NKS-290

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Abstract Book of the NKS Workshop on Radioanalytical Chemistry, Risø, Roskilde, Denmark, 2-6th Sept. 2013

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August 2013

Abstract

This report compiled all abstracts presented in the NKS Workshop on Radioanalytical Chemistry at Risø, Roskilde, Denmark in 2-6th Sept. 2013. Total 35 participants registered to the workshop, among them 18 from Sweden, 5 from Denmark, 3 from Finland, and 3 from Norway, there are also 6 participants from Germany, France, Slovenia, Korea, Turkey, and China. The workshop consists two part, 3 days lab practices and 2 days lectures/presentation. 3 lab practices were organized, i.e. (1) Radiochemical separation of Pu and ICP-MS measurement of Pu isotopes; (2) Radiochemical separation of ^{210}Po and ^{226}Ra and their alpha spectrometry measurement; and (3) Radiochemical separation of ^{55}Fe, ^{63}Ni, ^{90}Sr and their LSC measurement. Among them, each participant can participate in 2 lab practices. 15 invited lectures are given by the experts in their specific fields, 8 oral and 6 poster presentations are given by the participants. The abstracts of all presentations are included in this report.

Key words

Radioanalysis, radionuclides, workshop, lab practice

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2^{nd} NKS-B Workshop on Radioanalytical Chemistry

2-6th Sept. 2013, Risø, Roskilde, Denmark

The workshop organizers:

Technical University of Denmark, Center for Nuclear Technologies (DTU Nutech), Denmark

University of Helsinki, Finland

Royal Institute of Technology, Sweden

Norwegian University of Life Sciences, Norway

Swedish Radiation Safety Authority, Sweden

Organizing Committee:

Xiaolin Hou, Technical University of Denmark (Chairman)

Sven P. Nielsen, Technical University of Denmark

Jukka Lehto, University of Helsinki, Finland

Mats Jonsson, Royal Institute of Technology, Sweden

Lindis Skipperud, Norwegian University of Life Sciences

Mats Eriksson, Swedish Radiation Safety Authority, Sweden

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The workshop sponsor:

Nordic Nuclear Safety Research (NKS), Program B (Manager: Kasper Andersson)

Workshop Programme

Monday, 2nd Sept. 2013

8:30-9:00	Welcome and Introduction (Xiaolin Hou, Per Roos, Jixin Qiao) Risø, Building 201 meeting room (Big)
9:00-16:30	Lab Practice
	Group1-1: Radiochemical separation of Pu and ICP-MS measurement of Pu isotopes (Tutor: Jixin Qiao), Building 204 at Risø.
	Group2-1: Radiochemical separation of ^{210}Po and ^{226}Ra and their alpha spectrometry measurement (tutor: Per Roos), Building 204 at Risø.
	Group3-1: Radiochemical separation of ^{55}Fe, ^{63}Ni, ^{90}Sr and their LSC measurement (Tutor: Xiaolin Hou), Building 202 at Risø.

Tuesday, 3rd Sept. 2013

8:30-12:00	Lab Practice
	Group1-1(continued): Radiochemical separation of Pu and ICP-MS measurement of Pu isotopes (Tutor: Jixin Qiao), Building 204 at Risø.
	Group2-1 (Continued): Radiochemical separation of ^{210}Po and ^{226}Ra and their alpha spectrometry measurement (tutor: Per Roos), Building 204 at Risø.
	Group3-1 (Continued): Radiochemical separation of ^{55}Fe, ^{63}Ni, ^{90}Sr and their LSC measurement (Tutor: Xiaolin Hou), Building 202 at Risø.
13:30-16:30	Group1-2: Radiochemical separation of Pu and ICP-MS measurement of Pu isotopes (Tutor: Jixin Qiao), Building 204 at Risø.
	Group2-2: Radiochemical separation of ^{210}Po and ^{226}Ra and their alpha spectrometry measurement (tutor: Per Roos), Building 204 at Risø.
	Group3-2: Radiochemical separation of ^{55}Fe, ^{63}Ni, ^{90}Sr and their LSC measurement (Tutor: Xiaolin Hou), Building 202 at Risø.

Wednesday, 4th Sept. 2013

8:30-16:30 (Lab practice)	Lab Practice
	Group1-2 (Continued): Radiochemical separation of Pu and ICP-MS measurement of Pu isotopes (Tutor: Jixin Qiao), Building 204 at Risø.
	Group2-2 (Continued): Radiochemical separation of ^{210}Po and ^{226}Ra and their alpha spectrometry measurement (tutor: Per Roos), Building 204 at Risø.
	Group3-2 (Continued): Radiochemical separation of ^{55}Fe, ^{63}Ni, ^{90}Sr and their LSC measurement (Tutor: Xiaolin Hou), Building 202 at Risø.

Note: Each lab practice will take 1.5 days, and will be repeated once. Each participate can participate in two lab practices. The second lab practice will start from Tuesday afternoon, 3rd Sept. 13:30.

Thursday, 5th Sept. 2013 (Risø, Building 115 Meeting room)

Date/Time	Presentator	Title of lecture
Opening session	Welcome and introduction (Chaired by Prof. Xiaolin Hou)	
8:30-9:00	Jens-Peter Lynlov (Director of DTU-Nutech) Sven P. Nielsen (Head of RAS, DTU-Nutech, workshop organizer) Kasper Ansdersson (NKS-B manager)	
Sesssion-1	General Aspect of Radioanalytical Chemistry, (Chaired by Dr. Sven P. Nielsen)	
9:00-9:35	Jukka Lehto (University of Helsinki, Finland)	The chemistry behind analytical actinide separations
9:35-10:10	Xiaolin Hou (DTU Nutech)	Radioanalytical Chemistry in decommissioning of nuclear facilities
10:10-10:30	Coffee Break	
Session-2	**Radioanalytical methods-1**, Chaired by Prof. Jukka Lehto	
10:30-11:05 **(Invited)**	Ian W. Croudace (University of Southampton, UK	Application of thermal desorption techniques in the characterisation of nuclear decommissioning wastes
11:05-11:40 **(Invited)**	Jinzhou Du (East China Normal University, China)	Analysis of radionuclides in marine environment and their implications
11:40-12:15	Lindis Skipperud (University of Life Sciences, Norway)	Analysis and results of Po-210 and Pb-210 in fish – study of bioavailability
12:15-13:45	Lunch	
Session-3	**Radioanalytical methods-2,** Chaired by Prof. Jukka Lehto	
13:45-14:20 **(Invited)**	Phil E. Warwick (National Oceanography Center, UK)	Methodologies for determination of radionuclides in environmental or waste samples
14:20-14:55	Xiaolin Hou (DTU Nutech)	Updated analytical methods on determination of ^{63}Ni, ^{55}Fe, and ^{90}Sr
14:55-15:30	Aude Bombard (Triskem, France)	New Extraction chromatographic resin -CL resin for separation of ^{36}Cl and ^{129}I
15.30-15:50	Coffee Break	
Session-4	**Mass spectrometry methods**, Chaired by Dr. Lindis Skipperud	
15:50-16:25 **(Invited)**	Peter Steier (University of Vienna, Austria)	Determination of radionculides using AMS
16 :25-16 :55	Per Roos (DTU Nutech)	Determination of radioisotopes using ICP-MS – the coupling between chemistry and instrumentation
16 :55-17 :30	Mats Eriksson (Swedish Radiation Safety Authority, SSM, Sweden)	Nuclear fingerprinting of radioactive material in a radiological and nuclear safeguard perspective
18 :00-21 :30	**Workshop dinner on board of Roskilde Fjord tour**	

Friday, 6th Sept. 2013 (Risø, Building 115 Meeting room)

Date/Time	Presentator	Title of lecture
Session-5	**Radiometric methods**, Chaired by Dr. Mats Eriksson	
8:30-9:05 **(Invited)**	Jost Eikenberg (PSI, Switzerland)	Principles of liquid scintillation counting: theories and applications
9:05-9:40	Sven P. Nielsen (DTU Nutech)	Alpha, beta and gamma measurements of environmental radioactivity
Session-6	**Speciation analysis,** Chaired by Dr. Per Roos	
9:40-10:15 **(Invited)**	Clemens Walther (Leibniz University Hannover)	Actinide speciation analysis using mass spectrometry and laser spectroscopy
10:15-10:45 **(Invited)**	Brit Salbu (University of Life Sciences, Norway)	Speciation analysis of radionuclides in the environment
10:45-11:05	Coffee break	
Session-6	**Automated methods,** Chaired by Dr. Kasper Andersson	
11:05-11:40	Jay W. Grate (Pacific Northwest National Laboratory, USA)	Methodology and Application of Automation in Radiochemical Separations and Analysis
11:40-12:15	Jixin Qiao (DTU Nutech)	Automation and Methodology Development for Environmental and Biological Determination of Pu, Np, U and Tc
12:15-13:45	Lunch	
Session-7	**Presentation by participants,** Chaired by Prof. Xiaolin Hou	
13:45-14:00	Michael Granfors (Studsvik, Sweden)	Isotope dilution analysis of ^{90}Sr using dynamic reaction cell inductively coupled plasma mass spectrometry (IDA-DRC-ICP-MS)
14:00-14:15	Hyuncheol Kim(Korean Atomic Energy Research Institute)	A simultaneous pre-concentration procedure of Sr and Pu from seawater using precipitation
14:15-14:30	Mattias Olsson (Forsmarks kraftgrupp AB)	An Overview of Methods for Nuclide Specific NPP Release Monitoring
14:30-14:45	Daniela Pittauerova (University of Bremen)	Mathematical efficiency calibration in gamma spectrometry for analysis in natural and fallout radionuclides in sediments
14:45-15:00	Miao Yang (KTH, Sweden)	Inhibition of radiation induced dissolution of UO_2 by sulfide - a comparison with the hydrogen effect
15:00-15:15	Maria Kaipiainen (University of Helsinki, Finland)	Advanced separation techniques for radionuckel
15:15-15:30	Sofie Englund (OKG AB, Sweden)	Evaluation of ”double separation” to avoid interference by Co in the analysis of Ni-63
15:30-15:45	Yihong Xu (DTU Nutech)	Plutonium isotopes in Chinese soils and its potential application for tracing soil erosion
Session-9 15:45-15:55	Open Discussion (Xiaolin Hou/Kasper Andersson)	
15:55-16:00	Close of workshop	

Friday, 6th Sept. 2013 (Risø, Building 115 Meeting room)

