used as input for modeling?).
   - Many quantities of interest are determined by processes bridging orders of magnitude in time and/or space. Thus coarse-graining of processes at several levels is mandatory for a (efficient) simulation approach. The resulting uncertainties are rarely assessed.
   - Required input parameters are often of limited or unknown accuracy. Methods for uncertainty propagation are reasonably well established, but an assessment of input uncertainties can be much more difficult.

2. Molecular dynamics (MD) simulation was identified as a key simulation technique for plasma-material-interaction. These simulation are capturing most of the physical (many-body) phenomena relevant for PMI, where details on the electron-level (eg. band-structure) are considered of secondary importance for most of the quantities of interest (eg. erosion, sputtering, retention) or as too complex (eg. mixed material phase formation in non-equilibrium conditions). There are two main limitations of MD simulations:
   - The need to resolve atomic vibrations (phonons) on the scale of femtoseconds, thus limiting the simulated period to the order of nanoseconds.
   - The dependence of MD simulations on externally provided interaction potentials (force fields) whose accuracy is often hard to assess.

   The first point requires multi-scale approaches (eg. using MD derived transition rates in KMC-codes) to bridge the time-scales. There is the inherent problem that some important,

however less frequent occurring events are missed. The second point raises the questions with respect to design and validation of MD potentials, which were intensively discussed (see next point and previous summaries).

3. The methods of designing MD potentials have changed within the last three decades. Earlier the focus was on a simple (physics inspired) analytical description which was fitted to known experimental data. With the possibility of cheap DFT calculations the large number of DFT-based energies allows to fit much more (form-free) complex models, eg. based on Gaussian processes. This leaves two issues:
    *   How accurate are the energies provided by DFT, which depend on the 'arbitrarily' chosen exchange functionals and can they be improved?
    *   How accurately do the fitted MD-energy surfaces reflect the underlying energy surface and what impact have the deviations on the quantities of interest?

    It was concluded that a parametric study to reveal P (observable | potential-parameters) may be the easiest (but by no means complete) way to identify crucial parameter space areas. Here definitely more research is needed (eg. exploiting ideas based on information-theory based measures like KL-divergence or expected utility) because the uncertainty is on a high-dimensional function, largely complicating matters.

    It was pointed out that a global sensitivity approach may also be suited (the only feasible way?) to multiscale-models, because tracing the uncertainties through all model stages is very challenging.

4. Another point was the potential role of IAEA in the process of promoting UQ and PMI modeling. A suggested data-base on MD-potentials does not appear necessary, since the LAMMPS code base is already fulfilling a similar purpose. However, a repository for DFTB (Slater-determinants) seems to be lacking and could be of use (however, the size of the potential user community remained unclear).
    It was also suggested to have a similar meeting with a slightly larger share of experimentalists, capable of assessing the reliability of experimental results and their uncertainty in the field of PMI. Here especially the techniques NRA, PALS and TDS were mentioned, because all these methods require ill-conditioned deconvolutions.

5. The compilation of a best practice sheet, e.g., for the preparation of tungsten and evaluation of TDS data could be very beneficial for the community to establish a common standard. The up-stream incorporation of uncertainty quantification techniques into the codes used for PMI-modeling (TRIDYN, SOLPS, WallDyn etc) should be emphasized and supported.

6. It must be recognized that the uncertainties in hydrogen transport in fusion materials are often entirely dominated by uncertainties in the material microstructure, notably the concentration and nature of defects that serve as trapping sites for hydrogen, and also in some cases the concentration of chemically relevant trace impurities.

It is noted that most discussions were relying to tungsten material and the tungsten-plasma interface. Although this metal is of high importance for ITER, there are a large number of fusion tokamak experiments that do not rely on tungsten as the plasma-facing material. Setting base for the PMI discussions and relevant UQ wider than tungsten was suggested.

It is apparent that the application of methods of UQ to meso-scale PMI modelling is lagging behind that of UQ in some other areas of computational science.

Overall, the coordinating effort of IAEA was very welcomed and the meeting was considered as very useful and productive by all the participants.

