Multi-model Quantification of Defects in Irradiated Lithium Titanate

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Abstract

Lithium based ceramics (Li_2TiO_3, Li_4SiO_4 etc.) in the packed pebble form, used in breeder blankets to breed tritium, are promising candidates for future fusion reactors. Neutron induced trap sites in the ceramic breeder affect the diffusion and inventory of tritium and thereby influence the quantity of tritium recovered. In the present work, a multi model framework has been employed to quantify the number of defects, which can be used to evaluate trap-sites, due to neutron irradiation in porous Li_2TiO_3 pebbles. This framework is based on a three stage calculation viz. (i) radiation transport, (ii) obtaining the primary knock-on atom (PKA) energy spectrum and (iii) quantifying the ensuing damage by collision cascade simulations. The output of one stage is coupled to the next stage and finally the defect distribution is obtained. We use radiation transport calculations to obtain the neutron spectrum in ceramic pebbles. This is used as an input to SPECTER, a code that uses evaluated cross-sections, to obtain the Primary Knock-on Atom (PKA) energy spectrum. The PKA energy spectrum is then used to find the number and distribution of vacancies and displacements in porous Li_2TiO_3 pebbles using binary collision approximation (BCA) based Monte Carlo simulations. The vacancy distribution and the number of vacancies in Li_2TiO_3 have been evaluated for the ITER neutron source using the above frame work. The PKA energy spectrum of Li, Ti and O PKAs, have been obtained using SPECTER, and their average energies are 680, 120 and 320 keV respectively. The collision cascade simulations shows the expected Bragg peak for the number of defects created and has a range of 6-8 microns for O and Li PKA respectively and a fraction of a micron for Ti PKA. The total number of displaced atoms which is equal to the number of vacancies, at the end of the collision cascades, obtained using the above framework has also been used to estimate the displacements per atom (dpa) in Li_2TiO_3 and this shows a good agreement with the dpa calculated using the NRT model.

Key words: *Lithium Ceramics*, *Breeding blanket, Radiation Transport, PKA, Binary Collision Approximation, dpa*

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1. **Introduction**

D-T plasma based fusion reactors need tritium as a fuel to sustain fusion reactions. The tritium will be bred in the blanket of the reactor. It will be extracted and fueled back to the plasma for the operation of the reactor. Tritium in the breeding blanket will be generated through the n(Li, α)T reaction. Lithium based ceramics such as Li_2TiO_3, Li_4SiO_4 etc. are promising candidates for the breeder materials to be used in the breeding blankets [1–8]. Neutron interactions with these ceramic breeders not only generate tritium but also produce energetic Primary Knock on Atoms (PKAs), which trigger collision cascades in the material. This leads to the creation of interstitials and vacancies. In Lithium based ceramics the Oxygen related interstitial and vacancies are of more importance. Generally in metal oxides, three pair of vacancy and interstitial are generated. One is neutral vacancy and Oxygen interstitial. The other two are vacancies with one & two electrons known as F+ and F centers and corresponding O- centers [9]. Specifically for neutron irradiation of Li2TiO3, F+ and O- centers are considered in tritium release modelling [10]. Activation energies for annihilation of F^+ and O^- centers have been reported to be of the order of 0.6-0.9 eV [10,11,12] and the activation energy for tritium diffusion in Li_2TiO_3 is reportedly 1.07 eV [10,13]. These O- centers also become trap sites and can combine with tritium to make hydroxyl group. This process can also affect the tritium diffusion and inventory in Lithium Ceramics. The tritium bred in the lithium ceramics pebbles can diffuse and get re-trapped at the trap sites produced by the PKA. The diffusing tritium can also react to form molecules that have different diffusion properties as compared to its constituents. Therefore, diffusion of tritium and its recovery is affected by the damage to the ceramic pebbles.

