



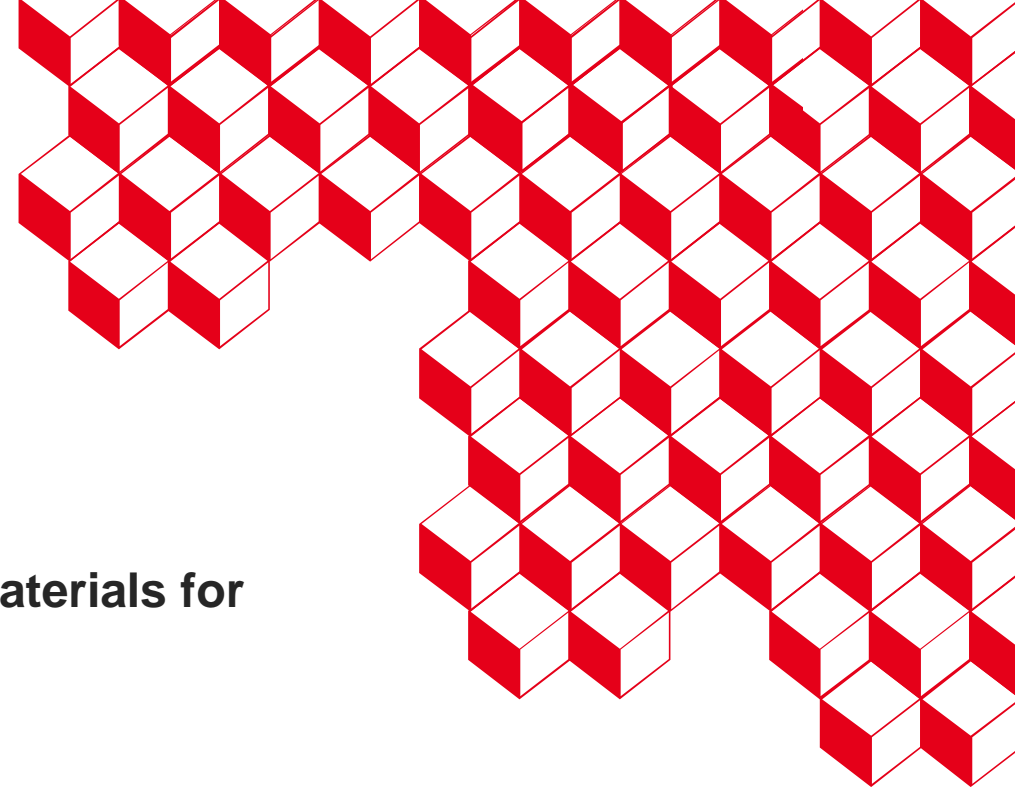
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**Technical Meeting on Compatibility Between Coolants and Materials for
Fusion Facilities and Advanced Fission reactors**

30th October – 3rd November 2023

Vienna, Austria



Improving the prediction of activity transfer in nuclear water circuits by integrating colloidal behaviour : experimental and modelling advances for the OSCAR code

C. Cherpin, F. Dacquait

Outline



1. Uniform corrosion and colloids



2. High temperature zeta potential determination



3. Modelling particles deposition



4. Simulation improvements



5. Conclusions et perspectives

1. Uniform corrosion and colloids

Activated corrosion products : 85 % of the collective dose (PWRs)

① Uniform corrosion

- Release of ionic metallic species and formation of double oxide structure (SS and Ni alloys)
- Particulate corrosion products in coolant (erosion of deposit / volume precipitation)

② Activation of corrosion products (**deposition under neutron flux**)

- ^{58}Co and ^{60}Co responsible for 90 % of the dose rates around the RCS of PWRs
- Transport of ionic and particulate ACPs toward out-of-flux regions

③ Activity build-up

- Volume contamination
- Surface contamination

Numerous stakes

- Radioprotection of nuclear workers
- Effluent management and dismantlement
- Availability of nuclear fleet
- Safety (source term)

1. Uniform corrosion and colloids

Important to understand and predict activity transfer by corrosion products

Conception
(i.e. choice of materials)

Operation
(i.e. chemistry optimization)

OSCAR code : TOol for Simulating ContAmination in Reactors

Paper by
F. Dacquait

- Spatio-temporal distribution of ACPs in water cooled circuits
- Unique prediction tool:
 - Wide range of conditions: 20 °C to 350 °C, reducing/oxidizing, acid/alkaline
 - More than 430 γ spectrometry measurements in 76 different PWRs