APPENDIX 1: Participants

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APPENDIX 2: Meeting Agenda

<u>**Wednesday 29 July 2015**</u>   Meeting Room: C0343

09:30–09:50   Opening, Introductions, Adoption of Agenda

09:50–10:00   Bas Braams: Meeting Objectives

10:00–10:40   Roberto Capote: Unified Monte Carlo: An evaluation method combining experimental and modelling uncertainties

10:40–11:00   Coffee break

11:00–11:30   H. Chung: IAEA Coordinated Activities on Uncertainty Quantification of Atomic and Molecular Data

11:30–12:10   Daiji Kato: DFT calculations of vacancy formation energies and interaction energies with hydrogen atoms in tungsten

12:10–14:00   Lunch and administrative formailites

14:00–14:40   Udo von Toussaint: Uncertainty quantification for Plasma and Plasma Wall Interaction models

14:40–15:20   Naohiko Otsuka: Experimental nuclear reaction data uncertainties - needs, concepts and documentation

15:20–15:40   Coffee break

15:40–16:20   Yves Ferro, Simulating PMI experiments from Density Functional Theory calculations: the example of tungsten

16:40–17:00   Bas Braams, Uncertainty assessment for calculated atomic, molecular and nuclear data and implications for GUM

19:00   Dinner

Thursday 30 July 2015

09:00–09:40   Gábor Csányi: Interatomic potentials with error bars

09:40–10:20   Takuji Oda: Uncertainties in modeling of diffusion of hydrogen interacting with traps in bcc metals

10:20–10:40   Coffee break

10:40–11:20   Kazuhito Ohsawa, Unique phenomena of tungsten associated with fusion reactor: uncertainties of stable hydrogen configuration trapped in a tungsten vacancy

11:20–12:00   Predrag Krstic: Quantum or Classical Molecular Dynamics?

12:10–13:40   Lunch

13:40–14:20   Duc Nguyen-Manh: Nano-cluster defects in irradiated W&W alloys: Uncertainty quantification assessment from multi-scale modelling

14:20–15:00	Brian Wirth: An approach to multiscale modeling plasma surface interactions in tungsten, with a framework for uncertainty quantification
15:00–15:30	Coffee break
15:30–16:15	Discussion on Uncertainty Quantification of Electronic Structure Calculations
16:15–17:00	Discussion on Uncertainty Quantification of Interatomic Potentials

Friday 31 July 2015

09:00–10:30	Discussion on Uncertainty Quantification of Molecular Dynamics
10:30–11:00	Coffee break
11:00–12:30	Discussion on Uncertainty Quantification of Multi-scale Modeling
12:30–13:50	Lunch
13:50–15:00	Summary and Discussion on Future work
15:00–	Adjournment of Meeting

APPENDIX 3: Abstracts

DFT calculations of vacancy formation energies and interaction energies with hydrogen atoms in tungsten

Daiji Kato, National Institute for Fusion Science, Japan

Since tungsten will be used for plasma facing component materials in ITER and is a candidate material also for DEMO reactors, tritium retention in neutron damaged tungsten is a key issue. Recently, experimental works with fission neutrons and high-energy heavy surrogate ions have been performed to evaluate the radiation effects. These works show significant enhancement of the retention in the damaged tungsten and primary roles of radiation induced vacancies and/or vacancy clusters (voids).

In divertor plasmas, the tungsten target would be exposed to low-energy and high-flux plasma particles as well as neutron. Strong enhancement of the hydrogen retention in single- and polycrystalline tungsten specimens has been observed at higher fluxes. A distinct feature of plasma irradiation from gas permeation obeying the Sieverts' law is that incident hydrogen atoms/ions from the plasma can readily overcome permeation barriers of the surface without high pressures. One can, therefore, assume that under the continuous high-flux implantation local concentration of hydrogen atoms can exceed the solubility limit of the tungsten. In such cases, the tungsten matrix may contain super-saturated hydrogen atoms and sustain extremely high stresses. Therefore, the blistering of tungsten surfaces may be induced by the exceedingly high local concentration of hydrogen atoms beyond the solubility limit of the tungsten.

