# LAMMPS Simulation in the Investigation of Dimensional Changes in Zirconium in the Presence of Alloying Elements and Hydrogen*

## LAMMPS User Group Meeting 2013

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*Work performed in collaboration with EPRI

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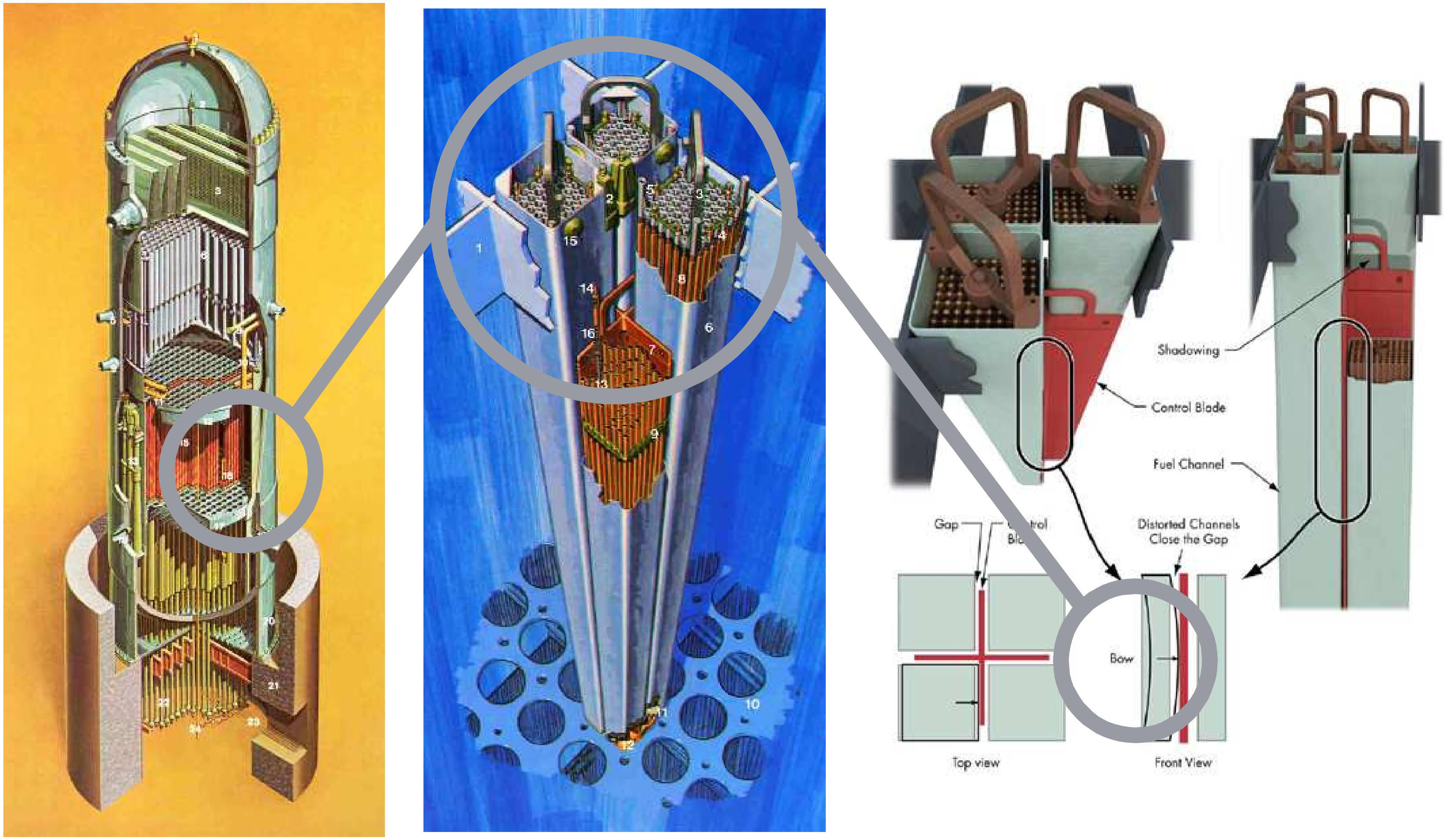
# Hydrogen in Metals

- Interaction of hydrogen with metals is important in
  - Embrittlement
  - Dimensional changes, irradiation growth, etc.
  - Batteries/hydrogen storage
- Understanding of such phenomena
  - First-principles (VASP)
  - Forcefields (LAMMPS)
  - Linkage between methods (MedeA®)
- For example: fuel channel bowing
  - Hydrogen content affects irradiation growth in Zr alloys
    - What is the role of hydrogen in modifying bowing?
    - What is the role of alloying elements, e.g. Nb?
    - What responses are possible?

# Background: Fuel Channel Bowing




- Fuel channel distortion or bowing caused by irradiation growth impacts reactor operation

