

Radioprotection and Radioactive Waste

Romão Trindade

The Radioprotection and Radioactive Waste Group (GRRR) has pursued in 2008 R&D activities with its involvement in the KADRWaste Project, in collaboration with Portuguese academia (Faculty of Sciences-U. Lisbon, Faculty of Sciences-U. Porto, U. Évora and ITN/Chemistry Unit). This Project has started in January, 2008. Still in terms of R&D, the GRRR and the Chemistry Unit of ITN, are involved in the European IP ACSEPT (“Actinide Recycling by Separation and Transmutation”) approved under the Euratom FP7-Fission-2007 Framework Program and started in March. Also in 2008, negotiations were initiated concerning ITN’ participation in PETRUS II (FP7) and GEOSAF (IAEA) projects.

Concerning Education & Training, a member of the Group has finished a Masters Degree thesis in Safety and Radiological Protection at (IST-Technical University of Lisbon), another member continued his work in the framework of a PhD degree in the field of radioactive waste management in collaboration with FC/UL (Faculty of Sciences, U. Lisbon). Two Post Graduation Thesis in Applied Geology were completed in collaboration with Faculty of Sciences of U. Lisbon (Department of Geology). A final year degree thesis in Environmental Engineering in the area of scrap metal as radioactive waste was also finished in collaboration with “Universidade Lusófona de Humanidades e Tecnologias”. Members of the GRRR have participated as lecturers and invited professors in high-level education activities: Advanced Post-Graduation Studies (DFA) on “Safety and Radiological Protection“. They have also participated in several professional training courses, in-house through the Training Centre and upon external request.

Members of the Group were also involved in several committees, working groups and task forces from the

EU, IAEA, OECD, OSPAR, CTBTO and national groups.

Considering the activities related to legal obligations such as licensing of sealed sources for medical, industrial, teaching and research applications, the Group has issued 454 licensing requested for analysis and authorization during 2008. Also during the last year and still considering legal obligations, about 156 requests for collecting and storing radioactive waste were received and processed.

Twelve events related to the detection of radioactive substances in scrap metal were carried out by GRRR during 2008. In terms of radiological protection and safety verifications, the GRRR has carried out eight actions at medical, military vessel and recycling facilities.

The Monitoring Programme of the radioactive liquid discharges from public and private nuclear medicine services into the public sewage of Lisbon was continued in 2008, in collaboration with Lisbon Borough City Council (CML).

The ITN Monitoring Programme in compliance with Articles 35° and 36° of the Euratom Treaty has been pursued with the setup of the ECoDELiR (Estação de Controlo das Descargas dos Efluentes Líquidos Radioactivos), to monitor all radioactive liquid discharges of ITN *Campus*. At the end of the 2008, work has started in order to renew and improve the ECoDELiR infrastructures.

Still in the framework of the aforementioned Euratom Articles, the *Campus* environmental gamma radiation dose has been assessed through the gamma monitoring network, GAMMANET, with detectors located in three strategic points of the Campus operated and maintained by this Group.

Radioprotection and radioactive waste staff

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KADRWaste – Study of the Adsorption Mechanisms and Kinetics in Geomaterials and Their Structural Characterisation: Implications for Processes of Natural Attenuation of Heavy Metal Contamination and Radioactive Wastes Confinement

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This project (PTDC/CTE-GEX/82678/2006) has started in January 2008 and aims at providing integrated knowledge on the adsorption/desorption mechanisms and kinetics of both heavy metals (Cu, Zn) and radionuclides (¹³⁷Cs, ⁹⁰Sr, ²⁴¹Am, and ²³⁸U) onto clay minerals and sapropels from the micro to macroscopic level. The objective is to will be such as to reflect natural environments and to propose remediation strategies integrated in radioactive waste confinement solutions. Special attention will be paid to interactions between metals and substrate, including

surface complexes, surface sites, sorption equilibrium and ion speciation. Sorption efficiency by means of kinetics of adsorption/desorption will be done for the long-term stability of metals. Assessment of adsorption competition between some radionuclides and these metals and the effects of radiation-induced damage on clay surfaces will be essayed. Radiological characterisation of sites for the long-term radioactive waste repository as an important step for monitoring actions will be performed.

