materials design®

LIEGE Université
Industry-academia day
on computer simulations of materials properties

Industrial Value of Atomistic Simulations: Successes and Challenges

Erich Wimmer

Materials Design
16 May 2018

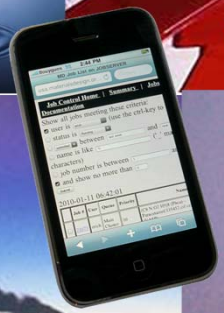
Outline

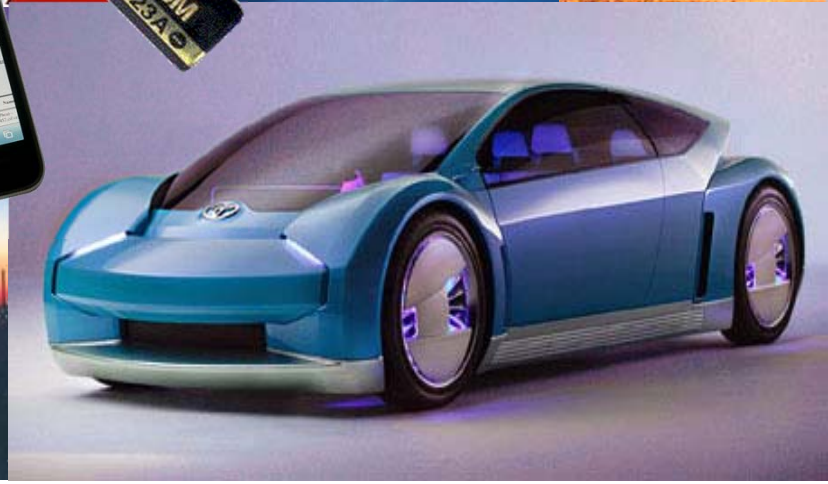
- Industrial values
- Examples of successes
  - Metal alloys
  - Batteries
  - Composite materials
  - Fluids
- Challenges

Industrial Value

Creation of products which are:

- Valuable for customer
- Innovative
- Cost-effective to manufacture
- Safe and reliable
- Environmentally responsible
- Meeting regulations

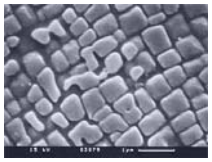


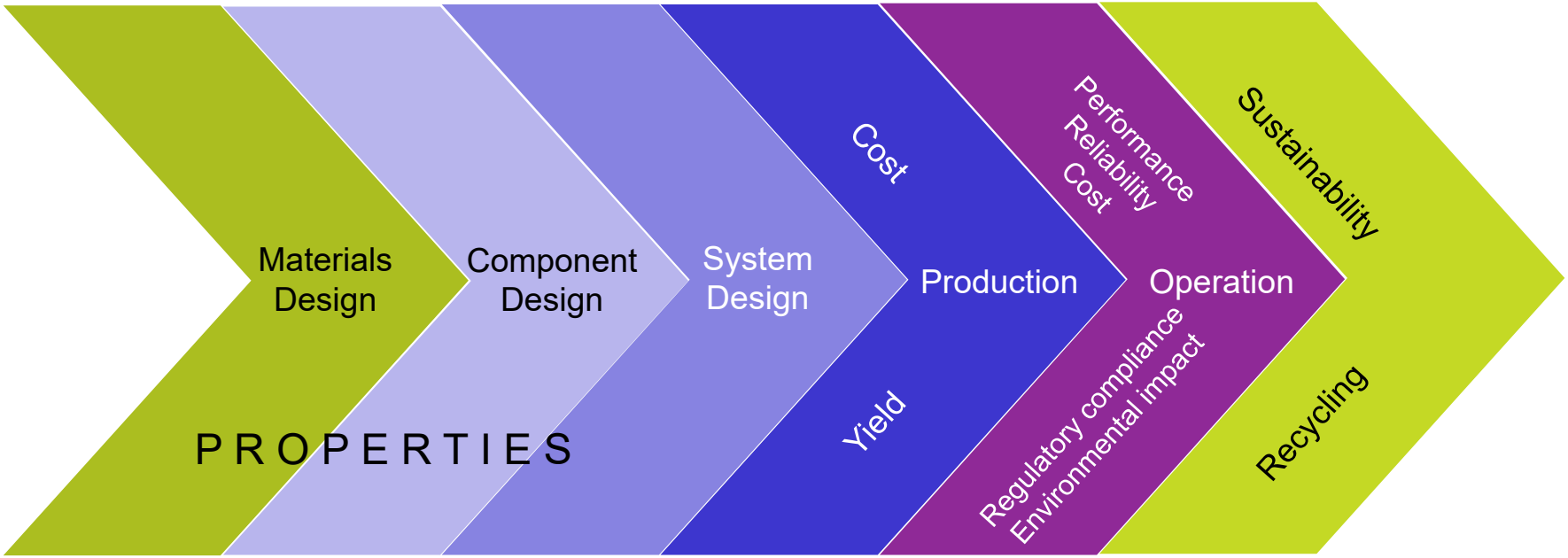


Value of Modeling and Simulation

- Understanding of mechanisms
- Prediction of properties

Optimal design and processing

Innovation Process

- Improvement of industrial products is a long-term iterative process
- Materials modeling and simulation are catalysts of this process

Safety of Nuclear Reactors

Nuclear Energy and Coal

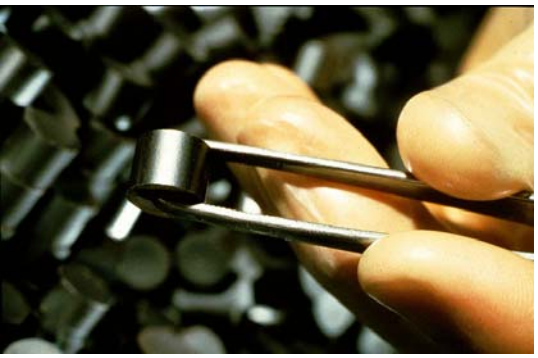
$U \rightarrow I, Cs, Sr, Xe, Ba, \ldots \quad \Delta E \approx 2\times\mathbf{10^{10}}$ kJ/mol

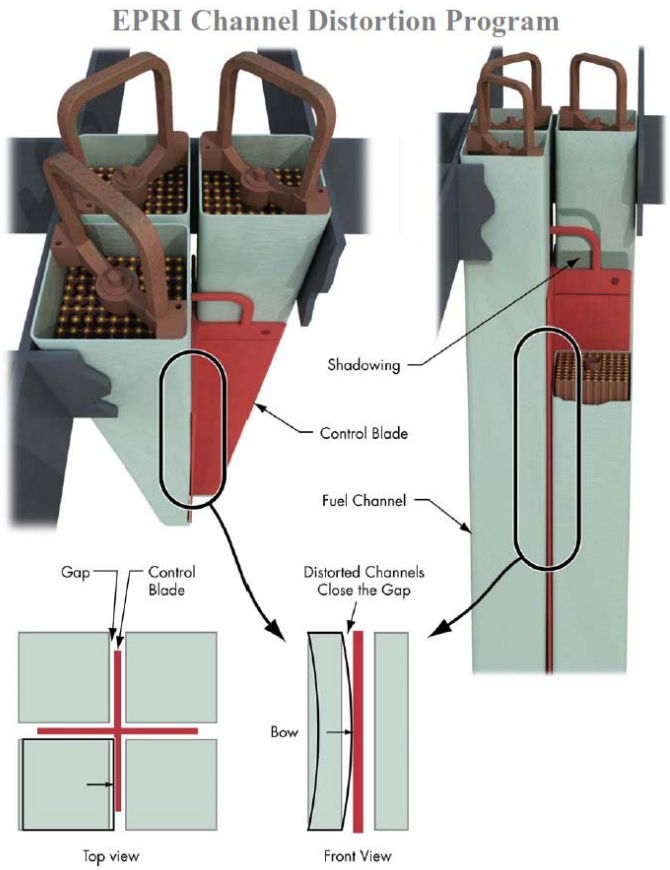
$C + O_2 \rightarrow CO_2 \quad \Delta_f H^0 = 3.935\times\mathbf{10^{2}}$ kJ/mol