For a better characterization of radiation damages in tungsten under the divertor condition, we examined vacancy formation energies by using density functional theories (DFT) with emphasis on surface energy correction and influence of hydrogen trapping. It is shown that DFT calculations using GGA-PBE tend to underestimate the vacancy formation energies. The present study supports other DFT results indicating that di-vacancy (V2) in tungsten have almost zero or negative binding energies (repulsive). However, it is shown that complexes of the di-vacancies trapping one hydrogen atom (V2H) become stable (attractive). It is also predicted by the present DFT calculations that VH6 complexes can trap 111crowdion-SIA and suppress the vacancy-SIA annihilation. Detrapping of hydrogen atoms from VHn complexes are investigated based on the hTST using DFT energies and normal mode frequencies. Calculated detrapping rates exhibit the double peak feature of thermal desorption spectra of D+ irradiated tungsten. Some uncertainty quantification issues are also discussed on DFT results of vacancy formation energies and interaction energies with hydrogen atoms in tungsten.

Simulating PMI experiments from Density Functional Theory calculations: the example of tungsten

Yves Ferro¸ Aix-Marseille Université, France

The scope of this talk is about how solving the electronic structure of an atomic system by Density Functional Theory calculations can provide the Fusion Material community with data of interest.

In a first part of this talk, I will try to classify and summarize the sources of uncertainty in DFT calculations, making the difference between *(i)* those coming from the approximations made to solve the many-body problem we are facing with, *(ii)* those coming from the technical or numerical parameters we apply in order run practicable calculations, and *(iii)* the ones arising from the real physics of the system we are studying.

In a second part, numerical-parameter-dependent uncertainties will be given within the specific example of metallic beryllium and beryllium in interaction with hydrogen.

In a last part, I will present the statistical model based on DFT we recently proposed[1] for hydrogen in tungsten. In such a model, the formation of vacancies is taken into account, which allows revising the solubility and diffusivity of hydrogen in tungsten. These results are compared with the experimental ones from Frauenfelder *et al[2]*. In the end, an improvement of the statistical model is proposed, which allows to derive the chemical potential of the system and to better understand trapping and de-trapping from vacancies in tungsten. These results are to be compared with TDS experiments[3].

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Uncertainties in modeling of diffusion of hydrogen interacting with traps in bcc metals

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Tungsten and its alloys are promising candidate materials for plasma-facing components in fusion reactors. Diffusion and solution behaviors of tritium in the materials determine tritium inventory in the components. Hence, it is important to establish appropriate models for the tritium behavior in irradiation-damaged tungsten. However, effects of irradiation damages make the tritium behavior complicated and non-linear. In addition, such models must be valid for a long term (~year) in a large system (~m), although fundamental processes occur in much smaller time and space scales. Therefore, decent quantification of uncertainty/error and its propagation in models is needed. In the present paper, we focus on hydrogen diffusivity in tungsten and discuss uncertainties in its quantity and its modeling.

Among several reported hydrogen diffusivities, the equation given by Frauenfelder [1] has been the most cited: $D = 4.1 \times 10^{-7}\exp(-0.39\ eV/kT)$ m^2 s^{-1} (Eq. 1). This formula was determined by degassing experiment of pre-loaded H$_2$ gas in 1100-2400 K [1]. However, the activation energy (0.39 eV) disagrees with the value determined by first-principles calculation based on the density functional theory (DFT), which is around 0.20 eV [2]. Heinola and Ahlgren [3] examined this disagreement, and showed that the diffusion coefficients obtained with DFT calculation are comparable with those of Frauenfelder's experiment, if experimental data at low temperatures (<1500 K) are excluded. The exclusion decreases the activation energy for hydrogen diffusion in Frauenfelder's experiment from 0.39 eV to 0.25 eV, as given in $D = 1.58 \times 10^{-7}\exp(-0.25\ eV/kT)$ m^2 s^{-1} (Eq. 2). This finding indicates significant influence of traps even at high temperatures like 1100-1500 K.

