Fission Product Diffusion in SiC: Computational Modeling and Experimental Measurement

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Outline

- Motivation
- *Ab initio* Calculations of Thermodynamic Properties
  - Interatomic potentials for defect configurations of fission products (Ag, Ru, I and Pd)
  - Defect formation and stable defect structures in SiC
  - Migration paths and energy barriers of fission products
- Dynamics Simulations
  - Fitting potentials for Ag-SiC system based on *ab initio* data
  - Formation energies and migration energy barriers as compared with *ab initio* calculations
- Experimental Approach
  - Diffusion in SiC
  - Planar diffusion couple construction
  - Ion implantation and irradiation
- Summary

NEUP project - Fission Product Transport in TRISO Fuel

- The objective of this research is to determine the diffusion coefficients for fission products (FP) in SiC under thermal and irradiation conditions, as well as synergistic effects of radiation damage, and fission products behavior at the IPyC/SiC interface.

- Experiments will utilize RBS and SIMS to determine diffusion parameters over a range of irradiation conditions and temperatures.

- First-principles approaches (*ab initio* calculations and molecular dynamics simulations) will be used to validate experimental data and determine the atomistic diffusion mechanisms to further improve empirical models used in the PARFUME code to predict FP release in TRISO fuel.

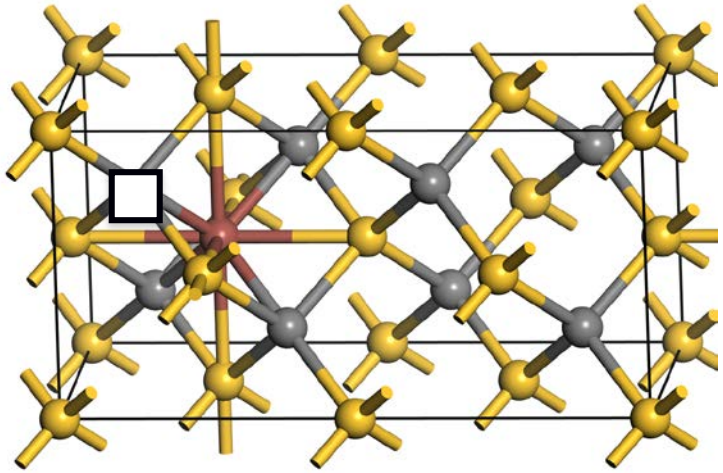
Ab Initio Calculations

- *Ab initio* calculation-VASP（Vienna *Ab initio* Simulation Package）

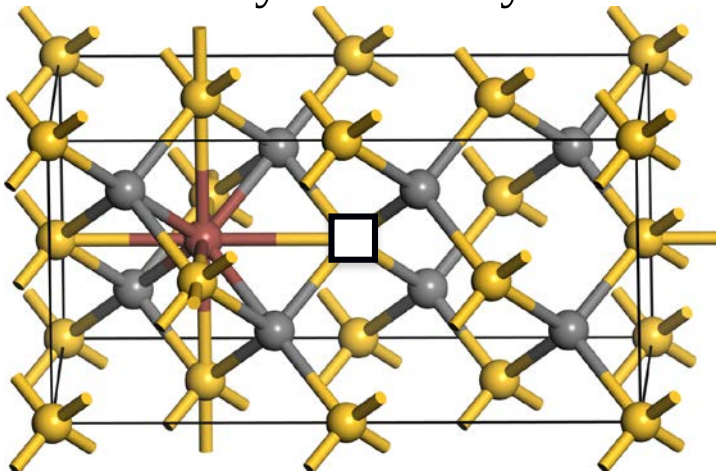
- Projected augmented wave (PAW) potentials and generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof (PBE) for exchange-correlation.
- Broyden-Fletcher-Goldfarb-Shanno (BFGS): achieve the minimum total energy of the systems.
- Cutoff energy for plane wave expansion: 500 eV
- SiC supercell: 3x3x3 (216 atoms)
- k-mesh: 4x4x4

Possible Defect Configurations

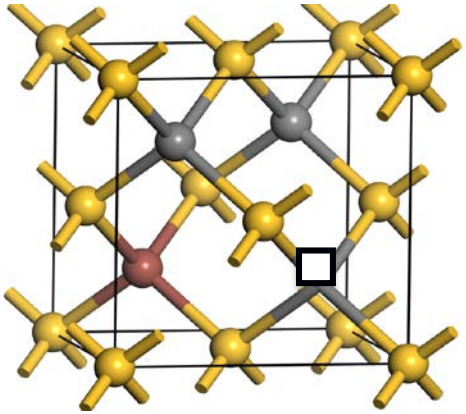
- Ag tetrahedral surrounding by four C atoms with a nearby Si vacancy



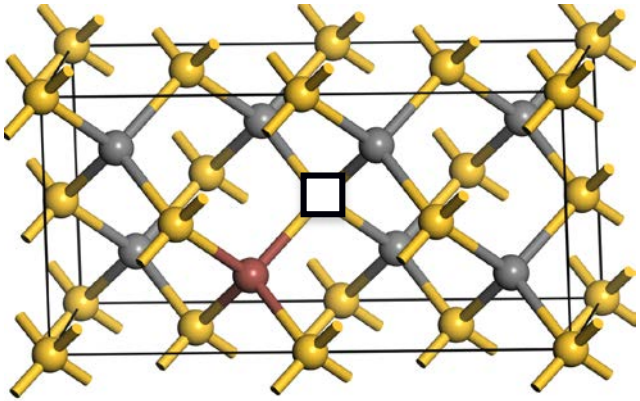
(6) Ag_Si+C_V



(7) Ag_Si+Si_V



(8) Ag_$T_C + C_V$



(9) Ag_$T_C + Si_V$

Ag

vacancy

Formation Energies

Defect	Si-rich (eV)	C-rich	Shrader 2011 (Si-rich)	Shrader 2011 (C-rich)
Ag_C	7.1392	7.6871	7.39	7.83
Ag_Si	6.6403	6.0924	6.60	6.16
Ag_Si+Cv	5.3131	5.3131	5.32	5.32
Ag_Si+Siv	13.9072	12.8114	13.53	12.65
Ag_C+Cv	8.0889	9.1874		
Ag_C+Siv	5.3131	5.3131		
Ag_T_C+Siv	6.6137	6.0658		
Ag_T_{Si}+C_v	7.0257	7.5736		
Ag_T_C	9.6098	9.6098	10.49	10.49
Ag_T_{Si}	10.6256	10.6256	11.38	11.38
Ag-C<100>	10.6204	10.6204		
Ag-C<111>	10.4699	10.4699		
Ag-Si<100>	9.5864	9.5864		
Ag-Si<110>	10.1734	10.1734	10.91	10.91
Ag-Si<111>	10.4728	10.4728		

- Ag_Si+Cv - 5.3131 eV; Ag_T_c+Siv – 6.61 eV; Ag-Si<100> - 9.59 eV; Ag_ T_C - 9.61 eV

Formation Energies

Defect	Si-rich (eV)	C-rich
Pd_C	4.2181	4.7660
Pd_Si	5.2006	4.6527
Pd_T_C	6.9943	6.9943
Pd_T_{Si}	8.8192	8.8192
Pd-C<100>	8.8147	8.8147
Pd-C<111>	10.0233	10.0233
Pd-Si<100>	6.9803	6.9803
Pd_C-Cv	5.5303	6.6261
Pd_Si-Cv	4.3458	4.3458
Pd_C-Siv	4.3428	4.3428
Pd_Si-Siv	12.1410	11.0452
Pd_T_C-Siv	5.1745	4.6266
Pd_T_{Si}-Cv	4.2142	4.7621

- Pd_Si<100> - 6.98 eV; Pd_T_C – 6.99 eV; Pd_Si-Siv – 4.34 eV; Pd_Si-Cv – 4.35 eV.
- Pd substitutions are also stable.

Formation Energies

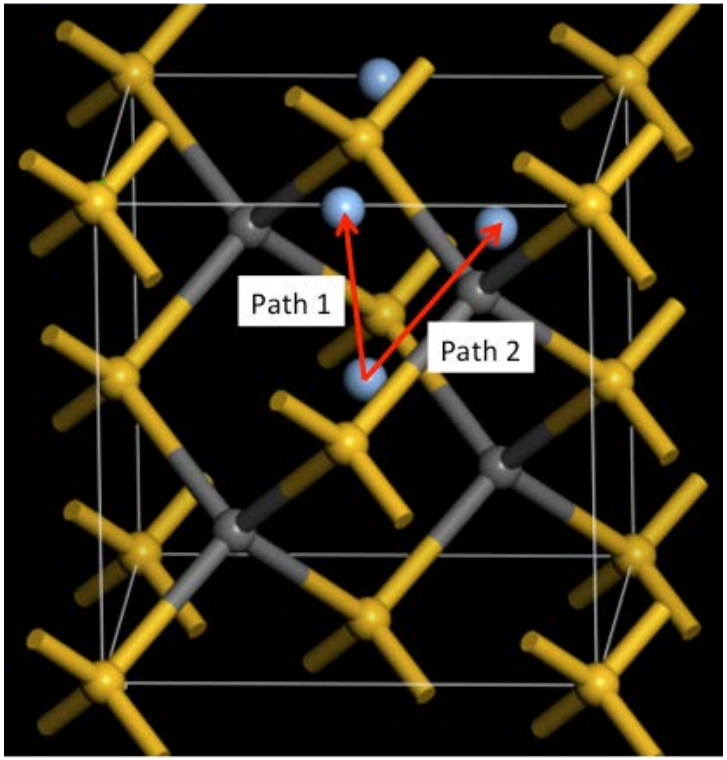
Defect	Si-rich	C-rich
Ru_C	4.1088	4.6567
Ru_Si	3.8055	3.2576
Ru_T_C	8.3264	8.3264
Ru_T_{Si}	7.5305	7.5305
Ru-C<100>	7.5207	7.5207
Ru-Si<100>	7.5304	7.5304
Ru_C-Cv	6.1058	7.2016
Ru_Si-Cv	5.5688	5.5688
Ru_C-Siv	5.5689	5.5689
Ru_Si-Siv	10.6273	9.5315
Ru_T_C-Siv	3.8046	3.2567
Ru_T_{Si}-Cv	4.1044	4.6523

- The different stable configurations of fission products suggest that their migration paths and mechanisms may be different.

- Ru_C<100> - 7.52 eV; Ru_Si<100> – 7.53 eV; Ru_Tsi – 7.53 eV; Ru_T_C-Siv – 3.80 eV; Ru_Tsi-Cv – 4.10 eV.
- Ru substitutions are also stable.