Tritium release behavior studies in Li_2TiO_3 is an important R & D area for D-T based fusion reactors. Several tritium release studies have been carried out and reported in literature. In some of the studies theoretical tritium release models are proposed and the kinetic parameters for tritium transport are obtained by curve fitting with experimental tritium release curves [14-16]. On the other hand in some other studies [10,17,18] it is discussed that the tritium diffusion in the Li_2TiO_3 material is affected by the presence of radiation induced defects. A tritium release model is presented in the ref. [10] which takes defect densities and trap densities as input for evaluation of the tritium release from Li_2TiO_3. Quantification of damage and its distribution plays an important role in estimating the tritium inventory and extraction from the solid breeder blankets. Radiation damage studies involve the evaluation of PKA energy spectra and average PKA energies for a given neutron spectrum. SPECTER [19] is widely employed to obtain PKA energy spectra in materials

[20-23]. Generally, the average energy of PKA or specific energy of PKA is picked and used for the cascade simulations [24-26].

We have developed a framework to evaluate the defect distribution, which can be used to obtain trap-sites, in lithium based ceramics. In this frame work, Monte Carlo neutron transport calculation provides the neutron spectrum in the ceramic pebbles. The neutron spectrum is used as inputs to SPECTER to obtain the PKA energy spectrum of Li, O and Ti. The PKA energy spectra of Li, O and Ti are then used to find the distribution of defects by Monte Carlo simulations of collision cascades based on the binary collision approximation (BCA) using SDTrimSP [27, 28]. The dpa has estimated using the NRT model [29] is compared with the dpa calculated by our calculations for validation. The defect evaluation framework is described in section 2. Results are discussed in section 3 and conclusions are presented in section 4.

2. **Frame work for defect evaluation**

Quantification of the number of defects has been carried out in three serial steps and in each step the calculations are carried out with separate codes. A framework as shown in fig-1 has been developed which couples these steps and gives the number of defects and their distributions. The output of one step becomes the input for the next step. These steps involve radiation transport, generation of PKAs and collisional cascade simulations to obtain the damage. The radiation transport simulation gives a neutron spectrum that is used to calculate the PKA spectrum. Binary Collision Approximation based Monte Carlo simulations are then carried out for the calculated PKA spectrum and the distribution of defects is obtained. The details of these steps are given below:

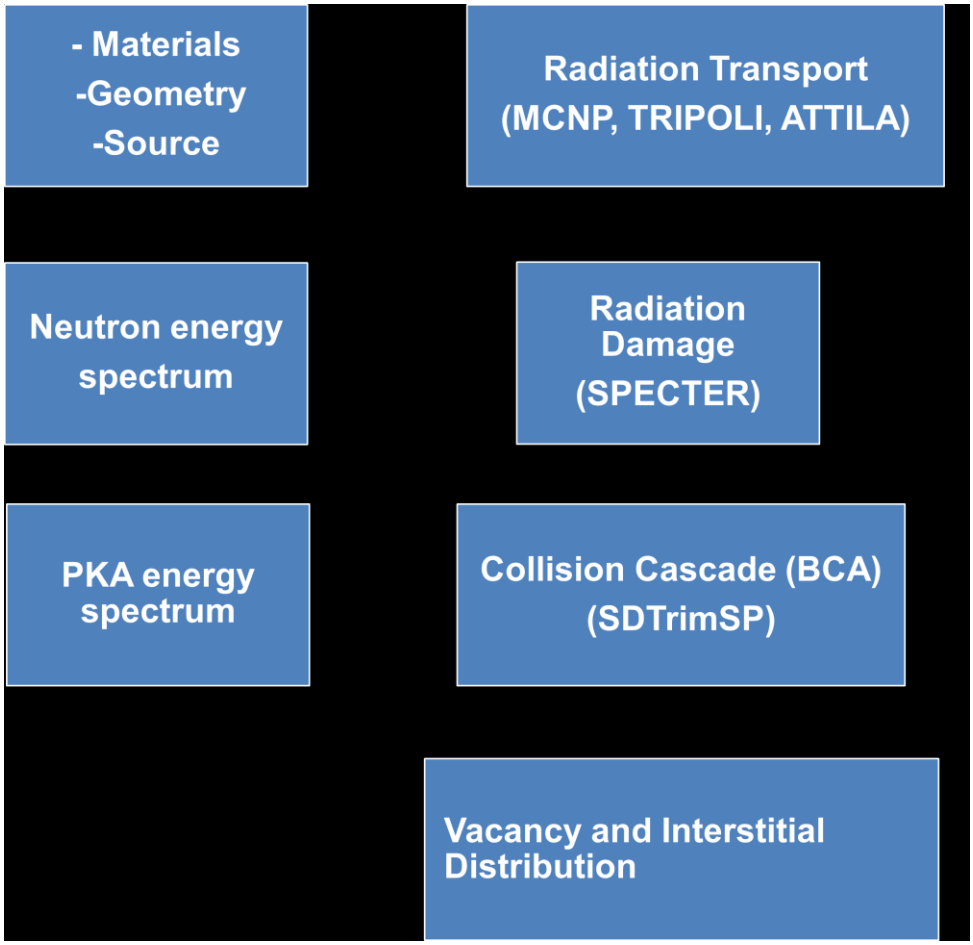

Fig-1. Frame Work for defect distribution evaluation

2.1 Radiation Transport simulations

A Mock-up of the Indian LLCB breeding blanket module [30-32] has Li_2TiO_3 as tritium breeder. A neutronic model of this blanket module is constructed and inserted in the 3-D ITER neutronic model [33]. The ITER neutronic model along with zoomed view of blanket module mock-up is shown fig-2. The Monte Carlo Radiation Transport code MCNP [34] along with Fusion Evaluated Nuclear Data Library (Fendl-2.1) [35] is used for the evaluation of nuclear responses such as neutron spectrum and fluxes in Li_2TiO_3. In the simulation, 50 million particle histories were tracked and the nuclear responses were normalized to 500 MW of fusion power.

