**Contribution aux exercices de prospective 2020-2030**

***Contribution to the 2020-2030 prospective reflection***

**Energie nucléaire et environnement**

*Nuclear energy and environment*

**1) Aperçu / *Overview***

Thème de recherche proposé :

*Research topic of the proposition :*

Complexation studies of radionuclides with ligands of environmental interest.

Axe principal concerné (**voir la liste des thèmes en fin de document**) :

*Main research topic (****see the list of research topics at the end of this document****) :*

Radioactivity and environment: acquisition of basic data (speciation, ligands), modelling, transfer processes, measurements of very low radioactivity. Application to the behaviour of radionuclides in the biotope, microorganisms, exploration of remediation processes.

Contributeur(s) (et affiliations) de la proposition :

*Proposition’s author(s) and affiliations :*

Sladkov Vladimir, IPNO/FLUO.

Email du contact de la proposition :

*E-mail of the corresponding author :*

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Résumé (500 caractères max., incluant les espaces) :

*Summary (500 characters maximum, including spaces) :*

The aim of this project is to characterize metal binding properties of siderophores in solution from structural and thermodynamic viewpoints for checking their efficiency in terms of affinity and selectivity. This information can be used for prediction on radionuclide migration in the environment and for creation of specific DGT (Diffusive Gradient in Thin-films) captors for plutonium (Pu4+) and other actinides (An4+), and of some tetravalent d-block metal cation surrogates (Zr4+, Hf4+).

**2) Description de la question/problématique scientifique rattachée au thème (1 page) / *Description of the scientific issue connected to the topic (1 page)***

The release into the environment of highly toxic actinides (An) and other radionuclides (RN) as a consequence of nuclear accidents, weapon tests, and waste processing/disposal all contribute to the radioactive contamination of soils and surface water. Knowledge of radionuclide transport in the environment is crucial, not only for fundamental geochemistry, but for assessing the risk posed by long-term storage of nuclear waste. The environmental impact strongly depends on the mobility of a given radionuclide in environmental aquatic systems.

Among a variety of possible geochemical processes (solid/water interface interactions, transport processes), complex formation with inorganic and organic species present in the aqueous phase plays an important role in the eventual dispersion of radionuclides. The formation of such complexes typically causes significant changes in their migration properties in the environment.

The interaction of radionuclides with a natural organic matter presents a particular interest. Humic acids are the principal components of humic substances, which are the major constituents of organic matter. They are composed of aliphatic and aromatic structural elements and a variety of different, mainly oxygen containing functional groups. The most simple model substances for humic acids represent low-molecular short chain organic acids which can occur as humic acid building blocks and simulate their structure and functionality. Siderophores, as natural iron-specific chelators, must be considered also in this context. These low molecular weight, hydrosoluble compounds are secreted by bacteria and fungi to overcome the limited bioavailability of iron under aerobic conditions by dissolution of iron oxohydroxides present in the soils. Obtained thermodynamic, kinetic and structural data may be used for prediction of actinide migration in the environment (waters, soils…).

The literature data on complexation of the metal ions with siderophores are very rare and they are not consistent.

The part of this project was granted by “NEEDS Environment” programme (2014-2018, interaction of uranyl and Ln(III) with siderophores) and by ANR (Project “Pluton”, 2018-2022).

The ANR “Pluton” deals with environmental problematic. Determination of trace levels of environmental plutonium, typically at femtomolar concentration levels, and other tetravalent metals requires a thorough radiochemical separation and very sensitive analytical methods. DGT (Diffusive Gradient in Thin-films) technique is widely used for sampling trace metals in various media (water, pore-water of soils and sediments). For the moment, there is no specific DGT able to discriminate different tetravalent species (An4+) from di- or trivalent transition metals. In the frame of this ANR it is proposed to develop specific An4+ resins usable in various physico-chemical environments (water, pore-water of soils and sediments). For this purpose, it is mandatory to design and select the optimal chelators, which should exhibit very high binding affinity and selectivity towards the tetravalent elements at the pH values of natural waters, over all other metals.

Merci d’indiquer le positionnement des objectifs dans l’état de l’art (échelle internationale), les liens avec des projets existants et/ou futurs, la pertinence du cadre académique dans la question abordée.

*Please include description of motivation against (international) state-of-the-art, as well as links to other projects (existing or foreseen), relevance of the academic frame for the issue suggested.*

Collaborateurs (personnes ou organismes) identifiés ou potentiels (dans et hors IN2P3) :

*Identified of potential collaborators (people or organizations, in- and outside IN2P3) :*

- Michel Meyer, Institut de Chimie Moléculaire de l'Université de Bourgogne (ICMUB, UMR 6302) ;

- Jean-Claude Chambron, Institut de Chimie de Strasbourg (UNISTA, UMR 7177) ;

- Laurelin Fevrier, IRSN ;

- Société « Triskem »

Instruments/Outils impliqués :

*Facilities/tools involved :*

Spectrophotometer

Capillary electrophoresis equipment

ICP-OES

Liquid Scintillation Counter

Calculation grid of IN2P3

**3) Suggestion de projet(s) pouvant répondre à la question/problématique proposée (1 page max.) / *Suggestion of project(s) addressing the issue proposed (1 page max)***

*Indiquer si possible l’envergure qu’auraient ce ou ces projets (manpower, budget, durée).*

*Indicate if possible the scale of this(these) project(s) (manpower, budget, duration).*

The aim of this project is to characterize metal binding properties of siderophores in solution from structural and thermodynamic viewpoints for checking their efficiency in terms of affinity and selectivity. This information can be used not only for prediction on radionuclide migration in the environment, but also for creation of specific DGT captors for plutonium (Pu4+) and other actinides (An4+), and of some tetravalent d-block metal cation surrogates (Zr4+, Hf4+). It should be stressed that all cited cations are labile with respect to the water exchange in their first coordination shell, thus fast reactions are anticipated. The new syntesised bio-inspired hydroxamic ligands (synthesized by our partners from ICMUB and UNISTA) with stronger chelating properties will be also studied in aqueous solutions following a multi-technique approach. The speciation of desferrioxamine B (natural siderophore) with Zr4+, Th4+, U4+, and Pu4+ and other radionuclides will be considered too, either because no data are available or for validation purposes (Pu4+). The main method used will be: spectrophotometry, affinity capillary electrophoresis, liquid-liquid extraction and ion exchange with ICP-OES detection or liquid scintillation counting. DFT calculations to confirm experimental data will be performed too.

The next step will be to immobilize these ligands onto organic polymers to afford original chelating resins (i) for analytical separation and (ii) for hitherto unavailable DGT-type devices. The pre-concentration capacities of the analytical DGT tool will be validated with respect to Pu(IV) and other tetravalent metals in laboratory (lab tests) and real environmental conditions (field tests). The industrial partner is committed to produce and commercialize the various resins and DGTs. This project will be performed also in close collaboration with IRSN (speciation in the environment, DGT design and tests) and industrial partner “Triskem”.

**Merci de renvoyer ce document à** [**prosp2020-GT11-copil-l@in2p3.fr**](mailto:prosp2020-GT11-copil-l@in2p3.fr) **avant le   
1er Novembre 2019**

***Please send this document to*** [***prosp2020-GT11-copil-l@in2p3.fr***](mailto:prosp2020-GT11-copil-l@in2p3.fr) ***before   
November 1rst, 2019***