

Progress in Materials Analysis for IFE Reactors at the Instituto de Fusión Nuclear

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- **Simulations on SiO₂: procedure and minimum energies for defect formations.**

Materials in Fusion

- Two main options are actually representing the lines of reactor systems in IFE: HYLIFE-II and SOMBRERO, together with the work developed by the ARIES team to arrive to conclusions in the specific components of such reactors.
- The blankets proposed in magnetic reactors are a group of combination of breeder material, coolant and specific structural materials that includes:
- **He or water cooled ceramic breeder, self-cooled Li, LiPb, Flibe blankets and ferritic, vanadium and composites structural materials.**

Materials in Fusion

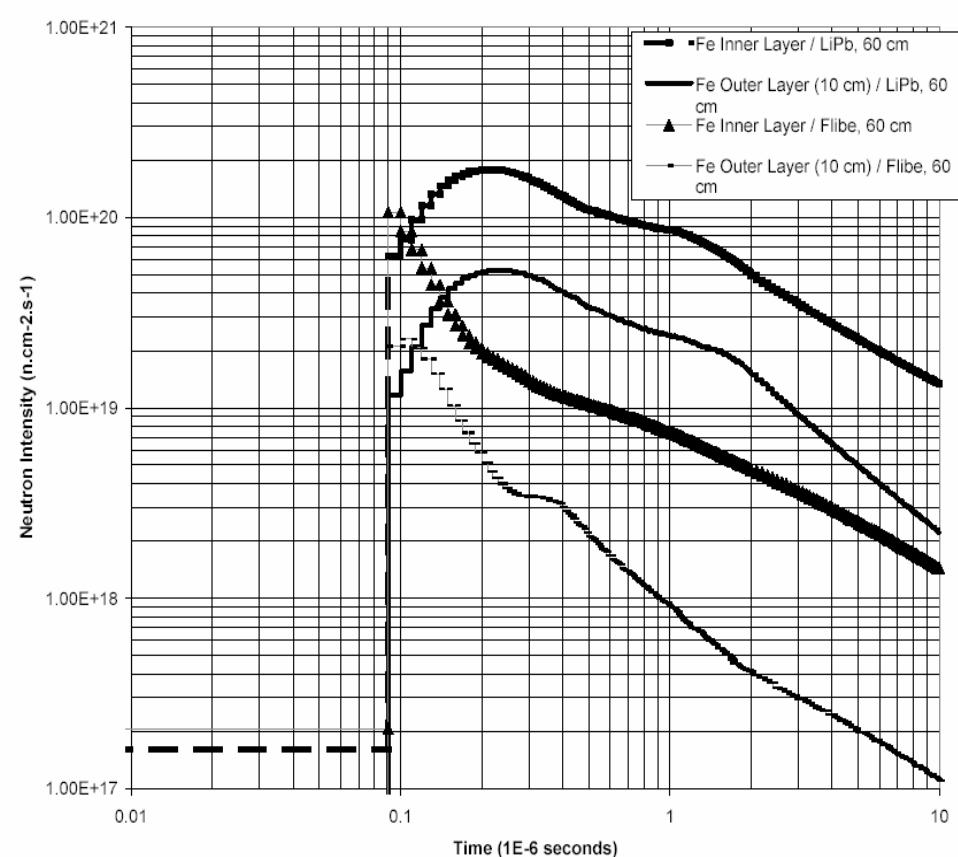
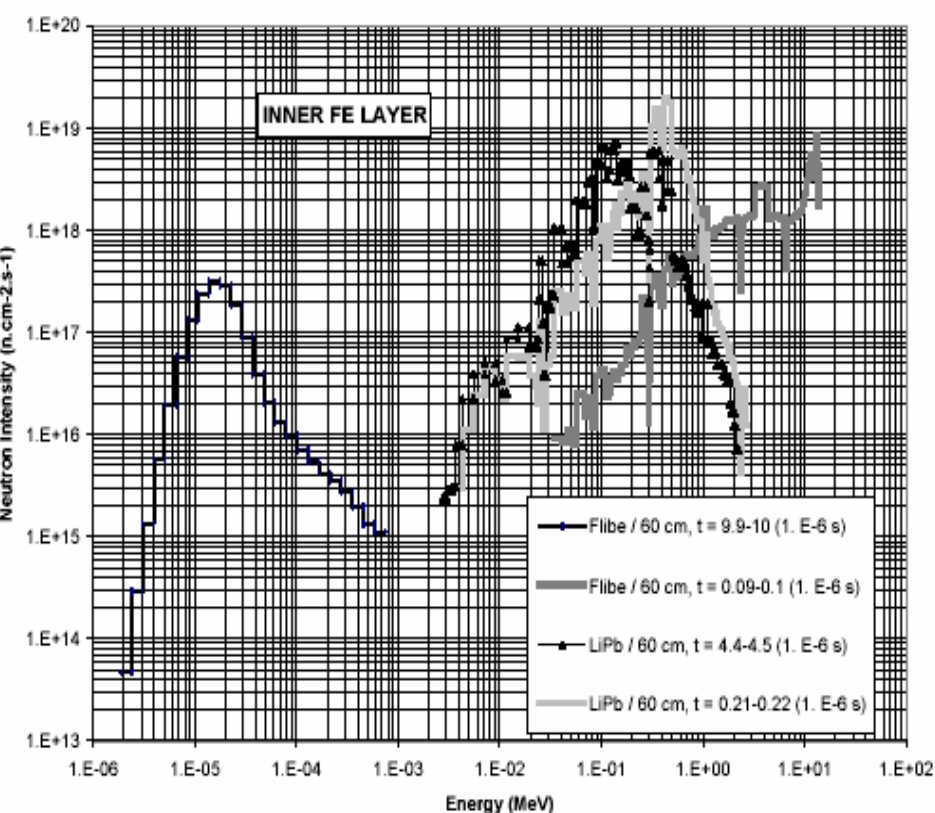
- Reduced-activation Ferritic/martensitic steels are the first option from the point of view of applicability:
F82H (7.5 Cr-2WVTa), JLF-1 (9Cr-2WVTa), EUROFER 97, and additional 7-9 Cr class of steels.
- The effect of **He**, generated by transmutation in fusion environments and producing severe embrittlement and swelling in these steels, is the **main question for final consideration**. Recent results (including molecular dynamics simulations) show that even at 600 appm He concentrations did not enhance embrittlement and hardening (specific experiment up to 80 appm by using B-10).
- It seems that in ion-implantation by Ni experiments the dominant effect came from the Ni itself, when trying to reproduce levels of He
- Limitation of these steels is the available temperature ($\approx 600^{\circ}\text{C}$)
 - $\Rightarrow$ **Oxide Dispersion Steels (ODS)**

Materials in Fusion

- The essential of ODS is the inclusion of **Y_2O_3 particles** (with concentrations 0.3 to 0.5 wt-%) in the range of **nanoscale** (2-3 nm diameter) which act as strong sinks of generated defects by irradiation at the particle-matrix interfaces. Specifically they pin dislocation and they are not dissolved for high temperature, not arriving to recrystallization up to 1373K. It is demonstrated a **gain in yield strength of 50 % which is maintained for high temperatures.**

Materials in Fusion

- Concerning **SiC, and composites from it**, their radiation stability is dominated by the differential swelling between the SiC fiber, that are not fully dense or crystalline, carbon interfaces and β SiC matrix. An increase in the deep/basic understanding of the damage mechanisms in this material is strongly necessary to complement the data bank emerging from experimental results in the fusion programs.
- **Vanadium alloys**, the main candidate is **V-4Cr-4Ti**, and a full description of impurity redistribution and precipitation, deformation and irradiation effects is going.



**Target nominal density 500 g.cm^{-3} . Two areas of density: 170 and 750 g.cm^{-3} .
 Carbon in the target with uniform density.
 4 m radius of chamber. 60 and 100 cm protections of Flibe and LiPb
 1 cm of Fe representing the Structural (Steel) Material.**

Neutron Intensities: First key magnitude to be assessed in time and energy

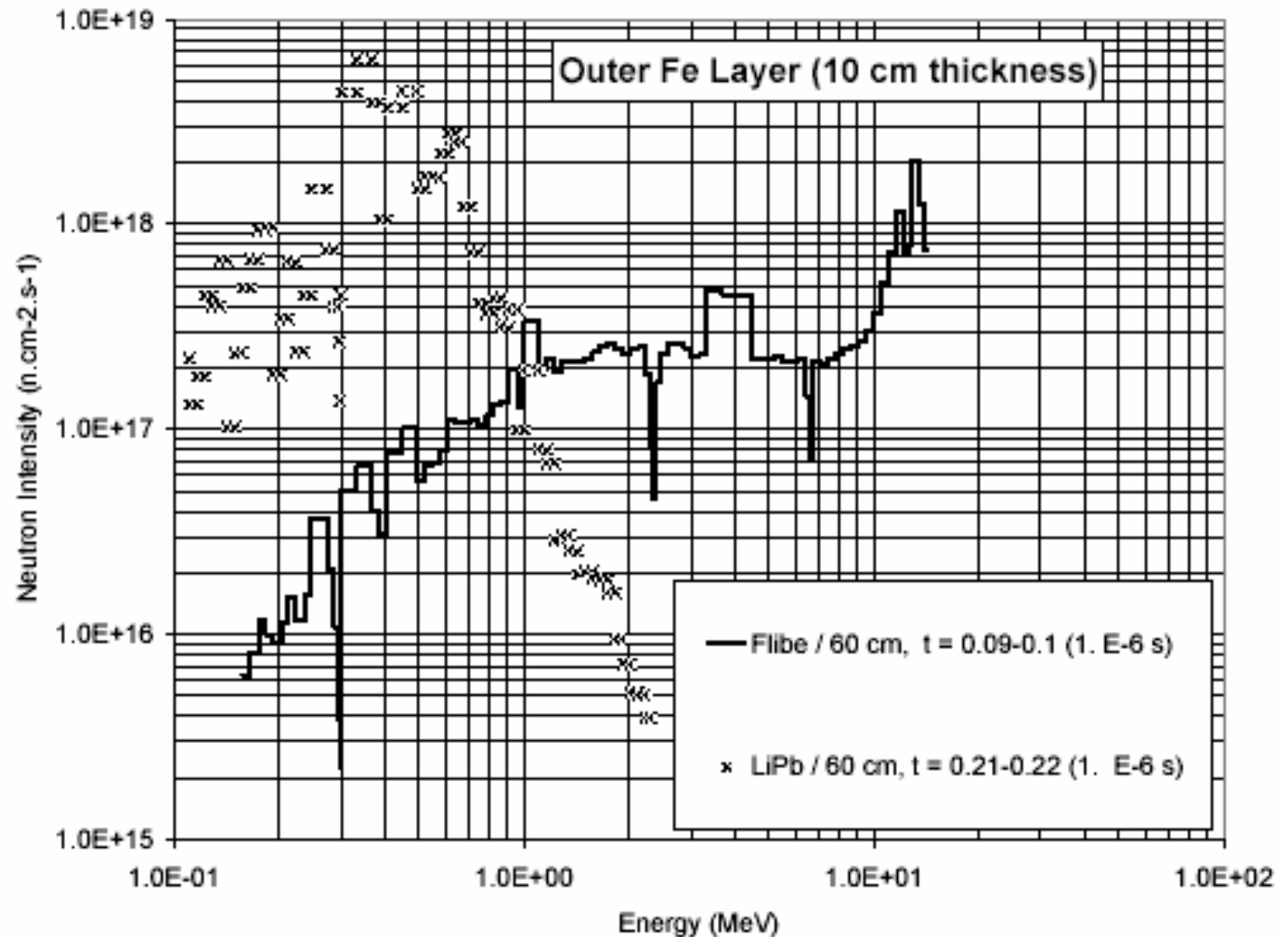
J. M. PERLADO, D. LODI, J. MARIAN, A. GONZÁLEZ PLATA, M. SALVADOR, L. COLOMBO, M.J. CATURLA, T. DÍAZ DE LA RUBIA, "Time-Dependent Neutronics in Structural Materials of Inertial Fusion Reactors and Simulation of Defect Accumulation in Pulsed Fe and SiC", *Fusion Science and Technology*, **43**, 384 (2003).

November
4 - 7, 2003

IAEA RCM on Elements of Power Plant Design for Inertial Fusion Energy, Vienna

Target nominal density 500 g.cm⁻³. Two areas of density: 170 and 750 g.cm⁻³.
 Carbon in the target with uniform density
 4 m radius of chamber 60 and 100 cm protections of Flibe and LiPb.
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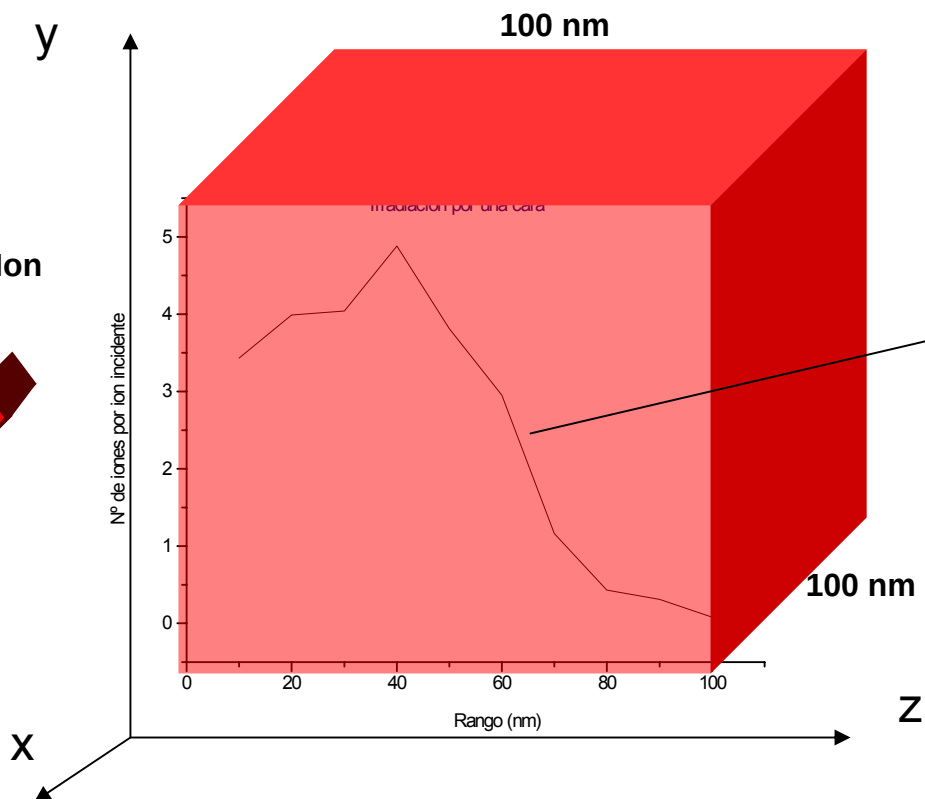
Neutron Intensity