Migration via Interstitial Mechanism

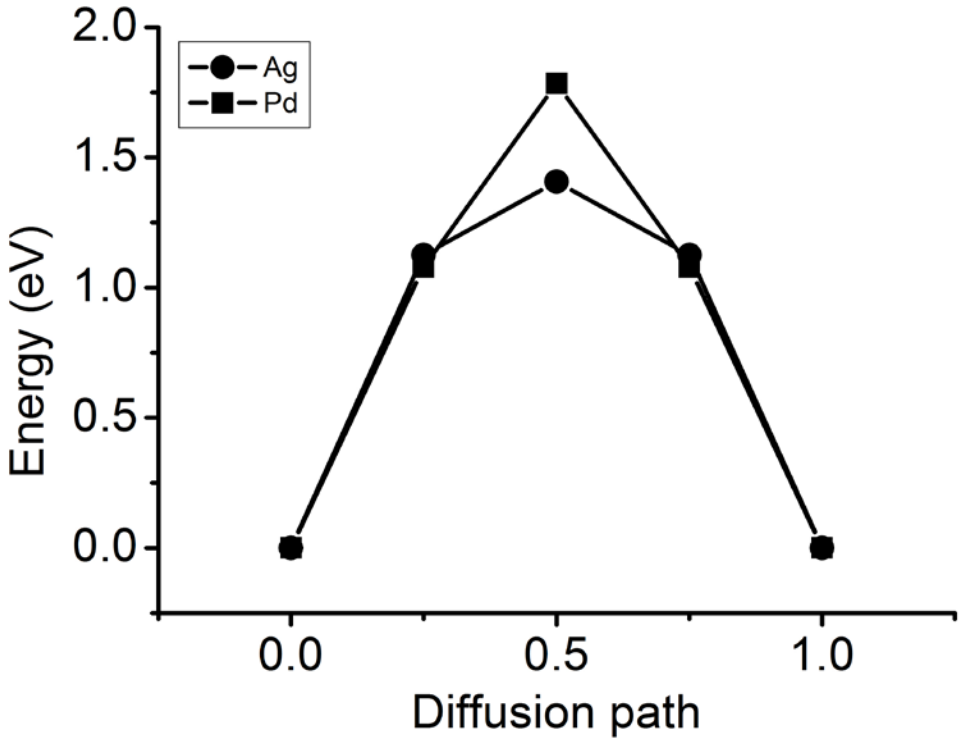
- The cell used in the present calculations 3x3x3 with 216 atoms
- GGA-PBE, energy cut off is 450 eV, 1 x 10^{-5} eV/cell for energy, -0.03 eV/angstrom for force
- K-mesh 6x6x6
- NEB (nudged elastic band)



- Path 1 – X (Ag, Ru, Pd) tetrahedral surrounding by Si atoms to a tetrahedral surrounding by Si atoms.
- Path 2 - X (Ag, Ru, Pd) tetrahedral surrounding by Si atoms to a tetrahedral surrounding by C atoms.

Migration via Interstitial Mechanism

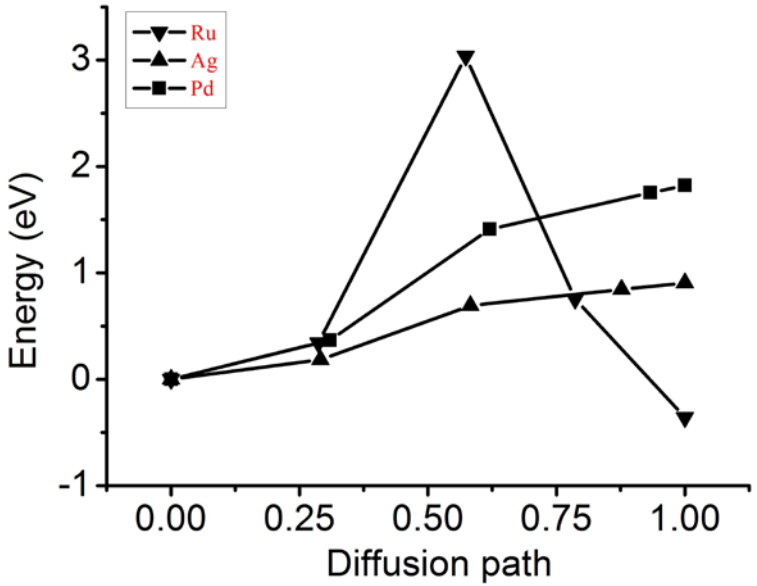
- Path 1

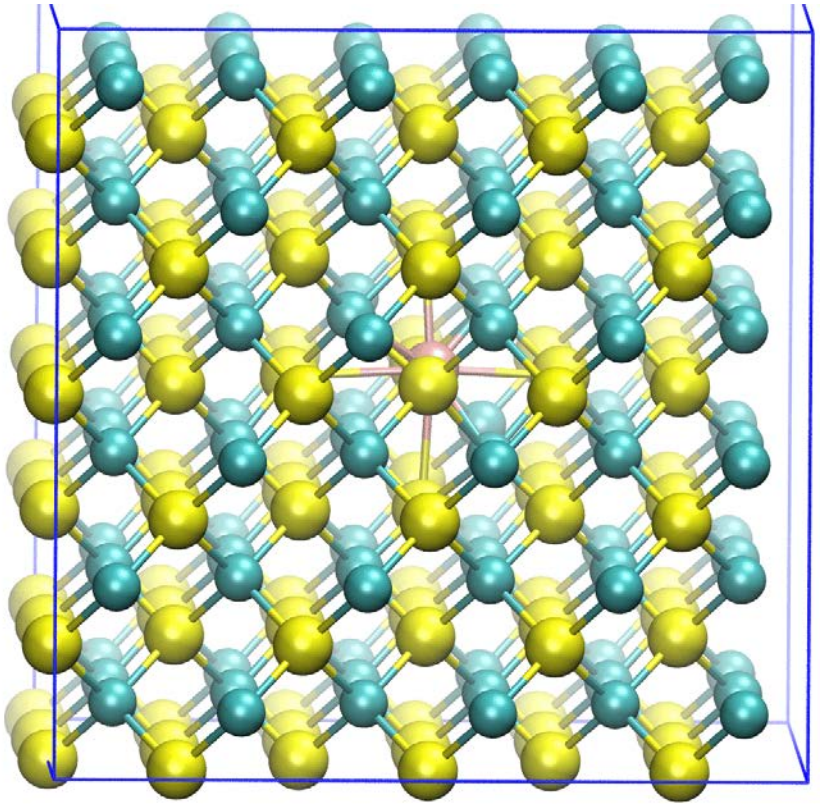


- Migration energy ranges between 1.4 and 1.8 eV, depending on fission products

Migration via Interstitial Mechanism

- Path 2





- The migration energy of Ru is very high (3.5 eV), while the migration energy of Pd (1.5eV) and Ag (.75eV) are much lower

Fitting Interatomic Potentials for Large Scale MD Simulations

- **Analytical Bond-Order Potential (ABOP)**

  - Accurate interatomic potential is crucial for performing large scale MD simulations for large scale diffusion coefficient calculations
  - The total energy is written as a sum of individual bond energies with pairwise repulsive and attractive contributions as well as a cutoff function
  - Three iterations of ABOP were calculated to find the potentials that most accurately describe physical properties as well as match previous DFT simulations in literature
  - ABOP I, II, and III refer to three valid solution sets to the potential that are closest to our expected value

Physical Properties (as compared with ab initio calculations)

- **Ag physical properties**

Ag		Exp.	Ab initio	ABOP
a_0 (Å)		4.09		4.11
E_c (eV/atom)		-2.85		-2.85
Elastic constant (0 K, GPa)				
B		108.7	115.17	113.93
C_{11}		131.5	132.99	131.4
C_{12}		97.3	106.26	105.2
C_{44}		51.1	61.38	62.07
E_{vac} (eV)		1.1,1.15	1.06,1.20	1.15
E^m_{vac} (eV)		0.66		0.62
HCP	E_c (eV)	0.0155, 0.003		0.004
	a /c (Å)	2.93/4.72		2.90/4.73
BCC	E_c (eV)	0.0334,0.039		0.020
	a_0 (Å)	3.29		3.269
Diamond	E_c (eV)	1.054		1.116
	a_0 (Å)			6.35
	E_c (eV)	0.428		0.351
	a_0 (Å)			2.77

Physical Properties (as compared with ab initio calculations)

- **Dimer properties**

Ag-Si	Exp.	DFT	ABOP I	ABOP II	ABOP III
E_b (eV)	1.88(10)		2.003	1.765	1.775
r_b (Å)	2.40,2.36	2.39	2.315	2.310	2.306
$\omega_0(cm^{-1})$	296.9		292.948	286.050	286.282
Ag-C					
E_b (eV)			2.020	2.047	2.056
r_b (Å)		2.051	2.048	2.096	2.151
$\omega_0(cm^{-1})$		477.2	427.365	426.664	442.207
Ag-Ag					
E_b (eV)	1.66	2.48		1.828	
r_b (Å)	2.530,2.56	2.533		2.582	
$\omega_0(cm^{-1})$	191.8			206.112	

Physical Properties (as compared with ab initio calculations)

- **Defect formation energies with an Ag atom in 3C-SiC (Si-rich)**

Defect	DFT	Shrader	ABOP I	ABOP II	ABOP III
Ag_C	7.1392	7.39	6.00	7.55	7.19
Ag_Si	6.6403	6.60	5.20	5.35	5.25
Ag_Si+Cv	5.3131	5.32	5.16	5.09	5.24
Ag_Si+Siv	13.9072	13.53	10.31	10.28	10.25
Ag_C+Cv	8.0889		8.02	9.47	9.06
Ag_C+Si_v	5.3131		5.16	5.09	5.24
Ag_T_C+Si_v	6.6137		5.20	5.35	5.25
Ag_T_{Si}+C_v	7.0257		9.87	10.69	7.19
Ag_T_C	9.6098	10.49	9.78	10.63	10.76
Ag_T_{Si}	10.6256	11.38	9.36	10.06	10.10
Ag-C<100>	10.6204		9.36	10.07	10.24
Ag-C<111>	10.4699		10.30	12.10	11.18
Ag-Si<100>	9.5864		10.56	11.41	11.42
Ag-Si<110>	10.1734	10.91	10.19	10.10	14.19
Ag-Si<111>	10.4728		9.35	10.06	10.09

*unit in eV

Migration Energy Barriers

❖ NEB (nudged elastic band method)

❖ Defect cluster migration pathway (Shrader et al.)