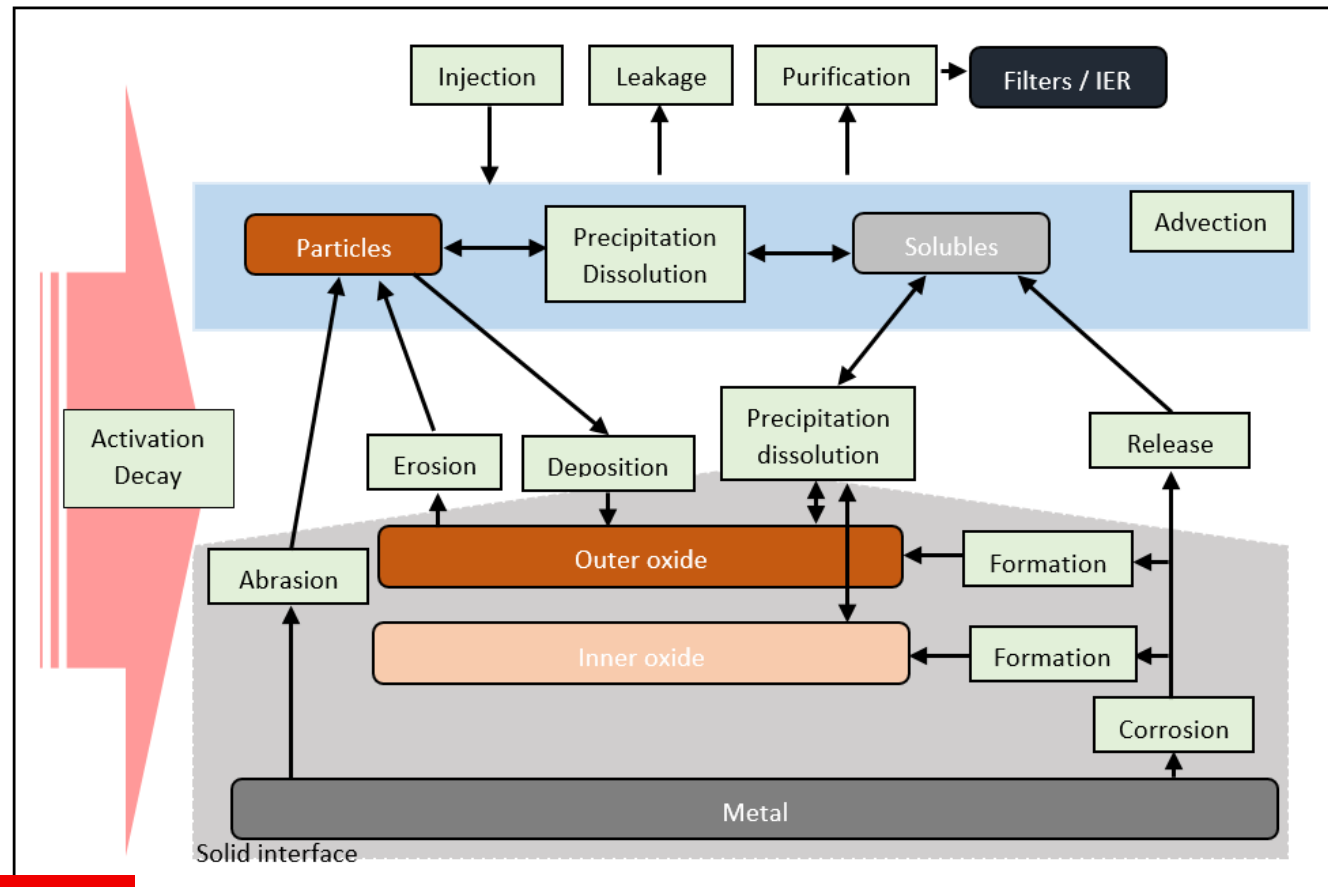
Global validation of the OSCAR code in PWRs conditions

1. Uniform corrosion and colloids

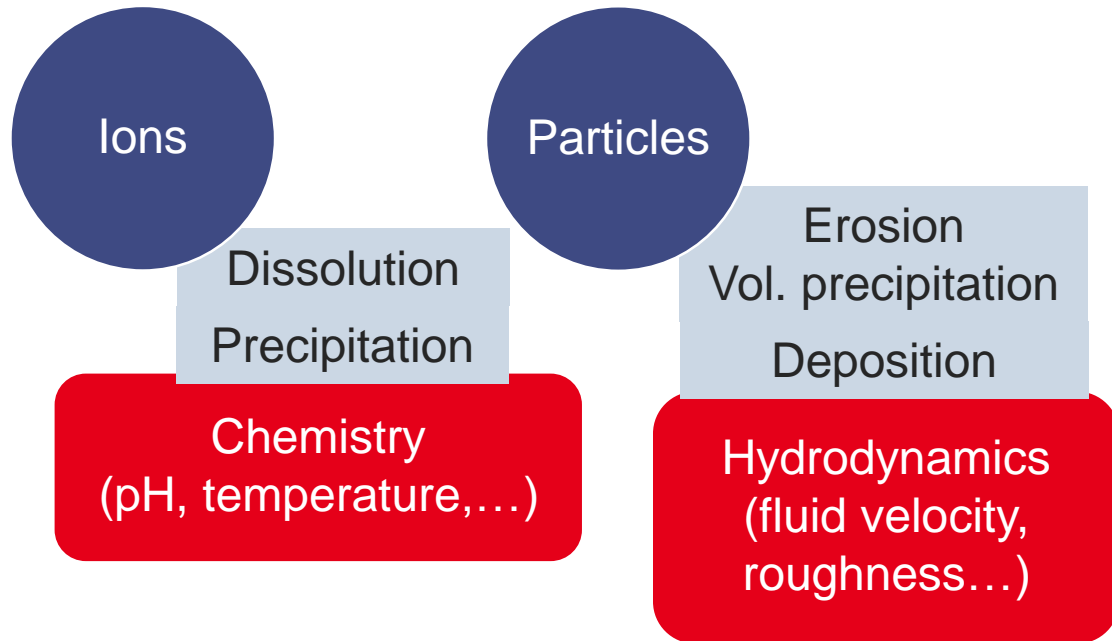


Description of interactions is based on physical models → phenomenological approach