# Computational Strategy

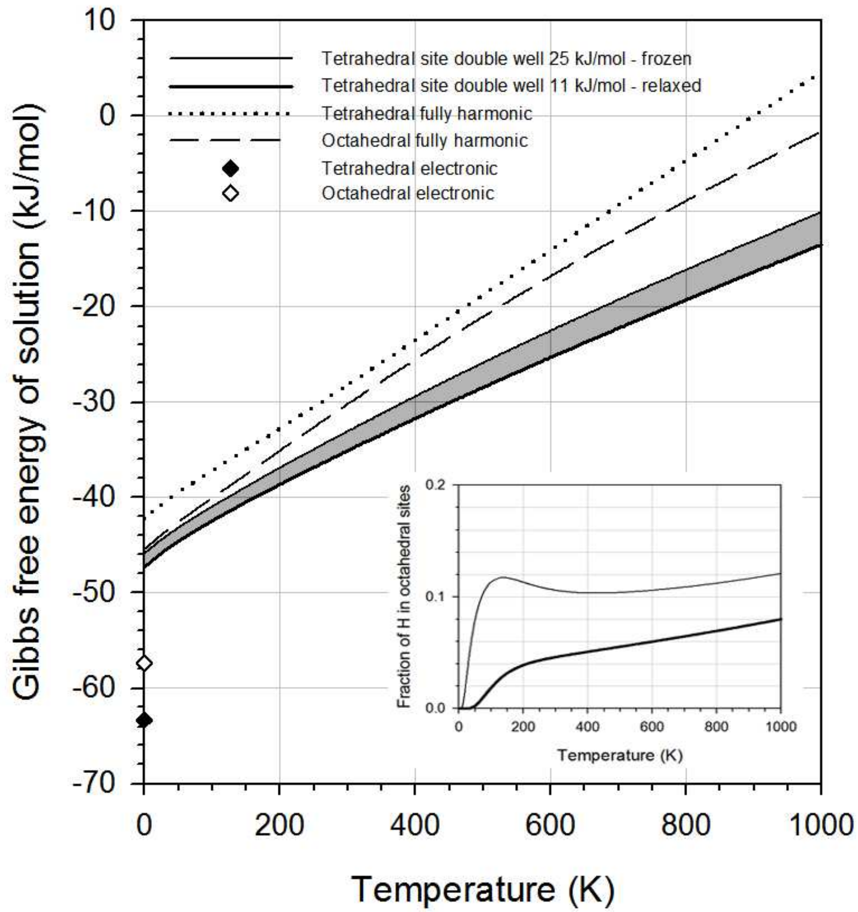
- VASP
  - Hydride and Zr structures and energies
  - Mechanical properties & Phonons
  - Forcefield parameters

MedeA® computational environment

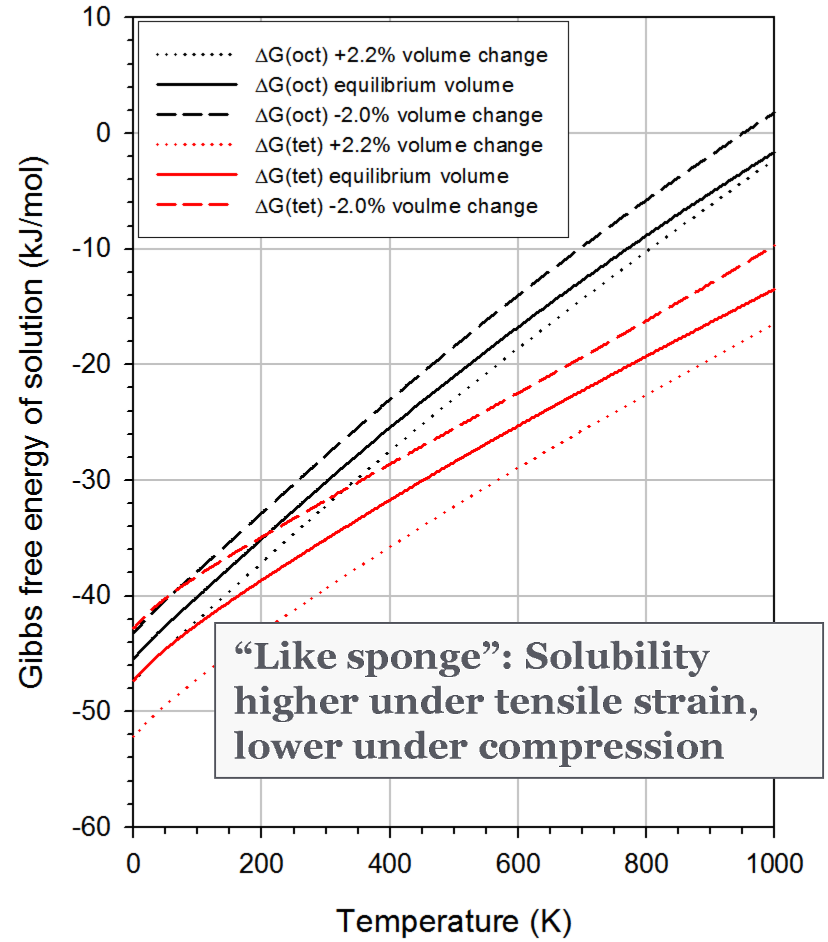
- LAMMPS
  - Structures and energies
  - Sampling
  - Diffusive properties