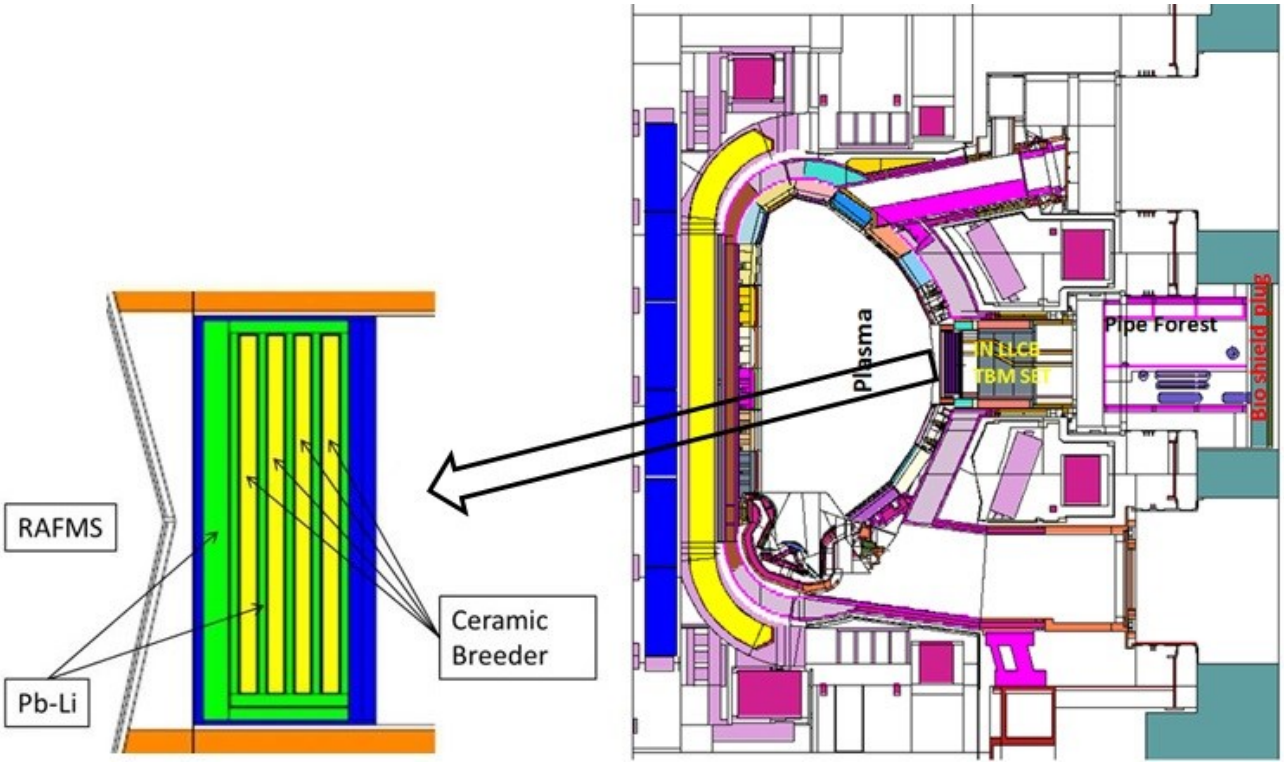

Fig-2. Neutronic model of ITER with LLCB TBM placed at equatorial port

2.2 PKA Energy Spectrum Calculation using SPECTER

Displacement of atoms and transmutations are two main primary radiation effects caused by neutron irradiation on materials. Neutrons can produce atomic displacement through two main channels: (1) direct displacement of an atom by neutrons which results in displacement cascades (2) absorption of neutron by atom and producing γ, α, β, p particles along with recoil which results in a displacement cascade. SPECTER is a neutron damage code which takes the neutron spectrum as input and calculates the displacement damage cross-section, displacement per atom (dpa), hydrogen & helium gas production and Primary Knock on Atom (PKA) energy spectrum. The SPECTER code includes scattering (elastic and inelastic), (n,xn), (n,p), (n,d), (n,d), (n,α), (n, γ) etc. reactions to estimate the neutron-induced damage in the given material. Group averaged cross-sections are prepared from the nuclear data and provided as data set with SPECTER. The user provides a neutron spectrum and based on this spectrum SPECTER calculates the spectral averaged quantities for various elements. We have obtained the neutron energy spectrum in Lithium Titanate from the previous step of our framework and used it in the SPECTER to evaluate PKA energy spectra and

number of PKAs for Lithium, Titanium and Oxygen. A packing fraction of 60 % with 10 % porosity is considered to calculate the atom densities of Lithium, Titanium and Oxygen in Li_2TiO_3. Using these atom densities we have quantified the PKAs of Lithium, Titanium and Oxygen.

2.3 Collisional Cascade Simulation using SDTrimSP

SDTrimSP is a Binary Collision Approximation based Monte Carlo code designed for simulating atomic collisions to calculate ranges, reflection coefficients and sputtering yields as well as more detailed information such as depth distribution of implanted and energy distribution of backscattered and sputtered atoms. Three separate runs for Lithium, Titanium and Oxygen PKAs were carried out. The whole energy spectrum of PKAs has been used as input to SDTRIM-SP which uses a rejection method to sample the PKA spectrum. 100, 1000 and 10000 PKA histories are tracked to sample the PKA energy distribution until the results did not change other than the statistical fluctuations. The values of the displacement energies for Lithium, Titanium and Oxygen are taken to be 25, 85 and 49 eV respectively [36]. The defects distributions produced by PKAs will be input to a Kinetic Monte Carlo (KMC) code that carries out tritium diffusion in the presence of defects. Therefore, in order to obtain a typical defect distribution for the generated PKA spectrum we initiated the PKA at 500 Angstroms and directed it away from the surface into the solid in SDTrimSP simulations. In order to account for the porosity (10%) and packing fraction of Li2TiO3 pebbles (60%), we applied a factor 0.54 to the theoretical density of Li_2TiO_3 and used a reduced density for our calculations. A depth of 500 Angstroms makes sure that no defect comes out of the surface (sputters out) of the system.