Date/Time	Presentator	Title of lecture
Session-8	**Poster session**	
13:00-13:45 5th & 6th Sept.	Alexandre Barreiro Fidalgo (KTH, Sweden)	Effect of bentonite on radiation induced dissolution of UO_2 in an aqueous system
	Jenna Knuutinen (University of Helsinki, Finland)	Sorption of europium(III), nickel(II) and calcium(II) on α-Al2O3, TiO2 and ZrO2
	Kristina Nilsson (KTH, Sweden)	ADOPT pellet leaching properties, a comparison with UO_2 pellet
	Risto Juvonen (Hidex Oy, Finland)	Quench correction using Triple to Double Coincidence Ratio (TDCR) method on Liquid Scintillation Counting (LSC)
	Akbar Abbasi (Eastern Mediterranean University)	Analysis and measurements of ^{226}Ra in drinking water used in Caspian area
	Luyuan Zhang (DTU Nutech)	Speciation Analysis of iodine Isotopes (^{129}I and ^{127}I) in Aerosol Samples
	Stefan B. Bengtsson	Methodologies for determination of different types of radionuclides in radiological emergencies and their behaviour

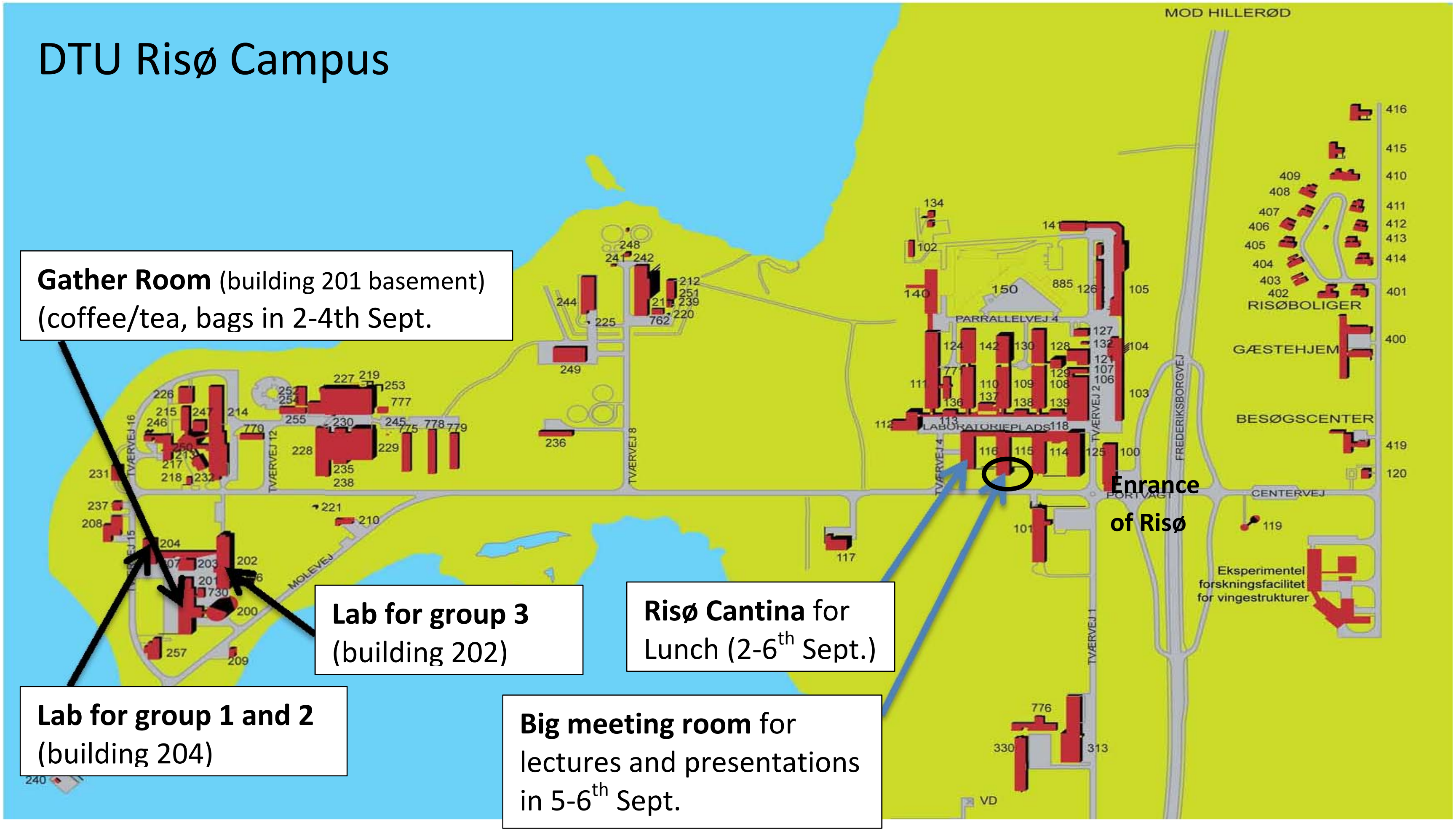
Note: The posters can be attached to the board from 9:00 am 5th Sept., and removed at 16:00 6th Sept. 2013. The authors can present the work during 13:00-13:40 on 5th and 6th Sept.

Groups of lab practice

Goup 1-1	Goup 1-2	Goup 2-1	Goup 2-2	Goup 3-1	Goup 3-2
Anna Ljungfalk	Akbar Abbasi	Akbar Abbasi	Asser N. Poulsen	Asser N. Poulsen	Anna Ljungfalk
Camilla Nordhei	Alexandre B. Fidalgo	Alexandre B. Fidalgo	Filippa S. Bruzell	Filippa S. Bruzell	Camilla Nordhei
Frances Burrell	Daniela Pittauerova	Daniela Pittauerova	Frances Burrell	Gustav Pettersson	Helene Öhlin
Helene Öhlin	Frank Jacobsen	Frank Jacobsen	Gustav Pettersson	Irene Boren	Hyuncheol Kim
Hyuncheol Kim	Jenna Knuutinen	Jenna Knuutinen	Irene Boren	Jasmina Znidersic	Kristina Nilsson
Kristina Nilsson	Katrin Ahlford	Katrin Ahlford	Jasmina Znidersic	Klas Källström	Maria Kaipiainen
Maria Kaipiainen	Luyuan Zhang	Luyuan Zhang	Klas Källström	Lene Valle	Miao Yang
Miao Yang	Stefan Bengtsson	Parvine Naghchbandi	Lene Valle	Lina Ekerljung	Parvine Naghchbandi
Qianqian Bi	Yihong Xu	Sara Ehrs	Lina Ekerljung	Maria Anderot	Qianqian Bi
Risto Juvonen	Frances Burrell	Sofie Englund,	Maria Anderot	Mattias Olsson	Risto Juvonen
		Stefan Bengtsson	Mattias Olsson	Olof Gottfridsson	Sara Ehrs
		Yihong Xu	Olof Gottfridsson	Frances Burrell	Sofie Englund,

Note: The lab practice will be carried out in 6 groups, and each participant will participate in 2 groups in 3 days (2-4th Sept.). The lab practice of groups 1-1, 2-1 and 3-1 will be implemented from 9:00 am 2nd Sept. until 12:00am 3rd Sept.; and the lab practice of group2 1-2, 2-2 and 3-2 will start from 13:30 3rd Sept until 16:30 4th Sept.

Locations of workshop in 2-6th Sept. 2013



Gather Room (building 201 basement) (coffee/tea, bags in 2-4th Sept.

Lab for group 3 (building 202)

Risø Cantina for Lunch (2-6th Sept.)

Lab for group 1 and 2 (building 204)

Big meeting room for lectures and presentations in 5-6th Sept.

THE CHEMISTRY BEHIND ANALYTICAL ACTINIDE SEPARATIONS

Jukka Lehto

Laboratory of Radiochemistry, Department of Chemistry, University of Helsinki, Finland

Both environmental and nuclear waste materials contain important actinides (Th, U, Np, Pu, Am, Cm) the concentration and isotopic composition of which are typically measured either by alpha spectrometry or by mass spectrometry. In most cases the actinides need to be chemically separated from interfering radionuclides and stable elements to enable their measurement. In solid geological samples (rock, soil, sediments) both thorium and uranium are abundant while in ground waters only uranium is relevant. For the measurement of thorium and uranium by alpha spectrometry they need to be removed from other emitting radionuclides, ^{226}Ra and ^{210}Po. Measurement of uranium concentration directly from natural waters by ICP-MS is practical but if accurate isotopic composition is needed mass spectrometric measurements also require radiochemical separations of Th and U.

The upper soil and surface waters, as well as nuclear waste, may also contain measureable amounts of transuranium actinides (TRU) neptunium, plutonium, americium and curium. The activities of neptunium and curium compared to other actinides are so low that they do not cause a problem in the measurement of other actinides but if their own activities need to be measured careful chemical isolation is required.

The selection of chemical separation procedure for the isolation of actinides for measurement is dependent on three main factors:

- Sample type:
    - environmental samples without TRU nuclides
    - environmental samples with TRU nuclides
    - waste samples with variable U and TRU composition but no Th, Ra and Po present
- Measurement method:
    - alpha spectrometry
    - mass spectrometry
- Target element/s, i.e. which actinide/s is/are to be determined

Actinides (An) are a group of elements characterized by filling of 5f orbitals. Actinium and actinides from americium on behave very much like lanthanides and are mostly at the trivalent oxidation state (An^{3+}). Elements between actinium and americium in turn behave like transition elements and have variable oxidation states between +III and +VI. In the higher oxidation states +V and +VII they occur as AnO_2^{+} and AnO_2^{2+} oxocations. Thorium occurs solely at the tetravalent state as Th^{4+} which is readily hydrolysable and highly insoluble and therefore cannot be practically found in natural waters. In the reducing conditions prevailing in deep geological conditions also uranium is in the tetravalent state U^{4+} and behaves much like thorium. Uranium, is, however, readily oxidizable to hexavalent uranyl ion UO_2^{2+}

in which form it is usually in radiochemical separations. Neptunium is also a redox-sensitive element occurring in the tetravalent form Np^{4+} in reducing and slightly oxidizing conditions and turns into the pentavalent NpO_2^+ in more oxidizing conditions. Plutonium is the most challenging element with respect to redox behavior. Its most stable oxidation state is +IV (Pu^{4+}) but it can simultaneously exist at four oxidation states, including also Pu^{3+}, PuO_2^+ and PuO_2^{2+}.

Most typically actinides are separated from each other as well as from interfering radionuclides (^{226}Ra, ^{210}Po) and stable elements by anion exchange chromatography, solvent extraction or extraction chromatography. The separations are carried out in rather concentrated nitric and hydrochloric solutions and are based on the stabilities of nitrate and chloride complexes of actinides. Actinides forming stable anionic complexes, such as $UO_2Cl_4^{2-}$, are efficiently retained in the anion exchanger while elements forming no or very weak complexes, such Th^{4+} with chloride, pass the column. In case of solvent extraction and extraction chromatography the forming complexes transferring into the organic phase are neutral, such as UO_2Cl_2.

In addition to the stability of chloride and nitrate complexes of actinides, as well as interfering radionuclides and elements, radiochemical separations make use of oxidation state adjustment because the stabilities of nitrate and chloride complexes differ much from oxidation state to another. The oxidation state adjustment applies to two actinides Pu and Np. In the radiochemical of plutonium its oxidation state is changed between Pu^{4+}, forming strong complexes, and Pu^{3+}, forming no or weak complexes. In case of Np it is adjusted to highly retaining tetravalent Np^{4+} for its separation.