In order to quantitatively confirm this finding, we evaluated the influence of traps in hydrogen diffusivity in tungsten by using kinetic Monte Carlo (KMC) simulation [4]. As a typical trap, mono-vacancy was considered in the simulation. Hydrogen-vacancy interaction energies were taken from DFT calculation results [5]. In results of the KMC simulation, hydrogen diffusion coefficients reported by Frauenfelder [2] were nicely reproduced if hydrogen and trap concentrations expected in the experiment were employed in the simulation. The effective diffusivity of hydrogen was evidently decreased by traps even at high temperatures like 1300 K. These results suggest that only high-temperature experimental data that are not significantly affected by traps should be fitted to, in order to accurately derive hydrogen diffusivity from the experimental data. Therefore, we recommend Eq. 2 as hydrogen diffusivity in tungsten, which was obtained by fitting only to experimental data at 1500-2400 K [3].

The above discussion on the hydrogen diffusivity indicates that simple utilization of classical equations (for example, Arrhenius plot to determine the activation energy) or inappropriate design of experiments (for example, determining a quantity at conditions where the quantity is sensitive to some experimental parameters that are difficult to control) cause non-negligible errors in the determined quantity, even when experiment itself was carefully performed. Comparison with results of KMC simulation (or rate equations) combined with kinetic parameters given by DFT calculation would warn us of a possible error in the experimental result. Specifically, if there is a significant difference between an experimental result and a simulation result, it is better to critically re-scrutinize both results.

Although DFT calculation is a powerful tool, we also need to recognize that results of DFT calculation are not free from uncertainty/error. One intrinsic source of uncertainty/error in DFT calculation is exchange-correlation functionals. For tungsten, we systematically calculated defect formation energies and migration energies with various functionals. It was confirmed that these energies can be different by up to around 10%, depending on a sort of the functionals and a kind of property to be calculated [6]. As a practical method, performing DFT calculations with several different functionals may indicate a reasonable estimate of uncertainty/error expected in DFT calculation results.

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Unique phenomena of tungsten associated with fusion reactor: uncertainties of stable hydrogen configuration trapped in a tungsten vacancy

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Back ground and motivation

In the fusion reactor, divertor armor tiles are exposed to extremely intense plasma particle irradiation. In order to protect divertor, its surface is planning to be covered by tungsten (W) or its alloy. So, study of interaction between W and plasma particle started. W is one of promising plasma facing materials because of high melting point, low hydrogen solubility, and low sputtering erosion. Besides, it has high thermal conductivity and low induced radioactivity. By the way, some unique and unexpected phenomena are observed associated with the interaction between W and plasma particles (H and He). (1)Negative formation energy for di-vacancy in W. (2)He cluster migrations more quickly than a single He atom. (3)Fibreform tungsten covering surface by He irradiation. (4) Abnormal H configuration in W mono-vacancy.

We will focus on the fourth topics, abnormal H configuration in W mono-vacancy, in the present work. Simulations are often performed on the basis of expectation and prediction. However, they sometimes cause failure due to the preconception. We suppose that uncertainties on simulations contain such human mistakes. The method for examining stable H configuration would be a good solution to avoid such mistakes.

Review of H in bcc metal

Typical interstitial sites of bcc metals are tetrahedral interstitial site (T-site) and octahedral interstitial site (O-site). According to the previous works, T-site is more favorable for an H atom than O-site in perfect bcc metals. On the other hand, an H atom trapped in a mono-vacancy is located close to an O-site. If multiple H atoms are trapped in the mono-vacancy, each H atom is also located close to an O-site. As a result, a maximum of 6 H atoms can be accommodated in the mono-vacancy because there are 6 O-sites next to the mono-vacancy in bcc lattice. This is standard model for H configuration trapped in a mono-vacancy in bcc metals.

