



CONSEIL SCIENTIFIQUE

Plenary meeting IN2P3 scientific council

- *Theoretical chemistry* -

Yves Pipon, MCF

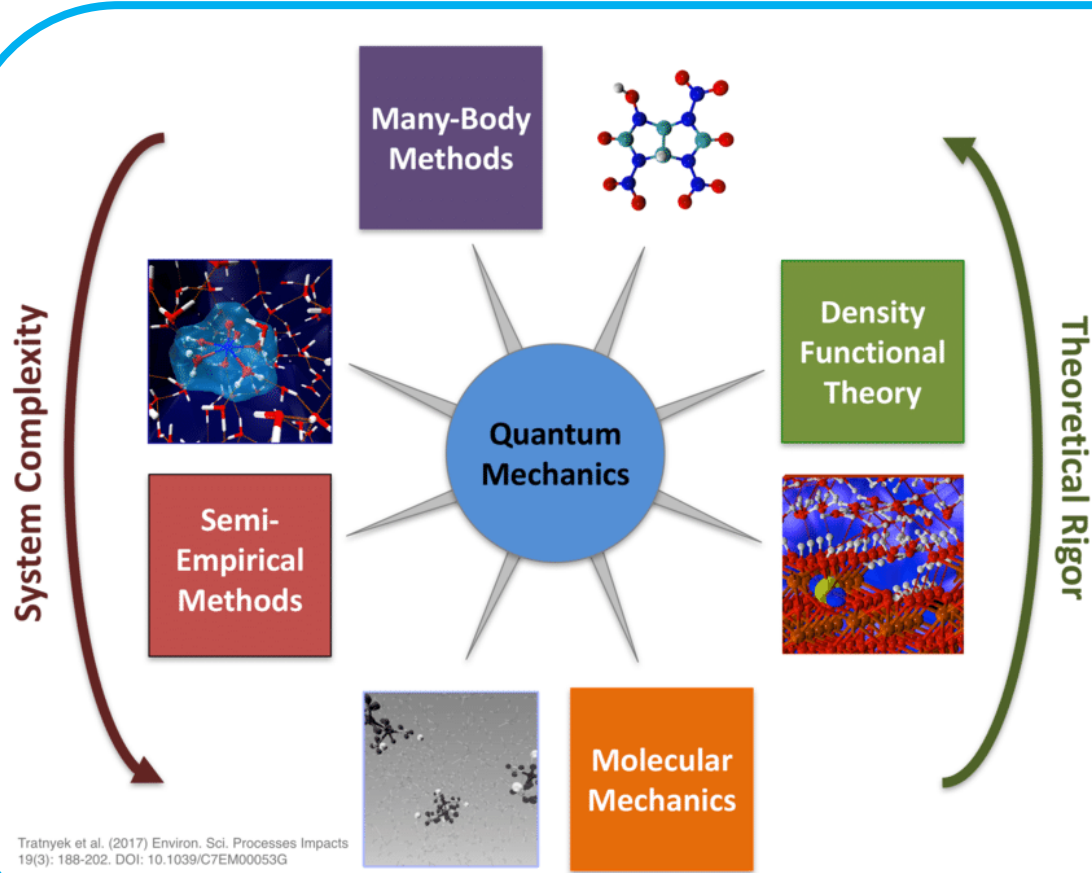
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What is « theoretical chemistry » ?

« All theoretical chemistry is really physics ; and all theoretical chemists know it ! »

- Richard P. Feynman -



Tratnyek et al. (2017) Environ. Sci. Processes Impacts
19(3): 188-202. DOI: 10.1039/C7EM00053G

