РОССИЙСКАЯ АКАДЕМИЯ НАУК
Институт проблем безопасного развития атомной энергетики

RUSSIAN ACADEMY OF SCIENCES
Nuclear Safety Institute (IBRAE)

Development of the Mechanistic Fuel Performance And Safety Code SFPR Using the Multi-Scale Approach

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MMSNF-2013
Chicago, IL
October 14-16, 2013

SFPR Code Structure

SFPR code

Single Fuel Rod ↔Performance

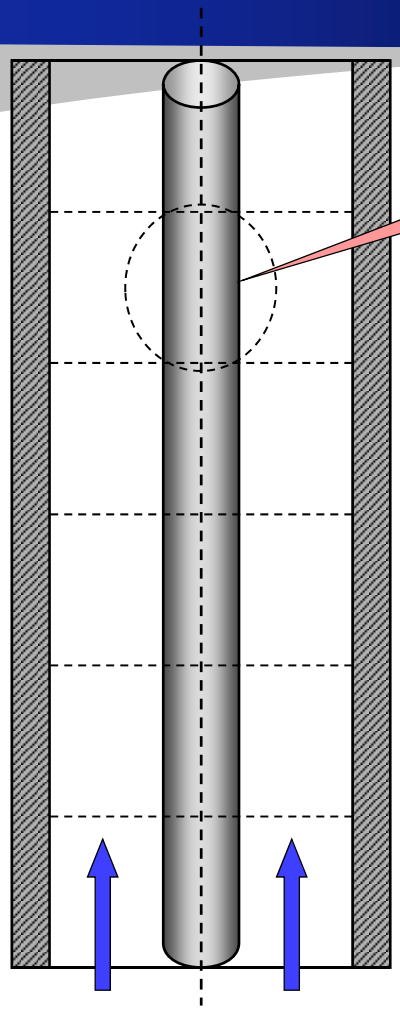
SVECHA code

- Advanced mechanistic code for modeling thermo-mechanical and physico-chemical behavior of a single LWR fuel rod
- Initially designed for several accident regimes, extended to normal operation conditions
- Collaboration of IBRAE with IRSN (France), FZK (Germany) JRC/IE, US NRC

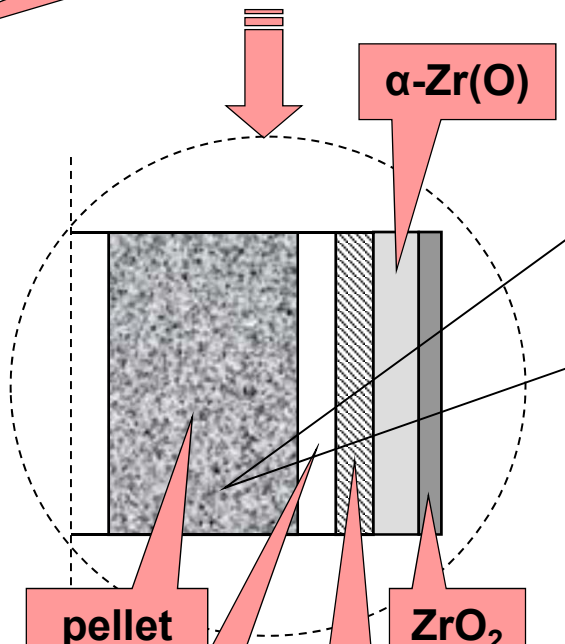
MFPR code

- Advanced mechanistic code for modelling fission product release from irradiated UO_2 fuel
- Various regimes of reactor operation, including severe accidents, and high burnups
- Collaboration of IBRAE with IRSN (France)

SFPR Meshing



vertical mesh



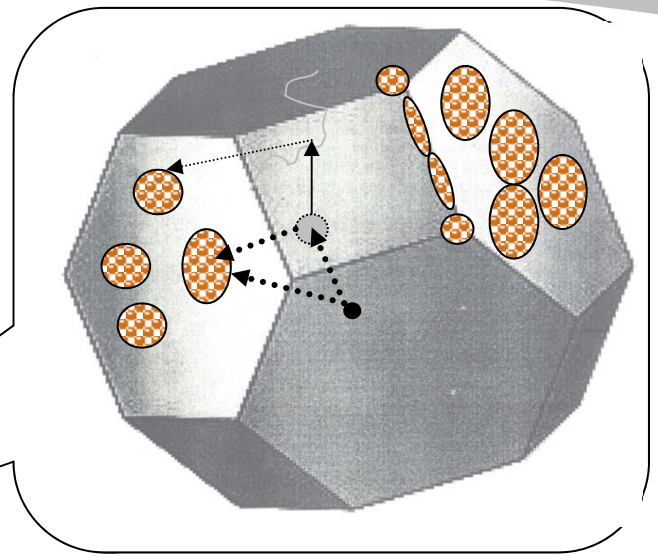
α-Zr(O)

pellet

gap

Zr cladding

ZrO_2



MFPR

Coolant flow:
$H_2O(l, g)$, O_2, Ar, H_2

SVECHA

Self-consistent description of thermo-mechanical and physico-chemical behaviour of single fuel rod

MFPR
Module for Fission Product Release

- **IBRAE-IRSN co-operation (since 1995)**
  - IBRAE: models and code development
  - IRSN: code application to interpretation of FP behavior (VERCORS, PHEBUS…), benchmarking of ELSA code (in ASTEC)

- **Project Realization**
  - Development of the initial code version as an upgraded replica of progenitor code VICTORIA
  - Further improvement of physical models, inclusion of new individual effects modelling

MFPR Development

- **Extensive validation**
  - Separate-effect tests
    (steady irradiation, transients, annealing)
  - Integral tests (HI-VI, VERCORS)
  - Reactor tests (SILOE, Ris∅)
- **Sensitivity study** (SUNSET)
  - Identify important parameters
- **External evaluation**
  (Hj. Matzke (ITU), R. Grimes (UK), 2005)
  - Largely positive and stimulating conclusions
  - Recommendations in agreement with internal plans (currently realized)

