He behavior in tantalum

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Outline

- ✓ Motivation
- ✓ Helium behavior in Ta
- ✓ The comparison of helium behavior in Ta and W
- ✓ Summary

Motivation

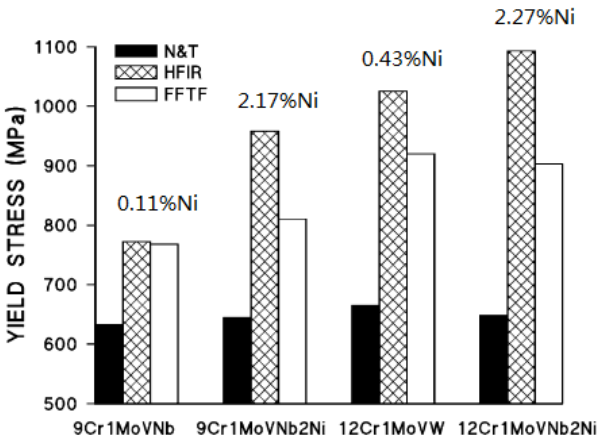
Why we are interested in He behavior?

Mechanical properties of neutron-irradiated nickel-containing martensitic steel: I Experimental study II Review and analysis of helium-effects studies R.L.Klueh, N.Hashimoto, M.A.Sokolov, et.al, JNM 357(2006)156-168 169-182

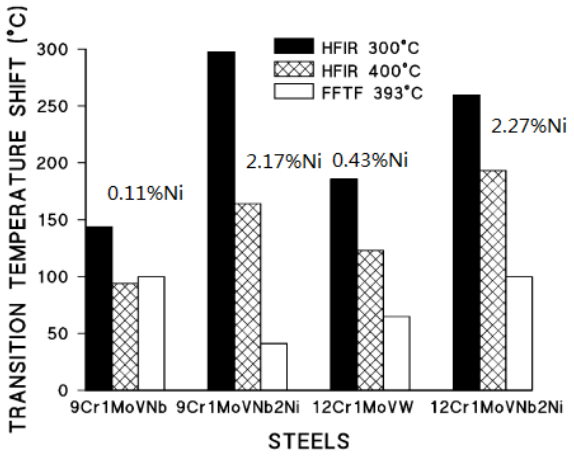
Composition of the steels (wt%)

Element[a]	9Cr–1MoVNb		12Cr–1MoVW	
	Standard	2% Ni	Standard	2% Ni
C	0.09	0.064	0.21	0.20
Mn	0.36	0.36	0.50	0.49
P	0.008	0.008	0.011	0.011
S	0.004	0.004	0.004	0.004
Si	0.08	0.08	0.18	0.14
Ni	0.11	2.17	0.43	2.27
Cr	8.62	8.57	11.99	11.71
Mo	0.98	0.98	0.93	1.02
V	0.209	0.222	0.27	0.31
Nb	0.063	0.066	0.018	0.015
Ti	0.002	0.002	0.003	0.003
Co	0.013	0.015	0.017	0.021
Cu	0.03	0.04	0.05	0.05
Al	0.013	0.015	0.030	0.028
W	0.01	0.01	0.54	0.54
As	<0.001	<0.001	<0.001	<0.002
Sn	0.003	0.003	0.003	0.002
N	0.050	0.053	0.020	0.017
O	0.007	0.006	0.005	0.007

$$Ni^{58}(n,\gamma) \rightarrow Ni^{59}(n,\alpha) \rightarrow Fe^{56}$$



A comparison of the yield stress of steel in the N&T condition and after irradiation in HFIR and FFTF.

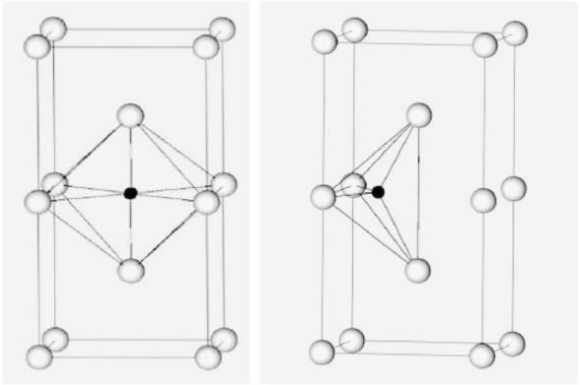


Shift in ductile-brittle transition temperature of steels after irradiation in HFIR at 300 and 400°C and in FFTF at 393°C