Our atmosphere is thin and vulnerable

(ash from coal is radioactive)

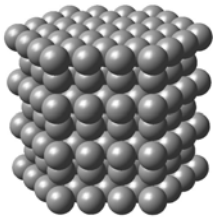
Safety of Nuclear Reactors



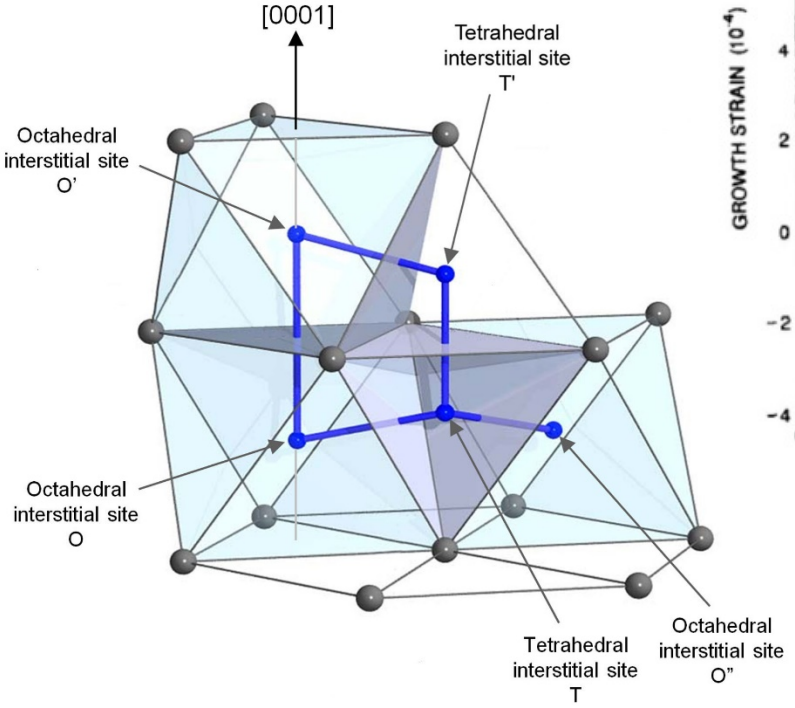


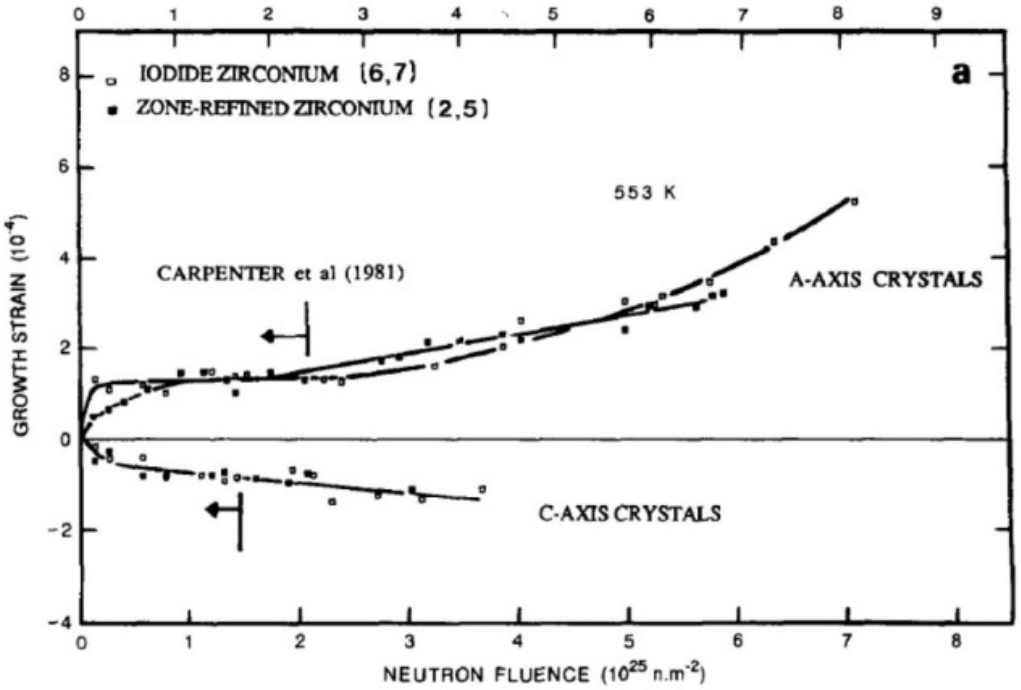
- Reliability of structural materials
  - Channel distortion
- Accident tolerant fuels
- Managing radioactive waste
- Alternative designs