❖ Ag_Si-Cvac

- hops that just move the accompanying defects around Ag. (reorientation hop)-2types
- hops that actually move Ag (Ag hops)

❖ Ag_Si-2Cvac

- hops that just move the accompanying defects around Ag. (reorientation hop)-1type
- hops that actually move Ag (Ag hops): ring mechanism & swap+Cmove

❖ Ag_Si-$V_{Si}V_C$

- hops that actually move Ag (Ag swap with V_{Si})

❖ Ag_TC (Ag at tetrahedral site surrounded by C)

- hops that actually move Ag to another tetrahedral site surrounded by C

D. Shrader, S. Khalil, T. Gerczak, T. Allen, A. Heim, I. Szlufarska, and D. Morgan, *J. Nucl. Mater.* 408, 257 (2010).

Overview of NEB Barrier

	Shrader	DFT	ABOP I	ABOP II	ABOP III
Ag_Si-Cvac					
I. direct swap	9.1		10.85	12.82	12.21
II. two Ag hops	9.2		11.64	12.83	12.72
III. reo1	8.02		6.68	6.62	6.50
IV. reo2	5.97		6.55	6.53	6.17
Ag_Si-2Cv					
I. ring	≥ 6.61		9.51	9.06	9.19
II. swap+Cmove	7.04		6.34	6.00	7.46
III. single reo	5.02		5.47	5.52	5.52
Ag_Si-VsiVc	0.58		0.20	0.37	0.55
Ag_TC	0.89		0.90	0.81	0.71
Ag_TSi					
I.Path 1		1.40	1.34	1.39	1.37
II. Path 2		0.75	0.90	0.81	0.71

D. Shrader, S. Khalil, T. Gerczak, T. Allen, A. Heim, I. Szlufarska, and D. Morgan, *J. Nucl. Mater.* 408, 257 (2010).

Summary

- Larger number of defect configurations of fission products are identified and investigated using *ab initio* approach in SiC;
- Most stable defect types for Ag, Ru,Pd and I are determined, and their migrations are initiated;
- Based on *ab initio* calculations, the interatomic potentials for Ag-SiC system are fitted for large scale MD simulations to study migrations of fission products and radiation enhanced diffusion
- Physical properties for Ag-SiC system are fairly reproduced as compared with both experiments and *ab initio* results;

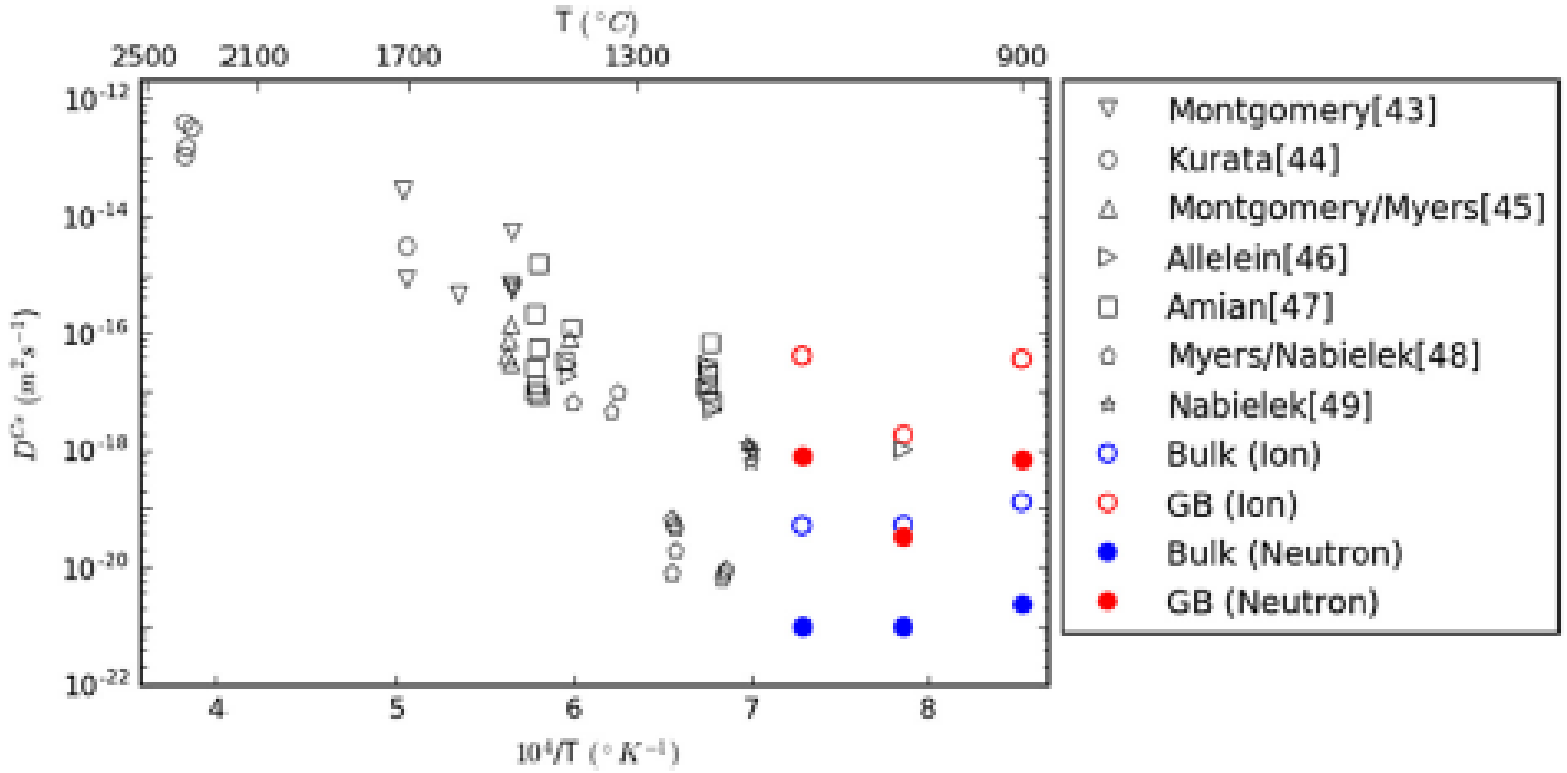
Experimental Objective

- The objective of this research is to measure radiation enhanced diffusion coefficients as well as calculate pre-exponential, and activation energies for silver, palladium, ruthenium, and iodine in silicon carbide.

- Ion Irradiation will be used to simulate 10dpa in the temperature range of 900-1300C

- Silicon ions will produce dose rates between $4.6\text{x}10^{-4}$ dpa/s and $4.6x10^{-5}$ dpa/s

Cesium Diffusion Data in SiC



Irradiation enhanced diffusion coefficient results from cesium implanted diffusion couples in past experiments. “Neutron” data points were scaled down from ion irradiated samples via dose rate.

Dwaraknath, S. S., & Was, G. S. (2016). *Journal of Nuclear Materials, 474*, 76-87.

Planar Diffusion Couple Construction

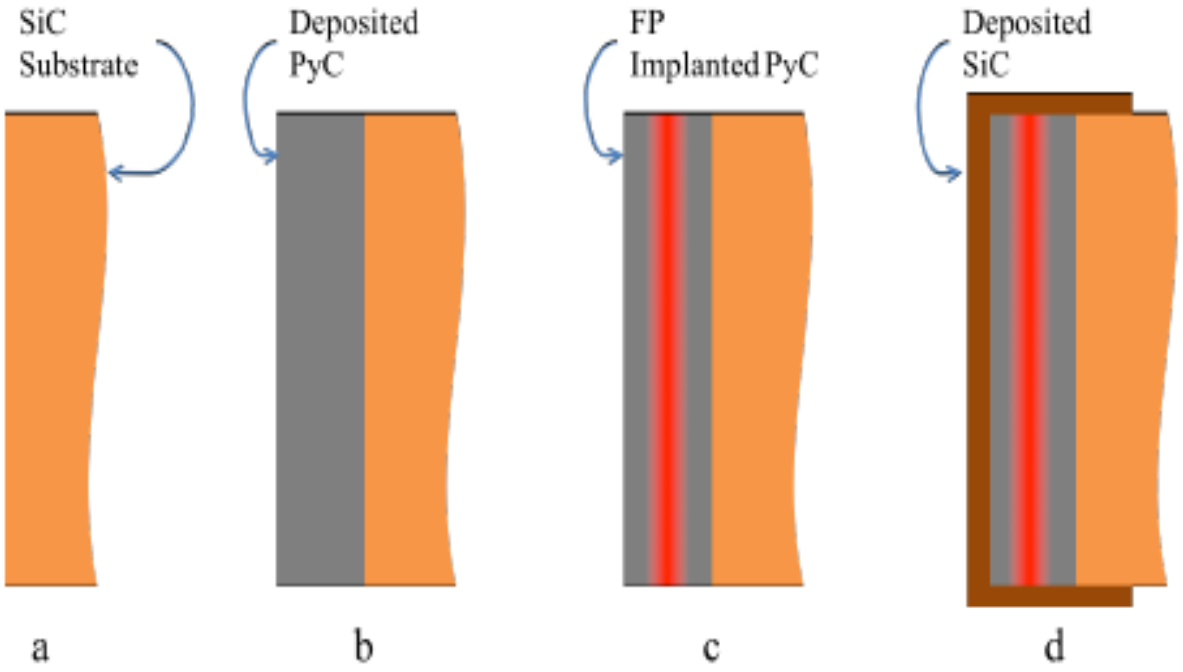
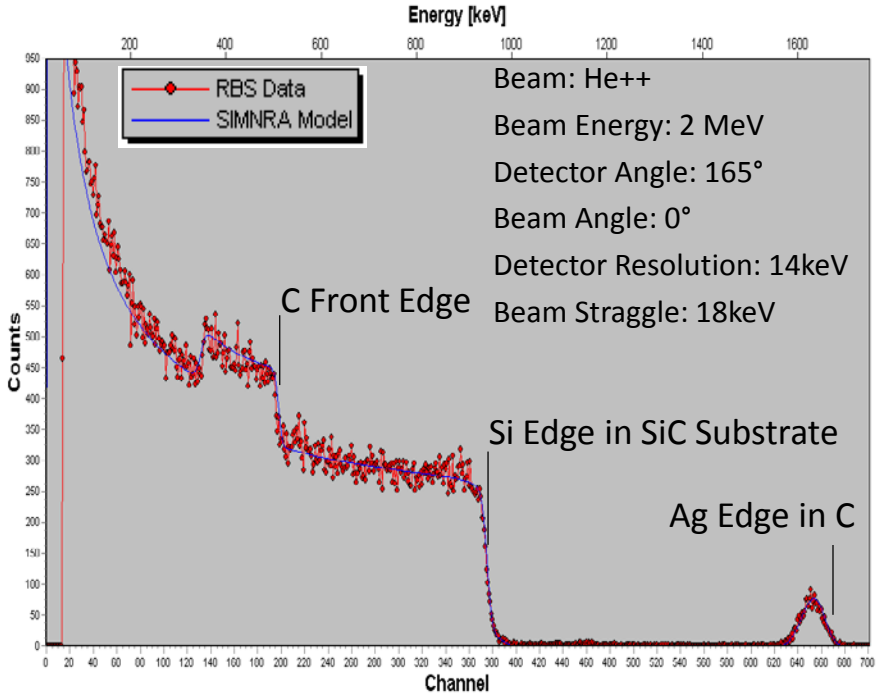
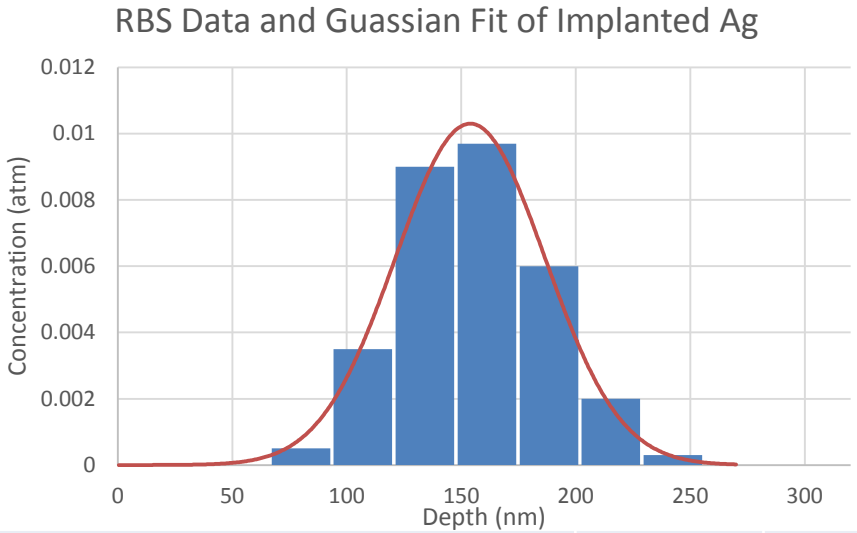