Motivation

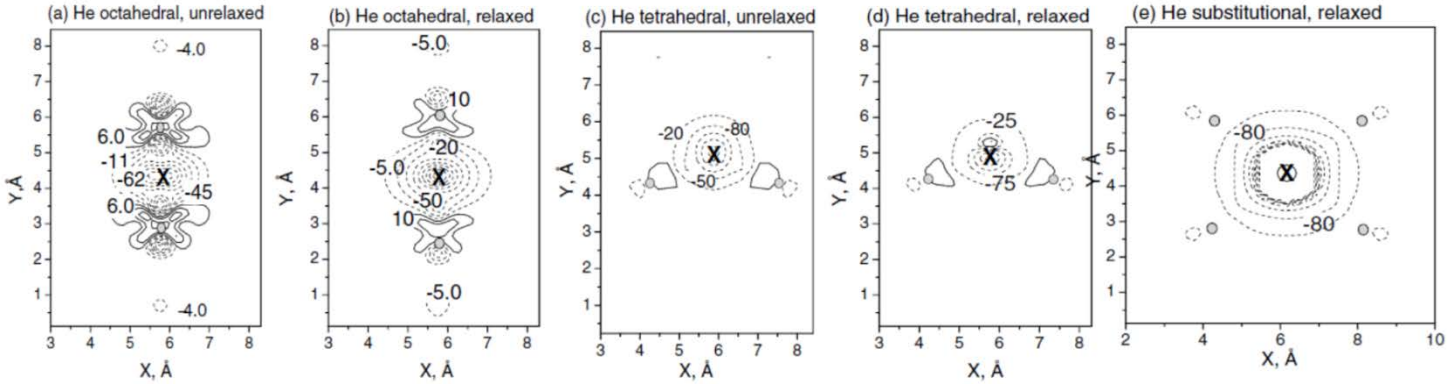
Ab initio Study of He behavior

T.Seletskaia, Y.Osetsky, R.E.Stoller and G.M.Stocks, PRL 94, 046403(2005) C.C.Fu and F.Willaime PRB 72, 064117(2005)



	He	1st Fe		2nd Fe	
	μ (μ_B)	r (Å)	μ (μ_B)	r (Å)	μ (μ_B)
octa, unrelaxed	0.012	1.41	1.67	1.99	2.17
octa, relaxed	0.015	1.66	2.01	2.09	2.24
tetra, unrelaxed	0.007	1.58	1.99	2.54	2.17
tetra, relaxed	0.012	1.76	2.15	2.57	2.21
sub, unrelaxed	0.000	2.45	2.25	2.83	2.08
sub, relaxed	0.000	2.47	2.29	2.85	2.11

He strongly resists spin polarization, however it does produce significant changes in magnetic moment of 1st Fe and 2nd Fe



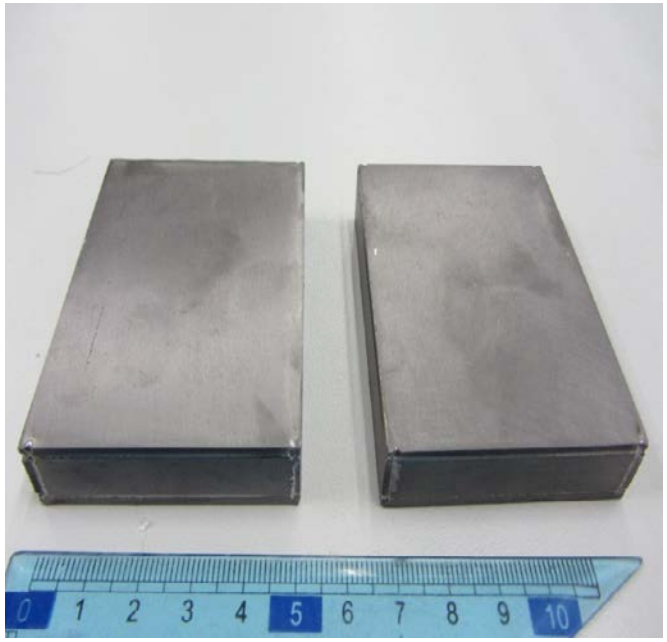
Both interstitials and their 1st neighbor Fe atoms are strongly polarized, and the trend is *Sub<Tetra<Octa*