Radiation-induced Swelling



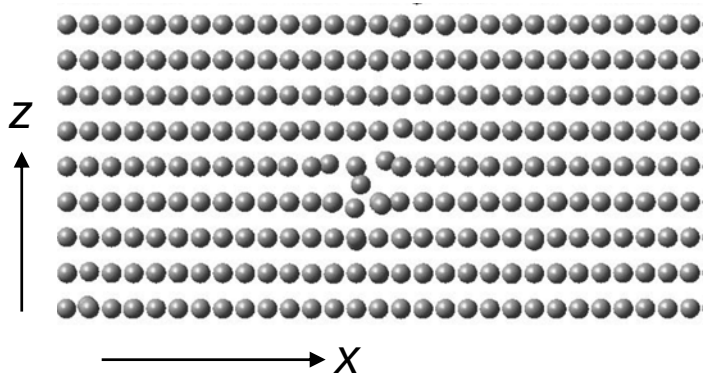
Hexagonal close-packed



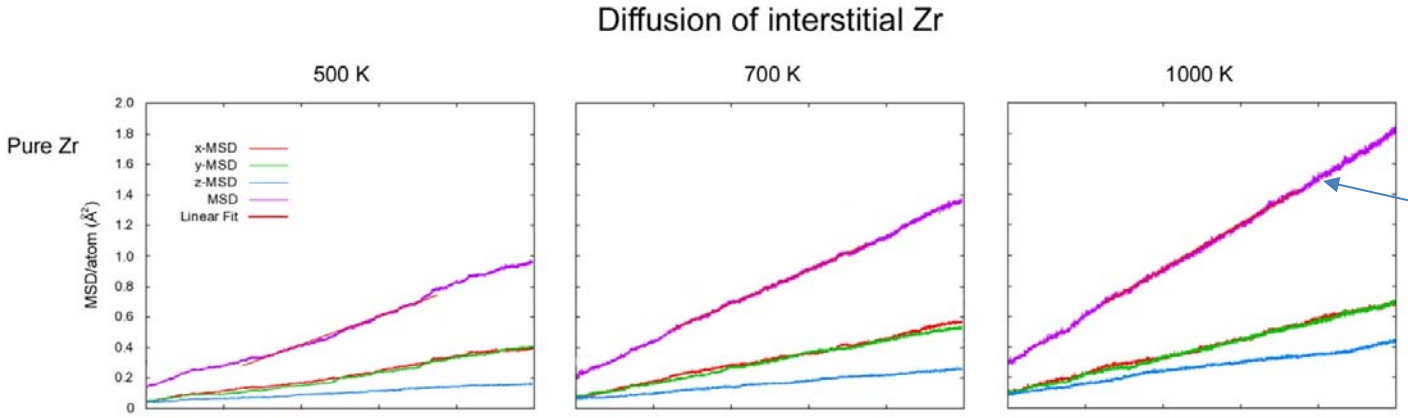


Diffusion of Interstitials in Zr

MedeA® LAMMPS/EAM



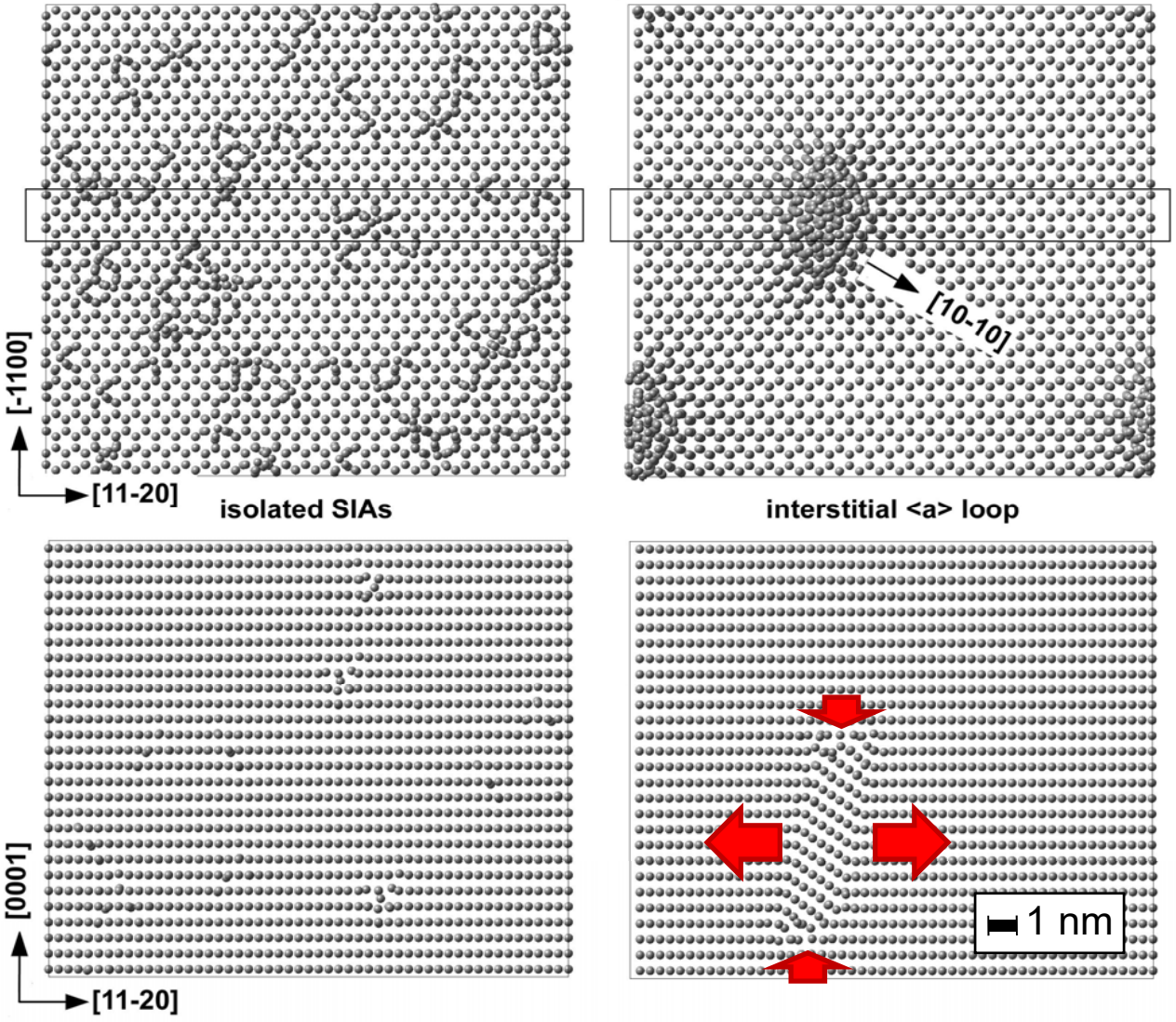
- Fast
- Anisotropic: faster in xy-direction
- Build-up of <a> dislocation loops



Slope of mean square displacement vs. time is proportional to diffusion coefficient