Illustration of the diffusion couple counstruction with (a) substrate (b) pyrocarbon layer (c) implanted fission product and (d) cap layer

Ion Implantation

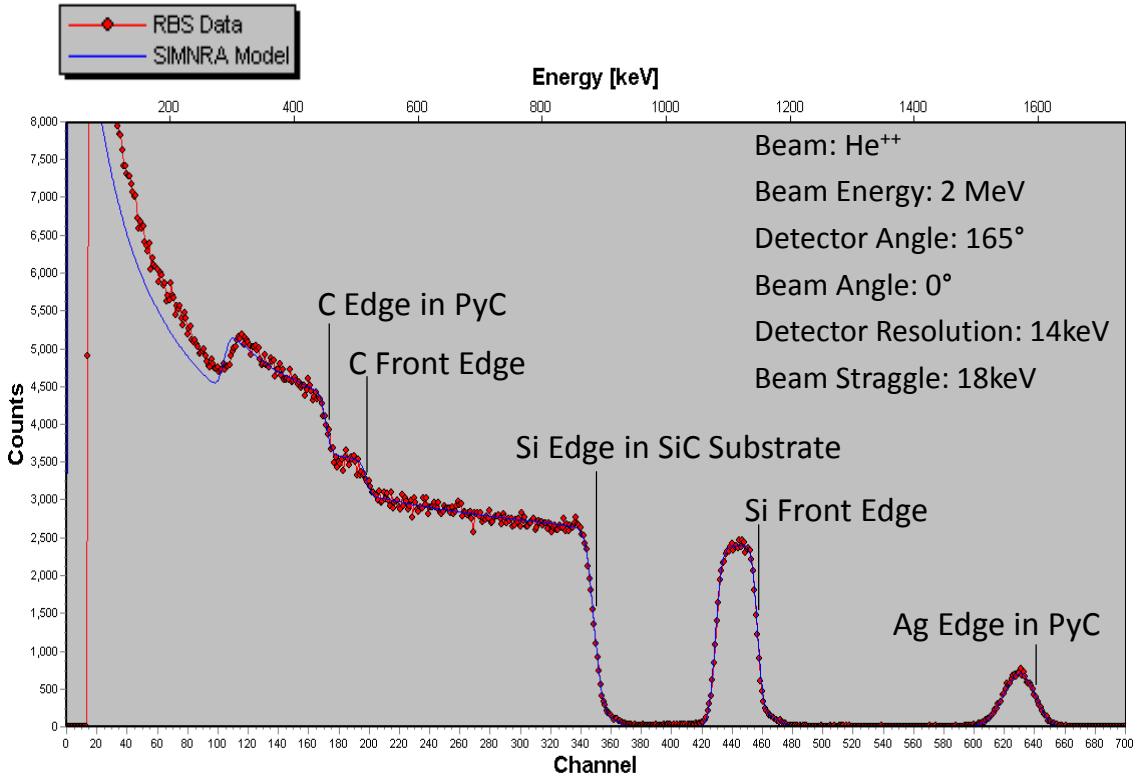
- Ion implnatation of ruthenium, silver, palladium, and iodine perfomed at Michigan Ion Beam Labratory (MIBL)
- 400keV Ag, Pd, I, and Ru ions implant into room temperature PyC layer of diffusion couple

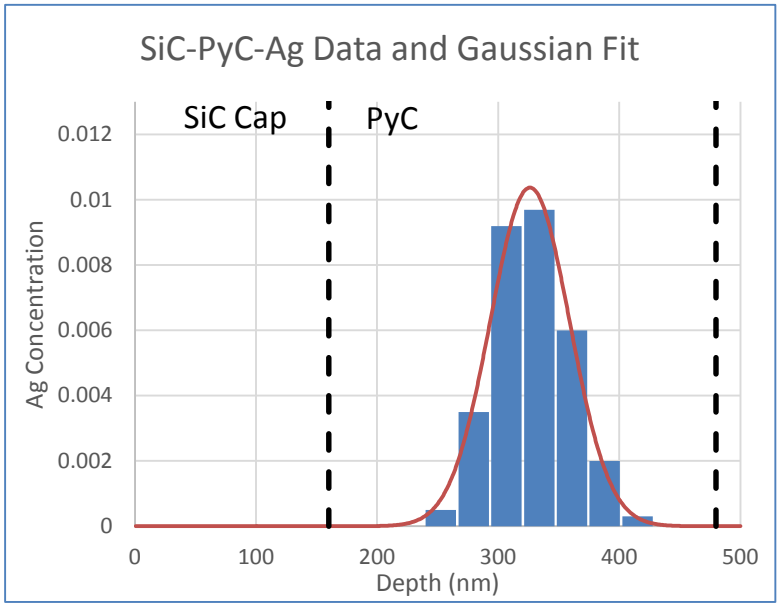


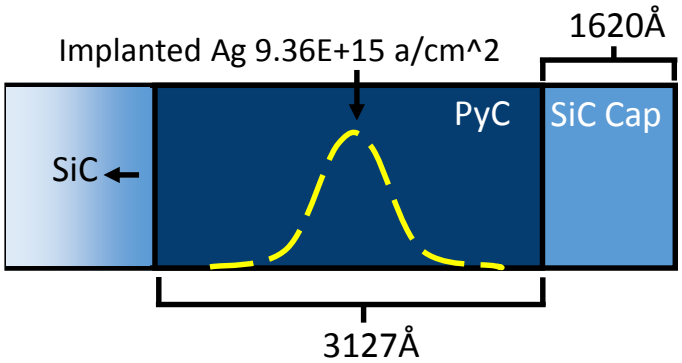


	Experiment	SRIM
Peak Concentration (atm%)	1.03	1.25
Peak Range (nm)	154	152
FWHM (nm)	76.9	68.2
Integrated Implanted Fluence (a/cm^2)	9.3E+15	1E+16

RBS Profile of Completed Diffusion Couple



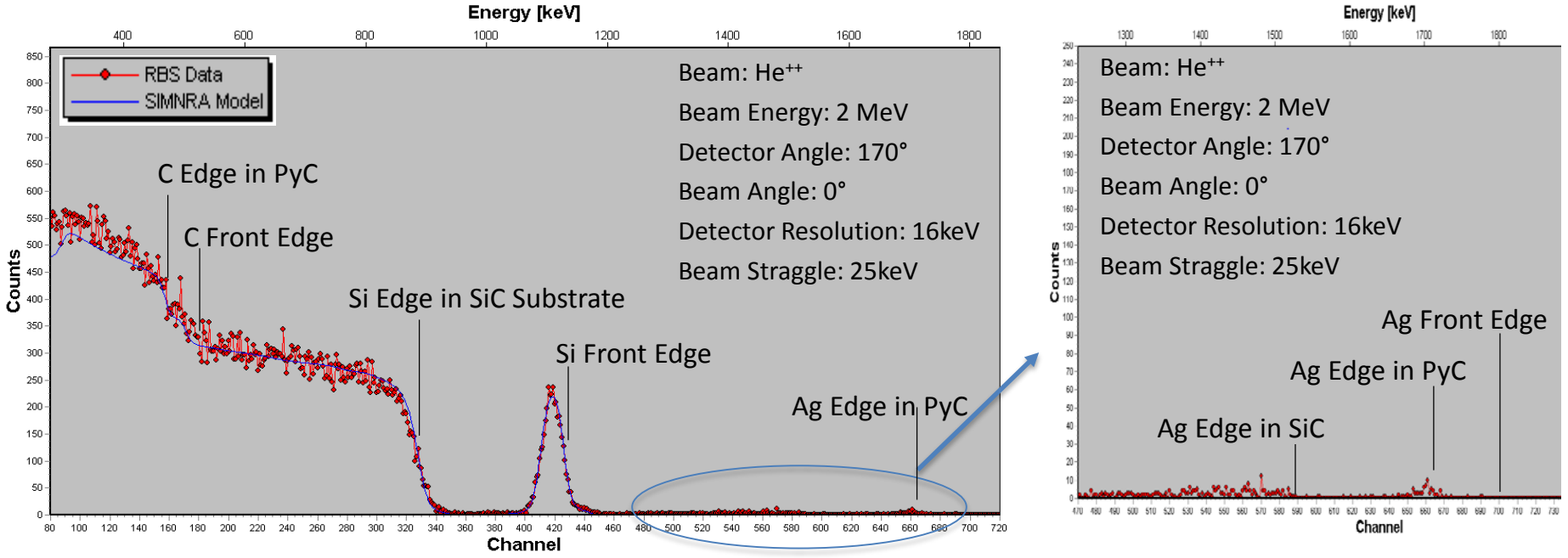




Peak Concentration (atm%)	1.04
Peak Range (nm)	326.22
Straggle (nm)	32.72
FWHM (nm)	77.066
Integrated Implanted Fluence [a/cm^2]	9.36E+15

