



Understanding the effect of crystal anisotropy on grain growth, texturing and transport via the orthorhombic β -U system

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Changing the World's Energy Future

Andrea M Jokisaari, Benjamin Beeler, Michael Tonks, Fidelma Giulia Di Lemma, Aashique Rezwan, Yuhao Wang, Tiankai Yao



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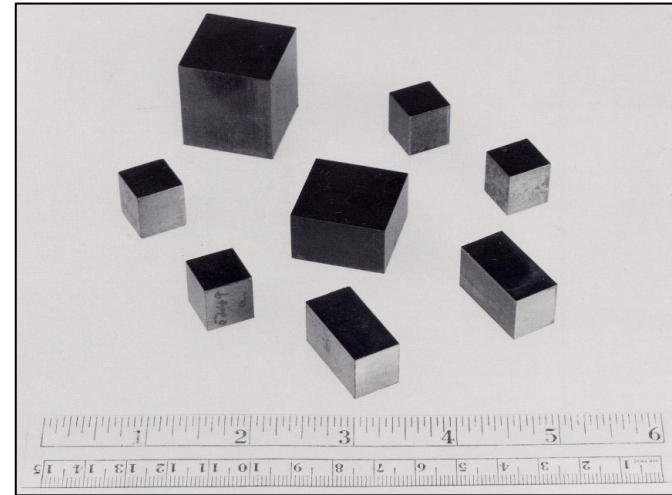
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Understanding the effect of crystal anisotropy on grain growth, texturing and transport via the orthorhombic α -U system

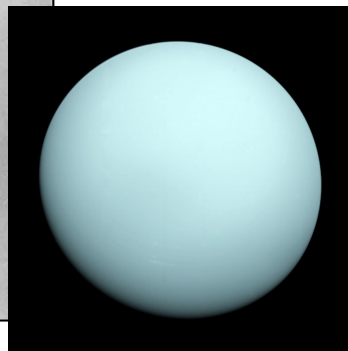
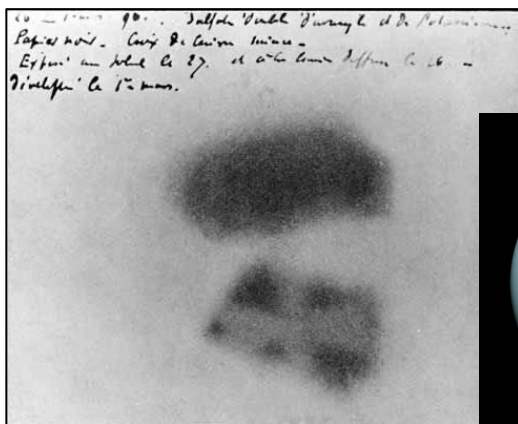
TMS 2023
March 19-23, 2023

A dive into the world of uranium

- Uranium is the last element on the periodic table to naturally occur on Earth
- Isotopes of U of engineering importance: U-238, U-235, U-233
- Density: 19.1 g/cm³
- Several physical properties of α -U are extremely anisotropic

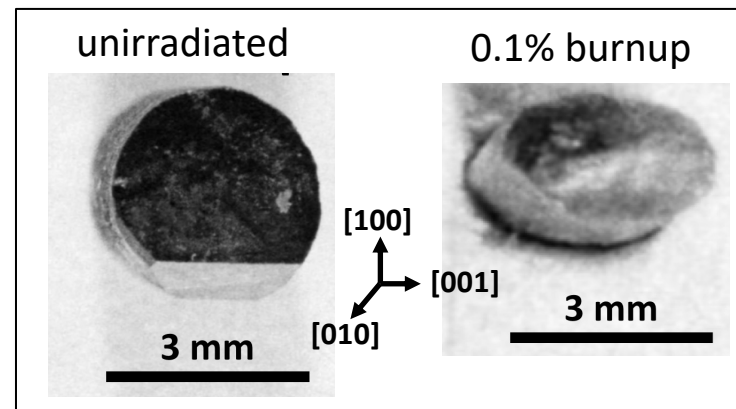
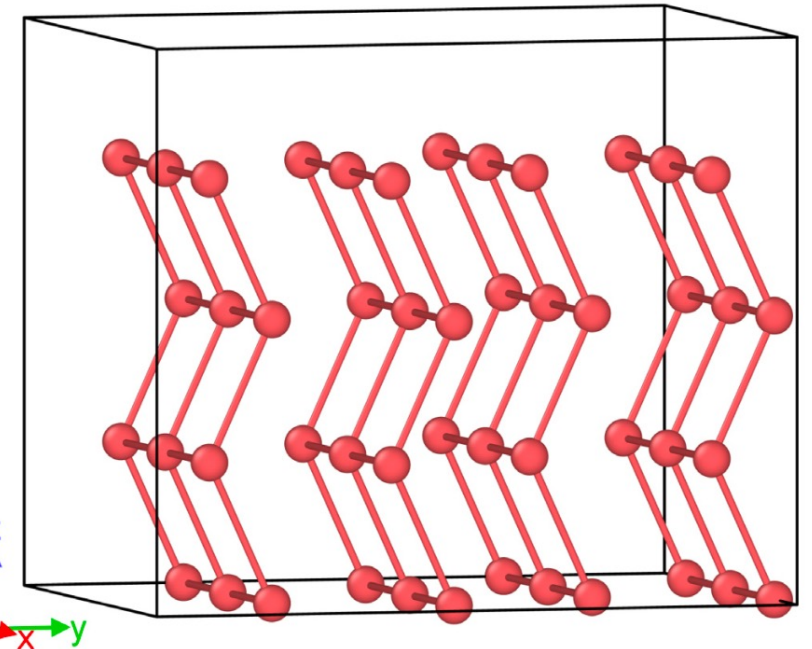
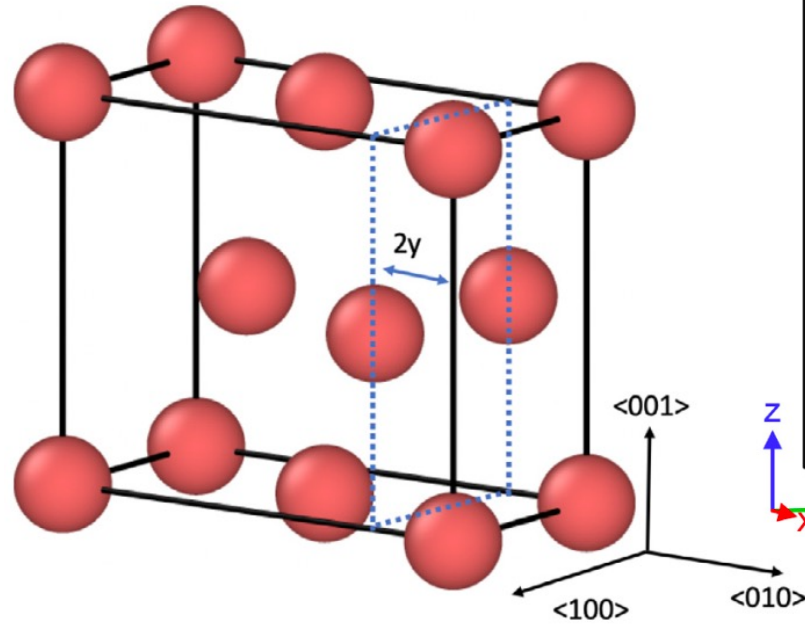


1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	* 71 Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	* 103 Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og
		* 57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb		
		* 89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No		