1 ns molecular dynamics

Nucleation of Dislocation Loops



MedeA®-LAMMPS/EAM

Nanoclusters of interstitial atoms cause

expansion in <a>

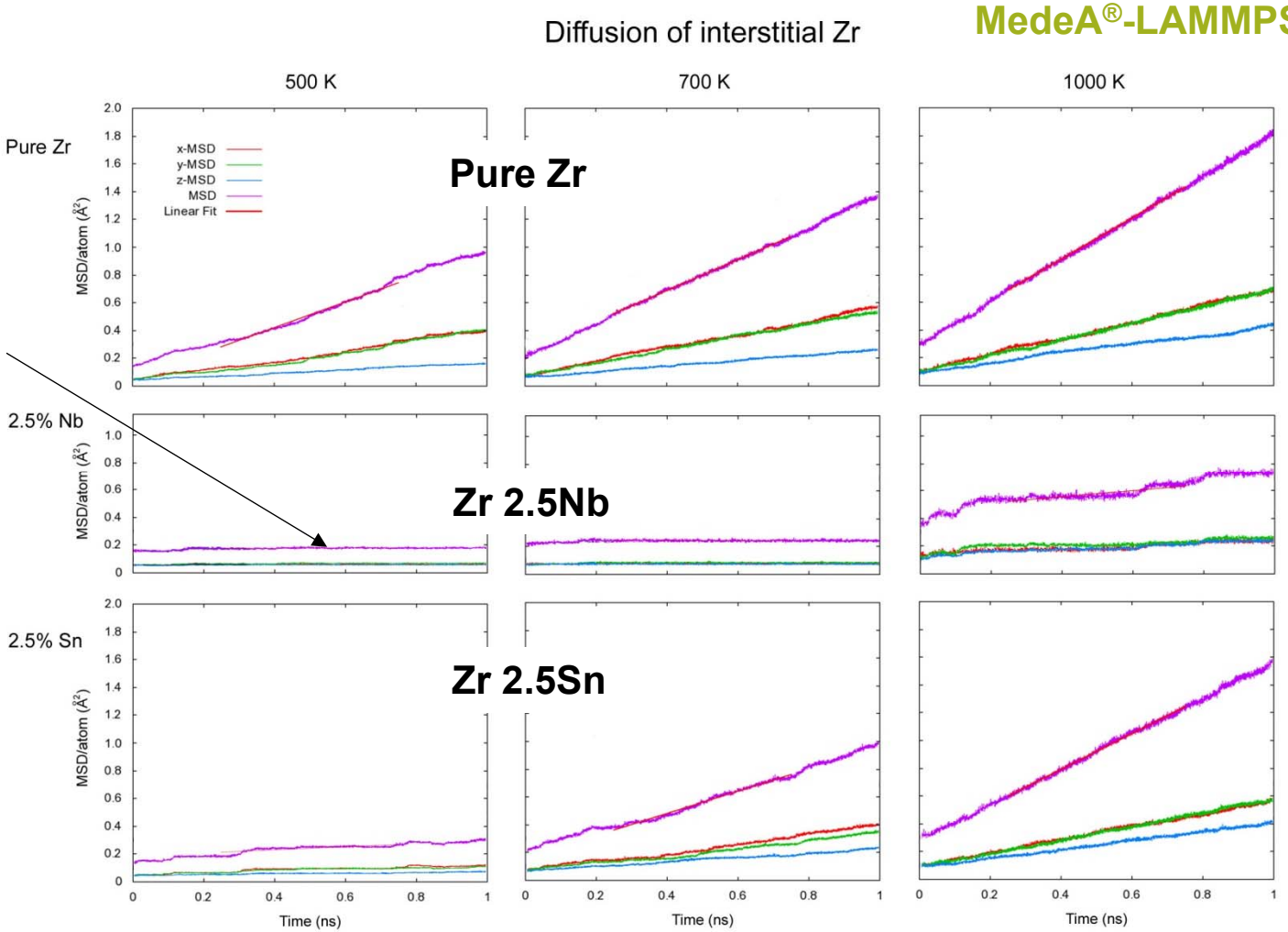
shrinkage in <c>

Consistent with experimental data on radiation-induced growth

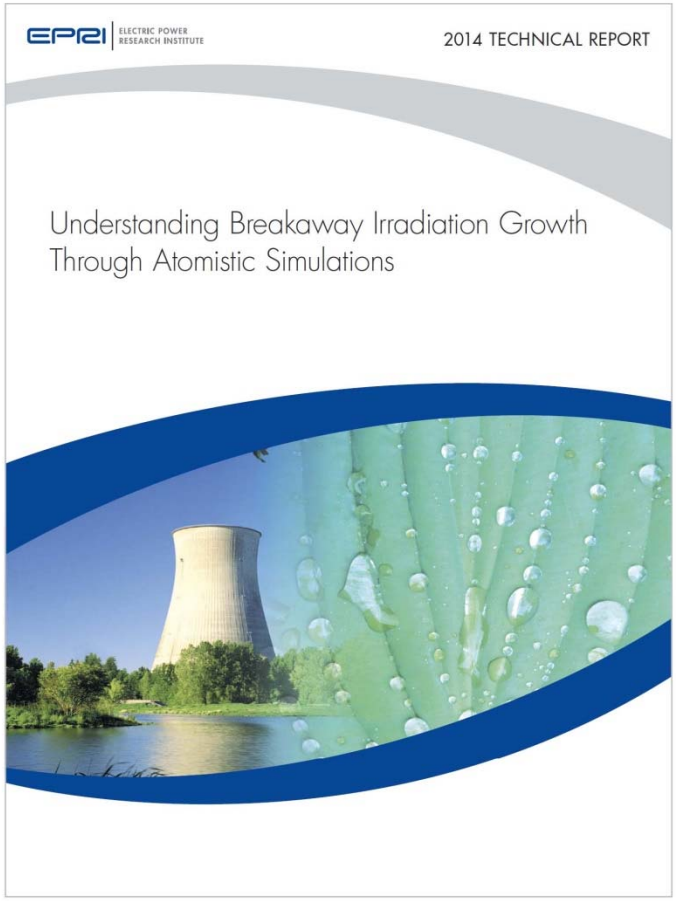
Effect of Alloying with Nb and Sn

MedeA®-LAMMPS/EAM

Nb suppresses interstitial diffusion



References



http://www.epri.com/abstracts/Pages/ProductAbstract.aspx?ProductId=000000003002003185

ASTM Conference, 1 Feb 2013 - Hyderabad, India

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. Freeman, E. Wimmer,
Materials Design Inc., Santa Fe, NM, USA
R. B. Adamson, *Zircology Plus, Freemont, CA, USA*
L. Hallstadius, *Westinghouse Electric Sweden, Västerås, Sweden*
P. Cantonwine, *Global Nuclear Fuels, Wilmington, NC, USA*
E. V. Mader, *Electric Power Research Institute (EPRI), Palo Alto, CA, USA*

John H. Schemel Best Paper Award

Journal of Nuclear Materials 445 (2014) 241–250

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

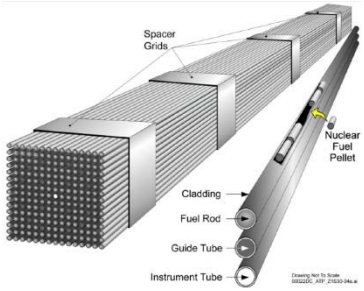
Effect of alloying elements on the properties of Zr and the Zr–H system

M. Christensen [a], W. Wolf [a,*], C.M. Freeman [a], E. Wimmer [a], R.B. Adamson [b], L. Hallstadius [c], P.E. Cantonwine [d], E.V. Mader [e]

[a] Materials Design, Inc., 343 West Manhattan Avenue, Santa Fe, NM 87501, USA
[b] Zircology Plus, 36848 Montecito Dr., Fremont, CA 94536, USA
[c] Westinghouse Electric Sweden AB, SE-721 63 Västerås, Sweden
[d] Global Nuclear Fuel – Americas, P.O. Box 780, M/C F12, Wilmington, NC 28402, USA
[e] Electric Power Research Institute (EPRI), 3420 Hillview Ave, Palo Alto, CA 94303, USA