3. Results and discussions

14 MeV neutron generated in the plasma will interact with first wall and other components in ITER. Multiple scattering and interaction with various materials will lead to a neutron spectrum within Li_2TiO_3. A neutron spectrum, resolved in 100 energy groups has been calculated in the Li_2TiO_3 zone. This neutron spectrum is shown in fig-3.

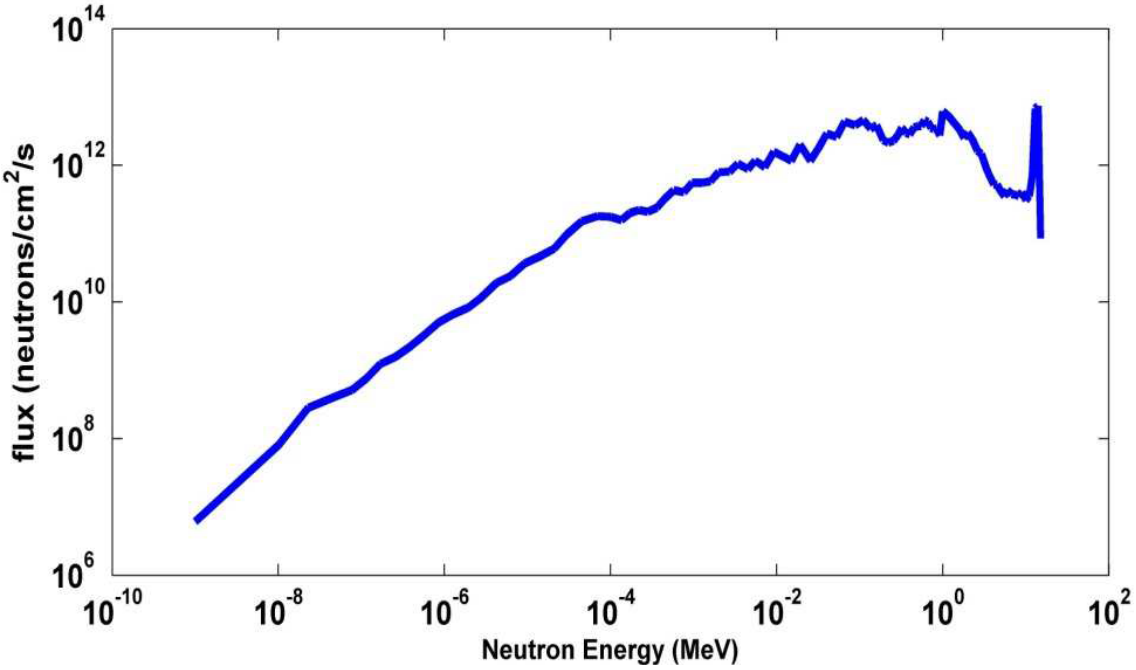

Fig-3. Neutron energy spectrum in Li_2TiO_3

These neutrons will generate PKA in Li_2TiO_3 and give rise to damage. Neutrons will interact with Li_2TiO_3 and transfer energy to Lithium, Titanium and Oxygen atoms. The energy spectra of PKA of Lithium, Titanium and Oxygen have been calculated by SPECTER and are given in fig-4. Specifically the numbers of PKAs within an energy bin are shown on the Y-axis. Note that the axes have a log-scale in fig-4. The PKA spectra show a discontinuous jump as each order of magnitude is crossed on the log-scale because the PKA energy bin width also increases by ten times. It is observed in fig-4 that the peak for Ti PKA occurs at lower energy (around 0.01 MeV) whereas the Li and O PKA peaks around 1 MeV. This is due to higher mass of Ti than that of Li and O.