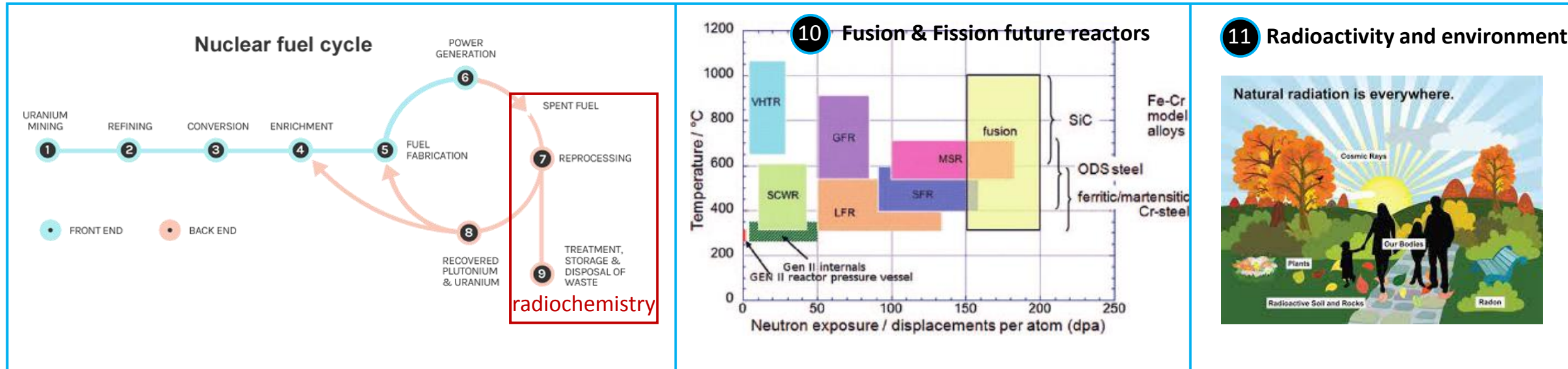
In silico methods to predict material properties:

- *Ab initio* (many body methods as CC, MBPT) → 100 atoms,
- Density Functional Theory (DFT) → 1,000 atoms,
- Semi-empirical methods → 10^3 - 10^5 atoms,
- Molecular mechanics (empirical interatomic potentials, force fields, ...) → $>10^6$ atoms

Development of **multiscale** methods in order to be closer to realistic conditions:

- Bottom-up approach (DFT → Monte Carlo methods) to access larger physical times,
- Concurrent multiscale scheme (mixed QM/MM approach) to access larger length scales,
- ...

Interdisciplinary research related to ionizing radiation in the fields of nuclear energy and environment.



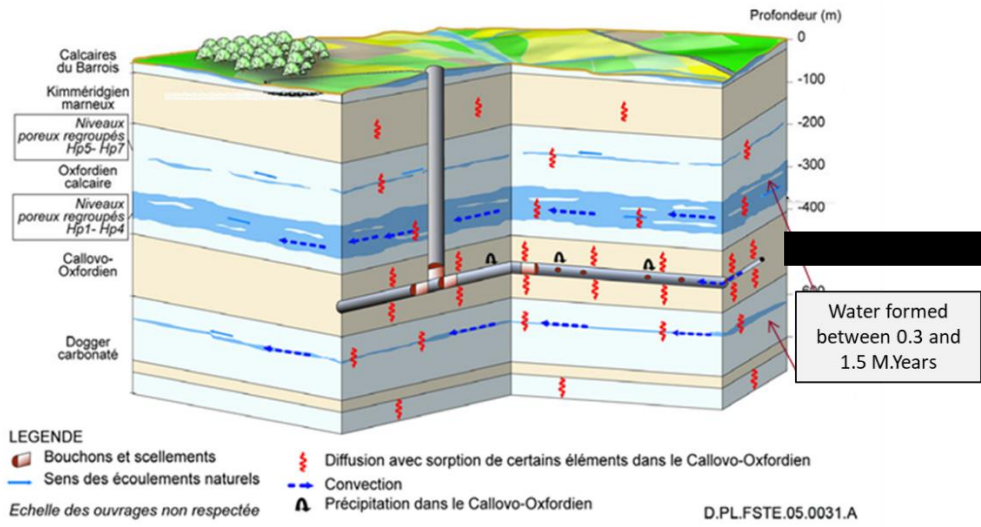
4 main activities:

- 1) Study of interactions between RNs or fluids with solid interfaces (cement & clay systems)
 → 9 & 11 // **DFT, MM**
- 2) Study of RN interactions in solution (basic science, PUREX process, ...)
 → 7, 9 & 11 // **ab initio, DFT, MM**
- 3) & 4) Study of nuclear solids (defect formation, diffusion of FP or RN or He, autodiffusion, ...)
 → 6 & 10 // **DFT, semi-empirical potentials, kMC, (DDD, phase field methods, ...)**

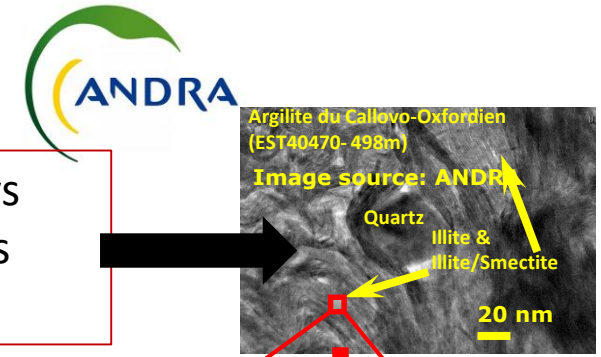
Clay/cement system interfaces with fluids