Below major chemical features of the separations are listed for the most important actinides:

- Thorium exists solely as Th^{4+} which forms a strong nitrate complex but no chloride complex.
- Uranium can exist as U^{4+} and UO_2^{2+} but only the latter one relevant in typical radiochemical separations. Uranyl ion forms a rather weak nitrate complex and a strong chloride complex.
- Neptunium occurs both as Np^{4+} and NpO_2^+ the latter one prevailing in most oxidizing conditions. For the radiochemical separation Np is reduced to Np^{4+} at which oxidation state it forms strong complexes especially with nitrate.
- Plutonium occurs at four oxidation states as Pu^{3+}, Pu^{4+}, PuO_2^+ and PuO_2^{2+}. In the tetravalent state Pu forms a very strong complex with nitrate and as such can be efficiently retained in the separation medium. To separate Pu from tetravalent (Th, Np) and hexavalent (U) actinides it is reduced to trivalent state which does not retain in most separation media while for the separation of Pu from trivalent actinides (Am, Cm) it is oxidized to Pu^{4+}.
- Americium and curium occur solely as trivalent Am^{3+} and Cm^{3+} which are not retained in separation media taking up tetra and hexavalent actinides.

Radioanalytical Chemistry in decommissioning of nuclear facilities

Xiaolin Hou

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With increasing request for decommissioning of nuclear facilities, characterization of various wastes is required for evaluation of the radioactivity inventory in various materials and decision making for management of the produced waste. This is carried out by quantitative determination of various radionuclides present in the materials. Among these radionuclides, the determination of pure beta and alpha emitters including ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{63}Ni, ^{90}Sr and some transuranics is the major challenges, because they could not be measured without separation from the matrix of the samples and from all other radionuclides, this entitles them as the radionuclides of difficult to measure.

In addition, the investigation of environmental and geological behaviors of radionuclides for radioecology and repository of nuclear waste also requests to measure various radionuclides in different environmental and geological matrix. In this case, the measurement of long-lived fission products and actinides, such as ^{129}I, ^{99}Tc, isotopes of plutonium and neptunium, is the major challenge.

In the past years, our laboratory has developed various radiochemical analytical methods aiming at characterization of various decommissioning waste by determination of various radionculides of difficult to measure [1-6]. Meanwhile we have also improved our analytical methods for determination long-lived radionuclides in the environment, focus on improving the detection limit, automation of analytical operation, and rapid determination. This presentation gives an overview of these analytical methods, mainly on the present status, progress and perspectives.

References:

1. Hou X.L., Østergaard L.F., Nielsen S.P., Determination of ^{36}Cl in Nuclear Waste from Reactor Decommissioning, *Anal. Chem.*, 2007, 79, 3126-3134.
2. Hou X.L, Radiochemical analysis of radionuclides difficult to measure, *J. Radioanal. Nucl. Chem.*, 2007, 273, 43-48
3. Hou Xiaolin, Radiochemical determination of ^{41}Ca in reactor concrete for decommissioning, Radiochim., Acta, 2005, 93:611-617.
4. Hou Xiaolin, Determination of C-14 and H-3 in Reactor Graphite and Concrete for Decommission, *Applied Radiation and Isotopes*, 2005, 62:871-882.
5. Hou X.L., Frøsig Østergaard L.; Nielsen S.P., Determination of Ni-63 and Fe-55 in nuclear waste and environmental samples, *Anal. Chim. Acta*, 2005, 535:297-307.

Application of thermal desorption techniques in the characterisation of nuclear decommissioning wastes

Ian W Croudace

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Thermal desorption-based techniques provide an efficient and rapid approach for the isolation of volatile radionuclides. Traditionally, such approaches have been routinely used for ^{3}H, ^{14}C and ^{35}S. However, thermal desorption / decomposition techniques are being increasingly applied to a wider range of radionuclides including ^{36}Cl, ^{99}Tc and ^{129}I. To ensure quantitative recovery of radionuclides during the desorption process, an in depth understanding of the association of the radionuclide with the bulk matrix is essential, particularly when considering the diverse range of matrices encountered during nuclear decommissioning. This review will summarise the findings of a recent programme of research undertaken by GAU-Radioanalytical into the association of ^{3}H and ^{14}C in decommissioning samples and its significance to sampling, sample storage and analytical recovery. The review will also consider how thermal desorption can be effectively extended to other radionuclides and discuss the associated analytical challenges.

Analysis of radionuclides in marine environment and their implications

Jinzhou Du

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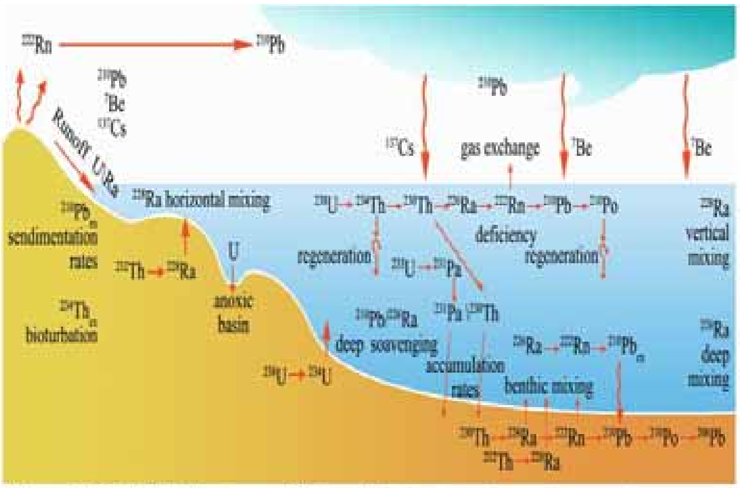

Figure 1. Nuclide behaviors and their application to marine environment processes

Radionuclides are an important tool to evaluate the ecological effects and good tracers to marine environment processes. This work describes radiometric determination of natural and anthropogenic radionuclides in marine environments (including suspended particles, sea water, sediment and biomass). Exception of most concerned anthropogenic radionuclides such as ^{137}Cs, ^{90}Sr, Pu, there are large amount of natural nuclides originally from uranium and thorium decay series such as ^{234}Th, Ra, ^{210}Po, etc. and cosmogenic nuclides such as ^{7}Be, ^{32}P,^{14}C, etc., which is broadly apply into marine environment processes(Figure 1). For example, the particle-reactive nuclides ^{7}Be, ^{234}Th, ^{137}Cs, ^{210}Pb can be used to study particle settling/resuspended processes, sediment dating, sediment dating. In the same time, the dissolved nuclides such as ^{222}Rn, Ra(^{223}Ra, ^{224}Ra, ^{226}Ra, ^{228}Ra) can used to diffusion proceeded from coast to offshore, submarine groundwater discharge(SGD), and interface exchange between atmosphere/surface seawater and sediment/bottom seawater (benthic layer) processes. Moreover, underwater gamma-ray spectrometry, a cost-effective technique for applications requiring surveys of large areas, an immediate assessment, or long-term observations of marine radioactivity, will also be discussed.

ANALYSIS OF Po-210 AND Pb-210 IN FISH – A STUDY OF BIOAVAILABILITY

SKIPPERUD, L., JØRGENSEN, A-G., HEIER, L.S., ROSSELAND, B.O. AND SALBU,B.

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In most countries, uranium mining is considered the most hazardous step of nuclear materials production, both in terms of radiation doses and in the number of people affected. Key problems have been associated with the transport of uranium and its daughters in aquatic and terrestrial ecosystems, where radionuclides are transferred from air, water and soils into plants, animals and finally to man. Special attention is paid to the most hazardous decay products of uranium; ^{210}Pb, ^{210}Po and ^{226}Ra. It is assumed that 40 % of the world reserves of uranium are situated in Kazakhstan, Kyrgyzstan and Tajikistan. Following the cold war, extensive uranium mining and production took place at numerous sites in the former Soviet Union.

Po-210 and ^{210}Pb in water and fish has been determined as part of a Joint project between Norway, Kazakhstan, Kyrgyzstan and Tajikistan. The project aims to assess long term consequences of TENORM waste from uranium mining and milling in the Central Asian region. The objective of the present work (Skipperud et al., 2013) was to determine the concentration and size distribution of the naturally occurring radioactive isotope ^{210}Po and ^{210}Pb in water and in different fish organs from an area of former uranium mining activity in Tajikistan.

Sampling of water was performed by *in situ* fractionation, separating radionuclides in terms of size (molecular mass) and charge. Fish were obtained from local fishermen. Determination of ^{210}Po and ^{210}Pb in water samples was performed by combining the method of Chen et al. (2001) and Suriyanarayanan (2008), while the analysis of fish organs (bone, liver and muscle) was performed in accordance with the method described by Chen et al. (2001). For total decomposition of fish tissue samples, acid decomposition by UltraCLAVE high performance reactor was used.

The total activity concentration of ^{210}Po in the Pit Lake was 5.6±0.7 mBq/L, and 210Po was mainly (60%) identified in the colloidal size fraction (<0.45µm – 10 kDa). In fish, the highest activity concentration of ^{210}Po was identified in the liver (3700±430 Bq kg^{-1} ww) of *C. auratus*, while the highest activity concentrations of ^{210}Pb were found in the bones (100±54 Bq kg^{-1}). The results show that the highest uptake of ^{210}Po was in the liver and that the distribution of ^{210}Po and ^{210}Pb in fish organs were different. Considering the standard BCF values presented by IAEA (2009); (High potential BCF >1000; Moderate Potential 1000 > BCF > 250; Low potential 250 >BCF), the BCF values indicated high potential BCF of 210Po in all organs of C. auratus. The activity concentration of ^{210}Po in liver showed a significant linear relation to δ^{15}N in *C. auratus* that could indicate a biomagnificating property of ^{210}Po.

References:

Skipperud, L., Jørgensen, A.G. Heier, L.S., Rosseland, B.O., Salbu, B., (2013) "Po-210 and Pb-210 in water and fish from Taboshar uranium mining Pit Lake, Tajikistan" Journal of Environmental Radioactivity, 123,82-89.

Chen, Q., Aarkrog, A., Nielsen, S.P., Dahlgaard, H., Lind, B., Kolstad, A.K., Yu, Y., (2001) "Procedures for Determination of ^{239,240}Pu, ^{241}Am, ^{237}Np, ^{234,238}U, ^{228,230,232}Th, ^{99}Tc and ^{210}Pb and ^{210}Po in Environmental Materials" Risø National Laboratory, Risø, Denmark.

Suriyanarayanan, S., Brahmanandhan, G.M., Malathi, J., Ravi Kumar, S., Masilamani, V., Shahul Hameed, P., Selvasekarapandian, S., (2008) "Studies on the distribution of ^{210}Po and ^{210}Pb in the ecosystem of Point Calimere Coast (Palk Strait), India" Journal of Environmental Radioactivity, 99 (4), 766-771.