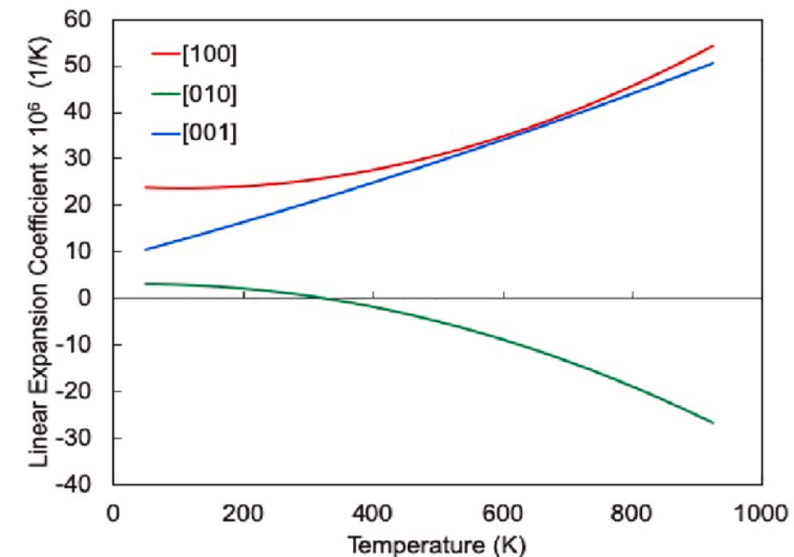


A dive into the world of uranium

- α -U crystal structure is orthorhombic and can be thought of as stacks of corrugated atom planes
- Several physical properties of α -U are extremely anisotropic
 - Thermal expansion
 - Electrical conductivity
 - Modulus of elasticity
 - Irradiation growth

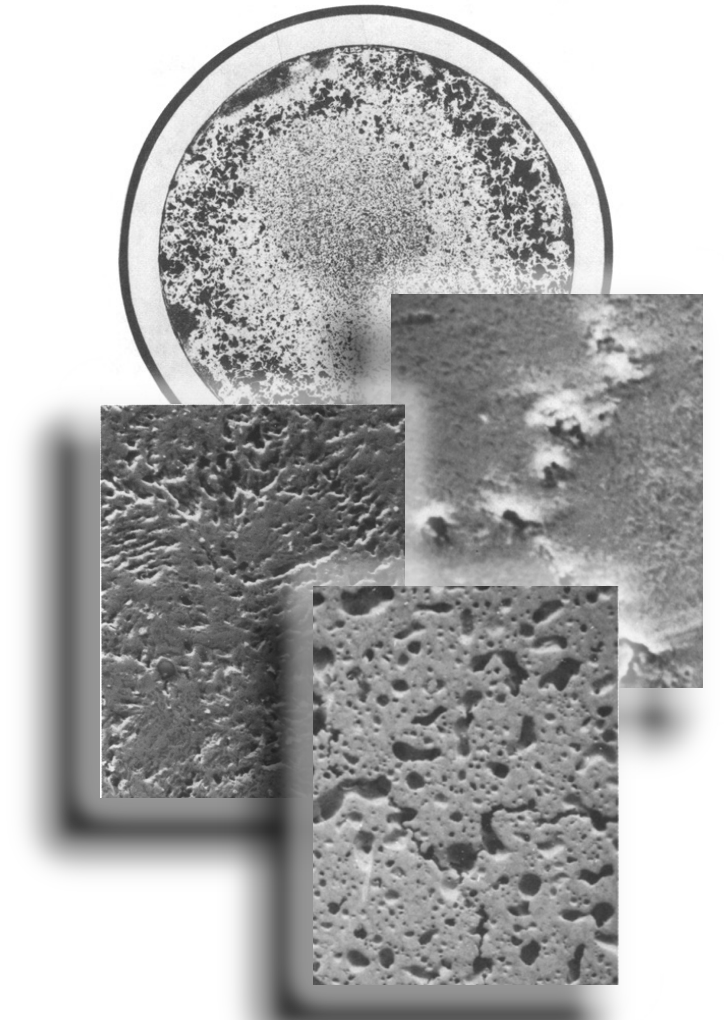


Paine, ANL-5676, 1958



α -Uranium is a strange metal, but why do we care?

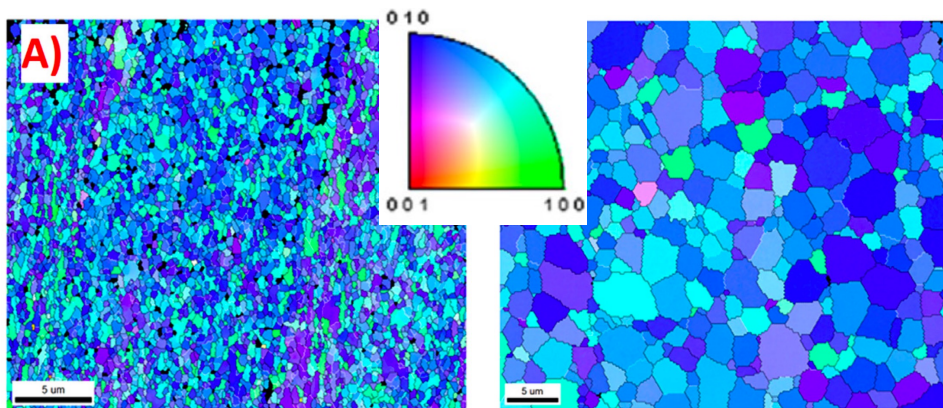
- U can be used in kinetic energy penetrators and armor plating
- Aircraft counterweights, radiation shielding, neutron reflectors
- α -U is one phase in certain metallic nuclear fuels (e.g., U-Zr)
 - Impacts fuel swelling behavior, which can limit fuel lifetime
 - Current predictive capability of its behavior is limited
 - Heyday of α -uranium research was in the 1960s
- Theoretical understanding of α -U thermophysical behavior is needed