Motivation

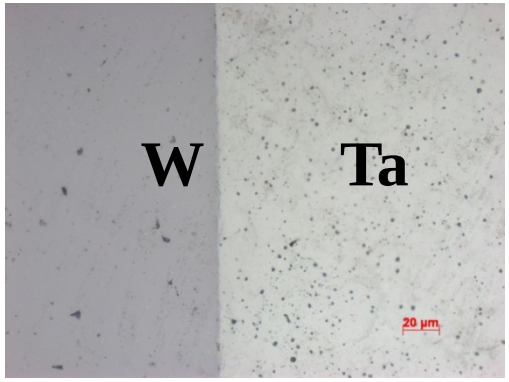
✓ Ta can yield many neutrons under spallation conditions that are comparable to tungsten.

✓Ta is renowned for its chemical stability; In fact, at temperature below 150ºC tantalum almost completely immune to attack by the normally aggressive acids.

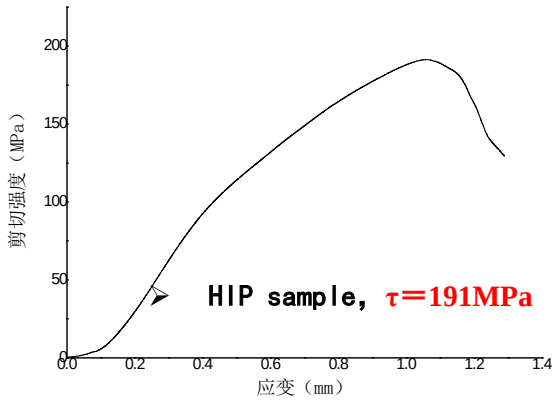
☐ The activity and afterheat of Ta is much higher than that of W under the same irradiation condition.



Hot isostatic Pressing sample



Interface between W target and Ta cladding

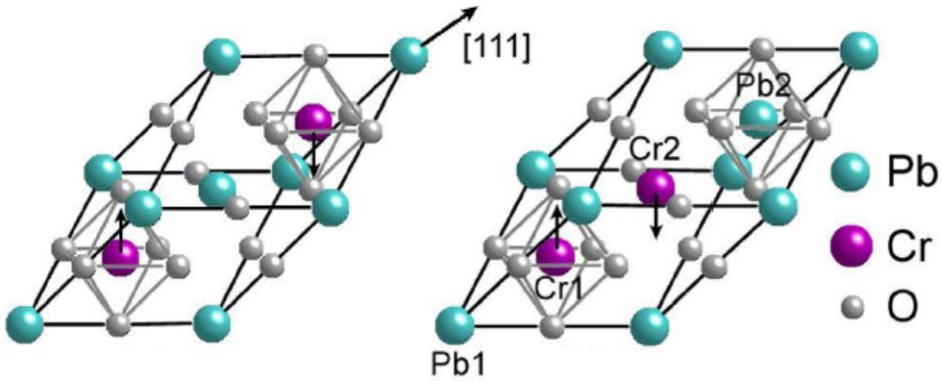


✓ 11 tungsten plates
✓ 170*70mm with different thickness
✓ 1.2mm cooling channel
✓ 0.3mm tantalum cladding
✓ Using HIP technology for cladding Ta