MFPR Models

- **Mechanistic description**

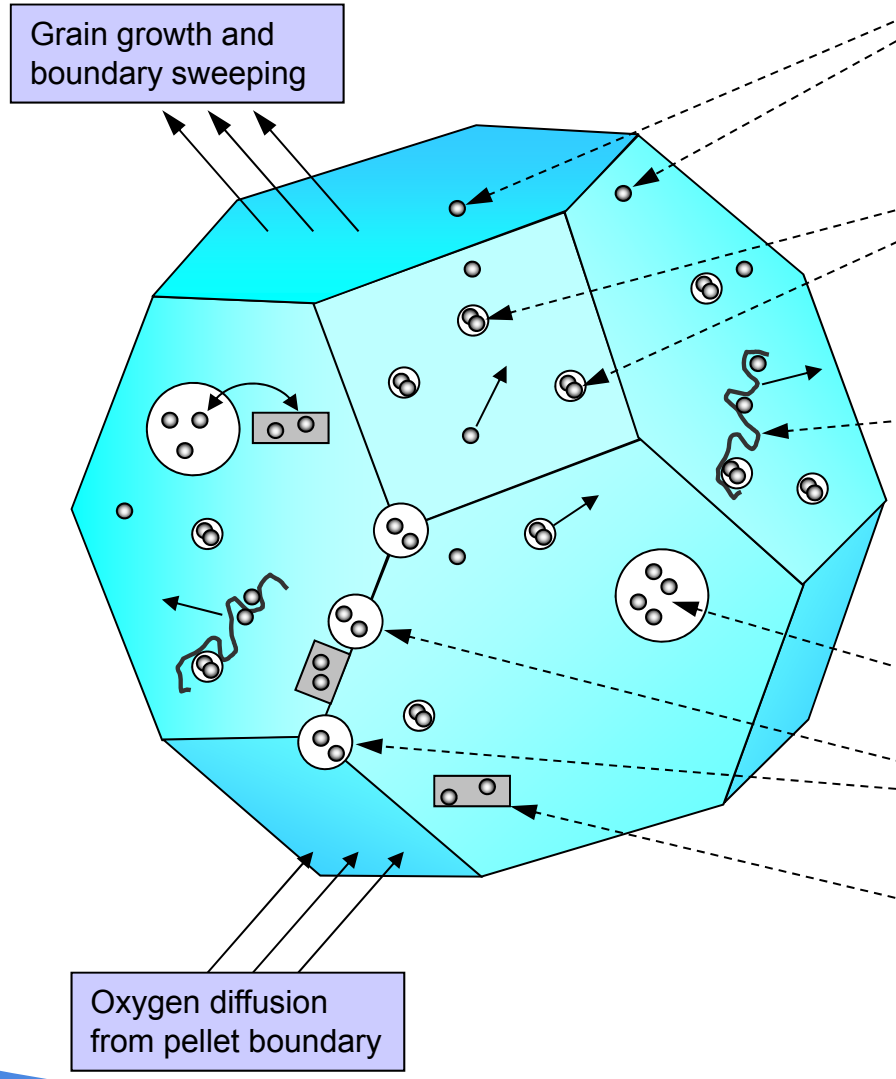
  of FP behavior in irradiated UO_2

  - In irradiation regime: steady state and transients
  - In annealing regime
  - In accidental conditions: LOCA, severe accidents

- **Three groups of models** (tightly coupled)

  - Fission products (~200–400 stable and radioactive) transport (intra- and inter-granular)
  - Evolution of fuel microscopic defect structure
  - Thermochemistry
    - Chemical compound and phase formation
    - Fuel oxidation/vaporisation in steam / hydrogen / inert mixtures

MFPR Basic Processes



Intragranular gas atoms
- diffusion toward the grain boundary
- biased migration in external gradients
- bubble nucleation
- capture by bubbles and pores
- sweeping by grain boundary and dislocations

Intragranular bubbles
- diffusion toward the grain boundary
- biased migration in external gradients
- bubble nucleation
- coalescence & capture of gas atoms
- loss of atoms by thermal and recoil resolution
- sweeping by grain boundary and dislocations

Dislocations
- creep toward grain boundaries
- trapping of intragranular gas and bubbles

Intragranular zone

Bubbles
- fed by diffusion gas and bubble flux out of grain vaporization of condensed phase species
- irradiation induced resolution of atoms
- chemical reactions in gas phase

Face bubbles
- growth & interconnection leading to formation of grain face channels toward edges

Edge bubbles
- swelling until a critical threshold
- release to open porosity

Precipitates
- fed by diffusion gas and bubble flux out of grain, flux from grain face bubbles
- formation of metallic (Ru, Mo), ternary (Cs_2MoO_4 etc) and CsI phases
- thermochemical equilibrium between precipitates and gas phase

Intergranular zone

Intra-granular Transport of Fission Gas

$$\frac{\partial \mathbf{X}(R,t)}{\partial t} = \mathbf{F}(\mathbf{X};R,t), \quad \mathbf{X} = \left\{C_g, Y_b, N_b, V_b, \rho_d, Y_d, C_v, C_i, C_{vl}, C_{il}, R_{il}\right\}$$

diffusion

Bubble interactions with:
gas
dislocations
vacancies

$$\frac{\partial C_g}{\partial t} = D_g \Delta C_g - F_{g\to b} - F_{g\to d} + \kappa G$$

$$\frac{\partial Y_b}{\partial t} = D_b \Delta Y_b - \omega_{bmg} Y_b + F_{g\to b} - F_{b\to d} + \nabla\left(\mathbf{v}_{vac} Y_b\right)$$

Bubble biased migration

$$\frac{\partial N_b}{\partial t} = k_{bb} Y_b - k_{gg} F_n \frac{C_g^2}{2C_b} + k_{gb}\left(C_g - C_g^{eq}\right) - bGN_b \varphi(N_b)$$

Bubble coalescence, nucleation, growth, thermal & irradiation resolution

$$F_{g\to b} = k_{gg} F_n C_g^2 + k_{gb}\left(C_g - C_g^{eq}\right)\frac{Y_b}{N_b} - bGY_b$$

JNM 277 (2000) 67
NED 236 (2006) 179
JNM 362 (2007) 327
JNM 376 (2008) 174

Inter-granular Transport of Fission Gas

- **Transport:** *grain face (gf)* → *grain edge (ge)* ("percolation" at coverage ≈ 50%)

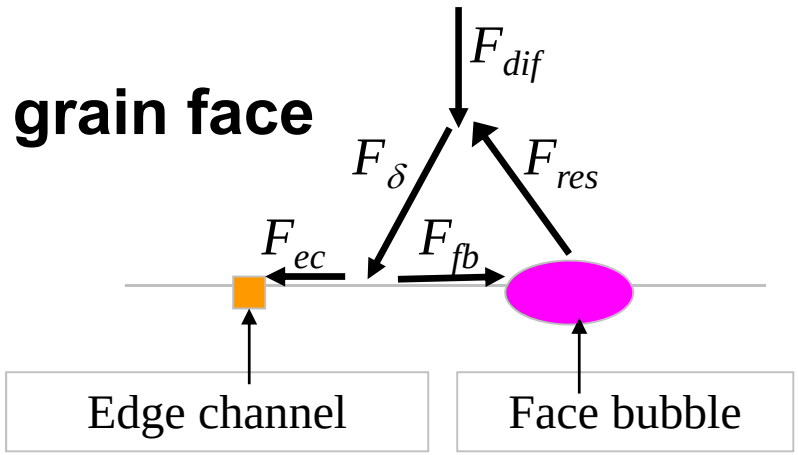
  grain edge → *open porosity (op)*

$$J_{gf\to ge} = \frac{N_{gf}}{\tau_{gf\to ge}}\left(1-\frac{N^{crit}_{gf(gas)}}{N_{gf(gas)}}\right),\quad J_{ge\to op} = \xi^{collapse}_{ge} J_{gr+gf\to ge} + \frac{N_{ge}}{\tau_{ge\to op}}\left(1-\frac{N^{crit}_{ge(gas)}}{N_{ge(gas)}}\right)$$

- **Gas atoms "circulation" over grain face**

  Resulting in notable gas release below percolation threshold (at coverage ≈ 25%)