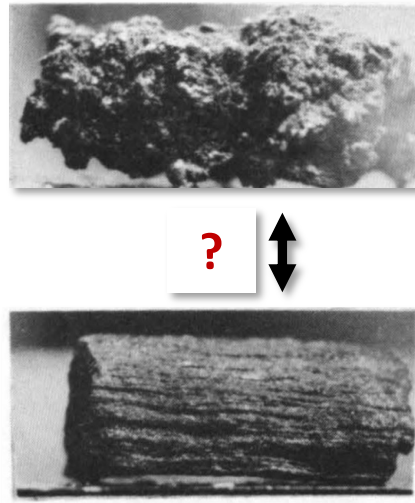
Met. Trans. A 21A (1990) 517

α -Uranium can really change in service

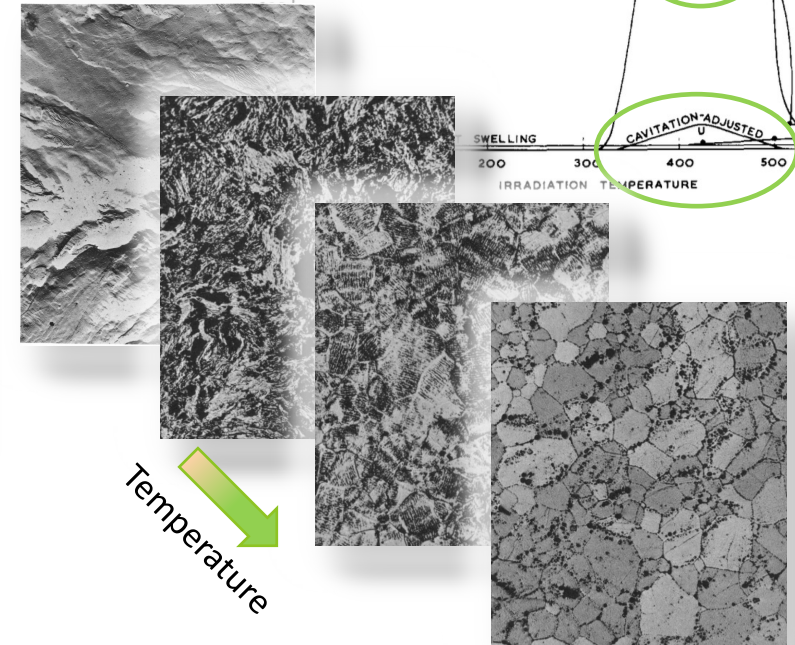
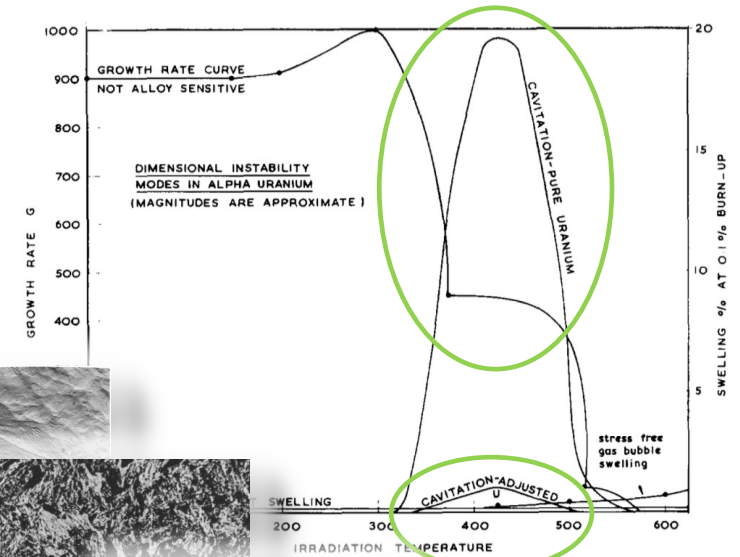
- Thermal cycling experiments on polycrystals demonstrated sample elongation and porosity development
- Irradiation of polycrystals shows an array of deformation and porosity development behavior
- Radiation annealing experiments show texture is maintained during grain growth



JNM 542 (2020) 152467



Leggett, HW-79559, 1963



Acta Metall. 23 (1975) 101

Mesoscale studies shed light on complex polycrystalline behaviors

- Irradiation growth and thermal expansion are two hypothesized causes of observed irradiation damage behavior in α -U materials
- Mesoscale modeling is particularly important to understand highly anisotropic materials
 - Directionally-dependent properties
 - Aggregate behaviors
 - Feedback
- Can still draw useful conclusions about microstructure evolution using macroscopic single-crystal properties without atomistic mechanisms

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Irradiation-induced internal stresses in polycrystalline α -uranium: a mesoscale mechanical approach

A.M. Jokisaari

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Modeling brittle fracture due to anisotropic thermal expansion in polycrystalline materials

Aashique A. Rezwani^{a,*}, Andrea M. Jokisaari^b, Michael R. Tonks^c

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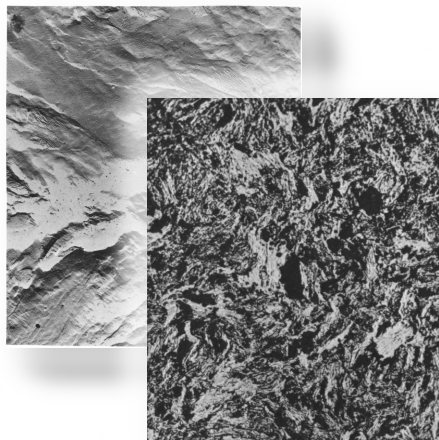
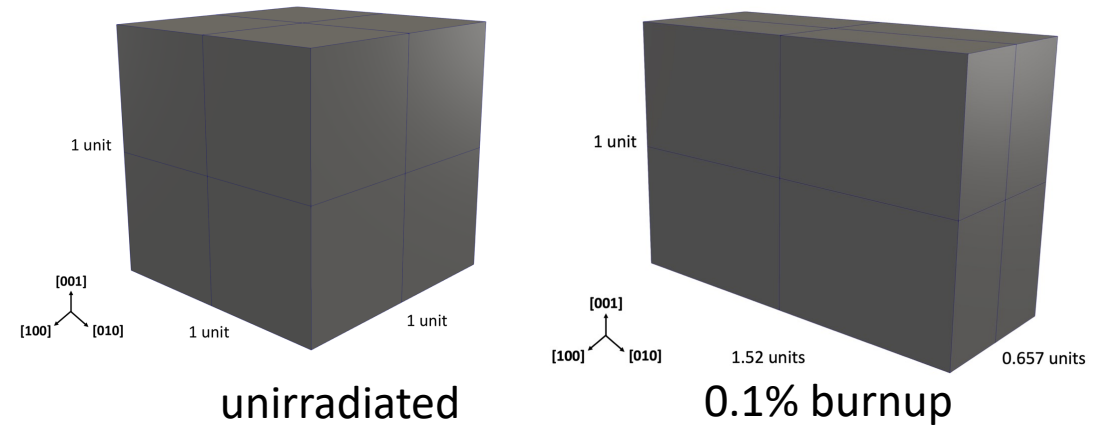
Andrea M. Jokisaari^{a,*}, Khadija Mahbuba^b, Yuhao Wang^c, Benjamin Beeler^b

^a Computational Mechanics and Materials Department, Idaho National Laboratory, Idaho Falls, ID 83415, United States of America
^b Nuclear Engineering Department, North Carolina State University, Raleigh, NC 27695, United States of America
^c Nuclear Engineering and Radiological Sciences Department, University of Michigan, Ann Arbor, MI 48109, United States of America

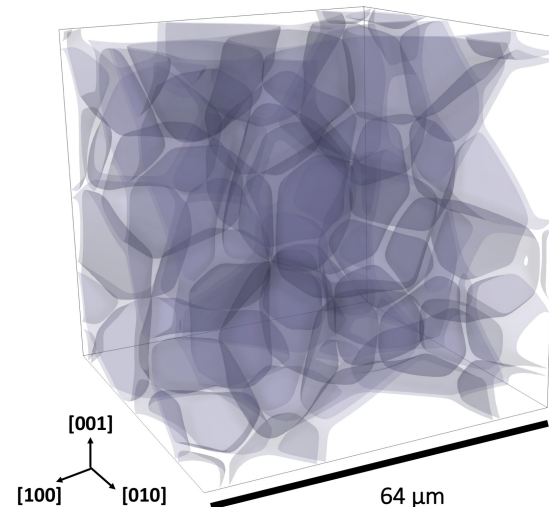
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Mesoscale study shows irradiation growth can cause large microstresses


- Irradiation growth in single-crystal α -U causes major dimensional distortions with small amounts of burnup
- Hypothesized that irradiation growth in polycrystalline material causes observed “marble cake” structure