Models developed for **PWRs primary circuit conditions** are applicable to **Primary Heat Transport Systems of ITER / DEMO conditions**



1. Uniform corrosion and colloids



OPEX from PWRs

→ Majority of volume activity transported as particles (from $< 1 \mu\text{m}$ up to several μm)

COLLOIDS

INERTIAL PARTICLES

→ Particle size distribution studies highly dependant on the PWR unit

→ Varying proportion $< 0.45 \mu\text{m}$

→ $^{110\text{m}}\text{Ag}$ is colloidal in reducing conditions

1. Coolant – material interactions

Litterature – Origin of colloids

[Matijevic, 1986]

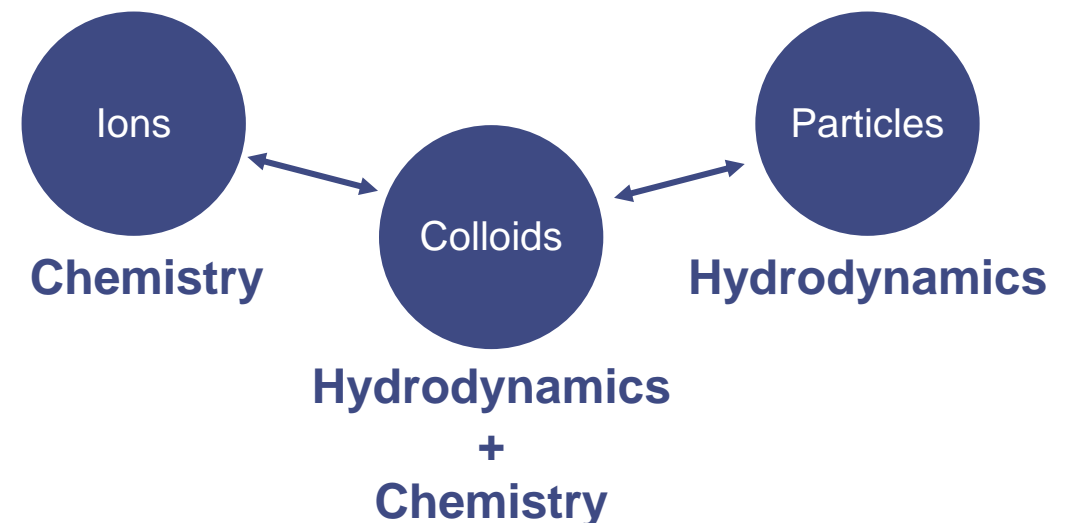
« Corrosion products [...] may appear as thin or thick films or as fine or coarse particle. Thus, **these products are either in colloidal state or must have existed in such state for shorter or longer periods of time** before they grew into coarse systems »

[Berger, 2015]

Dissolution of silver metal → production of Ag^+ → reduction by solvated electrons from water radiolysis
→ silver metal colloids produced

Litterature – Behavioral specificities of colloids

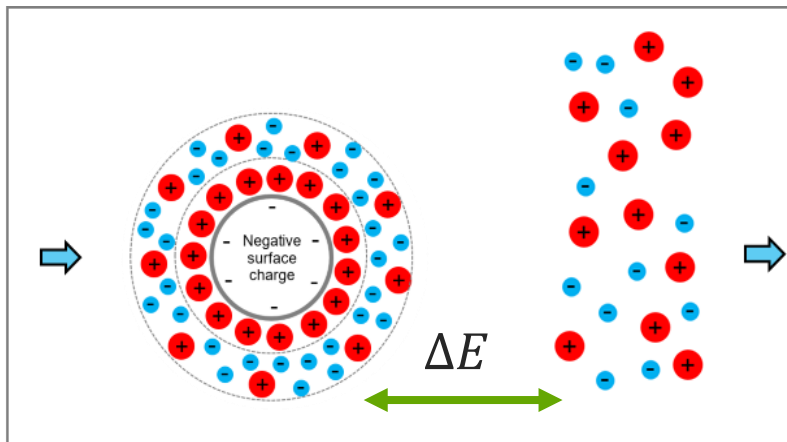
- Surface forces \gg Volume forces
 - Zeta potential: potential at the shear plane of the particle → Key parameter
- Depends on pH, temperature, ions in the solution...