  JNM 392 (2009) 78-84; 85-89



- **Bubbles coalescence** JNM 374 (2008) 44

  Two mechanisms: ***diffusion and growth impingement***

Model for Evolution of Defect Structure

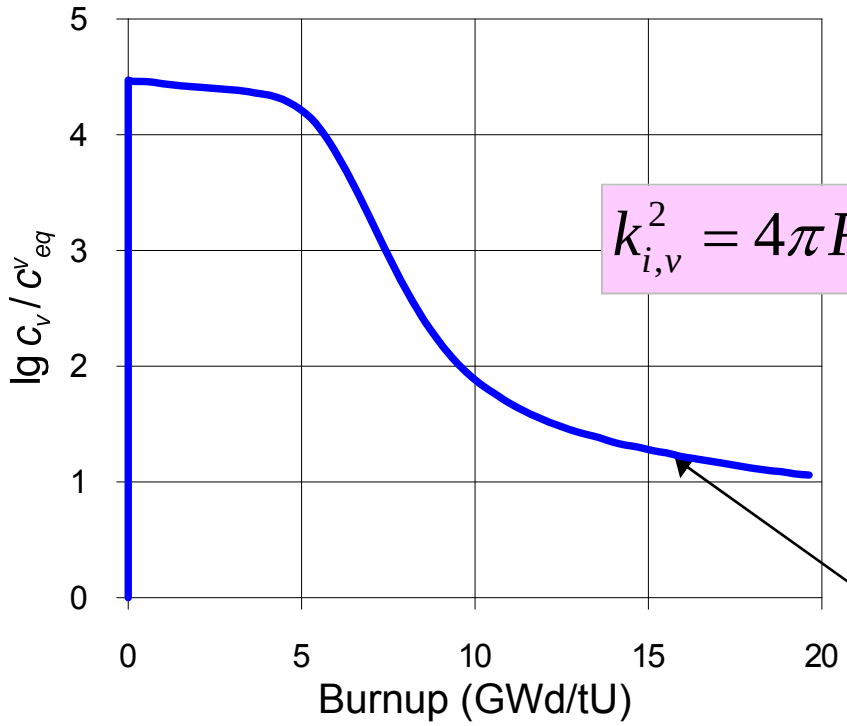
$$\frac{\partial c_v}{\partial t} = \begin{cases} -(k_v^2 + k_{vgb}^2) D_v c_v - \alpha D_i c_i c_v + K_e + K_b + K(1-\zeta_l), & irradiation \\ D_v \Delta c_v - k_v^2 D_v c_v + K_e, & annealing \end{cases}$$

Bubbles

Vacancy clusters

$$k_{i,v}^2 = 4\pi R_b C_b + Z_{i,v}\left(\rho_d + 2\pi R_{il} C_{il} + 2\pi R_{vl} C_{vl}\right)$$

Dislocations



Suppression of bubble nucleation

$$F_{nuc} = c_v$$

JNM 384 (2009) 12

Transport of Chemically Active FP

CLASSES OF CHEMICAL ELEMENTS

Cs Ce I Eu Mo Nd Ru Nb
Ba Sb Sr Te Zr Xe La

THE MAIN PHASES

SOLID SOLUTION

Cs Ba BaO Sr SrO Zr ZrO_2 Nb NbO NbO_2
La $LaO_{3/2}$ Ce $CeO_{3/2}$ CeO_2 Eu $EuO_{3/2}$ EuO
Nd $NdO_{3/2}$ Sb $SbO_{3/2}$ Mo MoO_2 Ru RuO_2

GAS PHASE

Xe Te I Cs CsI Cs_2MoO_4 MoO_3 $(MoO_3)_2$
$(MoO_3)_3$ RuO RuO_2 RuO_3 Ba Sr ZrO LaO
CeO NdO NbO O_2 …

”WHITE” (METAL) PHASE

Mo Ru ...

“GREY” PHASE (TERNARY COMPOUNDS)

$BaUO_4$ $SrUO_4$ Cs_2UO_4 $BaMoO_4$ $SrMoO_4$
Cs_2MoO_4 $BaZrO_3$ $SrZrO_3$ Cs_2ZrO_3

SOLID PHASE CsI

CsI

Intragranular FP Transport:

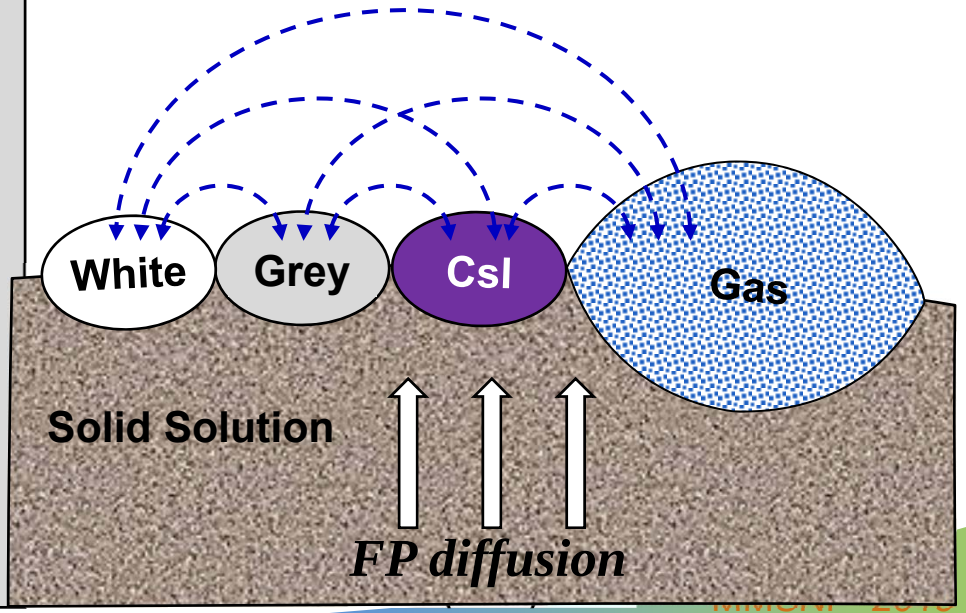
FP diffusion

Boundary conditions:

Thermochemical equilibrium between different phases at grain the boundary

FP release:

Transport to the open porosity via the gas phase



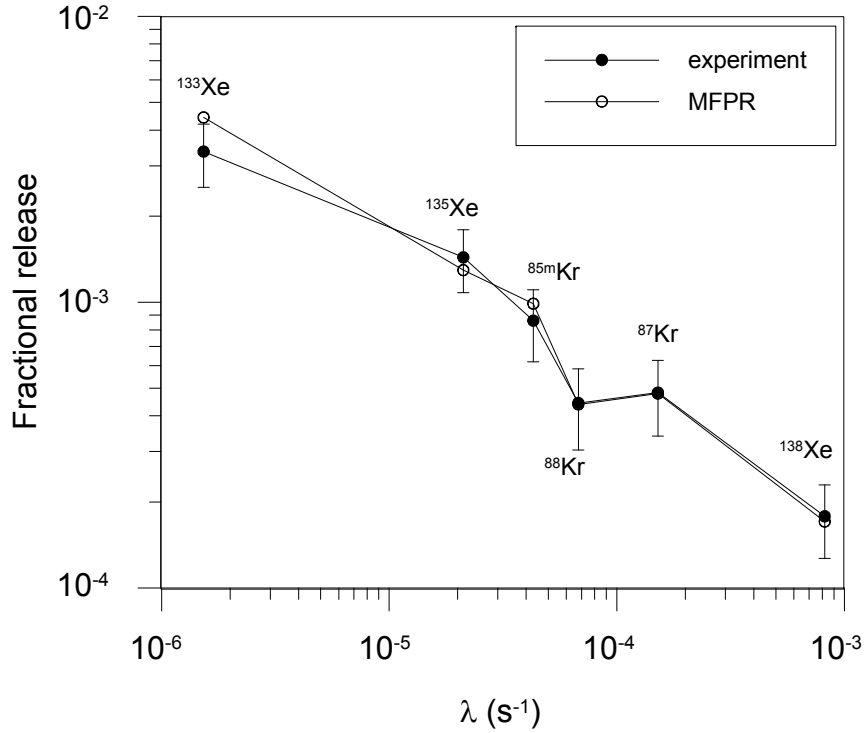
Transport of Radioactive FPs

$$\frac{\partial \mathbf{c}(r,t)}{\partial t} = \mathbf{D}\Delta\mathbf{c}(r,t) - \boldsymbol{\lambda}\mathbf{c}(r,t) + \mathbf{s},$$