IAEA, 2009. International Atomic Energy Agency (IAEA). Available at: iaea.org/ About/index.html (accessed 01.03.09.).

Methodologies for determination of radionuclides in environmental or waste samples

Phil E Warwick

University of Southampton, National Oceanography Centre, Southampton SO14 3ZH, UK

The transition of many nuclear sites to decommissioning has required radioanalytical laboratories to adapt to significantly different technical demands. Traditionally, laboratories were required to analyse a relatively limited range of sample types usually as part of an analytical programme in support of routine plant operations, effluent characterisation, health physics support or environmental monitoring. The onset of site decommissioning has given rise to a much more diverse range of sample matrices, many of which would not have been encountered during routine operational monitoring. Radionuclides in the samples may arise from contamination or direct irradiation of the sample, often over decades impacting on their association with the bulk matrix. In many cases, analytical schedules will include previously un-encountered radionuclides which are now significant for waste characterisation due to their long half lives and contribution to the long-term waste repository safety case. To meet these challenges, research has been undertaken into sample preparation procedures, novel radionuclide separation techniques, novel calibration and data processing techniques to extend the capability of liquid scintillation analysis, application of mass spectrometry and method validation approaches. Experience in applying this research in a routine radioanalytical laboratory is presented.

Updated analytical methods on determination of ^{63}Ni, ^{55}Fe and ^{90}Sr

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Both ^{63}Ni and ^{55}Fe are activation products through reactions of neutron with iron, nickel and copper. Due to high concentration of iron and nickel in construction materials of nuclear reactor, the inventory of ^{55}Fe and ^{63}Ni the nuclear waste, especially steel and alloy are relative high. Considering the relative long half-lives of these two radionuclides, ^{63}Ni and ^{55}Fe are two of the most important radionuclides in the management of nuclear waste, especially the metals from nuclear reactor which has been exposed to neutron irradiation. In addition, due to the corrosion of the metals in the water cooling system, ^{55}Fe and ^{63}Ni are also major contributors of radioactivity in exchange resin used for purification of water in nuclear reactor. ^{90}Sr is one of the most important fission products for both nuclear waste and environmental samples. All these radionuclides are pure beta emitter (or decay by electron capture), separation of them from sample matrix and other radionuclides is necessary before measurement. Many methods have been reported for the separation of these radionuclides, this work aim to overview these method, especially the newly reported method. The methods developed and applied in our laboratory for the analysis of waste and environmental samples are presented in detailed.

Separation and Determination of Cl-36 and I-129 using CL Resin

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Measurement techniques employed for the determination of Cl-36 and I-129 in environmental and decommissioning samples usually require thorough and selective sample preparation in order to give accurate and precise results; this is especially true for liquid scintillation counting. Methods employed for the separation and purification of these radionuclides are often tedious, time consuming and elaborate.

In order to facilitate the analysis of Cl-36 and I-129 an extraction chromatographic resin (CL Resin) that allows extraction, and subsequent separation, of Cl-36 and I-129 from pretreated environmental and decommissioning samples was developed and characterized [1]. Some results of the resin characterization, including weight distribution ratios (D_w values) of various cations on the CL resin, and of Cl^- and I^- on Ag^+ loaded CL resin, are presented.

Based on determined resin retention characteristics methods for the separation and determination of Cl-36 and I-129 from various matrices have been developed and tested on (spiked) real samples. Matrices analysed included drinking, sea and waste water, filter, concrete, soil, spent resin [2] and waste solution [3]; sample preparation methods applied included leaching and thermal decomposition of the samples. The results of these tests are presented.

References:

[1] A. Zulauf, S. Happel, M. B. Mokili, A. Bombard, H. Jungclas: Characterization of an extraction chromatographic resin for the separation and determination of ^{36}Cl and^{129}I. J. Radanal Nucl. Chem., 286(2), 2010, 539-54

[2] P E Warwick, A Zulauf, S Happel, I W Croudace: Determination of 36Cl in decommissioning samples using a Pyrolyser furnace and extraction chromatographic separations. Presented at the 11th ERA Symposium, September 2010, Chester (UK) *http://www.triskem-international.com/iso_album/11_era_chester_warwick_determination_of_36cl_in_decommissioning_samples_using_a_pyrolyser.pdf*

[3] C. Decamp, S. Happel: Utilization of a mixed-bed column for the removal of iodine from radioactive process waste solutions. J Radioanal Nucl Chem., DOI 10.1007/s10967-013-2503-1

Accelerator Mass Spectrometry - analysis of the rarest atom species for earth and environmental science

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Accelerator Mass Spectrometry (AMS) excels over other methods of analytical isotope research by its extraordinary abundance sensitivity, i.e. the ability to separate out the rarest atom species from abundant stable isotopes. This makes the method the perfect fit for long-lived radionuclides with half-lives between 1 kyr and 100 Myr, of both natural and anthropogenic origin. During the last 30 years, AMS has made significant contributions to practically all fields of science.

Dating based on natural C-14 for archaeology and palaeontology is still one major application. With increasing precision, radiocarbon dating starts to compete with traditional historical dating, e.g. connected to the ancient Thera eruption in the Eastern Mediterranean Sea. Anthropogenic C-14, produced by the atmospheric nuclear weapons tests, allows for so-called bomb-peak dating with a precision of about 1 year. This development has fallen on fertile ground in biomedicine. Dating of human tissue and especially DNA reveals new insights into turnover rates and regeneration. Pharmaceutical research benefits also from so called "microdosing" which studies the human metabolism with C-14 labeled, but nonhazardous small amounts of new substances. New dedicated C-14 AMS systems usually have reduced size and complexity, providing competitive performance with a footprint of only a few square meters.

A complementary trend to larger size accelerators is observed for the upcoming facilities aiming at the other "traditional" AMS isotopes Be-10, Al-26, Cl-36, and I-129. Their detection in surface rocks or in natural archives like ice cores allows to investigate the processes shaping our planet, and to assess their time scales, which is the basis for the prediction of the impact of anthropogenic activities on a global scale. The new facilities are often designated as working horses for earth and environmental science, and are not necessarily affiliated at physics departments.

Recently, U-236 and the plutonium isotopes from global fallout have demonstrated their potential as environmental tracers. U-236 shows conservative behavior in the ocean. Pu, on the other hand, binds to soil particles, and can thus serves as a tracer for sediment transport. Both uranium and plutonium can be used to detect releases of nuclear fuel.

The main technical challenge for AMS is imposed by stable isobars from other elements, which are always present at the ppm level even in purified samples. While the higher energy of AMS allows their suppression via the different energy loss in matter for the lighter masses, even the largest machines cannot generally separate isobars above Fe-60. Many isotopes in the middle mass range with suitable half-life are present in the environment as natural and anthropogenic fission products. New methods based on chemical reaction cells or laser detachment are developed to remove the interfering isobars. Once successful, a whole new spectrum of applications will be opened.

Determination of radioisotopes using ICP-MS – the coupling between chemistry and instrumentation

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With the introduction of inductively coupled plasma mass spectrometry in the 1980's there was a belief that analysis of several of the medium and long-lived radioisotopes soon may be done without the need of cumbersome and time-consuming chemical separations. With replacing of many radiometric methods by mass spectrometry the field of radiochemistry would only be found associated with analysis of short-lived radioisotopes. Similar thoughts made their way to the radioanalytical community with the introduction of collision cells in ICP-MS during the late 1990's. Experiences have however shown that the complexity with which disturbances in the obtained mass-spectra is produced by the combination of instrumental settings and sample composition is far greater than for radiometric methods. In order to be able to use the full potential of ICP-MS instrumentation in terms of sensitivity and isotope ratio measurements chemical isolation of the analyte is in general even more important than for radiometric methods. In this presentation an overview of ICP-MS as a tool for analysis of radioisotopes is given. Emphasis will be given on how the combination of chemistry and instrumentation influences the analyte signal.

Nuclear fingerprinting of radioactive material in a radiological and nuclear safeguard perspective

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The talk will show and discuss two special cases on the use of radio analytical techniques. It will focus on these techniques, applied on radioactive particles, to reveal a nuclear fingerprint in both a nuclear safeguard as well as a radiological perspective. Especially focus will be on the micro analytical characterization and speciation techniques, i.e. the use of radiation induced X-ray fluorescence techniques (synchrotron radiation based XRF, XANES and the accelerator based PIXE techniques) and the mass spectrometric technique, secondary ion mass spectrometry. A general discussion on the importance of studying radioactive particles in radiological investigations will also be given.

Principles of liquid scintillation counting: theory and applications

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Liquid scintillation spectrometry (LSC) is a widely applied counting technique in the fields of radiation protection, environmental radionuclide determination, geochemical and biochemical applications and, furthermore for standardization purposes of various radionuclides that are difficult to be measured by means of other counting techniques. LSC is based on the interaction of particle radiation (alpha, beta) with an organic scintillation liquid, the fluor molecule that will release photons (light) from an excited state back to the ground state. In contrast to other counting techniques for determining particle radiation, sample preparation is rather simple: a liquid phase containing the radioisotope to be measured is homogenized with a scintillation liquid in a liquid scintillation vial.

Light emitted from this cocktail solution is transferred to a photomultiplier tube, releasing an electronic pulse which is processed in a multichannel amplifier.

Starting from measurement of tritiated water and radiocarbon in the early 50ies, the LSC technique was continuously improved. Meanwhile background noise from cosmic radiation is largely reduced by electronic coincidence and anticoincidence circuits and alpha/beta separation techniques were developed by electronic pulse shape analysis and improvement of alpha/beta cocktails. The latest development of LSC is the application of triple to double coincidence ratio (TDCR) measurement, which allows in several cases direct quantification of measured count rates into activity. The presentation on LSC will focus on basic principles, latest developments such as TDCR measurement and alpha/beta separation and applications in radiochemistry as well as for radionuclide monitoring in the nuclear fuel cyce and the environment.

Alpha, beta and gamma measurements of environmental radioactivity

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Monitoring of environmental radioactivity started at Risø in 1956 and detection of radioactivity in samples was first based on commercial GM counters. These were later replaced by low-level GM counters produced at Risø and supplemented with NaI and Ge(Li) detectors. The present version of the Risø low-level multicounter system is a gas-flow unit incorporating five individual GM sample counter elements and a common guard counter. The guard counter reduces the cosmic-ray background using anti-coincidence technique. A sample slide allows five samples to be inserted into the multicounter where they are counted simultaneously. The counter is placed in a 10-cm lead shield to reduce the ambient background.

NaI well detectors are used in connection with analysis of ^{90}Sr and ^{99}Tc in environmental samples for the purpose of determining chemical yields of the radiotracers, ^{85}Sr and ^{99m}Tc. Determination of ^{90}Sr and ^{99}Tc is carried out by beta counting in multicounters.