2. High temperature zeta potential ζ determination

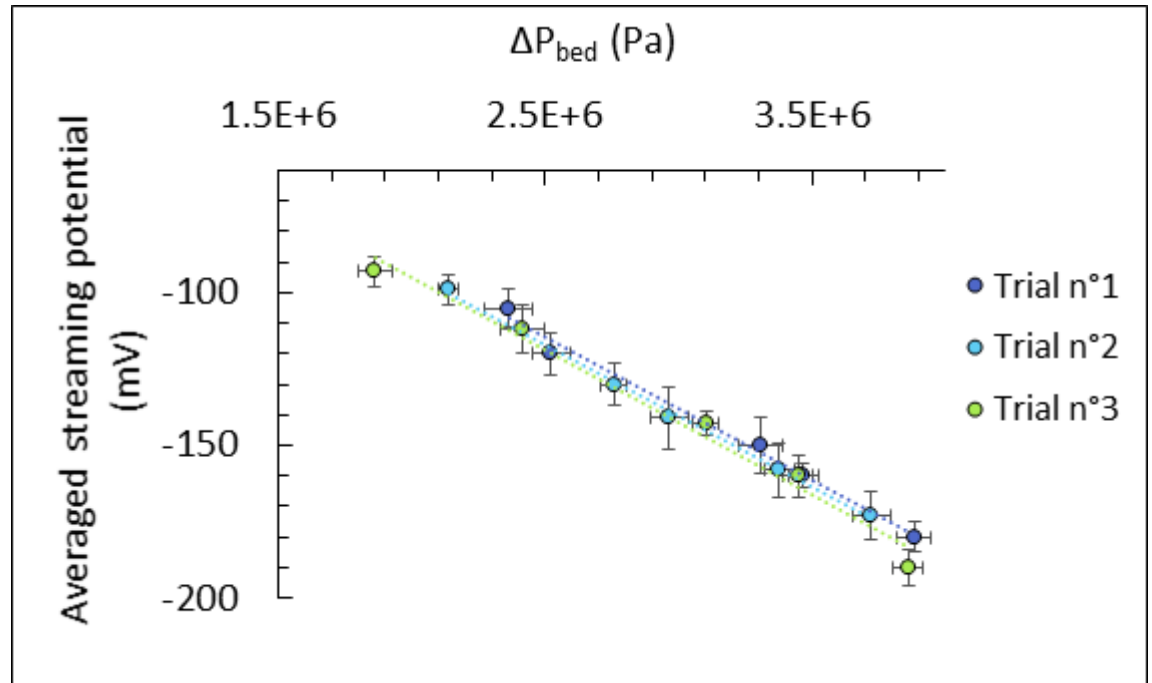
- Experimental difficulties related to operating at high temperatures

Streaming potential method



$$\frac{\Delta E}{\Delta P} = \frac{\varepsilon_0 \varepsilon_r R r^2}{4 \mu L} \zeta$$

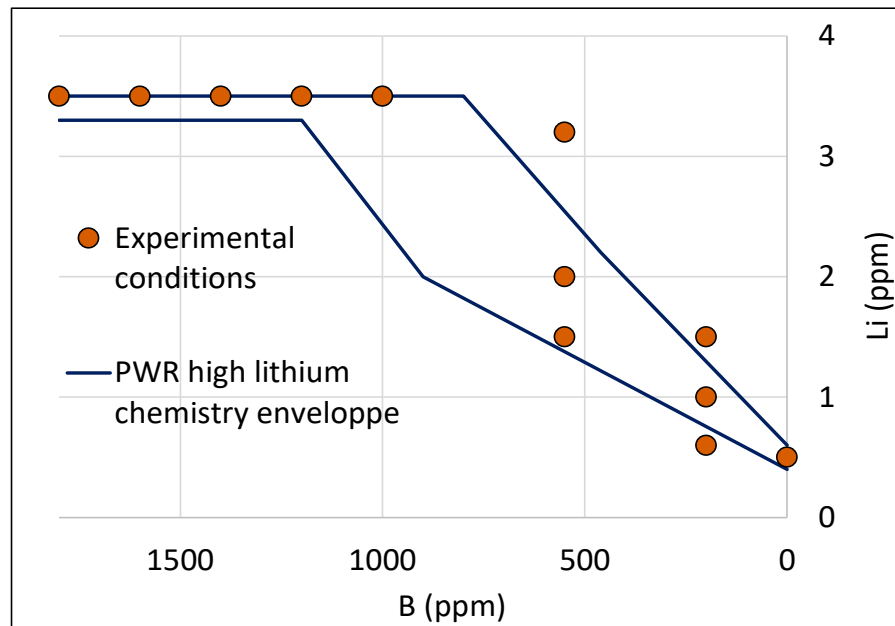
ΔE streaming potential, ΔP pressure drop, ε_0 permittivity of free space, ε_r relative permittivity of the solution, R electrical resistance of column, r column radius, μ dynamic viscosity, L length of column



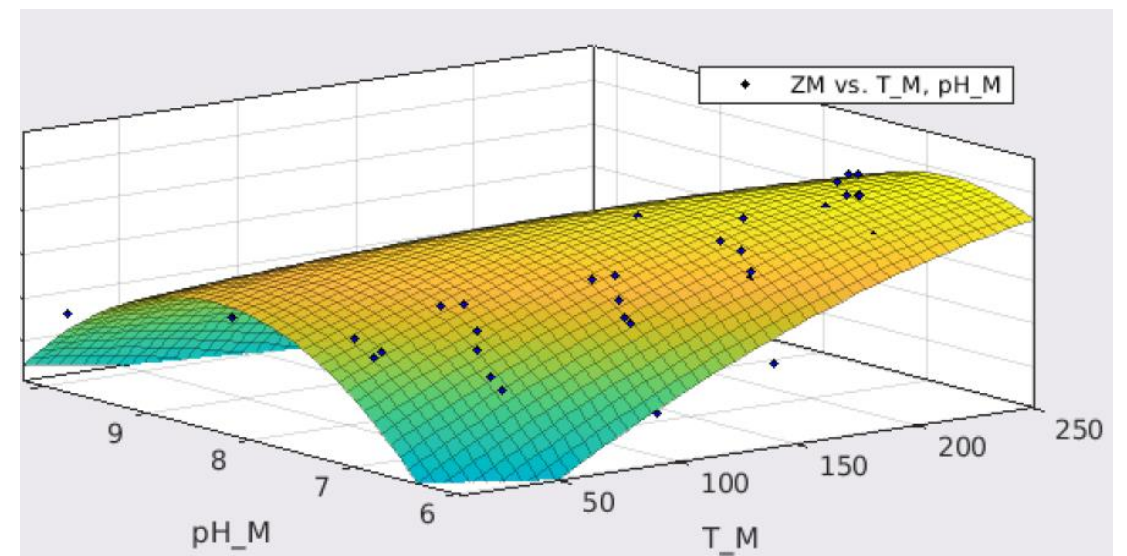
2. High temperature zeta potential ζ determination