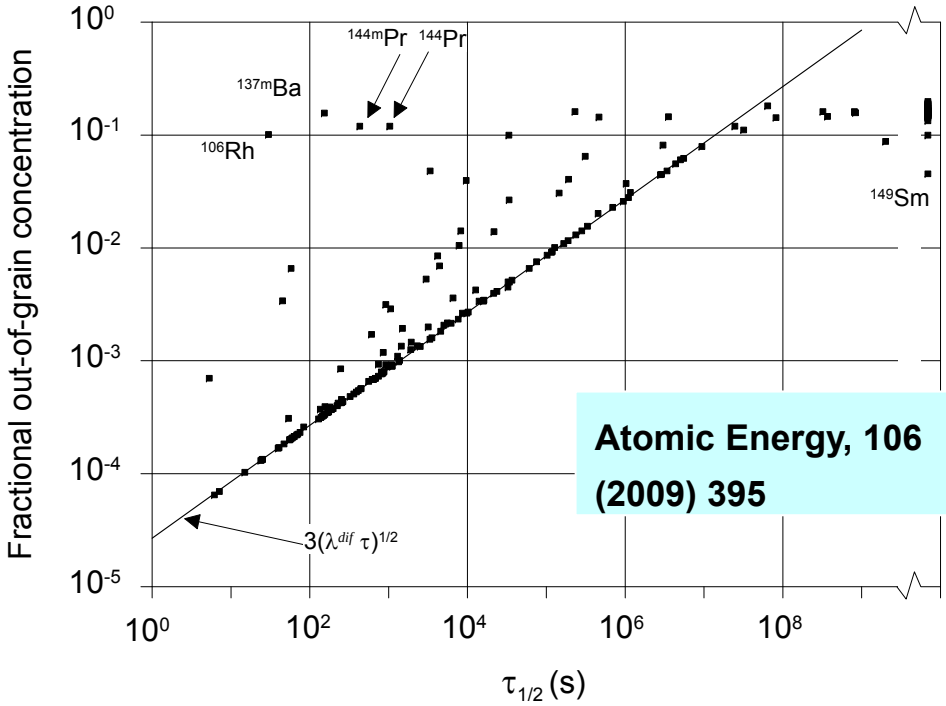
$$\mathbf{c}\big|_{r=R_{gr}} = \mathbf{c}^{bnd}(t), \quad \mathbf{c}\big|_{t=0} = \mathbf{c}^{0}(r),$$

$$\mathbf{c}(r,t) = \mathbf{c}^{bnd}(t) + \sum_{n=1}^{\infty} \mathbf{w}_n(t) L_n \frac{X_n(r)}{r}$$

$$\frac{d}{dt}\mathbf{w}_n(t) = -\boldsymbol{\lambda}_n \mathbf{w}_n + \mathbf{S} - \frac{d}{dt}\mathbf{c}^{bnd} - \boldsymbol{\lambda}\mathbf{c}^{bnd}$$



Verification against [Turnbull et al., 1982]



Atomic Energy, 106 (2009) 395

Predictions for reactor conditions

Verification Matrix (1)

Separate-effect tests (examples)

Test	Regime	Measured characteristics
Baker (UK) Cornell (UK) Une-Kashibe (Japan)	Steady irradiation High burn-up	Intragranular bubble sizes and concentrations
Turnbull (UK)	Steady irradiation	Grain growth, Xe release
Zimmermann (Germany)	Steady irradiation	Xe release, fuel swelling
Matzke (ITU)	Transient	Bubble size distribution
Zacharie (CEA) Une-Kashibe (Japan)	Post-irradiation annealing	Bubble size distribution, Xe release , fuel swelling

Verification Matrix (2)

Integral Tests

Test	Regime	Measured characteristics
MCE 1–5 (CRL)	annealing in inert atmosphere	FP release kinetics, fuel stoichiometry
UCE 9–1 (CRL)	annealing in steam	FP release kinetics, fuel stoichiometry
VERCORS (IRSN-EDF, CEA)	annealing in steam and steam/hydrogen	FP release kinetics, fuel stoichiometry
VI (NRC-ORNL)	annealing in steam and steam/hydrogen	FP release kinetics, fuel stoichiometry
Phebus FPT-0 Phebus FPT-1	annealing in steam and steam/hydrogen	FP release kinetics

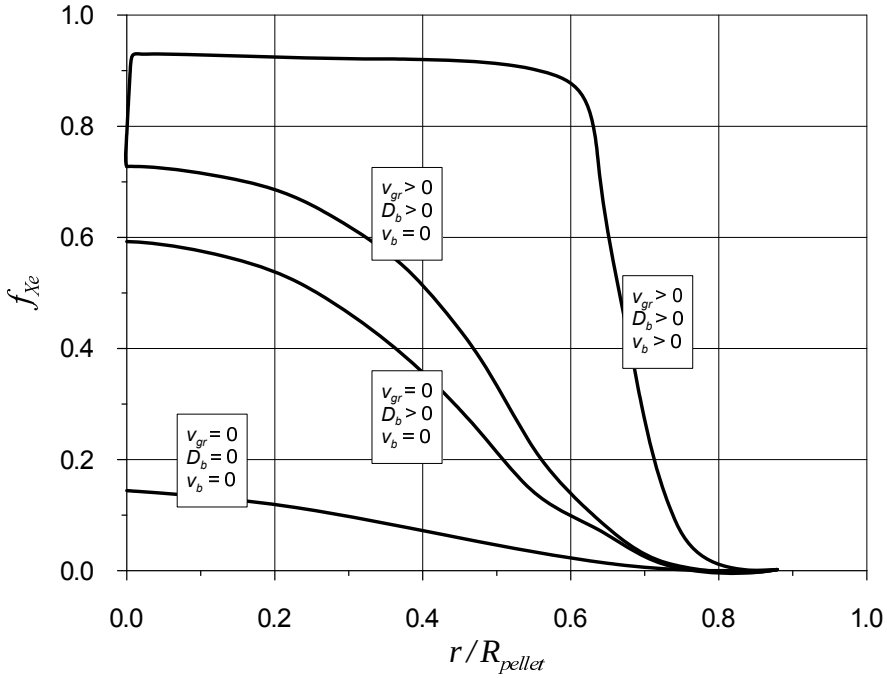
Verification Matrix (3)

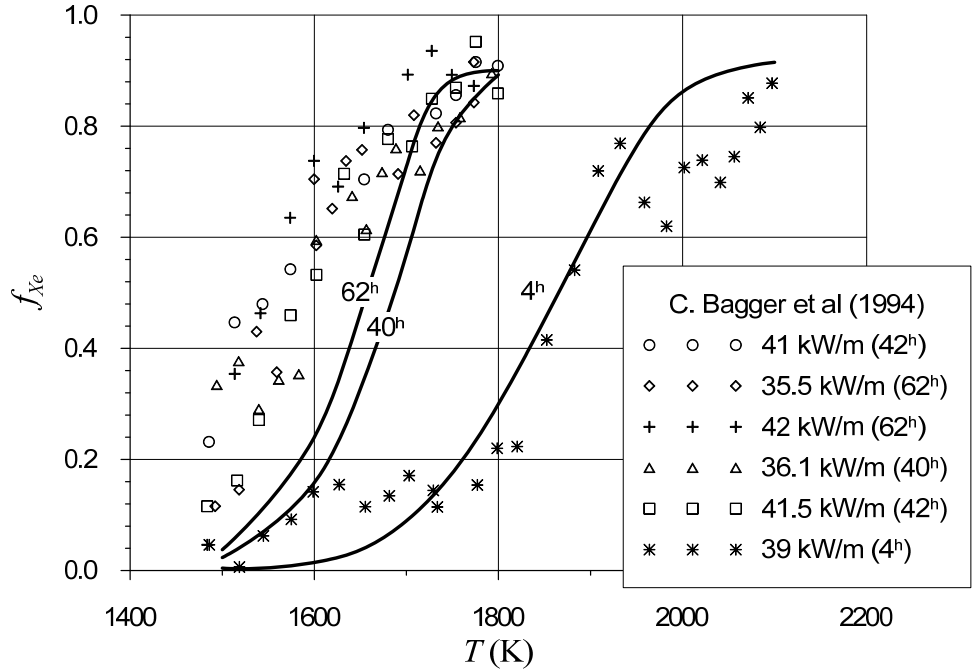
Reactor Tests

Test	Regime	Measured characteristics
SILOE: CONTACT1	Steady state	Stable (Xe) and radioactive FP release
RISØ (Mogensen et al., JNM)	Transient	Xe release, fuel swelling, grain growth