Gamma-emitting radionuclides in environmental samples are determined by gamma spectrometry using Ge detectors. The Ge detectors used for monitoring purposes are placed in 10-cm lead shields, cover efficiencies in the range 25-39% and include low gamma energies. Samples range by size and geometry from sub-gram amounts in well geometries to kilogram amounts in Marinelli beakers. Software for analysis of gamma spectra was developed in-house and calibration is based on measurement of mixed nuclide solutions. The gamma analysis includes corrections for true coincidence summing effects and sample density.

Alpha-emitting radionuclides are determined by alpha spectrometry using Si detectors. Vacuum chambers each hold 8 detectors.

Actinide speciation analysis using mass spectrometry and laser spectroscopy

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Many radionuclides have a very rich – some may call it complicated - solution chemistry. This is particularly true for some of the actinides. Under certain chemical conditions as many as four different oxidation states of Plutonium are present in solution simultaneously, each of them having unique properties. Consequently, quantification of the chemical species with regard to oxidation states, complexation, polymerization and also sorption onto or incorporation into solids needs to be investigated, and that in short is called speciation.

Chemical species can vary over a wide size range, from small ions or molecules with sizes of less than 1 nm via small molecules with simple ligands to macromolecules, organometallic compounds, polymeric species, nano-particles and colloids ranging from 1 nm all the way to macroscopic mineral particles and other solid phases with sizes of more than 1µm. The species of different size ranges are related to each other, often in terms of chemical equilibria. One simple example is the equilibrium between two oxidation states determined by the redox potential. An example from the upper size range is the solubility of a defined solid phase in contact with an (aqueous) solution, which determines the amount of atomic/ionic species in solution for defined chemical conditions of the liquid phase.

No single analytical technique is capable of measuring a complete speciation but typically a multitude of complementary characterization techniques are required. The presentation will introduce the speciation methods Electrospray Mass Sepctrometry (ESI-MS) for the quantification of molecules in solution, Time-Resolved Laser Spectroscopy (TRLFS) for chemical speciation of luminescent probes in liquid and solid phase, Laser-Induced Photoacoustic Spectroscopy (LPAS) for trace concentration oxidation state analysis and Laser-Induced Breakdown Spectroscopy (LIBD) for quantification of aquatic colloids.

Speciation analysis of radionuclides in the environment

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Naturally occurring and artificially produced radionuclides in the environment can be present in different physico-chemical forms (i. e. radionuclide species) varying in size (nominal molecular mass), charge properties and valence, oxidation state, structure and morphology, density, complexing ability etc. Low molecular mass (LMM) species are believed to be mobile and potentially bioavailable, while high molecular mass (HMM) species such as colloids, polymers, pseudocolloids and particles are considered inert. Following deposition in the environment, time dependent transformation processes influencing the radionuclide species will occur, such as particle weathering and mobilization of radionuclide species from solid phases or interactions of mobile and reactive radionuclide species with components in soils and sediments. Therefore, the original distribution of radionuclides deposited in ecosystems will change over time and influence the ecosystem behaviour.

To assess the environmental impact from radionuclide contamination, information on radionuclide species deposited, interactions within affected ecosystems and the time-dependent distribution of radionuclide species influencing mobility and biological uptake is essential. The development of speciation techniques to characterize radionuclide species in waters, soils and sediments should therefore be essential for improving the prediction power of impact and risk assessment models. The present paper will focus on analytical techniques which should be utilised for characterizing radionuclide species in the environment.

Methodology and Application of Automation in Radiochemical Separations and Analysis

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Radiochemical analysis relies on chemical separations to isolate the radionuclides of interest from the matrix and other interfering radionuclides. Chemical separations by classical methods such as solvent extraction or precipitation are time-consuming and difficult to automate. Column-based separation, using ion exchange, or more recently, extraction chromatography, provide more efficient separations and they can be automated. Automation approaches provide considerable value in the laboratory, in terms of reducing labor, costs and worker exposure, as well as increasing throughput and capacity. However, automation is also valuable as a means to make measurements at-site or *in situ* for monitoring nuclear processes or the environment. In this lecture, robotic, fluidic and hybrid automation approaches will be described, giving examples in fission product analysis and actinide analysis as they apply to laboratory, process and environmental settings.

Automation and Methodology Development for Environmental and Biological Determination of Pu, Np, U and Tc

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This presentation overviews the recent development in automation and methodology for determination of Pu, Np, U and Tc in environmental and biological samples in DTU Nutech. Anion exchange (AG-1, AG MP-1) and extraction (TEV, UTEVA) chromatography were used for chemical purification of target radionuclides followed by detection with inductively coupled plasma mass spectrometry (ICP-MS) or accelerates mass spectrometry (AMS). Flow based techniques including flow injections (FI), sequential injection (SI), lab-on-valve bead injection (LOV-BI) were exploited for automation of the analytical processes.

For environmental assays, several analytical methods have been established for Pu, Np and U determination with sufficient reliability and high sample throughput (3-6 samples/day), which show high potential applicability in emergency preparedness as well as environmental monitoring. Moreover, a joint method has recently been developed for Pu, Np, U and Tc simultaneous determination in large volume (200 L) of seawaters, wherein an FI system was constructed for multi-sample processing in order to improve the sample throughput.

For bioassays, analytical methods were established for Pu and Np determination in large volume (≥1 L) of urine samples, wherein efforts were contributed to develop optimal sample pre-concentration methods. Several pre-concentration techniques including evaporation, co-precipitation with calcium phosphate, bismuth phosphate, iron hydroxide, manganese hydroxide, etc. were investigated and compared in detail. Finally, the analytical protocol based on manganese hydroxide co-precipitation was selected as the optimal.

Isotope dilution analysis of Sr-90 using dynamic reaction cell inductively coupled plasma mass spectrometry (IDA-DRC-ICP-MS)

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Sr-90 appears as a radionuclide in the decay series of nuclear fission and can therefore be found in irradiated nuclear fuel. Current methods for the detection of this radionuclide are time consuming. In this work, inductively coupled plasma mass spectrometry (ICP-MS) is used for the determination of Sr-90. Such a measurement is however interfered by Zr-90, a fission product and a component of the UO_2 fuel cladding material, at the same mass to charge ratio m/z. A Perkin Elmer Elan 6100 DRC II ICP-MS equipped with the dynamic reaction cell (DRC) is used to suppress the isobaric overlap by using O_2 as a reaction gas. Sr-90 standard usually consists of naturally occurring Sr isotopes as a carrier in much higher concentrations than the Sr-90 concentration. The high presences of naturally Sr in the Sr-90 standard depress the plasma of the ICP-MS, giving a poor intensity for the standards compared to the samples. Therefore an isotopic dilution analysis (IDA) method was developed for the analysis of Sr-90 using the DRC-ICP-MS technique. A Sr-86 isotopic standard was used for spiking of the samples derived from irradiated UO_2 fuel leached in synthetic ground water or irradiated UO_2 fuel dissolved in acids.

A simultaneous pre-concentration procedure of Sr and Pu from seawater using precipitation

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Artificial radionuclides are released into the environment as a result of nuclear facility accidents and nuclear weapon testing. Among them, strontium and plutonium are the most frequently monitored in environmental studies. Strontium and Plutonium are beta and alpha emitters, respectively. The chemical separation is necessary to determine these radionuclides. In particular a large amount of seawater is needed, because these radionuclides exist at an ultra-trace concentration in the environment. Before the purification process, the volume of the sample should be reduced based on the optimal size of the instrument. For an analysis of strontium in seawater, carbonate precipitation or the cation exchange resin process is available. The co-precipitation process using $Fe(OH)_3$ or MnO_2 has been used for a Pu analysis in seawater. After these pre-treatments, the seawater sample is purified by Sr-resin, anion exchange resin and TEVA.

We studied a simultaneous pre-concentration process of Sr and Pu in a large seawater sample. After the pH of the sample was adjusted to pH 9, plutonium was coprecipitated with iron hydroxide and strontium was concentrated as a form of strontium carbonate (Fig 1). A 1L volume of the seawater sample was reduced by 20ml, having 70-80% recovery for strontium (n=3) and 60-80% for plutonium (n=2), within 3hr. Some Ca and Mg ion in seawater were precipitated as a form of carbonate precipitate, because K_{sp} of $CaCO_3$ and $MgCO_3$ are $5.16x10^{-6}$ and $6.8x10^{-6}$, which are higher than that of $SrCO_3$. After this pre-concentration process, Sr and Pu could be purified by an extraction chromatography using Sr-resin, TEVA, etc. This approach can be applied for larger volume of seawater such as 5-10L of seawater.

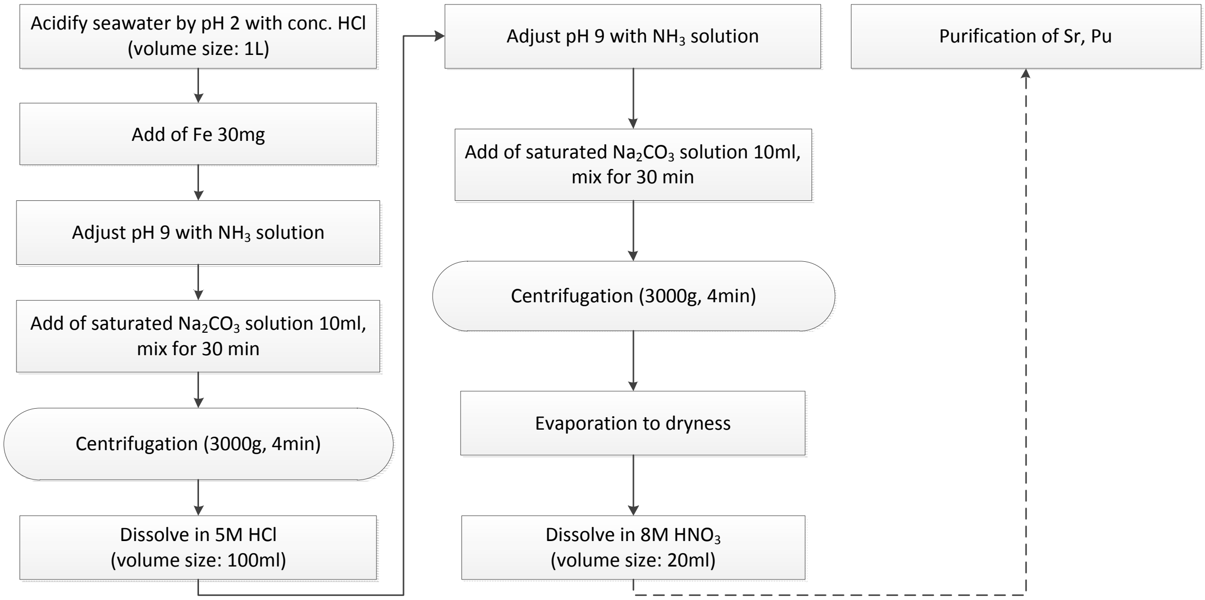

Figure 1 Procedures of pre-concentration of Sr and Pu in seawater

An Overview of Methods for Nuclide Specific NPP Release Monitoring

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Introduction

As a nuclear reactor is run, radioactive nuclides are generated. There are three main paths behind that generation in a light water reactor: the fission and activation of the nuclear fuel, the activation of corrosion products and other materials near the reactor core, and the activation of the reactor coolant (hydrogen and oxygen – water).