1st experimental campaign : 2020

- ζ values of Fe_3O_4 and NiFe_2O_4 up to 240 °C for 12 primary system chemistries
- Extrapolation of ζ values of Fe_3O_4 and NiFe_2O_4 to primary conditions



$$\zeta = p_{00} + p_{10}T + p_{01}pH + p_{20}T^2 + p_{11}TpH + p_{02}pH^2$$



ζ extrapolation for Fe_3O_4



2. High temperature zeta potential ζ determination

2nd experimental campaign : ongoing (CEA)

Objectives :

- $T \in [20,340] \text{ } ^\circ\text{C}$
- Phases : chromites, ferrites
- Chemistry changes

→ Knowledge of ζ is essential to model surface interactions

3. Modelling particles deposition

Modelling deposition considering surface interactions

- DLVO theory : Prediction of the variation of the interaction energy between a particle and a surface

$$\phi_T = \phi_A + \phi_R$$

London Van der Waals interaction

Born interaction

Electrical double layer interaction (ζ)

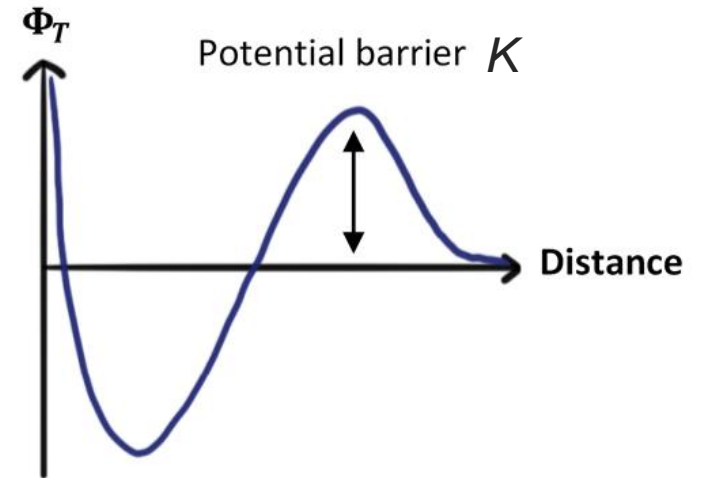
Attachment probability

- Urrutia - condition for deposition: $K \leq \frac{1}{2} m_p v_p^2$

- Rudolph

- definition of energy ratio $P_S = \frac{\frac{1}{2} m_p v_p^2}{K}$

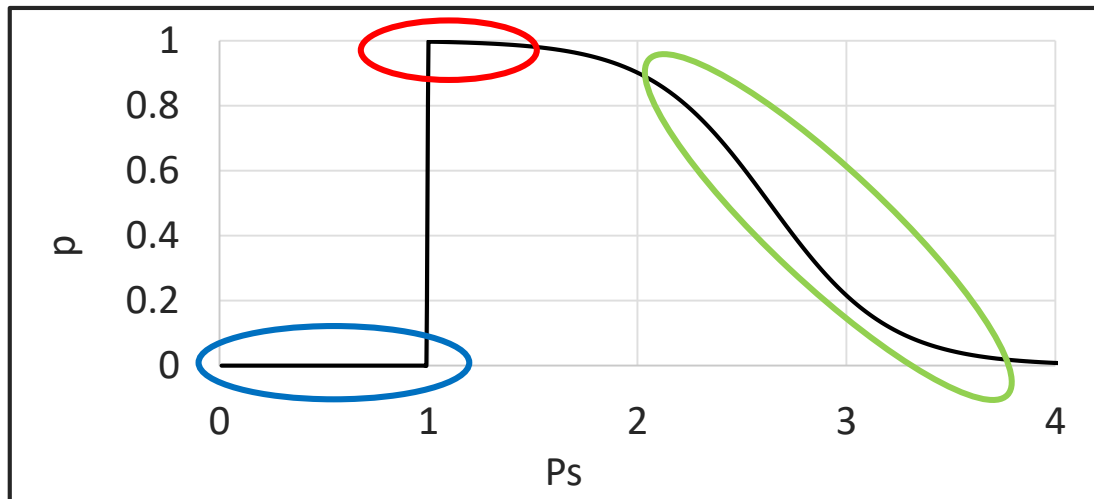
- definition of an attachment probability $p = \frac{1}{1 + A e^{B P_S}}$