Acta Metall. 23 (1975) 101



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
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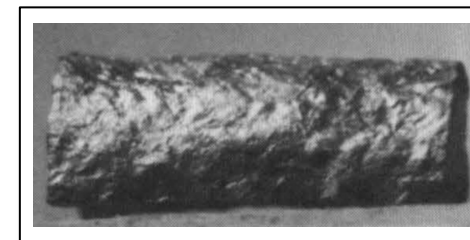
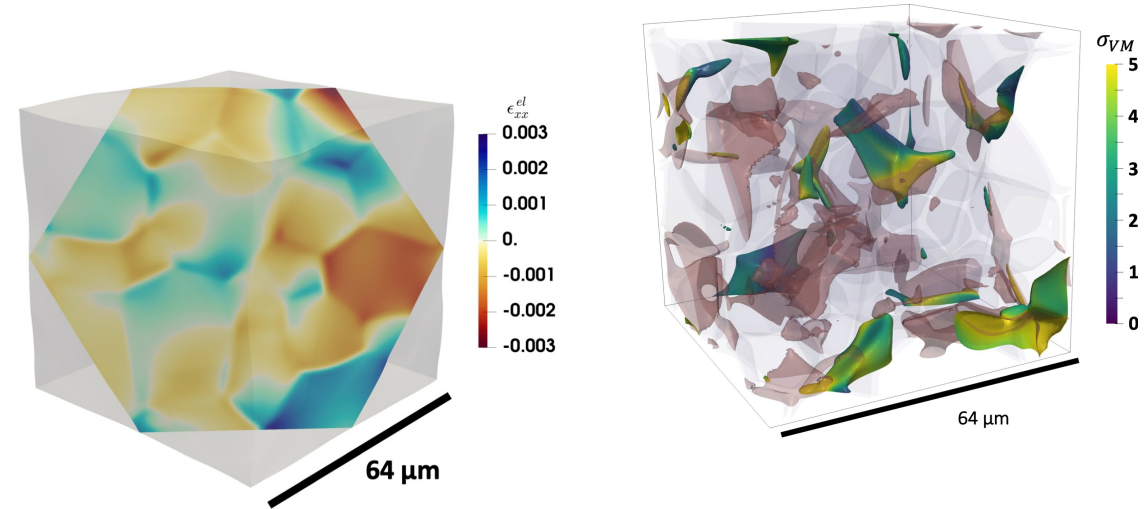
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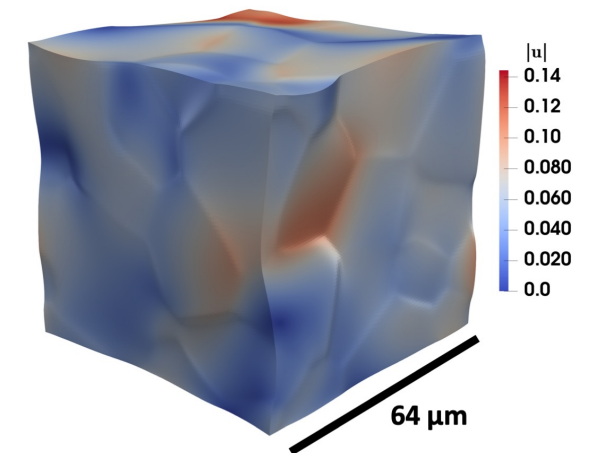


Mesoscale study shows irradiation growth can cause large microstresses

- Showed that large von Mises and hydrostatic stresses can form along grain boundaries and triple points, leading to either plastic strain or cavitation
- Surface roughening also results for polycrystalline specimens
- Applied tensile or compressive stresses up to 100 MPa do not appreciably counteract growth stresses



Leggett, HW-79559, 1963



Computational Materials Science 176 (2020) 109545

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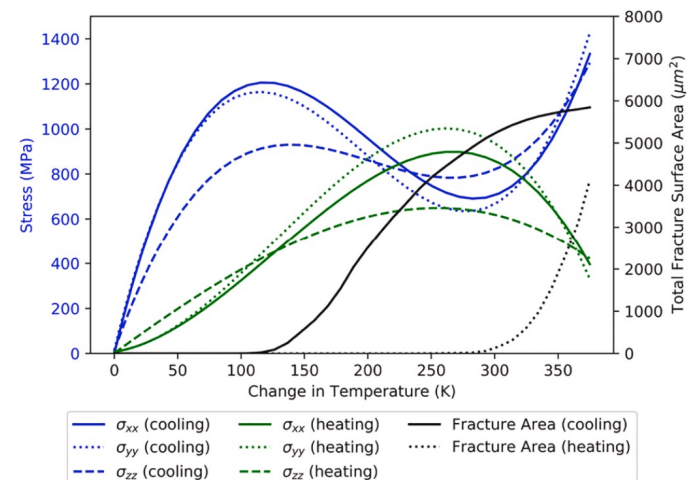
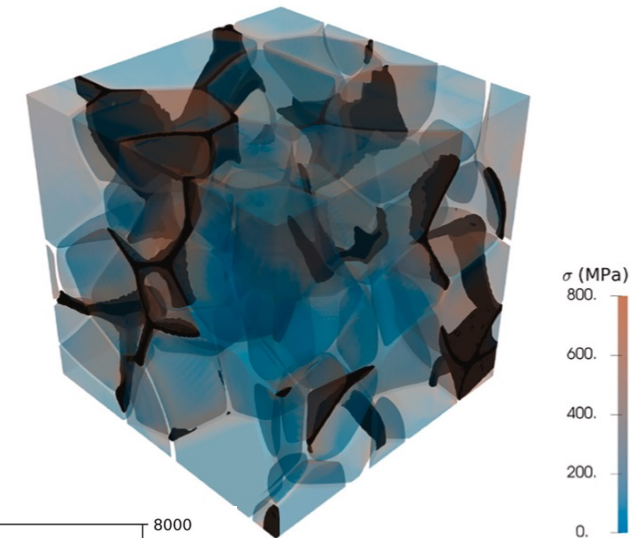
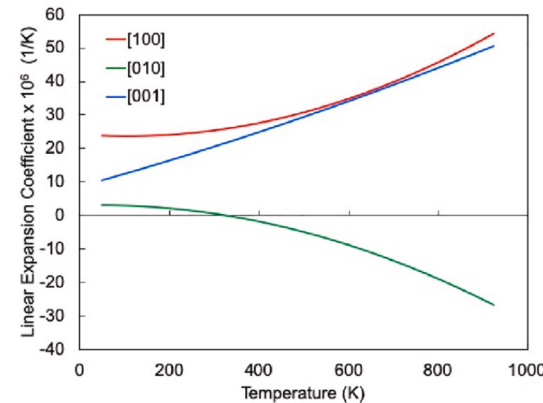
Irradiation-induced internal stresses in polycrystalline α -uranium: a mesoscale mechanical approach

A.M. Jokisaari

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Mesoscale study shows thermal expansion can cause large microstresses & fracture

- Thermal cycling of polycrystalline α -U has been shown to induce shape changes and porosity development
- Hypothesized that anisotropic thermal expansion in polycrystalline material is the cause
- Showed that [010] negative coefficient of thermal expansion can cause fracture with 100 K temperature change