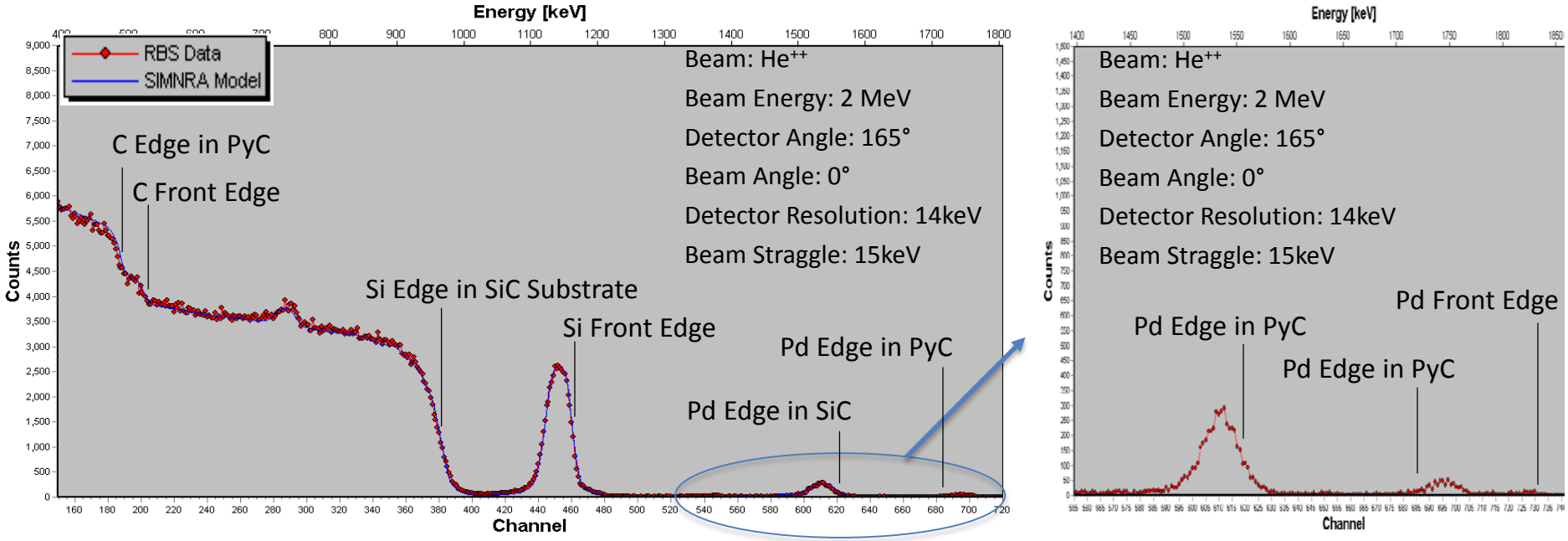
Annealed Ag Diffusion Couple

- Annealing Conditions:
  - Vertical flow graphite walled furnace, argon environment
  - Temperature measurement using type K thermocouple
  - 901C±8.6C 10 h anneal
- Silver Retention:
  - Initial diffusion couple construction retained only 10% of implanted silver



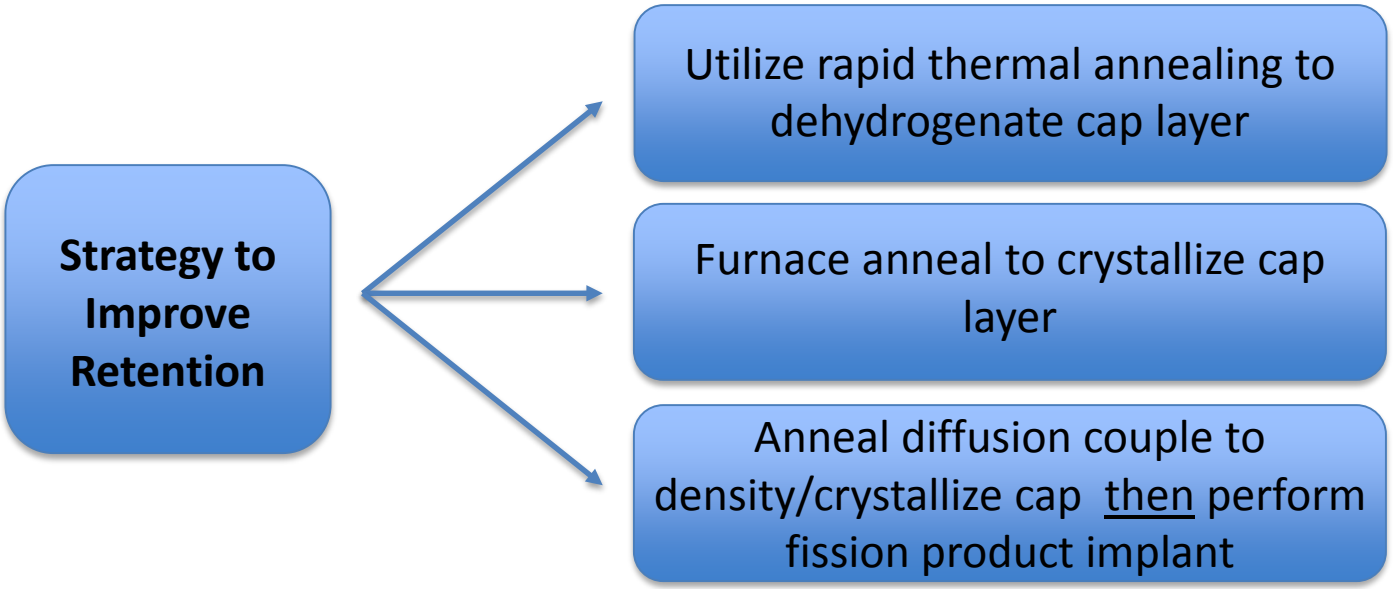
Annealed Pd Diffusion Couple

- Annealing Conditions:
  - Vertical flow graphite walled furnace, argon environment
  - Temperature measurement using type K thermocouple
  - 1101C±12.2C 10 h anneal
- Palladium Retention:
  - Initial diffusion couple construction retained only 25% of implanted palladium

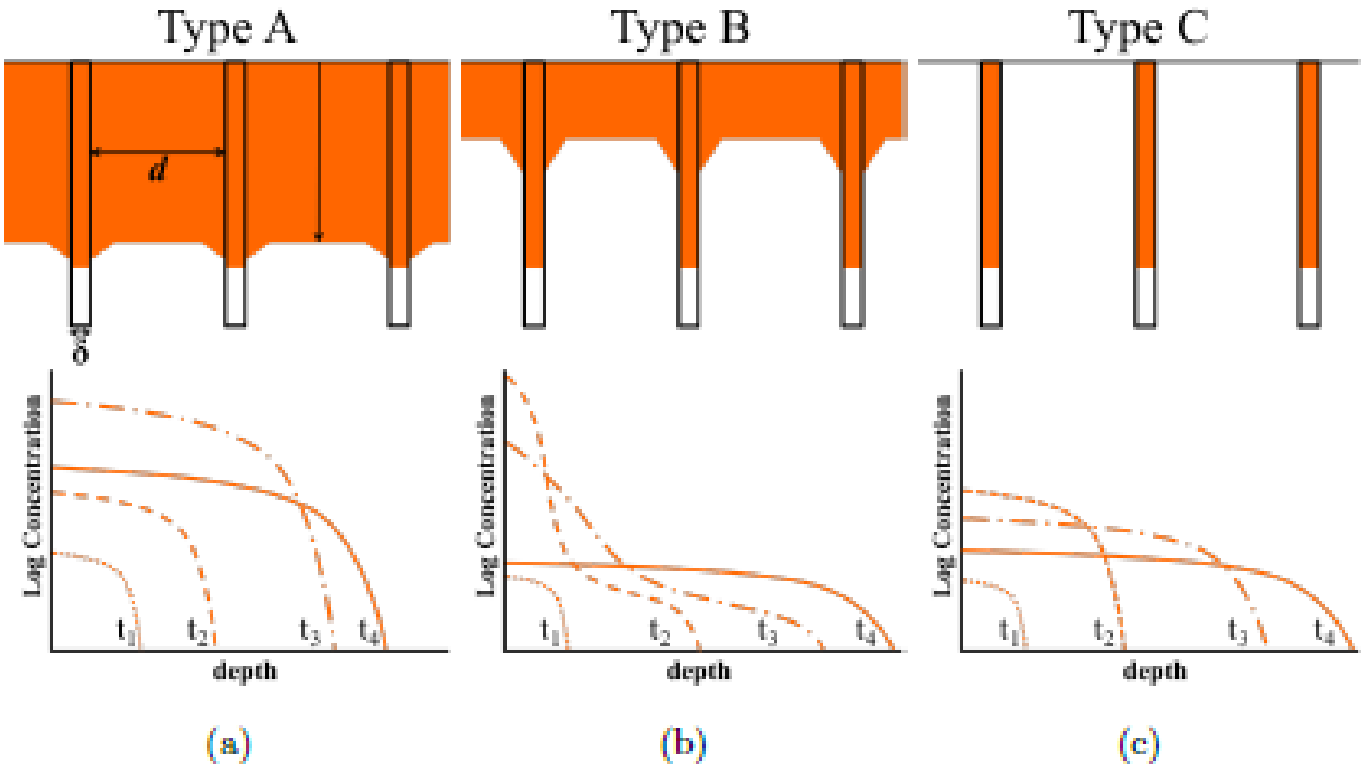


Strategy for Improving Retention

- Calculated density of as deposited SiC cap is very low, ~61% of theoretical mass density
- Uniformly densifying cap layer above 95% should provide necessary diffusion barrier for experiments to progress
- Previous experiments used similar process with SiC cap at 85% theoretical density to successfully perform experiments on Cs, Eu, Sr implanted diffusion couples



Diffusion Regimes



Schematic showing concentration profile for three diffusion regimes from Harrison model. (a) bulk dominant (b) grain boundary and bulk (c) grain boundary dominant