Simulation characteristics

Incoming Ion



Two impurity contents: 5 appm (Ultra High Pure Iron)
100 appm (Pure Iron)

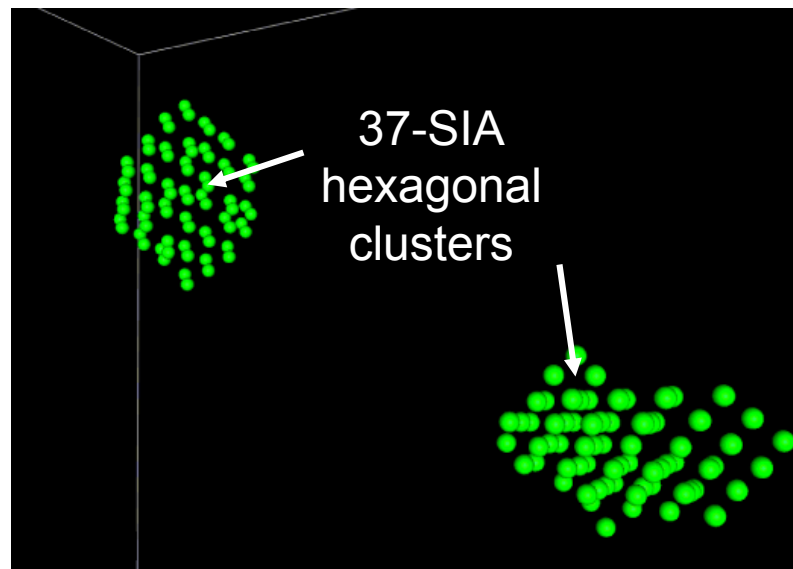
EXPERIMENTS FOR BASIC DATA IN
MULTISCALE MODELING



A new mechanism of interaction of original interstitial loops type $\langle 111 \rangle$ from cascade after irradiation to generate visible $\langle 100 \rangle$ has been discovered and assessed with TEM experiments. It has already implemented in our version of Kinetic Event MonteCarlo - BIGMAC code.

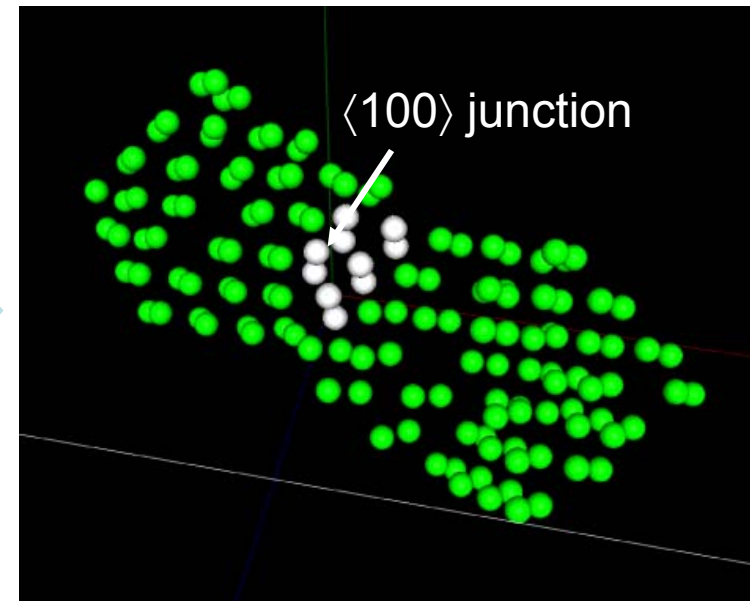
J. MARIAN, B. WIRTH, J.M. PERLADO, "On the Mechanisms of Formation and Growth of $\langle 100 \rangle$ Loops", *Physical Review Letters*, **88**, 25 255507-1/4 (2002)

$$\frac{1}{2} [111] + \frac{1}{2} [1\bar{1}\bar{1}] \rightarrow [100]$$



$\frac{1}{2} [111]$

$\frac{1}{2} [1\bar{1}\bar{1}]$

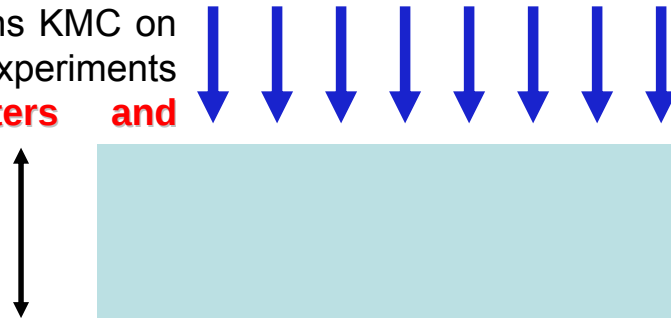


$[100]$

Modeling thin film Fe irradiation with 150keV Fe

Additional Simulations KMC on the same experiments and different parameters and physics of defects

100nm



150 keV Fe

EXPERIMENTS

Input data for MM modeling (KMC) / Conditions of MM simulation

- Cascade database from Roger Stoller (*Oak Ridge National Laboratory*)
Self-interstitial cluster evolution rules from Jaime Marian's MD (PRL **88**,25 (2002) 255507-1/4)

∅ Loops formed in cascade as $\langle 111 \rangle$ mobile

∅ Interaction between two loops:

If resulting BV is $\langle 100 \rangle$ and both > 15 defects $\rightarrow$ $\langle 100 \rangle$ loop

If resulting BV is $\circ \langle 100 \rangle$

$\langle 111 \rangle$ loop

∅ Interaction between loop and Impurities:

$\langle 111 \rangle$ loop + Impurity

$\langle 111 \rangle$ loop

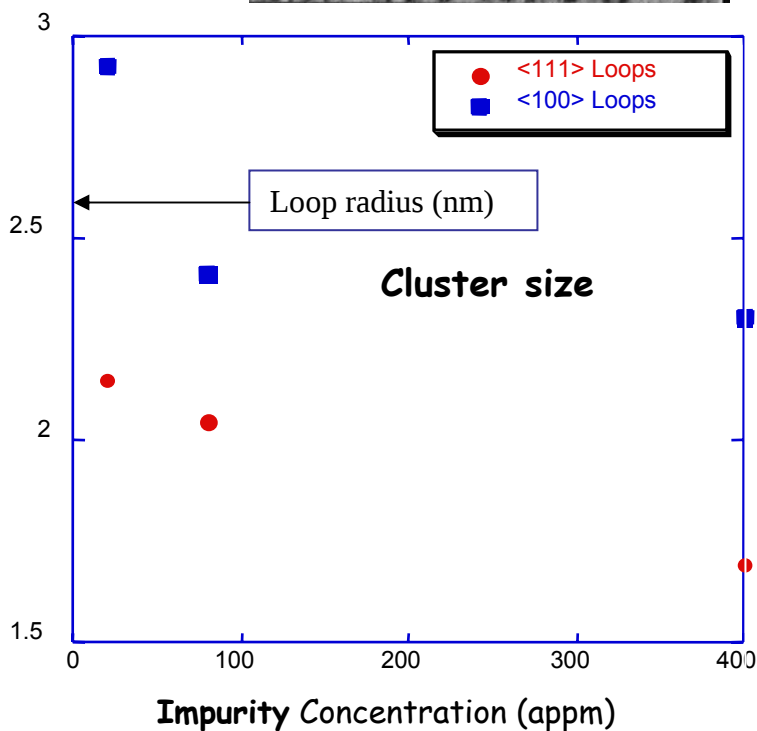
(trapped)

$\langle 111 \rangle$ trapped loop with other loops same rules as in (2)

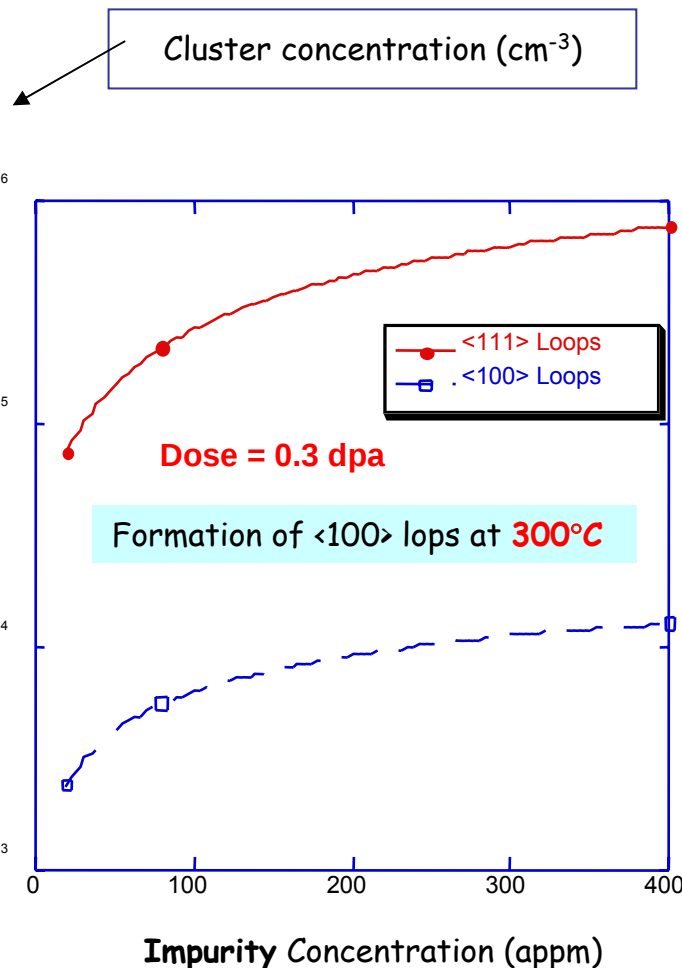
Binding energy $\langle 111 \rangle$ loop and Impurity between 0.4 and 1

150dus keV Fe⁺ Ions; 2 × 10⁻⁴ dpa/s; 1 dpa; T= 300 °C / new analysis from here !!

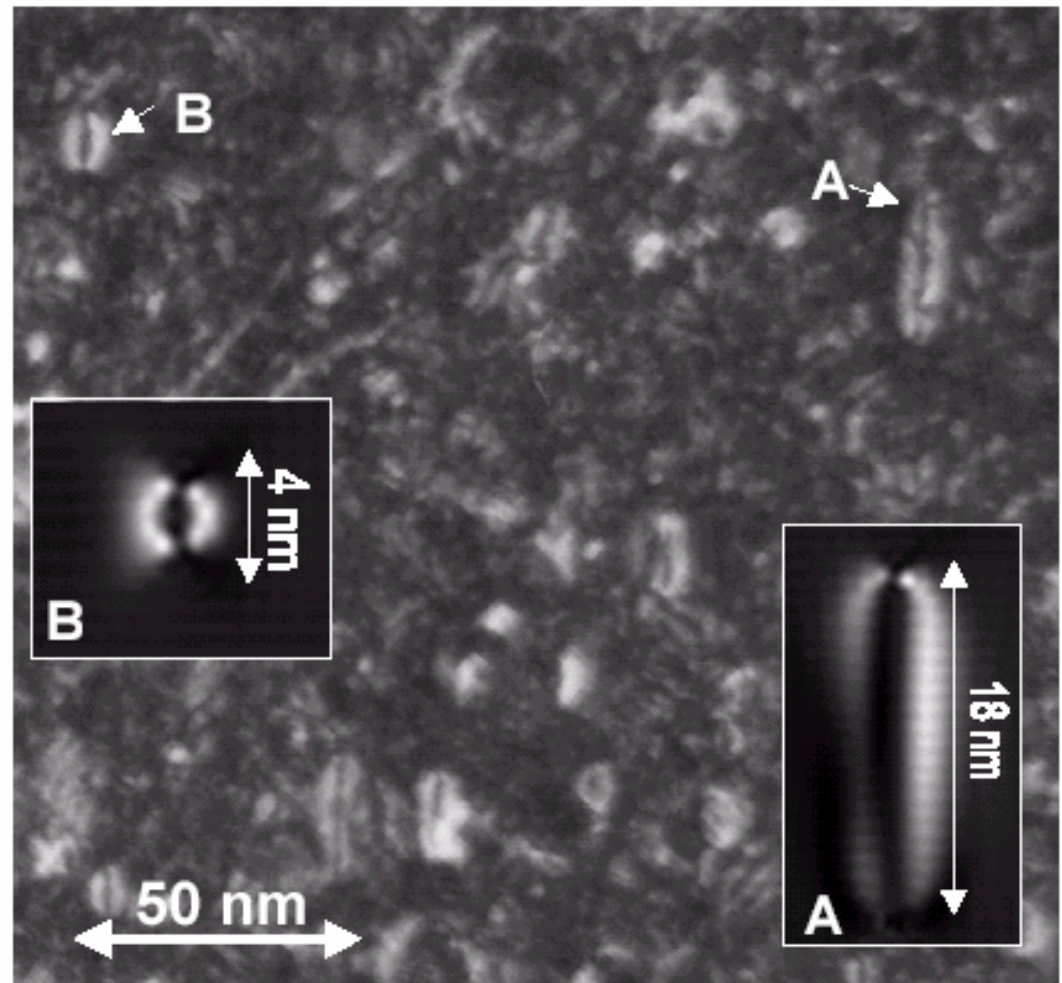
FINAL COMPARISON with TEM Experimental Results (M. Hernández, L.G. Briceño)



- If no impurities all <111> loops at surfaces and no <100> loops
- If binding energy with impurities = 0.4 eV all <111> loops at surfaces.
- <100> loops larger than <111> loops at all impurity concentrations
- Concentration of <100> loops lower than <111> sessile loops