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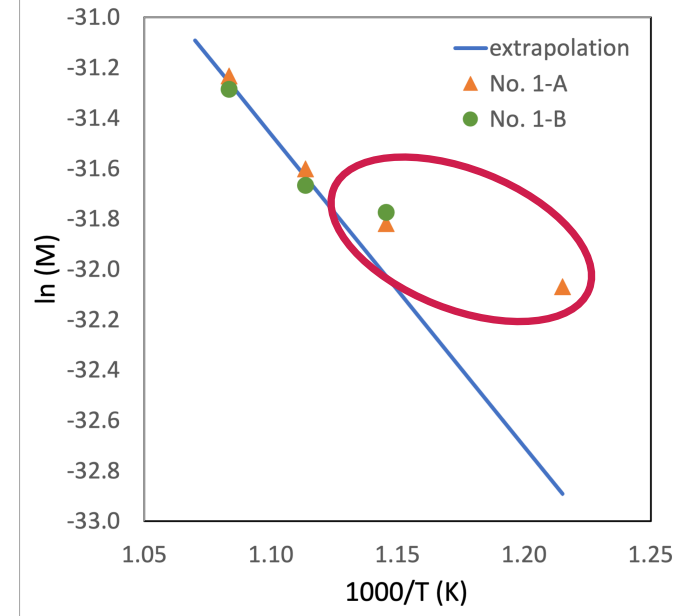
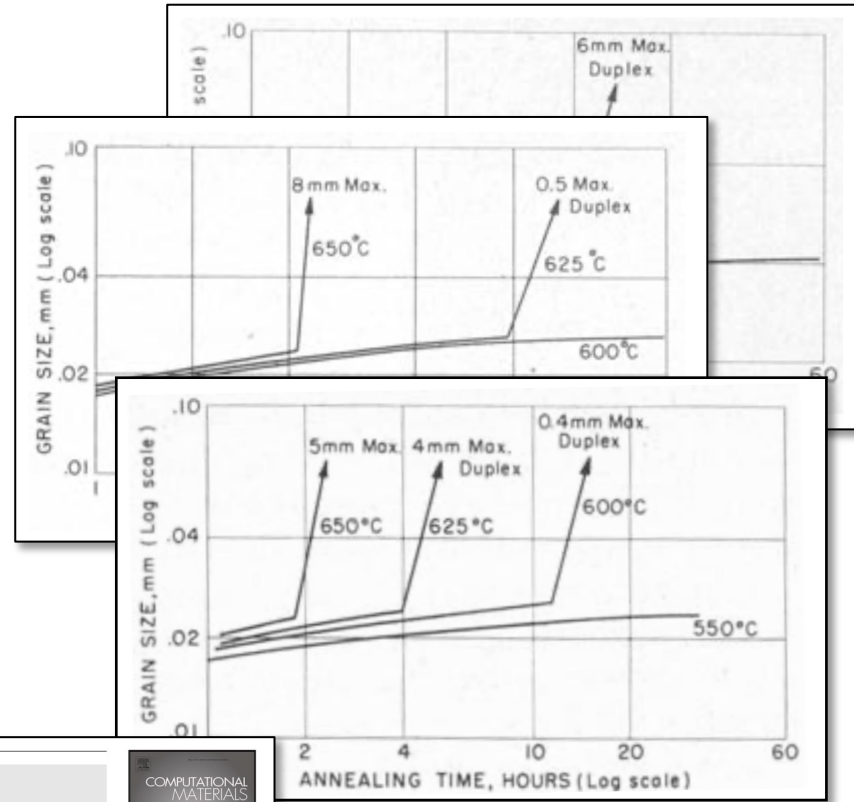
Modeling brittle fracture due to anisotropic thermal expansion in polycrystalline materials

Aashique A. Rezwan^{a,*}, Andrea M. Jokisaari^b, Michael R. Tonks^c

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Analysis of historical grain growth data brings a mystery

- Motivation: determine grain boundary mobility for α -U
- Found experimental grain growth data, fitted it
- Anomalously large mobility at low temperatures
- Hypothesized strain from thermal expansion could cause the observed behavior




$$M_0 = 1.85 \times 10^{-8} \text{ m}^4/\text{J s}$$

$$Q = 1.07 \text{ eV}$$

J. Metals (1957) 882-888

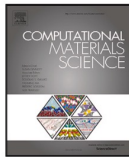
$$\bar{D}^2(t) = \bar{D}_0^2 + \frac{1}{2} \alpha \gamma M t$$




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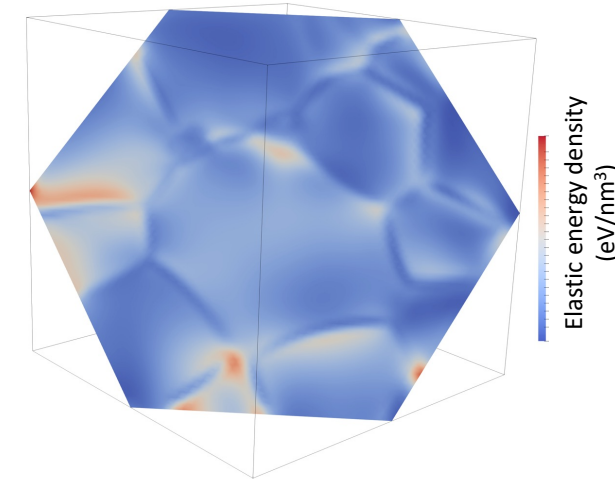
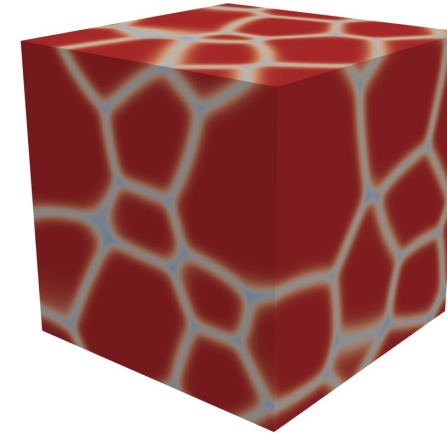
The impact of anisotropic thermal expansion on the isothermal annealing of polycrystalline α -uranium



Andrea M. Jokisaari ^{a,*}, Khadija Mahbuba ^b, Yuhao Wang ^c, Benjamin Beeler ^b

Microstructure evolution simulations are shedding light on thermomechanical behaviors

- Grain boundary mobility α -U has not been experimentally measured
- Phase field model includes strain energy from thermal expansion, where $T_0 = 933$ K
- Found that including thermal expansion in grain growth simulations resulted in qualitatively similar deviations from Arrhenius behavior



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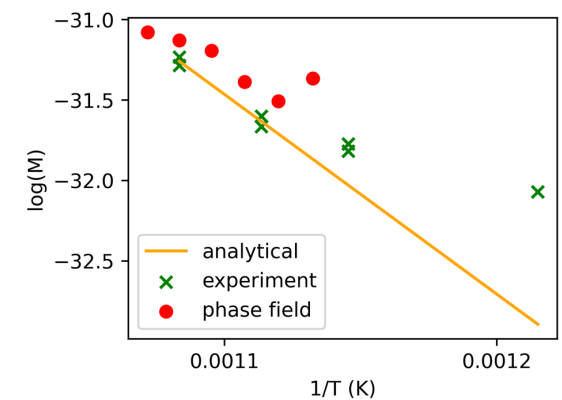
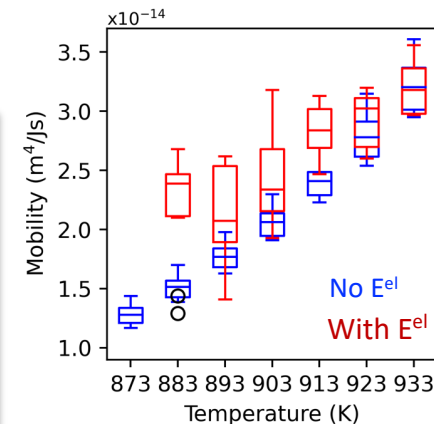
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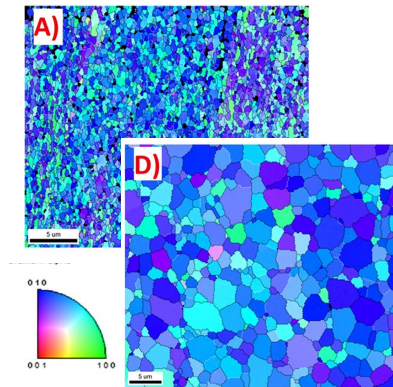
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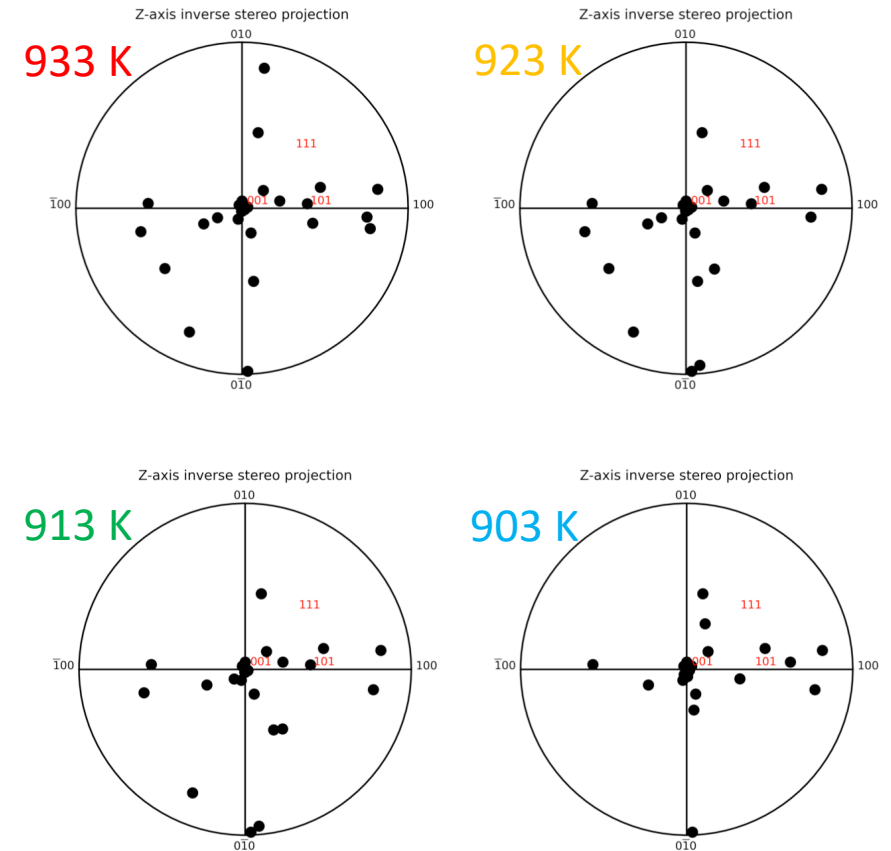