A nuclear power plant inevitably releases some amount of radioactive nuclides to the environment. These effluents are directed to defined release points, and are closely monitored and reported to the proper national authority. This presentation will provide an overview of the methods for sampling and analysis that are used at the Forsmark NPP (three BWR units) in Sweden. The annual dose consequence to a member of the public, because of the effluents from the three power units, is reported to the Swedish Radiation Safety Authority (SSM). A normal value for Forsmark is about 0,00015 mSv/year. Most of that dose is due to the release of C-14.

The radioactive nuclides that are monitored and released can be divided into the groups below.

Effluents to the air through the main stack

C-14 and H-3 are created from the neutron activation of the reactor coolant. The sampling is integrated by the continuous capture of C-14 and H-3 in bottles with NaOH solution and water respectively. The analysis is done with liquid scintillation counting.

Aerosols and Iodines are collected as integrated samples in filter cartridges: aerosols of activation products such as Co-60, Co-58 and Mn-54; transuranium nuclides such as Pu-239 and Cm isotopes; fission products such as I-131, Cs-137 and Sr-90. Analyses are mainly done with gamma and alpha spectrometry. Sr-90 requires chemical separation and time for radioactive equilibrium before repeated measurements are done on the Y-90 daughter to plot its decay.

Radioactive noble gases (mainly isotopes of Xe and Kr) are measured with on-line gamma spectrometry on stack air. The measurement time of evaluated spectra may be changed retroactively. For reporting purposes the evaluation is done from a spectrum with a one month measurement time.

Effluents to the water recipient (Baltic Sea)

The radioactive nuclides released to the water recipient are largely the same as to the air, except for noble gases and C-14 which are released almost exclusively to the air. For all liquid discharges, a proportional sample is collected. H-3 is the most prominent of the nuclides released to water, but its dose conversion factor is low.

Ni-63 and Fe-55 are additional nuclides that need to be reported for liquid discharges. Ni-63 is analyzed in the reactor coolant. It is separated from other beta emitters and measured with liquid scintillation counting. For reporting purposes it is assumed that the relation Ni-63/Co-60 is the same in the discharged water as in the reactor coolant. Theoretical vector calculations are done for Sr-89 and Fe-55 which are not analyzed at all.

Future needs

The reporting of effluent data to the SSM follows the regulation SSMFS 2008:23 and the recommendations of the European Union Commission, 2004/2/Euratom. The regulation is about to be updated with higher demands, and that development will continue in the revisions to come. Several of the analytical methods that are used at Forsmark NPP have been the same for 20–30 years. During that time better methods have become available. Future needs for improvement may include:

- Chemical separation before alpha spectrometry measurements. The current method does not allow the separation of interfering nuclides such as Pu-238/Am-241 and Po-210/Am-243.
- Improved separation of Ni and Co before the measurement of Ni-63. Co-58 and Co-60 cause interference that is difficult to compensate for without an increase of the combined uncertainty of the analysis.
- Improved method for Sr-90 analysis. The current method is based on liquid-liquid extractions of Y and sometimes suffers from interference, probably by Ba-140 and La-140.
- Capability to measure nuclides that are currently calculated by the use of nuclide vectors or other methods. Such nuclides could be Fe-55, Sr-89, Ni-59 and Kr-85.

Mathematical efficiency calibration in gamma spectrometry for analysis in natural and fallout radionuclides in sediments

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Traditional approach in gamma spectrometric efficiency calibration involves measurement of test sources in order to obtain efficiencies for given geometries. In the IUP laboratory experimental efficiency curves for each detector have been constructed for "standard geometries" (including Marinelli beakers, plastic cylindrical dishes, plastic bottles filled into defined heights and filters) using standards with gamma-emitters mixture with a wide range of energies (for densities 1 $g \cdot cm^{-3}$ and 1.5 $g \cdot cm^{-3}$).

Sediment samples are frequently measured in the IUP laboratory in order to provide recent chronologies of sediment cores based on natural and artificial fallout radionuclides. As the sediment material originates from different environments and from different sources, it develops enormous variability of compositions and densities. The matrix of the sample is then hardly comparable to the standard samples matrix, which may lead to introducing additional error to efficiency calibration.

For density correction of samples with density different from that of "standard geometries", it is possible to distribute the sample material in a larger volume of inactive material (paraffin powder) and prepare pressed pellets of geometries and densities close to those standard ones. This is a good approach for materials with higher activities, for which dilution does not play an important role, but it is a destructive method. Also it does not take into account changes of self absorption due to elemental composition of the sample matrix.

As an alternative non-destructive approach for sediment samples, the suitability of Canberra LabSOCS mathematical efficiency calibration for factory characterized detectors was tested. Here the efficiency is calculated based on precisely described sample geometry, density and elemental composition.

A series of validation tests involving point and voluminous sources in different geometries.

1. point sources of ^{137}Cs measured in different distances from the detector,
2. a voluminous source (^{137}Cs solution) measured in different distances from the detector,
3. ^{210}Pb and ^{137}Cs points sources and different absorbers used for transmission measurements and
4. a soil sample with known ^{137}Cs, ^{40}K and ^{226}Ra activity measured in different geometries.

The first two experiments were used in order to validate mainly the geometrical correctness of the calibration. The third test was designed to check mainly the absorption correction for different energies. The last test combines validation of both previously tested properties. The evaluation and results will be presented.

Inhibition of radiation induced dissolution of UO_2 by sulfide - a comparison with the hydrogen effect [1]

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Nuclear power has constituted a major fraction of the energy production in several countries for many years and will play an increasingly significant role in the future since many nuclear plants of high installed capacity are under construction. Nuclear power has a number of advantages in comparison with other forms of energy production; however, there is a major problem: namely, how to minimize the potential impact of the high level radioactive spent nuclear fuel. Reprocessing is one way of dealing with the problem.

In Sweden, the KBS-3 model, developed by SKB (The Swedish Nuclear Fuel and Waste Management Co) will be adopted to store the radioactive spent nuclear fuel in a deep geological repository. The copper canisters with an insert of steel and spent fuel inside will be surrounded by bentonite clay and placed 500 m under the ground. One of the fundamental issues for the safety assessment of the SNF (spent nuclear fuel) geological repository is the rate of dissolution of UO_2, which is the matrix of SNF (approximately 95% of the fuel), and the rest radioactive fission products and actinides. In case of canister damage, although it is not expected to occur in 1000 years, the highly radioactive SNF will induce radiolysis of the groundwater. Both oxidants ($OH\cdot$,H_2O_2, $HO_2\cdot$ and O_2) and reductants (e_{aq}^-,$H\cdot$ and H_2) will be produced in the radiolysis process. H_2O_2 oxidizes UO_2 to soluble UO_2^{2+} and therefore increases the rate of SNF dissolution. The UO_2^{2+} will form soluble complexes with carbonate present in the groundwater [2].

In this work we have studied the influence of the groundwater component H_2S/HS^- on radiation induced dissolution of spent nuclear fuel using simple model systems. The reaction between H_2O_2 and H_2S/HS^- has been studied experimentally as well as the effect of H_2S/HS^- on γ-radiation induced dissolution of a UO_2 pellet. The experiments clearly show that the reaction of H_2O_2 and H_2S/HS^- is fairly rapid and that H_2O_2/H_2S stoichiometry is favorable for inhibition. Radiolysis experiments show that H_2S/HS^- can effectively protect UO_2 from oxidative dissolution. The effect depends on sulfide concentration in combination with dose rate. Autoclave experiments were also conducted to study the role of H_2S in the reduction of U(VI) in the presence and absence of H_2 and Pd particles in anoxic aqueous solution. The aqueous solutions were pressurized with H_2 or N_2 and two different concentrations of H_2S/HS^- was used in the presence and absence of Pd. U(VI) reduction was found to be proportional to H_2S concentration in H_2 and N_2 atmosphere. It is clearly shown the Pd catalyzed H_2 effect is more powerful than the effect of H_2S/HS^-. H_2S/HS^- poisoning of the Pd catalyst is not observed.

References:

[1] M. Yang et al. J. Nucl. Mater., 434(2013)38-42
[2] S. Nilsson, M. Jonsson, J.Nucl. Mater., 374(2008)290.

ADVANCED SEPARATION TECHNIQUES FOR RADIONICKEL

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A large volume of radioactive waste waters is produced by nuclear power plants. One of these are decontamination solutions that typically contains large amount of inactive metal ions (K^+, Na^+, Fe^{3+}, Ca^{2+}) and only trace amount of radioactive metals, usually locked in some complexing agents such as oxalate. For reduction of negative impact on the environment it is needed to find efficient separation technique of these radionuclides.

The aim of this work was to develop advanced separation technique for removal of radionickel (Ni-63), which is one of the activation products in the reactor materials of nuclear power plants. The separation technique is based on the degradation of formed metal complexing agents like EDTA and adsorption of released metal on to the ion exchanger. EDTA forms a strong complex with many metals. These metal-EDTA complexes (Ni-EDTA also) are stable and quite inert to chemical treatments. The synergy of photocatalytic activity of an inorganic ion exchanger (like TiO_2, SnO_2) and UV/H_2O_2 has been seen as a possibility to find a specific method of separating radionickel.

Some metal oxides are tested for the removal of radionickel in the decontamination solution simulates. Preliminary results using SnO_2 and UV light irradiation resulted in high cobalt uptake from Co-EDTA containing solution. The oxidative destruction of nickel complexes is studied with this inorganic material as a photocatalytic semiconductor under UV light irradiation.

Plutonium isotopes in Chinese soils and its potential application for tracing soil erosion

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Based on high association of ^{137}Cs to soil particles (mainly minerals), global fallout ^{137}Cs has been widely used for soil erosion in the past decades. However, considering the relatively short half-life of 30.2 years, the application of ^{137}Cs for investigations of soil erosion, transportation and sedimentation will be impractical since more than 60% of the original inventory of global fallout ^{137}Cs has decayed. Plutonium isotopes (^{239}Pu and ^{240}Pu with half-lives of 24110 yr and 6561 yr) were suggested as ideal substitutes of ^{137}Cs for investigation of soil erosion, due to their long half-lives, dominating source of global fallout worldwide, as well as their high retention and low mobility in soil. Especially, with the rapid development of more sensitive measurement techniques of mass spectrometry including ICP-MS and AMS in the recent years, application of Pu isotopes for this investigation becomes more attractive and competitive.

In this work, surface soil samples and soil cores collected from northeast China were analyzed using radiochemical separation and ICP-MS for plutonium isotopes. The distribution, source term and environmental behavior of Pu in soils in northeast China were presented. Integrated with the data of ^{137}Cs, the feasibility of applying Pu isotopes for soil erosion, transport and sedimentation studies was explored. The data sets also provided background baseline data for further monitoring and evaluation of any unexpected contamination of anthropogenic radionuclides in the area in the future.