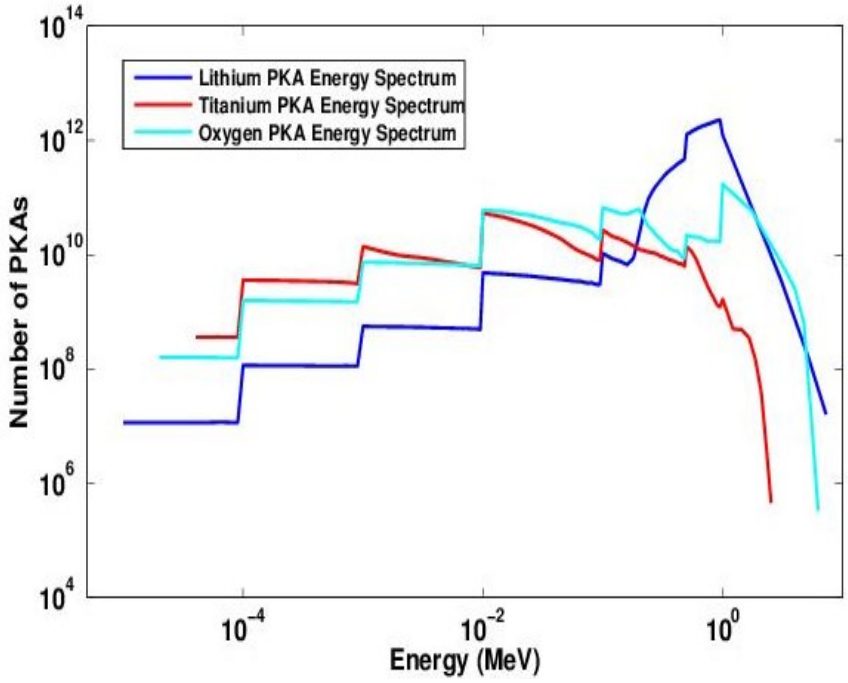

Fig-4. Lithium, Titanium and Oxygen PKA energy spectrum in Li_2TiO_3

The PKA spectrum of Lithium, Titanium and Oxygen will further trigger collision cascades and will displace atoms in Li_2TiO_3. The BCA-MC code SDTrimSP has been used for collision cascade simulations. PKA of Lithium, Titanium and Oxygen with their energy spectrum were launched inside Li_2TiO_3 at a distance of 500 Å. The vacancy distributions caused by Lithium, Titanium and Oxygen PKA as calculated by SDTrimSP are shown in fig-5 and the resultant total vacancy distribution is given in fig-6. These figures show that for given PKA specie, vacancy concentration increases towards the end of the range of the PKA (Bragg peak). PKA in materials lose their energy by two mechanisms, namely electronic and nuclear stopping. Nuclear stopping is responsible for the creation of vacancies whereas electronic stopping slows down the PKA. Electronic stopping dominates at higher energies of the PKA (greater than several tens of keV depending on the material). At lower energies, the PKA will have higher interaction probabilities because the interaction cross-section is inversely proportion to the velocity of PKA. These results in a higher concentration of vacancies towards the end of their range after the PKAs have lost their energies due to electronic stopping.

The nuclear energy loss of PKA energy is responsible of displacing the atoms in Li_2TiO_3 therefore the vacancy distribution in Li_2TiO_3 follows the nuclear energy loss profile. The nuclear energy loss profile in Li_2TiO_3 for Lithium PKAs is shown in fig-7. It is clear from fig-7 that as the

Li PKA travels in Li_2TiO_3, the nuclear energy loss increases, reaches a maximum towards the end of the range, and then decreases rapidly. The average energies of Li, Ti and O PKAs, as obtained from SPECTER, are 680, 120 and 320 keV respectively. Moreover, Ti is more massive and has many more electrons than either Li or O. Therefore, the range of Ti is smaller than either Li or O.

The total number of displaced atoms is the number of defects created during the collision cascade which is obtained from the third stage of the simulation using the SDTrimSP code. Using these displacements and the range of the PKA, the dpa for Li_2TiO_3 turns out to be 2.9 for one year of continuous irradiation in ITER. We have also employed the NRT model and used PKA energy spectrum to obtain the dpa with NRT model. The dpa in Li_2TiO_3 calculated by this model is 3.2. Both the dpa values are very close and this provides a check on the estimations of defect numbers and distribution in Li_2TiO_3.