Examples of MFPR Verification

Transient tests RISØ [Mogensen et al., 1994]





Fractional gas release as a function of pellet radial position for hold time of 62 h (different options of the code)

D_b>0 – bubble thermal diffusivity;
V_{gr}>0 – grain growth and sweeping;
V_b>0 – bubble mobility in ∇T

Fractional gas release in ramp tests with different hold times (4, 40 and 62 h)

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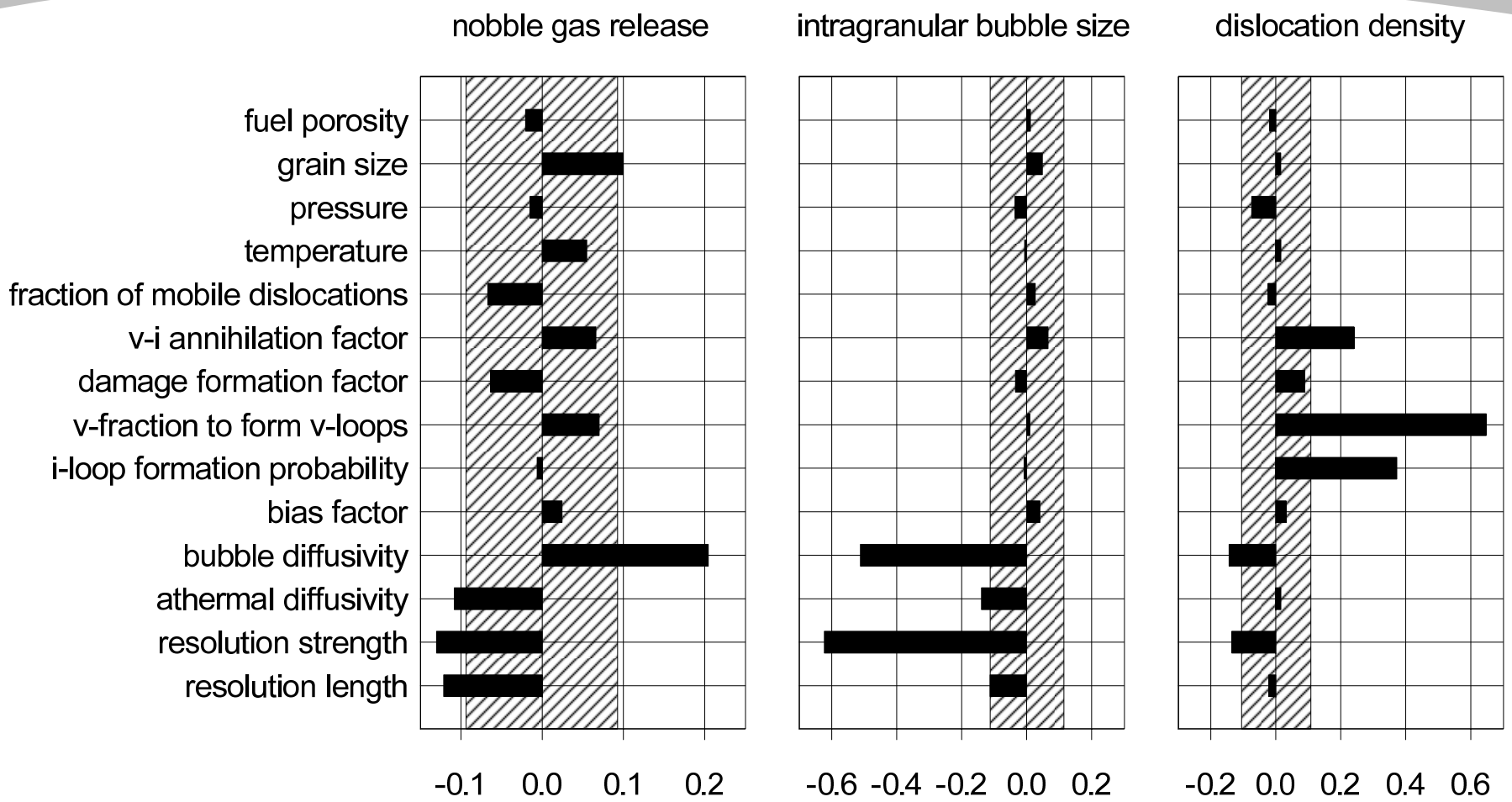
Sensitivity Studies



Sensitivity coefficients for three MFPR responses with respect to the microscopic model parameters (hatched regions - discrimination levels) with modified SUNSET tool (IRSN)

Multi-Scale Approach in MFPR (1/2)

- MFPR code is a meso-scale mechanistic code, which includes an extended set of microscopic parameters, characterising, e.g. the crystal defect structure, and being poorly measured (despite high *sensitivity* of the code predictions to their values)
- Up-to-now this was regarded as a serious disadvantage of the mechanistic approach, often criticized for this reason (see, e.g., Peer Review of the mechanistic approach in VICTORIA in 90-s) and thus retarding further development of mechanistic codes (all over the world)
- VICTORIA was chosen a progenitor code of MFPR (in the mid 90-s), in order to preserve the state-of-the-art attained in this area and to develop further the mechanistic, theory based approach

Multi-Scale Approach in MFPR (2/2)

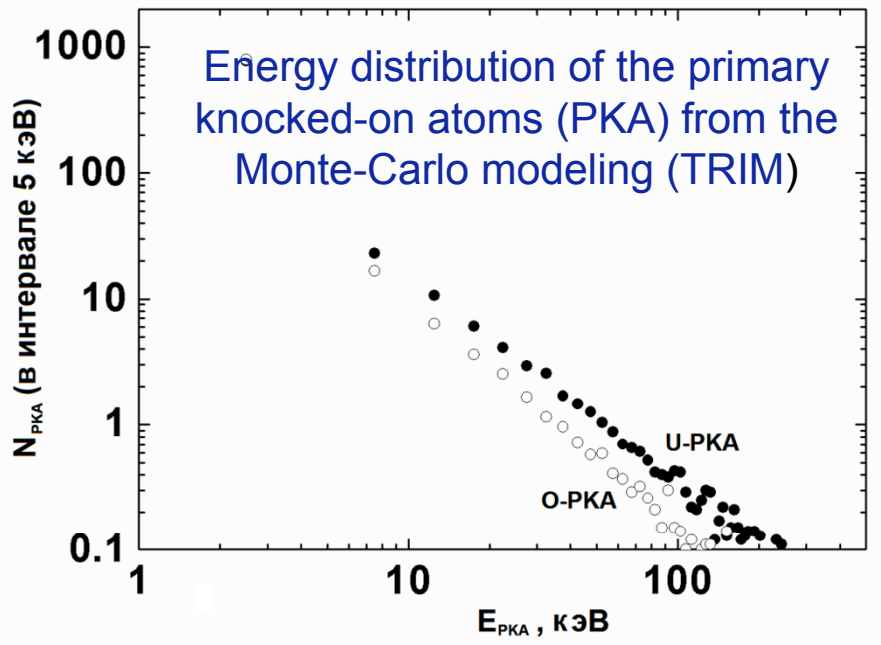
- Up to now, in MFPR new microscopic parameters being *physically grounded* were fixed from analyses of available experimental data (in course of extensive verification of the code) and then used without any artificial tuning (the main advantage of mechanistic approach)
- Recently collaboration with Joint Institute for High Temperatures (JIHT) RAS has been launched for atomic scale determination (DFT, MD, MC) of the *key microscopic parameters for input* in MFPR, realizing the multi-scale approach
- The priority list of MFPR microscopic parameters, which should be evaluated using the atomistic calculation techniques, is formed on the base of the sensitivities studies (presented above)