# Gibbs Free Energy of H Solution in Zr

H Solubility




Effect of Strain

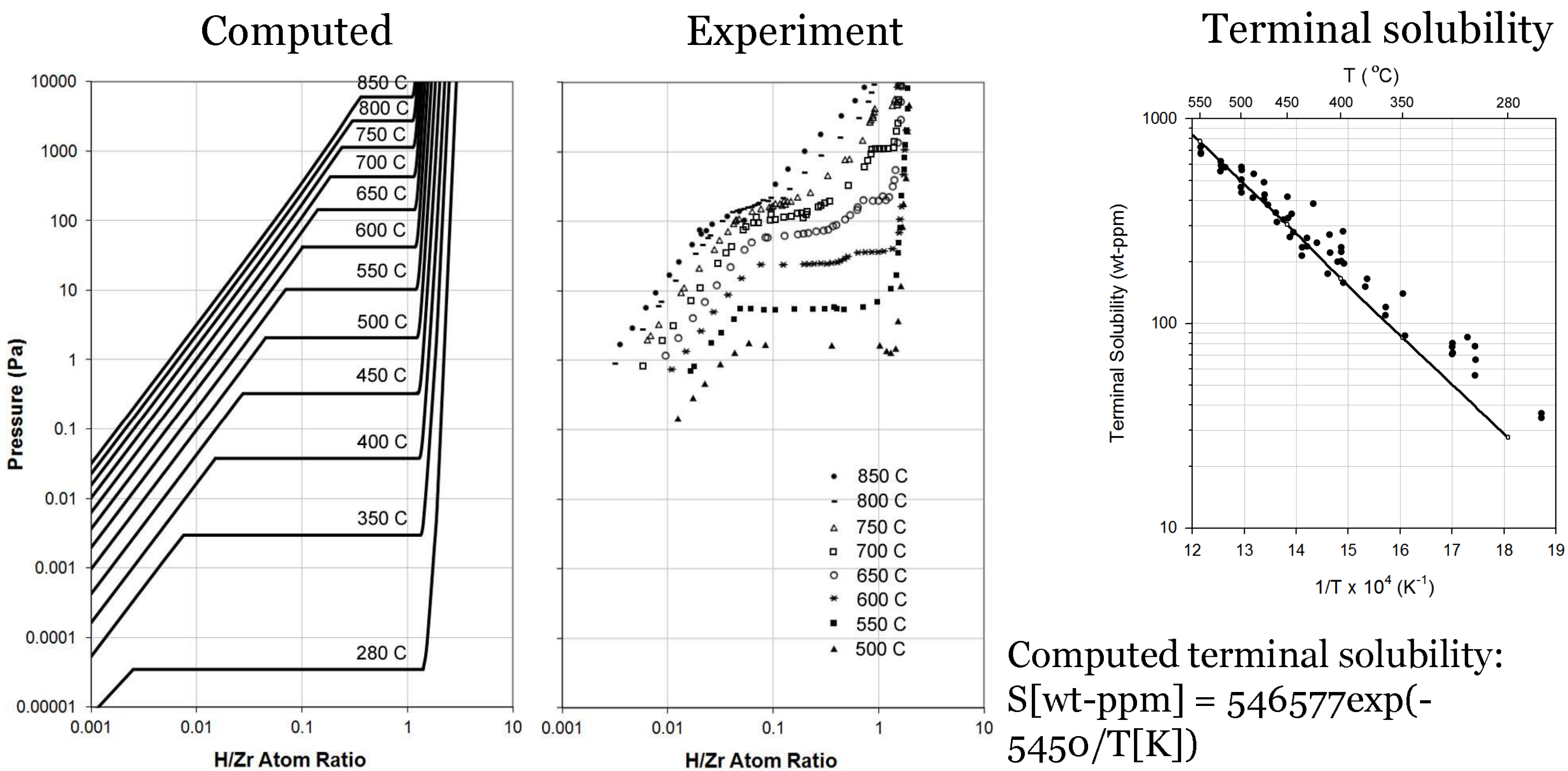



Computed $\Delta H^o_{298}$ = -41.3 kJ/mol
Experimental: -32.5 to -64 kJ/mol

From: 'H in alpha-Zr and in zirconium hydrides: solubility, effect on dimensional changes and the role of defects', M. Christensen, W. Wolf, C. M. Freeman, E. Wimmer, R. B. Adamson, L. Hallstadius, P. E. Cantonwine, E. V. Mader, In preparation

# H Solubility in α-Zr

## Solubility of H increases with temperature




Computed terminal solubility:
$S[\text{wt-ppm}] = 546577\exp(-5450/T[\text{K}])$

at 300 °C: S = 41 wt-ppm
at 25 °C: S = 0.006 wt-ppm

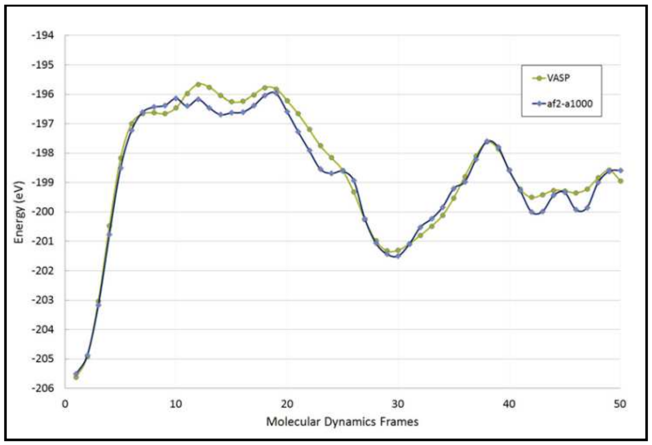
From: 'H in alpha-Zr and in zirconium hydrides: solubility, effect on dimensional changes and the role of defects', M. Christensen, W. Wolf, C. M. Freeman, E. Wimmer, R. B. Adamson, L. Hallstadius, P. E. Cantonwine, E. V. Mader, In preparation

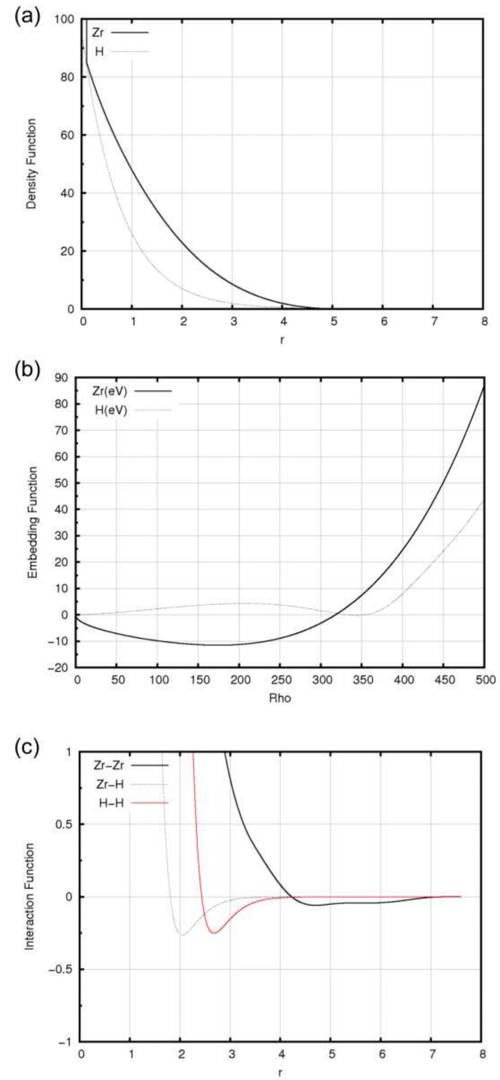
# Dimensional Changes

- First-principles (VASP) calculations provide an accurate description of thermodynamic properties
- But, channel bowing is a dynamic process
  - Each Zr atom in a reactor steel is displaced from its lattice site multiple times during the lifetime of the component
  - Irradiation growth and bowing occurs over component lifetime
- Questions
  - How do Zr atoms migrate through the hcp lattice?
  - How do alloying elements interact with Zr diffusion?
  - How does hydrogen affect Zr diffusion?
- Such questions require molecular dynamics (LAMMPS) calculations