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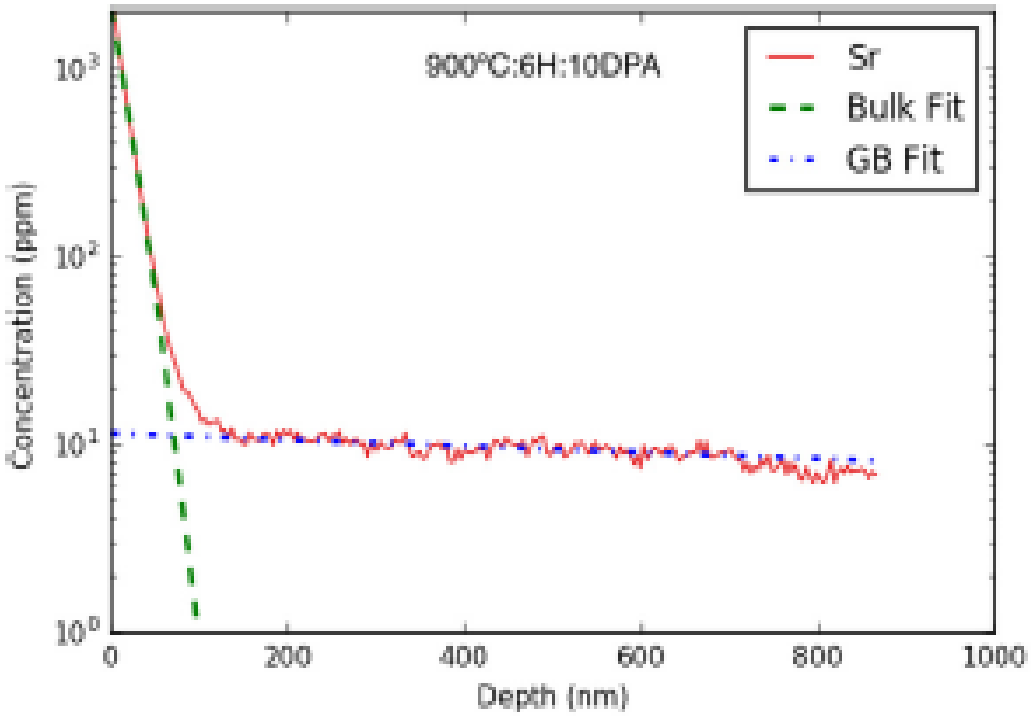
ToF-SIMS Data and Fit from Past Experiment



Measured concetnration curve of stronium into SiC in ion irradiated diffusion couple by ToF-SIMS

$$C_B(x) = C_o^B \exp\left(\frac{(x - x_o)^2}{4D_B t}\right)$$

$$C_{GB}(x) = C_o^{GB} \exp(-m * x^{\frac{6}{5}})$$

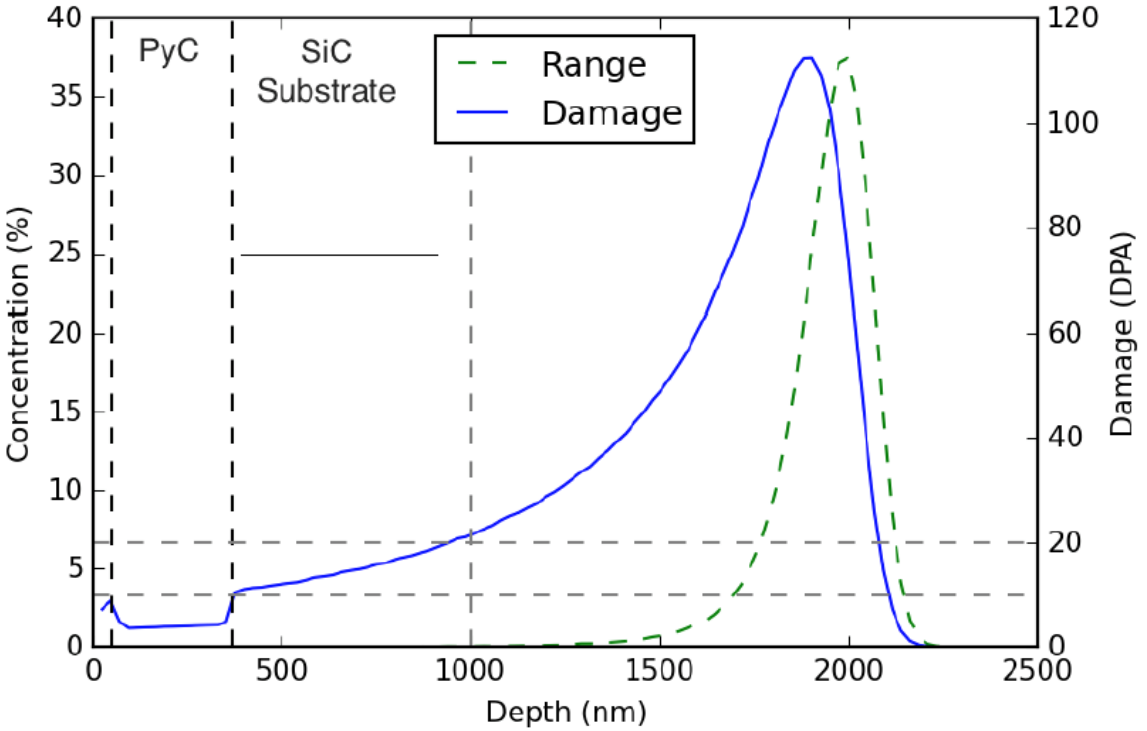
$$s\delta D_{GB} = 2\sqrt{\frac{D_B}{t}}\left(.77 + \frac{\delta}{\sqrt{8D_B t}}\right)^{5/3} m^{-5/3}$$

Dwaraknath, S. S., & Was, G. S. (2016). *Journal of Nuclear Materials, 474*, 76-87.

Ion Irradiation

- Completed diffusion couples will be irradiated at MIBL

- 4.5 MeV Si^{++} ions irradiate diffusion couples at temeperatures between 900 and 1300C
- The exaimnation area is limited to the first 500nm of SiC substrate material due to the damage curve rapidly increaseing thereafter



SRIM calculated damage curve and injected interstitials from 4.5MeV Si^{++} irradiation at MIBL

Comparison of Model and Measurement

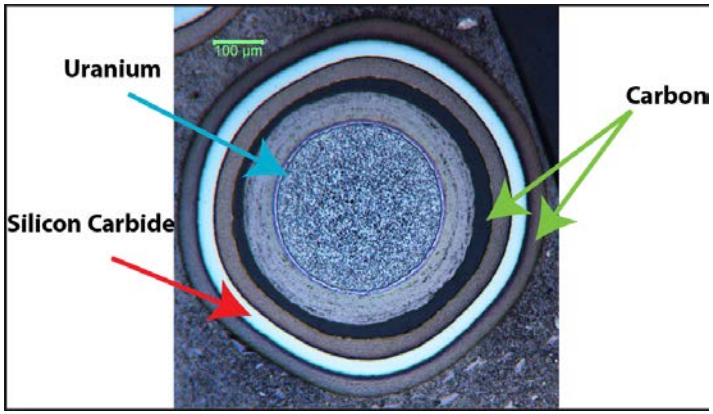
- Modeling efforts will use measured diffusion coefficients, pre-exponential, and activation energy terms to calibrate and verify the models.

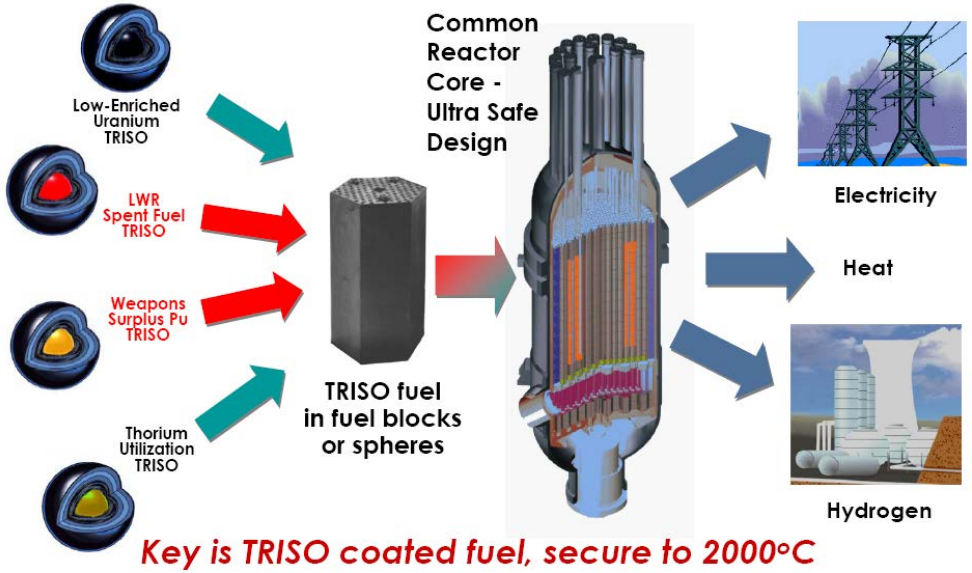
- Modeling will only consider bulk diffusion paths and not any of the proposed fast diffusion path mechanisms.

Thank you

Extra Slides:

TRISO Fuel

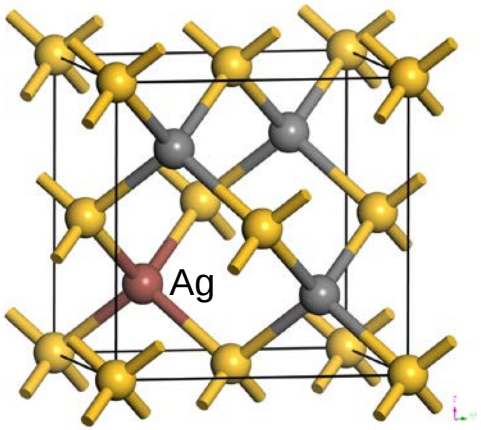




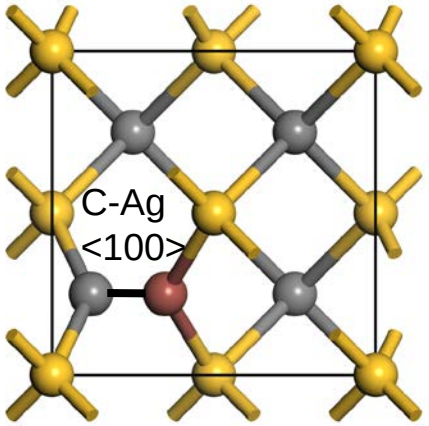
- Thermal and mechanical properties of PyC and SiC under irradiation
- Fission product release from PyC and SiC (i.e. diffusion)
- Interface structures and their changes under irradiation
- Self healing of radiation damage at high temperatures

Possible Defect Configurations

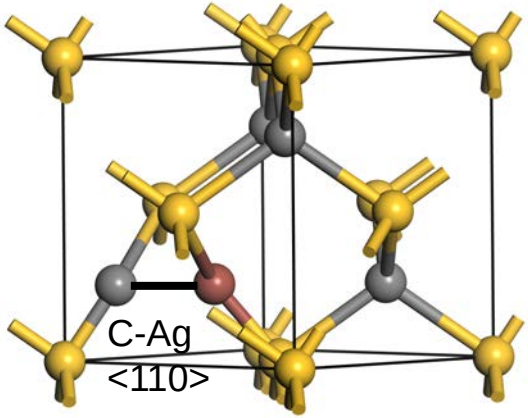
- Defect configurations for FPs in SiC (16 configurations)