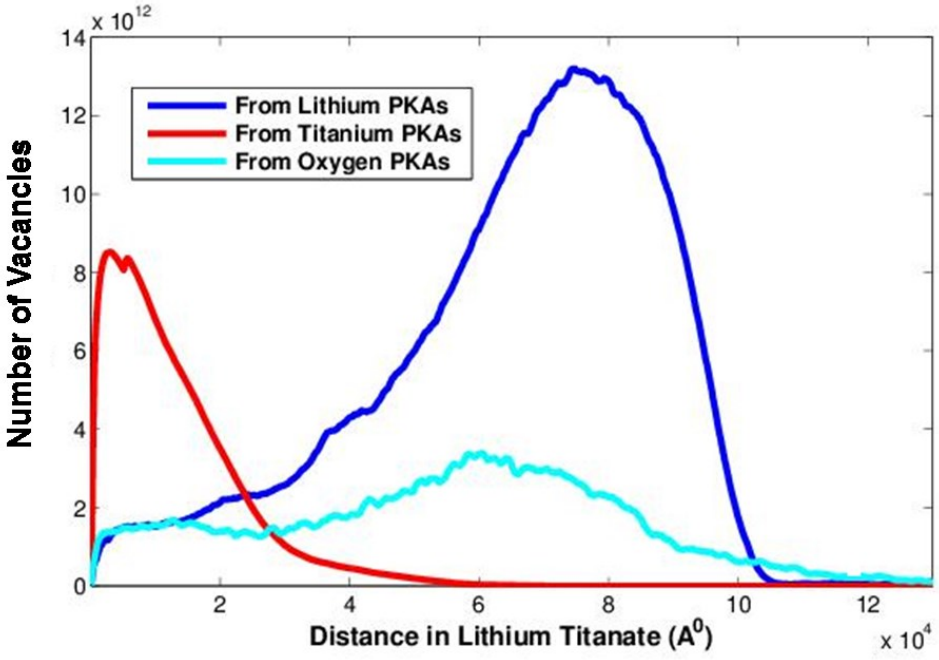

Fig-5. Vacancy distributions formed by Lithium, Titanium and Oxygen PKAs in Li_2TiO_3