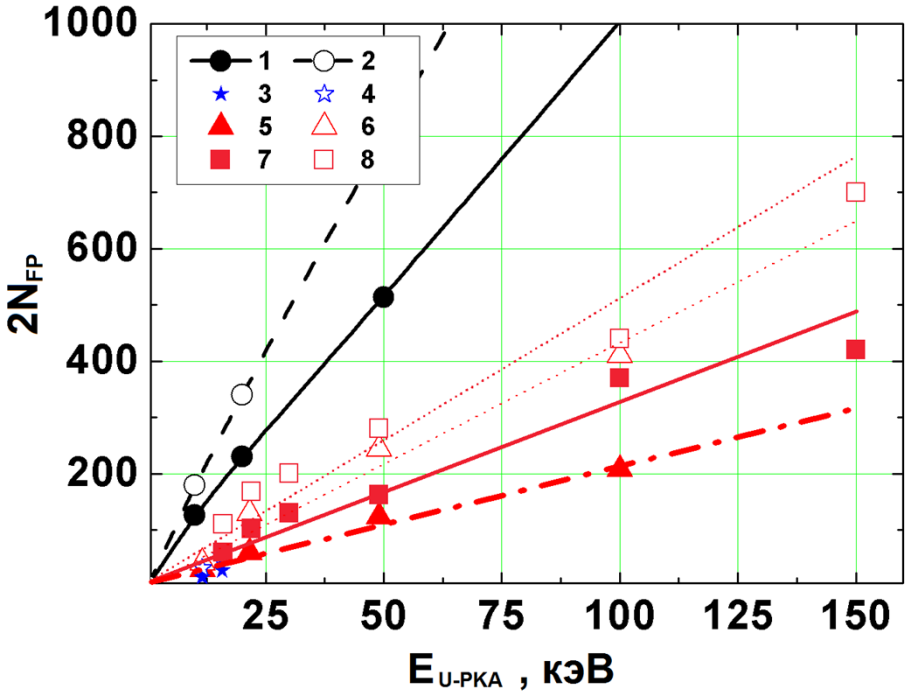
Atomistic Calculations of UO_2

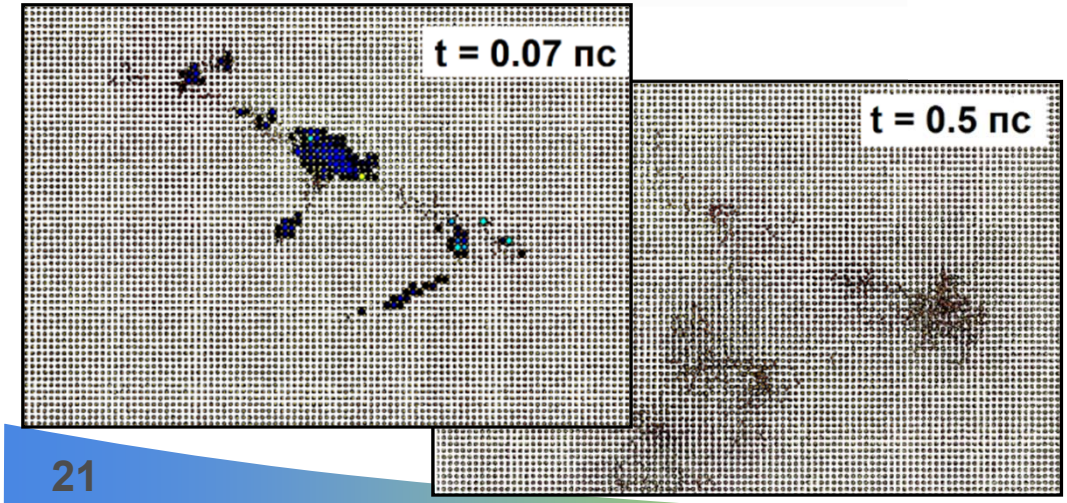
Examples of atomistic calculations

- Monte-Carlo (TRIM)
- Classical Molecular Dynamics (LAMMPS) using various inter-ionic potentials:
  - Morelon et al. (2003)
  - Tiwary *et al.* (2009)
  - Yakub et al. (2007)
  - Potashnikov et al. (2011)
  - Geng et al. (2008) for Xe-U and Xe-O