Accident Tolerant Fuel Systems

Fukushima Daiichi Reactor 12 March 2011

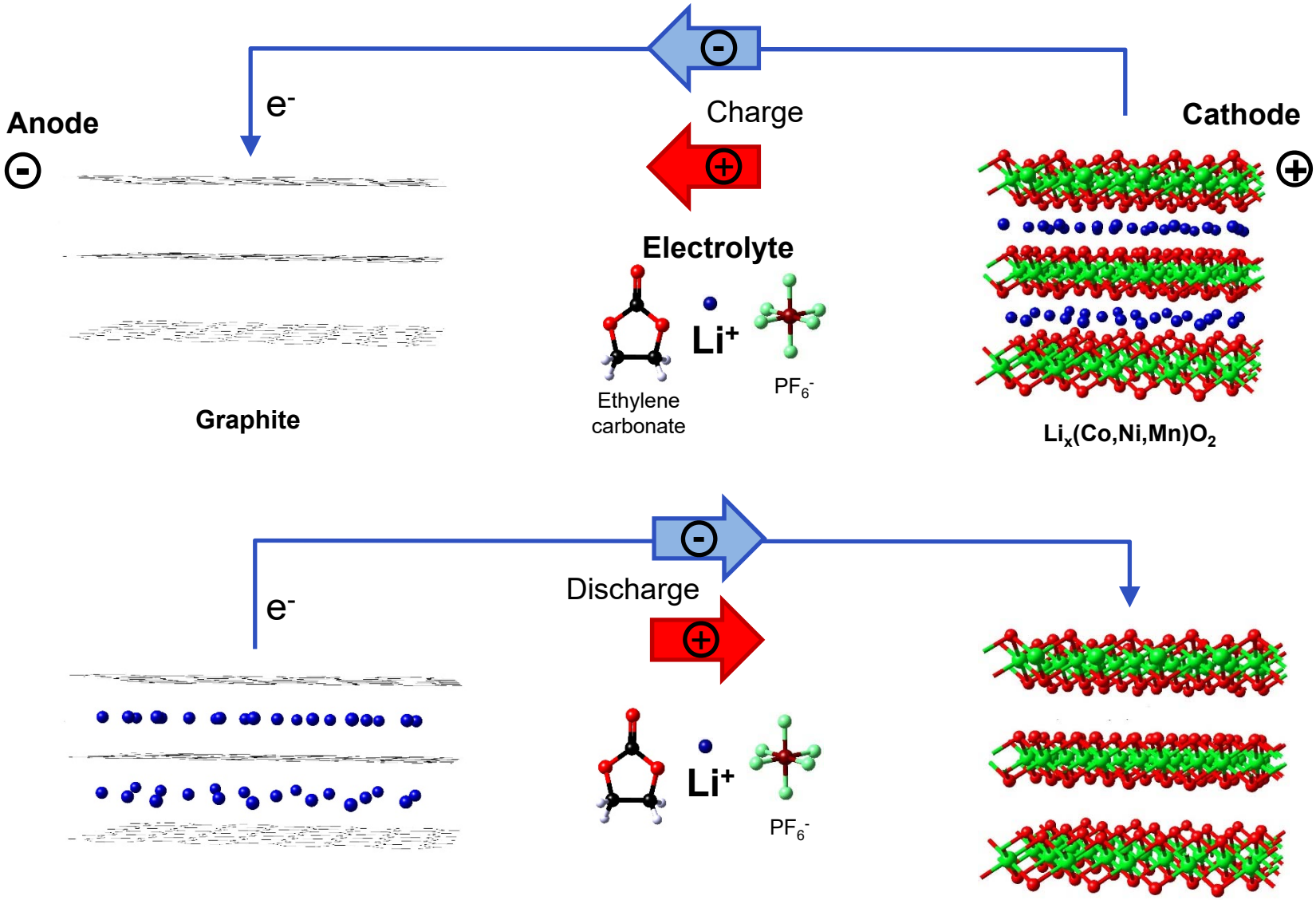


$$Zr + H_2O \rightarrow ZrO_2 + H_2 + heat$$
$$H_2 + ½\ O_2 \rightarrow H_2O + heat$$

- The explosion was caused by a chemical reaction of Zr with water
- Consequence: Cladding of Zr or replacement of Zr by other materials
  - Ni-free steel
  - SiC
  - Other

Batteries

Li-Ion Battery



Issues

- Limited capacity: Li_xCoO_2 is unstable if more than half of Li is removed. Practical operation is restricted to $0.5 \leq x \leq 1$; half of the capacity is not used.
- Li_xCoO_2 expands when Li is removed and contracts when Li is inserted → degradation
- The liquid electrolyte is flammable
- Each cell behaves differently, which is a serious issue in battery packs

Low-strain Cathode Materials

Computational Design and Experimental Verification of Zero- and Low-strain Cathode Materials for Solid-State Li-ion batteries

Fabio Rosciano[1], Mikael Christensen[2], Volker Eyert[2], Alexander Mavromaras[2], Erich Wimmer[2]

[1]Toyota Motor Europe, Advanced Technology 1, Hoge Wei 33, Zaventem, Belgium
[2]Materials Design S.A.R.L., Montrouge, France

TOYOTA

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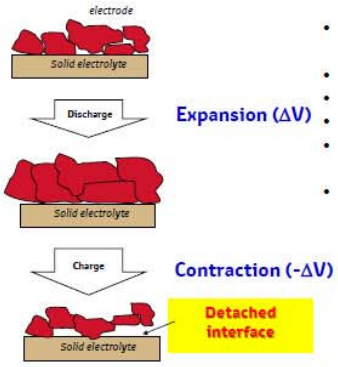
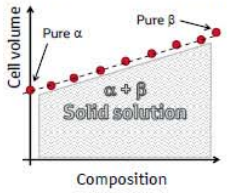
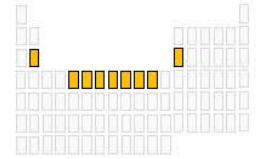
Background



- One of the major degradation factors in Li-ion batteries is the volume change occurring upon charge and discharge in electrode materials [1].
- This volume change can be easily accommodated in conventional composite electrodes, but in the case of a rigid, solid-state battery the stress generated at the grain interfaces will lead to the destruction of the device.
- For the anode, $Li_4Ti_5O_{12}$ is a strong candidate for solid-state Li-ion batteries, giving its zero-strain properties [2].
- For the cathode, on the other hand, no viable zero-strain candidate has been described in the literature yet.
- Finding a zero-strain cathode material with high-voltage by trial-and-error is clearly not a viable strategy → ab-initio Material Design
- We employed DFT calculations to link the volume change, the composition and the lithium content of various precursor materials with the spinel structure.
- Using Vegard's law [3] we mixed the precursors to obtain materials with the desired strain properties

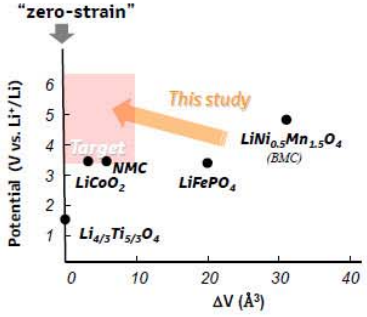
$$\alpha \rightarrow V\alpha$$
$$\beta \rightarrow V\beta$$
$$V(x\alpha + y\beta) = x\,V\alpha + y\,V\beta$$





$$x\,LiM^1{}_2O_4 + y\,LiM^2{}_2O_4 + z\,LiM^3{}_2O_4 = Li(M^1{}_xM^2{}_yM^3{}_z)O_4$$