3. Modelling particles deposition

Attachment probability

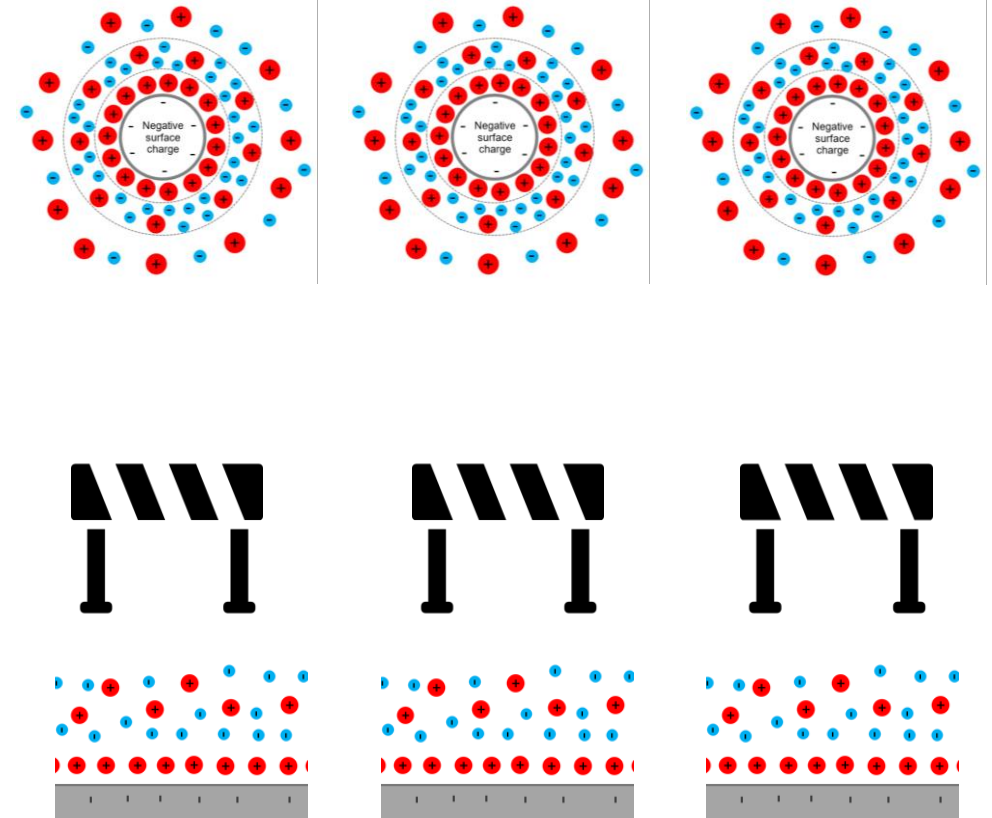
$$\text{attachment probability } p = \frac{1}{1 + Ae^{BPs}}$$



Case 1

Case 2

Case 3

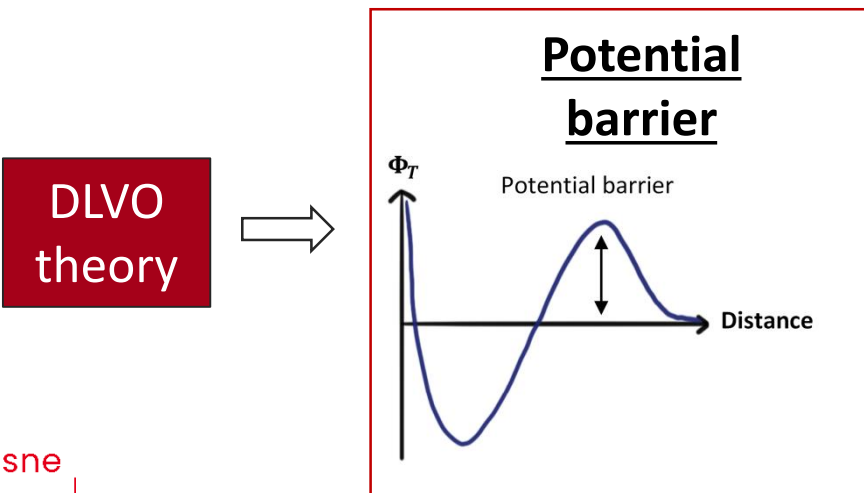


3. Modelling particles deposition

Deposition velocity

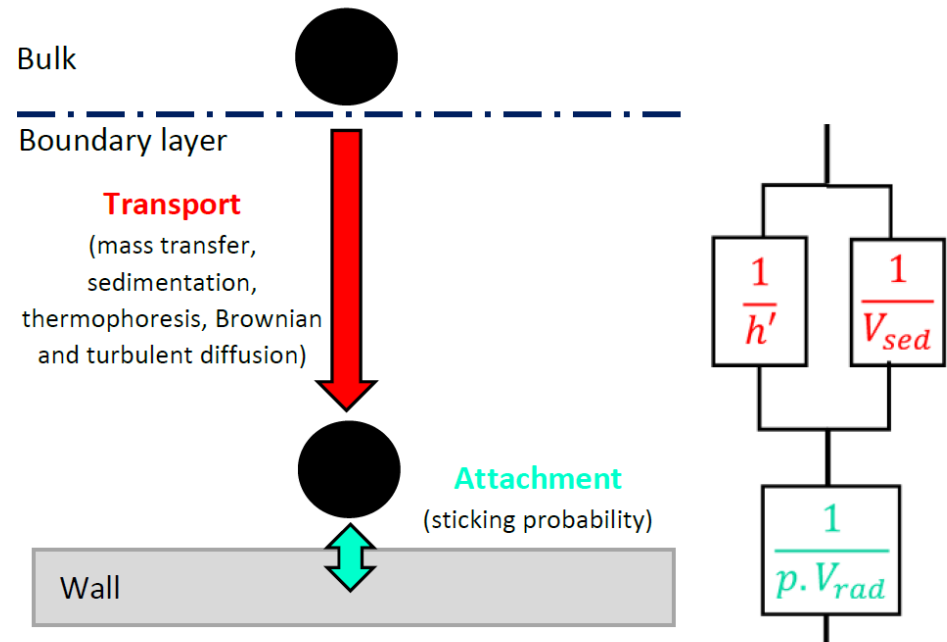
$$\frac{1}{V_{dep}} = \frac{1}{h'} + \frac{1}{V_{sed}} + \frac{1}{p \cdot V_{rad}}$$