# Forcefields for Zr, H, and Nb

- EAM forcefields
- Two forms developed
  - One based on Mendelev & Ackland*
  - One based on a fit to VASP MD results

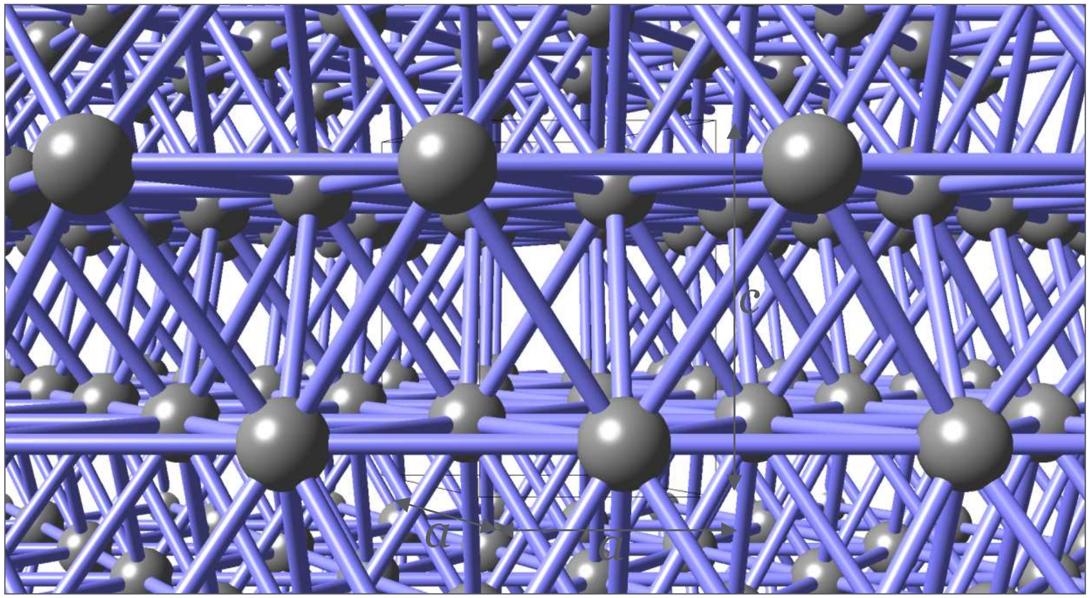


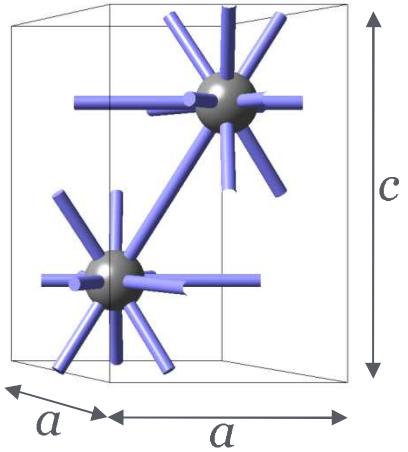




- Agreement between first-principles and forcefields (shown above for an example inorganic system) satisfactory
- Forcefields permit large system simulation (LAMMPS)

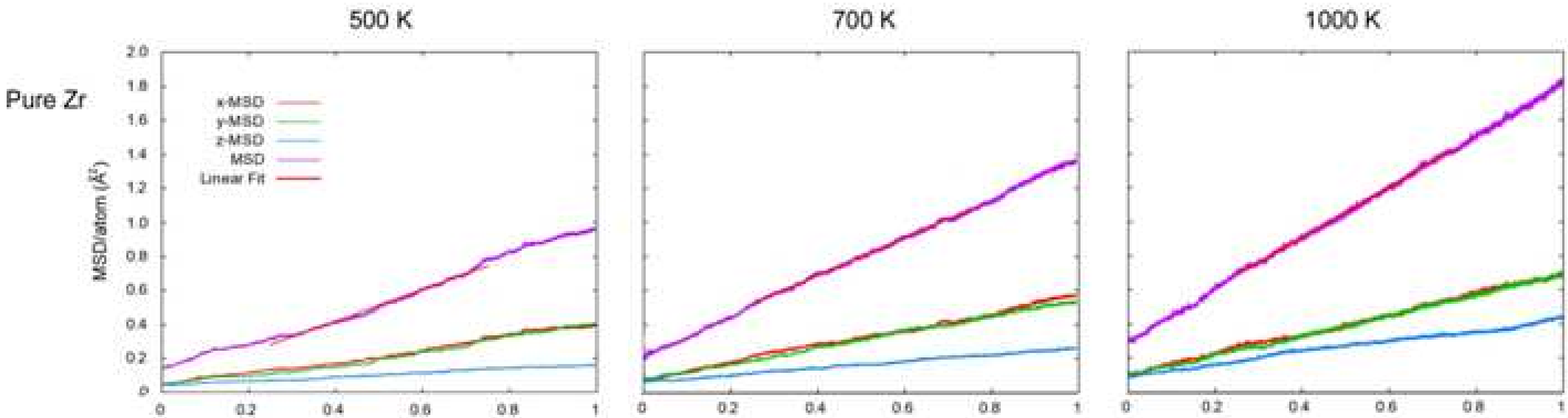
*M.I. Mendelev and G.J. Ackland, Phil. Mag. Lett., 87, 349-359 (2007)