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ACSEPT-Actinide reCycling by SEParation and Transmutation (7th Programme EURATOM- FP7-Fission2007)

I. Paiva, J. Marçalo, P. Vaz, R. Trindade

Actinide recycling by separation and transmutation is considered worldwide and particularly in several European countries as one of the most promising strategies to reduce the inventory of radioactive waste, thus contributing to make nuclear energy more sustainable. The ACSEPT Consortium (FP7-Euratom Fission) has started in March of 2008 and ITN preliminary experimental results are already available. The overall objective in this project is to study the kinetics and energetics of the association reactions of Ln(III) and An(III) ions with ligands effective in Ln/An separations and in use within ACSEPT, by means of mass spectrometry techniques. An experimental methodology has been setup to carry out studies of the gas-phase reactivity of Ln(III) and An(III) ions with ligands provided by ACSEPT partners, using FT-ICR mass spectrometry.

In the FT-ICR/MS technique, metal ions generated by laser desorption/ionisation (LDI) are injected and confined in the ICR cell, where they are selected and collisions with neutral species can occur. Resulting reactions can be monitored with time and the ionic species formed can be detected. The main feature of gas-phase reactions is that, especially at low pressure, the physical and chemical properties of ionic species can be studied in the absence of disturbing factors such as the solvent and lattice inherent to the condensed phases (solution or solid). Ion-molecule reactions in the gas phase can be up to two orders of magnitude faster than in solution due to the long-range attractive forces that confer enough energy to the ion-neutral complex to quickly proceed to the reaction products.

Preliminary studies of the gas-phase association of the Me-BTBP ligand (from CEA) with Gd(III) and Cm(III) as MCl_2^+ ions were performed. The experiments were carried out using a Finnigan FT/MS 2001-DT FT-ICR mass spectrometer, equipped with a 3 T superconductivity magnet, a Spectra-Physics Quanta-Ray GCR-11 Nd:YAG laser (1064 nm) and a Finnigan Venus Odyssey data system. The formation of the MCl_2^+ ions from the reactions of M^+ or MO^+ with CH_2Cl_2 was studied in detail. These gas-phase reactions have not been studied previously and bear interest in themselves for gas-phase f-element chemistry. The kinetics of reactions 1-3 was studied and the reaction efficiencies (k/k_{COL}) determined was as follows: Gd^+ (45%), GdO^+ (6.5%), Cm^+ (39%), CmO^+ (24%). Experimental procedures are being optimized.

In the ESI-QIT/MS technique, metal ion complexation in solution can be probed. Very preliminary studies with the Me-BTBP and TEMA ligands (from CEA) in solution, to study the competitive complexation with Pr, Nd and Sm (as the nitrates) and the effect of ionic radii, were carried out, using a Bruker HCT ESI-QIT mass spectrometer.

Characterization of Suitable Areas for a Long-Term Radioactive Waste Repository Facility in PortugalP. Duarte, I. Paiva, A. Mateus¹, R. Trindade

The work in progress is part of a PhD thesis being carried out in collaboration with Department of Geology/FCUL. Following a preliminary study, two specific potential suitable areas to host near-surface repositories (geologically composed by peridotites and schists), have been chosen to be characterized in more detail. Radiological mapping of both areas was performed, using a based Geographic Information System and gamma radiation data from 980 measuring points along several profiles. These data was obtained with portable NaI (Tl) gamma detectors. Statistical treatment of the data (interpolation by kriging) obtained from the radiometric profiles pointed out significant and representative sampling locals. In these locals, soils (through vertical profiles in three different depths), rocks, and vegetable covers were collected and were taken to the lab for ongoing analysis by various techniques: Gamma spectrometry for the identification and quantification of natural and artificial gamma emitters (total sample and fraction $<63\mu m$); XRD for mineralogical analysis (with emphasis on granulometric fraction $<10\mu m$ due to the importance of clay minerals in radionuclide retention processes). Organic matter content was also quantified in soil samples and, rock samples mineralogy is in progress.