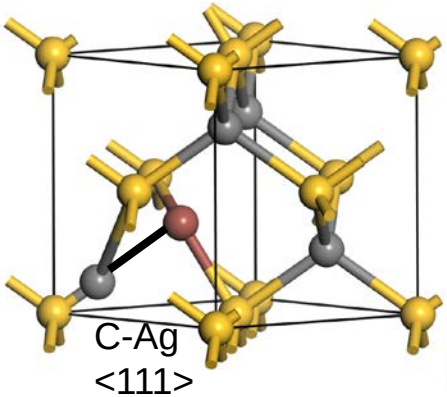
(1) Ag substituting C atom



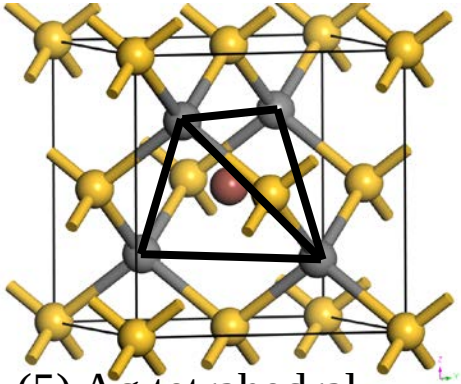
(2) C-Ag <100> interstitial



(3) C-Ag <110> interstitial



(4) C-Ag <111> interstitial



(5) Ag tetrahedral surrounding by four C

- Similar configurations for Si-Ag

C Si Ag (Pd, I, Ru)

Formation Energies

$$\Delta E[V_X] = E_{defect} - E_{perfect} + \mu_X \quad (1)$$

$$\Delta E[Inter] = E_{defect} - E_{perfect} - \mu_I \quad (2)$$

$$\Delta E[I_X] = E_{defect} - E_{perfect} - \mu_I + \mu_X \quad (3)$$

$$\Delta E[I_Y V_X] = E_{defect} - E_{perfect} - \mu_I + \mu_X + \mu_Y \quad (4)$$

$$\Delta E[I_{inter} V_X] = E_{defect} - E_{perfect} - \mu_I + \mu_X \quad (5)$$

Inter: interstitial

I_Y: atom type I substitutes atom Y site (Si or C)

- E_{defect} and $E_{perfect}$ are the total energies of defective (containing defect) and perfect 3C-SiC supercells, respectively.
- The $\mu_{X,Y}$ (X,Y = Si or C) and μ_I (I = Ag, I, Pd and Ru) are the corresponding atomic chemical potentials.

Fitting Interatomic Potentials for Large Scale MD Simulations

❖ Analytical Bond-Order Potential (ABOP)

- The total energy is written as a sum of individual bond energies

$$V = \sum_{i,j} V_{ij} = \frac{1}{2}\sum_{i,j} f_C(r_{ij})\left[V_R(r_{ij}) - b_{ij}V_A(r_{ij})\right],$$

- The pairwise repulsive and attractive contributions

$$V_R(r_{ij}) = \frac{D_0}{S-1}\exp\left[-\beta\sqrt{2S}(r-r_0)\right],$$

$$V_A(r_{ij}) = \frac{SD_0}{S-1}\exp\left[-\beta\sqrt{2/S}(r-r_0)\right],$$

D_0 : dimer energy
r_0 : bond length
R and D specify the cutoff region

- The cutoff function

$$f_C(r_{ij}) \begin{cases} 1, & r < R - D \\ \frac{1}{2} - \frac{1}{2}\sin\left(\frac{\pi}{2}\frac{r-R}{D}\right), & |R-r| \le D \\ 0, & R + D < r \end{cases}$$

- The bond-order contains three-body contributions and angularity

$$b_{ij} = (1 + \chi_{ij})^{-1/2}$$

$$\chi_{ij} = \sum_{k(\neq i,j)} f_C(r_{ik})\exp\left[\alpha_{ijk}(r_{ij} - r_{ik})\right] g(\theta_{ijk})$$

$$g(\theta_{ijk}) = \gamma\left(1 + \frac{c^2}{d^2} - \frac{c^2}{d^2 + \left[h + \cos\theta_{ijk}\right]^2}\right)$$

Fitting Interatomic Potentials for Large Scale MD Simulations

Potential parameters I for Ag-SiC system

	Ag	Ag-Si	Ag-C	Si *	Si-C *	C *
D_0 (eV)	1.82805675551	2.00361170132	2.02026077226	3.24	4.36	6.00
r_0 (Å)	2.58163271708	2.31460474819	2.04753165865	2.232	1.79	1.4276
S	3.68316712324	1.69016805103	3.84175420199	1.842	1.847	2.167
β	1.51806072803	1.32471791917	1.33976920179	1.4761	1.6991	2.0099
γ	0.44489696600	0.14753865159	0.07312191123	0.114354	0.011877	0.11233
c	1.03289678667	21.0375388795	1.10814959544	2.00494	273987	181.910
d	0.25330171412	60.3175649975	0.77396268822	0.81472	180.314	6.28433
h	-0.2099199277	0.21868016212	0.92541289514	0.259	0.68	0.5556
R (Å)	3.56075816554	3.03639741646	3.51882471597	2.82	2.40	2.00
D (Å)	0.28147532635	0.74749292842	0.37543761698	0.14	0.20	0.15
α ($Å^{-1}$)	0.38016638168	See below	See below	0.0	0.0	0.0

α Value

	α ($Å^{-1}$)		α ($Å^{-1}$)		α ($Å^{-1}$)
Ag-Si-Si	0.90875582360	Ag-C-C	0.70452852140	Ag-Si-C	0.13703028988
Si-Ag-Si	0.86747096045	C-Ag-C	-0.41177361740	Ag-C-Si	-0.00435439830
Si-Si-Ag	-4.48446346689	C-C-Ag	1.40005035436	Si-Ag-C	3.24603971382
Ag-Ag-Si	/	Ag-Ag-C	/	C-Ag-Si	0.85566412456
Ag-Si-Ag	/	Ag-C-Ag	/	Si-C-Ag	0.61791114755
Si-Ag-Ag	/	C-Ag-Ag	/	C-Si-Ag	1.41610646065

*P. Erhart and A. Karsten. *Physical Review B* **71** 035211 (2005).

Fitting Interatomic Potentials for Large Scale MD Simulations

Potential parameters II for Ag-SiC system

	Ag	Ag-Si	Ag-C	Si *	Si-C *	C *
D_0 (eV)	1.82805675551	1.76530700202	2.04676834038	3.24	4.36	6.00
r_0 (Å)	2.58163271708	2.31004494932	2.09569535931	2.232	1.79	1.4276
S	3.68316712324	1.59731414291	3.79038110462	1.842	1.847	2.167
β	1.51806072803	1.37806767557	1.32888262245	1.4761	1.6991	2.0099
γ	0.44489696600	0.14443642296	0.07481070830	0.114354	0.011877	0.11233
c	1.03289678667	13.7168847218	1.14093201064	2.00494	273987	181.910
d	0.25330171412	31.0911197658	0.82596547844	0.81472	180.314	6.28433
h	-0.2099199277	0.53294763514	0.91112477995	0.259	0.68	0.5556
R (Å)	3.56075816554	2.99592317204	3.53670395711	2.82	2.40	2.00
D (Å)	0.28147532635	0.75625242892	0.41712091595	0.14	0.20	0.15
α ($Å^{-1}$)	0.38016638168	See below	See below	0.0	0.0	0.0

α Value

	α ($Å^{-1}$)		α ($Å^{-1}$)		α ($Å^{-1}$)
Ag-Si-Si	0.31933455028	Ag-C-C	0.33490170717	Ag-Si-C	0.19620102134
Si-Ag-Si	5.20015949840	C-Ag-C	0.19340750853	Ag-C-Si	1.29133813232
Si-Si-Ag	-5.83803531968	C-C-Ag	1.96551206210	Si-Ag-C	3.41432591389
Ag-Ag-Si	/	Ag-Ag-C	/	C-Ag-Si	1.04300805038
Ag-Si-Ag	/	Ag-C-Ag	/	Si-C-Ag	0.60673325872
Si-Ag-Ag	/	C-Ag-Ag	/	C-Si-Ag	1.1088644175

*P. Erhart and A. Karsten. *Physical Review B* **71** 035211 (2005).

Fitting Interatomic Potentials for Large Scale MD Simulations

Potential parameters III for Ag-SiC system

	Ag	Ag-Si	Ag-C	Si *	Si-C *	C *
D_0 (eV)	1.82805675551	1.77460786961	2.05591711295	3.24	4.36	6.00
r_0 (Å)	2.58163271708	2.30648177310	2.15069165013	2.232	1.79	1.4276
S	3.68316712324	1.58058143760	3.77395223051	1.842	1.847	2.167
β	1.51806072803	1.37556514236	1.37422386740	1.4761	1.6991	2.0099
γ	0.44489696600	0.14375484933	0.07436272452	0.114354	0.011877	0.11233
c	1.03289678667	13.8110154592	1.13652646306	2.00494	273987	181.910
d	0.25330171412	40.5313077227	0.85750986240	0.81472	180.314	6.28433
h	-0.2099199277	0. 5356310440	0.91020523039	0.259	0.68	0.5556
R (Å)	3.56075816554	2.97511183183	3.50726464574	2.82	2.40	2.00
D (Å)	0.28147532635	0.75061127085	0.41141565201	0.14	0.20	0.15
α ($Å^{-1}$)	0.38016638168	See below	See below	0.0	0.0	0.0

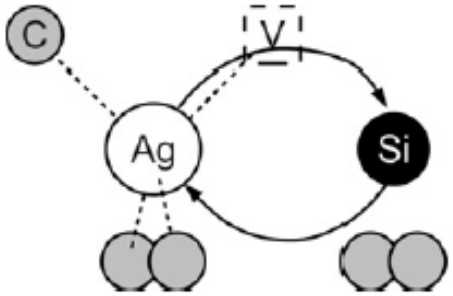
α Value

	α ($Å^{-1}$)		α ($Å^{-1}$)		α ($Å^{-1}$)
Ag-Si-Si	0.24808988231	Ag-C-C	0.32341524608	Ag-Si-C	0.50643415266
Si-Ag-Si	4.77766170545	C-Ag-C	0.725877408010	Ag-C-Si	1.10701484269
Si-Si-Ag	-4.5548576851	C-C-Ag	1.85770117131	Si-Ag-C	3.50873767128
Ag-Ag-Si	/	Ag-Ag-C	/	C-Ag-Si	2.15170649873
Ag-Si-Ag	/	Ag-C-Ag	/	Si-C-Ag	0.82367476374
Si-Ag-Ag	/	C-Ag-Ag	/	C-Si-Ag	1.23959010268

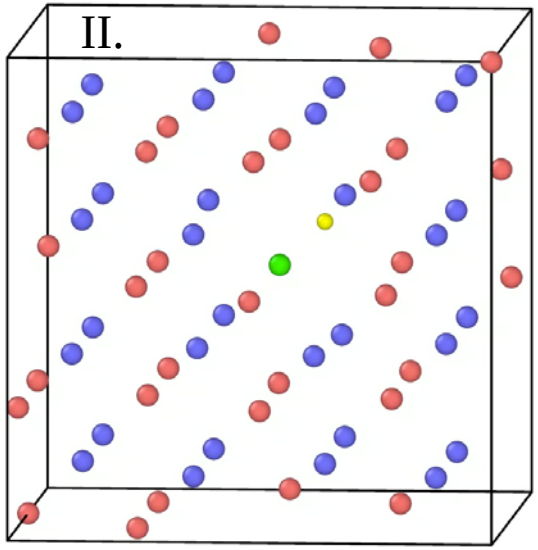
*P. Erhart and A. Karsten. *Physical Review B* **71** 035211 (2005).