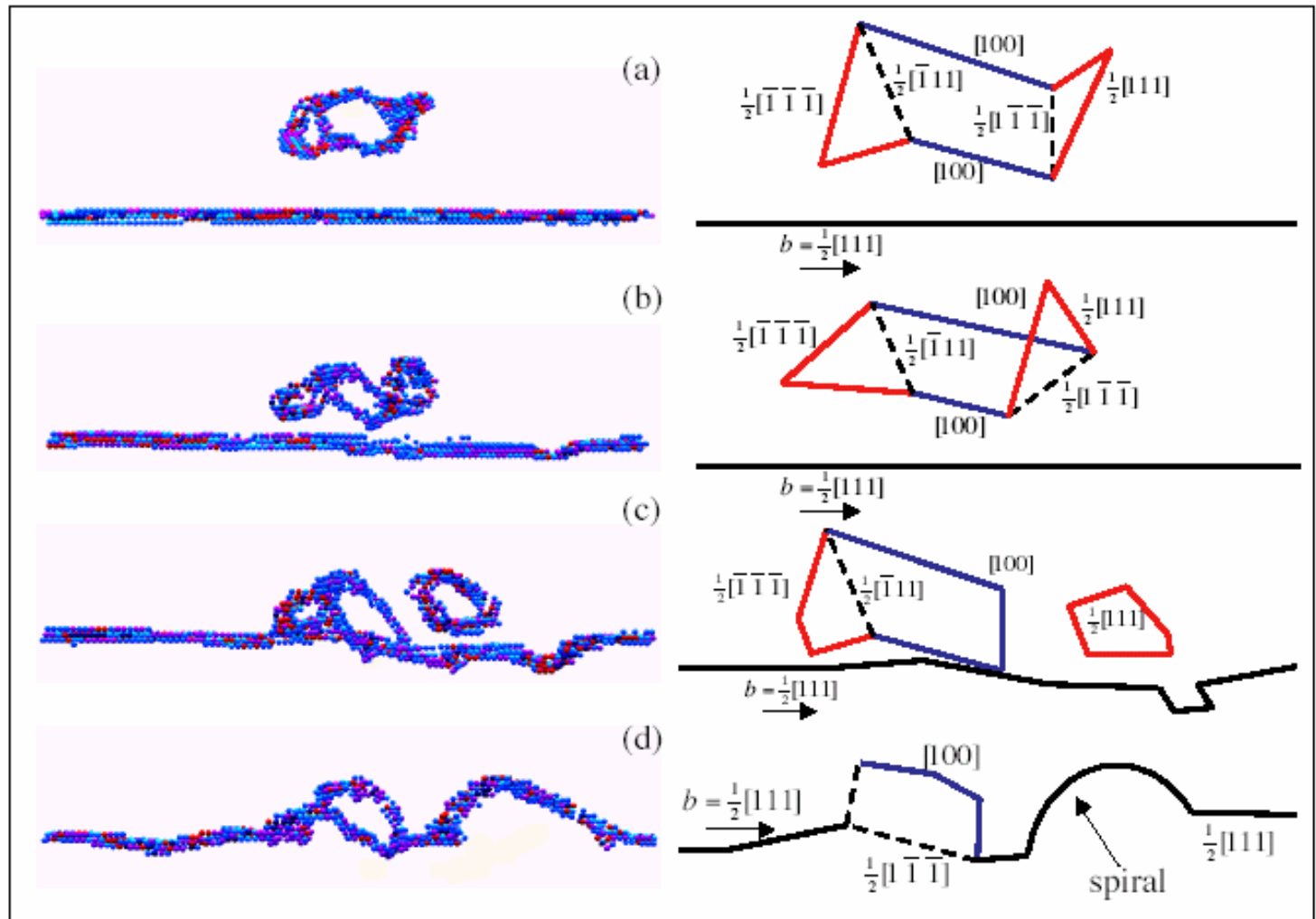


Approaching simulations to observations



Experimental TEM weak beam image of a Fe-9Cr crystal irradiated with neutrons to a dose of 8.8 dpa at 302°C. The two insets represent TEM simulated images of (A) an 18-nm, rectangular [100] loop and (B) a 4-nm, hexagonal, [100] loop. A number of features can be observed in the TEM micrograph, among which two (A and B) interstitial loops with Burgers vector [100], sitting on {100} planes, can be identified. The qualitative agreement with the simulated loops in both contrast and shape is excellent.

Close to
macroscopic
magnitudes

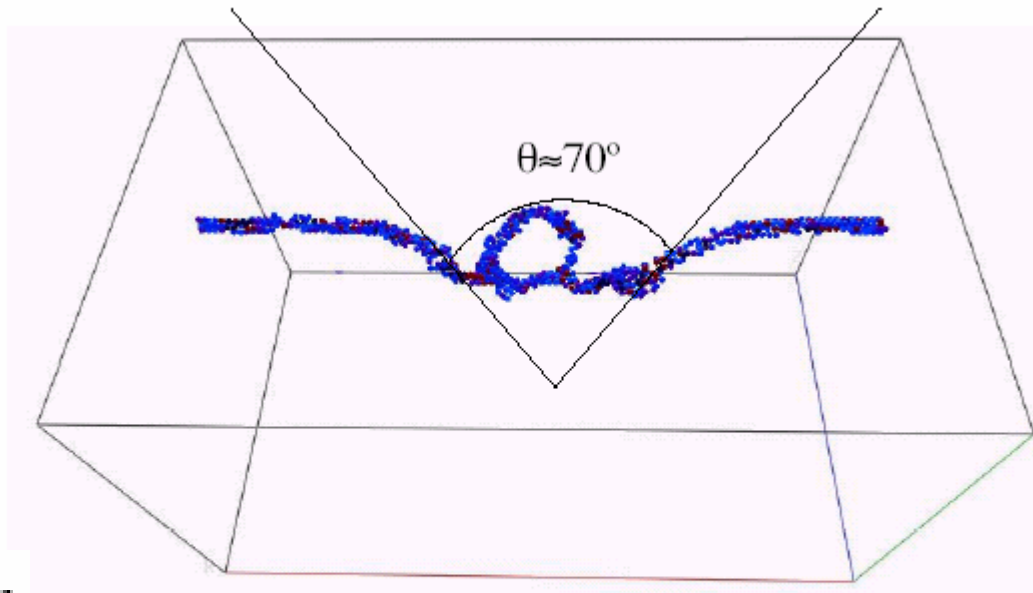


Sequence of MD snapshots (left) and equivalent dislocation interpretation (right) of the interaction between a 113-SIA, rhombic [100] dislocation loop and a $\frac{1}{2} \langle 111 \rangle$ screw dislocation at 100 K and **750 MPa of applied shear stress**. The dislocation and loop cores have been visualized using the centro-symmetry deviation parameter.

Orowan Theory
continuum

$$\Delta\tau_s = \alpha Gb(Nd)^{1/2}$$

$$\sigma_{||} = T \tau_s$$



$$\theta \approx 70^\circ$$

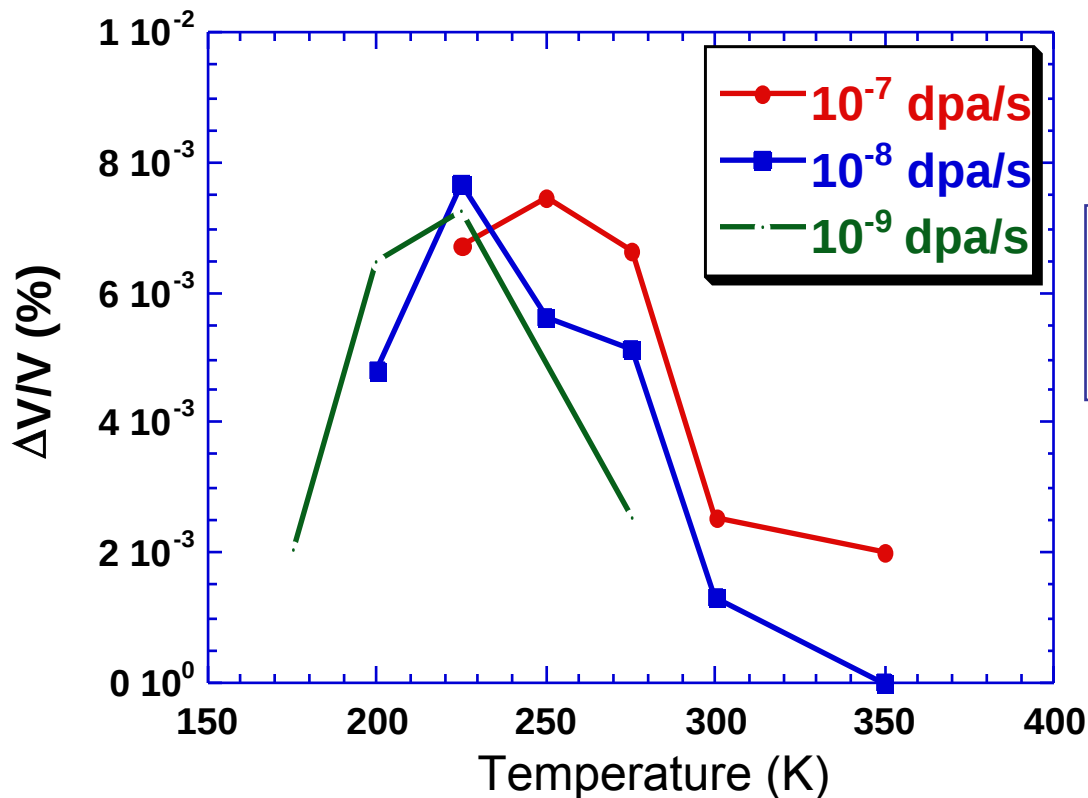
$$\cos\left(\frac{\theta}{2}\right) = \alpha \approx 0.8$$

Last snapshot before the dislocation breaks through the obstacle, leaving a [100] loop behind and two heavily-curved spirals along the dislocation line. The critical angle is $\theta=70^\circ$, necessary to estimate the obstacle strength, $\cos(\theta/2) = \alpha = 0.8$.

$N = 1.2 \times 10^{23} \text{ m}^{-3}$ (simulations) $d = 2 \text{ nm}$; $G \approx 75 \text{ MPa} \Rightarrow \tau_s \approx 200 \text{ MPa} \Rightarrow \sigma_u \approx 590 \text{ MPa} \Rightarrow$

50 % from experiments

Dose Rate dependence of void swelling can be extracted from kMC simulations: other key effect in Fusion from MM



Change in Volume due to He-V clusters at 0.03 dpa as a function of dose rate and temperature

Formulation:

$$(\Delta V/V(\phi))_{\text{voids}} = C_V^{\text{void}(n)}(\phi) \times \Omega_r(n)$$

From MM (KMC) calculations:

C_V^{voids} = concentration of Vacancies in Voids

Ω_r = relaxation volume of a Vacancy in a Void

n = number of defects in Void

versus dose ϕ

Characteristic swelling curve for FCC metals is obtained from kMC

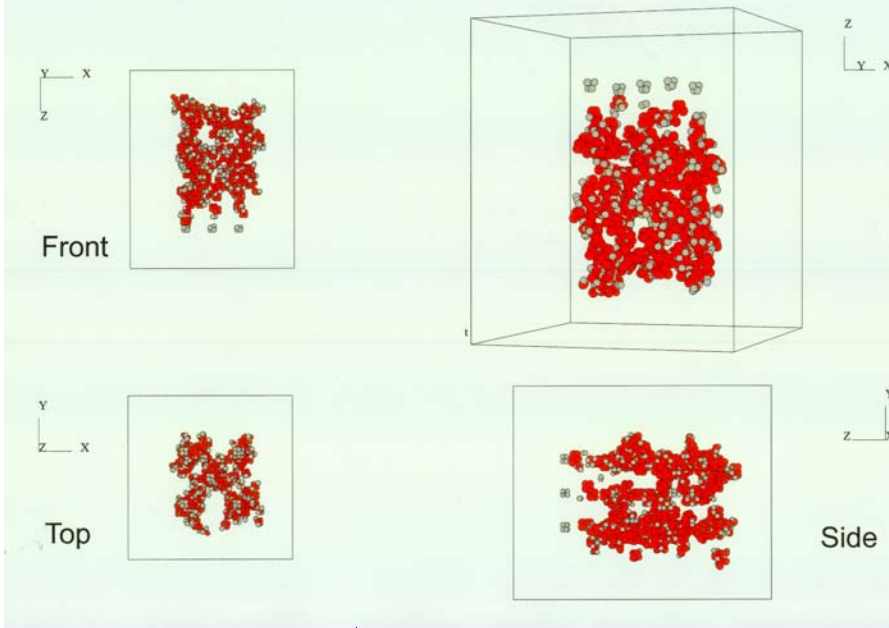
Mechanisms clearly envisioned from MM: Swelling peak shift with

dose rate

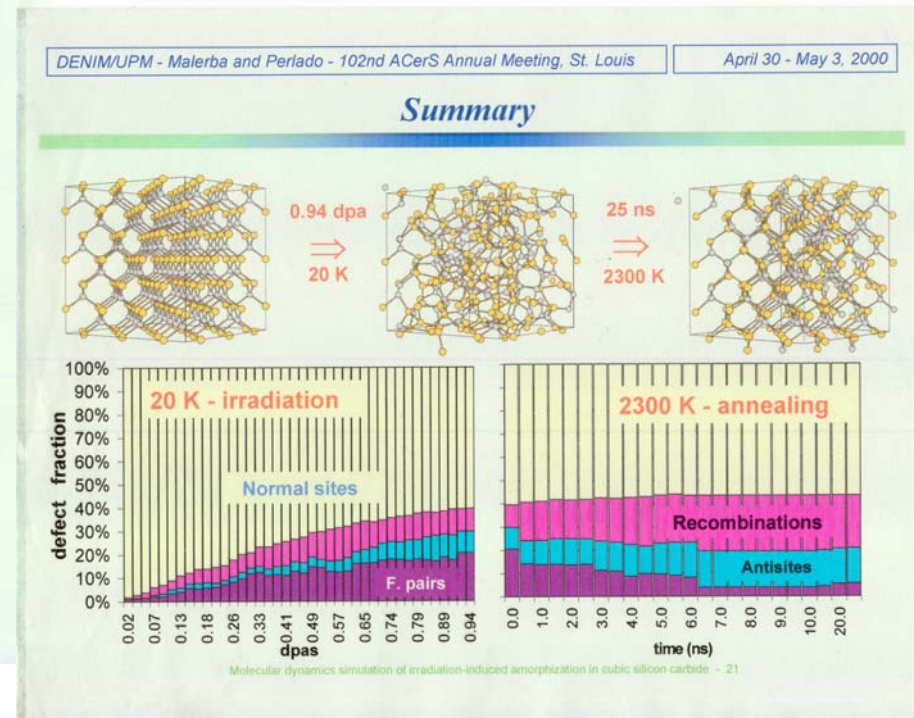
SiC

Work using Molecular Dynamics: identifying mechanisms but **what more on defect diffusion ????**

Accumulation of 25 1 keV Si-recoils at 1300 K - Excited Atoms



J.M. Perlado, L. Malerba, T. Díaz de la Rubia,
Fusion Technology, **34**, n3, part 2 (1998) 840-
847



Molecular dynamics simulation of irradiation-induced amorphization of cubic silicon carbide.
L. Malerba, J.M. Perlado
Journal of Nuclear Materials **289** (2001) 57-70.