Effect of bentonite on radiation induced dissolution of UO_2 in an aqueous system

Alexandre Barreiro Fidalgo, Sara Sundin, Mats Jonsson

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In order to elucidate the impact of bentonite on the process of radiation induced oxidative dissolution of UO_2 in an aqueous system, the dissolution of U(VI) and consumption of H_2O_2 over time has been studied. In addition, γ-irradiation experiments were performed to study a more relevant and complex system, serving as a comparison with the previously stated system. In both cases, the experiments revealed that the presence of bentonite in water could either delay or prevent in part the release of uranium to the environment. The cause is mainly attributed to the scavenging of radiolytic oxidants rather than to the adsorption of uranium onto bentonite.

Sorption of europium(III), nickel(II) and calcium(II) on α-Al_2O_3, TiO_2 and ZrO_2

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Understanding radionuclide sorption mechanisms on mineral surfaces is important when considering the long-term safety of nuclear waste disposal. The main two sorption mechanisms of metals on a mineral surface are cation exchange and surface complexation, i.e. formation of outer- and inner-sphere complexes, respectively [1]. Outer-sphere complexation of a positively charged metal cation occurs on a negatively charged surface, while inner-sphere complexes form on amphoteric surface hydroxyl groups through chemical bond formation between the metal cation and surface oxygen atoms.

In the present study, the sorption of europium(III), nickel(II) and calcium(II) onto α-Al2O3, TiO2 and ZrO2 have been studied as a function of pH. The aim of the study was to determine how pH, metal ion properties, and surface characteristics of the different minerals affect the sorption behavior of the chosen metals. Trivalent europium is used as a chemical analogue to the trivalent actinides Pu3+, Am3+, and Cm3+. The divalent activation product 59Ni is an important long-lived radionuclide in the spent nuclear fuel, whereas Ca2+ is abundant in the environment.

The different metal oxides, α-Al2O3, TiO2 and ZrO2 were selected for the study due to their different surface properties. The surface of TiO2 becomes negatively charged due to dissociation of surface hydroxyl protons in the acidic pH-range yielding ≡Ti-O- groups, while α-Al2O3 retains a positive surface charge due to protonated aluminol ≡Al-OH2+ groups even in the alkaline pH range. Thus, outer-sphere complexes could potentially form on the negative TiO2 surface, while this metal ion complexation mechanism is very unlikely to occur on the positive α-Al2O3 surface. The isoelectric point (IEP), where the net surface charge equals zero, was determined for each mineral by measuring the zeta potential as a function of pH in three different ionic strengths (MilliQ-water, 0.01M NaClO4 and 0.1M NaClO4). The value of pHIEP was measured to be at 9.7 ± 0.2 for α-Al2O3, 6.7 ± 0.5 for ZrO2, and 3.8 ± 0.3 for TiO2.

The batch experiments on Eu(III), Ni(II) and Ca(II) sorption on α-Al2O3, TiO2 and ZrO2 were done in a glove box under N2 atmosphere to exclude the formation of metal-carbonate complexes in the studied mineral suspensions. The samples were prepared using constant electrolyte and mineral concentrations of 0.01M NaClO4 and 2 g/l, respectively. The metal ion concentration was adjusted to 1·10-7 mol/l in the europium and nickel experiments, while the calcium concentration was higher at 25·10-4 mol/l. The pH of all three sample series was adjusted from pH 3 to 11. Figure 1 presents the surface area normalized sorption distribution coefficients log(Kd/As) as a function of pH for all three metal ions on the different mineral oxides. Sorption occurs first on titanium oxide, thereafter on zirconium oxide and finally on aluminum oxide, following the same order as the values of pHIEP. Furthermore, the higher retention of Eu3+ and Ni2+ on TiO2 in comparison to the other oxide minerals in the acidic pH range points to the existence of outer-sphere complexes on the negative TiO2 surface. The metal ions retain on the surfaces in the order Eu3+ > Ni2+ > Ca2+ following the trend of metal ion hydrolysis in solution. According to hydrolysis constants found in the NAGRA/PSI database [2] europium is hydrolyzed in solutions exceeding pH 6, while nickel hydrolysis begins only above pH 8. Calcium does not hydrolyze at all. Thus, both metal ion properties and mineral surface characteristics seem to play an important role in the retention of radionuclides on solid surfaces.

ADOPT pellet leaching properties, a comparison with UO_2 pellet

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According to the KBS-3 method, the spent nuclear fuel will be stored in a deep repository, 500 meters below ground level in the bedrock; kept in copper canisters that are surrounded by bentonite clay. In the event of a copper canister failure and groundwater diffusing through the bentonite clay, the spent nuclear fuel will come in contact with the groundwater. The UO_2 matrix will in that case govern the release of radionuclides into the biosphere. Understanding the oxidative dissolution behavior of UO_2 in water solution is of importance when assessing the safety of a deep repository [1].

ADOPT (Advanced Doped Pellet Technology) is a new type of fuel developed by Westinghouse with properties such as enlarged grain size and reduced Fission Gas Release (FGR). To achieve this UO_2 is doped with chromium and aluminum oxides [2]. These dopants could influence the leaching properties of the fuel compared to fuel based on pure UO_2.

In this work we have performed a kinetic and mechanistic study of ADOPT pellet leaching by evaluating the reactivity of H_2O_2 towards ADOPT pellets in aqueous solutions containing HCO_3^-. The dissolution of uranium has been studied in parallel with H_2O_2 consumption. The catalytic decomposition of H_2O_2 on ADOPT fuel has been quantified and compared to pure UO_2. In addition, studies of γ-irradiation induced dissolution of ADOPT pellets as well as the influence of H_2 on the oxidative dissolution process have been carried out. The Arrhenius parameters for the reaction between H_2O_2 and ADOPT fuel have also been determined.

Furthermore, leaching experiments of irradiated ADOPT fuel have been performed. The results of these leaching studies are compared to numerical simulations of radiation induced dissolution of spent nuclear fuel taking the results from the model studies above into account.

References:

[1] Jonsson, M., Radiation Effects on Materials Used in Geological Repositories for Spent Nuclear Fuel. ISRN Materials Science, 2012. 2012: p. 13.

[2] Arborelius, J., et al., Advanced doped UO2 pellets in LWR applications. Journal of Nuclear Science and Technology, 2006. 43(9): p. 967-976.

Quench correction using Triple to Double Coincidence Ratio (TDCR) method on Liquid Scintillation Counting (LSC)

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In LSC measurements signal is often quenched by color or chemicals such as salts or by water, yielding in need for quench correction. Until recently the most typical quench correction method has been External standard method requiring counter equipped with radioactive gamma source, and preparation of quench curve with radioisotope on interest in same conditions as the unknown samples. In the presentation I introduce triple-to-double-coincidence-ratio (TDCR) method, which is an absolute counting method for obtaining the results automatically quench corrected without external radioactive source. The method requires triple coincidence detector and it's been studied by metrology instituted since late 70's when it was published by two Polish scientists. TDCR method has been available commercially since launching of HIDEX 300 SL™ TDCR Liquid Scintillation counter on 2008.

Analysis and measurements of ^{226}Ra in drinking water used in Caspian area

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Analysis and measurements of radium concentrations in drinking water are important from the human health of view. Therefore simple and reliable analytical methods must be available. In this research we have determined the radium activity concentration in 22 points used in Caspian area. For this purpose we used radon precipitation technique and also alpha spectrometry method. This work gives details of sample preparation and concentration between 2.1 mBq/L to38.2 mBq/L. These values were compared with the recommended safe limits for drinking water.

Evaluation of ”double separation” to avoid interference by Co in the analysis of Ni-63

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Ni-63 is measured at the nuclear power plant in Oskarshamn, Sweden, since 2011. Reactor water, spent fuel pool water and waste water are monitored for e.g. Ni-63 quarterly and are reported to the Swedish Radiation Safety Authority in the annual reports. Preliminary results have earlier been presented at the annual Mätmöte with participants from nuclear power plants in the Nordic countries, March 2013.

The analytical method for Ni-63 is based on double solid phase extraction. In a first step iron and actinides are removed using a TRU-resin from Eichrom and in a second step a nickel specific resin is used to selectively separate nickel from the rest of the elements in the samples. Ni-63 is measured using liquid scintillation and the yield of the analysis is determined by addition of inactive nickel and measurements by ICP-OES. Interference from Co-60 in the liquid scintillation measurement is corrected for using a quench correction curve, whereas no corrections for other nuclides are done.

Co-58 was early identified as a potential interfering nuclide in the original method that was developed in cooperation with FOI in Umeå. The results for quarter four in 2011 showed significantly higher activity levels for Ni-63 than earlier samples. Co-58 decays by beta and has a half-life of 71 days. Three options were identified to handle the situation:

- To wait with the analyses until most of the Co-58 has decayed
- To adjust for Co-58 with a quench correction curve in the same way as for Co-60
- To use a so called “double separation” in which the samples are allowed to pass the nickel specific column twice instead of once in combination with a reasonable time between sampling and analysis.

The first option was not considered as possible since the authorities want their reports on time. The second option, although a bit complicated due to the relative short half-life, would be possible but increases the total uncertainty of the analyses. The third option was investigated in this work.

Two different samples were analyzed using single and double separation. In the first sample the content of Co-60 was high and Co-58 was low. In the second sample the opposite situation prevailed. Double separation results in that a smaller amount of Co-58 is accounted for as Ni-63. This is especially important for samples with a high amount of Co-58. Also the correction done for Co-60 using the quench correction curve will be smaller and thus giving less uncertainty to the final determination of Ni-63.

In addition three samples were analyzed using single separation but at four different occasions with approximately one year in between the first and last analyses. The results from the long-time test shows that the activity levels of Ni-63 in the samples are directly proportional to the Co-58 activity levels in the eluate.

The combination of waiting with the analysis until some of the Co-58 has decayed (at least six weeks) and using double separation ensures that the results for Ni-63 are not overestimated by more than 20 %. This recommendation has now been introduced at OKG. The extra time needed in each analysis is approximately two hours.

Methodologies for determination of different types of radionuclides in radiological emergencies and their behavior

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The large number and construction of Spallation Sources emphasizes the requirement to develop rapid and reliable methodologies for determination of different types of radionuclides related to Spallation Sources. Radionuclides such as radioberyllium (^{7}Be), tritium (^{3}H), radioiron (^{55}Fe) and radiosodium (^{22}Na), referred as hard-to-detect (HTD) radionuclides are produced by the target station inside the Spallation Neutron Source. These HTD radionuclides are found in the concrete and in the first 20 cm of the neighbouring soil used for shielding of the accelerator tunnel. The HTD radionuclides could be dispersed to the surroundings by some type of accident. The areas close to Spallation Sources are frequently this type used for intensive agricultural production and there are concerns if a release of these HTD radionuclides could contaminate these areas. On that account, there is a significant importance to develop rapid techniques for determination of HTD radionuclides.