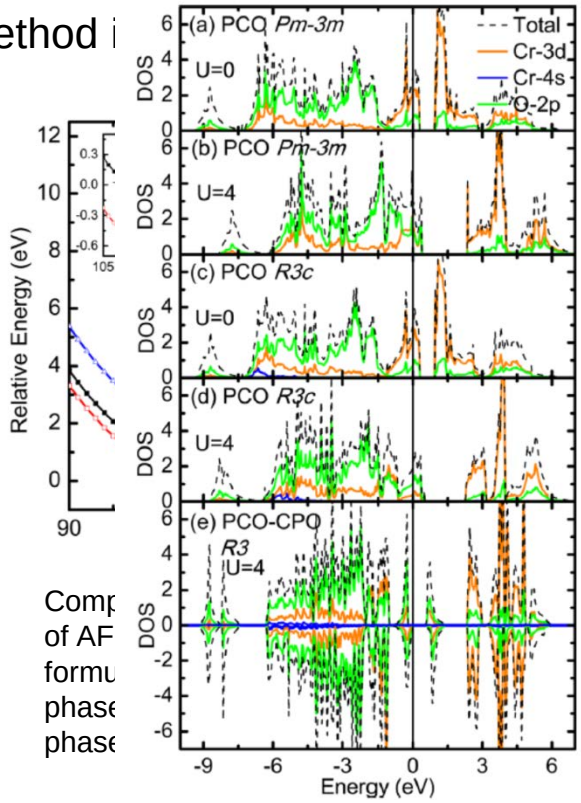
He behavior in Ta

Calculation Method

- The calculations had been based on the frozen-core projected augmented wave (PAW) method within the Vienna *ab initio* Simulation Package VASP.
- The exchange and correlation effects are described by the Density Functional Theory (DFT) within the Generalized Gradient Approximation (GGA) of Perdew and Wang.
- The climbing image nudged elastic band (CINEB) method i ...imum energy path (MEP).



Cubic PCO in AFM configuration



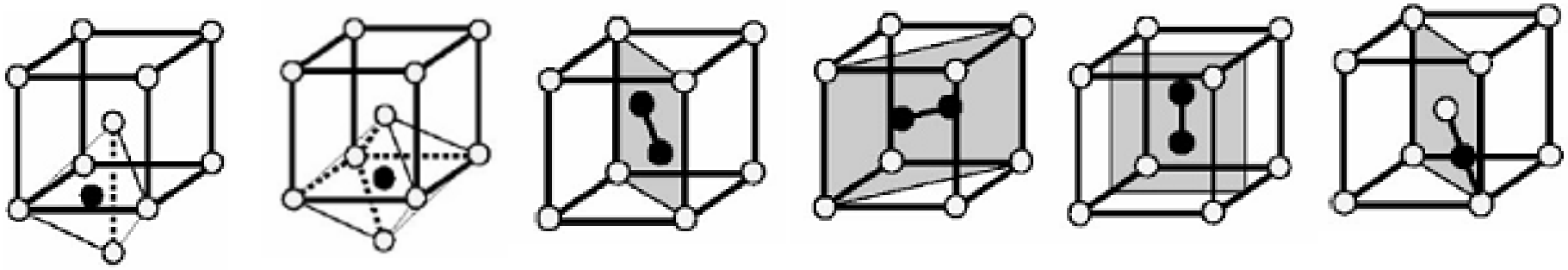
Cubic PCO-CPO in AFM configuration

Comp ...s cells
of AF ...
formu ...nd R3
phase ...m-3m
phase ...

Bao-Tian Wang, Wen Yin, Wei-dong Li, and Fang-Wei Wang, J. Appl. Phys. 111,013503 (2012)

He behavior in Ta

Defect formation energies in tantalum



Tetrahedral | Octahedral | <111> SIA | <110> SIA | <100> SIA | Crowdion

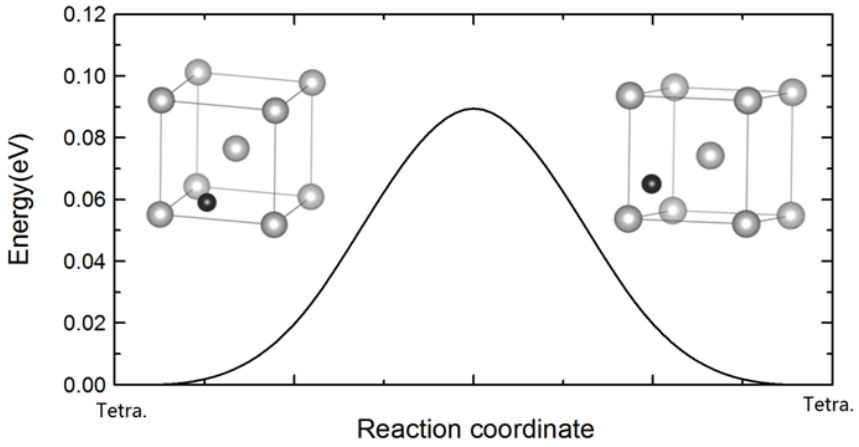
Formation energies (in eV) of He, H, vacancy and self-interstitial defects with the different configurations in tantalum. The crowdion SIA (not represented) is nearly degenerate with the <111> configuration. The calculation were done using 54 atom super-cell and 125 k points and 128 atom and 27 k points