# Anisotropy of Zr Diffusion



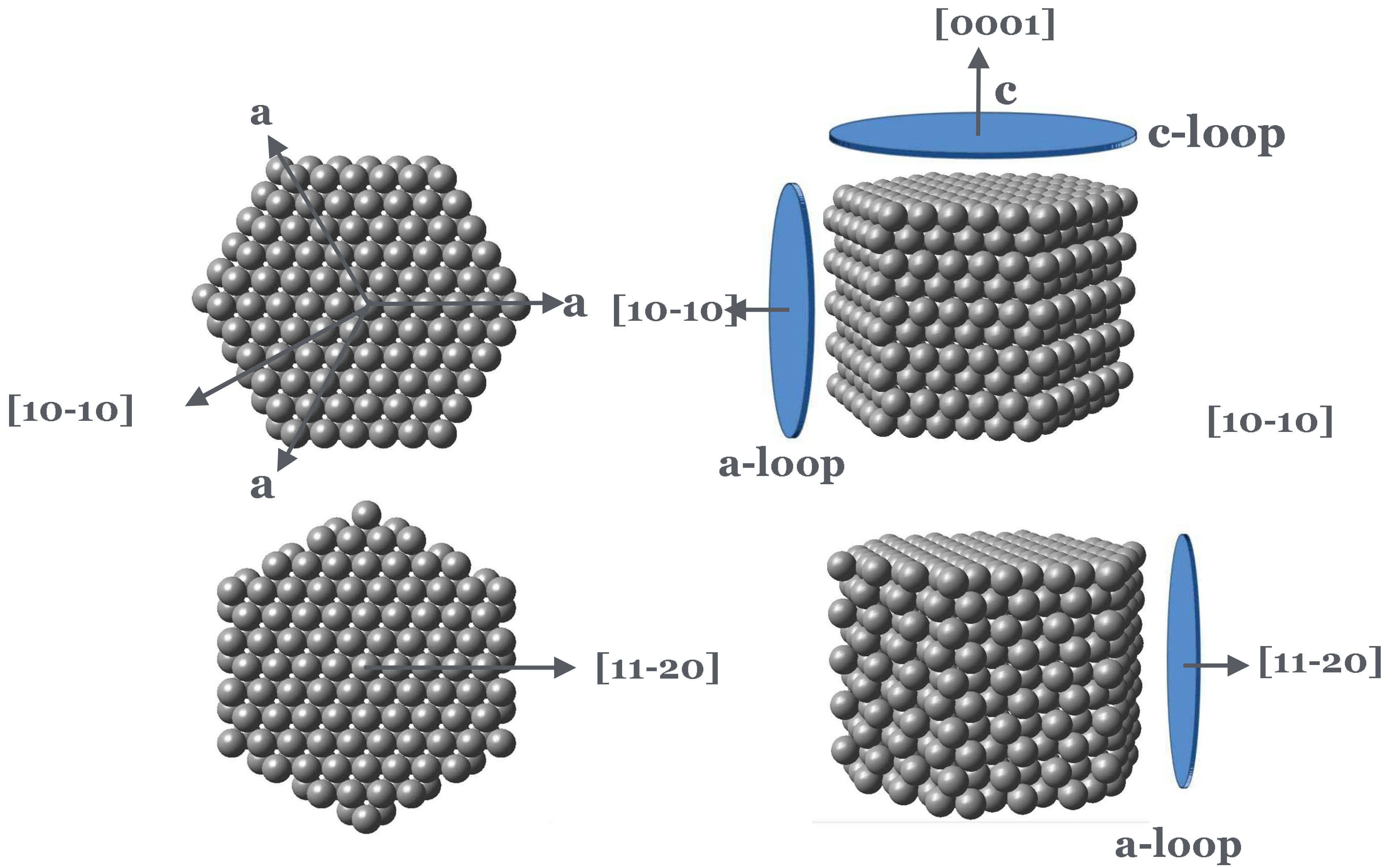



# Self Interstitial Diffusion MSD Plots

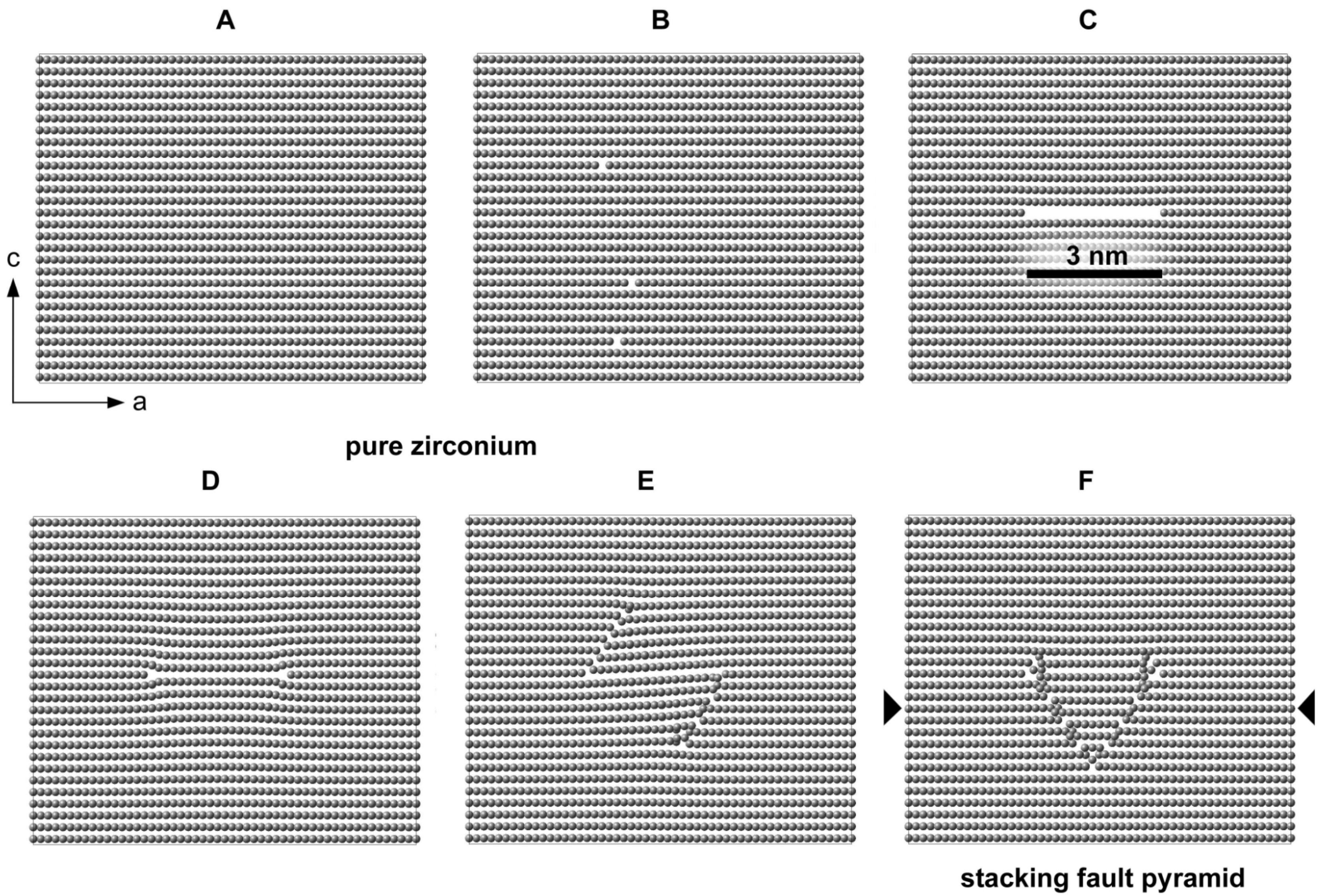



- Interstitial Zr diffusion is anisotropic
- Forcefield calculations also provide information on H diffusion