Simulation method

In the present simulation, we don't assume that H atoms in a W mono-vacancy are located close to the O-sites. Instead, initial H positions for ionic relaxation are randomly generated to find unexpected and the most stable configuration. We performed first-principles calculations based on DFT using Vienna ab-initio simulation package (VASP). PBE potential is used. We employed supper cell composed of 54 atoms. Plane wave cut-off energy is 350 eV. Electronic and ionic relaxations are iterated until a break condition (0.003 eV/angstrom) is satisfied for every atom.

Results and discussion

Figure 1 shows typical abnormal and stable configurations of H atoms in W mono-vacancy. Stable configuration of 4 H atoms is tetrahedral structure but planar one. Stable configuration of 6 H atoms is complicated. Symmetric 6 H structure is unstable. A maximum of 12 H atoms can be accommodated in the vacancy and they are located close to T-sites. Figure 2 shows the total binding energies of single and multiple H atoms to a mono-vacancy in bcc transition metals. A mono-vacancy in W and Mo can accommodate 12 H atoms. However, 6 H atoms are trapped in a vacancy in other bcc metals, which is good agreement with the standard model. The abnormal and stable H configurations in W mono-vacancy are obtained from a special scheme of initial H arrangements. Careful examinations for stable atomic configurations are supposed to be important for lattice relaxation simulations.
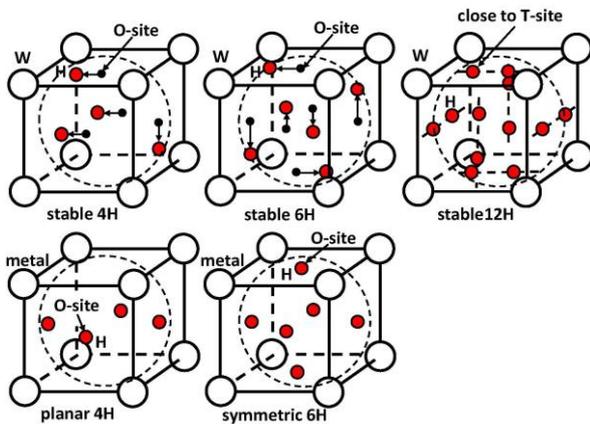

Fig. 1: Schematic view of abnormal and stable H configurations in W vacancy for 4, 6, and 12 H atoms (upper 3), and standard H configurations for 4 and 6 H atoms (lower 2).
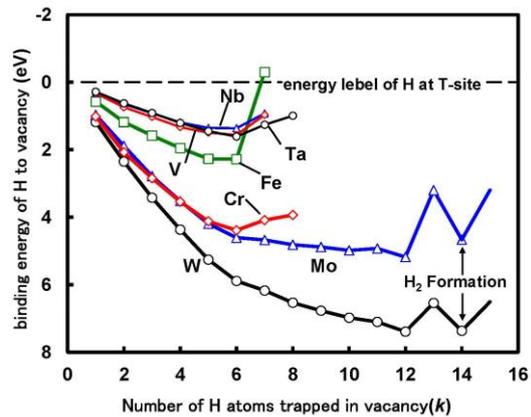
Fig 2: Total binding energies of single and multiple H atoms to mono-vacancy in bcc transition metals (V, Nb, Ta, Cr, Mo, W, and Fe).

Quantum or Classical Molecular Dynamics?

Predrag Krstic, Institute for Advanced Computational Science, Stony Brook University, USA, di

Plasma-Material Interface (PMI) mixes materials of the two worlds, plasma and material, creating in between a new entity, a dynamical surface which communicates between the two, creating one of the

most challenging areas of multidisciplinary science, which has many fundamental processes and synergies. The surface science is not the material science, it has its own temporal and spatial scales which depend on the impact plasma particles energies, and requires specific scientific approaches, not necessary typical for either material or plasma sciences. The understanding of the phenomenology of this interface to bridge the epistemic UQ, endemic in this sciences,