Configuration	Octahedral	Tetrahedral	substitution	<111>	<110>	<100>
Helium	3.65	3.34	4.58			
Hydrogen	-3.70	-3.37	-0.08			
SIA	5.49	5.30		4.32	4.96	5.41
Vacancy			3.27			

- ✓ The predicted favored interstitial site of helium in Ta is tetrahedral interstitial site (TIS) and the energy differences between octahedral and tetrahedral is 0.31 eV.
- ✓ The formation energy of hydrogen in Ta is negative, which means tantalum can absorb hydrogen atoms.

He behavior in Ta

He migrations in tantalum



✓ A tetrahedral He may migrate between two equivalent sites without passing through an octahedral one.
✓ Such a low migration energy 0.09 eV means that the migration of interstitial He is almost athermal.
✓ It will encounter other defect such as another He atom, hydrogen atom, vacancy or SIA.

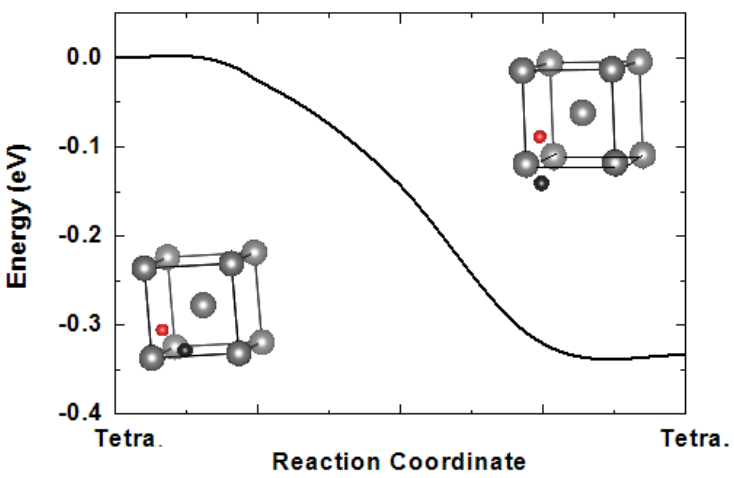
The binding energies (in eV) and distance variation between helium and other defects. ***a*** is the lattice parameters.

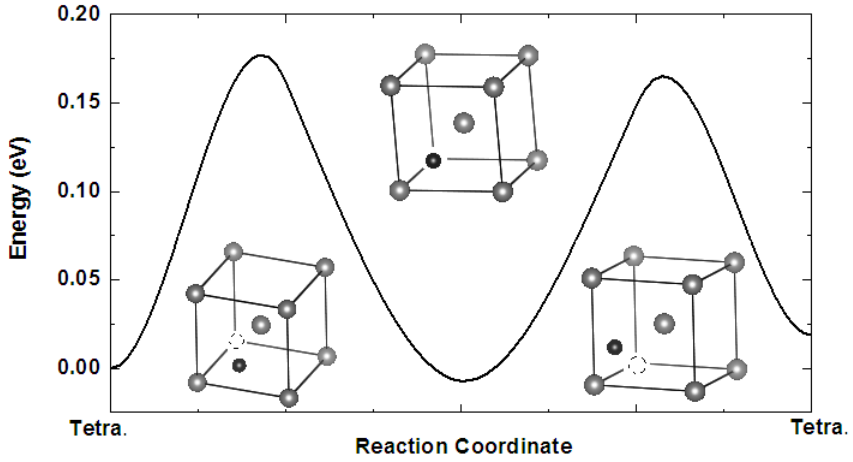
The binding energy between helium and the other defects			
Helium	Hydrogen	Vacancy	SIA
-0.14	-0.48	2.03	-0.31
The initial distance between helium and the other defects			
Helium	Hydrogen	Vacancy	SIA
0.354a	0.354a	0.559a	0.560a
The final distance between helium and the other defects after relaxation			
Helium	Hydrogen	Vacancy	SIA
0.464a	0.584a	0.0110a	1.030a