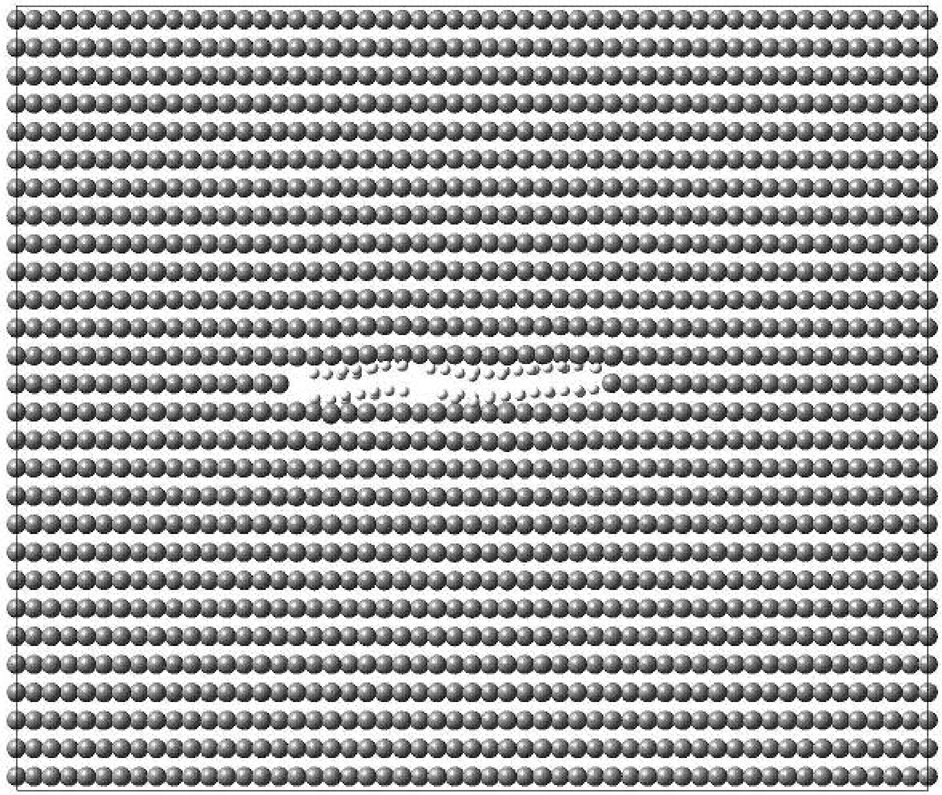
# Formation of Loops in hcp α-Zr



# Vacancy c-Loops



# Vacancy c-Loop with Hydrogen

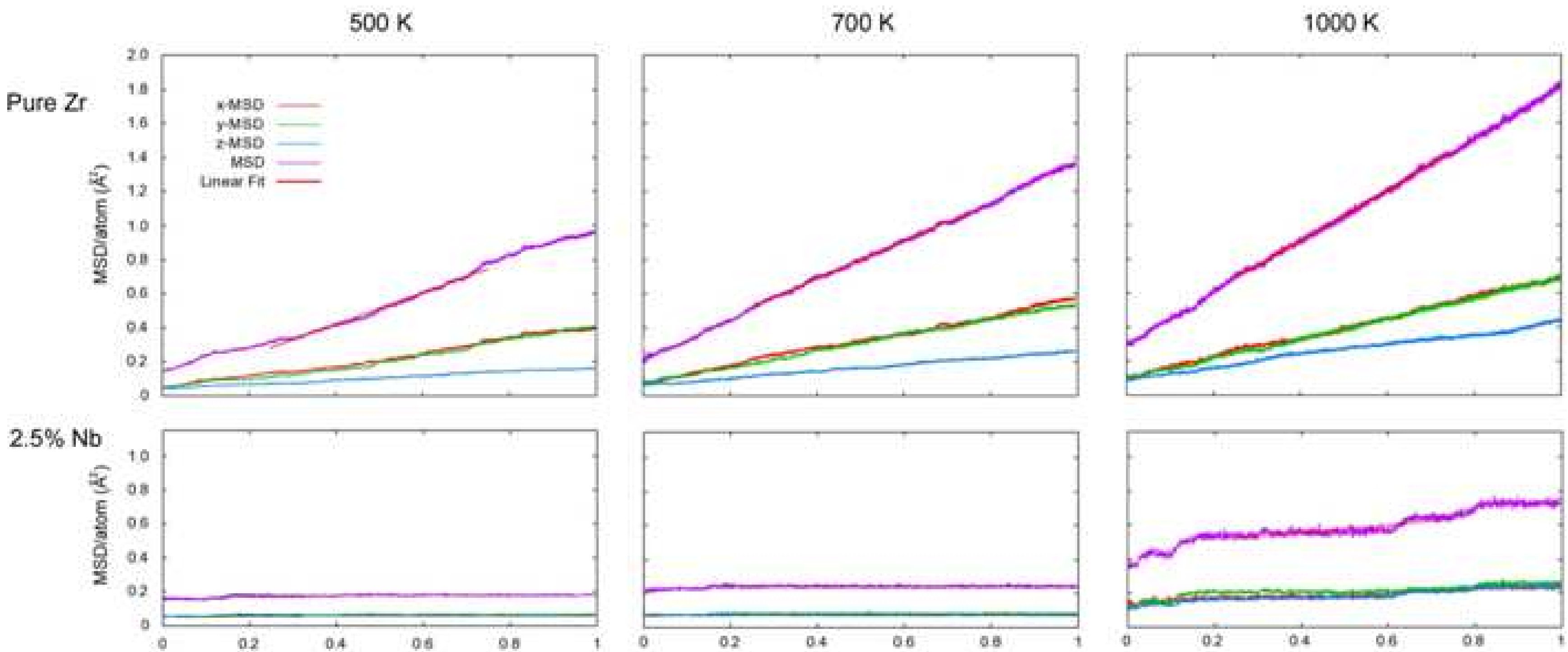