attachment probability (surface effects)
 radial velocity of particles
 sedimentation velocity
 Mass transfer coefficient



Kinetic energy of particle

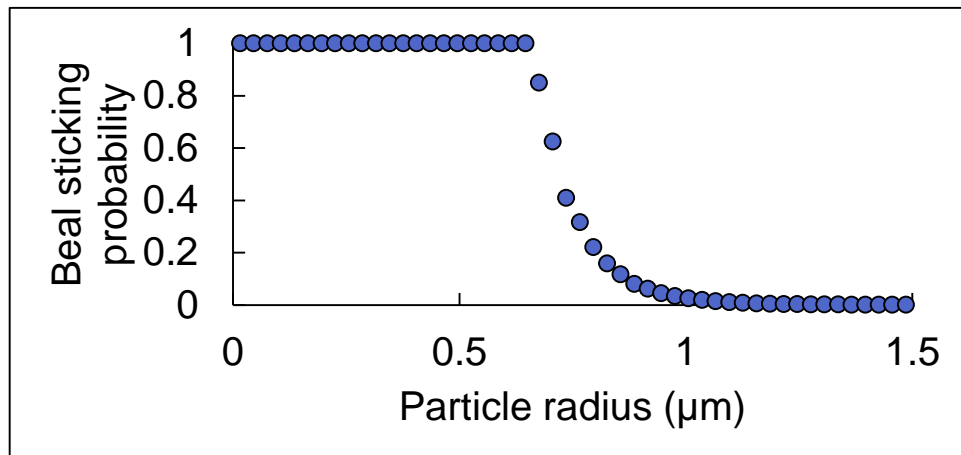
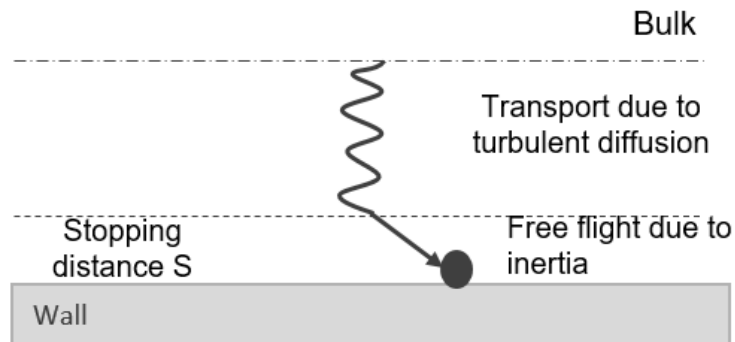
→ P_s → $p = \frac{1}{1 + Ae^{BP_s}}$



3. Modelling particle deposition

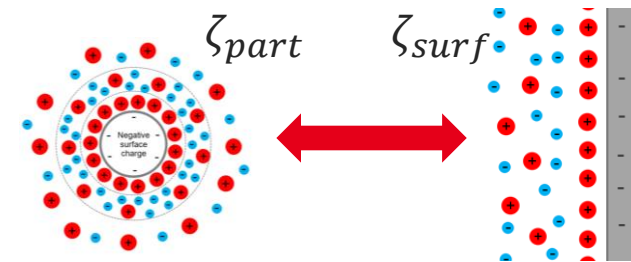
Previous deposition model
OSCAR v1.4.a

- Macroscopic approach [Beal, 1978]

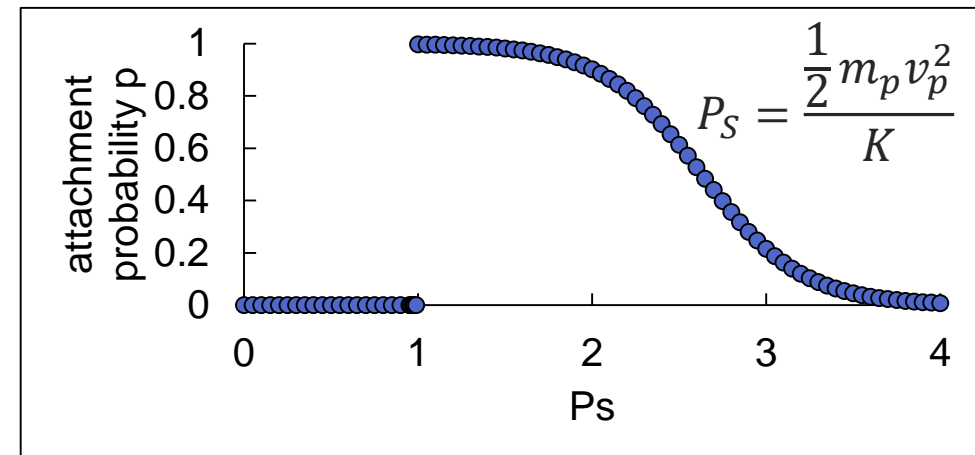


New deposition model
OSCAR v2.0

- DLVO theory → surface interactions



If potential barrier exists :