The understanding of the dynamic surface response and evolution in mixed material environments (D, W, He, O, Be, Li, C, B, O, Mo, TZM, Fe, Re, Ta) of the present and future fusion devices (ITER, DEMO, NSTX,…) with impingement of plasma particles in the energy range below 100 eV, but also a few keV (ELMs, disruptions), including microstructure changes, erosion, surface chemistry, deuterium and helium implantation and permeation, redeposition, diffusion, etc. at various surface temperatures, with their aleatoric and epistemic uncertainties, are parts of the big picture and main underlined goal of this meeting. It is understood that the resolution of the uncertainties, endemic in this PMI research, will come from application of the developed computational tools to existing tokamaks, both for the purpose of model validation and to assist in experimental planning. Besides predicting and understanding the phenomenology of these processes, being atomistic in its nature, and can be provided by atomistic approaches like is MD, probability rates for the processes and reactions to be used in the source and sink terms of the mesoscopic approaches (that are used to connect various temporal and spatial scales, as described by B Wirth) are also product of the MD approaches.

How to build a theoretical approach integrated with experiment? To start, it is worth noting that, for example, a flux of 10^{25} particles/m^{2}s at the hot gas-material interface means that, statistically, one particle is impinging on a surface of 10 nm^2 every 10 ns. This gives to the system enough time to evolve freely from the external influx of energy and particles, still staying far from equilibrium. With an impact particle energy of 100 eV this would result in heat flux of ~160 MW/m^2. However, a typical chemistry-based process, like is chemical sputtering, at impact energies $\leq$ 100 eV, fully evolves at the interface for less than 50 ps, implying that each impact is independent, uncorrelated, and discrete. However, each impact changes the surface, functionalizing it for the subsequent collision, i.e. building a dynamic surface with memory, and depositing energy.

At the high impact energies of a gas-solid interface Monte Carlo binary potential methods (SRIM, are an acceptable alternative for situations such as material dislocation dynamics. However, classical molecular dynamics is only as good as the interatomic multibody potential model used. *Creation, calibration and validation of these multibody, semi-empirical potentials is an important computational/experimental challenge which was extensively discussed at the meeting by Gabor, comparing Finnis-Sinclair and Tersoff-like potentials for W.* In addition, for simulations of metallic systems, the Embedded Atom Method is a relatively accurate technique that is related to the second moment of tight binding theory. Essentially, in addition to pair-wise interactions among the atomic species, there is an additional term called the embedding function, which approximates the energy required to insert an atom in the existing cloud of electrons. Validated EAM potentials can be very accurate for a variety of mechanical properties of metallic systems, but tend to be complex and not easily mixed. In other words, it is generally necessary to specifically parameterize a given alloy, rather than being able to calculate the interactions between components through a generic mixing rule, as is often the case for Lennard-Jones type potentials. The issue here is further complicated in that EAM, and MD in general, does not treat variable charge on atomic species and may not be applicable to these systems. This is a particular challenge because of the nonlocalized character of metallic coupling. Such potentials will likely require further validation for the properties in which we are interested, which will be available from the experimental aspect of the research. The candidate potentials can be identified, various EAM and ReaxFF, that may be applicable, but their accuracy and applicability are yet to be determined. However, to simulate effects of chemical reactions with

contaminants EAM potentials are not satisfactory, requiring development of Reactive Bond Order Potentials (RBOP, which exist for a number of mixed materials, including metals, and are being further developed, using DFT parameters inputs). Additional complications arise due to the polarization features and long-range bonding interactions, specifically in the case of species with low (Li) or high (O, N) electronegativity interacting with species with intermediate electronegativity (H, C, Sn, W, Fe, He, and other materials). In these cases classical molecular dynamics might not provide physically believable answers because of its intrinsic inability to treat the coordinate-dependent charging/polarizations. A potential solution lies in more complex interaction potentials that treat many-body interactions (and explicitly including variable charges, including bond-order potentials, ReaxFF and COMB, or in explicit development of semi-classical parameterization which is capable of computing the atomic charges dynamically, as the coordinates change (Electronegativity Equalization Method, EEM). These methods lose their accuracy with the increase of a system mixing order. In addition, the added complexity of these methods can increase computation time by two orders of magnitude, which can adversely affect the time and length scales accessible to the simulations. If these methods become a necessity, it will be useful to use them in tandem with the faster, less accurate techniques with each method applied in the proper realms.