M = Mg, V, Cr, Mn, Fe, Co, Ni, Cu, Al



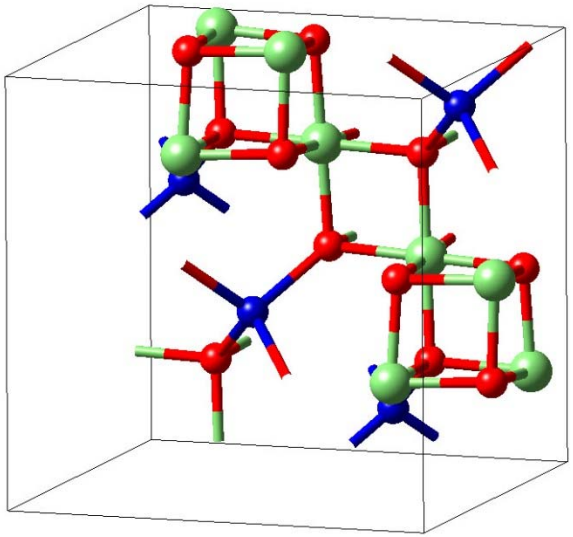
International Battery Association (IBA) Conference, Brisbane, Australia, 3-7 March 2014
Patent WO 2014/191018 A1

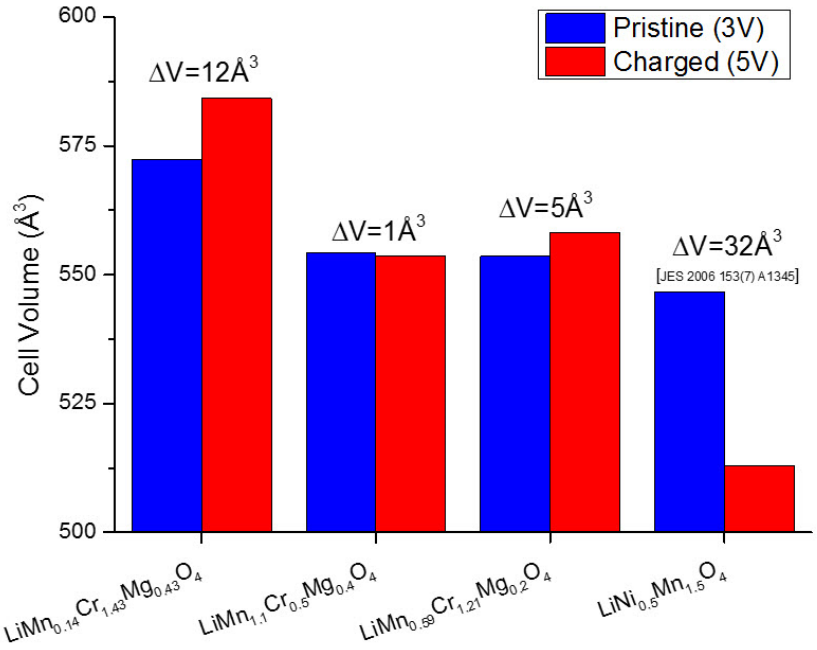
Computations

- Using *ab initio* methods, compute equilibrium volume of spinel structures and identify those compounds which shrink and which expand when Li is removed

$$\mathrm{Li}(\mathrm{M}_x^1, \mathrm{M}_y^2, \mathrm{M}_z^3)\mathrm{O}_4, \quad \mathrm{M} = \mathrm{Mg, V, Cr, Mn, Fe, Co, Ni, Cu, Al}$$

- Find compounds close to zero expansion when Li atoms are inserted





Patent

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
4 December 2014 (04.12.2014)

WIPO | PCT

(10) International Publication Number
WO 2014/191018 A1

(51) International Patent Classification:
C01G 45/12 (2006.01) *H01M 10/052* (2010.01)
H01M 4/505 (2010.01) *H01M 10/0562* (2010.01)

(21) International Application Number:
PCT/EP2013/060881

(22) International Filing Date:
27 May 2013 (27.05.2013)

(25) Filing Language: English

(26) Publication Language: English

(71) Applicant: **TOYOTA MOTOR EUROPE NV/SA** [BE/BE]; Avenue du Bourget 60, B-1140 Brussels (BE).

(72) Inventors: **ROSCIANO, Fabio**; Milcampslaan 127, B-1030 Schaarbeek (BE). **CHRISTENSEN, Mikael**; Branningevagen 1, S-120 54 Arsta (SE). **EYERT, Volker**; Baumschulenweg 6A, 14469 Potsdam (DE). **MAVRO-MARAS, Alexander**; Bergandsgatan 43, S-133 41 Saltsjobaden (SE). **WIMMER, Erich**; 3 avenue du Commerce, F-78000 Versailles (FR).

(74) Agents: **HART-DAVIS, Jason** et al.; Cabinet Beau de Lomenie, 158 rue de l'Université, F-75340 Paris Cedex 07 (FR).

(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

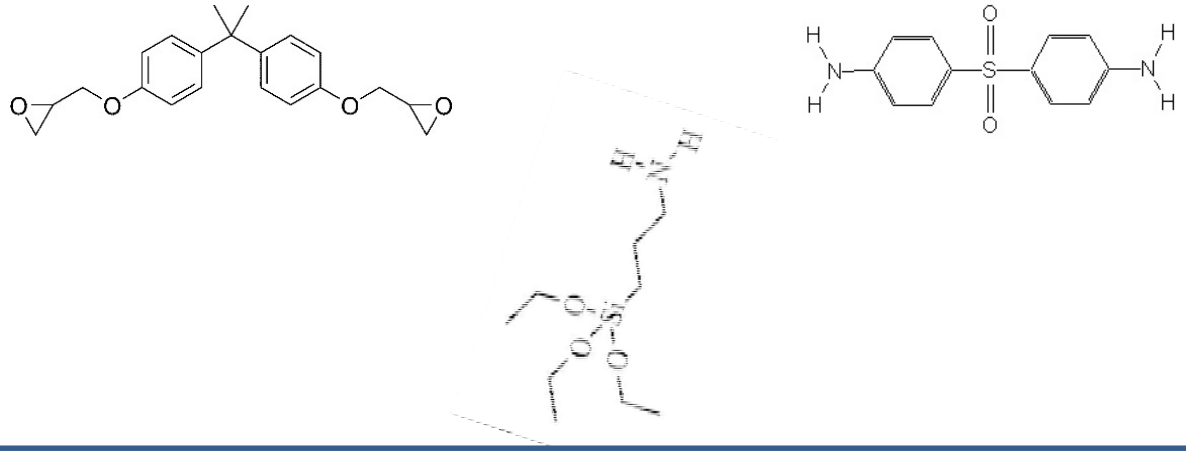
Published:

— *with international search report (Art. 21(3))*

Composite materials

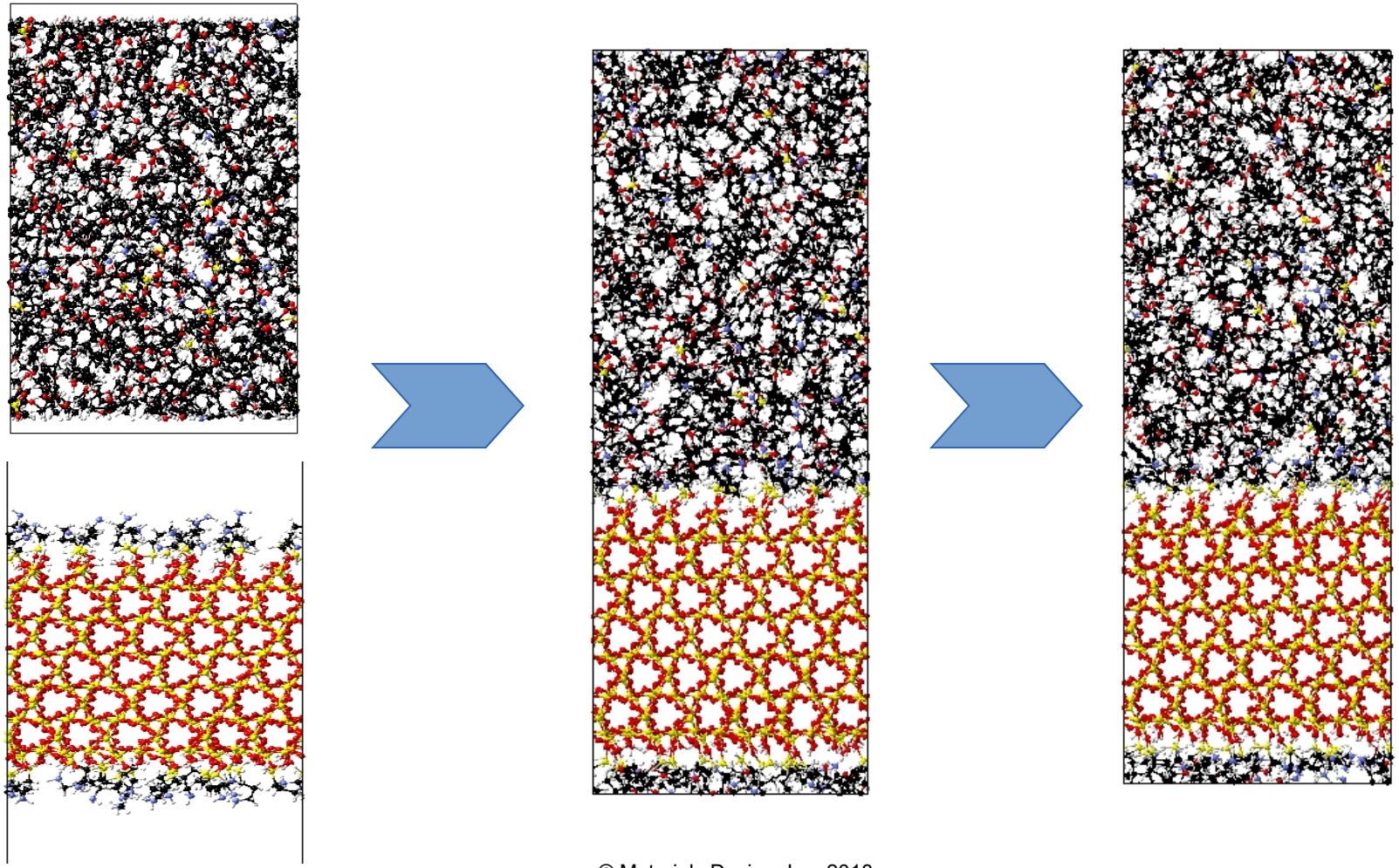
Epoxy-Oxide Interfacial Systems

How much primer (coupling between SiO_2 surface and polymer) is optimal?



Epoxy-SiO_2 Interfaces – Model Building

Primed SiO_2+ unreacted epoxy, crosslinking and equilibration



Epoxy-SiO_2 Interfaces – Mechanical Properties

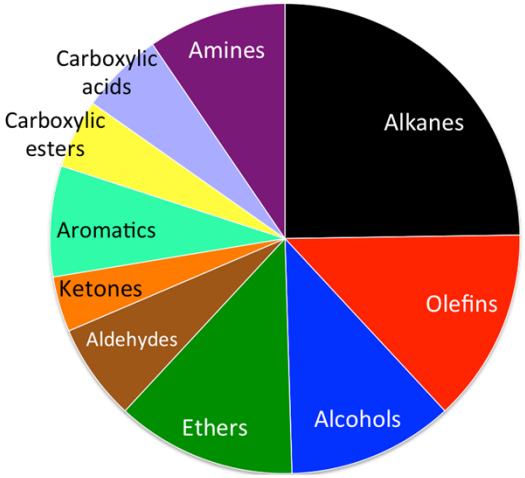
Stiffness coefficient C_{33} for SiO_2 - epoxy composite layer systems with various levels of primer coverage:

Primed surface sites (%)	C_{33} (GPa)
0	7.2
12.5	7.8
25	8.8
50	5.0
Reference: Bulk Cured Resin	5.97 +/-0.29

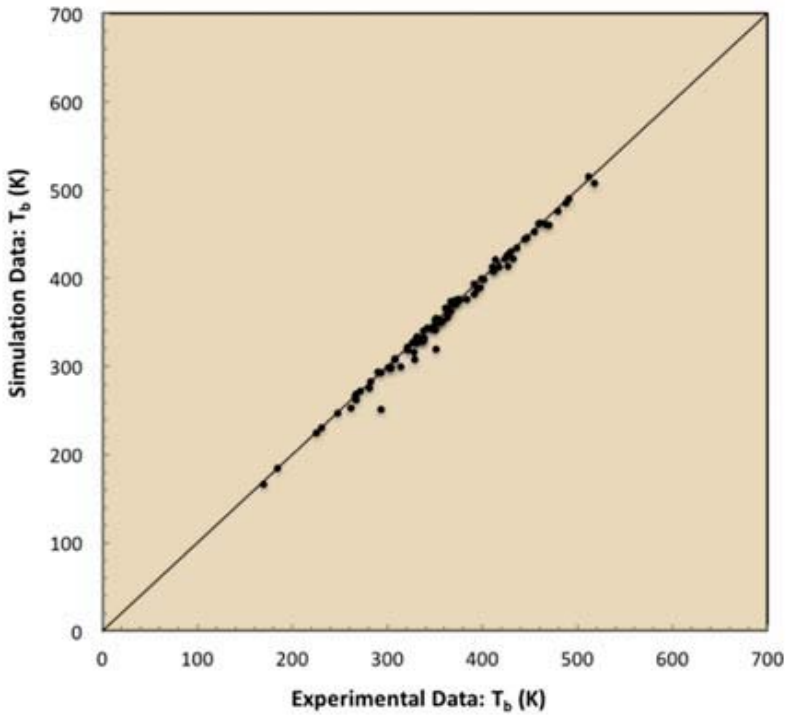
Intermediate primer coverage produces optimal enhancement in small strain mechanical behavior.

Fluids

Predicting Boiling Point Temperature



- ~100 compounds
- Average absolute error of the T_b calculated by Gibbs-ensemble Monte Carlo simulation from the DIPPR [1] data, is 1.4%.
- More than half of the compounds have an absolute deviation of the T_b below 1.0%.