MC and MD simulations of defect generation in fission tracks



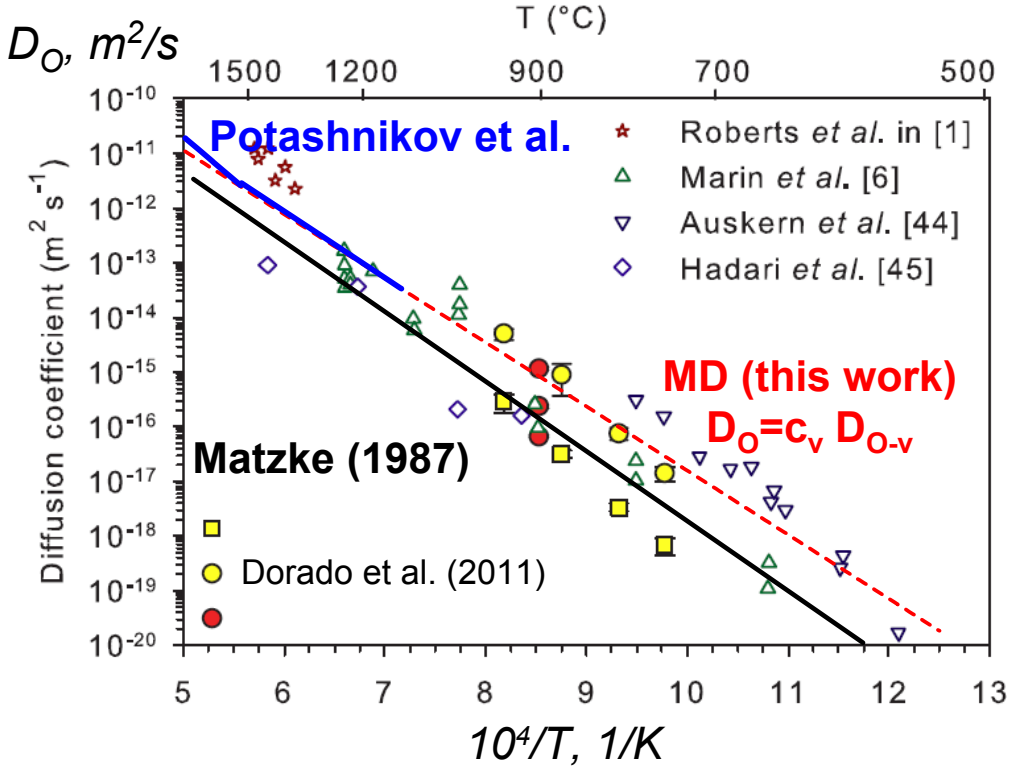




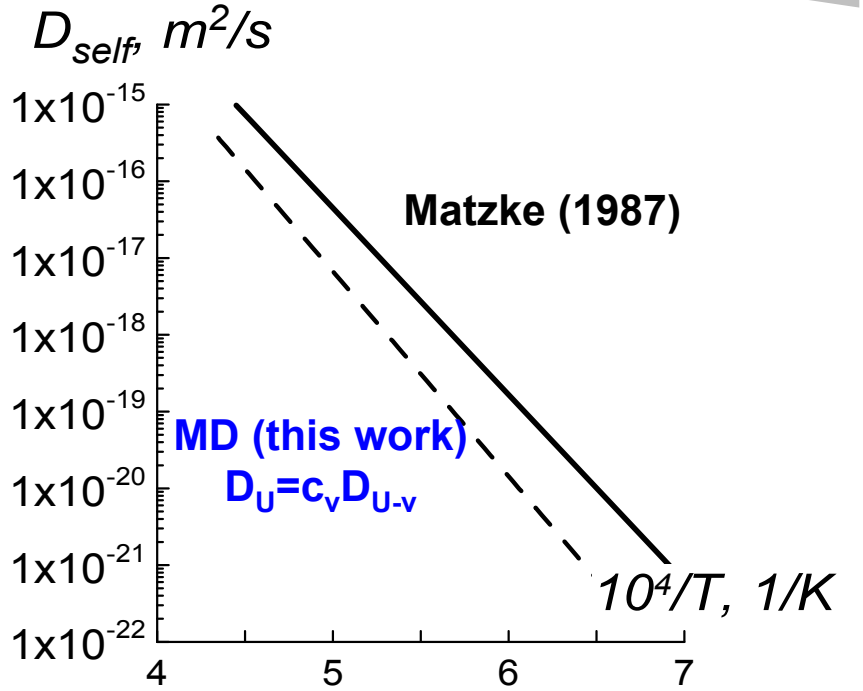
Number of oxygen (open symbols) and uranium (closed symbols) point defects produced in collision cascades initiated by U-PKA (with different interatomic potentials)

Total amount of U Frenkel pairs:
$\approx 10^4$ (MFPR parameter)

MD calculation of O and U diffusivity in UO_2



Calculated O diffusivity
in comparison with experimental
data and other calculations



Calculated U self-diffusivity
in comparison with experimental
data

U Frenkel pairs recombination rate (1/3)

First step: MD calculations (≈10^3 ps)

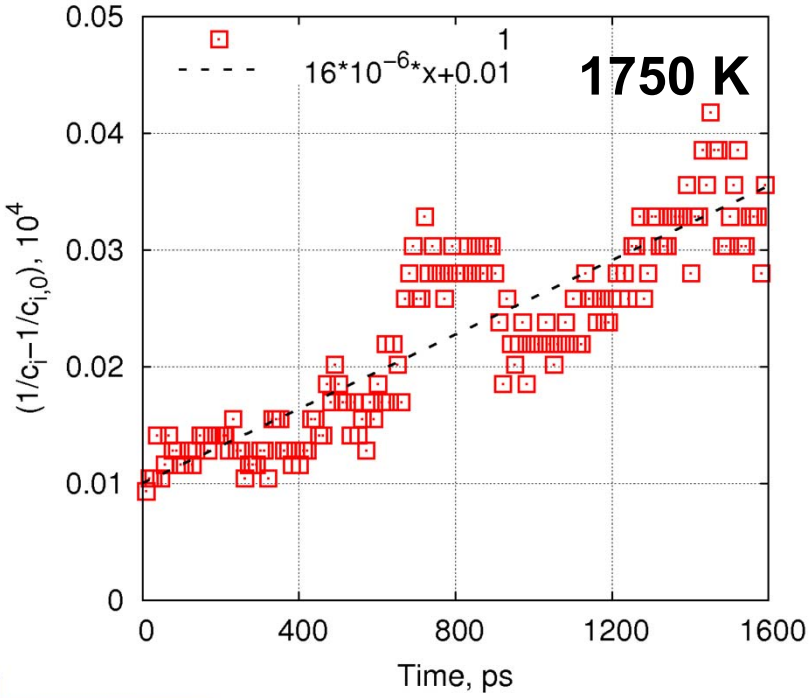
32x32x32 unit cells (393 216 U atoms) C_{FP} = 2x10^{-4}