Table 1.- Comparison of different potentials for defects energetic calculations.

Energy	Value (eV)				
	Pearson <i>et al</i> ¹	MEAM ²	Tersoff I ³	Tersoff II ⁴	First Principles ⁵
E _f (V _{Si})	6.70	14.28	12.46	12.92	12.60
E _f (V _C)	5.26	9.58	11.61	11.58	11.70
E _f (I _{Si} T _C)	3.23	5.96	4.08	4.09	8.00
E _f (I _C T _C)	3.73	3.07	2.29	2.38	4.40
E _f (I _{Si} T _{Si})	10.55	2.09	8.00	7.90	8.30
E _f (I _C T _{Si})	3.80	3.04	0.23	1.18	1.90
E _f (Si in C antisite)	3.66	7.21	5.55	5.50	6.40
E _f (C in Si antisite)	2.33	2.17	0.61	0.35	0.20

J.M. Perlado, J. Marian *et al*, Mathematics and Computation Reactor Physics and Environmental Analysis in Nuclear Applications, vol. 2, (pag. 1151-1162) J.M. Aragonés, C. Ahnert, O. Cabellos (Eds.), Senda Editorial S.A., ISBN: 84-699-0943-6 (1999).

¹ Pearson E, Takai T, Halicioglu T, Tiller W 1984 *J. Cryst. Growth* **70** 33-40

² Huang H, Ghoniem N M, Wong J K, Baskes M I 1995 *Modelling Simul. Mater. Sci Eng.* **3** 615-627

³ Huang H, Ghoniem N M, Wong J K, Baskes M I 1995 *Modelling Simul. Mater. Sci Eng.* **3** 615-627

⁴ DENIM

⁵ Wang C, Bernholc J, Davis R 1988 *Phys. Rev. B* **38** 12752-12755; Bernholc J, Kajihara S, Wang C, Antonelli A, Davis R 1992 *Mater. Sci. Eng. B* **11** 265-272

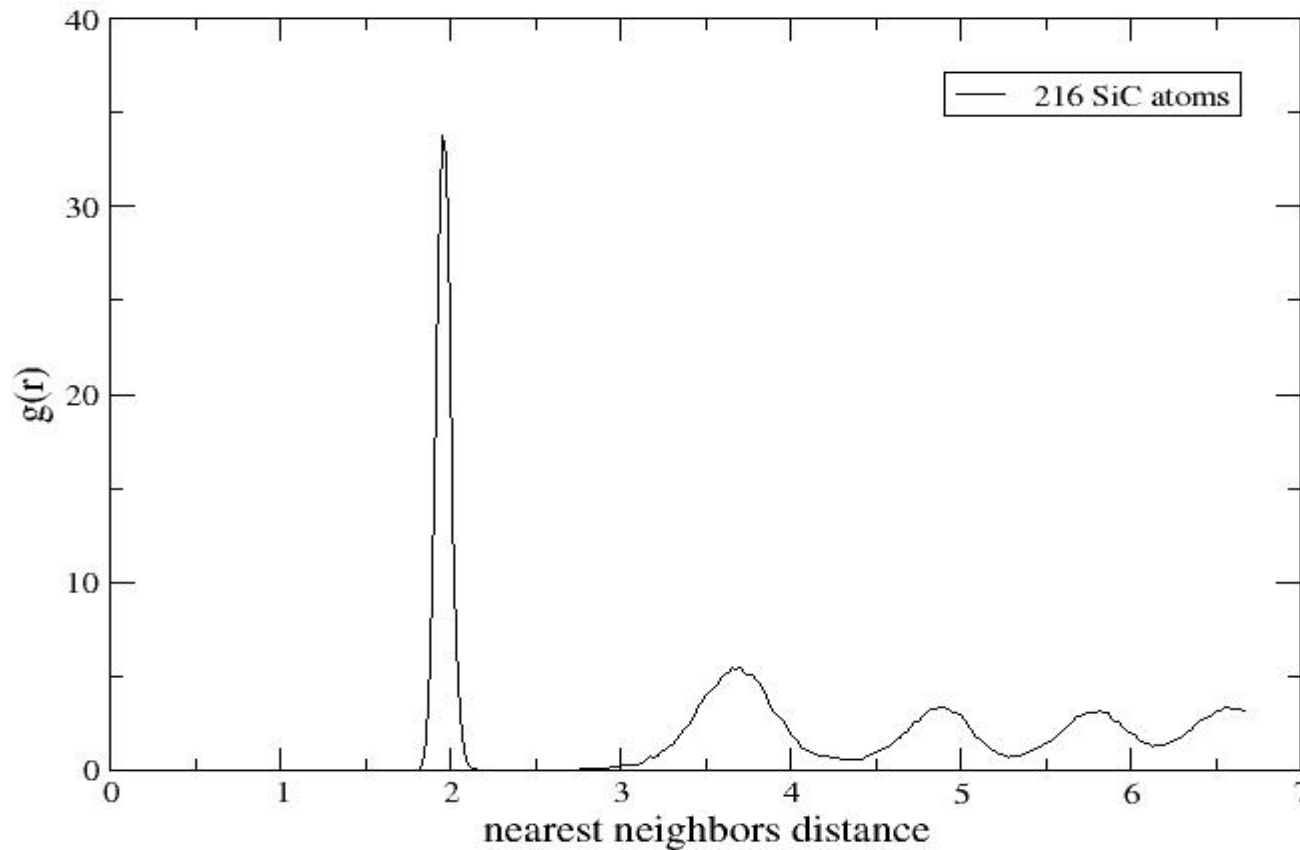
Tight binding silicon carbide code TBSiC

Our tight binding model uses a quantum resolution equations to obtain the band structure energy and the Hellmann-Feynman contribution forces.

The semi-empirical character of our model is localized in the **repulsive potential equation**. We establish a weighted average due to the different atomic species in the β -SiC crystal.

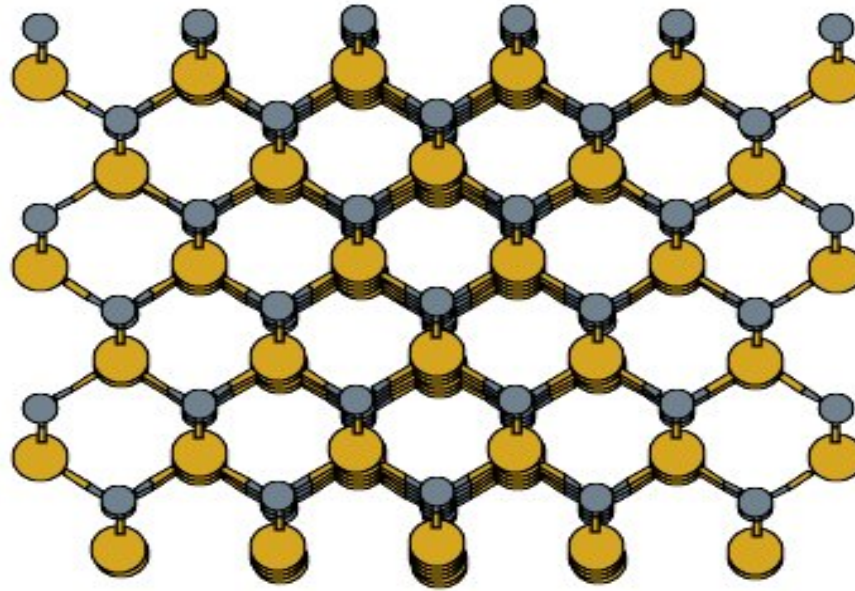
Each new weighted average applied in the calibration of our tight binding repulsive potential force, was tested to obtain: **accurate total cohesive energy in the silicon carbide simulation box with its respectively lattice parameter.**

Pair correlation function



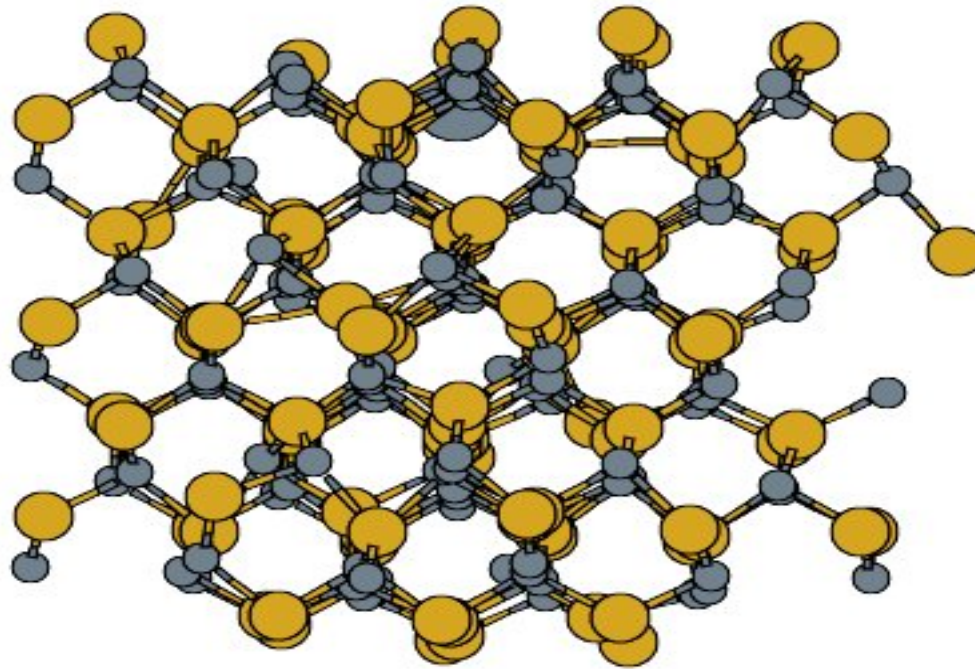
Tight
Binding
MD
code
for
SiC

We can manage different quantities of atoms in our simulations, in the graphic, we show the pair correlation function into a SiC crystal with 216 atoms, and we can see the nearest neighbor distances which are correctly assessed to theory and experiments



Tight
Binding
MD
code
for
SiC

We can simulate diverse studies at different temperatures, the number maximum of atoms in our simulations is 1000, here we show you a perfect crystal of SiC, 216 atoms. With our new parameterization in the coefficients into the repulsive term.

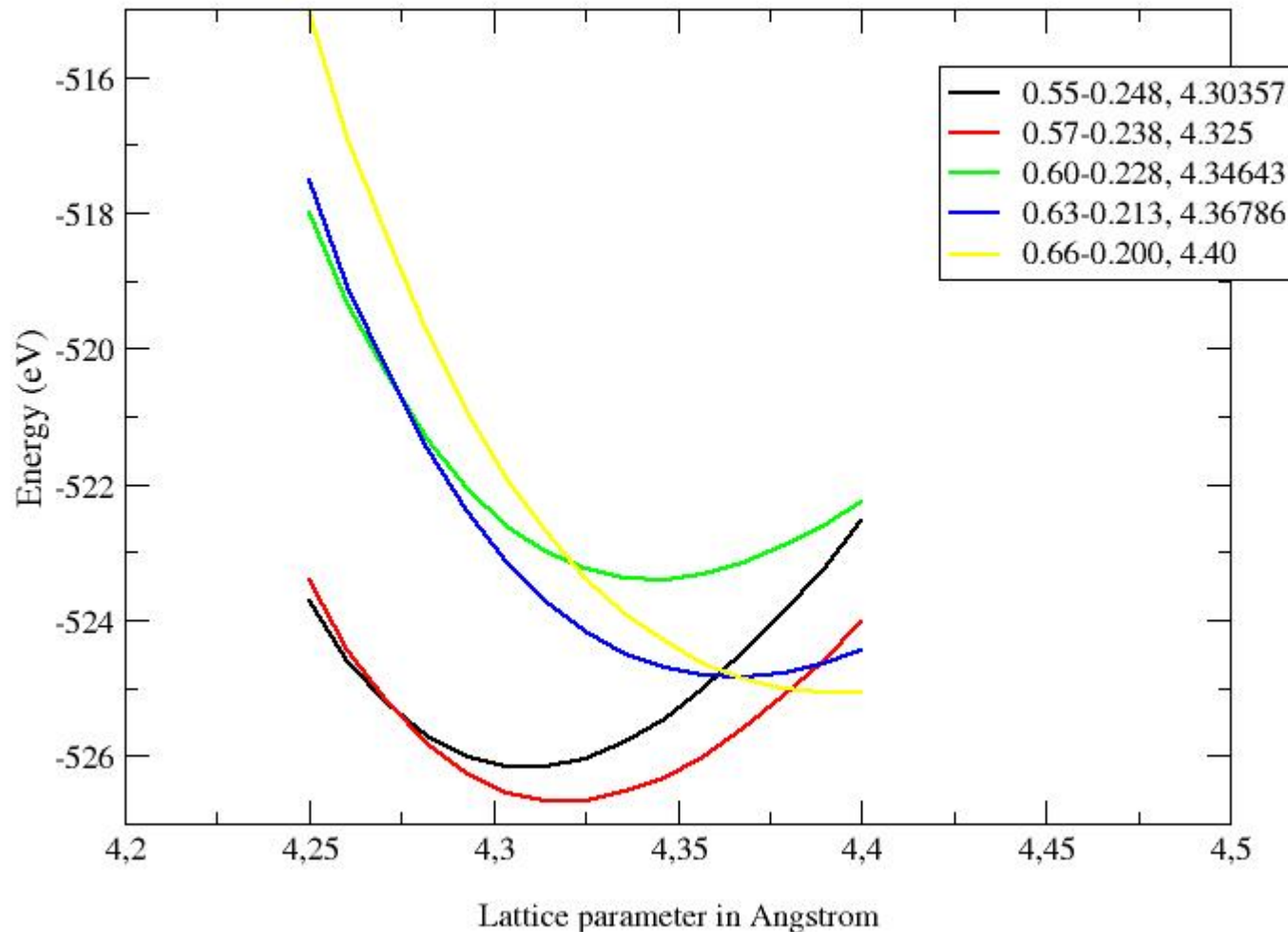


Tight
Binding
MD
code
for
SiC

This result, was obtained after a canonical (NVT) simulation, we find that the crystalline structure exist at high temperatures 1600 K, this configuration was obtained in 8000 time steps NVT simulation

Adjusting coefficients for right cohesive energy

Cohesive total energy in a SiC crystal



We use the damped dynamics technique to find, in one time-step simulation, the minimum cohesive energy value at different values of the lattice parameter silicon carbide box.

For the energetics native defects:

Carbon and Silicon vacancies, Carbon and Silicon Interstitials, Carbon and Silicon antisites

we use considerations of carbon rich and silicon rich conditions, and chemical potential formula for averaging cell conditions.