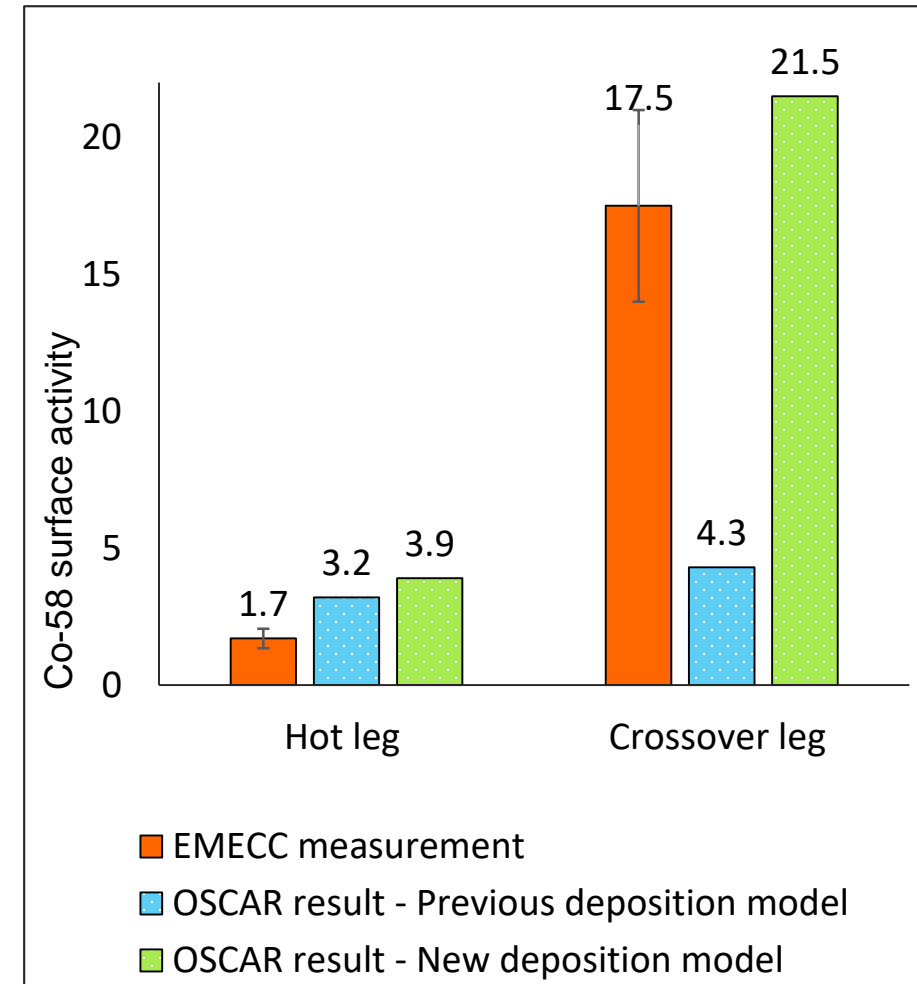
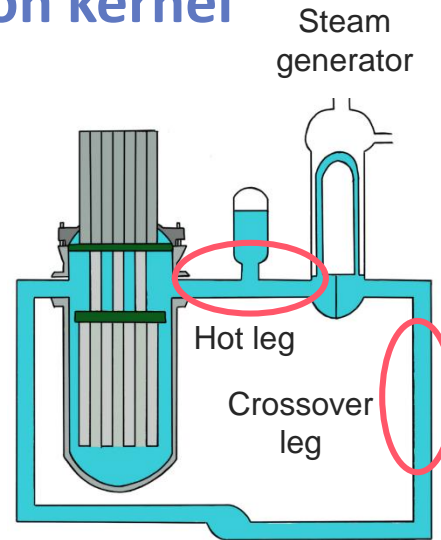
Else : attachment probability = 1

4. Simulation improvements

Implementation in OSCAR calculation kernel

Simulation of a specific case

- 1300 MWe PWR with steam generator replacement (SGR)
- EMECC measurements during shutdown at cycle SGR + 1
- ^{58}Co preferential contamination in the crossover leg
- Simulation results:
 - Previous deposition model (without surface interactions)
 - New deposition model (with surface interactions, no fitted parameter)



→ **Taking into account surface interactions leads to significant improvements of the description obtained using OSCAR**



Conclusion and perspectives

- Colloidal species are essential to comprehensively describe activity transfer in nuclear systems
- Main challenge: ζ values in the conditions studied → ongoing work

Perspectives for fusion reactors:

- ζ values of oxides formed on RAFM steels (Eurofer, F82H...) and Cu-base alloys in PHTSs conditions
- Particle size distribution measurements in various PHTSs
- Coupled effect of chemistry and sub-cooled nucleate boiling on colloidal behavior
→ implementation in OSCAR-Fusion



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Thank you for your attention

Acknowledgements

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