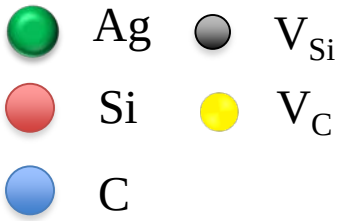
Ag_Si-Cvac

I. Direct swap

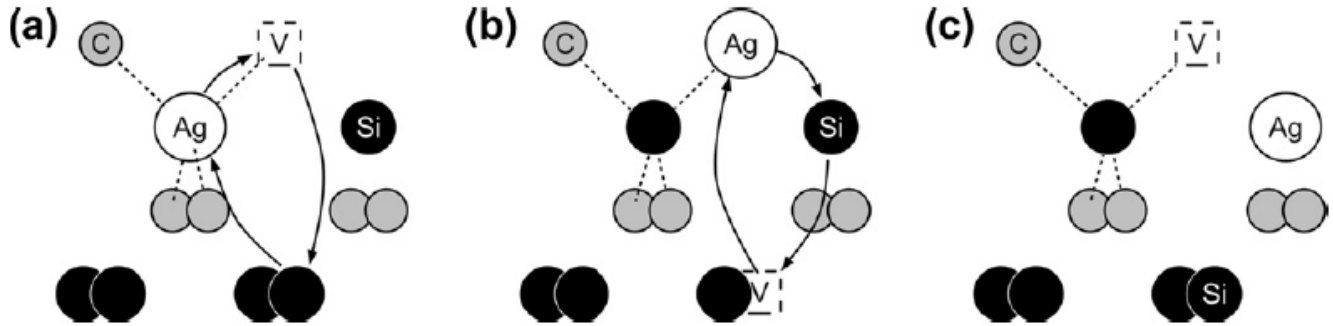


II.





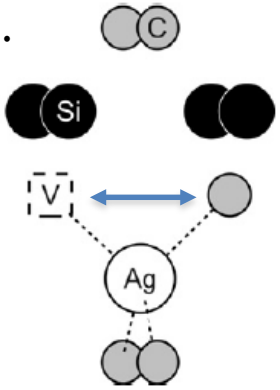
II. Two Ag hops



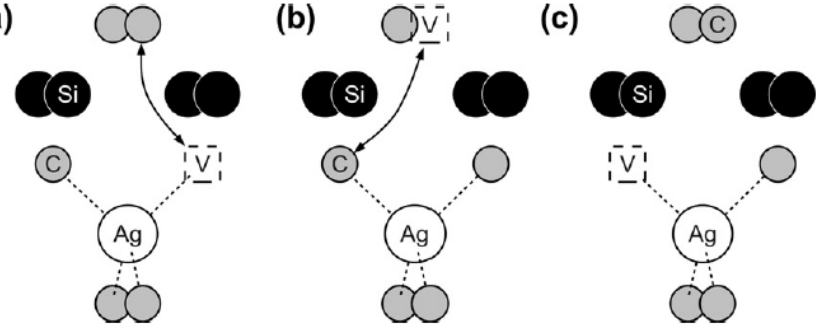
	Shrader	ABOP I	ABOP II	ABOP III
I.direct swap	9.1	10.85	12.82	12.21
II. two Ag hops	9.2	11.64	12.83	12.72

Ag_Si-Cvac

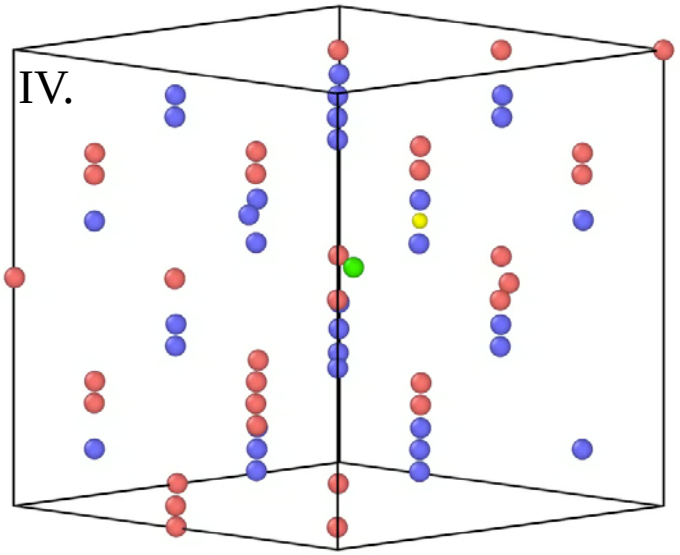
III.



IV.



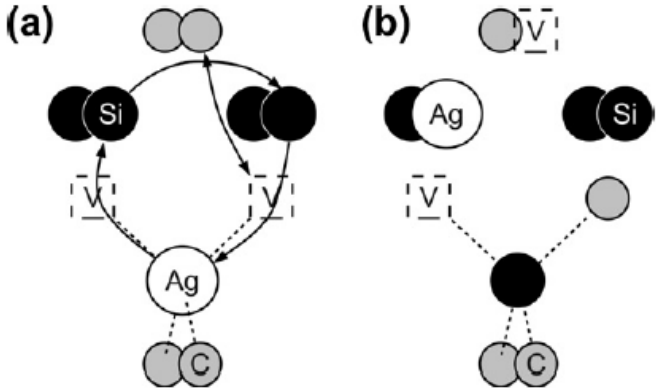
IV.



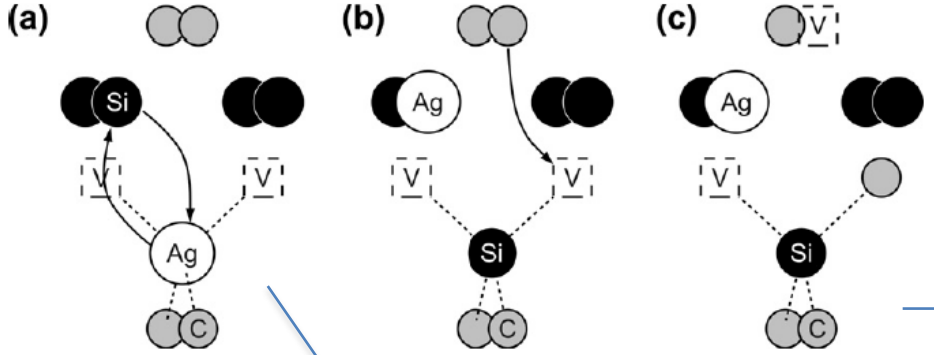
	Shrader	ABOP I	ABOP II	ABOP III
III.reo1	8.02	6.68	6.62	6.50
IV.reo2	5.97	6.55	6.53	6.17

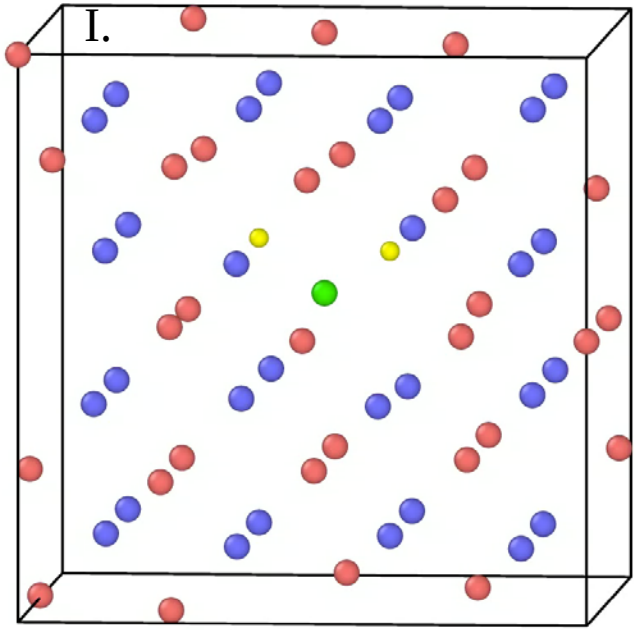
Ag_Si-2Cvac

I. Ring diffusion



II. Swap and C-Move





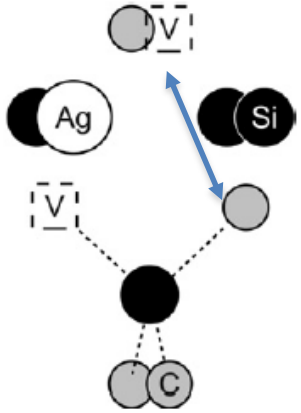
Barrier from first step. Consider as lower bound barrier

	Shrader	ABOP I	ABOP II	ABOP III
I. ring	≥ 6.61	9.51	9.06	9.19
II. swap+Cmove	7.04	6.34	6.00	7.46

Ag_Si-2Cvac

III. Single reorientation hop: moving a C vacancy around the Ag atom.

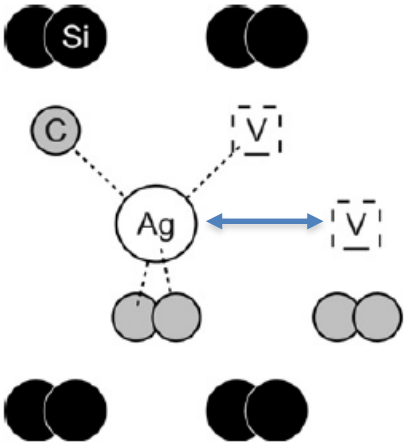
III.



	Shrader	ABOP I	ABOP II	ABOP III
III. single reo	5.02	5.47	5.52	5.52

Ag_Si-$V_{Si}V_C$

- Ag swaps with the V_{Si}



	Shrader	ABOP I	ABOP II	ABOP III
Ag swap with V_{Si}	0.58	0.20	0.37	0.55