(A.G. Kalinichev)

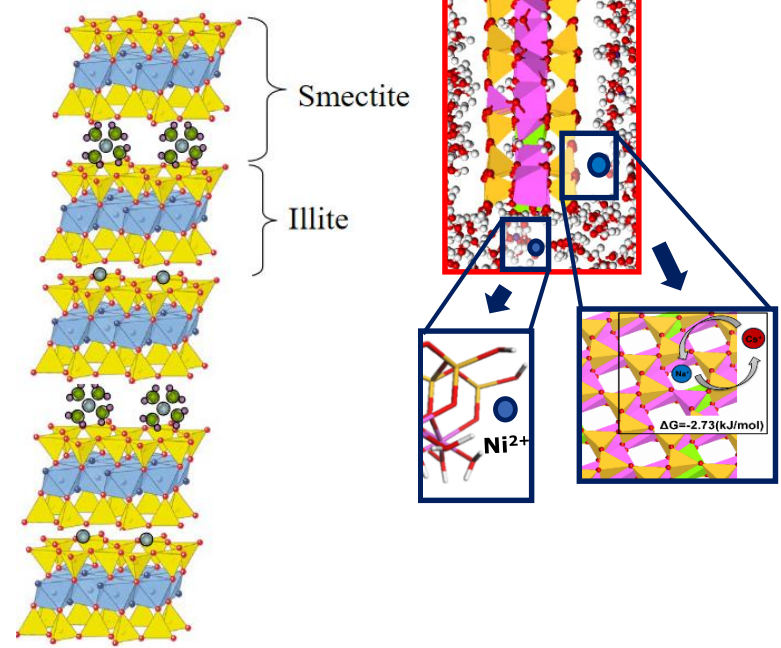
Research & Teaching industrial chair @ IMT Atlantique: "Storage and Disposal of Radioactive Waste"



Callovo-Oxfordian (Cox) clays as a rock host for HA wastes (CIGEO scenario)



- ✓ Development of realistic models of Illite / Smectite interbedded clays as a most accurate model of COx .
- ✓ Quantification of the effects of specific surface sites on the adsorption and transport of ions in clays and other disordered materials



Multiscale modeling of polonium chemistry in sol.

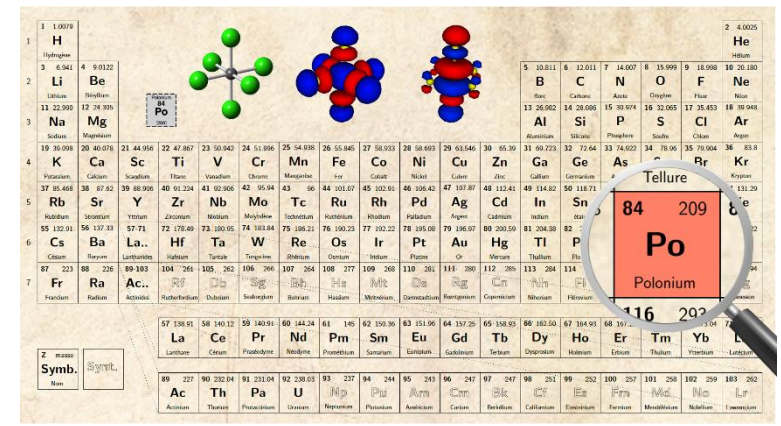
(R. Maurice)



via MITI (80|Prime) with INP (PhLAM, Lille) & INC (CEISAM, Nantes) (2020)

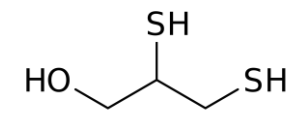
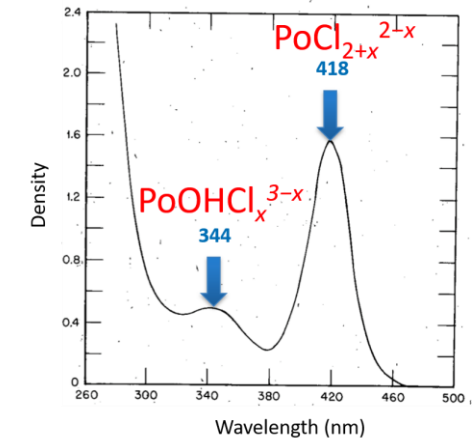


relativity & correlation + force-field development + conformational analysis



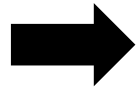
= complete *ab initio* approach to answer grand questions by theory itself & theory/expt. comparisons:

- What is the actual speciation of polonium in solution, beyond past speculations?
- Which conditions lead to single molecular species vs. mixtures of species?
- Can we interpret old and soon to be acquired spectroscopic data?
- Would there be a better antidote than the one for arsenic (BAL)?
- *etc.*



Mo nanoparticle irradiation (Y. Pison)

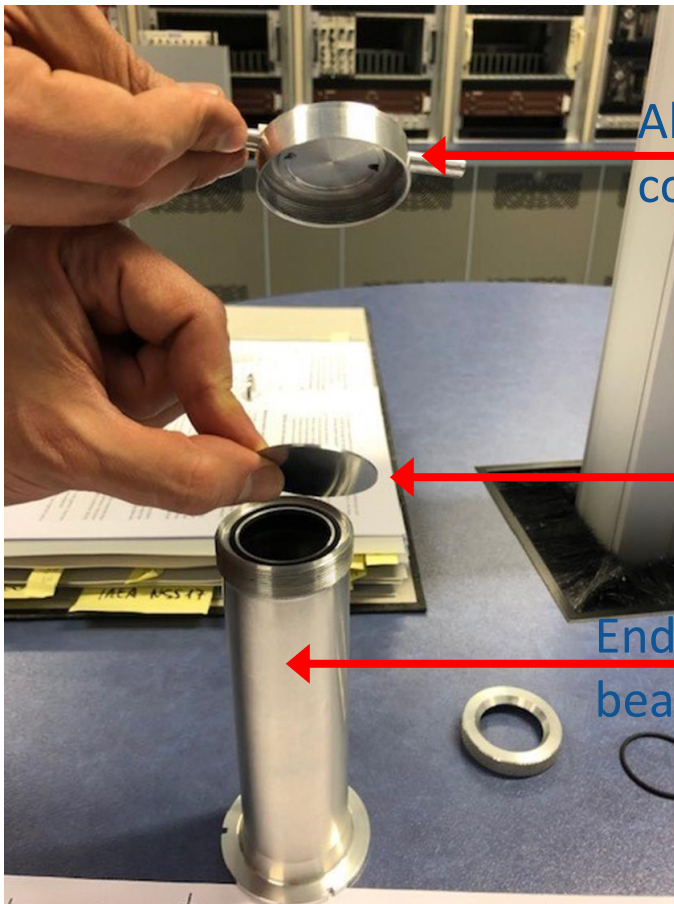
Context: new elaboration route to produce ^{99}Mo / $^{99\text{m}}\text{Tc}$ radiotracer via $^{100}\text{Mo}(n,2n)^{99}\text{Mo}$ or $^{100}\text{Mo}(\gamma, n)^{99}\text{Mo}$



Collaboration with



- to understand the irradiation of a Mo NP and,
- to guide the experimental setup → NP size? Nuclear reaction? ...