A more accurate solution, with comparable computational feasibility is in approximate quantum mechanics in combination with classical dynamics. Namely, the specifics of the coordinate-dependent charging are polarizations defined by electron dynamics. Since electrons are necessarily quantum-mechanical entities, the *Quantum-Classical Molecular Dynamics (QCMD) of large and mixed systems presents, in these cases, a first principles alternative to classical MD, and a scientific discovery challenge.* QCMD, whose quantum component is based on Density-Functional Theory or its approximations could make this approach orders of magnitude more computationally intensive then CMD. However, QCMD does not rely on predefined potentials, and can in principle handle important *electronic transitions in the dynamics process of the polarized materials (charge transfer), which have been so far largely neglected. This presents the largest challenge for the discovery computational science.* While the accelerated molecular dynamics or coarse grain methods are conceptually difficult to apply to a quantum mechanical system, alternative approaches, like self-consistent-charge tight-binding (SCC-DFTB) or localized basis set approaches are the practical starting points for this endeavor. There has been already an extensive experience developed over a decade in using these methods in the mixed material dynamics. However, these approximate approaches contain their own challenges, specifically in the development and application of the pair-potentials for mixed materials used to accelerate the solution, which requires a further development. Although about three-orders of magnitude slower than classical MD, these are still about three orders of magnitude faster than QCMD based on DFT, thus falling in the ball-park of the current supercomputing capabilities.

The development of the atomistic potentials has been historically based on the parameters developed by the DFT approached to the atomic structure. Although DFT has played an important role in the material structure computations in the previous decades, it is currently possible through development of both hardware and software supercomputing capabilities to move at the next step and significantly improve quality of the classical MD potentials by using more advanced and more accurate methods of computational chemistry, with a significant positive impact to the UQ of the CMD. It is to be noted that there has been a lot of resistance and conservativism in the part of the material sciences community to this progress. This resistance is justified in case of the bulk-material research, but cannot be accepted for size of atomic clusters used for the development and parameterization of the CMD potentials.

Nano-cluster defects in irradiated W and W alloys: Uncertainty quantification assessments from multi-scale modelling

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Understanding of irradiation-induced vacancy behaviour in tungsten is one of outstanding and debate issues since the 1960s although numerous studies have been taken with regard to the generation and formation of defect structures formed under irradiation. On one hand, a nucleation and growth of vacancies into voids and dislocation loops can be observed in damage microstructures in W and W alloys using neutron and ion irradiation. On the other hand, the vacancy population produced by irradiation was unaccounted for using TEM due to nano-scale sizesin but small vacancy clusters can be detected by Positron Annihilation Spectroscopy (PAS).

By employing multi-scale approach based on first-principles calculations for which the inhomogeneous and low electron density in vacancy-cluster regions has been taken into account, we have performed a systematic assessment of quantification uncertainty at different multi-scale levels of energetic and kinetic properties of nano-vacancy in bcc TMs with a particular focus on tungsten and origin of the observed anomalous effects of solute segregation in W alloys. The formation and migration energies of vacancy clusters in nanometer-size scale as well as different dissociation pathway of vacancy clusters have been investigated in order to assess the stability and transformation of different defect configurations by using density functional theory (DFT) calculations in combination with molecular relaxation and kinetic Monte-Carlo simulations at various ranges of temperature and time. Importantly, our ab-initio data base for vacancy cluster calculations have been used to designing a new EAM-type interatomic potentials for which the surface corrections to the embedding functional have been implemented. By cross-checking both formation and migration energies for nanoclusters of vacancies, the agreement between new corrected EAM potentials with DFT calculations is very satisfactory. The predicted temperature of vacancy-cluster dissociation is consistent not only with observation of the voids but also with the variation of dislocation length in high-temperature annealing of self-ion irradiated tungsten [1]. Finally, we describe briefly the extension of a combined DFT-MC formalism by considering vacancy as a new element in a multi-component alloys within the Cluster Expansion (CE) in order to study the origin of coherent precipitates in W-2%Re alloys under irradiation [2]. The quantification uncertainty within this DFT/CE/MC approach is mainly related to cross-validation of effective cluster interactions between DFT and CE that is only 5 meV/atom in the W-Re-vacancy systems.