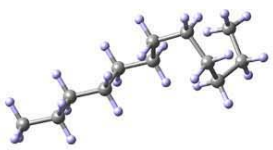


MedeA®-GIBBS simulations (AUA & TraPPE-UA FF) by M. Yiannourakou and P. Ungerer

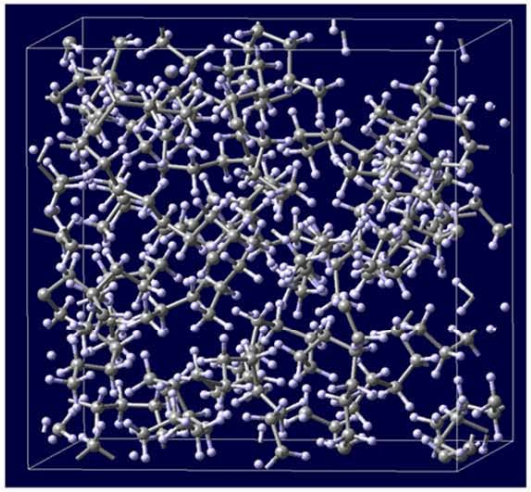
1. Wilding WV, Rowley RL, Oscarson JL. Dippr project 801 evaluated process design data. Fluid Phase Equilib. 1998;150–151:413–420

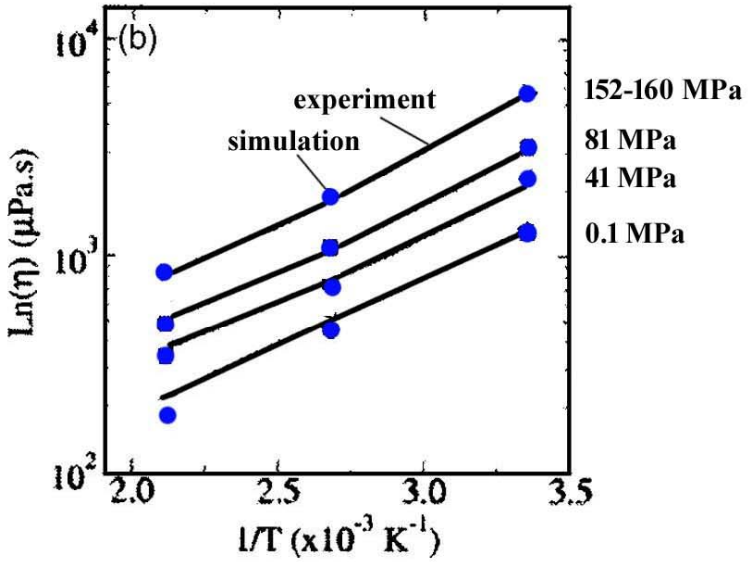
Viscosity of Diesel Fuel

- Industrial question: What is the viscosity of a Diesel fuel at high pressure and temperature (fuel injection)



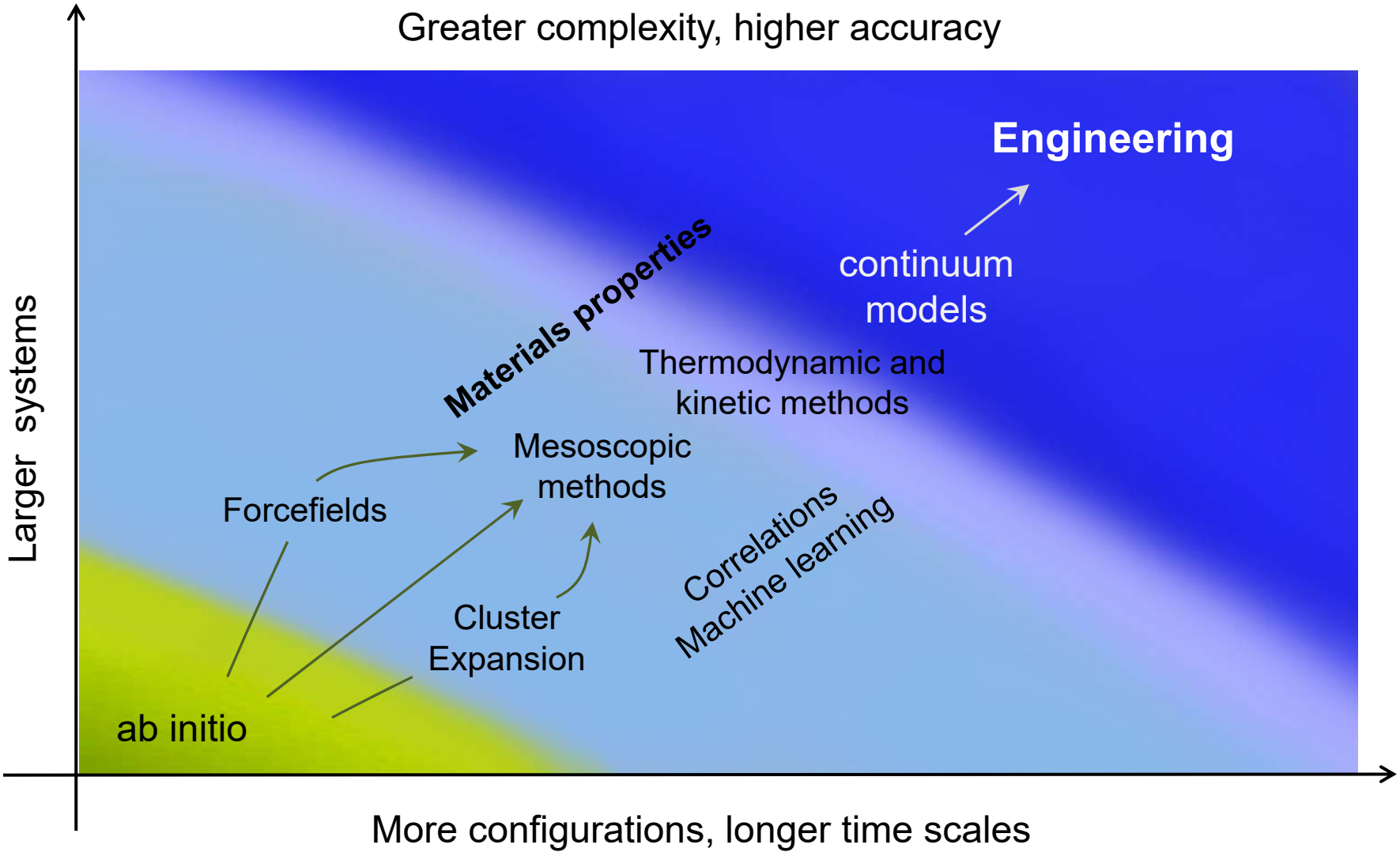
n-dodecane





after C. Nieto-Draghi, P. Ungerer, and B. Rousseau, J. Chem. Phys. **125**, 044517 (2006)

Scientific Challenges



Practical Challenges

- What can be easily computed is usually industrially not important, and what is industrially important is not easy to compute
- We need translators: industrial problems ↔ simulations
- We need simulation software
  - Applicable to variety of complex systems
  - Validated
  - Easy to use
  - Interoperable
  - Well supported - over many decades

European Materials Modelling Council
www.emmc.info

Conclusions

- Atomic-scale modeling and simulations are becoming an integral part of industrial R&D
- Value
  - Understanding mechanisms
  - Prediction of properties

Innovation
Better products & processes
Higher efficiency

- Examples
  - Zr alloys
  - Batteries
  - Composites
  - Fluids
- Need higher accuracy and better coupling in multi-scale models: theoretical approaches, algorithms, software
- Highly skilled scientists and engineers