Studies of the $<63\mu m$ soil fraction adsorption capacity for radionuclides, mainly ^{137}Cs , have started (with the involvement of Group of Environmental Radioactivity of DPRSN). ED-XRF analyses were carried out with the collaboration of the Environmental Analytical Chemistry Group of the ITN's Chemistry Department; Gamma Spectrometry studies were developed in collaboration with the Measurement Unit of ITN's DPRSN; FT-IR measurements were performed in the Department of Geology of FCUP.

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SERVICES

1. Radioactive waste management

During 2008 about 156 requests for radioactive waste collection were received and carried out. These radioactive wastes were collected, segregated, transported for interim storage at the “Pavilhão de Armazenamento Interino de Resíduos Radioactivos”, (PAIRR) located at ITN *Campus*. In this year only one (1) technician is working at PAIRR, which is not enough to carry out all present duties. It is very important and urgent to define and to establish a national plan related to radioactive wastes produced in Portugal, according international directives.

2. Sealed sources licensing

According Decree-Law n° 38/2007 and Decree-Law n° 165/2002, 454 sealed sources licensing were issued: national territory introduction licences (150), transfer licences (47), transport licences (90) and ownership licences (167). Only one person is related with this activity.

3. Gamma Monitoring Network (GAMMANET) of Instituto Tecnológico e Nuclear (ITN)

The environmental dose gamma radiation at ITN *Campus* is continuously being measured by the gamma network, GAMMANET. The data are collected, analysed and reported to the EU, according to articles 35° and 36° of the Euratom Treaty. During 2008 the network was reviewed and calibrated at Germany.

4. Radioactive liquid discharges from Instituto Tecnológico e Nuclear (ITN)

Radioactive liquid wastes originated at ITN are analysed and measured at “Estação de Controlo das Descargas dos Efluentes Líquidos Radioactivos” (ECoDELiR) before being discharged into Estação de Águas Residuais (ETAR). The data are reported to the EU according Articles 35° and 36° of Euratom Treaty and to the Radioactive Substances Committee of OSPAR Convention. By the end of 2008, the work to repair and to improve ECoDELiR has started.

5. Radioactive liquid discharges from Instituto Português de Oncologia (IPO), Coimbra

In 2008 and as requested by IPO-CROC, EPE, Coimbra, the radiological survey of radioactive liquid effluents from the IPO’s Medicine Nuclear Retention Tanks, was carried out by the Group before discharge into the public sewage.

6. Nuclear vessels radiological monitoring

In 2008, two nuclear vessels stayed at Portinho da Costa harbour and estuary of Rio Tejo. Environmental radioactivity survey programmes consisting on continuous monitoring of radioactive aerosols and airborne radioiodine, sampling of water, sediments and biological species for gamma spectrometry analysis were carried out. Sampling was done before, during and after the stay of the vessel. Reports were sent to Ministry of Defence.

7. Radioactivity in scrap metal

In 2008, and as result of radiological surveys requested by the smelting industry, twelve events related to the detection of radioactive materials in scrap metal at smelting factories have been reported. The material collected has been stored at Pavilhão de Armazenamento Interino de Resíduos Radioactivos, (PAIRR) as radioactive waste. Reports were sent to the smelting company.

8. Radiological protection and safety verifications

During 2008, GRRR has carry out six verifications concerning radiological protection and safety at medical facilities, in collaboration with GDR. Two radiological monitoring actions were carry out in a military vessel and in a recycling facility.

9. Radioactive Liquid Discharges from Hospitals in Public Sewage of Lisbon Borough Council (CML)

Radioactive liquid discharges from private and state owned nuclear medicines in Lisbon public sewage as well as residual effluents from Lisbon’s four Water Treatment Plants (ETARs), have been monitored by UPSR/ITN in 2008. Sampling was carried out in order to identify the radionuclides present and their activities. About 100 samples of liquid effluents were collected and analysed by quantitative and qualitative gamma spectrometry. This monitoring programme, requested by CML, was divided in two different programmes. Programme I involved sequential collection of 4 discrete samples in 5 sampling points from nuclear medicine facilities. In Programme II, 4 discrete samples were taken at one single discharge point of each Lisbon’s ETARs.