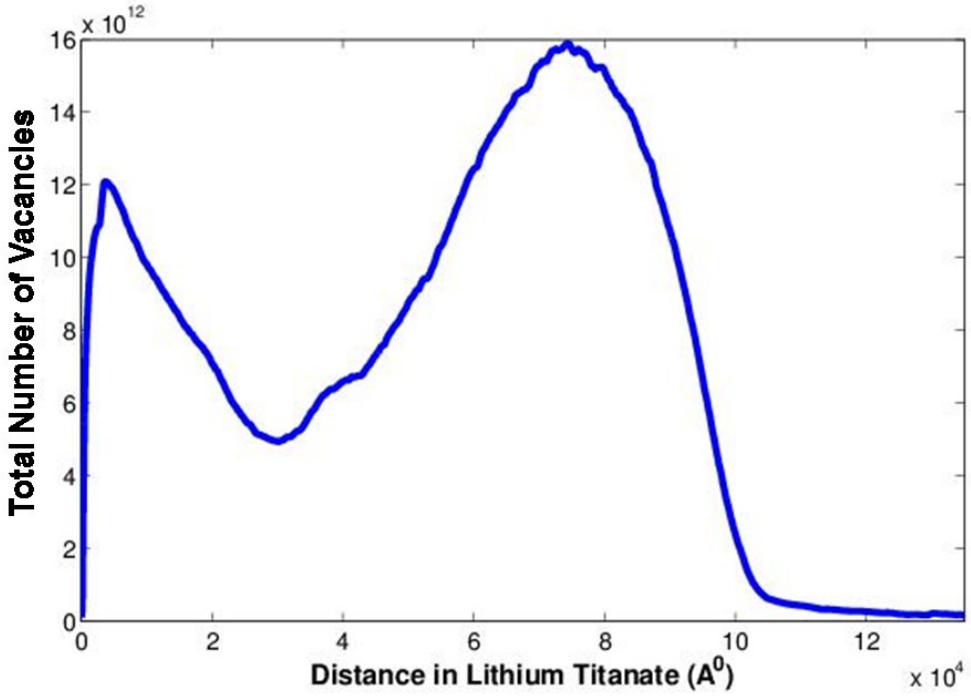

Fig-6. Total vacancy distribution due to Li, Ti and O PKA in Li_2TiO_3

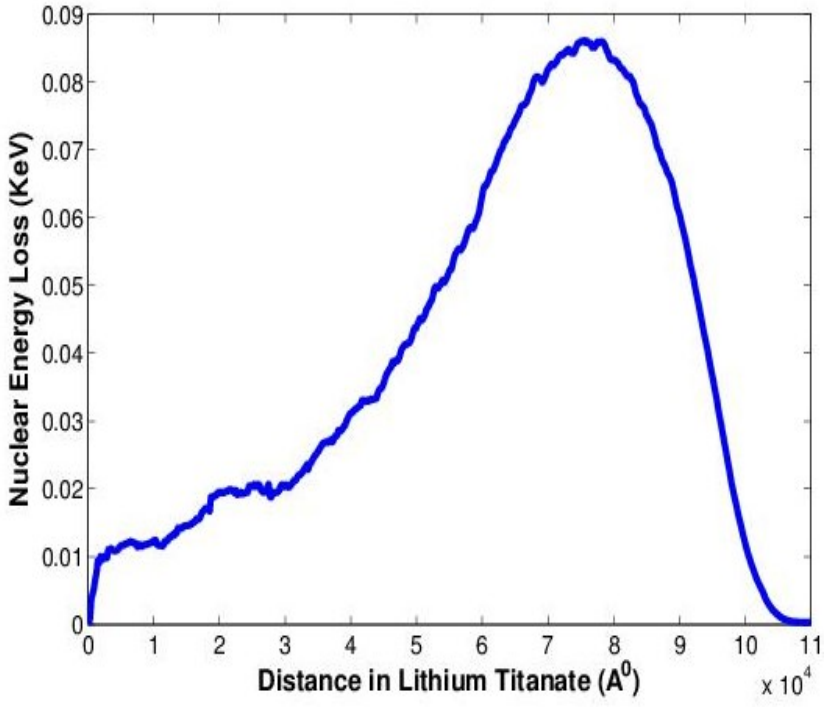

Fig-7. Nuclear energy loss profile of Lithium PKAs in Li_2TiO_3

4. **Conclusion**

A framework to obtain the defect distribution in materials under neutron irradiation is presented. Suitable coupling of the output of one stage of calculation to input of the next stage calculation has been carried out. The framework has been applied to obtain the defect distribution in Li_2TiO_3 under ITER irradiation conditions. The diffusion controlled recombination of tritium in porous lithium ceramic pebbles is important to estimate the tritium recovery from blankets. Therefore, these results are essential inputs to the tritium diffusion studies in Li_2TiO_3. The Li and O PKA spectra have peaks at around 1 MeV and show Bragg peaks around 7 microns away from the origin of the PKA in the corresponding vacancy distribution profile. The Ti PKA has a peak at around 0.01 MeV and creates vacancies much closer to the origin of PKA. Displacement Per Atom (dpa) has been calculated in Li_2TiO_3 using the frame work described above and the NRT model. Both the dpa are in reasonably good agreement with each other. This shows that the framework forms a good basis for the defect distribution evaluations in irradiated materials. We use a wide band of neutron energies, from 10-8 to 14 MeV in this work (Fig.3 of the manuscript) which produces energetic primary knock-on atoms in the energy range 10-4 to a few MeV (fig.4 of the manuscript) which will create defects with varying local energy deposition. This local annealing can give rise to different defect structures which may be important for tritium diffusion and inventory. Studying the structures and their survival probability is outside the scope of this work.

Acknowledgment

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