✓ The attraction between a vacancy and a helium atom is about 2.03 eV.
✓ The helium atom is trapped by the vacancy, which may explain why very small diffusion constants of helium in Ta are obtained in the previous experiments. (F.Zielinski, J.M.Costantini, J.Haussy, and F.Durbin, Journal of Nuclear Materials 312 (2003) 141)

He behavior in Ta

He migrations in tantalum in presence of other defects

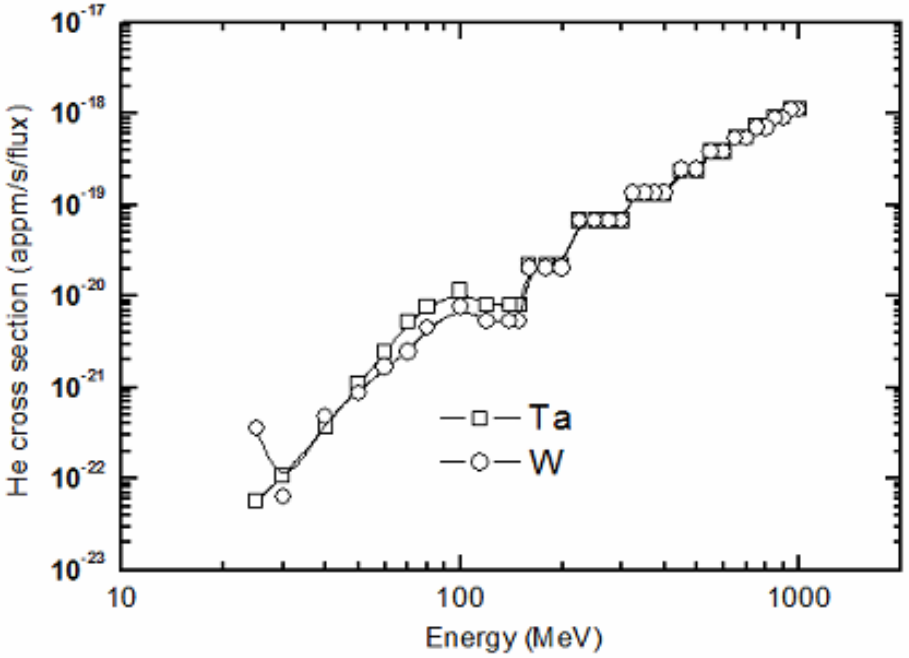




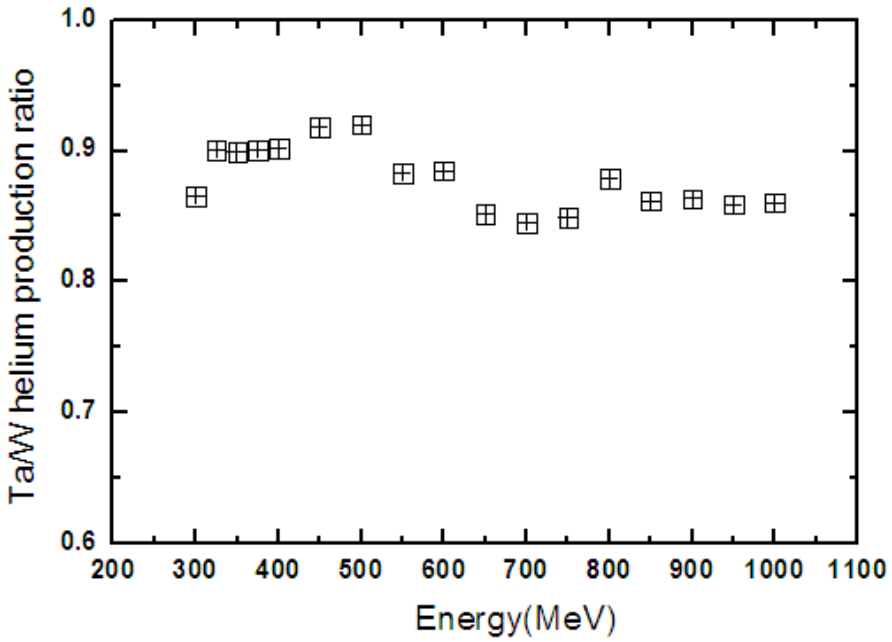
Migration path of He atom in presence of a hydrogen atom (left) or a vacancy (right)

- The repulsive energy between He and H atoms pushes He atom away from the H atom; He atom shows it will jump into the nearest TIS without any energy barrier.
- The strong attractive interaction energy between He and the Vacancy makes the vacancy is a trap site.