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An approach to multiscale modeling plasma surface interactions in tungsten, with a framework for uncertainty quantification

Brian Wirth and colleagues

The presentation began with a view of an information-passing multiscale materials modeling approach, which is hierarchical in nature and integrates *ab initio* electronic structure calculations, molecular dynamics (MD) simulations, kinetic Monte Carlo (KMC), and kinetic reaction-diffusion rate theory simulations with thermodynamics and kinetics through the passing of information about the controlling physical mechanisms over the relevant length and time scales to model the fates of gas

atoms and defects relevant to plasma surface interactions. The presentation then summarized observations from large-scale MD simulations elucidate the mechanisms of helium behavior following implantation into tungsten that are likely responsible for the early stages of tungsten surface evolution leading to the nanometer-scale fuzz, as well as have the potential to significantly modify tritium retention behavior.

The atomistic modeling of the tungsten – helium system have clearly shown the insolubility of helium, it's high mobility as an interstitial atom which leads to rapid diffusion in a more-or-less random trajectory that can result in returning to the free surface and escaping, or diffusing deeper into the solid until it encounters a defect or another helium atom. Due to the essentially repulsive nature of the helium – tungsten interactions, combined with the shorter range and lower magnitude of repulsive helium-helium interactions, the helium has a strong driving force to cluster. Small helium clusters are also mobile, as long as all of the helium reside in interstitial positions in the BCC tungsten lattice, and any clusters that form continue to perform an essentially random walk with fast diffusivity. As the migrating helium clusters grow larger, they eventually reach a condition in which the effective pressure generated is sufficient to create a tungsten vacancy and self-interstitial (Frenkel) pair, in a process called trap mutation. The size at which trap mutation first occurs depends on a number of factors, including the temperature and the proximity of the helium cluster to the free surface, as well as the surface crystallographic orientation. Growing, over-pressurized helium bubbles continue to grow and release pressure by dislocation loop punching and eventually bubble rupture. Such bubble growth processes lead to significant surface roughening. Results were then shown at a helium implantation flux of $4x10^{25}$ $m^{-2}s^{-1}$ to fluence on the order of $3x10^{19}$ m^{-2}, which showed a clear influence of surface orientation on the formation and evolution of a near-surface layer enriched in helium, as well as a distribution of deeper bubbles. Multiscale results showing kinetic Monte Carlo and then a new, spatially dependent reaction-diffusion-advection cluster dynamics model, which were parameterized by the thermodynamics and kinetics of helium-vacancy clusters from the atomistic simulations and the elastic drift forces and the modified trap mutation behavior observed near tungsten surfaces were used to demonstrate an outstanding initial benchmarking of the continuum model predictions in comparison to the MD simulation results.

The presentation next focused on demonstrating an approach using Bayesian analysis to fit the atomistic data for the Helium – vacancy cluster formation energies, and used this approach to both interpolate and extrapolate the limited atomistic data, and to use polynomial chaos expansion of the resulting Legendre polynomial fitting functions of formation energies as a function of number of helium and vacancies to perform a global sensitivity analysis. While that analysis indicated no impact of the helium – vacancy cluster binding energies on the quantities of interest for helium retention and sub-surface helium bubble evolution for low-energy helium implantation at a temperature around 900 K. However, the framework is in place for further global sensitivity analysis associated with the helium kinetics and the reaction rate constants controlling helium clustering.

Finally, the presentation presented preliminary atomistic simulation results for H partitioning to sub-surface, over-pressurized helium bubbles that indicate a strong interaction between H and the He bubbles that traps a significant amount of hydrogen at the bubble interface with the matrix, and substantial hydrogen retention to very high temperatures. These initial results raise concerns about possible tritium retention in helium bubble containing divertors and motivate additional high-fidelity density functional theory calculations.