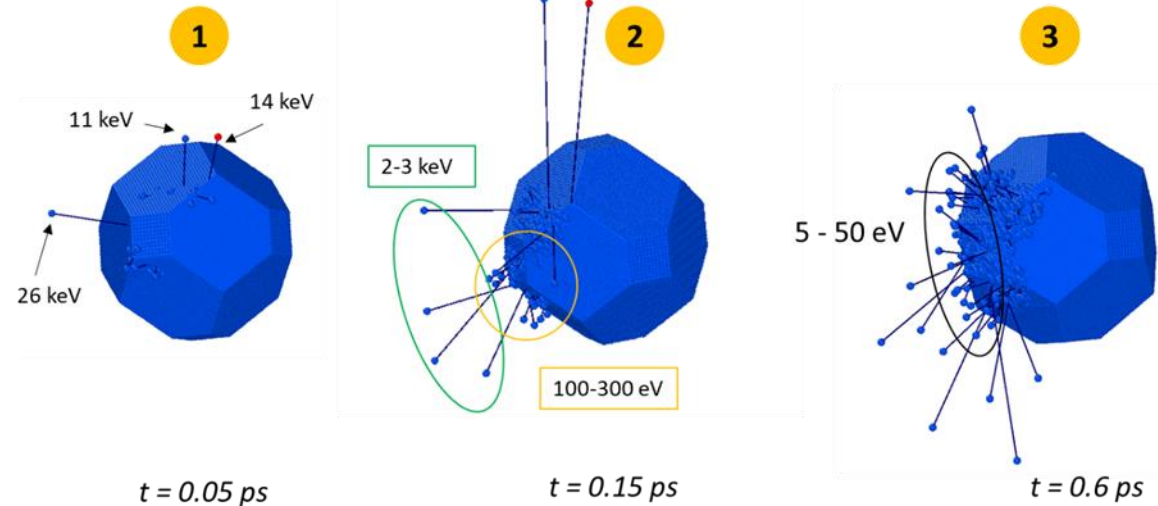


Al cap with cooling

Tritium target

End of proton beam path

Semi-empirical potentials with MD to model the collision cascades inside Mo NP:

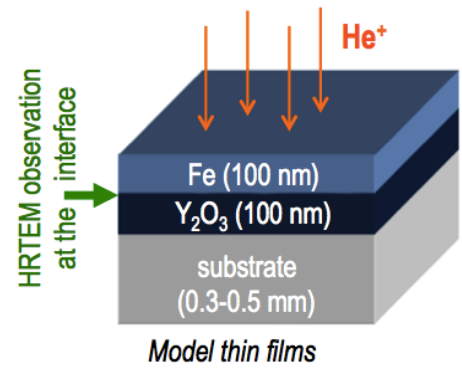


He diffusion in ODS* steels (J. Roques)

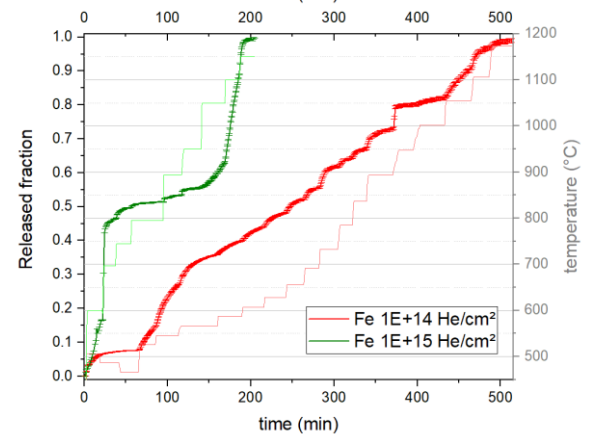
ODS steels = candidate materials for structural components in fusion (or GEN IV) reactors for their improved mechanical properties, higher operating temperatures, ... **BUT: diffusion of Helium ? Bubble formation ?**

Collaboration between experimenters (CENBG / IJCLab) and theorist (IJCLab):

Irradiation @ Jannus-Scalp

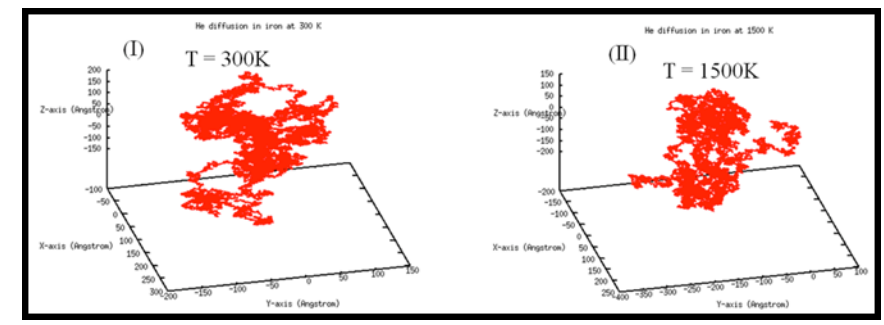
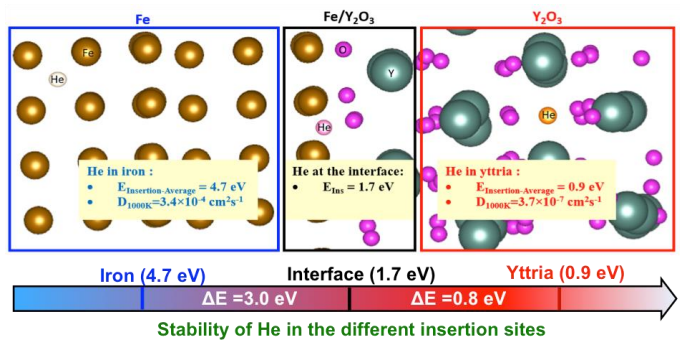