The comparison of the He production rate between tantalum and tungsten



He production cross sections in Ta and W (data from HILO2K)



He production ratio when the two metals are bombarded by the different energies neutrons (calculated by MCNPX 2.5.0)

Tantalum has almost the same He production under the different neutron energies.

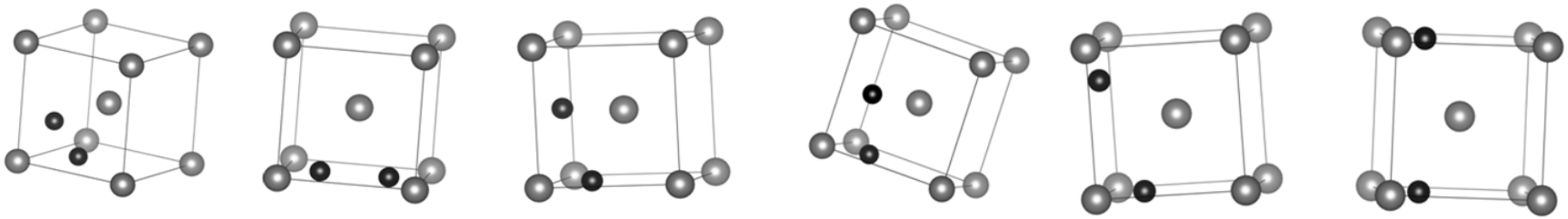
The comparison of the He formation energy between tantalum and tungsten

Formation energy (eV) for a single He in Octa. or Tetra. Interstitial sites as well as in substitution. The calculation were done 128 atom and 27 k points , * data from Ref. Charlotte S. Becquart and Christophe Domain, PRL97,196402 (2006)

Configuration	Octahedral	Tetrahedral	Substitution	Migration energy
Tantalum	3.65	3.34	4.58	0.09
Tungsten	6.34	6.11	4.65	0.06
Tungsten*	6.38	6.16	4.70	0.06

- ✓ The predicted favored interstitial sites of helium in Ta and W are tetrahedral sites.
- ✓ The migration energies of He in Ta and W are lower than 0.1 eV which means He is easy to move in these two metals.

The comparison of the He-He interaction between tantalum and tungsten



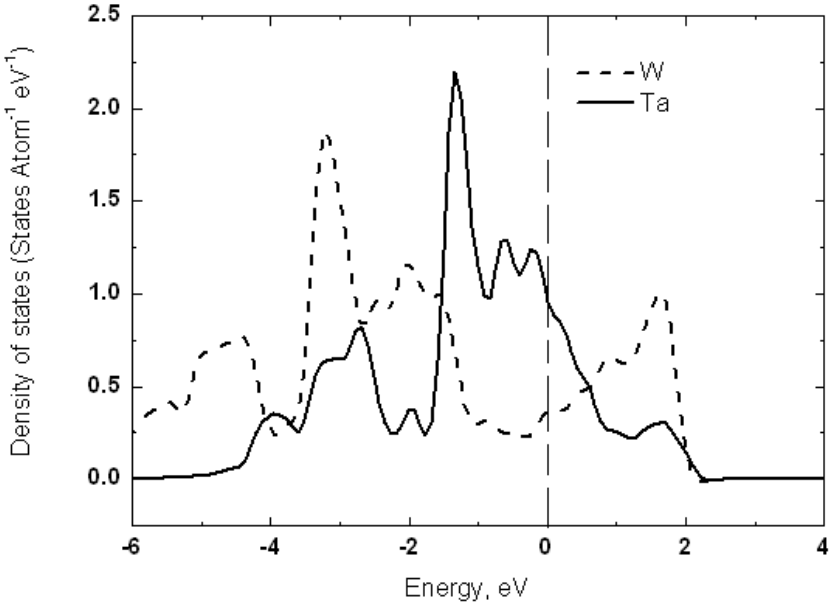
0.354a *0.5a* *0.612a* *0.707a* *0.791a* *a*