In the example of the ESS in Lund the estimated amount of HTD radionuclides that will be produced in the concrete that are used for shielding are; 2.45 Bq g^{-1} for ^{7}Be, 44.7 Bq g^{-1} for ^{3}H, 5.1 Bq g^{-1} for ^{55}Fe and 3.92 Bq g^{-1} for ^{22}Na. In the surrounding first 20 cm soil the corresponding numbers are estimated to be; 0.06 Bq g^{-1} for ^{7}Be, 0.60 Bq g^{-1} for ^{3}H, 1.21 Bq g^{-1} for ^{55}Fe and 0.096 Bq g^{-1} for ^{22}Na. These HTD radionuclides could be scattered to the surroundings by some kind of accident. Areas close to these types of spallation facilities are often used for intensive agricultural productions and there can be concerns if any kind of release of these HTD radionuclides could contaminate these areas.

The areas used for agricultural production can be of high risk for a potential release of HTD radionuclides to them. There is a need to acquire better understanding on the colloidal transport and migration of HTD radionuclides in agricultural soils. This can be achieved by study the transportation of HTD radionuclides by using *e.g.* soil columns (lysimeters), where the HTD radionuclides are placed on the soil surface and can then be studied for their behaviour on migration.

In earlier studies the focus have been on radionuclides that can be released from nuclear power plant accidents or nuclear bomb explosions *e.g.* radiocaesium (^{134,137}Cs), plutonium (^{239}Pu), americium (^{241}Am), neptunium (^{237}Np) and radiostrontium ($^{89,\ 90}$Sr). For those radionuclides a great number of preparedness methods have been developed *e.g.* preparedness methods for ^{89}Sr and ^{90}Sr by Tovedal *et al.* (2009). These radionuclides are also rather understood for their behaviour in the environment, *e.g.* migration in different soils. However, there is a lack of knowledge of preparedness methods and the behaviour of HTD radionuclides at a release to the environment.

The aim of the presented project is to develop and to study the behaviour of radioberyllium, tritium, radioiron and radiosodium. The research objectives of the project proposal are to: (1) Develop and test methods for estimation of these HTD radionuclides after accidental release to the environment. (2) Estimate the robustness of the methods at a “real” release of radioberyllium, tritium, radioiron and radiosodium to the environment. (3) Get an understanding of the behaviour of radioberyllium, tritium, radioiron and radiosodium in agricultural soils.

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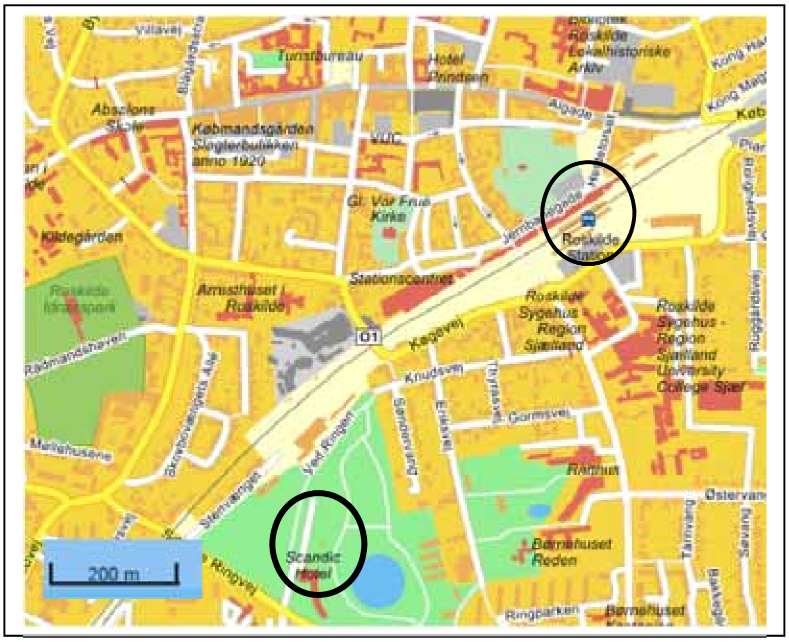
Transport to Workshop at Risø, Roskilde, Denmark

From Airport to Roskilde:

There is train connection from Copenhagen Airport to Roskilde station (Railway) through Copenhagen Central Station, it takes 40-50 min. The train departs from the airport (Kastrup Lufthavn) 3-4 times per hour, and cost about 100 DKK. You can find your time schedule from the website: http://www.rejseplanen.dk/bin/query.exe/en. The cost of a taxi from Copenhagen Airport to Roskilde is approx. DKK 700.



From Roskilde Station to your Hotel:

Scandic Hotel Roskilde is about 1.2 km to Roskilde station, about 15 min. walk (see the map, you can also find a big map from Google map).

From Hotel to Risø

A bust transport will be arranged for take the participants from the Scandic Hotel every morning at 8:00am to Risø, and send back from Risø every afternoon at 16:30 to the Hotel (16:00 on Friday). In Thursday afternoon, the bus will leave at 17:30 from Risø to Roskilde harbor for a tour and dinner in the Roskilde Fjord, and send the participants back to Hotel at 21:00.

Meeting place at Risø

The workshop on 5th and 6th Sept. will be hold in big meeting room in building 115 at Risø, and the lab practice in 2-4th Sept. will be hold in building 202 and 204 at Risø.

Transport to Workshop at Risø, Roskilde, Denmark

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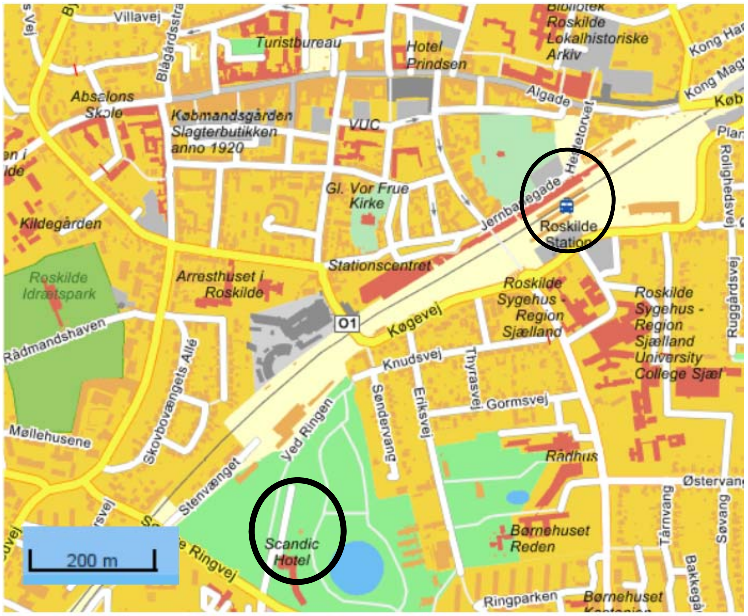

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Meeting place at Risø

The workshop on 5th and 6th Sept. will be hold in big meeting room in building 115 at Risø, and the lab practice in 2-4th Sept. will be hold in building 202 and 204 at Risø. The bus will directly transport all participants to the building 201 where they can leave their bags and clothes in the meeting room.

Time schedule of bus transport for workshop

Date	From Hotel to Risø (1)	Lunch at Risø Cantina *	From Risø to Hotel (3)
Monday/2nd Sept	8:00 am	12:00-13:30	16:30
Tuesday/3rd Sept.	8:00 am	12:00-13:30	16:30
Wednesday/4th Sept.	8:00 am	12:00-13:30	16:30
Thursday/5th Sept.	8:00 am	12:15-13:45	17:30 (2)
Friday/6th Sept.	8:00 am	12:15-13:45	16:00 (4)

1) All participants should meet in the front of the Scandic Hotel Roskilde before 8:00 am every morning and take bus to Risø.

2) In Thursday afternoon, the bus will leave at 17:30 from Risø to Roskilde harbor for a tour and dinner in the Roskilde Fjord, and send the participants back to Hotel at 21:30.

3) We will walk to the Risø Cantina from the laboratory building for lunch; it is about 1 km distance and takes about 10 min. for walk.

4) In Friday afternoon, the bus will leave Risø at 16:00 to the Roskilde station, then to the Scandic Hotel.

Note: To those do not stay in the Scandic Hotel and will transport to Risø themselves, please get you entrance card at Risø Gate (already registered to the Workshop on Radioanalytical Chemistry) . In 2-4th Sept, please go to the meeting room in building 201 before 8:30, all participants will get together in the meeting room first every morning, and start the lab practice afterwards. In 5-6th Sept., please directly go to the meeting room in the building 115 (nearby the entrance gate).

Hotel Information

The hotel rooms have been reserved to all participants in the Scandic Hotel Roskilde, and bus transport between Scandic Hotel and Risø have been arranged during workshop (2-6th Sept.). The participants have to book their own room in the hotel using the booking code (RIS010913) by e-mail or phone with a price of 1004 DKK per night for single room and 200 DKK more for double room. You might also book your room through internet with a lower price (950 DKK) , no booking code is needed for this booking, because the organizer could not make this agreement with the hotel, in this case you have to consider the charge for change and cancelation. The hotel rooms have already been booked for invited lecturers.

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Web: scandichotels.com
Booking code: RIS010913

Social Activities

A Roskilde Fjord tour will be organized in Thursday evening 18:00-21:30. The bus will leave from Risø at 17:30 directly to the Roskilde harbor, we will board about 18:00 to sailing in the beautiful Roskilde Fjord and have a dinner on the board. All participants are invited to join in for free. The temperature might be relative low in the evening (12-15°C), it will be nice to

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Project	NKS-B RADIOANALYSIS
No. of pages	50
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No. of illustrations	0
No. of references	20
Abstract max. 2000 characters	This report compiled all abstracts presented in the NKS Workshop on Radioanalytical Chemistry at Risø, Roskilde, Denmark in 2-6th Sept. 2013. Total 35 participants registered to the workshop, among them 18 from Sweden, 5 from Denmark, 3 from Finland, and 3 from Norway, there are also 6 participants from Germany, France, Slovenia, Korea, Turkey, and China. The workshop consists two part, 3 days lab practices and 2 days lectures/presentation. 3 lab practices were organized, i.e. (1) Radiochemical separation of Pu and ICP-MS measurement of Pu isotopes; (2) Radiochemical separation of ^{210}Po and ^{226}Ra and their alpha spectrometry measurement; and (3) Radiochemical separation of ^{55}Fe, ^{63}Ni, ^{90}Sr and their LSC measurement. Among them, each participant can participate in 2 lab practices. 15 invited lectures are given by the experts in their specific fields, 8 oral and 6 poster presentations are given by the participants. The abstracts of all presentations are included in this report.
Key words	Radioanalysis, radionuclides, workshop, lab practice

Available on request from the NKS Secretariat, P.O.Box 49, DK-4000 Roskilde, Denmark.
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