TDS @ CENBG



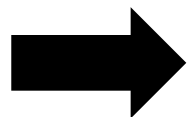
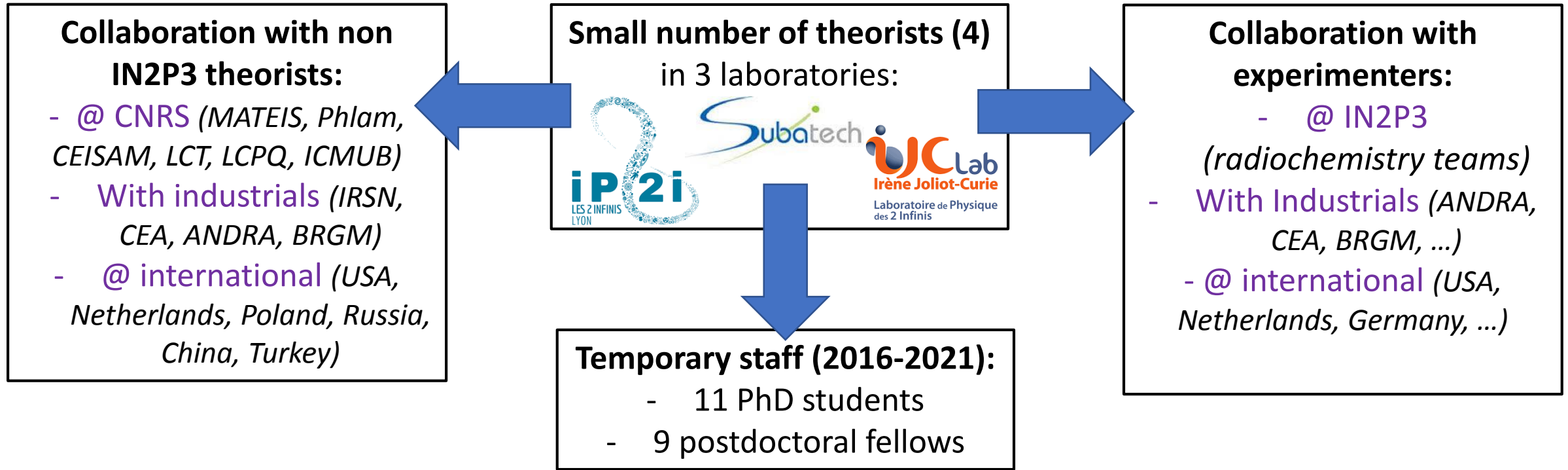
- Diffusion coefficients
- Activation energy from Arrhenius law

DFT + kMC calculations (IJCLab):



*ODS: Oxide Dispersion Strengthened

Theoretical chemistry @ IN2P3/CNRS: *manpower*



- Lots of different research topics to investigate,
- Increasing demand for coupling experimental data and computational chemistry,
- Large collaboration network.

- **Multiscale approach:**

- Recommendation of the working group (GT11) in its “2020-2030 French Strategic Plan for Nuclear Physics, Particle Physics, Astroparticle Physics, associated technologies & applications” report.
- Growing interest from industrials (ANDRA for nuclear wastes, Framatome and others for advanced structural materials used in GEN IV / fusion reactors).



- Already in use in our topics.
- Could be intensified in the next years

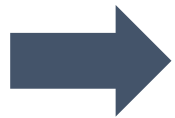


RF Thémósia which could be an excellent place of exchanges between code developers and users

Short-term perspectives (2/2)

- **Close interactions with experimenters:**

- Strengthening of the theory-experiment link in molecular radiochemistry through R. Maurice projects (main goal of the RCT master project).
- Strong demand from IN2P3 experimenters (or outside) with a lot of multi-disciplinary projects already done or in the pipelines (ANR BENEFICIA, ...).
- Several structures (GDR SCINEE – IN2P3 & INC // NEEDS programme // ...) exist to favor the interactions between theorists and experimenters and to initiate « big » projects.



- Theory-experiment link in molecular radiochemistry @ IN2P3 could be weakened after the departure of R. Maurice to ISCR Rennes (INC).
- Activities on solids (nuclear wastes, future materials, ...) should increase in the next years.

THANK YOU FOR YOUR
ATTENTION

A decorative graphic consisting of several parallel white lines of varying thicknesses, slanted diagonally from the bottom-left towards the top-right, located in the lower right quadrant of the slide.