We use for the defects energetics results the weighted average of carbon and silicon: 0.50 and 0.28 respectively and lattice parameter of 4.295 Å with a $\Delta H = -0.45832$ eV.

Carbon rich analysis

We use for the next defects energetics results the weighted average to carbon and silicon: 0.50

and 0.28 respectively and lattice parameter of 4.295 Å , with a $\Delta H = -0.45832$ eV.

Defect	Ab-initio VASP	Tight Binding	Tersoff
A _C	2.65	2.1522	1.8791
A _{Si}	5.06	4.5515	4.8349
V _C	4.06	5.2326	4.0463
V _{Si}	8.06	5.3040	3.1347
C _{TC}	9.90	4.9656	7.0503
C _{TSi}	9.50	3.9532	4.2366
Si _{TC}	7.34	6.8478	17.8458
Si _{TSi}	9.45	7.5426	16.0587

Silicon rich analysis

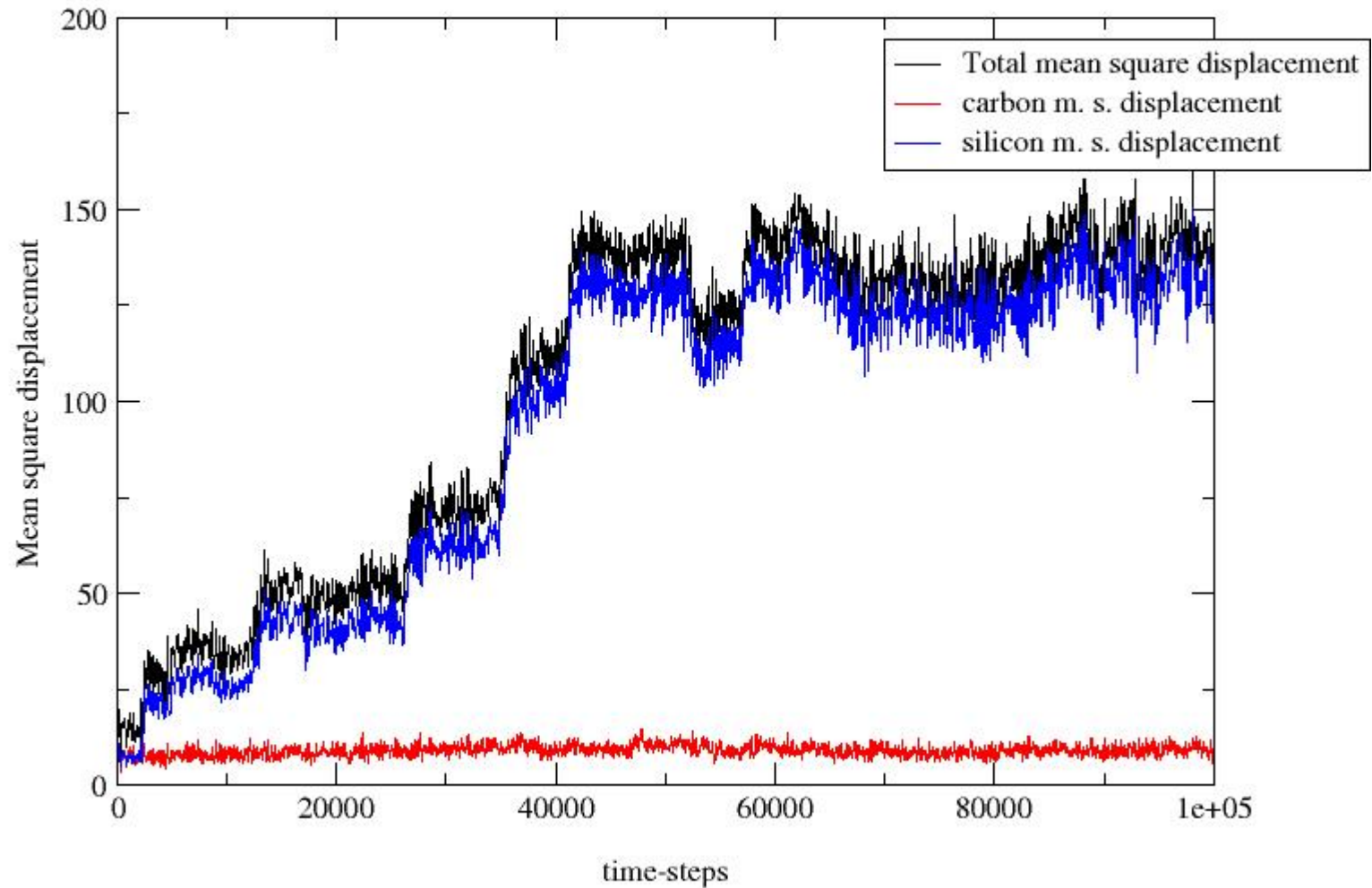
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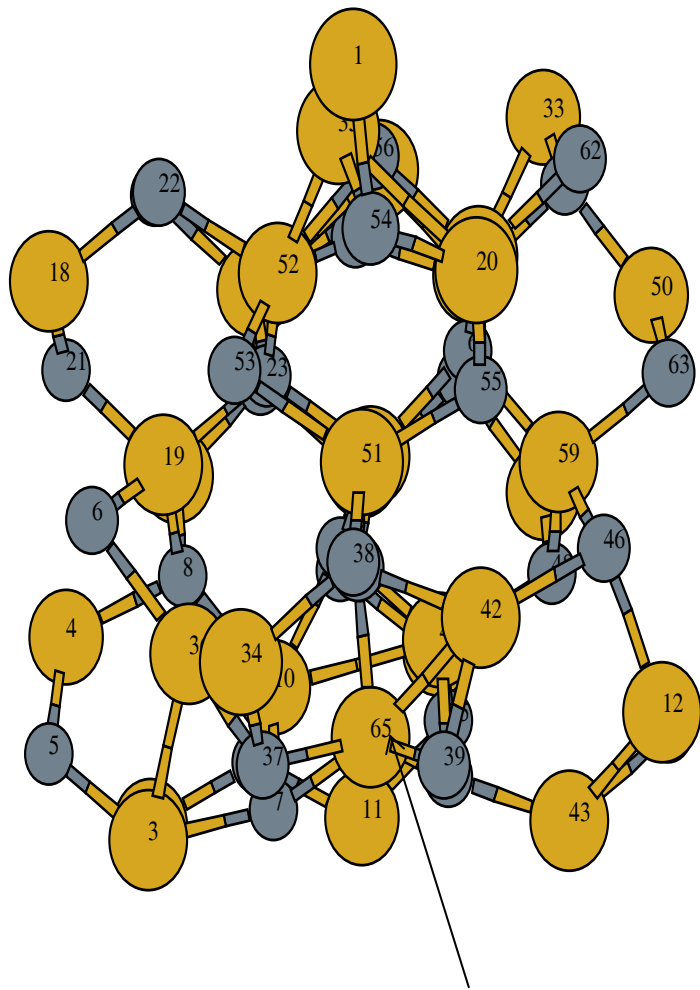
Defect	Ab-initio VASP	Tight Binding	Tersoff
A _C	3.91	3.1916	2.5235
A _{Si}	3.79	3.5121	4.1905
V _C	3.42	4.7130	3.7241
V _{Si}	8.70	5.8237	3.4569
C _{TC}	10.53	5.4852	7.3725
C _{TSi}	10.13	4.4728	4.5588
Si _{TC}	6.70	6.3281	17.5235
Si _{TSi}	8.81	7.0229	15.7365

We observed in this simulation silicon carbide sample, at 2000 K, the *kick-out* phenomena.

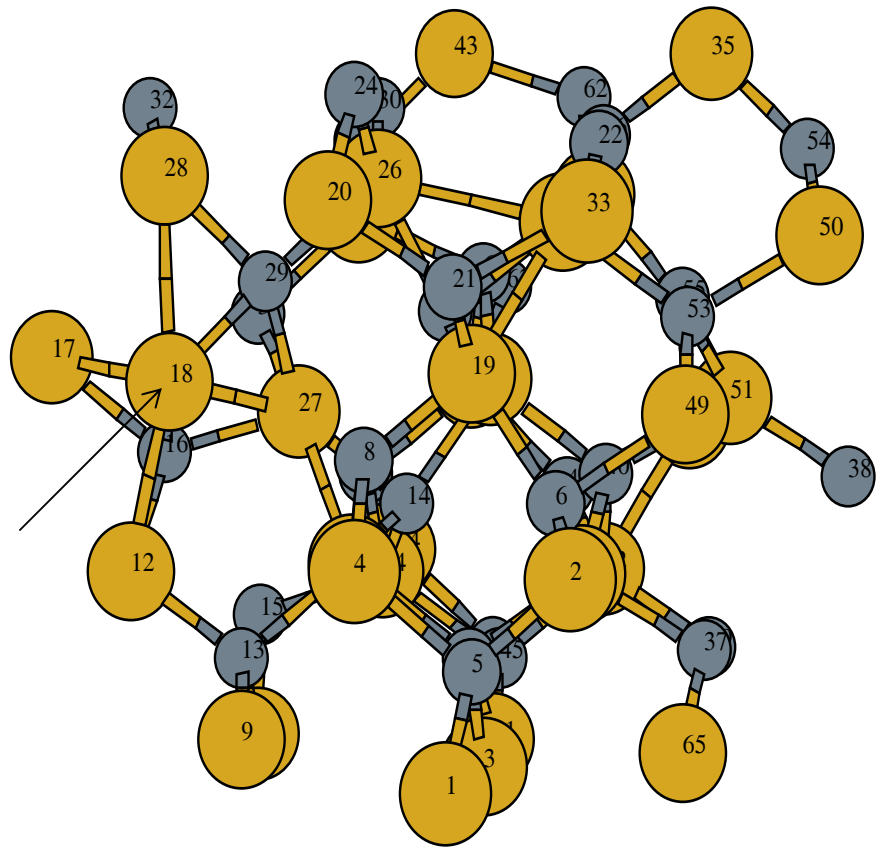
This phenomena occurs when into the SiC crystal, the silicon self-interstitial implanted defect (65 number atom), *kick-out* another silicon atom (18 number atom) into the silicon carbide box, and the initial interstitial would be established into the place of the ejected atom, like its shown in the next figures.

Kick-out phenomena. Si self-interstitial. 2000K





Initial configuration



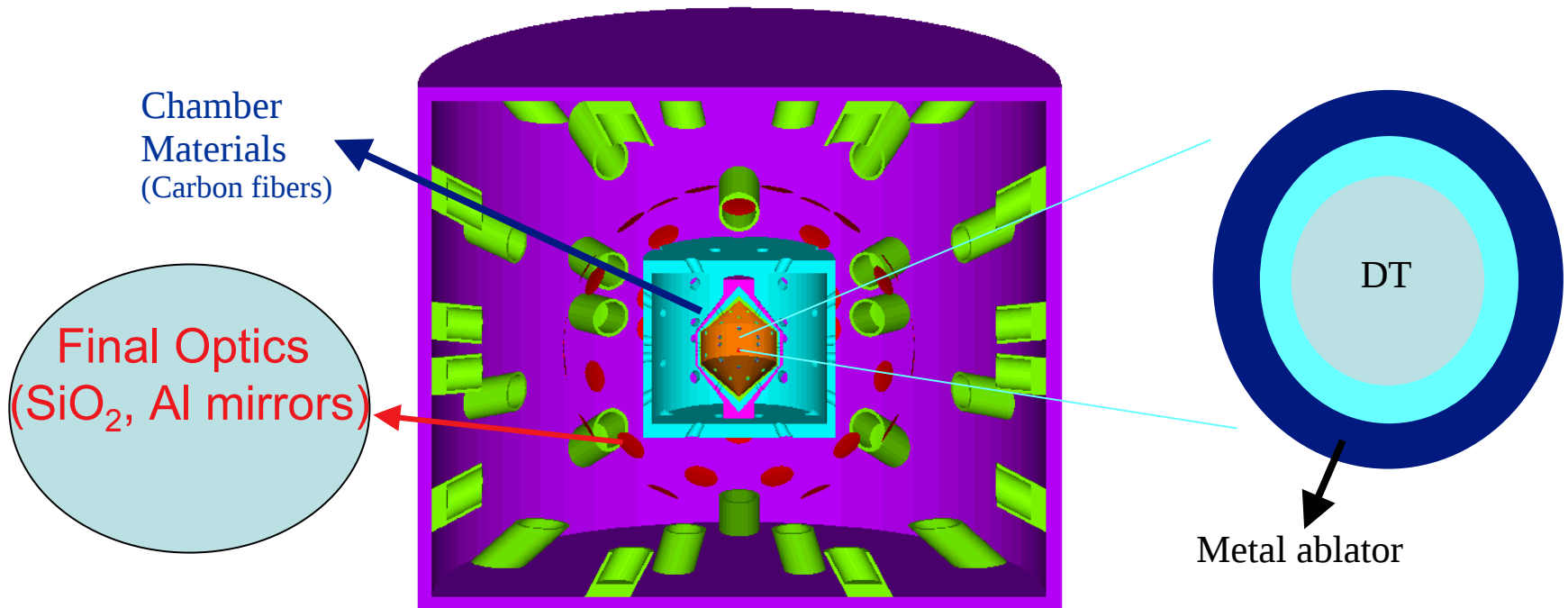
Final configuration

The knowledge of the migration defects into one material like the silicon carbide at different temperatures, let us determine the diffusion coefficients $D(T)$ by the Einstein relation:

$$D(T) = \lim_{t \rightarrow \infty} \frac{N_{at}}{\sum_{\alpha=1} \frac{[(R\alpha(t) - R\alpha(t=0))]^2}{6t}}$$

Laser or ions used to focus energy on a small DT target

A conceptual model: the SOMBRERO reactor



Courtesy of S. Reyes

Materials from metals to ceramics will be exposed to high energy particles (14MeV neutrons) and high doses

What do we know from experiments about the behaviour of fused silica under irradiation?"