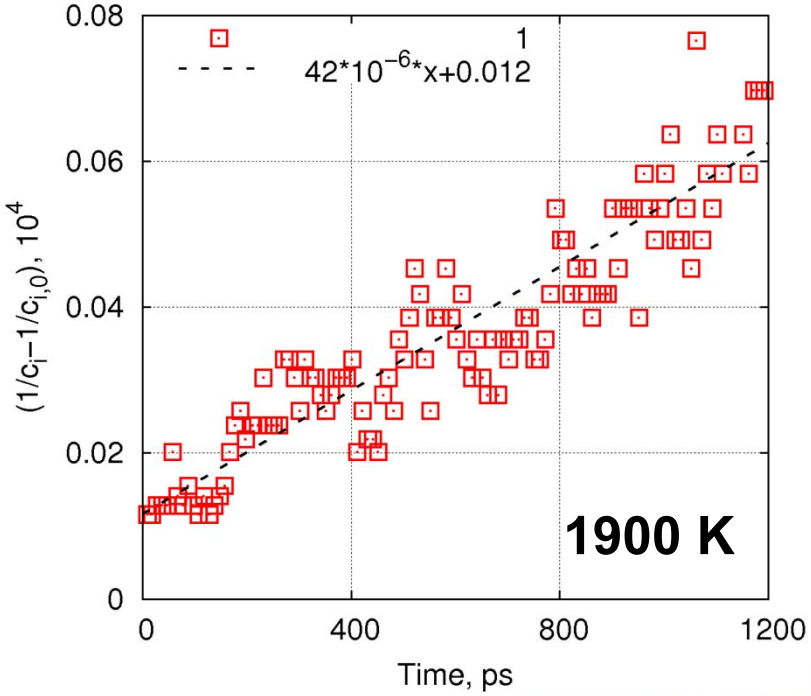
$$\dot{c}_i = -\alpha D_i c_i c_v$$

$$1/c_v = 1/c_{v0} + \alpha_{iv} t$$

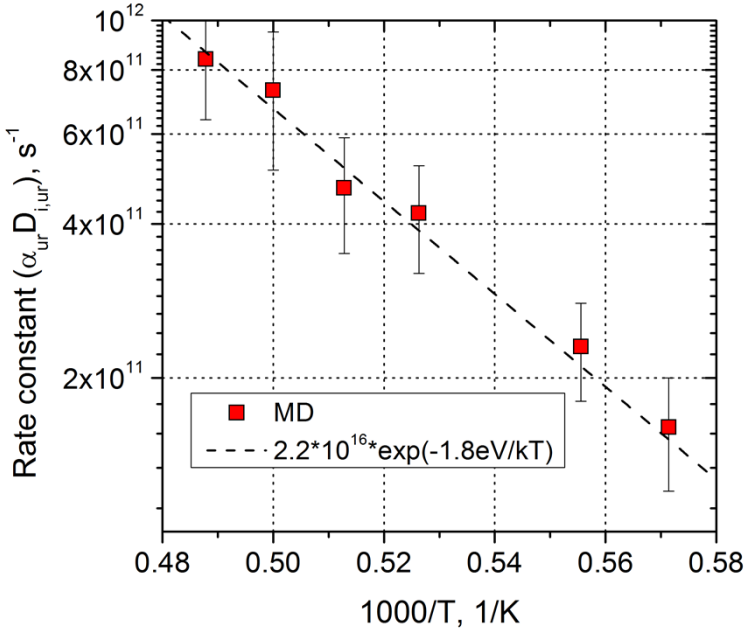
$$\alpha = 4\pi r_c / \Omega$$

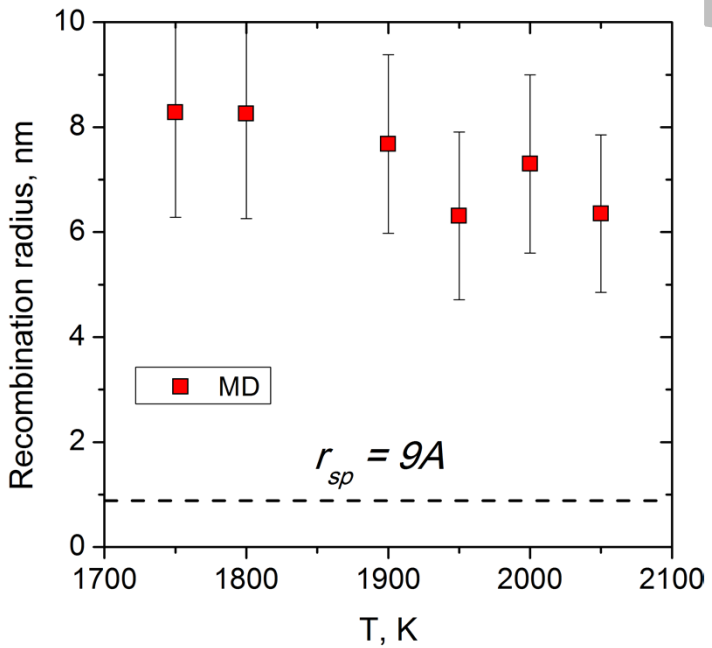
(following Insepov, Rest, Yacout, Kuksin, Norman et al., 2011 for Mo)





U Frenkel pairs recombination rate (2/3)

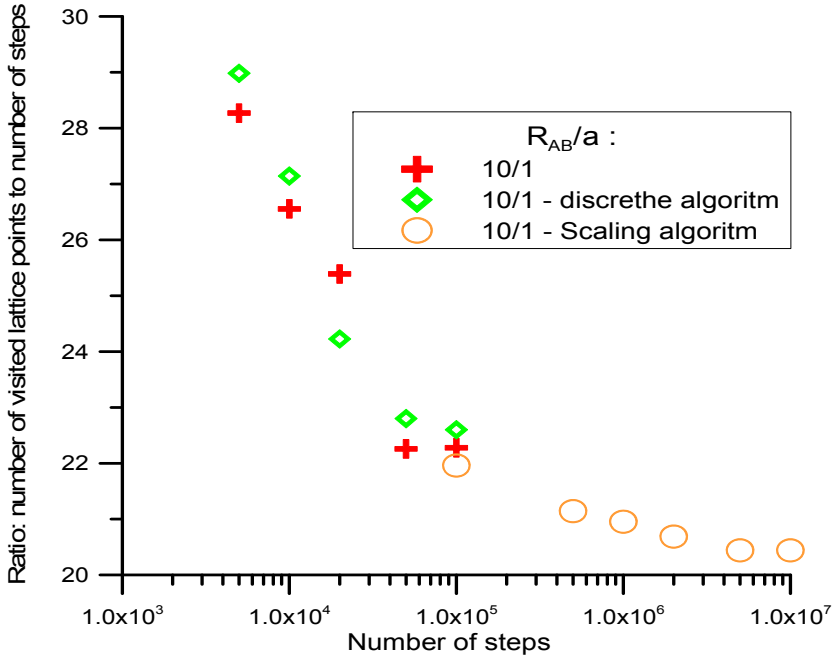




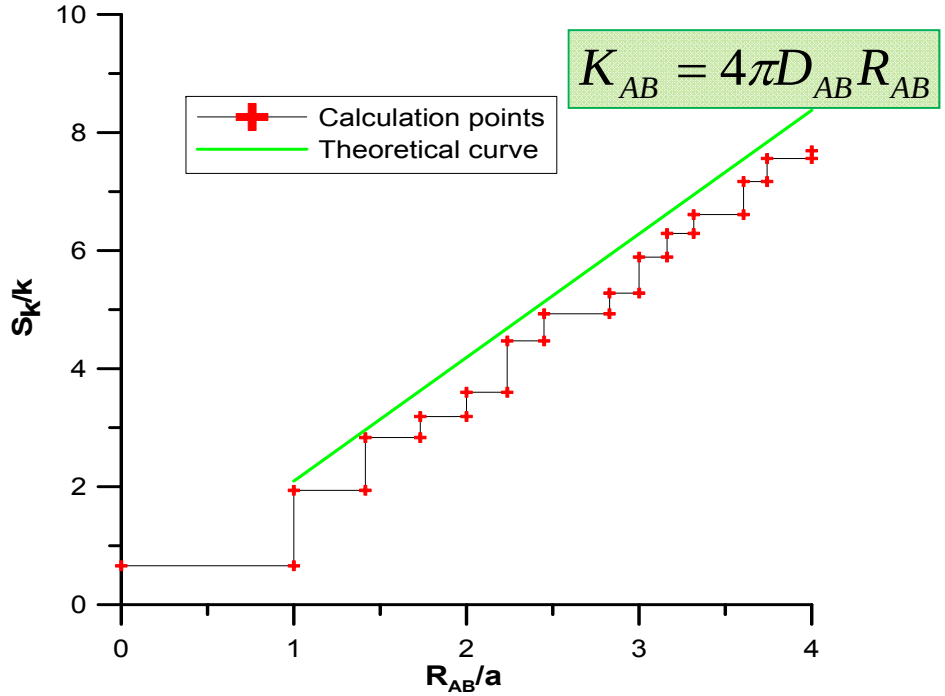
T	D_i	$\tau = a^2/6D_i$	r_c	Number of jumps in MD
1850 K	1.35×10^{-10} m²/c	3.73×10^{-10} s	≈ 6-8 nm	5
2000 K	3.46×10^{-10} m²/c	1.45×10^{-10} s		28
2100 K	6.02×10^{-10} m²/c	8.37×10^{-11} s		48

U Frenkel pairs recombination rate (3/3)

Second step: kinetic calculations (stand-alone module for point defects migrating on discrete cubic lattice)



Attainment of steady state collision rate ($\approx 5 \cdot 10^6$ jumps)



Modification of Smolukhowski collision kernel for discrete lattice

Result:

if $r_c \approx 7$nm after 50 jumps (MD) $\Rightarrow$ $r_c \approx 2.5$ nm after $\approx 5 \cdot 10^6$ jumps

SFPR Code: Development Plan

- The first version of the fuel performance code SFPR, designed by coupling of MFPR with the mechanistic thermo-mechanical module SVECHA, was released and tested in 2008
- Validation (by IBRAE) and commercial applications to operation conditions of the new **VVER-2006** reactor design (by ATOMENERGOPROECT) started in 2009
- Implementation of SFPR module in the advanced version of the Russian integral best-estimate safety code **SOCRAT** (for LWRs), started in 2010 (Russian collaboration under IBRAE coordination)
- Extension to the Fast Reactor (FR) fuel and implementation in the new generation integral code **EUCLID** (developed under IBRAE coordination) for FR design and safety justification, within the new **Russian Federal Target Program** "Nuclear Power Technologies of a New Generation for the years 2010-2015 with a perspective to 2020"