Microstructure evolution simulations are shedding light on thermomechanical behaviors

- Texture development is observed for polycrystalline α -U and remains with annealing
- Hypothesized that thermal expansion stresses can drive texture evolution
- Strain-free temperature at 933 K, thermal eigenstrain increases with decreasing temperature
- Observed decrease in randomness of grain orientation during grain growth with increasing ΔT



JNM 542 (2020) 152467



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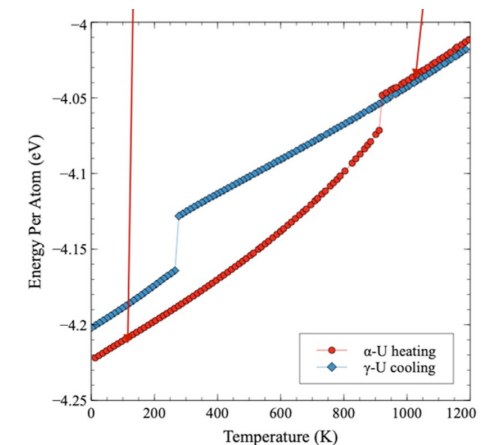
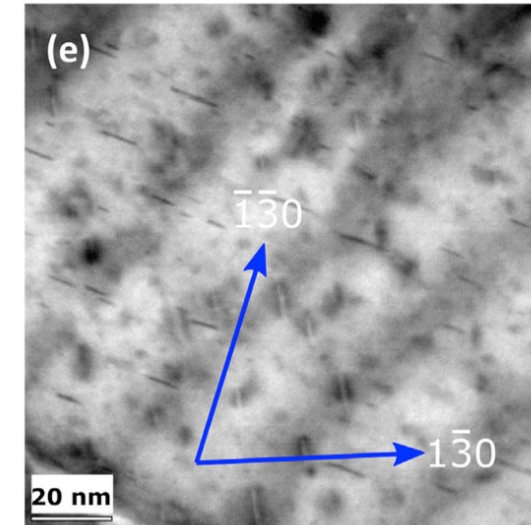
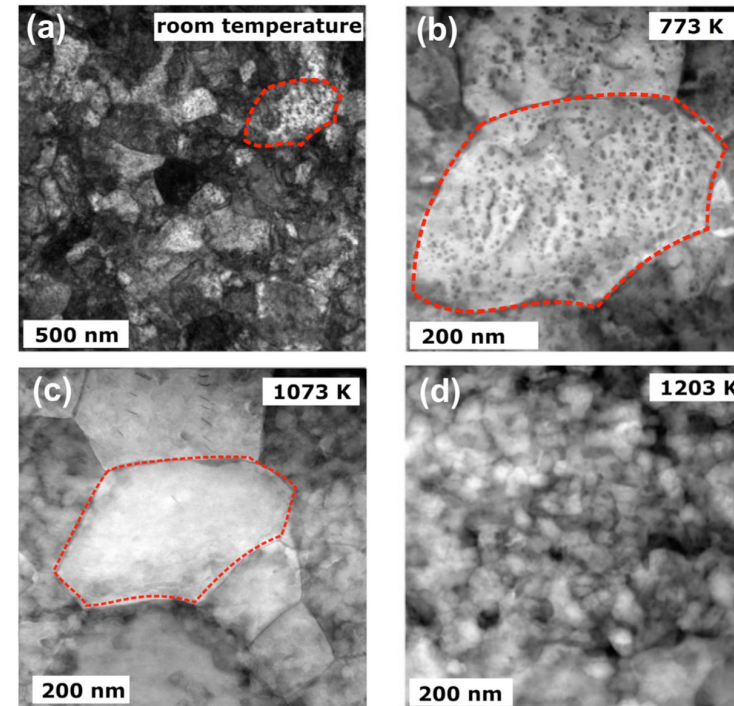
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Andrea M. Jokisaari ^{a,*}, Khadija Mahbuba ^b, Yuhao Wang ^c, Benjamin Beeler ^b

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Novel *in-situ* studies reveal defect organization and phase change behavior

- In collaboration with Fidelma Di Lemma (PI)
- In-situ TEM observation of heating and cooling of TREAT-irradiated U
- Molecular dynamics simulations help interpret experimental results



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Microstructural and phase changes in alpha uranium investigated via *in-situ* studies and molecular dynamics



Fidelma G. Di Lemma^{a,*}, Tiankai Yao^a, Daniele Salvato^a, Luca Capriotti^a, Fei Teng^a,
Andrea M. Jokisaari^a, Benjamin W. Beeler^{a,b}, Yuhao Wang^c, Colby J. Jensen^a

Atomistic studies quantify anisotropic energetic and kinetic properties

- Molecular dynamics simulations (*ab initio* and classical) to investigate a range of fundamental materials properties of α -U
 - Grain boundary and surface energies
 - Work of adhesion
 - Directional diffusion coefficients
 - Defect formation and binding energies
 - Defect-induced lattice distortions
- Ab initio studies provide a basis of comparison to results from classical MD potential



An atomistic study of fundamental bulk and defect properties in α -uranium

Yuhao Wang^a, Benjamin Beeler^b, Andrea Jokisaari^{c,*}



Evaluation of the anisotropic grain boundaries and surfaces of α -U via molecular dynamics

Khadija Mahbuba^{a,*}, Benjamin Beeler^{a,b}, Andrea Jokisaari^b

^aNorth Carolina State University, Raleigh, NC 27607, United States
^bIdaho National Laboratory, Idaho Falls, ID 83415, United States