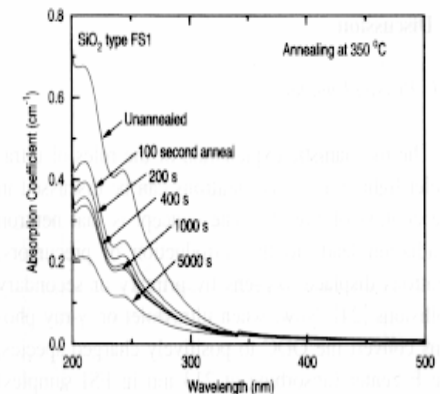
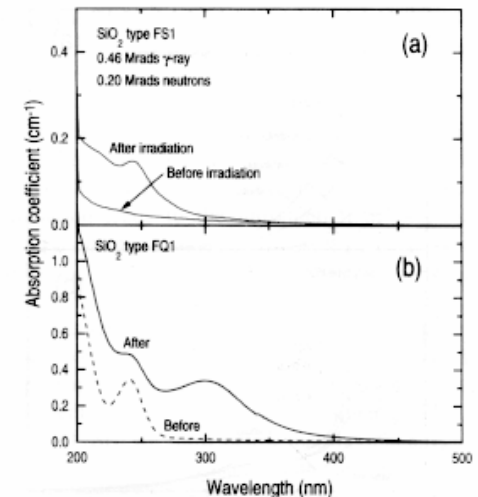
Neutron irradiation can induce obscuration of the optics through color centers

Spectroscopic observations show increase in defect densities (NBOHC, ODC, E') with MeV neutron irradiation.

These defect concentrations are shown to decrease with annealing, though the annealing mechanism is not well understood.

There are some suggestions that cascade overlap can also contribute to reduced defect densities

Additional details on more recent experiments appears in J.Latkowski, A. Kubota, M.J. Caturla, S.N. Dixit, J.A. Speth, S.A. Payne, Fused Silica Final Optics for Inertial Fusion Energy: radiation studies and system-level analysis, Fusion Science and Technology (2003)

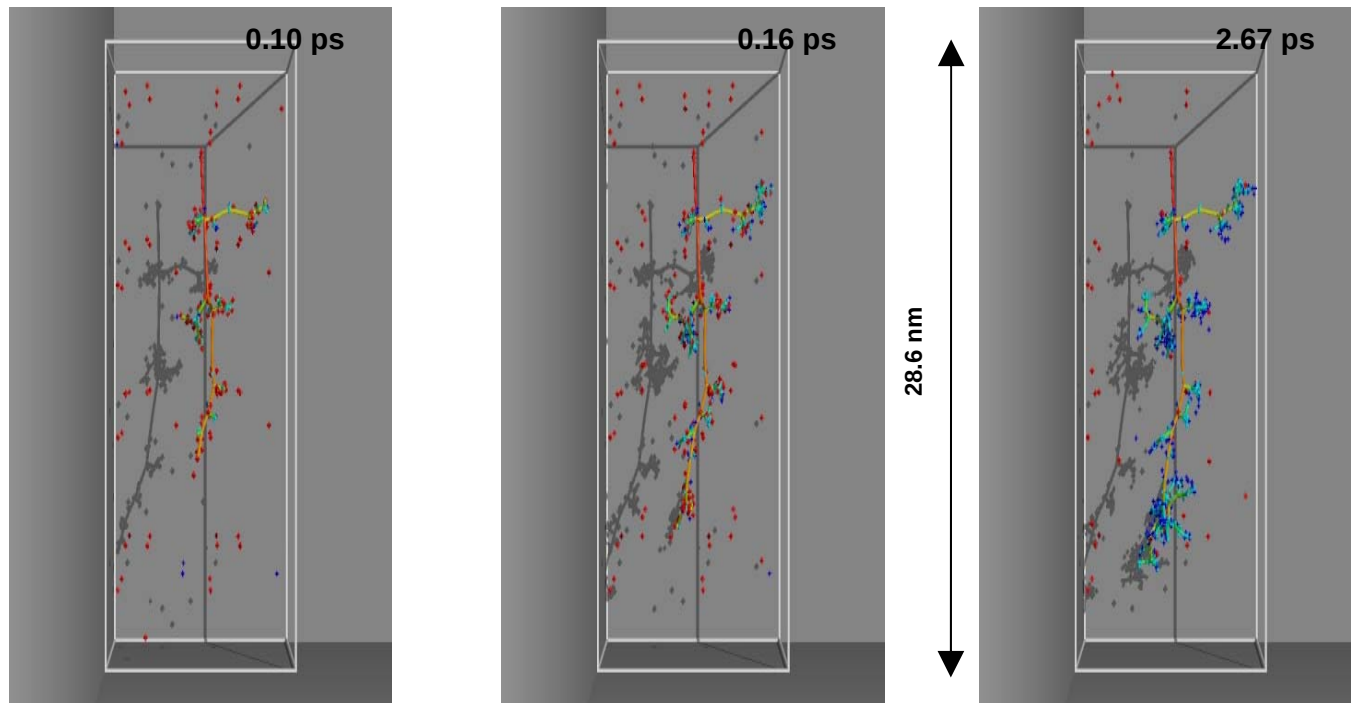


C. D. Marshall, J. A. Speth, S. A. Payne, Non-Crystalline Solids, 212 (1997) 59

Previous work in Multiscale Modeling looking for cascade effects

Attention with statistic !!

**Modeling damage in Ceramics:
5 keV recoil in fused silica**



Large production of Oxygen Deficient Centers produced along the cascade tracks during the cascade. Residual defects observed after the cascade.

New Challenges: amorphous material, 2-component (Oxygen-Silicon), implications of defect production in changes in optical properties

Interatomic potential for fused silica

Molecular Dynamics for processes far-from-equilibrium, with atomic-scale detail. MD involves the integration of Newton's Equation,

$$dx_i^2/dt^2 = -\nabla_i V(r_1, \dots, r_n)$$

with $V(r_1, \dots, r_n)$ taken as modified Born-Mayer-Huggins potentials of Garofalini for Si-O systems,

$$V_{ij}^2 = A_{ij} \exp(-r_{ij}/\rho_{ij}) + Z_i Z_j / r_{ij} \operatorname{erfc}(r_{ij}/\beta_{ij})$$

Dispersion Term Screened Coulomb Term

V_{ijk}^3 = Si-O-Si and O-Si-O Bond-Angle-Dependent Term

The Garofalini Potentials have been used in numerous studies examining the bulk, surface and interfacial properties of fused silica.

Simulations run with MDCASK LLNL software on ASCI White

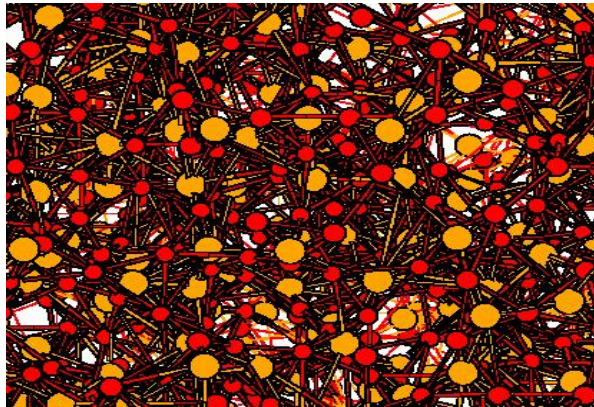
DENIM objectives

- Minimum energy to produce a defect in fused silica
- Neutron and Gamma Radiation Damage produced by PKA energies between 1 and 20 keV with a large statistics.... **New types of defects ????????**
- Recovery of damage through annealing

Basic Understanding: What is fused silica?

☒ The Structure of SiO_2

- Basic Building Unit: $(\text{SiO}_4)^{-4}$ Tetrahedron
4-fold Coordinated Si Atoms, 2-fold Coordinated O Atoms
- Fully Connected 3D Network of Corner-Shared $(\text{SiO}_4)^{-4}$ Tetrahedra

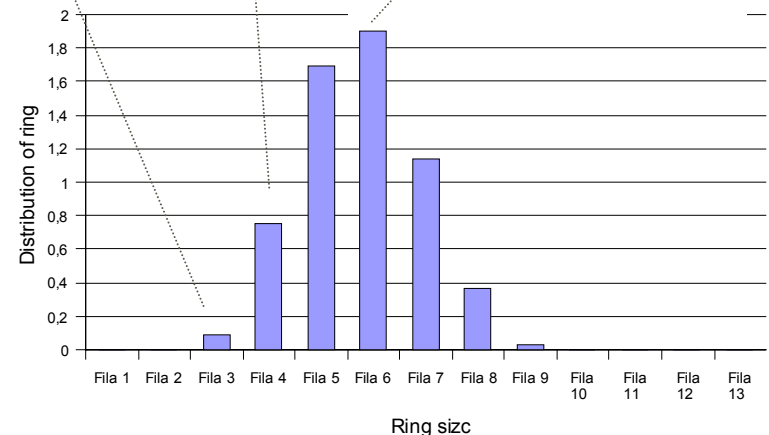
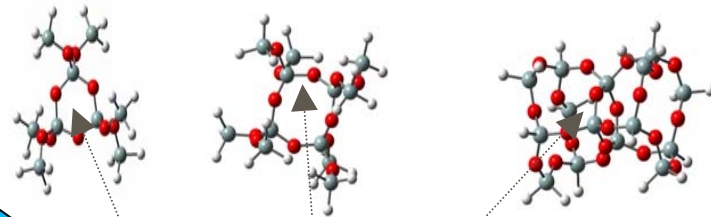


Amorphous silica structure

Note that this amorphous state presents in any case some order of structure that will make the analysis under irradiation more visible.

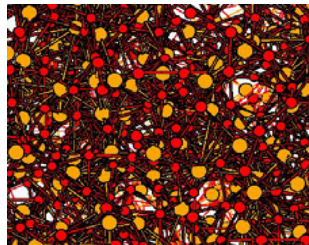
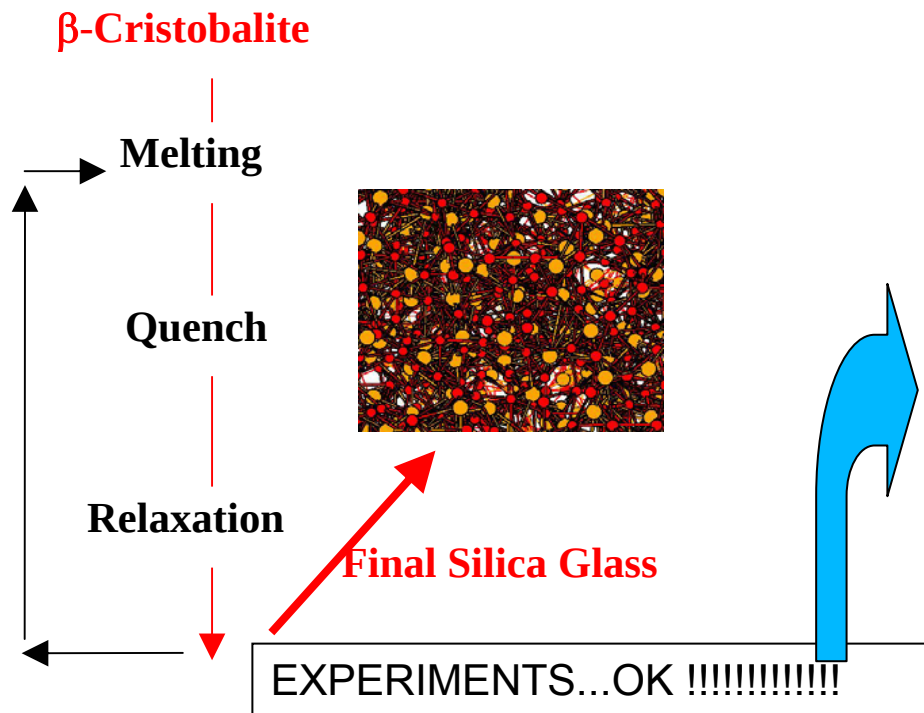
• Ring Structures

- Average 6-Member Ring (Loop)

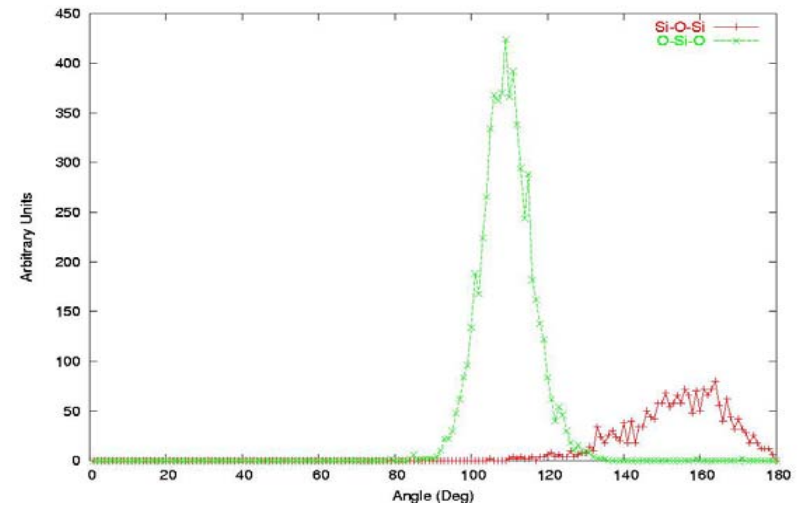


How do we generate silica glass with MD?

- Beta Cristobalite was melted at 7000K for 25 psec
- Temperature stepped down at 1000K increments and run for 25 psec, until T=1000K.
- T=300K, periodic boundary condition released in one direction. Run for 25 psec.



Bond angle distribution



Bond Angle

	Model	Exp.
Si-O-Si Peak	158 °	160°
Si-OSi FWHM	32	38
O-Si-O Peak	109°	109°
O-Si-O FWHM	12	14

Problems that need to be solved

- Identification of defects: *not as simple as in perfect lattices*
- High variations in the number of defects due to temperature fluctuations
- Large simulation boxes are needed (long range of ions)
- Possible solutions: new codes to identify defects / and calculate defects through average of positions.

Approach of the problem

- We have created codes to identify defect. These codes identify defects in two different forms, calculating the coordination of the atoms and calculating whatever they have been displaced from their positions.
- In order to reduce the time of computational processing, the codes use a box of atoms smaller than the total box. The size of the box is the maximum distance that crosses the PKA more three units of cell.
- MDCASK code has been modified, to make the average of the positions of atoms each a certain number of steps thus to avoid the noise in the detection of defects due to the vibration of atoms.