Zr with H

α-Zr supercell with 20384 lattice sites
with 85 vacancies

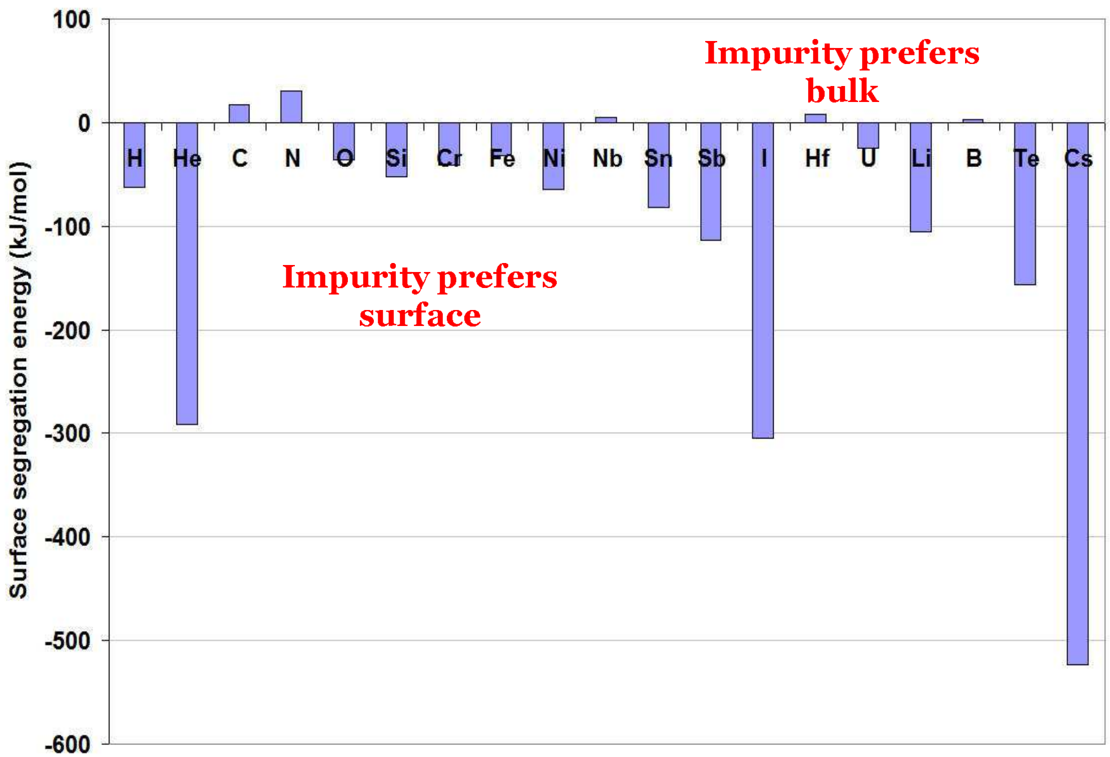
- Hydrogen diffuses rapidly & stabilizes vacancy structures