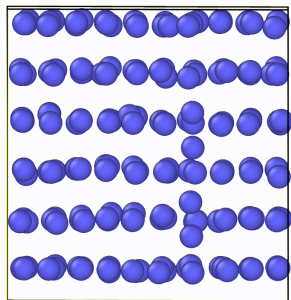
Determination of Thermal Expansion, Defect Formation Energy, and Defect-Induced Strain of α -U Via *ab Initio* Molecular Dynamics

Benjamin Beeler^{1,2*}, Khadija Mahbuba¹, Yuhao Wang³ and Andrea Jokisaari²

¹Department of Nuclear Engineering, North Carolina State University, Raleigh, NC, United States, ²Idaho National Laboratory, Idaho Falls, ID, United States, ³Department of Nuclear Engineering and Radiological Sciences, University of Michigan, Ann Arbor, MI, United States

Ab-initio studies show individual point defect behaviors are not responsible for α -U irradiation growth

- Hypothesized that irradiation growth is a result of single defect behaviors (rather than multi-defect organization)
- AIMD studies α -U from 100 – 800 K
- Fundamental properties match well with experimental data
- Lattice strains of the individual defects do not qualitatively correspond to the irradiation growth strains \rightarrow due to extended defects?



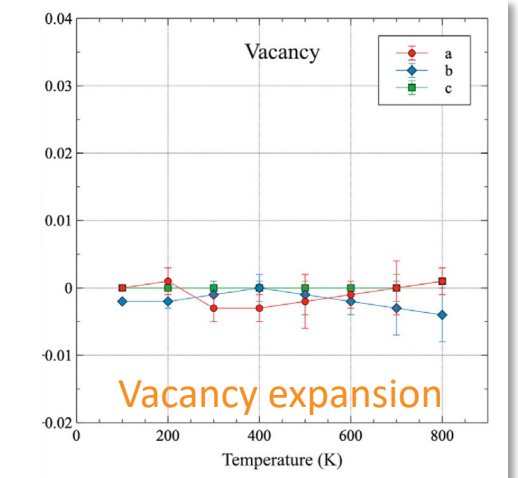
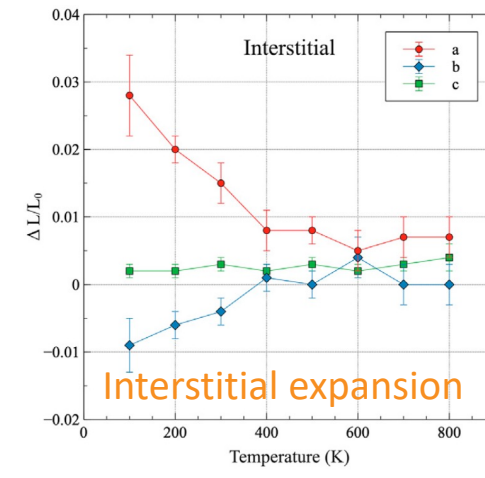
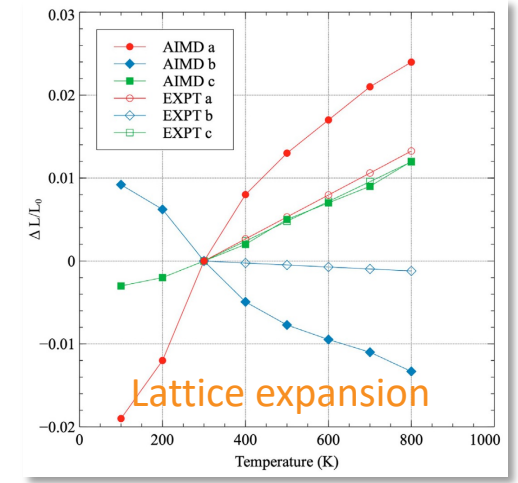
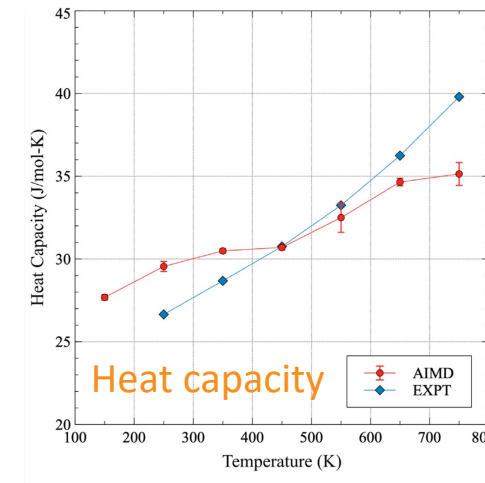
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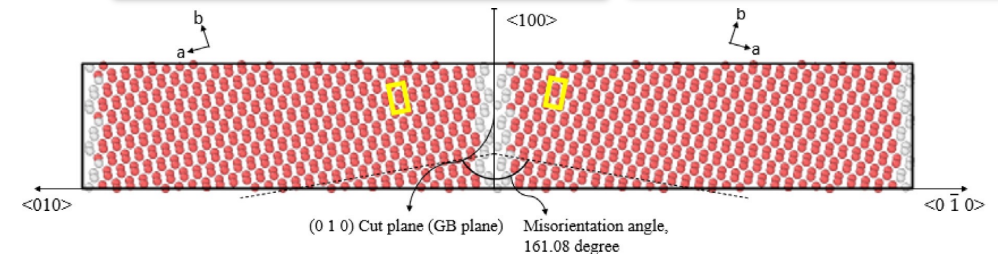
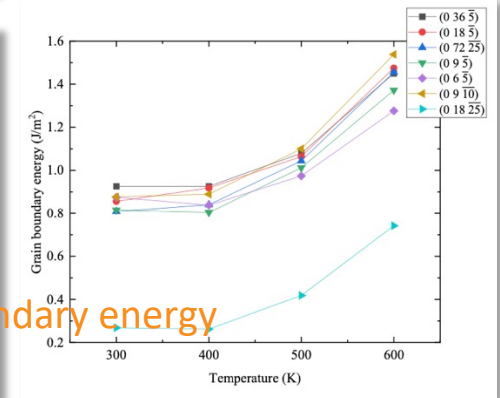
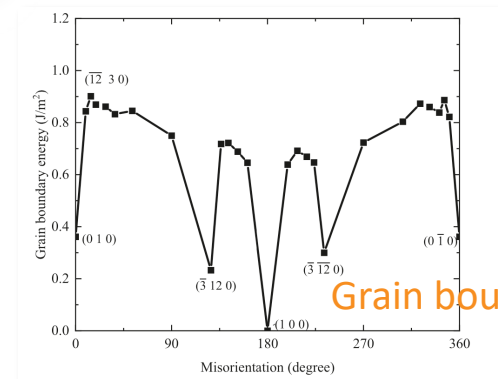
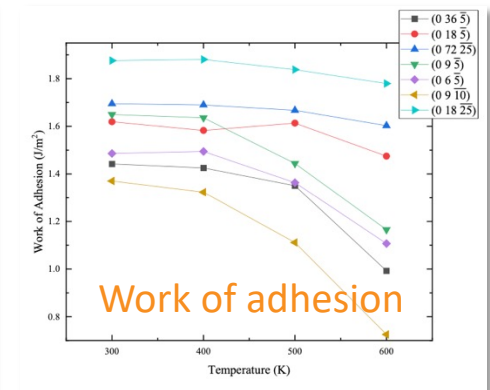
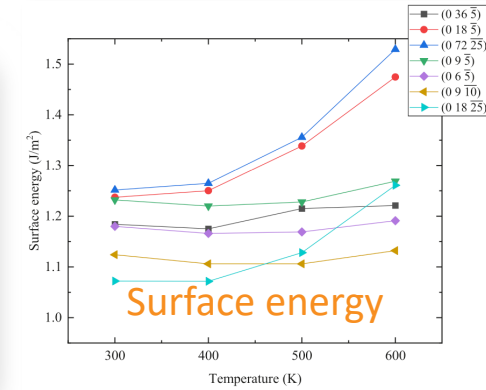
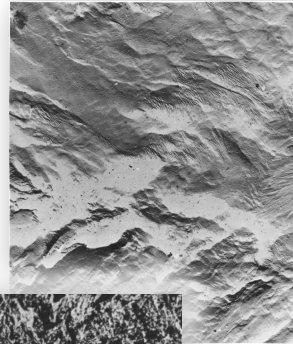
Determination of Thermal Expansion, Defect Formation Energy, and Defect-Induced Strain of α -U Via *ab Initio* Molecular Dynamics