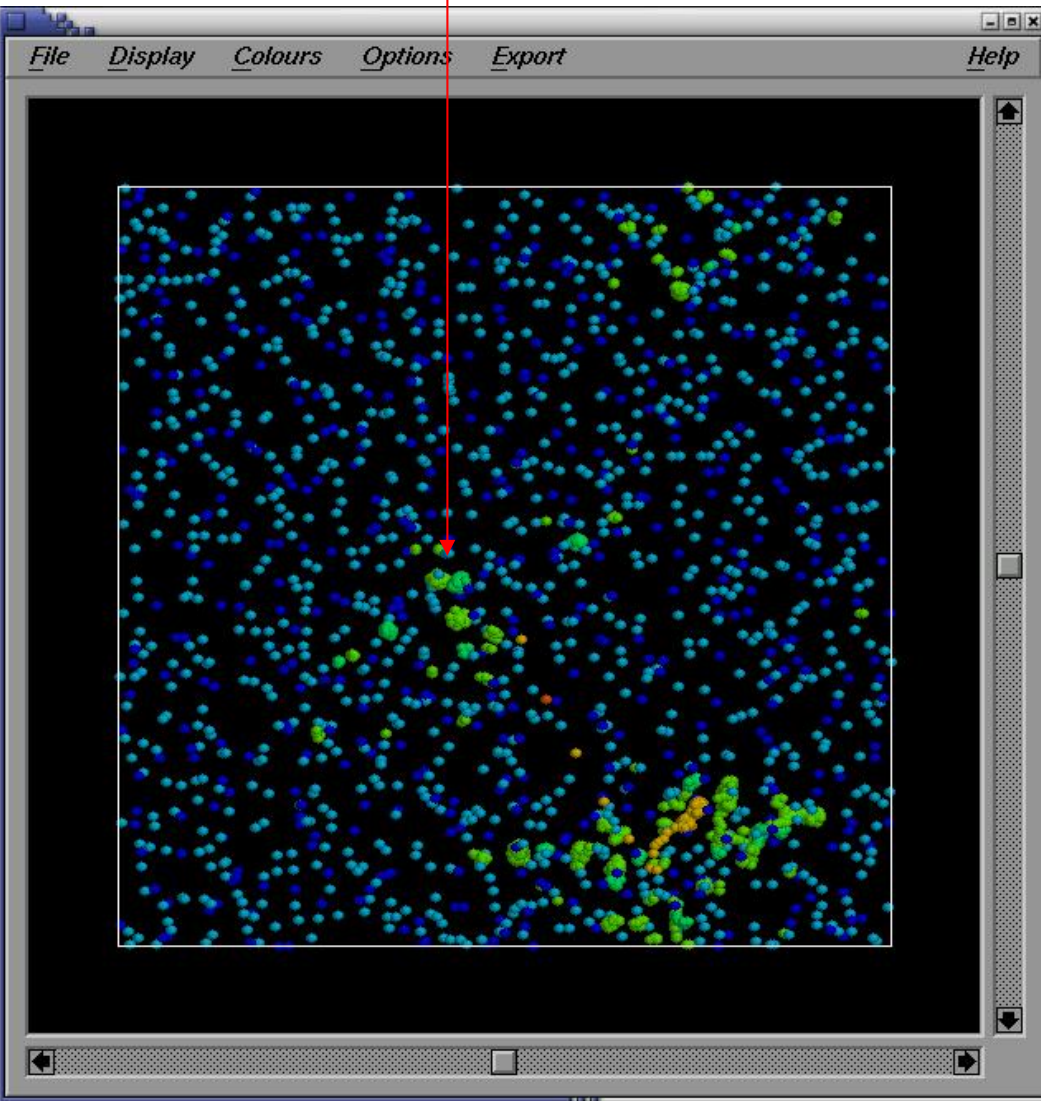
- - We are making simulations with recoil energies in steps of 10 eV starting at 40 eV until we find stable defects. Thus we will find the minimum energy to produce defects.

This knowledge will give us more information about this amorphous material; for example to find out if there is some dependency with the angle of backward movement of the PKA, that is, to have one better understanding of the behavior of this material opposed the radiation.

- We will study the production of defects with recoil energies of 1 keV until 20 keV.

Results with the analysis codes

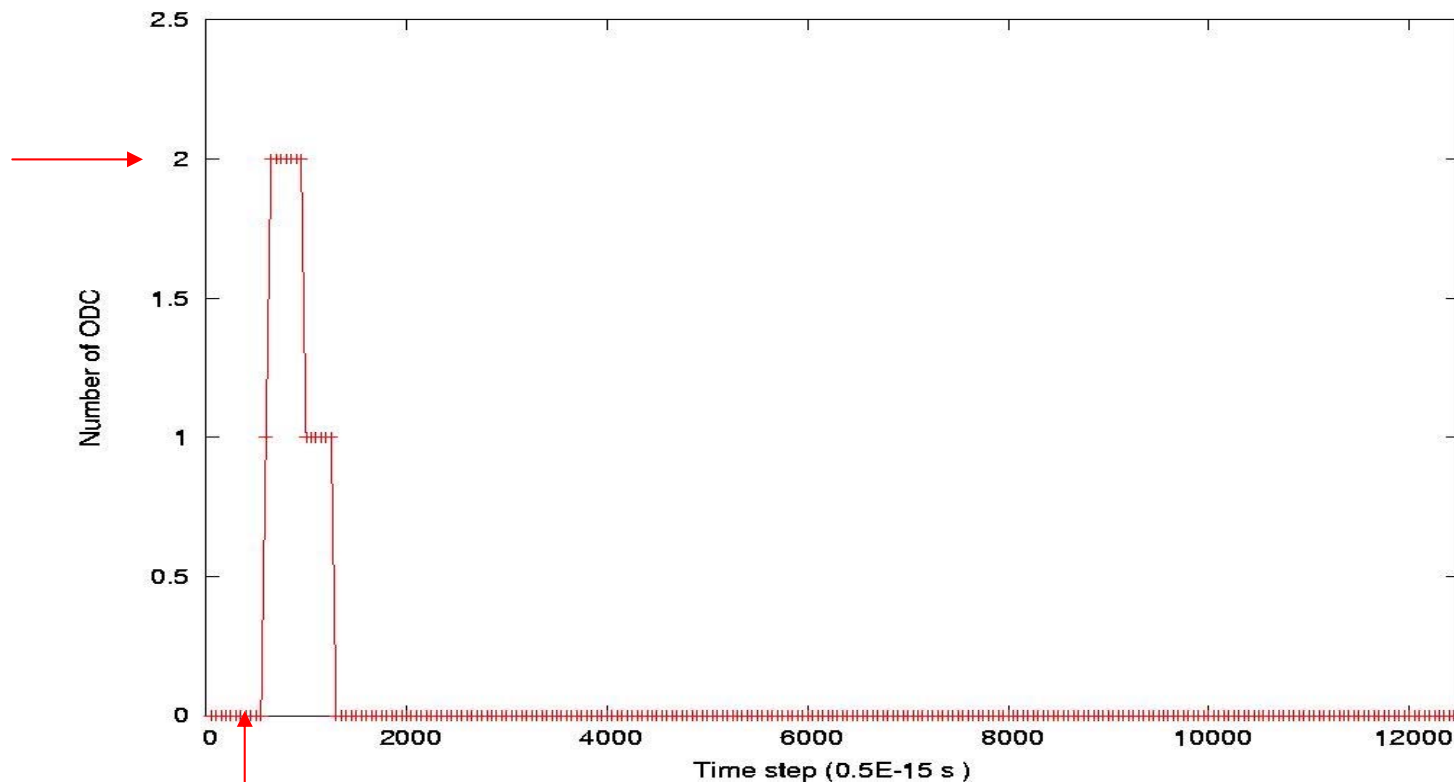
Initial position of PKA



This image is a superposition of frames for different times of a Simulation of PKA with a recoil energy of 90 eV in a simulation box of 1536 atoms.

- In orange we have the path of PKA
- In green we have the path of other atoms influenced by the energy of PKA
- In dark blue Si atoms
- In blue O atoms.

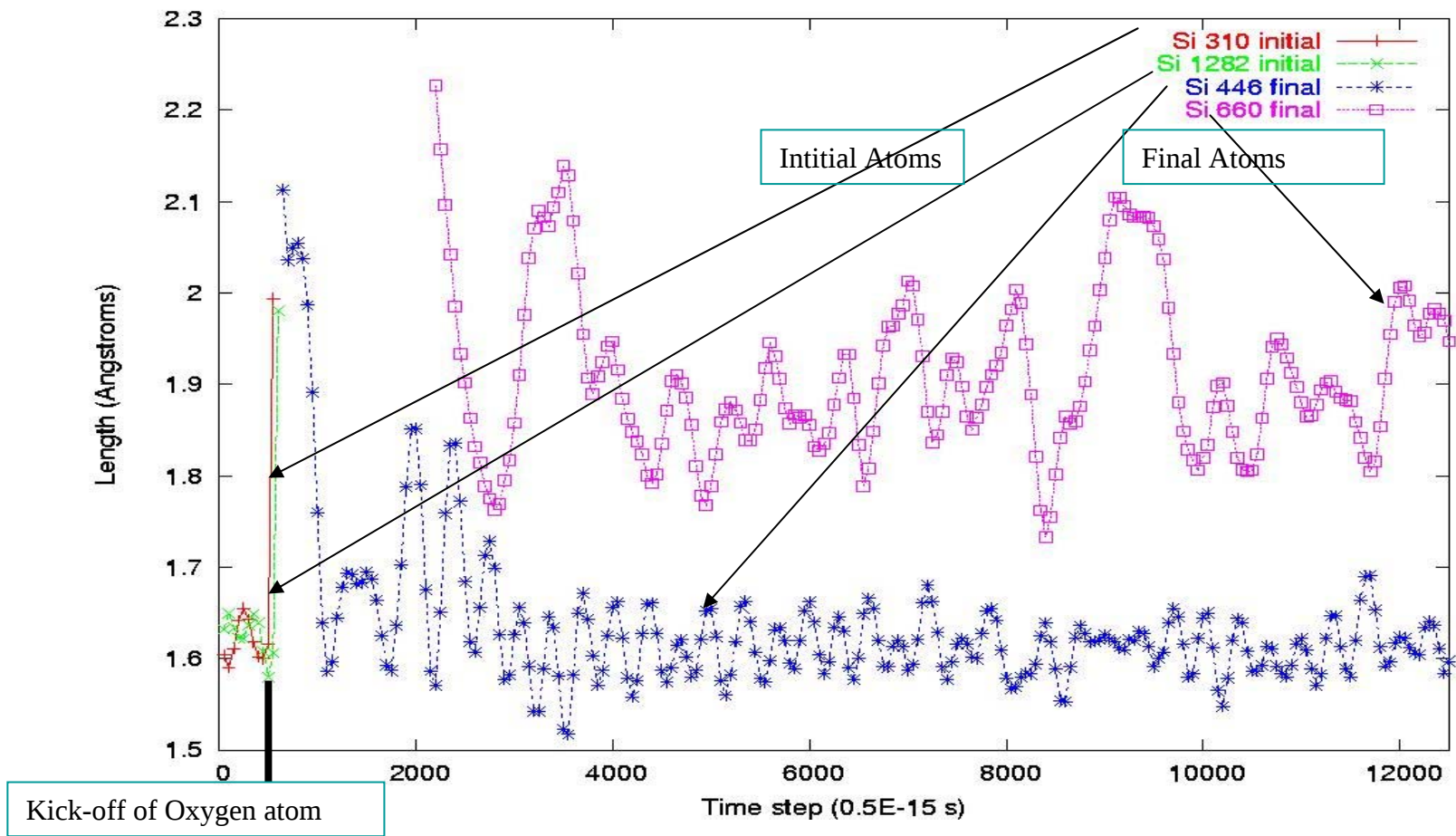
As you can see in the figure the PKA almost does not cause any effect in its first stages, and finally it causes all the damage in the material.



Numbers of ODCs created by Pka with 28.5 eV

Kick off

As you can see in this figure when the energy is communicated to the PKA, two ODCs are produced. After, these ODCs disappear because of PKA has not got enough energy to produce stable defects.



Distance from PKA (Oxygen) to the Silicon atoms which are getting closer in its path, whenever they are to a distance smaller than 2.35 Å