The binding energies (in eV) in tantalum and tungsten. ***a*** is the lattice parameters. * data from PRL97,196402 (2006)

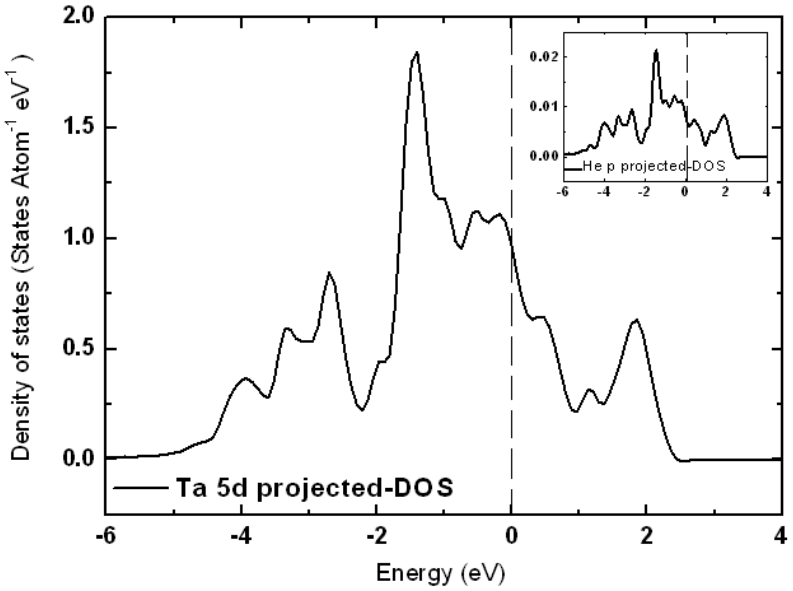
	The He-He binding energy					
He-He Initial distance	0.354*a*	0.5*a*	0.612*a*	0.707*a*	0.791*a*	*a*
Tantalum/ 54atoms	-0.14	0.012	-0.048	-0.15	-0.13	-0.23
Tantalum/ 128atoms	-0.10	0.050	0.022	-0.090	-0.080	-0.17
Tungsten/ 54atoms	0.72	0.82	1.14	0.48	1.11	0.07
Tungsten/ 54atoms*	0.68	0.87	0.98	0.29	0.96	-0.06
Tunagsten/ 128atoms	0.71	0.91	1.00	0.30	1.00	-0.05
Tunagsten/128 atoms*	0.74	0.94	1.01	0.31	1.03	-0.04

The interaction between the two He atoms in tantalum shows repulsive or weak repulsive interaction other than attractive interaction in tungsten. The repulsive or weak repulsive interaction indicates the two He atoms are more difficult to become a cluster in tantalum than in tungsten and the He cluster is much less stable in tantalum than in tungsten.

The comparison of the He-He interaction between tantalum and tungsten



Comparison of the 5d site-projected DOS between tantalum and tungsten. The Fermi energy is set as 0 eV.



5d site-projected DOS of Ta atom. The *p* site-projected DOS of the corresponding He atoms are shown in the inset of the figures. The Fermi energy is set as 0 eV.

✓ For pure tantalum, there exists a sharp decrease of the DOS just around the Fermi level and the Fermi level lies at the maximum of the sharp peak;

✓ The overall similarity in the shapes of Ta *5d* and He *p* projected DOS reflects the strong hybridization of Ta and He atoms, which suggests there is a strong electronic interaction between Ta and He atoms.

Summary

- ✓ He atoms will stay in the tetrahedral site in tantalum.
- ✓ The tetrahedral He atoms will migrate between two equivalent sites, which imply low effective migration energy for tantalum.
- ✓ The strong attractive interaction energy between He and the Vacancy makes the vacancy in tantalum is a trap site.
- ✓ The interaction between He atoms in tantalum are much weaker than that in tungsten, which maybe due to the sharp drop of the Ta *5d* projected density of states around Fermi level . It means although the production of He atoms in these two heavy metals are similar, it is more difficult for tantalum to form the He clusters.

Thanks for your attention!