# Self Interstitial Diffusion in Presence of Nb from Molecular Dynamics




- The diffusion of self interstitial Zr is impeded by Nb

# Surface Segregation of Alloying Elements




Sn and Ni have significant thermodynamic driving force to segregate to surface

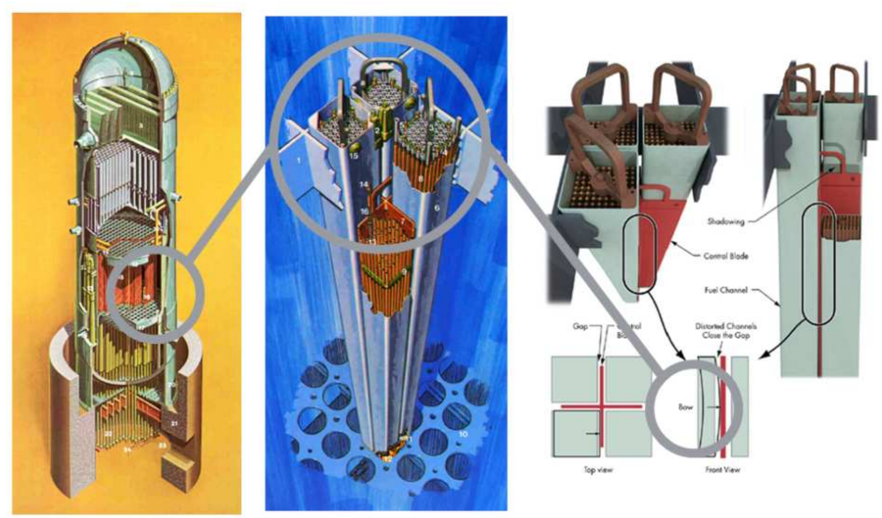
Fe and Cr have smaller driving force

Nb prefers bulk

M. Christensen, T. M. Angeliu, J. D. Ballard, J. Vollmer, R. Najafabadi, and E. Wimmer, J. Nucl. Mat. **404**, 121 (2010)

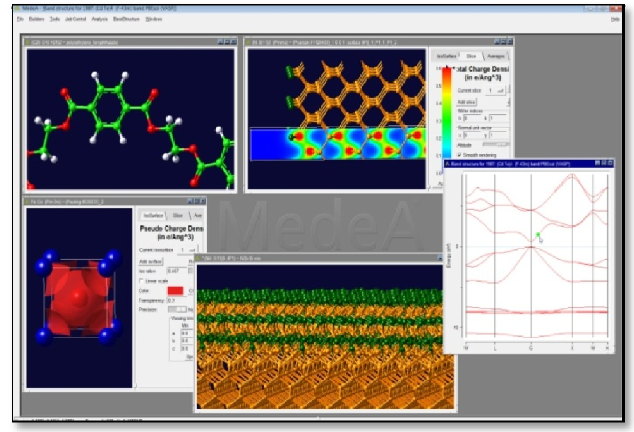
# Summary of Key Findings

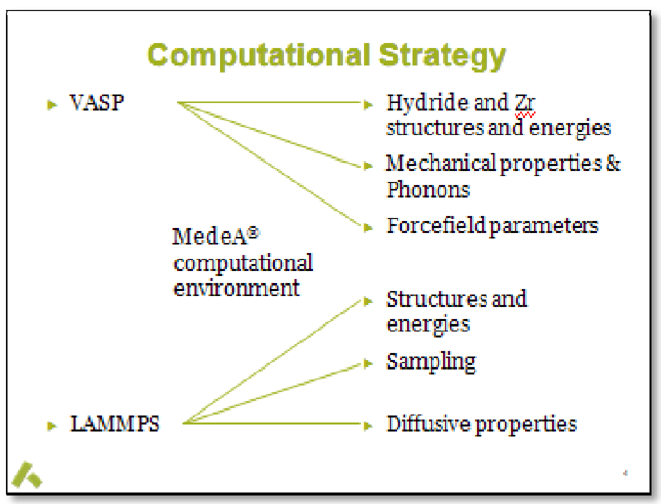
- Zr lattice
  - Vacancy and interstitial loop behavior depends on orientation
  - Rapid and anisotropic Zr interstitial diffusion
  - Loops restructure to reduce anisotropy
- Hydrogen
  - Increases Zr interstitial concentration by reducing recombination of interstitials and vacancies
  - Inhibits restructuring increasing anisotropic affect of c-loop formation
- Niobium
  - Resides in bulk
  - Traps interstitial Zr
- Understanding
  - Alloying elements
  - Operational measurement



# Basis of Calculations: LAMMPS

- Combination of LAMMPS and VASP linked through forcefields, and high-performance and high-throughput computing in the MedeA® environment





# Acknowledgements


- Work carried out with EPRI (Electric Power Research Institute) and its member organizations

- References:

  - ‘Effect of hydrogen on dimensional changes of zirconium and the influence of alloying elements: first-principles and classical simulations of point defects, dislocation loops, and hydrides’, M. Christensen, W. Wolf, C. M. Freeman, E. Wimmer, R. B. Adamson, L. Hallstadius, P. E. Cantonwine, E. V. Mader, ASTM conference proceedings, ASTM Conference, Feb 6 2013, Hyderabad, India
  - ‘H in alpha-Zr and in zirconium hydrides: solubility, effect on dimensional changes and the role of defects’, M. Christensen, W. Wolf, C. M. Freeman, E. Wimmer, R. B. Adamson, L. Hallstadius, P. E. Cantonwine, E. V. Mader, In preparation
  - ‘Diffusion of H and Zr in alpha-Zr and the formation of dislocation loops’, M. Christensen, W. Wolf, C. M. Freeman, E. Wimmer, R. B. Adamson, L. Hallstadius, P. E. Cantonwine, E. V. Mader, Jounal of Nuclear Materials, Submitted, 2013
  - ‘Effect of alloying elements and oxygen on hydrogen-induced dimensional changes in alpha zirconium in the presence of radiation-induced defects’, M. Christensen, W. Wolf, C. M. Freeman, E. Wimmer, R. B. Adamson, L. Hallstadius, P. E. Cantonwine, E. V. Mader, In preparation

- Additional information:
  - www.MaterialsDesign.com