Benjamin Beeler^{1,2*}, Khadija Mahbuba¹, Yuhao Wang³ and Andrea Jokisaari²



Grain boundary and surface characterization shed light temperature-dependent behavior

- Grain boundary tearing is responsible for ~50% α -U swelling
- Hypothesize that behavior variation with temperature is result of changing grain boundary characteristics
- Molecular dynamics studies of STGBs from 300 -- 600 K
- The work of adhesion decreases with increasing temperature, suggesting reason for grain boundary tearing

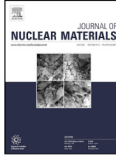




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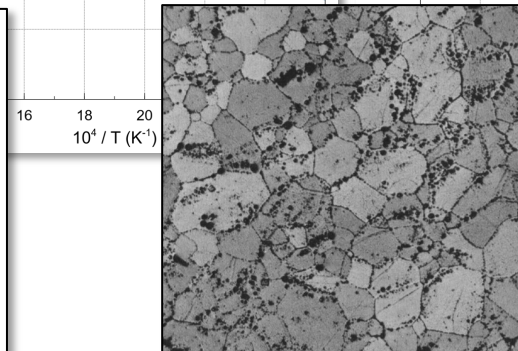
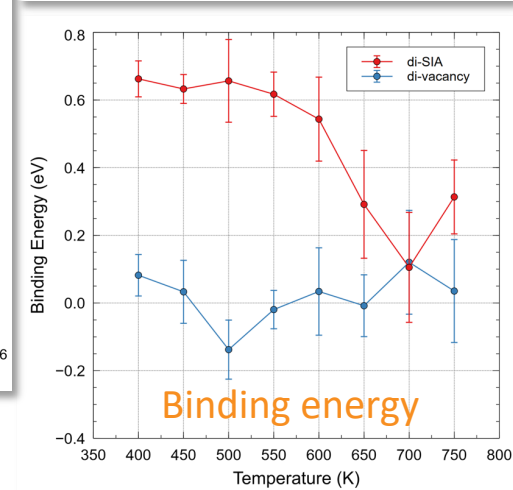
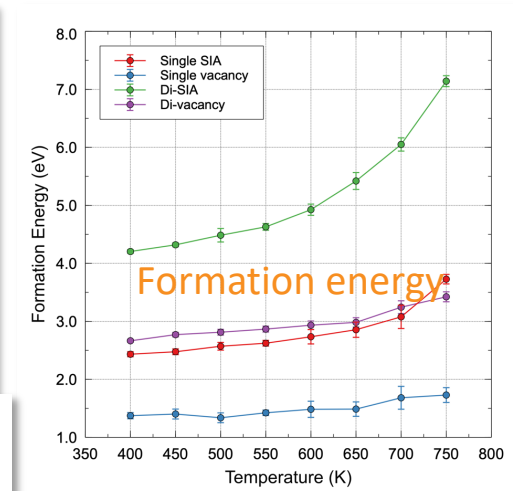
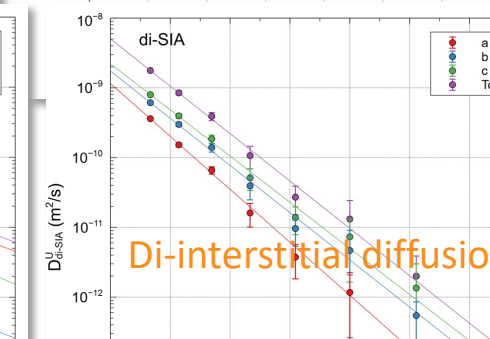
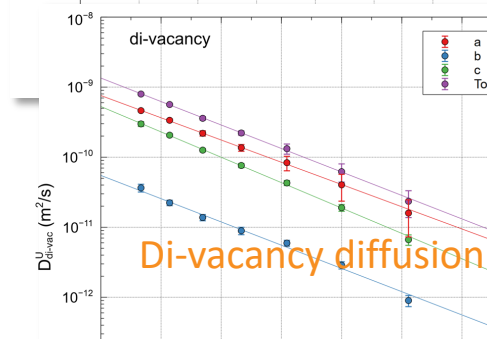
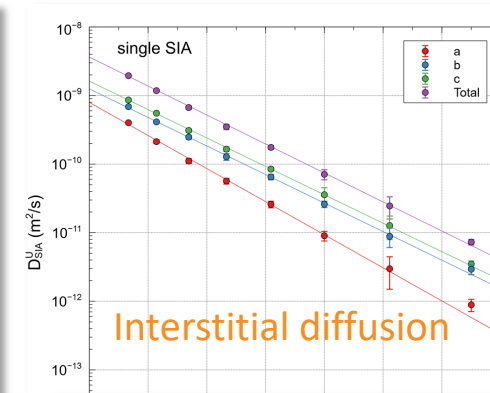
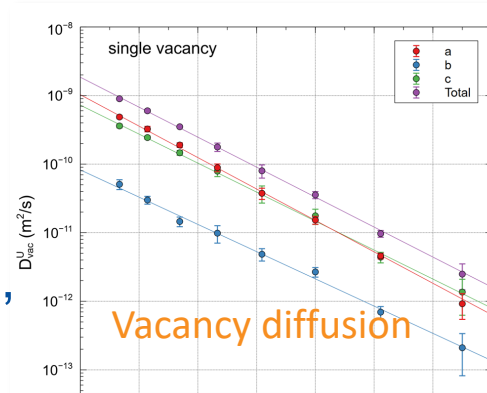
Evaluation of the anisotropic grain boundaries and surfaces of α -U via molecular dynamics

Khadija Mahbuba^{a,*}, Benjamin Beeler^{a,b}, Andrea Jokisaari^b



α -U defect energetics and transport may explain the varied irradiation behavior with temperature

- Molecular dynamics studies from 400 – 750 K
- Characterized anisotropic diffusivities of vacancies, interstitials, and di-defects, formation and binding energies
- Increasing defect formation energy with temperature may explain reduced microstructure damage



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An atomistic study of fundamental bulk and defect properties in α -uranium

Yuhao Wang^a, Benjamin Beeler^b, Andrea Jokisaari^{c,*}

Conclusions and future work

- Multiscale modeling is providing new insights into the behavior of α -uranium
 - Thermal expansion on microstructure
 - Irradiation growth stresses
 - Bulk properties
 - Defect energetics, kinetics, and elastic characteristics
- Thermal expansion strains can significantly drive microstructure evolution during, e.g., thermomechanical processing
- Irradiation and thermal expansion are both important factors controlling irradiation behavior
- Decreasing work of adhesion with increasing temperature could explain cavitation
- Increasing defect formation energy and lower binding energies at higher temperatures could explain the reduced irradiation swelling at high temperature
- Individual defects strain the lattice significantly, but does not qualitatively compare to irradiation growth – likely due to organization of extended defects into loops