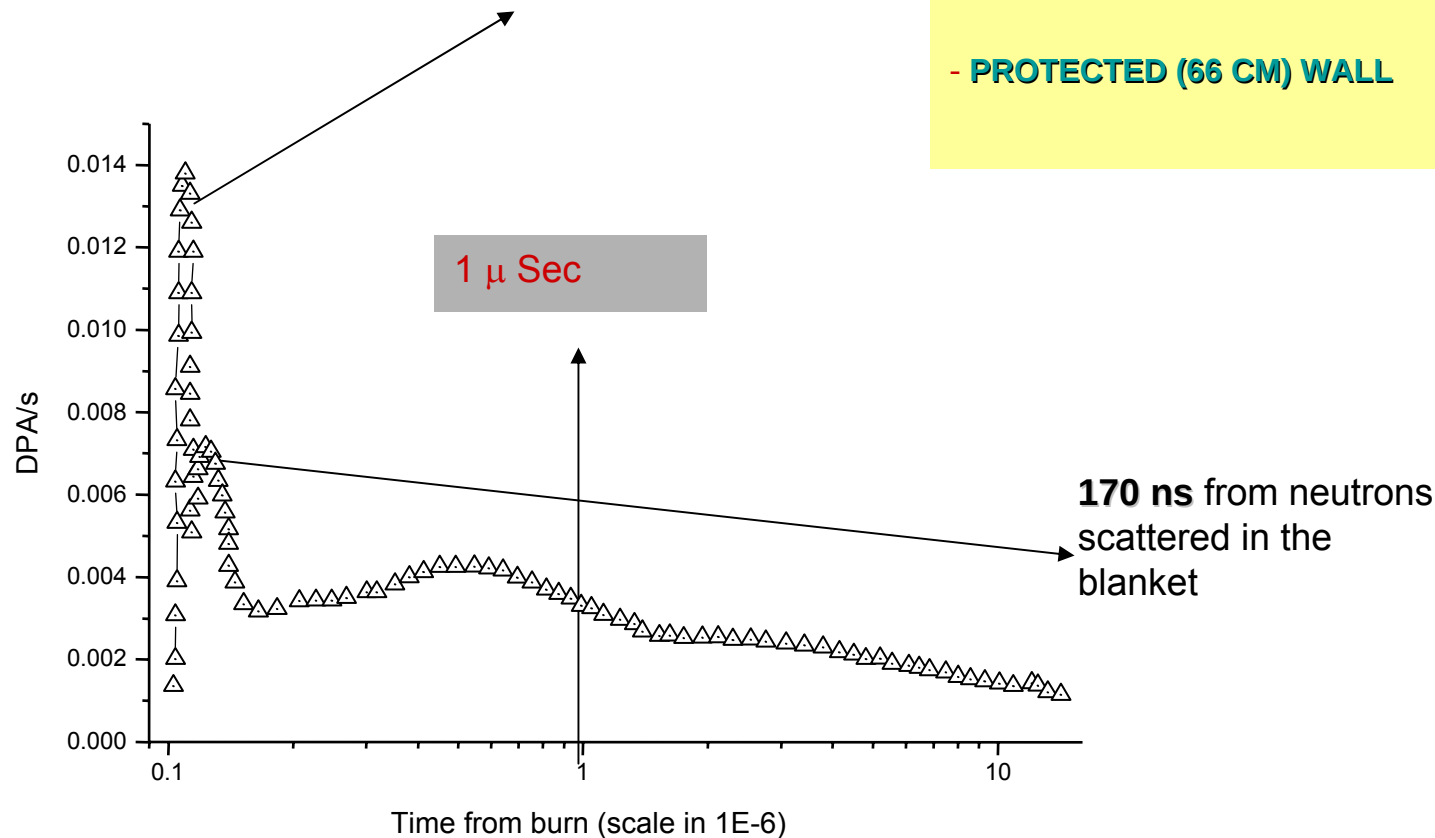
Duration of the pulse in the wall

According to transport
calculation

130 ns from 14 MeV
unscattered neutron

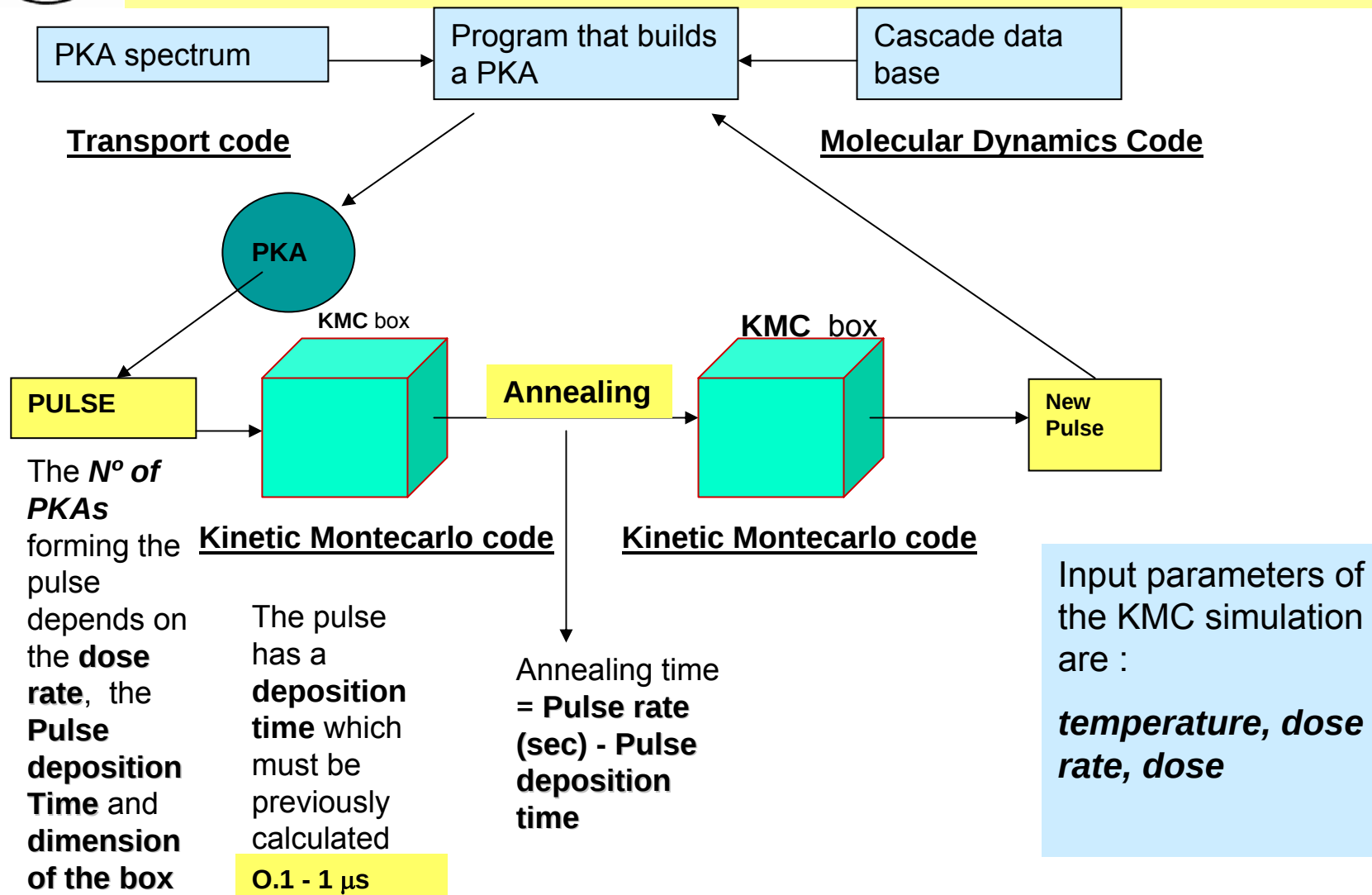
- ASSUMING TARGET SPECTRAL
CONDITIONS

- PROTECTED (66 CM) WALL





Multiscale approach for Pulsed Irradiation

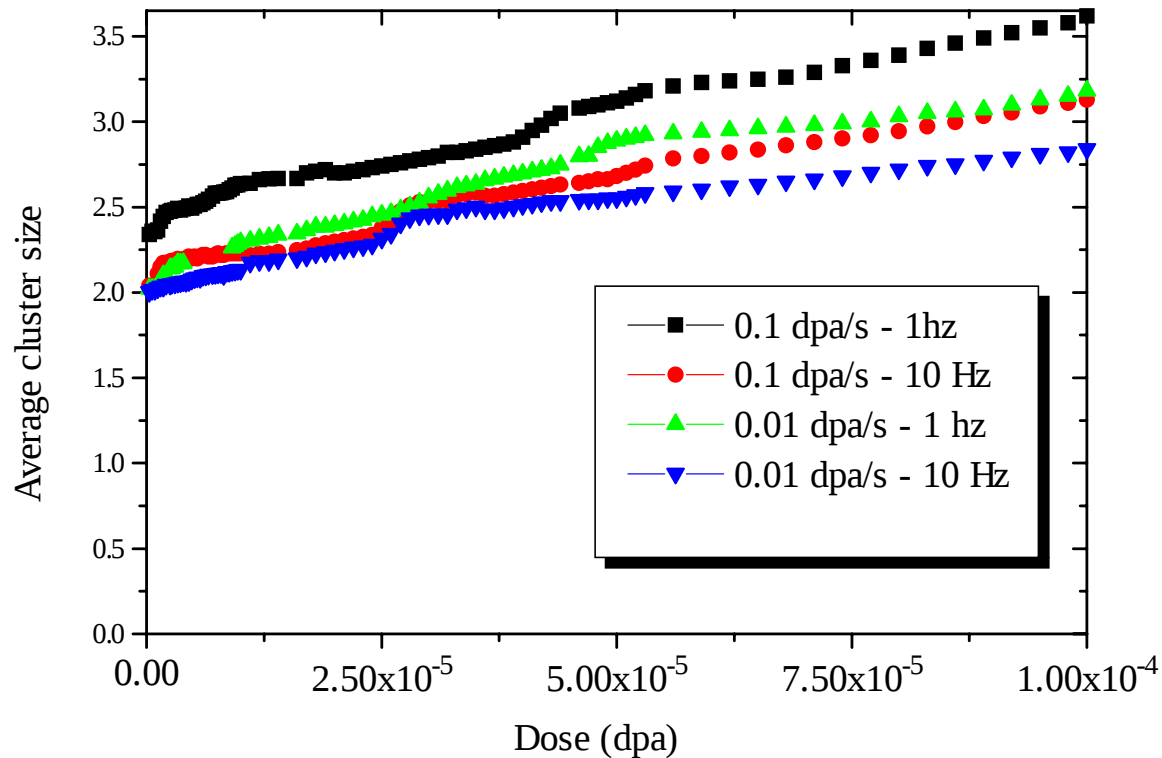




Vacancies

For a given dose rate, frequency control
vacancy cluster size

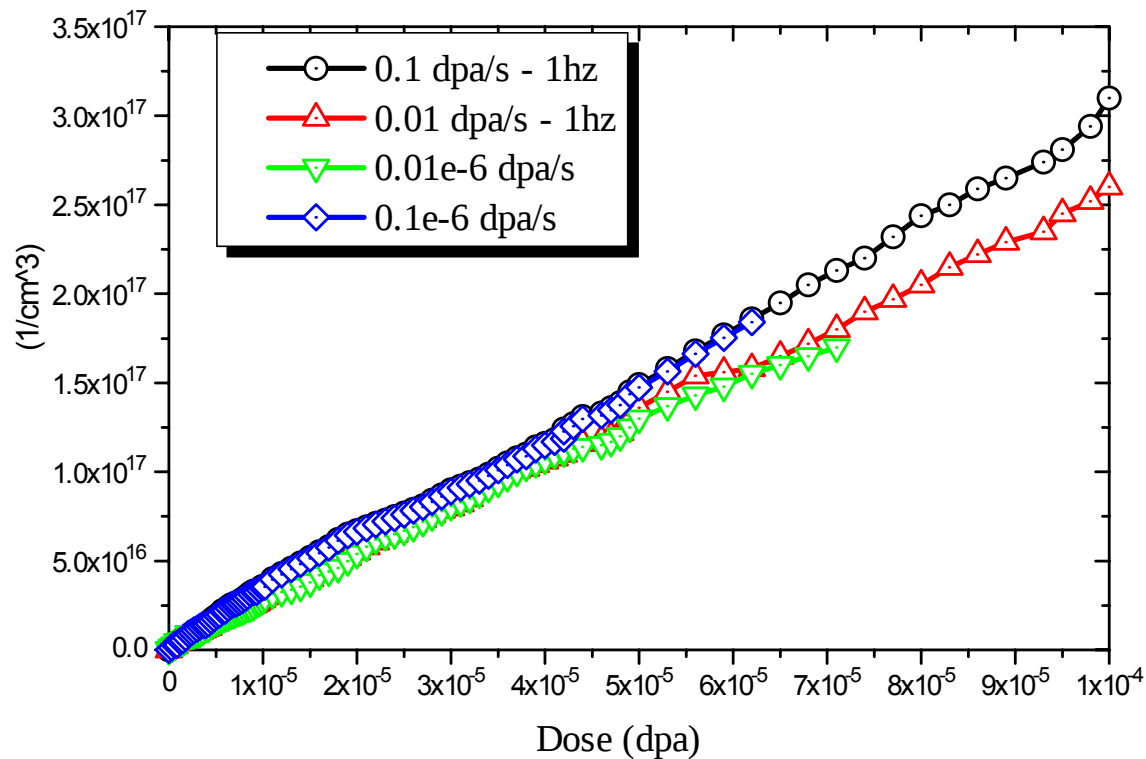
Lower frequency = larger average size



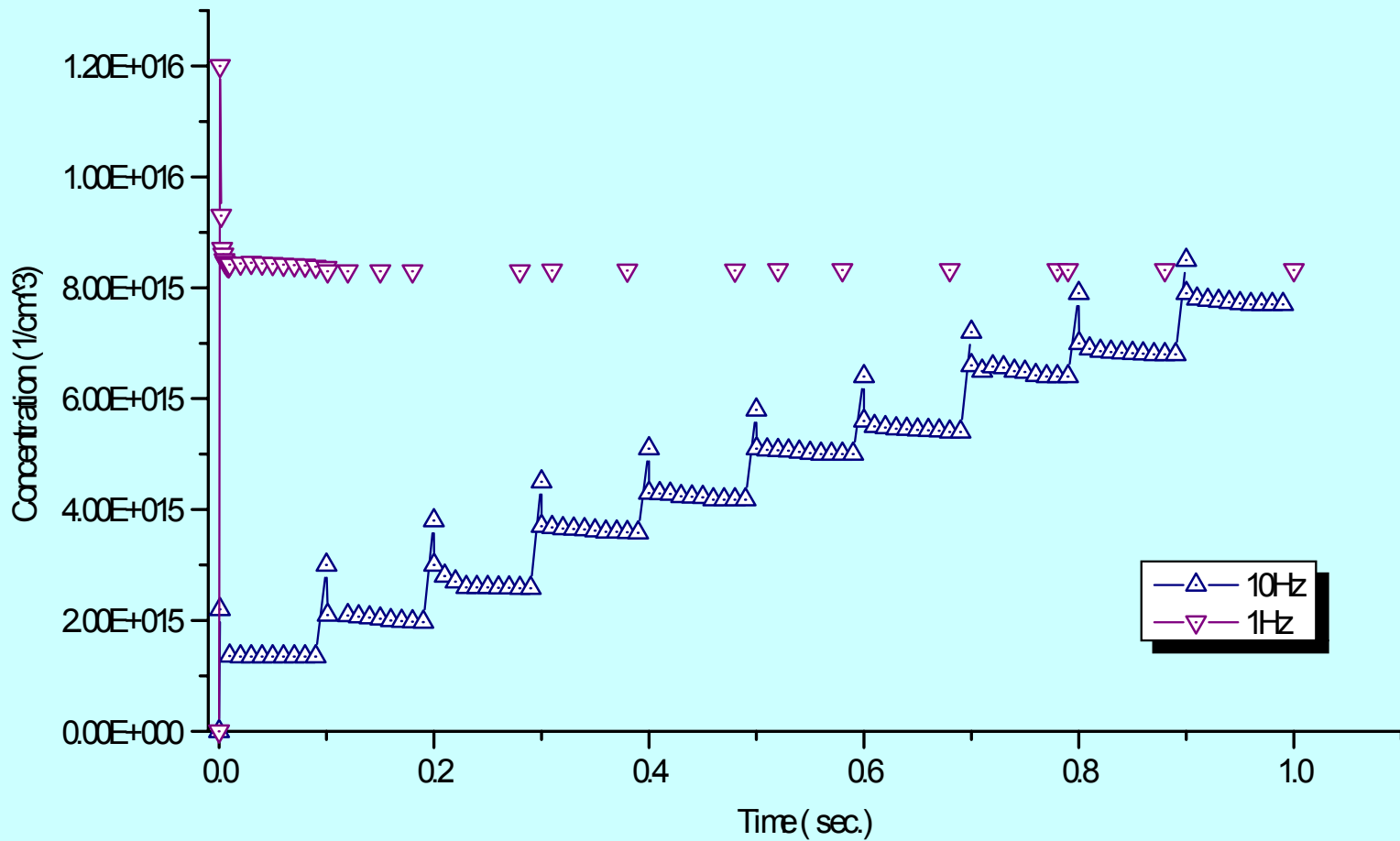


Continuous vs Pulsed

Comparison between Pulsed and Continuous irradiation leads to the conclusion that damage accumulation is almost identical as regard to vacancy clusters density



Vacancy Clusters Concentration Vs Time





Vacancy clusters Concentration 1Hz pulse

